4. Industrial Processes and Product Use

The Industrial Processes and Product Use (IPPU) chapter includes greenhouse gas emissions occurring from industrial processes and from the use of greenhouse gases in products. The industrial processes and product use categories included in this chapter are presented in Figure 4-1. Greenhouse gas emissions from industrial processes can occur in two different ways. First, they may be generated and emitted as the byproducts of various non-energy-related industrial activities. Second, they may be emitted due to their use in manufacturing processes or by end-consumers.

In the case of byproduct emissions, the emissions are generated by an industrial process itself, and are not directly a result of energy consumed during the process. For example, raw materials can be chemically or physically transformed from one state to another. This transformation can result in the release of greenhouse gases such as carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and fluorinated greenhouse gases (e.g., HFC-23). The greenhouse gas byproduct generating processes included in this chapter include iron and steel production and metallurgical coke production, cement production, lime production, other process uses of carbonates (e.g., flux stone, flue gas desulfurization, and glass manufacturing), ammonia production and urea consumption, petrochemical production, aluminum production, HCFC-22 production, soda ash production and use, titanium dioxide production, ferroalloy production, glass production, zinc production, phosphoric acid production, lead production, silicon carbide production and consumption, nitric acid production, adipic acid production, and caprolactam production.

Greenhouse gases that are used in manufacturing processes or by end-consumers include man-made compounds such as hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), sulfur hexafluoride (SF_6), and nitrogen trifluoride (NF_3). The present contribution of HFCs, PFCs, SF_6, and NF_3 gases to the radiative forcing effect of all anthropogenic greenhouse gases is small; however, because of their extremely long lifetimes, many of them will continue to accumulate in the atmosphere as long as emissions continue. In addition, many of these gases have high global warming potentials; SF_6 is the most potent greenhouse gas the Intergovernmental Panel on Climate Change (IPCC) has evaluated. Use of HFCs is growing rapidly since they are the primary substitutes for ozone depleting substances (ODS), which are being phased-out under the Montreal Protocol on Substances that Deplete the Ozone Layer. Hydrofluorocarbons, PFCs, SF_6, and NF_3 are employed and emitted by a number of other industrial sources in the United States, such as semiconductor manufacture, electric power transmission and distribution, and magnesium metal production and processing. Carbon dioxide is also consumed and emitted through various end-use applications. In addition, nitrous oxide is used in and emitted by semiconductor manufacturing and anesthetic and aerosol applications.

In 2017, IPPU generated emissions of 358.9 million metric tons of CO_2 equivalent (MMT CO_2 Eq.), or 5.6 percent of total U.S. greenhouse gas emissions.[1] Carbon dioxide emissions from all industrial processes were 166.9 MMT CO_2 Eq. (166,872 kt CO_2) in 2017, or 3.2 percent of total U.S. CO_2 emissions. Methane emissions from industrial processes resulted in emissions of approximately 0.3 MMT CO_2 Eq. (11 kt CH_4) in 2017, which was less than 1 percent of U.S. CH_4 emissions. Nitrous oxide emissions from IPPU were 22.6 MMT CO_2 Eq. (76 kt N_2O) in 2017, or 6.3 percent of total U.S. N_2O emissions. In 2017 combined emissions of HFCs, PFCs, SF_6, and NF_3 totaled 169.1

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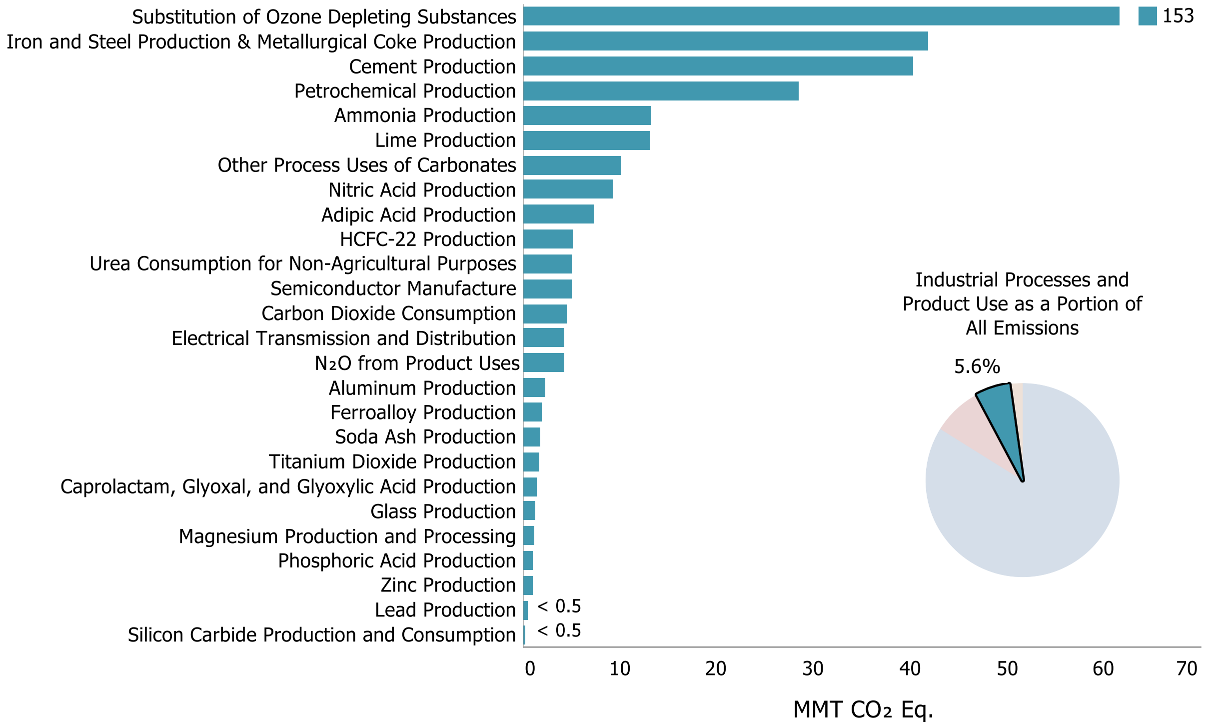
[1] Emissions reported in the IPPU Chapter include those from all 50 states, including Hawaii and Alaska, as well as from U.S. Territories to the extent of which industries are occurring.

MMT CO_2 Eq. Total emissions from IPPU in 2017 were 4.9 percent more than 1990 emissions. Indirect greenhouse gas emissions also result from IPPU, and are presented in Table 4-112 in kilotons (kt).

Figure 4-1: 2017 Industrial Processes and Product Use Chapter Greenhouse Gas Sources (MMT CO_2 Eq.)



The increase in overall IPPU emissions since 1990 reflects a range of emission trends among the emission sources. Emissions resulting from most types of metal production have declined significantly since 1990, largely due to production shifting to other countries, but also due to transitions to less-emissive methods of production (in the case of iron and steel) and to improved practices (in the case of PFC emissions from aluminum production). Similarly, CO_2 and CH_4 emissions from many chemical production sources have either decreased or not changed significantly since 1990, with the exception of petrochemical production which has steadily increased. Emissions from mineral sources have either increased (e.g., cement manufacturing) or not changed significantly (e.g., glass and lime manufacturing) since 1990 but largely follow economic cycles. Hydrofluorocarbon emissions from the substitution of ODS have increased drastically since 1990, while the emissions of HFCs, PFCs, SF_6, and NF_3 from other sources have generally declined. Nitrous oxide emissions from the production of adipic and nitric acid have decreased, while N_2O emissions from product uses have remained nearly constant over time. Some emission sources exhibit varied interannual trends. Trends are explained further within each emission source category throughout the chapter. Table 4-1 summarizes emissions for the IPPU chapter in MMT CO_2 Eq. using *IPCC Fourth Assessment Report* (AR4) GWP values, following the requirements of the current United Nations Framework Convention on Climate Change (UNFCCC) reporting guidelines for national inventories (IPCC 2007).[2] Unweighted native gas emissions in kt are also provided in Table 4-2. The source descriptions that follow in the chapter are presented in the order as reported to the UNFCCC in the Common Reporting Format (CRF) tables, corresponding generally to: mineral products, chemical production, metal production, and emissions from the uses of HFCs, PFCs, SF_6, and NF_3.

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[2] See <http://unfccc.int/resource/docs/2013/cop19/eng/10a03.pdf>.

Table 4-1: Emissions from Industrial Processes and Product Use (MMT CO_2 Eq.)

Gas/Source	1990	2005	2013	2014	2015	2016	2017
CO_2	**208.8**	**191.7**	**173.1**	**179.2**	**173.0**	**164.5**	**166.9**
Iron and Steel Production & Metallurgical Coke Production	101.6	68.2	53.5	58.4	47.8	42.3	41.8
Iron and Steel Production	*99.1*	*66.2*	*51.6*	*56.3*	*45.0*	*41.0*	*41.2*
Metallurgical Coke Production	*2.5*	*2.1*	*1.8*	*2.0*	*2.8*	*1.3*	*0.6*
Cement Production	33.5	46.2	36.4	39.4	39.9	39.4	40.3
Petrochemical Production	21.2	26.8	26.4	26.5	28.1	28.1	28.2
Ammonia Production	13.0	9.2	9.5	9.4	10.6	10.8	13.2
Lime Production	11.7	14.6	14.0	14.2	13.3	12.9	13.1
Other Process Uses of Carbonates	6.3	7.6	11.5	13.0	12.2	11.0	10.1
Urea Consumption for Non-Agricultural Purposes	3.8	3.7	4.6	1.8	4.6	5.1	5.0
Carbon Dioxide Consumption	1.5	1.4	4.2	4.5	4.5	4.5	4.5
Ferroalloy Production	2.2	1.4	1.8	1.9	2.0	1.8	2.0
Soda Ash Production	1.4	1.7	1.7	1.7	1.7	1.7	1.8
Titanium Dioxide Production	1.2	1.8	1.7	1.7	1.6	1.7	1.7
Glass Production	1.5	1.9	1.3	1.3	1.3	1.2	1.3
Aluminum Production	6.8	4.1	3.3	2.8	2.8	1.3	1.2
Phosphoric Acid Production	1.5	1.3	1.1	1.0	1.0	1.0	1.0
Zinc Production	0.6	1.0	1.4	1.0	0.9	0.9	1.0
Lead Production	0.5	0.6	0.5	0.5	0.5	0.5	0.5
Silicon Carbide Production and Consumption	0.4	0.2	0.2	0.2	0.2	0.2	0.2
Magnesium Production and Processing	+	+	+	+	+	+	+
CH_4	**0.3**	**0.1**	**0.1**	**0.2**	**0.2**	**0.3**	**0.3**
Petrochemical Production	0.2	0.1	0.1	0.1	0.2	0.2	0.3
Ferroalloy Production	+	+	+	+	+	+	+
Silicon Carbide Production and Consumption	+	+	+	+	+	+	+
Iron and Steel Production & Metallurgical Coke Production	+	+	+	+	+	+	+
Iron and Steel Production	+	+	+	+	+	+	+
Metallurgical Coke Production	*0.0*	*0.0*	*0.0*	*0.0*	*0.0*	*0.0*	*0.0*
N_2O	**33.3**	**24.9**	**21.0**	**22.8**	**22.3**	**23.6**	**22.6**
Nitric Acid Production	12.1	11.3	10.7	10.9	11.6	10.1	9.3
Adipic Acid Production	15.2	7.1	3.9	5.4	4.3	7.0	7.4
N_2O from Product Uses	4.2	4.2	4.2	4.2	4.2	4.2	4.2
Caprolactam, Glyoxal, and Glyoxylic Acid Production	1.7	2.1	2.0	2.0	2.0	2.0	1.4
Semiconductor Manufacture	+	0.1	0.2	0.2	0.2	0.2	0.2
HFCs	**46.6**	**122.3**	**146.1**	**150.7**	**153.8**	**155.0**	**158.3**
Substitution of Ozone Depleting Substances[a]	0.3	102.1	141.7	145.2	149.2	151.7	152.7
HCFC-22 Production	46.1	20.0	4.1	5.0	4.3	2.8	5.2
Semiconductor Manufacture	0.2	0.2	0.3	0.3	0.3	0.3	0.4
Magnesium Production and Processing	0.0	0.0	0.1	0.1	0.1	0.1	0.1
PFCs	**24.3**	**6.7**	**5.9**	**5.6**	**5.1**	**4.4**	**4.1**
Semiconductor Manufacture	2.8	3.2	2.9	3.1	3.1	3.0	3.0
Aluminum Production	21.5	3.4	3.0	2.5	2.0	1.4	1.1
Substitution of Ozone Depleting Substances	0.0	+	+	+	+	+	+
SF_6	**28.8**	**11.8**	**6.3**	**6.3**	**5.8**	**6.3**	**6.1**
Electrical Transmission and Distribution	23.1	8.3	4.4	4.6	4.1	4.4	4.3

Magnesium Production and Processing	5.2	2.7	1.3	0.9	1.0	1.1	1.1
Semiconductor Manufacture	0.5	0.7	0.7	0.7	0.7	0.9	0.7
NF_3	**+**	**0.5**	**0.5**	**0.5**	**0.6**	**0.6**	**0.6**
Semiconductor Manufacture	+	0.5	0.5	0.5	0.6	0.6	0.6
Total	**342.1**	**358.0**	**353.1**	**365.2**	**360.8**	**354.6**	**358.9**

+ Does not exceed 0.05 MMT CO_2 Eq.

[a] Small amounts of PFC emissions also result from this source.

Note: Totals may not sum due to independent rounding.

Table 4-2: Emissions from Industrial Processes and Product Use (kt)

Gas/Source	1990	2005	2013	2014	2015	2016	2017
CO_2	**208,834**	**191,654**	**173,073**	**179,191**	**172,963**	**164,524**	**166,872**
Iron and Steel Production & Metallurgical Coke Production	101,630	68,210	53,471	58,353	47,825	42,309	41,782
Iron and Steel Production	*99,126*	*66,160*	*51,641*	*56,332*	*44,981*	*40,983*	*41,204*
Metallurgical Coke Production	*2,504*	*2,050*	*1,830*	*2,020*	*2,843*	*1,327*	*578*
Cement Production	33,484	46,194	36,369	39,439	39,907	39,439	40,324
Petrochemical Production	21,222	26,810	26,395	26,496	28,062	28,110	28,225
Ammonia Production	13,047	9,196	9,480	9,377	10,634	10,838	13,216
Lime Production	11,700	14,552	14,028	14,210	13,342	12,942	13,145
Other Process Uses of Carbonates	6,297	7,644	11,524	12,954	12,182	10,969	10,139
Urea Consumption for Non-Agricultural Purposes	3,784	3,653	4,556	1,807	4,578	5,132	4,958
Carbon Dioxide Consumption	1,472	1,375	4,188	4,471	4,471	4,471	4,471
Ferroalloy Production	2,152	1,392	1,785	1,914	1,960	1,796	1,975
Soda Ash Production	1,431	1,655	1,694	1,685	1,714	1,723	1,753
Titanium Dioxide Production	1,195	1,755	1,715	1,688	1,635	1,662	1,688
Glass Production	1,535	1,928	1,317	1,336	1,299	1,249	1,315
Aluminum Production	6,831	4,142	3,255	2,833	2,767	1,334	1,205
Phosphoric Acid Production	1,529	1,342	1,149	1,038	999	998	1,023
Zinc Production	632	1,030	1,429	956	933	925	1,009
Lead Production	516	553	546	459	473	450	455
Silicon Carbide Production and Consumption	375	219	169	173	180	174	186
Magnesium Production and Processing	1	3	2	2	3	3	3
CH_4	**12**	**4**	**4**	**6**	**9**	**11**	**11**
Petrochemical Production	9	3	3	5	7	10	10
Ferroalloy Production	1	+	+	1	1	1	1
Silicon Carbide Production and Consumption	1	+	+	+	+	+	+
Iron and Steel Production & Metallurgical Coke Production	1	1	+	+	+	+	+
Iron and Steel Production	*1*	*1*	+	+	+	+	+
Metallurgical Coke Production	*0*	*0*	*0*	*0*	*0*	*0*	*0*
N_2O	**112**	**84**	**71**	**77**	**75**	**79**	**76**
Nitric Acid Production	41	38	36	37	39	34	31
Adipic Acid Production	51	24	13	18	14	23	25
N_2O from Product Uses	14	14	14	14	14	14	14
Caprolactam, Glyoxal, and Glyoxylic Acid Production	6	7	7	7	7	7	5
Semiconductor Manufacture	+	+	1	1	1	1	1
HFCs	**M**	**M**	**M**	**M**	**M**	**M**	**M**
Substitution of Ozone Depleting Substances[a]	M	M	M	M	M	M	M

HCFC-22 Production	3	1	+	+	+	+	+
Semiconductor Manufacture	M	M	M	M	M	M	M
Magnesium Production and Processing	0	0	+	+	+	+	+
PFCs	**M**	**M**	**M**	**M**	**M**	**M**	**M**
Semiconductor Manufacture	M	M	M	M	M	M	M
Aluminum Production	M	M	M	M	M	M	M
Substitution of Ozone Depleting Substances	0	+	+	+	+	+	+
SF_6	**1**	**1**	**+**	**+**	**+**	**+**	**+**
Electrical Transmission and Distribution	1	+	+	+	+	+	+
Magnesium Production and Processing	+	+	+	+	+	+	+
Semiconductor Manufacture	+	+	+	+	+	+	+
NF_3	**+**	**+**	**+**	**+**	**+**	**+**	**+**
Semiconductor Manufacture	+	+	+	+	+	+	+

+ Does not exceed 0.5 kt.
M (Mixture of gases)
[a] Small amounts of PFC emissions also result from this source.
Note: Totals may not sum due to independent rounding.

The UNFCCC incorporated the *2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines)* as the standard for Annex I countries at the Nineteenth Conference of the Parties (Warsaw, November 11-23, 2013). This chapter presents emission estimates calculated in accordance with the methodological guidance provided in these guidelines. For additional detail on IPPU sources that are not estimated in this Inventory report, please review Annex 5, Assessment of the Sources and Sinks of Greenhouse Gas Emissions Not Included. These sources are not estimated due to various national circumstances, such as that emissions from a source may not currently occur in the United States, data are not currently available for those emission sources (e.g., ceramics, non-metallurgical magnesium production, glyoxal and glyoxylic acid production, CH_4 from direct reduced iron production), emissions are included elsewhere within the Inventory report, or data suggest that emissions are not significant (e.g., various fluorinated gas emissions from the electronics industry and other produce uses). Information on planned improvements for specific IPPU source categories can be found in the Planned Improvements section of the individual source category.

In addition, as mentioned in the Energy chapter of this report (Box 3-6), fossil fuels consumed for non-energy uses for primary purposes other than combustion for energy (including lubricants, paraffin waxes, bitumen asphalt, and solvents) are reported in the Energy chapter. According to the *2006 IPCC Guidelines*, these non-energy uses of fossil fuels are to be reported under IPPU, rather than Energy; however, due to national circumstances regarding the allocation of energy statistics and carbon (C) balance data, the United States reports non-energy uses in the Energy chapter of this Inventory. Reporting these non-energy use emissions under IPPU would involve making artificial adjustments to the non-energy use C balance. These artificial adjustments would also result in the C emissions for lubricants, waxes, and asphalt and road oil being reported under IPPU, while the C storage for lubricants, waxes, and asphalt and road oil would be reported under Energy. To avoid presenting an incomplete C balance, double-counting, and adopting a less transparent approach, the entire calculation of C storage and C emissions is therefore conducted in the Non-Energy Uses of Fossil Fuels category calculation methodology and reported under the Energy sector. For more information, see the Methodology section for CO_2 from Fossil Fuel Combustion and Section 3.2, Carbon Emitted from Non-Energy Uses of Fossil Fuels.

Finally, as stated in the Energy chapter, portions of the fuel consumption data for seven fuel categories—coking coal, distillate fuel, industrial other coal, petroleum coke, natural gas, residual fuel oil, and other oil—are reallocated to the IPPU chapter, as they are consumed during non-energy related industrial process activity. Emissions from uses of fossil fuels as feedstocks or reducing agents (e.g., petrochemical production, aluminum production, titanium dioxide and zinc production) are reported in the IPPU chapter, unless otherwise noted due to specific national circumstances. More information on the methodology to adjust for these emissions within the Energy chapter is described in the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion. Additional information is listed within each IPPU emission source in which this approach applies.

QA/QC and Verification Procedures

For IPPU sources, a detailed QA/QC plan was developed and implemented for specific categories. This plan is consistent with the U.S. Inventory QA/QC plan outlined in Annex 8, but was tailored to include specific procedures recommended for these sources. The IPPU QA/QC Plan does not replace the Inventory QA/QC Plan, but rather provides more context for the IPPU sector. The IPPU QA/QC Plan provides the completed QA/QC forms for each inventory reports, as well as, for certain source categories (e.g., key categories), more detailed documentation of quality control checks and recalculations due to methodological changes.

Two types of checks were performed using this plan: (1) general (Tier 1) procedures consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines* that focus on annual procedures and checks to be used when gathering, maintaining, handling, documenting, checking, and archiving the data, supporting documents, and files; and (2) source category specific (Tier 2) procedures that focus on checks and comparisons of the emission factors, activity data, and methodologies used for estimating emissions from the relevant industrial process and product use sources. Examples of these procedures include: checks to ensure that activity data and emission estimates are consistent with historical trends to identify significant changes; that, where possible, consistent and reputable data sources are used and specified across sources; that interpolation or extrapolation techniques are consistent across sources; and that common datasets, units, and conversion factors are used where applicable. The IPPU QA/QC plan also checked for transcription errors in data inputs required for emission calculations, including activity data and emission factors; and confirmed that estimates were calculated and reported for all applicable and able portions of the source categories for all years.

General, or Tier 1, QA/QC procedures and calculation-related QC (category-specific, Tier 2) have been performed for all IPPU sources. Consistent with the *2006 IPCC Guidelines*, additional category-specific QC procedures were performed for more significant emission categories (such as the comparison of reported consumption with modeled consumption using EPA's Greenhouse Gas Reporting Program (GHGRP) data within Substitution of Ozone Depleting Substances) or sources where significant methodological and data updates have taken place. The QA/QC implementation did not reveal any significant inaccuracies, and all errors identified were documented and corrected. Application of these procedures, specifically category-specific QC procedures and updates/improvements as a result of QA processes (expert, public, and UNFCCC technical expert reviews), are described further within respective source categories, in the Recalculations and Planned Improvement sections.

For most IPPU categories, activity data are obtained via aggregation of facility-level data from EPA's GHGRP, national commodity surveys conducted by U.S. Geologic Survey National Minerals Information Center, U.S. Department of Energy (DOE), U.S. Census Bureau, industry associations such as Air-Conditioning, Heating, and Refrigeration Institute (AHRI), American Chemistry Council (ACC), and American Iron and Steel Institute (AISI) (specified within each source category). The emission factors used include those derived from the EPA's GHGRP and application of IPCC default factors. Descriptions of uncertainties and assumptions for activity data and emission factors are included within the uncertainty discussion sections for each IPPU source category.

The uncertainty analysis performed to quantify uncertainties associated with the 2017 emission estimates from IPPU continues a multi-year process for developing credible quantitative uncertainty estimates for these source categories using the IPCC Tier 2 approach. As the process continues, the type and the characteristics of the actual probability density functions underlying the input variables are identified and better characterized (resulting in development of more reliable inputs for the model, including accurate characterization of correlation between variables), based primarily on expert judgment. Accordingly, the quantitative uncertainty estimates reported in this section should be considered illustrative and as iterations of ongoing efforts to produce accurate uncertainty estimates. The correlation among data used for estimating emissions for different sources can influence the uncertainty analysis of each individual source. While the uncertainty analysis recognizes very significant connections among sources, a more comprehensive approach that accounts for all linkages will be identified as the uncertainty analysis moves forward.

Box 4-1: Methodological Approach for Estimating and Reporting U.S. Emissions and Removals

In following the United Nations Framework Convention on Climate Change (UNFCCC) requirement under Article 4.1 to develop and submit national greenhouse gas emission inventories, the emissions and removals presented in this report and this chapter, are organized by source and sink categories and calculated using internationally-accepted methods provided by the Intergovernmental Panel on Climate Change (IPCC) in the *2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines)*. Additionally, the calculated emissions and removals in a given year for the United States are presented in a common manner in line with the UNFCCC reporting guidelines for the reporting of inventories under this international agreement. The use of consistent methods to calculate emissions and removals by all nations providing their inventories to the UNFCCC ensures that these reports are comparable. The presentation of emissions and removals provided in this Inventory do not preclude alternative examinations, but rather, this Inventory presents emissions and removals in a common format consistent with how countries are to report Inventories under the UNFCCC. The report itself, and this chapter, follows this standardized format, and provides an explanation of the application of methods used to calculate emissions and removals.

Box 4-2: Industrial Processes Data from EPA's Greenhouse Gas Reporting Program

On October 30, 2009, the U.S. EPA published a rule requiring annual reporting of greenhouse gas data from large greenhouse gas emission sources in the United States. Implementation of the rule, codified at 40 CFR Part 98, is referred to as EPA's GHGRP. The rule applies to direct greenhouse gas emitters, fossil fuel suppliers, industrial gas suppliers, and facilities that inject CO_2 underground for sequestration or other reasons and requires reporting by sources or suppliers in 41 industrial categories ("Subparts"). Annual reporting is at the facility level, except for certain suppliers of fossil fuels and industrial greenhouse gases. In general, the threshold for reporting is 25,000 metric tons or more of CO_2 Eq. per year, but reporting is required for all facilities in some industries. Calendar year 2010 was the first year for which data were reported for facilities subject to 40 CFR Part 98, though some source categories first reported data for calendar year 2011.

EPA's GHGRP dataset and the data presented in this Inventory are complementary. The GHGRP dataset continues to be an important resource for the Inventory, providing not only annual emissions information, but also other annual information such as activity data and emission factors that can improve and refine national emission estimates and trends over time. GHGRP data also allow EPA to disaggregate national inventory estimates in new ways that can highlight differences across regions and sub-categories of emissions, along with enhancing application of QA/QC procedures and assessment of uncertainties. EPA uses annual GHGRP data in a number of categories to improve the national estimates presented in this Inventory consistent with IPCC guidelines. While many methodologies used in EPA's GHGRP are consistent with IPCC, it should be noted that the definitions for source categories in EPA's GHGRP may differ from those used in this Inventory in meeting the UNFCCC reporting guidelines (IPCC 2011). In line with the UNFCCC reporting guidelines, the Inventory is a comprehensive accounting of all emissions from source categories identified in the *2006 IPCC Guidelines*. Further information on the reporting categorizations in EPA's GHGRP and specific data caveats associated with monitoring methods in EPA's GHGRP has been provided on the GHGRP website.[3]

For certain source categories in this Inventory (e.g., nitric acid production, lime production, cement production, petrochemical production, carbon dioxide consumption, ammonia production, and urea consumption for non-agricultural purposes), EPA has integrated data values that have been calculated by aggregating GHGRP data that are considered confidential business information (CBI) at the facility level. EPA, with industry engagement, has put forth criteria to confirm that a given data aggregation shields underlying CBI from public disclosure. EPA is only publishing data values that meet these aggregation criteria.[4] Specific uses of aggregated facility-level data are described in the respective methodological sections. For other source categories in this chapter, as indicated in the respective planned improvements sections, EPA is continuing to analyze how facility-level GHGRP data may be used to improve the national estimates presented in this Inventory, giving particular consideration to ensuring time-series consistency and completeness.

As stated previously in the Introduction chapter, this year EPA has integrated GHGRP information for various Industrial Processes and Product Use categories and also identified places where EPA plans to integrate additional

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[3] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[4] U.S. EPA Greenhouse Gas Reporting Program. Developments on Publication of Aggregated Greenhouse Gas Data, November 25, 2014. See <http://www.epa.gov/ghgreporting/confidential-business-information-ghg-reporting>.

GHGRP data in additional categories[5] (see those categories' Planned Improvements sections for details). EPA has paid particular attention to ensuring time-series consistency for major recalculations that have occurred from the incorporation of GHGRP data into these categories, consistent with *2006 IPCC Guidelines* and *IPCC Technical Bulletin on Use of Facility-Specific Data in National GHG Inventories.*[6]

EPA verifies annual facility-level reports through a multi-step process to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent.[7] All reports submitted to EPA are evaluated by electronic validation and verification checks. If potential errors are identified, EPA will notify the reporter, who can resolve the issue either by providing an acceptable response describing why the flagged issue is not an error or by correcting the flagged issue and resubmitting their annual greenhouse gas report. Additional QA/QC and verification procedures occur for each GHGRP subpart, but due to concerns regarding CBI data, specific subpart QA/QC and verification procedures are not available to include in this Inventory report. The GHGRP dataset is a particularly important annual resource and will continue to be important for improving emissions estimates from IPPU in future Inventory reports. Additionally, EPA's GHGRP has and will continue to enhance QA/QC procedures and assessment of uncertainties within the IPPU categories (see those categories for specific QA/QC details regarding the use of GHGRP data).

4.1 Cement Production (CRF Source Category 2A1)

Cement production is an energy- and raw material-intensive process that results in the generation of carbon dioxide (CO_2) from both the energy consumed in making the clinker precursor to cement and the chemical process itself to make the clinker. Emissions from fuels consumed for energy purposes during the production of cement are accounted for in the Energy chapter.

During the clinker production process, the key reaction is where calcium carbonate ($CaCO_3$), in the form of limestone or similar rocks, is heated in a cement kiln at a temperature range of about 700 to 1,000 degrees Celsius (1,300 to 1,800 degrees Fahrenheit) to form lime (i.e., calcium oxide or CaO) and CO_2 in a process known as calcination or calcining. The quantity of CO_2 emitted during clinker production is directly proportional to the lime content of the clinker. During calcination, each mole of $CaCO_3$ heated in the clinker kiln forms one mole of CaO and one mole of CO_2. The CO_2 is vented to the atmosphere as part of the kiln lime exhaust:

$$CaCO_3 + heat \rightarrow CaO + CO_2$$

Next, over a temperature range of 1000 to 1450 degrees Celsius, the CaO combines with alumina, iron oxide and silica that are also present in the clinker raw material mix to form hydraulically reactive compounds within white-hot semifused (sintered) nodules of clinker. Because one of these "sintering" reactions is highly exothermic, very little extra heat energy is required, and these sintering reactions have few process emissions of CO_2. The clinker is then rapidly cooled to maintain quality, then very finely ground with a small amount of gypsum and potentially other materials (e.g., ground granulated blast furnace slag, etc.), and used to make Portland and similar cements.[8]

Carbon dioxide emitted from the chemical process of cement production is the second largest source of industrial CO_2 emissions in the United States. Cement is produced in 34 states and Puerto Rico. Texas, California, Missouri, Florida, and Pennsylvania were the leading cement-producing states in 2017 and accounted for almost 50 percent of

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[5] Ammonia Production, Glass Production, Lead Production, and Other Fluorinated Gas Production.

[6] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[7] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf>.

[8] Approximately three percent of total clinker production is used to produce masonry cement, which is produced using plasticizers (e.g., ground limestone, lime, etc.) and Portland cement (USGS 2011). Carbon dioxide emissions that result from the production of lime used to create masonry cement are included in the Lime Manufacture source category.

total U.S. production (USGS 2018a). Based on both GHGRP data (EPA 2018) and USGS reported data, clinker production in 2017 increased approximately 2 percent from 2016 levels as cement sales increased modestly (less than 2 percent) in 2017, with imports stagnant in 2017 (USGS 2018a). In 2017, U.S. clinker production totaled 77,500 kilotons (EPA 2018). The resulting CO_2 emissions were estimated to be 40.3 MMT CO_2 Eq. (40,324 kt) (see Table 4-3).

Table 4-3: CO_2 Emissions from Cement Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	33.5	33,484
2005	46.2	46,194
2013	36.4	36,369
2014	39.4	39,439
2015	39.9	39,907
2016	39.4	39,439
2017	40.3	40,324

Greenhouse gas emissions from cement production increased every year from 1991 through 2006 (with the exception of a slight decrease in 1997) but decreased in the following years until 2009. Since 1990, emissions have increased by 20 percent. Emissions from cement production were at their lowest levels in 2009 (2009 emissions are approximately 28 percent lower than 2008 emissions and 12 percent lower than 1990), due to the economic recession and associated decrease in demand for construction materials. Since 2010, emissions have increased by roughly 28 percent due to increasing consumption. In 2017, emissions from cement production increased by 2 percent from 2016 levels. Cement continues to be a critical component of the construction industry; therefore, the availability of public and private construction funding, as well as overall economic conditions, have considerable impact on the level of cement production.

Methodology

Carbon dioxide emissions were estimated using the Tier 2 methodology from the *2006 IPCC Guidelines*. The Tier 2 methodology was used because detailed and complete data (including weights and composition) for carbonate(s) consumed in clinker production are not available, and thus a rigorous Tier 3 approach is impractical. Tier 2 specifies the use of aggregated plant or national clinker production data and an emission factor, which is the product of the average lime fraction for clinker of 65 percent and a constant reflecting the mass of CO_2 released per unit of lime. The U.S. Geological Survey (USGS) mineral commodity expert for cement has confirmed that this is a reasonable assumption for the United States (Van Oss 2013a). This calculation yields an emission factor of 0.510 tons of CO_2 per ton of clinker produced, which was determined as follows:

$$EF_{clinker} = 0.650 \text{ CaO} \times [(44.01 \text{ g/mole } CO_2) \div (56.08 \text{ g/mole CaO})] = 0.510 \text{ tons } CO_2/\text{ton clinker}$$

During clinker production, some of the raw materials, partially reacted raw materials and clinker enters the kiln line's exhaust system as non-calcinated, partially calcinated, or fully calcinated cement kiln dust (CKD). To the degree that the CKD contains carbonate raw materials which are then calcined, there are associated CO_2 emissions. At some plants, essentially all CKD is directly returned to the kiln, becoming part of the raw material feed, or is likewise returned to the kiln after first being removed from the exhaust. In either case, the returned CKD becomes a raw material, thus forming clinker, and the associated CO_2 emissions are a component of those calculated for the clinker overall. At some plants, however, the CKD cannot be returned to the kiln because it is chemically unsuitable as a raw material, or chemical issues limit the amount of CKD that can be so reused. Any clinker that cannot be returned to the kiln is either used for other (non-clinker) purposes or is landfilled. The CO_2 emissions attributable to the non-returned calcinated portion of the CKD are not accounted for by the clinker emission factor. The IPCC recommends that these additional CKD CO_2 emissions should be estimated as two percent of the CO_2 emissions calculated from clinker production (when data on CKD generation are not available). Total cement production emissions were calculated by adding the emissions from clinker production to the emissions assigned to CKD (IPCC 2006).

Furthermore, small amounts of impurities (i.e., not calcium carbonate) may exist in the raw limestone used to produce clinker. The proportion of these impurities is generally minimal, although a small amount (1 to 2 percent) of magnesium oxide (MgO) may be desirable as a flux. Per the IPCC Tier 2 methodology, a correction for MgO is not used, since the amount of MgO from carbonate is likely very small and the assumption of a 100 percent carbonate source of CaO already yields an overestimation of emissions (IPCC 2006).

The 1990 through 2012 activity data for clinker production (see Table 4-4) were obtained from USGS (Van Oss 2013a, Van Oss 2013b). Clinker production data for 2013 were also obtained from USGS (USGS 2014). The data were compiled by USGS (to the nearest ton) through questionnaires sent to domestic clinker and cement manufacturing plants, including the facilities in Puerto Rico. During the 1990 through 2015 Inventory report cycle, EPA began incorporating clinker production data from EPA's GHGRP to estimate emissions in these respective years. Clinker production values in the current Inventory report utilize GHGRP data for the years 2014 through 2017 (EPA 2018). Details on how this change compares to USGS reported data can be found in the section on Uncertainty and Time-Series Consistency.

Table 4-4: Clinker Production (kt)

Year	Clinker
1990	64,355
2005	88,783
2013	69,900
2014	75,800
2015	76,700
2016	75,800
2017	77,500

Notes: Clinker production from 1990 through 2017 includes Puerto Rico.

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates are primarily due to uncertainties in the lime content of clinker and in the percentage of CKD recycled inside the cement kiln. Uncertainty is also associated with the assumption that all calcium-containing raw materials are $CaCO_3$, when a small percentage likely consists of other carbonate and non-carbonate raw materials. The lime content of clinker varies from 60 to 67 percent; 65 percent is used as a representative value (Van Oss 2013a). The amount of CO_2 from CKD loss can range from 1.5 to 8 percent depending upon plant specifications. Additionally, some amount of CO_2 is reabsorbed when the cement is used for construction. As cement reacts with water, alkaline substances such as calcium hydroxide are formed. During this curing process, these compounds may react with CO_2 in the atmosphere to create calcium carbonate. This reaction only occurs in roughly the outer 0.2 inches of the total thickness. Because the amount of CO_2 reabsorbed is thought to be minimal, it was not estimated. However, see Planned Improvements described below to reassess this assumption by conducting a review to identify recent studies that may provide information or data on reabsorption rates of cement products.

Total U.S. clinker production is assumed to have low uncertainty. USGS takes a number of manual steps to review clinker production reported through their voluntary surveys. EPA also continues to review reported clinker production data required by GHGRP Subpart H facilities for current and future Inventory reports. EPA verifies annual facility-level reports through a multi-step process (e.g., combination of electronic checks and manual reviews by staff) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent.

Based on the results of the verification process, the EPA follows up with facilities to resolve mistakes that may have occurred.[9] Facilities are also required to monitor and maintain records of monthly clinker production.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-5. Based on the uncertainties associated with total U.S. clinker production, the CO_2 emission factor for clinker production, and the emission factor for additional CO_2 emissions from CKD, 2017 CO_2 emissions from cement production were estimated to be between 38.0 and 42.7 MMT CO_2 Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 6 percent below and 6 percent above the emission estimate of 40.3 MMT CO_2 Eq.

Table 4-5: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Cement Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate (MMT CO_2 Eq.)		Uncertainty Range Relative to Emission Estimate (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Cement Production	CO_2	40.3	38.0	42.7	-6%	+6%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure time-series consistency from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above. More information on the consistency in clinker production data and emissions across the time series with the use of GHGRP clinker data for 2014 through 2017 can be found in the Uncertainty and Time-Series Consistency section.

QA/QC and Verification

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see QA/QC and Verification Procedures section in the introduction of the IPPU chapter. EPA relied upon the latest guidance from the IPCC on the use of facility-level data in national inventories and applied a category-specific QC process to compare activity data from EPA's GHGRP with existing data from USGS surveys. This was to ensure time-series consistency of the emission estimates presented in the Inventory. For the year 2014, USGS and GHGRP clinker production data showed a difference of approximately 2 percent, while in 2015, 2016 and in 2017 that difference decreased to less than 1 percent between the two sets of activity data. This difference resulted in an increase of emissions compared to USGS data (USGS 2017, 2018a, 2018b) by less than 0.1 MMT CO_2 Eq. in 2015, 2016, and in 2017. The information collected by the USGS National Minerals Information center surveys continue to be an important data source. See the Uncertainty and Time- Series Consistency section for information on how GHGRP data are verified.

Planned Improvements

In response to prior comments from the Portland Cement Association (PCA) and UNFCCC expert technical reviews, EPA is continuing to evaluate and analyze data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Cement Production source category. EPA held a technical meeting with PCA in August 2016 to review Inventory methods and available data from the GHGRP data set. Most cement production facilities reporting under EPA's GHGRP use Continuous Emission Monitoring Systems (CEMS) to monitor and report CO_2 emissions, thus reporting combined process and combustion emissions from kilns. In implementing further improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon, in addition to category-specific QC methods

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[9] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf>.

recommended by the *2006 IPCC Guidelines*.[10] EPA's long-term improvement plan includes continued assessment of the feasibility of using additional GHGRP information beyond aggregation of reported facility-level clinker data, in particular disaggregating the combined process and combustion emissions reported using CEMS, to separately present national process and combustion emissions streams consistent with IPCC and UNFCCC guidelines. This long-term planned analysis is still in development and has not been updated for this current Inventory.

During the Public Review comment period of the current (i.e., 1990 through 2017) Inventory report, EPA received comments on the characterization of cement and clinker production processes described in this chapter. As a result, EPA updated the description of these processes and comparison of available datasets (QA/QC and Verification section).

Finally, in response to feedback from PCA during the Public Review comment period of a previous Inventory in 2017, EPA plans to work with PCA to discuss additional long-term improvements to review methods and data used to estimate CO_2 emissions from cement production to account for both organic material and magnesium carbonate in the raw material, and to discuss the carbonation that occurs later in the cement product lifecycle. Priority will be to identify data and studies on the average MgO content of clinker produced in the United States, the average carbon content for organic materials in kiln feed in the United States, and CO_2 reabsorption rates via carbonation for various cement products. This information is not reported by facilities subject to report to GHGRP.

4.2 Lime Production (CRF Source Category 2A2)

Lime is an important manufactured product with many industrial, chemical, and environmental applications. Lime production involves three main processes: stone preparation, calcination, and hydration. Carbon dioxide (CO_2) is generated during the calcination stage, when limestone—mostly calcium carbonate ($CaCO_3$)—is roasted at high temperatures in a kiln to produce calcium oxide (CaO) and CO_2. The CO_2 is given off as a gas and is normally emitted to the atmosphere.

$$CaCO_3 \rightarrow CaO + CO_2$$

Some of the CO_2 generated during the production process, however, is recovered at some facilities for use in sugar refining and precipitated calcium carbonate (PCC) production.[11] Emissions from fuels consumed for energy purposes during the production of lime are accounted for in the Energy chapter.

For U.S. operations, the term "lime" actually refers to a variety of chemical compounds. These include CaO, or high-calcium quicklime; calcium hydroxide ($Ca(OH)_2$), or hydrated lime; dolomitic quicklime ([CaO•MgO]); and dolomitic hydrate ([$Ca(OH)_2$•MgO] or [$Ca(OH)_2$•$Mg(OH)_2$]).

The current lime market is approximately distributed across five end-use categories as follows: metallurgical uses, 37 percent; environmental uses, 31 percent; chemical and industrial uses, 22 percent; construction uses, 9 percent; and refractory dolomite, 1 percent (USGS 2018b). The major uses are in steel making, flue gas desulfurization systems at coal-fired electric power plants, construction, and water treatment, as well as uses in mining, pulp and paper and precipitated calcium carbonate manufacturing. Lime is also used as a CO_2 scrubber, and there has been experimentation on the use of lime to capture CO_2 from electric power plants.

Lime production in the United States—including Puerto Rico—was reported to be 18,000 kilotons in 2017 (USGS 2018a). At year-end 2017, there were 74 operating primary lime plants in the United States, including Puerto Rico.[12]

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[10] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[11] PCC is obtained from the reaction of CO_2 with calcium hydroxide. It is used as a filler and/or coating in the paper, food, and plastic industries.

[12] In 2017, 75 operating primary lime facilities in the United States reported to the EPA Greenhouse Gas Reporting Program.

Principal lime producing states in descending order of production are Missouri, Alabama, Ohio, Texas, and Kentucky (USGS 2018a).

U.S. lime production resulted in estimated net CO_2 emissions of 13.1 MMT CO_2 Eq. (13,145 kt) (see Table 4-6 and Table 4-7). The trends in CO_2 emissions from lime production are directly proportional to trends in production, which are described below.

Table 4-6: CO_2 Emissions from Lime Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	11.7	11,700
2005	14.6	14,552
2013	14.0	14,028
2014	14.2	14,210
2015	13.3	13,342
2016	12.9	12,942
2017	13.1	13,145

Table 4-7: Potential, Recovered, and Net CO_2 Emissions from Lime Production (kt)

Year	Potential	Recovered[a]	Net Emissions
1990	11,959	259	11,700
2005	15,074	522	14,552
2013	14,495	467	14,028
2014	14,715	505	14,210
2015	13,764	422	13,342
2016	13,312	370	12,942
2017	13,546	401	13,145

[a] For sugar refining and PCC production.

Note: Totals may not sum due to independent rounding.

In 2017, lime production increased compared to 2016 levels (increase of about 2 percent) at 18,000 kilotons, owing primarily to an increase in hydrated lime output (USGS 2018a; USGS 2017).

Methodology

To calculate emissions, the amounts of high-calcium and dolomitic lime produced were multiplied by their respective emission factors using the Tier 2 approach from the *2006 IPCC Guidelines*. The emission factor is the product of the stoichiometric ratio between CO_2 and CaO, and the average CaO and MgO content for lime. The CaO and MgO content for lime is assumed to be 95 percent for both high-calcium and dolomitic lime (IPCC 2006). The emission factors were calculated as follows:

For high-calcium lime:

$$[(44.01\ \text{g/mole}\ CO_2) \div (56.08\ \text{g/mole CaO})] \times (0.9500\ \text{CaO/lime}) = 0.7455\ \text{g}\ CO_2\text{/g lime}$$

For dolomitic lime:

$$[(88.02\ \text{g/mole}\ CO_2) \div (96.39\ \text{g/mole CaO})] \times (0.9500\ \text{CaO/lime}) = 0.8675\ \text{g}\ CO_2\text{/g lime}$$

Production was adjusted to remove the mass of chemically combined water found in hydrated lime, determined according to the molecular weight ratios of H_2O to ($Ca(OH)_2$ and [$Ca(OH)_2 \bullet Mg(OH)_2$]) (IPCC 2006). These factors set the chemically combined water content to 24.3 percent for high-calcium hydrated lime, and 27.2 percent for dolomitic hydrated lime.

The *2006 IPCC Guidelines* (Tier 2 method) also recommends accounting for emissions from lime kiln dust (LKD) through application of a correction factor. LKD is a byproduct of the lime manufacturing process typically not recycled back to kilns. LKD is a very fine-grained material and is especially useful for applications requiring very small particle size. Most common LKD applications include soil reclamation and agriculture. Currently, data on annual LKD production is not readily available to develop a country-specific correction factor. Lime emission estimates were multiplied by a factor of 1.02 to account for emissions from LKD (IPCC 2006). See the Planned Improvements section associated with efforts to improve uncertainty analysis and emission estimates associated with LKD.

Lime emission estimates were further adjusted to account for the amount of CO_2 captured for use in on-site processes. All the domestic lime facilities are required to report these data to EPA under its GHGRP. The total national-level annual amount of CO_2 captured for on-site process use was obtained from EPA's GHGRP (EPA 2018) based on reported facility level data for years 2010 through 2017. The amount of CO_2 captured/recovered for on-site process use is deducted from the total potential emissions (i.e., from lime production and LKD). The net lime emissions are presented in Table 4-6 and Table 4-7. GHGRP data on CO_2 removals (i.e., CO_2 captured/recovered) was available only for 2010 through 2017. Since GHGRP data are not available for 1990 through 2009, IPCC "splicing" techniques were used as per the *2006 IPCC Guidelines* on time-series consistency (IPCC 2006, Volume 1, Chapter 5).

Lime production data (by type, high-calcium- and dolomitic-quicklime, high-calcium- and dolomitic-hydrated, and dead-burned dolomite) for 1990 through 2017 (see Table 4-8) were obtained from the U.S. Geological Survey (USGS) (USGS 2017 and 2018a) annual reports and are compiled by USGS to the nearest ton. The high-calcium quicklime and dolomitic quicklime values were estimated using the ratio of the 2015 quicklime values to the 2017 total values. The 2015 values for high-calcium hydrated, dolomitic hydrated, and dead-burned dolomite were used since there is less fluctuation in their production from year to year. Natural hydraulic lime, which is produced from CaO and hydraulic calcium silicates, is not manufactured in the United States (USGS 2018b). Total lime production was adjusted to account for the water content of hydrated lime by converting hydrate to oxide equivalent based on recommendations from the IPCC, and is presented in Table 4-9 (IPCC 2006). The CaO and CaO•MgO contents of lime were obtained from the IPCC (IPCC 2006). Since data for the individual lime types (high calcium and dolomitic) were not provided prior to 1997, total lime production for 1990 through 1996 was calculated according to the three-year distribution from 1997 to 1999.

Table 4-8: High-Calcium- and Dolomitic-Quicklime, High-Calcium- and Dolomitic-Hydrated, and Dead-Burned-Dolomite Lime Production (kt)

Year	High-Calcium Quicklime	Dolomitic Quicklime	High-Calcium Hydrated	Dolomitic Hydrated	Dead-Burned Dolomite
1990	11,166	2,234	1,781	319	342
2005	14,100	2,990	2,220	474	200
2013	13,800	2,850	2,050	260	200
2014	14,100	2,740	2,190	279	200
2015	13,100	2,550	2,150	279	200
2016	12,615	2,456	2,150	279	200
2017	12,866	2,505	2,150	279	200

Table 4-9: Adjusted Lime Production (kt)

Year	High-Calcium	Dolomitic
1990	12,466	2,800
2005	15,721	3,522

2013	15,297	3,252
2014	15,699	3,135
2015	14,670	2,945
2016	14,185	2,851
2017	14,436	2,900

Note: Minus water content of hydrated lime.

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates can be attributed to slight differences in the chemical composition of lime products and CO_2 recovery rates for on-site process use over the time series. Although the methodology accounts for various formulations of lime, it does not account for the trace impurities found in lime, such as iron oxide, alumina, and silica. Due to differences in the limestone used as a raw material, a rigid specification of lime material is impossible. As a result, few plants produce lime with exactly the same properties.

In addition, a portion of the CO_2 emitted during lime production will actually be reabsorbed when the lime is consumed, especially at captive lime production facilities. As noted above, lime has many different chemical, industrial, environmental, and construction applications. In many processes, CO_2 reacts with the lime to create calcium carbonate (e.g., water softening). Carbon dioxide reabsorption rates vary, however, depending on the application. For example, 100 percent of the lime used to produce precipitated calcium carbonate reacts with CO_2; whereas most of the lime used in steel making reacts with impurities such as silica, sulfur, and aluminum compounds. Quantifying the amount of CO_2 that is reabsorbed would require a detailed accounting of lime use in the United States and additional information about the associated processes where both the lime and byproduct CO_2 are "reused" are required to quantify the amount of CO_2 that is reabsorbed. Research conducted thus far has not yielded the necessary information to quantify CO_2 reabsorption rates.[13] However, some additional information on the amount of CO_2 consumed on site at lime facilities has been obtained from EPA's GHGRP.

In some cases, lime is generated from calcium carbonate byproducts at pulp mills and water treatment plants.[14] The lime generated by these processes is included in the USGS data for commercial lime consumption. In the pulping industry, mostly using the Kraft (sulfate) pulping process, lime is consumed in order to causticize a process liquor (green liquor) composed of sodium carbonate and sodium sulfide. The green liquor results from the dilution of the smelt created by combustion of the black liquor where biogenic carbon (C) is present from the wood. Kraft mills recover the calcium carbonate "mud" after the causticizing operation and calcine it back into lime—thereby generating CO_2—for reuse in the pulping process. Although this re-generation of lime could be considered a lime manufacturing process, the CO_2 emitted during this process is mostly biogenic in origin, and therefore is not included in the industrial processes totals (Miner and Upton 2002). In accordance with IPCC methodological guidelines, any such emissions are calculated by accounting for net C fluxes from changes in biogenic C reservoirs in wooded or crop lands (see the Land Use, Land-Use Change, and Forestry chapter).

In the case of water treatment plants, lime is used in the softening process. Some large water treatment plants may recover their waste calcium carbonate and calcine it into quicklime for reuse in the softening process. Further research is necessary to determine the degree to which lime recycling is practiced by water treatment plants in the United States.

Another uncertainty is the assumption that calcination emissions for LKD are around 2 percent. The National Lime Association (NLA) has commented that the estimates of emissions from LKD in the United States could be closer to 6 percent. They also note that additional emissions (approximately 2 percent) may also be generated through

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[13] Representatives of the National Lime Association estimate that CO_2 reabsorption that occurs from the use of lime may offset as much as a quarter of the CO_2 emissions from calcination (Males 2003).

[14] Some carbide producers may also regenerate lime from their calcium hydroxide byproducts, which does not result in emissions of CO_2. In making calcium carbide, quicklime is mixed with coke and heated in electric furnaces. The regeneration of lime in this process is done using a waste calcium hydroxide (hydrated lime) [$CaC_2 + 2H_2O \rightarrow C_2H_2 + Ca(OH)_2$], not calcium carbonate [$CaCO_3$]. Thus, the calcium hydroxide is heated in the kiln to simply expel the water [$Ca(OH)_2 + \text{heat} \rightarrow CaO + H_2O$] and no CO_2 is released.

production of other byproducts/wastes (off-spec lime that is not recycled, scrubber sludge) at lime plants (Seeger 2013). Publicly available data on LKD generation rates, total quantities not used in cement production, and types of other byproducts/wastes produced at lime facilities are limited. EPA initiated a dialogue with NLA to discuss data needs to generate a country-specific LKD factor and is reviewing the information provided by NLA. NLA compiled and shared historical emissions information and quantities for some waste products reported by member facilities associated with generation of total calcined byproducts and LKD, as well as methodology and calculation worksheets that member facilities complete when reporting. There is uncertainty regarding the availability of data across the time series needed to generate a representative country-specific LKD factor. Uncertainty of the activity data is also a function of the reliability and completeness of voluntarily reported plant-level production data. Further research and data is needed to improve understanding of additional calcination emissions to consider revising the current assumptions that are based on IPCC guidelines. More information can be found in the Planned Improvements section below.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-10. Lime CO_2 emissions for 2017 were estimated to be between 12.9 and 13.4 MMT CO_2 Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 2 percent below and 2 percent above the emission estimate of 13.1 MMT CO_2 Eq.

Table 4-10: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Lime Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Lime Production	CO_2	13.1	12.9	13.4	-2%	+2%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Future improvements involve finishing a review of data to improve current assumptions associated with emissions from production of LKD and other byproducts/wastes as discussed in the Uncertainty and Time-Series Consistency section per comments from the NLA provided during the Public Review comment period of the previous (1990 to 2015) Inventory. In response to comments, EPA met with NLA on April 7, 2015 to outline specific information required to apply IPCC methods to develop a country-specific correction factor to more accurately estimate emissions from production of LKD. In response to this technical meeting, in January and February 2016, NLA compiled and shared historical emissions information reported by member facilities on an annual basis under voluntary reporting initiatives over 2002 through 2011 associated with generation of total calcined byproducts and LKD (LKD reporting only differentiated starting in 2010). This emissions information was reported on a voluntary basis consistent with NLA's facility-level reporting protocol also recently provided. EPA needs additional time to review the information provided by NLA and plans to work with them to address needs for EPA's analysis, as there is limited information across the time series. Due to limited resources and need for additional QA of information, this planned improvement is still in process and has not been incorporated into this current Inventory report. This is a long-term improvement. As an interim step, EPA has updated the qualitative description of uncertainty to reflect the information provided by NLA.

In addition, EPA plans to review GHGRP emissions and activity data reported to EPA under Subpart S, and in particular, aggregated activity data on lime production by type. Particular attention will be made to also ensuring time-series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and

UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[15]

4.3 Glass Production (CRF Source Category 2A3)

Glass production is an energy and raw-material intensive process that results in the generation of carbon dioxide (CO_2) from both the energy consumed in making glass and the glass process itself. Emissions from fuels consumed for energy purposes during the production of glass are accounted for in the Energy sector.

Glass production employs a variety of raw materials in a glass-batch. These include formers, fluxes, stabilizers, and sometimes colorants. The major raw materials (i.e., fluxes and stabilizers) which emit process-related CO_2 emissions during the glass melting process are limestone, dolomite, and soda ash. The main former in all types of glass is silica (SiO_2). Other major formers in glass include feldspar and boric acid (i.e., borax). Fluxes are added to lower the temperature at which the batch melts. Most commonly used flux materials are soda ash (sodium carbonate, Na_2CO_3) and potash (potassium carbonate, K_2O). Stabilizers are used to make glass more chemically stable and to keep the finished glass from dissolving and/or falling apart. Commonly used stabilizing agents in glass production are limestone ($CaCO_3$), dolomite ($CaCO_3MgCO_3$), alumina (Al_2O_3), magnesia (MgO), barium carbonate ($BaCO_3$), strontium carbonate ($SrCO_3$), lithium carbonate (Li_2CO_3), and zirconia (ZrO_2) (OIT 2002). Glass makers also use a certain amount of recycled scrap glass (cullet), which comes from in-house return of glassware broken in the process or other glass spillage or retention such as recycling or cullet broker services.

The raw materials (primarily limestone, dolomite and soda ash) release CO_2 emissions in a complex high-temperature chemical reaction during the glass melting process. This process is not directly comparable to the calcination process used in lime manufacturing, cement manufacturing, and process uses of carbonates (i.e., limestone/dolomite use), but has the same net effect in terms of CO_2 emissions (IPCC 2006). The U.S. glass industry can be divided into four main categories: containers, flat (window) glass, fiber glass, and specialty glass. The majority of commercial glass produced is container and flat glass (EPA 2009). The United States is one of the major global exporters of glass. Domestically, demand comes mainly from the construction, auto, bottling, and container industries. There are over 1,500 companies that manufacture glass in the United States, with the largest being Corning, Guardian Industries, Owens-Illinois, and PPG Industries.[16]

In 2017, 763 kilotons of limestone and 2,360 kilotons of soda ash were consumed for glass production (USGS 2016a; USGS 2017). Dolomite consumption data for glass manufacturing was reported to be zero for 2017. Use of limestone and soda ash in glass production resulted in aggregate CO_2 emissions of 1.3 MMT CO_2 Eq. (1,315 kt) (see Table 4-11). Overall, emissions have decreased 14 percent from 1990 through 2017.

Emissions in 2017 increased approximately 5 percent from 2016 levels while, in general, emissions from glass production have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. Specifically, the extended downturn in residential and commercial construction and automotive industries between 2008 and 2010 resulted in reduced consumption of glass products, causing a drop in global demand for limestone/dolomite and soda ash, and a corresponding decrease in emissions. Furthermore, the glass container sector is one of the leading soda ash consuming sectors in the United States. Some commercial food and beverage package manufacturers are shifting

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[15] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[16] Excerpt from Glass & Glass Product Manufacturing Industry Profile, First Research. Available online at: <http://www.firstresearch.com/Industry-Research/Glass-and-Glass-Product-Manufacturing.html>.

from glass containers towards lighter and more cost-effective polyethylene terephthalate (PET) based containers, putting downward pressure on domestic consumption of soda ash (USGS 1995 through 2015b).

Table 4-11: CO_2 Emissions from Glass Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.5	1,535
2005	1.9	1,928
2013	1.3	1,317
2014	1.3	1,336
2015	1.3	1,299
2016	1.3	1,249
2017	1.3	1,315

Note: Totals may not sum due to independent rounding

Methodology

Carbon dioxide emissions were calculated based on the *2006 IPCC Guidelines* Tier 3 method by multiplying the quantity of input carbonates (limestone, dolomite, and soda ash) by the carbonate-based emission factor (in metric tons CO_2/metric ton carbonate): limestone, 0.43971; dolomite, 0.47732; and soda ash, 0.41492.

Consumption data for 1990 through 2017 of limestone, dolomite, and soda ash used for glass manufacturing were obtained from the U.S. Geological Survey (USGS) *Minerals Yearbook: Crushed Stone Annual Report* (1995 through 2016a), 2016 and 2017 preliminary data from the USGS Crushed Stone Commodity Expert (Willett 2018a, Willett 2018b), the *USGS Minerals Yearbook: Soda Ash Annual Report* (1995 through 2015) (USGS 1995 through 2015b), *USGS Mineral Industry Surveys for Soda Ash* in February 2018 (USGS 2018) and the U.S. Bureau of Mines (1991 and 1993a), which are reported to the nearest ton. During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; or (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years.

There is a large quantity of limestone and dolomite reported to the USGS under the categories "unspecified–reported" and "unspecified–estimated." A portion of this consumption is believed to be limestone or dolomite used for glass manufacturing. The quantities listed under the "unspecified" categories were, therefore, allocated to glass manufacturing according to the percent limestone or dolomite consumption for glass manufacturing end use for that year.[17] For 2017, the unspecified uses of both limestone and dolomite consumption were not available at the time of publication, so 2016 values were used to proxy these values.

Based on the 2017 reported data, the estimated distribution of soda ash consumption for glass production compared to total domestic soda ash consumption is 48 percent (USGS 1995 through 2015b, 2018).

Table 4-12: Limestone, Dolomite, and Soda Ash Consumption Used in Glass Production (kt)

Activity	1990		2005		2013	2014	2015	2016	2017
Limestone	430		920		693	765	699	472	763

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[17] This approach was recommended by USGS.

Dolomite	59		541		0	0	0	0	0
Soda Ash	3,177		3,050		2,440	2,410	2,390	2,510	2,360
Total	**3,666**		**4,511**		**3,133**	**3,175**	**3,089**	**2,982**	**3,123**

Uncertainty and Time-Series Consistency

The uncertainty levels presented in this section arise in part due to variations in the chemical composition of limestone used in glass production. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals (potassium carbonate, strontium carbonate and barium carbonate, and dead burned dolomite). Similarly, the quality of the limestone (and mix of carbonates) used for glass manufacturing will depend on the type of glass being manufactured.

The estimates below also account for uncertainty associated with activity data. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the manufacturer of the input carbonates (limestone, dolomite and soda ash) and not the end user. For 2017, there has been no reported consumption of dolomite for glass manufacturing. These data have been reported to USGS by dolomite manufacturers and not end-users (i.e., glass manufacturers). There is a high uncertainty associated with this estimate, as dolomite is a major raw material consumed in glass production. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. The uncertainty of the estimates for limestone and dolomite used in glass making is especially high. Lastly, much of the limestone consumed in the United States is reported as "other unspecified uses;" therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses. Further research is needed into alternate and more complete sources of data on carbonate-based raw material consumption by the glass industry.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-13. In 2017, glass production CO_2 emissions were estimated to be between 1.3 and 1.4 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 4 percent below and 5 percent above the emission estimate of 1.3 MMT CO_2 Eq.

Table 4-13: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Glass Production (MMT CO_2 Eq. and Percent)

Source	**Gas**	**2017 Emission Estimate (MMT CO_2 Eq.)**	**Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)**		**(%)**	
			Lower Bound	**Upper Bound**	**Lower Bound**	**Upper Bound**
Glass Production	CO_2	1.3	1.3	1.4	-4%	+5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

As noted in the previous reports, current publicly available activity data shows consumption of only limestone and soda ash for glass manufacturing. While limestone and soda ash are the predominant carbonates used in glass manufacturing, there are other carbonates that are also consumed for glass manufacturing, although in smaller quantities. EPA has initiated review of available activity data on carbonate consumption by type in the glass industry from EPA's Greenhouse Gas Reporting Program (GHGRP) reported annually since 2010, as well as USGS publications. This is a long-term planned improvement.

EPA has initiated review of EPA's GHGRP data and anticipates finalizing assessment for future integration of data in the spring of 2019. This assessment will help to understand the completeness of emission estimates and facilitate category-specific QC per Volume 1 of the *2006 IPCC Guidelines* for the Glass Production source category. EPA's GHGRP has an emission threshold for reporting, so the assessment will consider the completeness of carbonate consumption data for glass production in the United States. Particular attention will also be made to also ensuring time-series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[18] These planned improvements are ongoing and EPA may also initiate research into other sources of activity data for carbonate consumption by the glass industry.

4.4 Other Process Uses of Carbonates (CRF Source Category 2A4)

Limestone ($CaCO_3$), dolomite ($CaCO_3MgCO_3$),[19] and other carbonates such as soda ash, magnesite, and siderite are basic materials used by a wide variety of industries, including construction, agriculture, chemical, metallurgy, glass production, and environmental pollution control. This section addresses only limestone and dolomite use. For industrial applications, carbonates such as limestone and dolomite are heated sufficiently enough to calcine the material and generate CO_2 as a byproduct.

$$CaCO_3 \rightarrow CaO + CO_2$$

$$MgCO_3 \rightarrow MgO + CO_2$$

Examples of such applications include limestone used as a flux or purifier in metallurgical furnaces, as a sorbent in flue gas desulfurization (FGD) systems for utility and industrial plants, and as a raw material for the production of glass, lime, and cement. Emissions from limestone and dolomite used in other process sectors such as cement, lime, glass production, and iron and steel, are excluded from this section and reported under their respective source categories (e.g., Section 4.3, Glass Production). Emissions from soda ash consumption associated with glass manufacturing are reported under Section 4.3 Glass Production (CRF Source Category 2A3). Emissions from fuels consumed for energy purposes during these processes are accounted for in the Energy chapter.

Limestone is widely distributed throughout the world in deposits of varying sizes and degrees of purity. Large deposits of limestone occur in nearly every state in the United States, and significant quantities are extracted for industrial applications. In 2016, the leading limestone producing states were Texas, Florida, Missouri, Ohio, and Illinois, which contributed 50 percent of the total U.S. output (USGS 2018). Similarly, dolomite deposits are also widespread throughout the world. Dolomite deposits are found in the United States, Canada, Mexico, Europe, Africa, and Brazil. In the United States, the leading dolomite producing states are Illinois, Pennsylvania, and New York, which currently contribute more than half of the total U.S. output (USGS 1995a through 2017).

In 2017, 19,851 kt of limestone, 2,088 kt of dolomite, and 2,550 kt of soda ash were consumed for these emissive applications, excluding glass manufacturing (Willett 2018a). Usage of limestone, dolomite and soda ash resulted in aggregate CO_2 emissions of 10.1 MMT CO_2 Eq. (10,139 kt) (see Table 4-14 and Table 4-15). While 2017 emissions have decreased 8 percent compared to 2016, overall emissions have increased 61 percent from 1990 through 2017.

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[18] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[19] Limestone and dolomite are collectively referred to as limestone by the industry, and intermediate varieties are seldom distinguished.

Table 4-14: CO_2 Emissions from Other Process Uses of Carbonates (MMT CO_2 Eq.)

Year	Flux Stone	FGD	Magnesium Production	Soda Ash Consumption[a]	Other Miscellaneous Uses[b]	Total
1990	2.6	1.4	0.1	1.4	0.8	6.3
2005	2.6	3.0	0.0	1.3	0.7	7.6
2013	2.3	6.3	0.0	1.1	1.8	11.5
2014	2.9	7.1	0.0	1.1	1.8	13.0
2015	2.9	7.3	0.0	1.1	0.9	12.2
2016	2.6	6.2	0.0	1.1	1.1	11.0
2017	2.6	5.9	0.0	1.1	0.5	10.1

[a] Soda ash consumption not associated with glass manufacturing.

[b] "Other miscellaneous uses" include chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining.

Note: Totals may not sum due to independent rounding.

Table 4-15: CO_2 Emissions from Other Process Uses of Carbonates (kt)

Year	Flux Stone	FGD	Magnesium Production	Soda Ash Consumption[a]	Other Miscellaneous Uses[b]	Total
1990	2,592	1,432	64	1,390	819	6,297
2005	2,649	2,973	0	1,305	718	7,644
2013	2,307	6,309	0	1,109	1,798	11,524
2014	2,911	7,111	0	1,143	1,790	12,954
2015	2,901	7,335	0	1,075	871	12,182
2016	2,585	6,164	0	1,082	1,137	10,969
2017	2,645	5,904	0	1,058	532	10,139

[a] Soda ash consumption not associated with glass manufacturing.

[b] "Other miscellaneous uses" include chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining.

Note: Totals may not sum due to independent rounding.

Methodology

Carbon dioxide emissions were calculated based on the *2006 IPCC Guidelines* Tier 2 method by multiplying the quantity of limestone or dolomite consumed by the emission factor for limestone or dolomite calcination, respectively – limestone: 0.43971 metric ton CO_2/metric ton carbonate, and dolomite: 0.47732 metric ton CO_2/metric ton carbonate.[20] This methodology was used for flux stone, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining. Flux stone used during the production of iron and steel was deducted from the Other Process Uses of Carbonates source category estimate and attributed to the Iron and Steel Production source category estimate. Similarly, limestone and dolomite consumption for glass manufacturing, cement, and lime manufacturing are excluded from this category and attributed to their respective categories.

Historically, the production of magnesium metal was the only other significant use of limestone and dolomite that produced CO_2 emissions. At the end of 2001, the sole magnesium production plant operating in the United States

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[20] *2006 IPCC Guidelines*, Volume 3: Chapter 2, Table 2.1.

that produced magnesium metal using a dolomitic process that resulted in the release of CO_2 emissions ceased its operations (USGS 1995b through 2012; USGS 2013).

Consumption data for 1990 through 2017 of limestone and dolomite used for flux stone, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining (see Table 4-16) were obtained from the U.S. Geological Survey (USGS) *Minerals Yearbook: Crushed Stone Annual Report* (1995a through 2017), preliminary data for 2016 and 2017 from USGS Crushed Stone Commodity Expert (Willett 2018a, 2018b), American Iron and Steel Institute limestone and dolomite consumption data (AISI 2018), and the U.S. Bureau of Mines (1991 and 1993a), which are reported to the nearest ton. For 2017, the unspecified uses of both limestone and dolomite consumption were not available at the time of publication, so 2016 values were used to proxy these values. The production capacity data for 1990 through 2017 of dolomitic magnesium metal also came from the USGS (1995b through 2012; USGS 2013) and the U.S. Bureau of Mines (1990 through 1993b). During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years; or (3) the average fraction of total limestone or dolomite for the end-use over the entire time period.

There is a large quantity of crushed stone reported to the USGS under the category "unspecified uses." A portion of this consumption is believed to be limestone or dolomite used for emissive end uses. The quantity listed for "unspecified uses" was, therefore, allocated to each reported end-use according to each end-use's fraction of total consumption in that year.[21]

Table 4-16: Limestone and Dolomite Consumption (kt)

Activity	1990	2005	2013	2014	2015	2016	2017
Flux Stone	6,737	7,022	6,345	7,599	7,834	7,092	7,302
Limestone	5,804	3,165	4,380	4,243	4,590	4,118	5,214
Dolomite	933	3,857	1,965	3,356	3,244	2,973	2,088
FGD	3,258	6,761	14,347	16,171	16,680	14,019	13,427
Other Miscellaneous Uses	1,835	1,632	3,973	4,069	1,982	2,587	1,210
Total	**11,830**	**15,415**	**24,665**	**27,839**	**26,496**	**23,698**	**21,939**

Once produced, most soda ash is consumed in chemical production, with minor amounts in soap production, pulp and paper, flue gas desulfurization, and water treatment (excluding soda ash consumption for glass manufacturing). As soda ash is consumed for these purposes, additional CO_2 is usually emitted. In these applications, it is assumed that one mole of carbon is released for every mole of soda ash used. Thus, approximately 0.113 metric tons of carbon (or 0.415 metric tons of CO_2) are released for every metric ton of soda ash consumed. The activity data for soda ash consumption (see Table 4-17) for 1990 to 2017 were obtained from the U.S. Geological Survey (USGS) *Minerals Yearbook for Soda Ash* (1994 through 2015b) and USGS *Mineral Industry Surveys for Soda Ash* (USGS 2017a, 2018). Soda ash consumption data[22] were collected by the USGS from voluntary surveys of the U.S. soda ash industry.

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[21] This approach was recommended by USGS, the data collection agency.

[22] EPA has assessed feasibility of using emissions information (including activity data) from EPA's GHGRP; however, at this time, the aggregated information associated with production of soda ash did not meet criteria to shield underlying confidential business information (CBI) from public disclosure.

Table 4-17: Soda Ash Consumption Not Associated with Glass Manufacturing (kt)

Activity	1990	2005	2013	2014	2015	2016	2017
Soda Ash[a]	3,351	3,144	2,674	2,754	2,592	2,608	2,550
Total	**3,351**	**3,144**	**2,674**	**2,754**	**2,592**	**2,608**	**2,550**

[a] Soda ash consumption is sales reported by producers which exclude imports. Historically, imported soda ash is less than 1 percent of the total U.S. consumption (Kostick 2012).

Uncertainty and Time-Series Consistency

The uncertainty levels presented in this section account for uncertainty associated with activity data. Data on limestone and dolomite consumption are collected by USGS through voluntary national surveys. USGS contacts the mines (i.e., producers of various types of crushed stone) for annual sales data. Data on other carbonate consumption are not readily available. The producers report the annual quantity sold to various end-users and industry types. USGS estimates the historical response rate for the crushed stone survey to be approximately 70 percent, and the rest is estimated by USGS. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the producer/mines and not the end user. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. Lastly, much of the limestone consumed in the United States is reported as "other unspecified uses;" therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses. This year, EPA reinitiated dialogue with the USGS National Minerals Information Center Crushed Stone commodity expert to assess the current uncertainty ranges associated with the limestone and dolomite consumption data compiled and published by USGS. During this discussion, the expert confirmed that EPA's range of uncertainty was still reasonable (Willett 2017a).

Uncertainty in the estimates also arises in part due to variations in the chemical composition of limestone. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals. The exact specifications for limestone or dolomite used as flux stone vary with the pyrometallurgical process and the kind of ore processed.

For emissions from soda ash consumption, the primary source of uncertainty results from the fact that these emissions are dependent upon the type of processing employed by each end-use. Specific emission factors for each end-use are not available, so a Tier 1 default emission factor is used for all end uses. Therefore, there is uncertainty surrounding the emission factors from the consumption of soda ash. Additional uncertainty comes from the reported consumption and allocation of consumption within sectors that is collected on a quarterly basis by the USGS. Efforts have been made to categorize company sales within the correct end-use sector.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-18. Carbon dioxide emissions from other process uses of carbonates in 2017 were estimated to be between 9.0 and 11.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below and 15 percent above the emission estimate of 10.1 MMT CO_2 Eq.

Table 4-18: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Other Process Uses of Carbonates (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Other Process Uses of Carbonates	CO_2	10.1	9.0	11.8	-12%	+15%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

EPA plans to continue the dialogue with USGS to assess uncertainty ranges for activity data used to estimate emissions from other process use of carbonates. This planned improvement is currently planned as a medium-term improvement.

4.5 Ammonia Production (CRF Source Category 2B1)

Emissions of carbon dioxide (CO_2) occur during the production of synthetic ammonia, primarily through the use of natural gas, petroleum coke, or naphtha as a feedstock. The natural gas-, naphtha-, and petroleum coke-based processes produce CO_2 and hydrogen (H_2), the latter of which is used in the production of ammonia. The brine electrolysis process for production of ammonia does not lead to process-based CO_2 emissions. Due to national circumstances, emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter. More information on this approach can be found in the Methodology section, below.

In the United States, the majority of ammonia is produced using a natural gas feedstock; however, one synthetic ammonia production plant located in Kansas is producing ammonia from petroleum coke feedstock. In some U.S. plants, some of the CO_2 produced by the process is captured and used to produce urea rather than being emitted to the atmosphere. In 2017, there were approximately 15 companies operating 32 ammonia producing facilities in 16 states. Roughly 50 percent of domestic ammonia production capacity is concentrated in the states of Louisiana, Oklahoma, and Texas. In 2016, upgrades came online to increase ammonia capacity at one facility in the United States and in 2017 two new ammonia facilities became operational (USGS 2018).

There are five principal process steps in synthetic ammonia production from natural gas feedstock. The primary reforming step converts methane (CH_4) to CO_2, carbon monoxide (CO), and H_2 in the presence of a catalyst. Only 30 to 40 percent of the CH_4 feedstock to the primary reformer is converted to CO and CO_2 in this step of the process. The secondary reforming step converts the remaining CH_4 feedstock to CO and CO_2. The CO in the process gas from the secondary reforming step (representing approximately 15 percent of the process gas) is converted to CO_2 in the presence of a catalyst, water, and air in the shift conversion step. Carbon dioxide is removed from the process gas by the shift conversion process, and the hydrogen gas is combined with the nitrogen (N_2) gas in the process gas during the ammonia synthesis step to produce ammonia. The CO_2 is included in a waste gas stream with other process impurities and is absorbed by a scrubber solution. In regenerating the scrubber solution, CO_2 is released from the solution.

The conversion process for conventional steam reforming of CH_4, including the primary and secondary reforming and the shift conversion processes, is approximately as follows:

$$0.88CH_4 + 1.26Air + 1.24H_2O \rightarrow 0.88CO_2 + N_2 + 3H_2$$

$$N_2 + 3H_2 \rightarrow 2NH_3$$

To produce synthetic ammonia from petroleum coke, the petroleum coke is gasified and converted to CO_2 and H_2. These gases are separated, and the H_2 is used as a feedstock to the ammonia production process, where it is reacted with N_2 to form ammonia.

Not all of the CO_2 produced during the production of ammonia is emitted directly to the atmosphere. Some of the ammonia and some of the CO_2 produced by the synthetic ammonia process are used as raw materials in the production of urea [$CO(NH_2)_2$], which has a variety of agricultural and industrial applications.

The chemical reaction that produces urea is:

$$2NH_3 + CO_2 \rightarrow NH_2COONH_4 \rightarrow CO(NH_2)_2 + H_2O$$

Only the CO_2 emitted directly to the atmosphere from the synthetic ammonia production process is accounted for in determining emissions from ammonia production. The CO_2 that is captured during the ammonia production process and used to produce urea does not contribute to the CO_2 emission estimates for ammonia production presented in this section. Instead, CO_2 emissions resulting from the consumption of urea are attributed to the urea consumption or urea application source category (under the assumption that the carbon stored in the urea during its manufacture is released into the environment during its consumption or application). Emissions of CO_2 resulting from agricultural applications of urea are accounted for in the Agriculture chapter. Previously, these emission estimates from the agricultural application of urea were accounted for in the *Cropland Remaining Cropland* section of the Land Use, Land Use Change, and Forestry chapter. Emissions of CO_2 resulting from non-agricultural applications of urea (e.g., use as a feedstock in chemical production processes) are accounted for in Section 4.6 Urea Consumption for Non-Agricultural Purposes of this chapter.

Total emissions of CO_2 from ammonia production in 2017 were 13.2 MMT CO_2 Eq. (13,216 kt), and are summarized in Table 4-19 and Table 4-20. Ammonia production relies on natural gas as both a feedstock and a fuel, and as such, market fluctuations and volatility in natural gas prices affect the production of ammonia. Since 1990, emissions from ammonia production have increased by 1.3 percent. Emissions in 2017 have increased by approximately 22 percent from the 2016 levels. Agricultural demands continue to drive demand for nitrogen fertilizers (USGS 2018).

Table 4-19: CO_2 Emissions from Ammonia Production (MMT CO_2 Eq.)

Source	**1990**	**2005**	**2013**	**2014**	**2015**	**2016**	**2017**
Ammonia Production	13.0	9.2	9.5	9.4	10.6	10.8	13.2
Total	**13.0**	**9.2**	**9.5**	**9.4**	**10.6**	**10.8**	**13.2**

Table 4-20: CO_2 Emissions from Ammonia Production (kt)

Source	**1990**	**2005**	**2013**	**2014**	**2015**	**2016**	**2017**
Ammonia Production	13,047	9,196	9,480	9,377	10,634	10,838	13,216
Total	**13,047**	**9,196**	**9,480**	**9,377**	**10,634**	**10,838**	**13,216**

Methodology

For the U.S. Inventory, CO_2 emissions from the production of synthetic ammonia from natural gas feedstock are estimated using a country-specific approach modified from the *2006 IPCC Guidelines* (IPCC 2006) Tier 1 and 2 methods. In the country-specific approach, emissions are not based on total fuel requirement per the *2006 IPCC Guidelines* due to data disaggregation limitations of energy statistics provided by the Energy Information Administration (EIA). A country-specific emission factor is developed and applied to national ammonia production to estimate emissions. The method uses a CO_2 emission factor published by the European Fertilizer Manufacturers Association (EFMA) that is based on natural gas-based ammonia production technologies that are similar to those employed in the United States. This CO_2 emission factor of 1.2 metric tons CO_2/metric ton NH_3 (EFMA 2000a) is applied to the percent of total annual domestic ammonia production from natural gas feedstock.

Emissions of CO_2 from ammonia production are then adjusted to account for the use of some of the CO_2 produced from ammonia production as a raw material in the production of urea. The CO_2 emissions reported for ammonia production are reduced by a factor of 0.733 multiplied by total annual domestic urea production. This corresponds to a stoichiometric CO_2/urea factor of 44/60, assuming complete conversion of ammonia (NH_3) and CO_2 to urea (IPCC 2006; EFMA 2000b).

All synthetic ammonia production and subsequent urea production are assumed to be from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of ammonia production from petroleum coke feedstock at one plant located in Kansas. Annual ammonia and urea production are shown in Table 4-21. The CO_2 emission factor for production of ammonia from petroleum coke is based on plant-specific data, wherein all carbon contained in the petroleum coke feedstock that is not used for urea production is assumed to be emitted to the atmosphere as CO_2 (Bark 2004). Ammonia and urea are assumed to be manufactured in the same manufacturing complex, as both the raw materials needed for urea production are produced by the ammonia production process. The CO_2 emission factor of 3.57 metric tons CO_2/metric ton NH_3 for the petroleum coke feedstock process (Bark 2004) is applied to the percent of total annual domestic ammonia production from petroleum coke feedstock.

The emission factor of 1.2 metric ton CO_2/metric ton NH_3 for production of ammonia from natural gas feedstock was taken from the EFMA Best Available Techniques publication, Production of Ammonia (EFMA 2000a). The EFMA reported an emission factor range of 1.15 to 1.30 metric ton CO_2/metric ton NH_3, with 1.2 metric ton CO_2/metric ton NH_3 as a typical value (EFMA 2000a). Technologies (e.g., catalytic reforming process, etc.) associated with this factor are found to closely resemble those employed in the United States for use of natural gas as a feedstock. The EFMA reference also indicates that more than 99 percent of the CH_4 feedstock to the catalytic reforming process is ultimately converted to CO_2.

The consumption of natural gas and petroleum coke as fossil fuel feedstocks for NH_3 production are adjusted for within the Energy chapter as these fuels were consumed during non-energy related activities. More information on this methodology is described in Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion. See the Planned Improvements section on improvements of reporting fuel and feedstock CO_2 emissions utilizing EPA's GHGRP data to improve consistency with *2006 IPCC Guidelines*.

The total ammonia production data for 2011 through 2017 were obtained from American Chemistry Council (ACC 2018). For years before 2011, ammonia production data (see Table 4-21) were obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011, and 2012) and the Census Bureau of the U.S. Department of Commerce (U.S. Census Bureau 1991 through 1994, 1998 through 2011) as reported in Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports. Urea-ammonia nitrate production from petroleum coke for years through 2011 was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011, and 2012), and from *CVR Energy, Inc. Annual Report* (CVR 2012, 2013, 2014, 2015, 2016, and 2017) for 2012 through 2017. Urea production data for 1990 through 2008 were obtained from the *Minerals Yearbook: Nitrogen* (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Census Bureau (U.S. Census Bureau 2010 and 2011). The U.S. Census Bureau ceased collection of urea production statistics in 2011.

For the current Inventory (i.e., 1990 through 2017), EPA began utilizing urea production data from EPA's GHGRP Subpart G to estimate emissions. Urea production values in the current Inventory report utilize GHGRP data for the years 2011 through 2017 (EPA 2018). Details on QA checks and how this change compares to previously used USGS *Minerals Yearbook: Nitrogen* (USGS 2014 through 2016) reported data can be found in the Uncertainty and Time-Series Consistency section, below. GHGRP urea production data for 2017 were not yet published and so 2016 data were used as a proxy.

Table 4-21: Ammonia Production, Recovered CO_2 Consumed for Urea Production, and Urea Production (kt)

Year	Ammonia Production	Total CO_2 Consumption for Urea Production	Urea Production
1990	15,425	5,463	7,450
2005	10,143	3,865	5,270
2013	10,930	4,501	6,137
2014	10,515	4,078	5,561
2015	11,765	4,312	5,880
2016	12,305	5,419	7,390
2017	14,070	5,419	7,390

Uncertainty and Time-Series Consistency

The uncertainties presented in this section are primarily due to how accurately the emission factor used represents an average across all ammonia plants using natural gas feedstock. Uncertainties are also associated with ammonia production estimates and the assumption that all ammonia production and subsequent urea production was from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of one ammonia production plant located in Kansas that is manufacturing ammonia from petroleum coke feedstock. Uncertainty is also associated with the representativeness of the emission factor used for the petroleum coke-based ammonia process. It is also assumed that ammonia and urea are produced at collocated plants from the same natural gas raw material. The uncertainty of the total urea production activity data, based on USGS *Minerals Yearbook: Nitrogen* data, is a function of the reliability of reported production data and is influenced by the completeness of the survey responses. In addition, due to the fact that 2017 nitrogen data has yet to be published, 2016 is used as a proxy which may result in greater uncertainty.

Recovery of CO_2 from ammonia production plants for purposes other than urea production (e.g., commercial sale, etc.) has not been considered in estimating the CO_2 emissions from ammonia production, as data concerning the disposition of recovered CO_2 are not available. Such recovery may or may not affect the overall estimate of CO_2 emissions depending upon the end use to which the recovered CO_2 is applied. Further research is required to determine whether byproduct CO_2 is being recovered from other ammonia production plants for application to end uses that are not accounted for elsewhere. However, for reporting purposes, CO_2 consumption for urea production is provided in this chapter.

EPA relied upon the latest guidance from the IPCC on the use of facility-level data in national inventories and applied a category-specific QC process to compare activity data from GHGRP with existing data from USGS. This was to ensure time-series consistency of the emission estimates presented in the Inventory. For the year 2011, USGS and GHGRP urea production data showed a difference of approximately 27 percent in 2011, 11 percent in 2012, 12 percent in 2013, 6 percent in 2014 and 2015, and 12 percent in 2016. The use of GHGRP data (EPA 2018) resulted in an decrease of emissions compared to USGS data (USGS 2011 through 2018) ranging from 0.24 to 1.06 MMT CO_2 Eq. for 2011 through 2016.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-22. Carbon dioxide emissions from ammonia production in 2017 were estimated to be between 12.6 and 13.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below and 5 percent above the emission estimate of 13.2 MMT CO_2 Eq.

Table 4-22: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Ammonia Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Ammonia Production	CO_2	13.2	12.6	13.8	-5%	+5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Recalculations Discussion

This current Inventory (i.e., 1990 through 2017) has been updated to include estimates of CO_2 emissions from urea consumption for the years 2011 through 2017) based on urea production data available from the GHGRP. Urea production data available from USGS was used in estimating emissions in prior inventories.

Urea production based on USGS data ranged from a low of 5,220 kt to a high of 6,610 kt over the period from 2011 through 2017. Urea production based on GHGRP data ranged from a low of 5,561 kt to a high of 7,390 kt over the same time period. These data show that total annual urea production based on GHGRP data averaged 12 percent higher than the total annual urea production based on USGS data during the same time period.

The estimated CO_2 emissions from ammonia production in 2011 through 2017 are approximately 5 percent lower in the current Inventory (i.e., 1990 through 2017) than the previous Inventory (i.e., 1990 through 2016).

Planned Improvements

Future improvements involve continuing to evaluate and analyze data reported under EPA's GHGRP to improve the emission estimates for the Ammonia Production source category, in particular new data from updated reporting requirements finalized in October of 2014 (79 FR 63750) and December 2016 (81 FR 89188),[23] that include facility-level ammonia production data and feedstock consumption. This data will first be reported by facilities in 2018 and available post-verification to assess in early 2019 for use in future Inventories (e.g., 2021 Inventory report) if the data meets GHGRP CBI aggregation criteria. Particular attention will be made to ensure time-series consistency of the emission estimates presented in future Inventory reports, along with application of appropriate category-specific QC procedures consistent with IPCC and UNFCCC guidelines. For example, data reported in 2018 will reflect activity in 2017 and may not be representative of activity in prior years of the time series. This assessment is required as the new facility-level reporting data from EPA's GHGRP associated with new requirements are only applicable starting with reporting of emissions in calendar year 2017, and thus are not available for all inventory years (i.e., 1990 through 2017) as required for this Inventory.

In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[24] Specifically, the planned improvements include assessing the anticipated new data to update the emission factors to include both fuel and feedstock CO_2 emissions to improve consistency with *2006 IPCC Guidelines,* in addition to reflecting CO_2 capture and storage practices (beyond use of CO_2 for urea production). Methodologies will also be updated if additional ammonia production plants are found to use hydrocarbons other than natural gas for ammonia production. Due to limited resources and ongoing data collection efforts, this planned improvement is still in development and so is not incorporated into this Inventory. This is a long-term planned improvement.

4.6 Urea Consumption for Non-Agricultural Purposes

Urea is produced using ammonia and carbon dioxide (CO_2) as raw materials. All urea produced in the United States is assumed to be produced at ammonia production facilities where both ammonia and CO_2 are generated. There were 32 plants producing ammonia in the United States during 2017, with two additional plants sitting idle for the entire year (USGS 2018).

The chemical reaction that produces urea is:

$$2NH_3 + CO_2 \rightarrow NH_2COONH_4 \rightarrow CO(NH_2)_2 + H_2O$$

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[23] See <https://www.epa.gov/ghgreporting/historical-rulemakings>.

[24] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

This section accounts for CO_2 emissions associated with urea consumed exclusively for non-agricultural purposes. Carbon dioxide emissions associated with urea consumed for fertilizer are accounted for in the Agriculture chapter.

Urea is used as a nitrogenous fertilizer for agricultural applications and also in a variety of industrial applications. The industrial applications of urea include its use in adhesives, binders, sealants, resins, fillers, analytical reagents, catalysts, intermediates, solvents, dyestuffs, fragrances, deodorizers, flavoring agents, humectants and dehydrating agents, formulation components, monomers, paint and coating additives, photosensitive agents, and surface treatments agents. In addition, urea is used for abating nitrogen oxide (NO_x) emissions from coal-fired power plants and diesel transportation motors.

Emissions of CO_2 from urea consumed for non-agricultural purposes in 2017 were estimated to be 5.0 MMT CO_2 Eq. (4,958 kt), and are summarized in Table 4-23 and Table 4-24. Net CO_2 emissions from urea consumption for non-agricultural purposes in 2017 have increased by approximately 31 percent from 1990. The significant decrease in emissions during 2014 can be attributed to a decrease in the amount of urea imported by the United States during that year.

Table 4-23: CO_2 Emissions from Urea Consumption for Non-Agricultural Purposes (MMT CO_2 Eq.)

Source	1990	2005	2013	2014	2015	2016	2017
Urea Consumption	3.8	3.7	4.6	1.8	4.6	5.1	5.0
Total	**3.8**	**3.7**	**4.6**	**1.8**	**4.6**	**5.1**	**5.0**

Table 4-24: CO_2 Emissions from Urea Consumption for Non-Agricultural Purposes (kt)

Source	1990	2005	2013	2014	2015	2016	2017
Urea Consumption	3,784	3,653	4,556	1,807	4,578	5,132	4,958
Total	**3,784**	**3,653**	**4,556**	**1,807**	**4,578**	**5,132**	**4,958**

Methodology

Emissions of CO_2 resulting from urea consumption for non-agricultural purposes are estimated by multiplying the amount of urea consumed in the United States for non-agricultural purposes by a factor representing the amount of CO_2 used as a raw material to produce the urea. This method is based on the assumption that all of the carbon in urea is released into the environment as CO_2 during use, and consistent with the *2006 IPCC Guidelines*.

The amount of urea consumed for non-agricultural purposes in the United States is estimated by deducting the quantity of urea fertilizer applied to agricultural lands, which is obtained directly from the Agriculture chapter (see Table 5-24) and is reported in Table 4-25, from the total domestic supply of urea. In previous Inventory reports, the quantity of urea fertilizer applied to agricultural lands was obtained directly from the *Cropland Remaining Cropland* section of the Land Use, Land Use Change, and Forestry chapter. The domestic supply of urea is estimated based on the amount of urea produced plus the sum of net urea imports and exports. A factor of 0.733 tons of CO_2 per ton of urea consumed is then applied to the resulting supply of urea for non-agricultural purposes to estimate CO_2 emissions from the amount of urea consumed for non-agricultural purposes. The 0.733 tons of CO_2 per ton of urea emission factor is based on the stoichiometry of producing urea from ammonia and CO_2. This corresponds to a stoichiometric CO_2/urea factor of 44/60, assuming complete conversion of NH_3 and CO_2 to urea (IPCC 2006; EFMA 2000).

Urea production data for 1990 through 2008 were obtained from the *Minerals Yearbook: Nitrogen* (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Census Bureau (2011). The U.S. Census Bureau ceased collection of urea production statistics in 2011. For the current Inventory (i.e., 1990 through 2017), EPA began utilizing urea production data from EPA's GHGRP to estimate emissions. Urea production values in the current Inventory report utilize GHGRP data for the years 2011 through 2017 (EPA 2018). Details on QA checks and how this change compares to previously used USGS *Minerals Yearbook: Nitrogen*

(USGS 2014 through 2016) reported data can be found in the Uncertainty and Time-Series Consistency section, below.

Urea import data for 2017 are not yet publicly available and so 2016 data have been used as proxy. Urea import data for 2013 to 2016 were obtained from the *Minerals Yearbook: Nitrogen* (USGS 2016). Urea import data for 2011 and 2012 were taken from U.S. Fertilizer Import/Exports from the United States Department of Agriculture (USDA) Economic Research Service Data Sets (U.S. Department of Agriculture 2012). USDA suspended updates to this data after 2012. Urea import data for the previous years were obtained from the U.S. Census Bureau *Current Industrial Reports Fertilizer Materials and Related Products* annual and quarterly reports for 1997 through 2010 (U.S. Census Bureau 2001 through 2011), The Fertilizer Institute (TFI 2002) for 1993 through 1996, and the United States International Trade Commission Interactive Tariff and Trade DataWeb (U.S. ITC 2002) for 1990 through 1992 (see Table 4-25).

Urea export data for 2017 are not yet publicly available and so 2016 data have been used as proxy. Urea export data for 2013 to 2016 were obtained from the *Minerals Yearbook: Nitrogen* (USGS 2016). Urea export data for 1990 through 2012 were taken from U.S. Fertilizer Import/Exports from USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2012). USDA suspended updates to this data after 2012.

Table 4-25: Urea Production, Urea Applied as Fertilizer, Urea Imports, and Urea Exports (kt)

Year	Urea Production	Urea Applied as Fertilizer	Urea Imports	Urea Exports
1990	7,450	3,296	1,860	854
2005	5,270	4,779	5,026	536
2013	6,137	6,059	6,470	335
2014	5,561	6,156	3,510	451
2015	5,880	6,447	7,190	380
2016	7,390	6,651	6,580	321
2017	7,390	6,888	6,580	321

Uncertainty and Time-Series Consistency

There is limited publicly-available data on the quantities of urea produced and consumed for non-agricultural purposes. Therefore, the amount of urea used for non-agricultural purposes is estimated based on a balance that relies on estimates of urea production, urea imports, urea exports, and the amount of urea used as fertilizer. The primary uncertainties associated with this source category are associated with the accuracy of these estimates as well as the fact that each estimate is obtained from a different data source. Because urea production estimates are no longer available from the USGS, there is additional uncertainty associated with urea produced beginning in 2011. There is also uncertainty associated with the assumption that all of the carbon in urea is released into the environment as CO_2 during use.

EPA relied upon the latest guidance from the IPCC on the use of facility-level data in national inventories and applied a category-specific QC process to compare activity data from GHGRP with existing data from USGS. This was to ensure time-series consistency of the emission estimates presented in the Inventory. For the year 2011, USGS and GHGRP urea production data showed a difference of approximately 27 percent in 2011, 11 percent in 2012, 12 percent in 2013, 6 percent in 2014 and 2015, and 12 percent in 2016. The use of GHGRP data (EPA 2018) resulted in an increase of emissions compared to USGS data (USGS 2011 through 2018) ranging from 0.24 to 1.06 MMT CO_2 Eq. for 2011 through 2016.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-26. Carbon dioxide emissions associated with urea consumption for non-agricultural purposes during 2017 were estimated to be between 4.4 and 5.5 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below and 12 percent above the emission estimate of 5.0 MMT CO_2 Eq.

Table 4-26: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Urea Consumption for Non-Agricultural Purposes (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Urea Consumption for Non-Agricultural Purposes	CO_2	5.0	4.4	5.5	-12%	+12%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Recalculations Discussion

This current Inventory (i.e., 1990 through 2017) has been updated to include estimates of CO_2 emissions from urea consumption for the years 2011 through 2017 based on urea production data available from the GHGRP. Urea production data available from USGS was used in estimating emissions in prior inventories.

Urea production based on USGS data ranged from a low of 5,220 kt to a high of 6,610 kt over the period from 2011 through 2017. Urea production based on GHGRP data ranged from a low of 5,561 kt to a high of 7,390 kt over the same time period. These data show that total annual urea production based on GHGRP data averaged 12 percent higher than the total annual urea production based on USGS data during the same time period.

Taking into account the fluctuations in urea application for agricultural uses, urea imports, and urea exports, the estimated CO_2 emissions from urea consumption in 2011 through 2017 are approximately 13 percent higher in the current Inventory (i.e., 1990 through 2017) than the previous Inventory (i.e., 1990 through 2016).

4.7 Nitric Acid Production (CRF Source Category 2B2)

Nitrous oxide (N_2O) is emitted during the production of nitric acid (HNO_3), an inorganic compound used primarily to make synthetic commercial fertilizers. It is also a major component in the production of adipic acid—a feedstock for nylon—and explosives. Virtually all of the nitric acid produced in the United States is manufactured by the high-temperature catalytic oxidation of ammonia (EPA 1998). There are two different nitric acid production methods: weak nitric acid and high-strength nitric acid. The first method utilizes oxidation, condensation, and absorption to produce nitric acid at concentrations between 30 and 70 percent nitric acid. High-strength acid (90 percent or greater nitric acid) can be produced from dehydrating, bleaching, condensing, and absorption of the weak nitric acid. The basic process technology for producing nitric acid has not changed significantly over time. Most U.S. plants were built between 1960 and 2000. As of 2017, there were 31 active nitric acid production plants, including one high-strength nitric acid production plant in the United States (EPA 2010; EPA 2018).

During this reaction, N_2O is formed as a byproduct and is released from reactor vents into the atmosphere. Emissions from fuels consumed for energy purposes during the production of nitric acid are accounted for in the Energy chapter.

Nitric acid is made from the reaction of ammonia (NH_3) with oxygen (O_2) in two stages. The overall reaction is:

$$4NH_3 + 8O_2 \rightarrow 4HNO_3 + 4H_2O$$

Currently, the nitric acid industry controls emissions of NO and NO_2 (i.e., NO_x). As such, the industry in the United States uses a combination of non-selective catalytic reduction (NSCR) and selective catalytic reduction (SCR) technologies. In the process of destroying NO_x, NSCR systems are also very effective at destroying N_2O. However, NSCR units are generally not preferred in modern plants because of high energy costs and associated high gas temperatures. NSCR systems were installed in nitric plants built between 1971 and 1977 with NSCRs installed at approximately one-third of the weak acid production plants. U.S. facilities are using both tertiary (i.e., NSCR) and secondary controls (i.e., alternate catalysts).

Nitrous oxide emissions from this source were estimated to be 9.3 MMT CO_2 Eq. (31 kt of N_2O) in 2017 (see Table 4-27). Emissions from nitric acid production have decreased by 23 percent since 1990, while production has increased by 8 percent over the same time period. Emissions have decreased by 35 percent since 1997, the highest year of production in the time series.

Table 4-27: N_2O Emissions from Nitric Acid Production (MMT CO_2 Eq. and kt N_2O)

Year	MMT CO_2 Eq.	kt N_2O
1990	12.1	41
2005	11.3	38
2013	10.7	36
2014	10.9	37
2015	11.6	39
2016	10.1	34
2017	9.3	31

Methodology

Emissions of N_2O were calculated using the estimation methods provided by the *2006 IPCC Guidelines* and country-specific methods from EPA's GHGRP. The *2006 IPCC Guidelines* Tier 2 method was used to estimate emissions from nitric acid production for 1990 through 2009, and a country-specific approach similar to the IPCC Tier 3 method was used to estimate N_2O emissions for 2010 through 2017.

2010 through 2017

Process N_2O emissions and nitric acid production data were obtained directly from EPA's GHGRP for 2010 through 2017 by aggregating reported facility-level data (EPA 2018). In the United States, all nitric acid facilities producing weak nitric acid (30 to 70 percent in strength) are required to report annual greenhouse gas emissions data to EPA as per the requirements of its GHGRP. As of 2017, there were 31 facilities that reported to EPA, including the known single high-strength nitric acid production facility in the United States (EPA 2018). All nitric acid (weak acid) facilities are required to calculate process emissions using a site-specific emission factor developed through annual performance testing under typical operating conditions or by directly measuring N_2O emissions using monitoring equipment.[25] The high-strength nitric acid facility also reports N_2O emissions associated with weak acid production and this may capture all relevant emissions, pending additional further EPA research. More details on the calculation, monitoring and QA/QC methods applicable to nitric acid facilities can be found under Subpart V: Nitric Acid Production of the regulation, Part 98.[26] EPA verifies annual facility-level GHGRP reports through a multi-step

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[25] Facilities must use standard methods, either EPA Method 320 or ASTM D6348-03 and must follow associated QA/QC procedures consistent during these performance test consistent with category-specific QC of direct emission measurements.

[26] See <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>.

process (e.g., combination of electronic checks and manual reviews) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent. Based on the results of the verification process, EPA follows up with facilities to resolve mistakes that may have occurred.[27]

To calculate emissions from 2010 through 2017, the GHGRP nitric acid production data are utilized to develop weighted country-specific emission factors used to calculate emissions estimates. Based on aggregated nitric acid production data by abatement type (i.e., with, without) provided by EPA's GHGRP, the percent of production values and associated emissions of nitric acid with and without abatement technologies are calculated. These percentages are the basis for developing the country-specific weighted emission factors which vary from year to year based on the amount of nitric acid production with and without abatement technologies.

1990 through 2009

Using GHGRP data for 2010,[28] country-specific N_2O emission factors were calculated for nitric acid production with abatement and without abatement (i.e., controlled and uncontrolled emission factors), as previously stated. The following 2010 emission factors were derived for production with abatement and without abatement: 3.3 kg N_2O/metric ton HNO_3 produced at plants using abatement technologies (e.g., tertiary systems such as NSCR systems) and 5.99 kg N_2O/metric ton HNO_3 produced at plants not equipped with abatement technology. Country-specific weighted emission factors were derived by weighting these emission factors by percent production with abatement and without abatement over time periods 1990 through 2008 and 2009. These weighted emission factors were used to estimate N_2O emissions from nitric acid production for years prior to the availability of GHGRP data (i.e., 1990 through 2008 and 2009). A separate weighted factor is included for 2009 due to data availability for that year. At that time, EPA had initiated compilation of a nitric acid database to improve estimation of emissions from this industry and obtained updated information on application of controls via review of permits and outreach with facilities and trade associations. The research indicated recent installation of abatement technologies at additional facilities.

Based on the available data, it was assumed that emission factors for 2010 would be more representative of operating conditions in 1990 through 2009 than more recent years. Initial review of historical data indicates that percent production with and without abatement can change over time and also year over year due to changes in application of facility-level abatement technologies, maintenance of abatement technologies, and also due to plant closures and start-ups (EPA 2012, 2013; Desai 2012; CAR 2013). The installation dates of N_2O abatement technologies are not known at most facilities, but it is assumed that facilities reporting abatement technology use have had this technology installed and operational for the duration of the time series considered in this report (especially NSCRs).

The country-specific weighted N_2O emission factors were used in conjunction with annual production to estimate N_2O emissions for 1990 through 2009, using the following equations:

$$E_i = P_i \times EF_{weighted,i}$$

$$EF_{weighted,i} = \left[\left(\%P_{C,i} \times EF_c\right) + \left(\%P_{unc,i} \times EF_{unc}\right)\right]$$

where,

E_i = Annual N_2O Emissions for year i (kg/yr)
P_i = Annual nitric acid production for year i (metric tons HNO_3)
$EF_{weighted,i}$ = Weighted N_2O emission factor for year i (kg N_2O/metric ton HNO_3)
$\%P_{c,i}$ = Percent national production of HNO_3 with N_2O abatement technology (%)
EF_c = N_2O emission factor, with abatement technology (kg N_2O/metric ton HNO_3)
$\%P_{unc,i}$ = Percent national production of HNO_3 without N_2O abatement technology (%)
EF_{unc} = N_2O emission factor, without abatement technology (kg N_2O/metric ton HNO_3)
i = year from 1990 through 2009

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[27] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf>.

[28] National N_2O process emissions, national production, and national share of nitric acid production with abatement and without abatement technology was aggregated from the GHGRP facility-level data for 2010 to 2017 (i.e., percent production with and without abatement).

- For 2009: Weighted N_2O emission factor = 5.46 kg N_2O/metric ton HNO_3.
- For 1990 through 2008: Weighted N_2O emission factor = 5.66 kg N_2O/metric ton HNO_3.

Nitric acid production data for the United States for 1990 through 2009 were obtained from the U.S. Census Bureau (U.S. Census Bureau 2008, 2009, 2010a, 2010b) (see Table 4-28). Publicly-available information on plant-level abatement technologies was used to estimate the shares of nitric acid production with and without abatement for 2008 and 2009 (EPA 2012, 2013; Desai 2012; CAR 2013). EPA has previously conducted a review of operating permits to obtain more current information due to the lack of publicly-available data on use of abatement technologies for 1990 through 2007, as stated previously; therefore, the share of national production with and without abatement for 2008 was assumed to be constant for 1990 through 2007.

Table 4-28: Nitric Acid Production (kt)

Year	kt
1990	7,200
2005	6,710
2013	7,580
2014	7,660
2015	7,210
2016	7,810
2017	7,780

Uncertainty and Time-Series Consistency

Uncertainty associated with the parameters used to estimate N_2O emissions includes the share of U.S. nitric acid production attributable to each emission abatement technology over the time series (especially prior to 2010), and the associated emission factors applied to each abatement technology type. While some information has been obtained through outreach with industry associations, limited information is available over the time series (especially prior to 2010) for a variety of facility level variables, including plant-specific production levels, plant production technology (e.g., low, high pressure, etc.), and abatement technology type, installation date of abatement technology, and accurate destruction and removal efficiency rates. Production data prior to 2010 were obtained from National Census Bureau, which does not provide uncertainty estimates with their data. Facilities reporting to EPA's GHGRP must measure production using equipment and practices used for accounting purposes. At this time EPA does not estimate uncertainty of the aggregated facility-level information. As noted in the Methodology section, EPA verifies annual facility-level reports through a multi-step process (e.g., combination of electronic checks and manual reviews by staff) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent. The annual production reported by each nitric acid facility under EPA's GHGRP and then aggregated to estimate national N_2O emissions is assumed to have low uncertainty.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-29. Nitrous oxide emissions from nitric acid production were estimated to be between 8.9 and 9.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the 2017 emissions estimate of 9.3 MMT CO_2 Eq.

Table 4-29: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from Nitric Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		Uncertainty Range Relative to Emission Estimate[a] (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Nitric Acid Production	N_2O	9.3	8.9	9.8	-5%	+5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017.

To maintain consistency across the time series and with the rounding approaches taken by other data sets, a new rounding approach was performed for the GHGRP Subpart V: Nitric Acid data. This resulted in production data changes across the time series of 2010 to 2017, in which EPA's GHGRP data have been utilized. The results of this update have had an insignificant impact on the emission estimates across the 2010 to 2017 time series. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Pending resources, EPA is considering both near-term and long-term improvement to estimates and associated characterization of uncertainty. In the short-term, with 8 years of EPA's GHGRP data, EPA anticipates completing updates of category-specific QC procedures to potentially also improve both qualitative and quantitative uncertainty estimates. Longer term, in 2020, EPA anticipates having information from EPA's GHGRP facilities on the installation date of any N_2O abatement equipment, per recent revisions finalized in December 2016 to EPA's GHGRP. This information will enable more accurate estimation of N_2O emissions from nitric acid production over the time series.

4.8 Adipic Acid Production (CRF Source Category 2B3)

Adipic acid is produced through a two-stage process during which nitrous oxide (N_2O) is generated in the second stage. Emissions from fuels consumed for energy purposes during the production of adipic acid are accounted for in the Energy chapter. The first stage of manufacturing usually involves the oxidation of cyclohexane to form a cyclohexanone/cyclohexanol mixture. The second stage involves oxidizing this mixture with nitric acid to produce adipic acid. Nitrous oxide is generated as a byproduct of the nitric acid oxidation stage and is emitted in the waste gas stream (Thiemens and Trogler 1991). The second stage is represented by the following chemical reaction:

$$(CH_2)_5CO(cyclohexanone) + (CH_2)_5CHOH(cyclohexanol) + wHNO_3 \\ \rightarrow HOOC(CH_2)_4COOH(adipic\ acid) + xN_2O + yH_2O$$

Process emissions from the production of adipic acid vary with the types of technologies and level of emission controls employed by a facility. In 1990, two major adipic acid-producing plants had N_2O abatement technologies in place and, as of 1998, three major adipic acid production facilities had control systems in place (Reimer et al. 1999).

In 2017, catalytic reduction, non-selective catalytic reduction (NSCR) and thermal reduction abatement technologies were applied as N_2O abatement measures at adipic acid facilities (EPA 2017).

Worldwide, only a few adipic acid plants exist. The United States, Europe, and China are the major producers, with the United States accounting for the largest share of global adipic acid production capacity in recent years. In 2017, the United States had two companies with a total of two adipic acid production facilities (one in Texas and one in Florida) following the ceased operations of a third major production facility at the end of 2015 (EPA 2017).

Adipic acid is a white crystalline solid used in the manufacture of synthetic fibers, plastics, coatings, urethane foams, elastomers, and synthetic lubricants. Commercially, it is the most important of the aliphatic dicarboxylic acids, which are used to manufacture polyesters. Eighty-four percent of all adipic acid produced in the United States is used in the production of nylon 6,6; 9 percent is used in the production of polyester polyols; 4 percent is used in the production of plasticizers; and the remaining 4 percent is accounted for by other uses, including unsaturated polyester resins and food applications (ICIS 2007). Food grade adipic acid is used to provide some foods with a "tangy" flavor (Thiemens and Trogler 1991).

National adipic acid production has increased by approximately 10 percent over the period of 1990 through 2017, to approximately 830,000 metric tons (ACC 2018). Nitrous oxide emissions from adipic acid production were estimated to be 7.4 MMT CO_2 Eq. (25 kt N_2O) in 2017 (see Table 4-30). Over the period 1990 through 2017, emissions have been reduced by 51 percent due to both the widespread installation of pollution control measures in the late 1990s and plant idling in the late 2000s. Very little information on annual trends in the activity data exist for adipic acid.

Table 4-30: N_2O Emissions from Adipic Acid Production (MMT CO_2 Eq. and kt N_2O)

Year	MMT CO_2 Eq.	kt N_2O
1990	15.2	51
2005	7.1	24
2013	3.9	13
2014	5.4	18
2015	4.3	14
2016	7.0	23
2017	7.4	25

Methodology

Emissions are estimated using both Tier 2 and Tier 3 methods consistent with the *2006 IPCC Guidelines.* Due to confidential business information (CBI), plant names are not provided in this section. Therefore, the four adipic acid-producing facilities that have operated over the time series will be referred to as Plants 1 through 4. Overall, as noted above, the two currently operating facilities use catalytic reduction, NSCR and thermal reduction abatement technologies.

2010 through 2017

All emission estimates for 2010 through 2017 were obtained through analysis of GHGRP data (EPA 2010 through 2013; EPA 2014 through 2016; EPA 2017), which is consistent with the *2006 IPCC Guidelines* Tier 3 method. Facility-level greenhouse gas emissions data were obtained from EPA's GHGRP for the years 2010 through 2017 (EPA 2010 through 2013; EPA 2014 through 2016; EPA 2017) and aggregated to national N_2O emissions. Consistent with IPCC Tier 3 methods, all adipic acid production facilities are required to calculate emissions using a facility-specific emission factor developed through annual performance testing under typical operating conditions or by directly measuring N_2O emissions using monitoring equipment.[205] More information on the calculation, monitoring and QA/QC methods for process N_2O emissions applicable to adipic acid production facilities under Subpart E can be found in the electronic code of federal regulations.[206] EPA verifies annual facility-level GHGRP reports through a multi-step process (e.g., combination of electronic checks and manual reviews) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent.[207]

1990 through 2009

For years 1990 through 2009, which were prior to EPA's GHGRP reporting, for both Plants 1 and 2, emission estimates were obtained directly from the plant engineers and account for reductions due to control systems in place at these plants during the time series. These prior estimates are considered CBI and hence are not published (Desai 2010, 2011). These estimates were based on continuous process monitoring equipment installed at the two facilities.

For Plant 4, 1990 through 2009 N_2O emissions were estimated using the following Tier 2 equation from the *2006 IPCC Guidelines*:

$$E_{aa} = Q_{aa} \times EF_{aa} \times (1 - [DF \times UF])$$

where,

E_{aa} = N_2O emissions from adipic acid production, metric tons
Q_{aa} = Quantity of adipic acid produced, metric tons
EF_{aa} = Emission factor, metric ton N_2O/metric ton adipic acid produced
DF = N_2O destruction factor
UF = Abatement system utility factor

The adipic acid production is multiplied by an emission factor (i.e., N_2O emitted per unit of adipic acid produced), which has been estimated, based on experiments that the reaction stoichiometry for N_2O production in the preparation of adipic acid, to be approximately 0.3 metric tons of N_2O per metric ton of product (IPCC 2006). The "N_2O destruction factor" in the equation represents the percentage of N_2O emissions that are destroyed by the installed abatement technology. The "abatement system utility factor" represents the percentage of time that the abatement equipment operates during the annual production period. Plant-specific production data for Plant 4 were obtained across the time series through personal communications (Desai 2010, 2011). The plant-specific production data were then used for calculating emissions as described above.

For Plant 3, 2005 through 2009 emissions were obtained directly from the plant (Desai 2010, 2011). For 1990 through 2004, emissions were estimated using plant-specific production data and the IPCC factors as described above for Plant 4. Plant-level adipic acid production for 1990 through 2003 was estimated by allocating national adipic acid production data to the plant level using the ratio of known plant capacity to total national capacity for all U.S. plants (ACC 2018; CMR 2001, 1998; CW 1999; C&EN 1992 through 1995). For 2004, actual plant production data were obtained and used for emission calculations (CW 2005).

Plant capacities for 1990 through 1994 were obtained from *Chemical & Engineering News*, "Facts and Figures" and "Production of Top 50 Chemicals" (C&EN 1992 through 1995). Plant capacities for 1995 and 1996 were kept the same as 1994 data. The 1997 plant capacities were taken from *Chemical Market Reporter*, "Chemical Profile: Adipic Acid" (CMR 1998). The 1998 plant capacities for all four plants and 1999 plant capacities for three of the plants were obtained from *Chemical Week*, Product Focus: Adipic Acid/Adiponitrile (CW 1999). Plant capacities for the year 2000 for three of the plants were updated using *Chemical Market Reporter*, "Chemical Profile: Adipic Acid" (CMR 2001). For 2001 through 2003, the plant capacities for three plants were held constant at year 2000 capacities. Plant capacity for 1999 to 2003 for the one remaining plant was kept the same as 1998.

National adipic acid production data (see Table 4-31) from 1990 through 2017 were obtained from the American Chemistry Council (ACC 2018).

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[205] Facilities must use standard methods, either EPA Method 320 or ASTM D6348-03, and must follow associated QA/QC procedures during these performance tests consistent with category-specific QC of direct emission measurements.

[206] See <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>.

[207] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf>.

Table 4-31: Adipic Acid Production (kt)

Year	kt
1990	755
2005	865
2013	980
2014	1,025
2015	1,055
2016	860
2017	830

Uncertainty and Time-Series Consistency

Uncertainty associated with N_2O emission estimates includes the methods used by companies to monitor and estimate emissions. While some information has been obtained through outreach with facilities, limited information is available over the time series on these methods, abatement technology destruction and removal efficiency rates and plant-specific production levels.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-32. Nitrous oxide emissions from adipic acid production for 2017 were estimated to be between 7.0 and 7.7 MMT CO_2 Eq. at the 95 percent confidence level. These values indicate a range of approximately 5 percent below to 5 percent above the 2017 emission estimate of 7.4 MMT CO_2 Eq.

Table 4-32: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from Adipic Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		Uncertainty Range Relative to Emission Estimate[a] (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Adipic Acid Production	N_2O	7.4	7.0	7.7	-5%	+5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Data presented in Table 4-31 are for informational purposes only. As previously reported in the Methodology section, adipic acid production data was obtained from EPA's GHGRP and used to estimate emissions between 2010 and 2017. The GHGRP Subpart E adipic acid production data are CBI and therefore not presented in this Inventory report. As a result, those using Table 4-31 values to calculate implied emission factors may occur variable IEFs across the time-series.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

4.9 Caprolactam, Glyoxal and Glyoxylic Acid Production (CRF Source Category 2B4)

Caprolactam

Caprolactam ($C_6H_{11}NO$) is a colorless monomer produced for nylon-6 fibers and plastics, with a substantial proportion of the fiber used in carpet manufacturing. Commercial processes for the manufacture of caprolactam are based on either toluene or benzene. The production of caprolactam can give rise to emissions of nitrous oxide (N_2O).

During the production of caprolactam, emissions of N_2O can occur from the ammonia oxidation step, emissions of carbon dioxide (CO_2) from the ammonium carbonate step, emissions of sulfur dioxide (SO_2) from the ammonium bisulfite step, and emissions of non-methane volatile organic compounds (NMVOCs). Emissions of CO_2, SO_2 and NMVOCs from the conventional process are unlikely to be significant in well-managed plants. Modified caprolactam production processes are primarily concerned with elimination of the high volumes of ammonium sulfate that are produced as a byproduct of the conventional process (IPCC 2006).

Where caprolactam is produced from benzene, the main process, the benzene is hydrogenated to cyclohexane which is then oxidized to produce cyclohexanone ($C_6H_{10}O$). The classical route (Raschig process) and basic reaction equations for production from cyclohexanone are (IPCC 2006):

$$\begin{gathered}
\text{Oxidation of } NH_3 \text{ to } NO/NO_2\\
\downarrow\\
NH_3 \text{ reacted with } CO_2/H_2O \text{ to yield ammonium carbonate } (NH_4)_2CO_3\\
\downarrow\\
(NH_4)_2CO_3 \text{ reacted with } NO/NO_2 \text{ (from } NH_3 \text{ oxidation) to yield ammonium nitrite } (NH_4NO_2)\\
\downarrow\\
NH_3 \text{ reacted with } SO_2/H_2O \text{ to yield ammonium bisulphite } (NH_4HSO_3)\\
\downarrow\\
NH_4NO_2 \text{ and } (NH_4HSO_3) \text{ reacted to yield hydroxylamine disulphonate } (NOH(SO_3NH_4)_2)\\
\downarrow\\
(NOH(SO_3NH_4)_2) \text{ hydrolised to yield hydroxylamine sulphate } ((NH_2OH)_2.H_2SO_4) \text{ and}\\
\text{ammonium sulphate } ((NH_4)_2SO_4)\\
\downarrow\\
\text{Cylohexanone reaction:}\\
C_6H_{10}O\ + \frac{1}{2}(NH_2OH)_2.H_2SO_4(+NH_3 \text{ and } H_2SO_4) \rightarrow C_6H_{10}NOH\ + (NH_4)_2SO_4\ + H_2O\\
\downarrow\\
\text{Beckmann rearrangement:}\\
C_6H_{10}NOH\ \ (+H_2SO_4 \text{ and } SO_2) \rightarrow C_6H_{11}NO.H_2SO_4\ (+4NH_3 \text{ and } H_2O) \rightarrow C_6H_{11}NO\ + 2(NH4)_2SO_4
\end{gathered}$$

In 1999, there were four caprolactam production facilities in the United States. As of 2017, the United States had 3 companies with a total of 3 caprolactam production facilities: AdvanSix in Virginia (AdvanSix 2018), BASF in Texas (BASF 2018), and Fibrant LLC in Georgia (Fibrant 2018; TechSci n.d. 2017).

Nitrous oxide emissions from caprolactam production in the United States were estimated to be 1.4 MMT CO_2 Eq. (5 kt N_2O) in 2017 (see Table 4-33). National emissions from caprolactam production have decreased by approximately 16 percent over the period of 1990 through 2017. Emissions in 2017 have decreased by approximately 30 percent from the 2016 levels.

Table 4-33: N_2O Emissions from Caprolactam Production (MMT CO_2 Eq. and kt N_2O)

Year	MMT CO_2 Eq.	kt N_2O
1990	1.7	6
2005	2.1	7
2013	2.0	7
2014	2.0	7
2015	2.0	7
2016	2.0	7
2017	1.4	5

Glyoxal

Glyoxal is mainly used as a crosslinking agent for acrylic resins, disinfectant, gelatin hardening agent, and textile finishing agent etc. It is produced from oxidation of acetaldehyde with concentrated nitric acid, or from the catalytic oxidation of ethylene glycol, and N_2O is emitted in the process of oxidation of acetaldehyde.

Glyoxal (ethanedial) ($C_2H_2O_2$) is produced from oxidation of acetaldehyde (ethanal) (C_2H_4O) with concentrated nitric acid (HNO_3). Glyoxal can also be produced from catalytic oxidation of ethylene glycol (ethanediol) (CH_2OHCH_2OH). Glyoxal is used as a crosslinking agent for vinyl acetate/acrylic resins, disinfectant, gelatin hardening agent, textile finishing agent (permanent-press cotton, rayon fabrics), wet-resistance additive (paper coatings) (IPCC 2006).

Glyoxylic Acid

Glyoxylic acid is produced by nitric acid oxidation of glyoxal. Glyoxylic acid is used for the production of synthetic aromas, agrochemicals and pharmaceutical intermediates (IPCC 2006).

EPA does not currently estimate the emissions associated with the production of Glyoxal and Glyoxylic Acid due to data availability and a lack of publicly available information on the industry in the United States.

Methodology

Emissions of N_2O were calculated using the estimation methods provided by the *2006 IPCC Guidelines*. The *2006 IPCC Guidelines* Tier 1 method was used to estimate emissions from caprolactam production for 1990 through 2017, as shown in this formula:

$$E_{N_2O} = EF \ x \ CP$$

where,

E_{N2O} = Annual N_2O Emissions (kg)
EF = N_2O emission factor (default) (kg N_2O/metric ton caprolactam produced)
CP = Caprolactam production (metric tons)

During the caprolactam production process, N_2O is generated as a byproduct of the high temperature catalytic oxidation of ammonia (NH_3), which is the first reaction in the series of reactions to produce caprolactam. The amount of N_2O emissions can be estimated based on the chemical reaction shown above. Based on this formula, which is consistent with an IPCC Tier 1 approach, approximately 111.1 metric tons of caprolactam are required to generate one metric ton of N_2O, or an emission factor of 9.0 kg N_2O per metric ton of caprolactam (IPCC 2006). When applying the Tier 1 method, the *2006 IPCC Guidelines* state that it is good practice to assume that there is no abatement of N_2O emissions and to use the highest default emission factor available in the guidelines. In addition, EPA did not find support for the use of secondary catalysts to reduce N_2O emissions, like those employed at nitric acid plants. Thus, the 525 thousand metric tons (kt) of caprolactam produced in 2017 (ACC 2018) resulted in N_2O emissions of approximately 1.4 MMT CO_2 Eq. (5 kt).

The activity data for caprolactam production (see Table 4-34) from 1990 to 2017 were obtained from the ACC *Guide to the Business of Chemistry* report (ACC 2018). EPA will continue to analyze and assess alternative sources of production data as a quality control measure.

Table 4-34: Caprolactam Production (kt)

Year	kt
1990	626
2005	795
2013	750
2014	755
2015	760
2016	755
2017	525

Carbon dioxide and methane (CH_4) emissions may also occur from the production of caprolactam but currently the IPCC does not have methodologies for calculating these emissions associated with caprolactam production.

Uncertainty and Time-Series Consistency

Estimation of emissions of N_2O from caprolactam production can be treated as analogous to estimation of emissions of N_2O from nitric acid production. Both production processes involve an initial step of NH_3 oxidation, which is the source of N_2O formation and emissions (IPCC 2006). Therefore, uncertainties for the default values in the *2006 IPCC Guidelines* is an estimate based on default values for nitric acid plants. In general, default emission factors for gaseous substances have higher uncertainties because mass values for gaseous substances are influenced by temperature and pressure variations and gases are more easily lost through process leaks. The default values for caprolactam production have a relatively high level of uncertainty due to the limited information available (IPCC 2006).

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-35. Nitrous oxide emissions from Caprolactam, Glyoxal and Glyoxylic Acid Production for 2017 were estimated to be between 1.0 and 1.9 MMT CO_2 Eq. at the 95 percent confidence level. These values indicate a range of approximately 31 percent below to 32 percent above the 2017 emission estimate of 1.4 MMT CO_2 Eq.

Table 4-35: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from Caprolactam, Glyoxal and Glyoxylic Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Caprolactam Production	N_2O	1.4	1.0	1.9	-31%	+32%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Pending resources, EPA will research other available datasets for caprolactam production and industry trends, including facility-level data. EPA will also research the production process and emissions associated with the production of glyoxal and glyoxylic acid. During the Expert Review comment period for the current Inventory report, EPA sought expert solicitation on data available for these emissions source categories. EPA did not receive information regarding these industries during Expert Review but will continue to research alternative datasets. This planned improvement is subject to data availability and will be implemented in the medium- to long-term.

4.10 Silicon Carbide Production and Consumption (CRF Source Category 2B5)

Carbon dioxide (CO_2) and methane (CH_4) are emitted from the production of silicon carbide (SiC), a material used as an industrial abrasive. Silicon carbide is produced for abrasive, metallurgical, and other non-abrasive applications in the United States. Production for metallurgical and other non-abrasive applications is not available and therefore both CO_2 and CH_4 estimates are based solely upon production estimates of silicon carbide for abrasive applications. Emissions from fuels consumed for energy purposes during the production of silicon carbide are accounted for in the Energy chapter.

Carbon dioxide and CH_4 are also emitted during the production of calcium carbide, a chemical used to produce acetylene. Carbon dioxide is implicitly accounted for in the storage factor calculation for the non-energy use of petroleum coke in the Energy chapter. However, CH_4 emissions from calcium carbide production are not included as data are not available to apply the Tier 3 methodology prescribed by the *2006 IPCC Guidelines*. EPA is continuing to investigate the inclusion of these emissions in future Inventory reports.

To produce SiC, silica sand or quartz (SiO_2) is reacted with C in the form of petroleum coke. A portion (about 35 percent) of the carbon contained in the petroleum coke is retained in the SiC. The remaining C is emitted as CO_2, CH_4, or carbon monoxide (CO). The overall reaction is shown below (but in practice it does not proceed according to stoichiometry):

$$SiO_2 + 3C \rightarrow SiC + 2CO \, (+ O_2 \rightarrow 2CO_2)$$

Carbon dioxide is also emitted from the consumption of SiC for metallurgical and other non-abrasive applications.

Markets for manufactured abrasives, including SiC, are heavily influenced by activity in the U.S. manufacturing sector, especially in the aerospace, automotive, furniture, housing, and steel manufacturing sectors. The U.S. Geological Survey (USGS) reports that a portion (approximately 50 percent) of SiC is used in metallurgical and other non-abrasive applications, primarily in iron and steel production (USGS 1991a through 2015). As a result of the economic downturn in 2008 and 2009, demand for SiC decreased in those years. Low cost imports, particularly from China, combined with high relative operating costs for domestic producers, continue to put downward pressure on the production of SiC in the United States. However, demand for SiC consumption in the United States has recovered somewhat from its low in 2009 (USGS 1991a through 2015). Abrasive-grade silicon carbide was manufactured at one facility in 2016 in the United States (USGS 2018a).

Carbon dioxide emissions from SiC production and consumption in 2017 were 0.2 MMT CO_2 Eq. (186 kt CO_2) (see Table 4-36 and Table 4-37). Approximately 49 percent of these emissions resulted from SiC production while the remainder resulted from SiC consumption. Methane emissions from SiC production in 2017 were 0.01 MMT CO_2 Eq. (0.4 kt CH_4) (see Table 4-36 and Table 4-37). Emissions have not fluctuated greatly in recent years, but 2017 emissions are about 53 percent lower than emissions in 1990.

Table 4-36: CO_2 and CH_4 Emissions from Silicon Carbide Production and Consumption (MMT CO_2 Eq.)

Year	1990	2005	2013	2014	2015	2016	2017
CO_2	0.4	0.2	0.2	0.2	0.2	0.2	0.2
CH_4	+	+	+	+	+	+	+
Total	**0.4**	**0.2**	**0.2**	**0.2**	**0.2**	**0.2**	**0.2**

+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-37: CO_2 and CH_4 Emissions from Silicon Carbide Production and Consumption (kt)

Year	1990	2005	2013	2014	2015	2016	2017
CO_2	375	219	169	173	180	174	186
CH_4	1	+	+	+	+	+	+

+ Does not exceed 0.5 kt.

Methodology

Emissions of CO_2 and CH_4 from the production of SiC were calculated[208] using the Tier 1 method provided by the *2006 IPCC Guidelines*. Annual estimates of SiC production were multiplied by the appropriate emission factor, as shown below:

$$E_{sc,CO2} = EF_{sc,CO2} \times Q_{sc}$$

$$E_{sc,CH4} = EF_{sc,CH4} \times Q_{sc} \times \left(\frac{1\ metric\ ton}{1000\ kg}\right)$$

where,

$E_{sc,CO2}$ = CO_2 emissions from production of SiC, metric tons
$EF_{sc,CO2}$ = Emission factor for production of SiC, metric ton CO_2/metric ton SiC
Q_{sc} = Quantity of SiC produced, metric tons
$E_{sc,CH4}$ = CH_4 emissions from production of SiC, metric tons
$EF_{sc,CH4}$ = Emission factor for production of SiC, kilogram CH_4/metric ton SiC

Emission factors were taken from the *2006 IPCC Guidelines*:

- 2.62 metric tons CO_2/metric ton SiC
- 11.6 kg CH_4/metric ton SiC

Emissions of CO_2 from silicon carbide consumption for metallurgical uses were calculated by multiplying the annual utilization of SiC for metallurgical uses (reported annually in the USGS *Minerals Yearbook: Silicon*) by the carbon content of SiC (31.5 percent), which was determined according to the molecular weight ratio of SiC.

Emissions of CO_2 from silicon carbide consumption for other non-abrasive uses were calculated by multiplying the annual SiC consumption for non-abrasive uses by the carbon content of SiC (31.5 percent). The annual SiC consumption for non-abrasive uses was calculated by multiplying the annual SiC consumption (production plus net imports) by the percent used in metallurgical and other non-abrasive uses (50 percent) (USGS 1991a through 2015) and then subtracting the SiC consumption for metallurgical use.

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[208] EPA has not integrated aggregated facility-level GHGRP information to inform these estimates. The aggregated information (e.g., activity data and emissions) associated with silicon carbide did not meet criteria to shield underlying confidential business information (CBI) from public disclosure.

The petroleum coke portion of the total CO_2 process emissions from silicon carbide production is adjusted for within the Energy chapter, as these fuels were consumed during non-energy related activities. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

Production data for 1990 through 2013 were obtained from the *Minerals Yearbook: Manufactured Abrasives* (USGS 1991a through 2015). Production data for 2014 through 2016 were obtained from the *Mineral Commodity Summaries: Abrasives (Manufactured)* (USGS 2018). Production data for 2017 were obtained from the *Mineral Industry Surveys: Manufactured Abrasives in the Second Quarter 2018* (USGS 2018b). Silicon carbide production data obtained through the USGS National Minerals Information Center has been previously been rounded to the nearest 5,000 metric tons to avoid disclosing company proprietary data. Silicon carbide consumption for the entire time series is estimated using USGS production data (USGS 1991b through 2015, USGS 2017c) and data from the U.S. International Trade Commission (USITC) database on net imports and exports of silicon carbide provided by the U.S. Census Bureau (2005 through 2018), see Table 4-38.

Table 4-38: Production and Consumption of Silicon Carbide (Metric Tons)

Year	Production	Consumption
1990	105,000	172,465
2005	35,000	220,149
2013	35,000	134,055
2014	35,000	140,733
2015	35,000	153,475
2016	35,000	142,104
2017	35,000	163,492

Uncertainty and Time-Series Consistency

There is uncertainty associated with the emission factors used because they are based on stoichiometry as opposed to monitoring of actual SiC production plants. An alternative would be to calculate emissions based on the quantity of petroleum coke used during the production process rather than on the amount of silicon carbide produced. However, these data were not available. For CH_4, there is also uncertainty associated with the hydrogen-containing volatile compounds in the petroleum coke (IPCC 2006). There is also uncertainty associated with the use or destruction of CH_4 generated from the process in addition to uncertainty associated with levels of production, net imports, consumption levels, and the percent of total consumption that is attributed to metallurgical and other non-abrasive uses.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-39. Silicon carbide production and consumption CO_2 emissions from 2017 were estimated to be between 9 percent below and 9 percent above the emission estimate of 0.19 MMT CO_2 Eq. at the 95 percent confidence level. Silicon carbide production CH_4 emissions were estimated to be between 9 percent below and 9 percent above the emission estimate of 0.01 MMT CO_2 Eq. at the 95 percent confidence level.

Table 4-39: Approach 2 Quantitative Uncertainty Estimates for CH_4 and CO_2 Emissions from Silicon Carbide Production and Consumption (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Silicon Carbide Production and Consumption	CO_2	0.19	0.17	0.20	-9%	+9%
Silicon Carbide Production	CH_4	+	+	+	-9%	+9%

+ Does not exceed 0.05 MMT CO_2 Eq.

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

4.11 Titanium Dioxide Production (CRF Source Category 2B6)

Titanium dioxide (TiO_2) is manufactured using one of two processes: the chloride process and the sulfate process. The chloride process uses petroleum coke and chlorine as raw materials and emits process-related carbon dioxide (CO_2). Emissions from fuels consumed for energy purposes during the production of titanium dioxide are accounted for in the Energy chapter. The chloride process is based on the following chemical reactions:

$$2FeTiO_3 + 7Cl_2 + 3C \rightarrow 2TiCl_4 + 2FeCl_3 + 3CO_2$$

$$2TiCl_4 + 2O_2 \rightarrow 2TiO_2 + 4Cl_2$$

The sulfate process does not use petroleum coke or other forms of carbon as a raw material and does not emit CO_2.

The C in the first chemical reaction is provided by petroleum coke, which is oxidized in the presence of the chlorine and $FeTiO_3$ (rutile ore) to form CO_2. Since 2004, all TiO_2 produced in the United States has been produced using the chloride process, and a special grade of "calcined" petroleum coke is manufactured specifically for this purpose.

The principal use of TiO_2 is as a pigment in white paint, lacquers, and varnishes; it is also used as a pigment in the manufacture of plastics, paper, and other products. In 2017, U.S. TiO_2 production totaled 1,260,000 metric tons (USGS 2018). There were a total five plants producing TiO_2 in the United States in 2017.

Emissions of CO_2 from titanium dioxide production in 2017 were estimated to be 1.7 MMT CO_2 Eq. (1,688 kt CO_2), which represents an increase of 41 percent since 1990 (see Table 4-40). Compared to 2016, emissions from titanium dioxide production increased by 2 percent in 2017 due to a 2 percent increase in production.

Table 4-40: CO_2 Emissions from Titanium Dioxide (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.2	1,195
2005	1.8	1,755
2013	1.7	1,715
2014	1.7	1,688
2015	1.6	1,635
2016	1.7	1,662
2017	1.7	1,688

Methodology

Emissions of CO_2 from TiO_2 production were calculated by multiplying annual national TiO_2 production by chloride process-specific emission factors using a Tier 1 approach provided in *2006 IPCC Guidelines*. The Tier 1 equation is as follows:

$$E_{td} = EF_{td} \times Q_{td}$$

where,

E_{td} = CO_2 emissions from TiO_2 production, metric tons
EF_{td} = Emission factor (chloride process), metric ton CO_2/metric ton TiO_2
Q_{td} = Quantity of TiO_2 produced

The petroleum coke portion of the total CO_2 process emissions from TiO_2 production is adjusted for within the Energy chapter as these fuels were consumed during non-energy related activities. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

Data were obtained for the total amount of TiO_2 produced each year. For years prior to 2004, it was assumed that TiO_2 was produced using the chloride process and the sulfate process in the same ratio as the ratio of the total U.S. production capacity for each process. As of 2004, the last remaining sulfate process plant in the United States closed; therefore, 100 percent of post-2004 production uses the chloride process (USGS 2005). The percentage of production from the chloride process is estimated at 100 percent since 2004. An emission factor of 1.34 metric tons CO_2/metric ton TiO_2 was applied to the estimated chloride-process production (IPCC 2006). It was assumed that all TiO_2 produced using the chloride process was produced using petroleum coke, although some TiO_2 may have been produced with graphite or other carbon inputs.

The emission factor for the TiO_2 chloride process was taken from the *2006 IPCC Guidelines*. Titanium dioxide production data and the percentage of total TiO_2 production capacity that is chloride process for 1990 through 2013 (see Table 4-41) were obtained through the U.S. Geological Survey (USGS) *Minerals Yearbook: Titanium Annual Report* (USGS 1991 through 2015). Production data for 2014 through 2017 were obtained from the *Minerals Commodity Summary: Titanium and Titanium Dioxide* (USGS 2018).[209] Data on the percentage of total TiO_2 production capacity that is chloride process were not available for 1990 through 1993, so data from the 1994 USGS *Minerals Yearbook* were used for these years. Because a sulfate process plant closed in September 2001, the chloride process percentage for 2001 was estimated based on a discussion with Joseph Gambogi (2002). By 2002, only one sulfate process plant remained online in the United States and this plant closed in 2004 (USGS 2005).

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[209] EPA has not integrated aggregated facility-level GHGRP information for Titanium Dioxide production facilities (40 CFR Part 98 Subpart EE). The relevant aggregated information (activity data, emission factor) from these facilities did not meet criteria to shield underlying CBI from public disclosure.

Table 4-41: Titanium Dioxide Production (kt)

Year	kt
1990	979
2005	1,310
2013	1,280
2014	1,260
2015	1,220
2016	1,240
2017	1,260

Uncertainty and Time-Series Consistency

Each year, the USGS collects titanium industry data for titanium mineral and pigment production operations. If TiO_2 pigment plants do not respond, production from the operations is estimated based on prior year production levels and industry trends. Variability in response rates varies from 67 to 100 percent of TiO_2 pigment plants over the time series.

Although some TiO_2 may be produced using graphite or other carbon inputs, information and data regarding these practices were not available. Titanium dioxide produced using graphite inputs, for example, may generate differing amounts of CO_2 per unit of TiO_2 produced as compared to that generated using petroleum coke in production. While the most accurate method to estimate emissions would be to base calculations on the amount of reducing agent used in each process rather than on the amount of TiO_2 produced, sufficient data were not available to do so.

As of 2004, the last remaining sulfate-process plant in the United States closed. Since annual TiO_2 production was not reported by USGS by the type of production process used (chloride or sulfate) prior to 2004 and only the percentage of total production capacity by process was reported, the percent of total TiO_2 production capacity that was attributed to the chloride process was multiplied by total TiO_2 production to estimate the amount of TiO_2 produced using the chloride process. Finally, the emission factor was applied uniformly to all chloride-process production, and no data were available to account for differences in production efficiency among chloride-process plants. In calculating the amount of petroleum coke consumed in chloride-process TiO_2 production, literature data were used for petroleum coke composition. Certain grades of petroleum coke are manufactured specifically for use in the TiO_2 chloride process; however, this composition information was not available.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-42. Titanium dioxide consumption CO_2 emissions from 2017 were estimated to be between 1.5 and 1.9 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 13 percent below and 13 percent above the emission estimate of 1.7 MMT CO_2 Eq.

Table 4-42: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Titanium Dioxide Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Titanium Dioxide Production	CO_2	1.7	1.5	1.9	-13%	+13%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Planned improvements include researching the significance of titanium-slag production in electric furnaces and synthetic-rutile production using the Becher process in the United States. Significant use of these production processes will be included in future Inventory reports. Due to resource constraints, this planned improvement is still in development by EPA and is not included in this report. This is a long-term improvement.

EPA continues to assess the potential of integrating aggregated facility-level GHGRP information for titanium dioxide production facilities based on criteria to shield underlying CBI from public disclosure. Pending available resources, EPA will also evaluate use of GHGRP data to improve category-specific QC consistent with both Volume 1, Chapter 6 of *2006 IPCC Guidelines* and the latest IPCC guidance on the use of facility-level data in national inventories.[210]

4.12 Soda Ash Production (CRF Source Category 2B7)

Carbon dioxide (CO_2) is generated as a byproduct of calcining trona ore to produce soda ash, and is eventually emitted into the atmosphere. In addition, CO_2 may also be released when soda ash is consumed. Emissions from soda ash consumption in chemical production processes are reported under Section 4.4 Other Process Uses of Carbonates (CRF Category 2A4) and emissions from fuels consumed for energy purposes during the production and consumption of soda ash are accounted for in the Energy sector.

Calcining involves placing crushed trona ore into a kiln to convert sodium bicarbonate into crude sodium carbonate that will later be filtered into pure soda ash. The emission of CO_2 during trona-based production is based on the following reaction:

$$2Na_2CO_3 \bullet NaHCO_3 \bullet 2H_2O(Trona) \rightarrow 3Na_2CO_3(Soda\ Ash) + 5H_2O + CO_2$$

Soda ash (sodium carbonate, Na_2CO_3) is a white crystalline solid that is readily soluble in water and strongly alkaline. Commercial soda ash is used as a raw material in a variety of industrial processes and in many familiar consumer products such as glass, soap and detergents, paper, textiles, and food. Emissions from soda ash used in glass production are reported under Section 4.3, Glass Production (CRF Source Category 2A3). Glass production is its own source category and historical soda ash consumption figures have been adjusted to reflect this change. After glass manufacturing, soda ash is used primarily to manufacture many sodium-based inorganic chemicals, including sodium bicarbonate, sodium chromates, sodium phosphates, and sodium silicates (USGS 2015a). Internationally, two types of soda ash are produced, natural and synthetic. The United States produces only natural soda ash and is second only to China in total soda ash production. Trona is the principal ore from which natural soda ash is made.

The United States represents about one-fifth of total world soda ash output (USGS 2018b). Only two states produce natural soda ash: Wyoming and California. Of these two states, only net emissions of CO_2 from Wyoming were calculated due to specifics regarding the production processes employed in the state.[211] Based on 2017 reported

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[210] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[211] In California, soda ash is manufactured using sodium carbonate-bearing brines instead of trona ore. To extract the sodium carbonate, the complex brines are first treated with CO_2 in carbonation towers to convert the sodium carbonate into sodium bicarbonate, which then precipitates from the brine solution. The precipitated sodium bicarbonate is then calcined back into sodium carbonate. Although CO_2 is generated as a byproduct, the CO_2 is recovered and recycled for use in the carbonation stage and is not emitted. A third state, Colorado, produced soda ash until the plant was idled in 2004. The lone producer of sodium

data, the estimated distribution of soda ash by end-use in 2017 (excluding glass production) was chemical production, 57 percent; soap and detergent manufacturing, 12 percent; distributors, 11 percent; flue gas desulfurization, 8 percent; other uses, 7 percent; water treatment, 3 percent, and pulp and paper production, 2 percent (USGS 2018).[212]

U.S. natural soda ash is competitive in world markets because the majority of the world output of soda ash is made synthetically. Although the United States continues to be a major supplier of world soda ash, China, which surpassed the United States in soda ash production in 2003, is the world's leading producer.

In 2017, CO_2 emissions from the production of soda ash from trona were approximately 1.8 MMT CO_2 Eq. (1,753 kt CO_2) (see Table 4-43). Total emissions from soda ash production in 2017 increased by approximately 2 percent from emissions in 2016, and have increased by approximately 22 percent from 1990 levels.

Emissions have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. The U.S. soda ash industry continued a trend of increased production and value in 2017 since experiencing a decline in domestic and export sales caused by adverse global economic conditions in 2009.

Table 4-43: CO_2 Emissions from Soda Ash Production (MMT CO_2 Eq. and kt CO_2)

Year	MMT CO_2 Eq.	kt CO_2
1990	1.4	1,431
2005	1.7	1,655
2013	1.7	1,694
2014	1.7	1,685
2015	1.7	1,714
2016	1.7	1,723
2017	1.8	1,753

Methodology

During the production process, trona ore is calcined in a rotary kiln and chemically transformed into a crude soda ash that requires further processing. Carbon dioxide and water are generated as byproducts of the calcination process. Carbon dioxide emissions from the calcination of trona can be estimated based on the chemical reaction shown above. Based on this formula, which is consistent with an IPCC Tier 1 approach, approximately 10.27 metric tons of trona are required to generate one metric ton of CO_2, or an emission factor of 0.0974 metric tons CO_2 per metric ton trona (IPCC 2006). Thus, the 18.0 million metric tons of trona mined in 2017 for soda ash production (USGS 2018) resulted in CO_2 emissions of approximately 1.8 MMT CO_2 Eq. (1,753 kt).

Once produced, most soda ash is consumed in chemical production, with minor amounts in soap production, pulp and paper, flue gas desulfurization, and water treatment (excluding soda ash consumption for glass manufacturing). As soda ash is consumed for these purposes, additional CO_2 is usually emitted. Consistent with the *2006 IPCC Guidelines for National Greenhouse Gas Inventories*, emissions from soda ash consumption in chemical production processes are reported under Section 4.4 Other Process Uses of Carbonates (CRF Category 2A4).

The activity data for trona production (see Table 4-44) for 1990 to 2017 were obtained from the U.S. Geological Survey (USGS) *Minerals Yearbook for Soda Ash* (1994 through 2015b) and USGS *Mineral Industry Surveys for*

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bicarbonate no longer mines trona in the state. For a brief time, sodium bicarbonate was produced using soda ash feedstocks mined in Wyoming and shipped to Colorado. Prior to 2004, because the trona was mined in Wyoming, the production numbers given by the USGS included the feedstocks mined in Wyoming and shipped to Colorado. In this way, the sodium bicarbonate production that took place in Colorado was accounted for in the Wyoming numbers.

[212] Percentages may not add up to 100 percent due to independent rounding.

Soda Ash (USGS 2017). Soda ash production[213] data were collected by the USGS from voluntary surveys of the U.S. soda ash industry. EPA will continue to analyze and assess opportunities to use facility-level data from EPA's GHGRP to improve the emission estimates for Soda Ash Production source category consistent with IPCC[214] and UNFCCC guidelines.

Table 4-44: Soda Ash Production (kt)

Year	Production[a]
1990	14,700
2005	17,000
2013	17,400
2014	17,300
2015	17,600
2016	17,700
2017	18,000

[a] Soda ash produced from trona ore only.

Uncertainty and Time-Series Consistency

Emission estimates from soda ash production have relatively low associated uncertainty levels in that reliable and accurate data sources are available for the emission factor and activity data for trona-based soda ash production. EPA plans to work with other entities to reassess the uncertainty of these emission factors and activity data based on the most recent information and data. Through EPA's GHGRP, EPA is aware of one facility producing soda ash from a liquid alkaline feedstock process. Soda ash production data was collected by the USGS from voluntary surveys. A survey request was sent to each of the five soda ash producers, all of which responded, representing 100 percent of the total production data (USGS 2016). One source of uncertainty is the purity of the trona ore used for manufacturing soda ash. The emission factor used for this estimate assumes the ore is 100 percent pure, and likely overestimates the emissions from soda ash manufacture. The average water-soluble sodium carbonate-bicarbonate content for ore mined in Wyoming ranges from 85.5 to 93.8 percent (USGS 1995).

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-45. Soda Ash Production CO_2 emissions for 2017 were estimated to be between 1.6 and 1.9 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 9 percent below and 8 percent above the emission estimate of 1.8 MMT CO_2 Eq.

Table 4-45: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Soda Ash Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Soda Ash Production	CO_2	1.8	1.6	1.9	-9%	+8%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

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[213] EPA has assessed feasibility of using emissions information (including activity data) from EPA's GHGRP program; however, at this time, the aggregated information associated with production of soda ash did not meet criteria to shield underlying confidential business information (CBI) from public disclosure.

[214] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

EPA plans to use GHGRP data for conducting category-specific QC of emission estimates consistent with both Volume 1, Chapter 6 of *2006 IPCC Guidelines* and the latest IPCC guidance on the use of facility-level data in national inventories.[215] This planned improvement is ongoing and has not been incorporated into this Inventory report. This is a medium-term planned improvement and expected to be completed by the 2021 Inventory submission.

4.13 Petrochemical Production (CRF Source Category 2B8)

The production of some petrochemicals results in the release of small amounts of carbon dioxide (CO_2) and methane (CH_4) emissions. Petrochemicals are chemicals isolated or derived from petroleum or natural gas. Carbon dioxide emissions from the production of acrylonitrile, carbon black, ethylene, ethylene dichloride, ethylene oxide, and methanol, and CH_4 emissions from the production of methanol and acrylonitrile are presented here and reported under IPCC Source Category 2B8. The petrochemical industry uses primary fossil fuels (i.e., natural gas, coal, petroleum, etc.) for non-fuel purposes in the production of carbon black and other petrochemicals. Emissions from fuels and feedstocks transferred out of the system for use in energy purposes (e.g., indirect or direct process heat or steam production) are currently accounted for in the Energy sector. The allocation and reporting of emissions from feedstocks transferred out of the system for use in energy purposes to the Energy Chapter is consistent with *2006 IPCC Guidelines*.

Worldwide more than 90 percent of acrylonitrile (vinyl cyanide, C_3H_3N) is made by way of direct ammoxidation of propylene with ammonia (NH_3) and oxygen over a catalyst. This process is referred to as the SOHIO process after the Standard Oil Company of Ohio (SOHIO) (IPCC 2006). The primary use of acrylonitrile is as the raw material for the manufacture of acrylic and modacrylic fibers. Other major uses include the production of plastics (acrylonitrile-butadiene-styrene [ABS] and styrene-acrylonitrile [SAN]), nitrile rubbers, nitrile barrier resins, adiponitrile, and acrylamide. All U.S. acrylonitrile facilities use the SOHIO process (AN 2014). The SOHIO process involves a fluidized bed reaction of chemical-grade propylene, ammonia, and oxygen over a catalyst. The process produces acrylonitrile as its primary product and the process yield depends on the type of catalyst used and the process configuration. The ammoxidation process also produces byproduct CO_2, carbon monoxide (CO), and water from the direct oxidation of the propylene feedstock, and produces other hydrocarbons from side reactions in the ammoxidation process.

Carbon black is a black powder generated by the incomplete combustion of an aromatic petroleum- or coal-based feedstock at a high temperature. Most carbon black produced in the United States is added to rubber to impart strength and abrasion resistance, and the tire industry is by far the largest consumer. The other major use of carbon black is as a pigment. The predominant process used in the United States is the furnace black (or oil furnace) process. In the furnace black process, carbon black oil (a heavy aromatic liquid) is continuously injected into the combustion zone of a natural gas-fired furnace. Furnace heat is provided by the natural gas and a portion of the carbon black feedstock; the remaining portion of the carbon black feedstock is pyrolyzed to carbon black. The resultant CO_2 and uncombusted CH_4 emissions are released from thermal incinerators used as control devices, process dryers, and equipment leaks. Carbon black is also produced in the United States by the thermal cracking of

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[215] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

acetylene-containing feedstocks (i.e., acetylene black process), by the thermal cracking of other hydrocarbons (i.e., thermal black process), and by the open burning of carbon black feedstock (i.e., lamp black process); each of these processes is used at only one U.S. plant (EPA 2000).

Ethylene (C_2H_4) is consumed in the production processes of the plastics industry including polymers such as high, low, and linear low density polyethylene (HDPE, LDPE, LLDPE); polyvinyl chloride (PVC); ethylene dichloride; ethylene oxide; and ethylbenzene. Virtually all ethylene is produced from steam cracking of ethane, propane, butane, naphtha, gas oil, and other feedstocks. The representative chemical equation for steam cracking of ethane to ethylene is shown below:

$$C_2H_6 \rightarrow C_2H_4 + H_2$$

Small amounts of CH_4 are also generated from the steam cracking process. In addition, CO_2 and CH_4 emissions are also generated from combustion units.

Ethylene dichloride ($C_2H_4Cl_2$) is used to produce vinyl chloride monomer, which is the precursor to polyvinyl chloride (PVC). Ethylene dichloride was used as a fuel additive until 1996 when leaded gasoline was phased out. Ethylene dichloride is produced from ethylene by either direct chlorination, oxychlorination, or a combination of the two processes (i.e., the “balanced process”); most U.S. facilities use the balanced process. The direct chlorination and oxychlorination reactions are shown below:

$$C_2H_4 + Cl_2 \rightarrow C_2H_4Cl_2 \text{ (direct chlorination)}$$

$$C_2H_4 + \tfrac{1}{2}O_2 + 2HCl \rightarrow C_2H_4Cl_2 + 2H_2O \text{ (oxychlorination)}$$

$$C_2H_4 + 3O_2 \rightarrow 2CO_2 + 2H_2O \text{ (direct oxidation of ethylene during oxychlorination)}$$

In addition to the byproduct CO_2 produced from the direction oxidation of the ethylene feedstock, CO_2 and CH_4 emissions are also generated from combustion units.

Ethylene oxide (C_2H_4O) is used in the manufacture of glycols, glycol ethers, alcohols, and amines. Approximately 70 percent of ethylene oxide produced worldwide is used in the manufacture of glycols, including monoethylene glycol. Ethylene oxide is produced by reacting ethylene with oxygen over a catalyst. The oxygen may be supplied to the process through either an air (air process) or a pure oxygen stream (oxygen process). The byproduct CO_2 from the direct oxidation of the ethylene feedstock is removed from the process vent stream using a recycled carbonate solution, and the recovered CO_2 may be vented to the atmosphere or recovered for further utilization in other sectors, such as food production (IPCC 2006). The combined ethylene oxide reaction and byproduct CO_2 reaction is exothermic and generates heat, which is recovered to produce steam for the process. The ethylene oxide process also produces other liquid and off-gas byproducts (e.g., ethane, etc.) that may be burned for energy recovery within the process. Almost all facilities, except one in Texas, use the oxygen process to manufacture ethylene oxide (EPA 2008).

Methanol (CH_3OH) is a chemical feedstock most often converted into formaldehyde, acetic acid and olefins. It is also an alternative transportation fuel, as well as an additive used by municipal wastewater treatment facilities in the denitrification of wastewater. Methanol is most commonly synthesized from a synthesis gas (i.e., “syngas” – a mixture containing H_2, CO, and CO_2) using a heterogeneous catalyst. There are a number of process techniques that can be used to produce syngas. Worldwide, steam reforming of natural gas is the most common method; most methanol producers in the United States also use steam reforming of natural gas to produce syngas. Other syngas production processes in the United States include partial oxidation of natural gas and coal gasification.

Emissions of CO_2 and CH_4 from petrochemical production in 2017 were 28.2 MMT CO_2 Eq. (28,225 kt CO_2) and 0.3 MMT CO_2 Eq. (10 kt CH_4), respectively (see Table 4-46 and Table 4-47). Since 1990, total CO_2 emissions from petrochemical production increased by 33 percent. Methane emissions from petrochemical (methanol and acrylonitrile) production reached a low of 1.8 kt CH_4 in 2011, given declining methanol production; however, CH_4 emissions have been increasing every year since 2011 and are now 14 percent greater than in 1990 (though still less than the peak in 1997) due to a rebound in methanol production.

Table 4-46: CO_2 and CH_4 Emissions from Petrochemical Production (MMT CO_2 Eq.)

Year	1990		2005		2013	2014	2015	2016	2017
CO_2	21.2		26.8		26.4	26.5	28.1	28.1	28.2

CH_4	0.2		0.1		0.1	0.1	0.2	0.2	0.3
Total	**21.4**		**26.9**		**26.5**	**26.6**	**28.2**	**28.4**	**28.5**

Table 4-47: CO_2 and CH_4 Emissions from Petrochemical Production (kt)

Year	**1990**		**2005**		**2013**	**2014**	**2015**	**2016**	**2017**
CO_2	21,222		26,810		26,395	26,496	28,062	28,110	28,225
CH_4	9		3		3	5	7	10	10

Methodology

Emissions of CO_2 and CH_4 were calculated using the estimation methods provided by the *2006 IPCC Guidelines* and country-specific methods from EPA's GHGRP. The *2006 IPCC Guidelines* Tier 1 method was used to estimate CO_2 and CH_4 emissions from production of acrylonitrile and methanol,[216] and a country-specific approach similar to the IPCC Tier 2 method was used to estimate CO_2 emissions from production of carbon black, ethylene oxide, and ethylene dichloride. The Tier 2 method for petrochemicals is a total feedstock C mass balance method used to estimate total CO_2 emissions, but is not applicable for estimating CH_4 emissions.

As noted in the *2006 IPCC Guidelines*, the total feedstock C mass balance method (Tier 2) is based on the assumption that all of the C input to the process is converted either into primary and secondary products or into CO_2. Further, the guideline states that while the total C mass balance method estimates total C emissions from the process but does not directly provide an estimate of the amount of the total C emissions emitted as CO_2, CH_4, or non-CH_4 volatile organic compounds (NMVOCs). This method accounts for all the C as CO_2, including CH_4.

Note, a small subset of facilities reporting under EPA's GHGRP use Continuous Emission Monitoring Systems (CEMS) to monitor CO_2 emissions, and these facilities are required to also report CH_4 and N_2O emissions from combustion of process off-gas in flares. Preliminary analysis of aggregated annual reports shows that these flared CH_4 and N_2O emissions are less than 500 kt/year. EPA's GHGRP is still reviewing this data across reported years to facilitate update of category-specific QC documentation and EPA plans to address this more completely in future reports.

Carbon Black, Ethylene, Ethylene Dichloride and Ethylene Oxide

2010 through 2017

Carbon dioxide emissions and national production were aggregated directly from EPA's GHGRP dataset for 2010 through 2016 (EPA 2017). The GHGRP data for 2016 were also used as a proxy for 2017 because the 2017 data were unavailable prior to preparation of this report. However, preliminary analysis of GHGRP Subpart X shows that emissions in 2017 were consistent, albeit slightly higher, with those reported in 2016. In 2016, data reported to the GHGRP included CO_2 emissions of 3,160,000 metric tons from carbon black production; 19,600,000 metric tons of CO_2 from ethylene production; 447,000 metric tons of CO_2 from ethylene dichloride production; and 1,100,000 metric tons of CO_2 from ethylene oxide production. These emissions reflect application of a country-specific approach similar to the IPCC Tier 2 method and were used to estimate CO_2 emissions from the production of carbon black, ethylene, ethylene dichloride, and ethylene oxide.

Since 2010, EPA's GHGRP, under Subpart X, requires all domestic producers of petrochemicals to report annual emissions and supplemental emissions information (e.g., production data, etc.) to facilitate verification of reported emissions. Under EPA's GHGRP, most petrochemical production facilities are required to use either a mass balance approach or CEMS to measure and report emissions for each petrochemical process unit to estimate facility-level

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[216] EPA has not integrated aggregated facility-level GHGRP information for acrylonitrile and methanol production. The aggregated information associated with production of these petrochemicals did not meet criteria to shield underlying CBI from public disclosure.

process CO_2 emissions; ethylene production facilities also have a third option. The mass balance method is used by most facilities[217] and assumes that all the carbon input is converted into primary and secondary products, byproducts, or is emitted to the atmosphere as CO_2. To apply the mass balance, facilities must measure the volume or mass of each gaseous and liquid feedstock and product, mass rate of each solid feedstock and product, and carbon content of each feedstock and product for each process unit and sum for their facility. To apply the optional combustion methodology, ethylene production facilities must measure the quantity, carbon content, and molecular weight of the fuel to a stationary combustion unit when that fuel includes any ethylene process off-gas. These data are used to calculate the total CO_2 emissions from the combustion unit. The facility must also estimate the fraction of the emissions that is attributable to burning the ethylene process off-gas portion of the fuel. This fraction is multiplied by the total emissions to estimate the emissions from ethylene production.

More details on the greenhouse gas calculation, monitoring and QA/QC methods applicable to petrochemical facilities can be found under Subpart X (Petrochemical Production) of the regulation (40 CFR Part 98).[218] EPA verifies annual facility-level GHGRP reports through a multi-step process (e.g., combination of electronic checks and manual reviews) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent.[219]

All non-energy uses of residual fuel and some non-energy uses of "other oil" are assumed to be used in the production of carbon black; therefore, consumption of these fuels is adjusted for within the Energy chapter to avoid double-counting of emissions from fuel used in the carbon black production presented here within IPPU sector. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (IPCC Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

1990 through 2009

Prior to 2010, for each of these 4 types of petrochemical processes, an average national CO_2 emission factor was calculated based on the GHGRP data and applied to production for earlier years in the time series (i.e., 1990 through 2009) to estimate CO_2 emissions from carbon black, ethylene, ethylene dichloride, and ethylene oxide production. For carbon black, ethylene, and ethylene dichloride, carbon dioxide emission factors were derived from EPA's GHGRP data by dividing annual CO_2 emissions for petrochemical type "i" with annual production for petrochemical type "i" and then averaging the derived emission factors obtained for each calendar year 2010 through 2016. For ethylene oxide, the carbon dioxide emission factor was derived in the same manner, except that only data from calendar years 2010 through 2013 were used to develop the average emission factor because process improvements in recent years have resulted in lower CO_2 emissions that are not representative of operation in 1990 through 2009. The average emission factors for each petrochemical type were applied across all prior years because petrochemical production processes in the United States have not changed significantly since 1990, though some operational efficiencies have been implemented at facilities over the time series.

The average country-specific CO_2 emission factors that were calculated from the GHGRP data are as follows:

- 2.63 metric tons CO_2/metric ton carbon black produced

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[217] A few facilities producing ethylene dichloride used CO_2 CEMS, those CO_2 emissions have been included in the aggregated GHGRP emissions presented here. For ethylene production processes, nearly all process emissions are from the combustion of process off-gas. Under EPA's GHGRP, Subpart X, ethylene facilities can report CO_2 emissions from burning of process gases using the optional combustion methodology for ethylene production processes, which requires estimating emissions based on fuel quantity and carbon contents of the fuel. This is consistent with the *2006 IPCC Guidelines* (p. 3.57) which recommends including combustion emissions from fuels obtained from feedstocks (e.g., off-gases) in petrochemical production under in the IPPU sector. In 2014, for example, this methodology was used by more than 20 of the 65 reporting facilities. In addition to CO_2, these facilities are required to report emissions of CH_4 and N_2O from combustion of ethylene process off-gas in both stationary combustion units and flares. Facilities using CEMS (consistent with a Tier 3 approach) are also required to report emissions of CH_4 and N_2O from combustion of petrochemical process-off gases in flares. Preliminary analysis of the aggregated reported CH_4 and N_2O emissions from facilities using CEMS and N_2O emissions from facilities using the optional combustion methodology suggests that these annual emissions are less than 500 kt/yr so not significant enough to prioritize for inclusion in the report at this time. Pending resources and significance, EPA may include these emissions in future reports to enhance completeness.

[218] See <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>.

[219] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf>.

- 0.77 metric tons CO_2/metric ton ethylene produced
- 0.041 metric tons CO_2/metric ton ethylene dichloride produced
- 0.46 metric tons CO_2/metric ton ethylene oxide produced

Annual production data for carbon black for 1990 through 2009 were obtained from the International Carbon Black Association (Johnson 2003 and 2005 through 2010). Annual production data for ethylene and ethylene dichloride for 1990 through 2009 were obtained from the American Chemistry Council's (ACC's) *Guide to the Business of Chemistry* (ACC 2002, 2003, 2005 through 2011). Annual production data for ethylene oxide were obtained from ACC's *U.S. Chemical Industry Statistical Handbook* for 2003 through 2009 (ACC 2014a) and from ACC's *Business of Chemistry* for 1990 through 2002 (ACC 2014b). As noted above, annual 2010 through 2016 production data for carbon black, ethylene, ethylene dichloride, and ethylene oxide, were obtained from EPA's GHGRP, and data from 2016 were used as a proxy for 2017.

Acrylonitrile

Carbon dioxide and methane emissions from acrylonitrile production were estimated using the Tier 1 method in the *2006 IPCC Guidelines*. Annual acrylonitrile production data were used with IPCC default Tier 1 CO_2 and CH_4 emission factors to estimate emissions for 1990 through 2017. Emission factors used to estimate acrylonitrile production emissions are as follows:

- 0.18 kg CH_4/metric ton acrylonitrile produced
- 1.00 metric tons CO_2/metric ton acrylonitrile produced

Annual acrylonitrile production data for 1990 through 2017 were obtained from ACC's *Business of Chemistry* (ACC 2018).

Methanol

Carbon dioxide and methane emissions from methanol production were estimated using the Tier 1 method in the *2006 IPCC Guidelines*. Annual methanol production data were used with IPCC default Tier 1 CO_2 and CH_4 emission factors to estimate emissions for 1990 through 2017. Emission factors used to estimate methanol production emissions are as follows:

- 2.3 kg CH_4/metric ton methanol produced
- 0.67 metric tons CO_2/metric ton methanol produced

Annual methanol production data for 1990 through 2017 were obtained from the ACC's *Business of Chemistry* (ACC 2018).

Table 4-48: Production of Selected Petrochemicals (kt)

Chemical	1990	2005	2013	2014	2015	2016	2017
Carbon Black	1,307	1,651	1,230	1,210	1,220	1,190	1,190
Ethylene	16,542	23,975	25,300	25,500	26,900	26,600	26,600
Ethylene Dichloride	6,283	11,260	11,500	11,300	11,300	11,700	11,700
Ethylene Oxide	2,429	3,220	3,150	3,140	3,240	3,210	3,210
Acrylonitrile	1,214	1,325	1,075	1,095	1,050	955	1,040
Methanol	3,750	1,225	1,235	2,105	3,065	4,250	4,295

As noted earlier in the introduction section of the Petrochemical Production chapter, the allocation and reporting of emissions from both fuels and feedstocks transferred out of the system for use in energy purposes to the Energy Chapter differs slightly from the *2006 IPCC Guidelines*. According to the *2006 IPCC Guidelines*, emissions from fuel combustion from petrochemical production should be allocated to this source category within the IPPU Chapter. Due to national circumstances, EIA data on primary fuel for feedstock use within the energy balance are presented by commodity only, with no resolution on data by industry sector (i.e. petrochemical production). In addition, under EPA's GHGRP, reporting facilities began reporting in 2014 on annual feedstock quantities for mass balance and CEMS methodologies (79 FR 63794), as well as the annual average carbon content of each feedstock (and molecular weight for gaseous feedstocks) for the mass balance methodology beginning in reporting year 2017 (81 FR 89260).[220] The United States is currently unable to report non-energy fuel use from petrochemical production under the IPPU chapter due to CBI issues. Therefore, consistent with *2006 IPCC Guidelines*, fuel consumption data reported by EIA are modified to account for these overlaps to avoid double-counting. More information on the non-energy use of fossil fuel feedstocks for petrochemical production can be found in Annex 2.3.

Uncertainty and Time-Series Consistency

The CH_4 and CO_2 emission factors used for acrylonitrile and methanol production are based on a limited number of studies. Using plant-specific factors instead of default or average factors could increase the accuracy of the emission estimates; however, such data were not available for the current Inventory report.

The results of the quantitative uncertainty analysis for the CO_2 emissions from carbon black production, ethylene, ethylene dichloride, and ethylene oxide are based on reported GHGRP data. Refer to the Methodology section for more details on how these emissions were calculated and reported to EPA's GHGRP. There is some uncertainty in the applicability of the average emission factors for each petrochemical type across all prior years. While petrochemical production processes in the United States have not changed significantly since 1990, some operational efficiencies have been implemented at facilities over the time series.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-49. Petrochemical production CO_2 emissions from 2017 were estimated to be between 26.7 and 29.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the emission estimate of 28.2 MMT CO_2 Eq. Petrochemical production CH_4 emissions from 2017 were estimated to be between 0.09 and 0.31 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 57 percent below to 45 percent above the emission estimate of 0.3 MMT CO_2 Eq.

Table 4-49: Approach 2 Quantitative Uncertainty Estimates for CH_4 Emissions from Petrochemical Production and CO_2 Emissions from Petrochemical Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Petrochemical Production	CO_2	28.2	26.7	29.7	-5%	+5%
Petrochemical Production	CH_4	0.34	0.09	0.31	-57%	+45%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see QA/QC and Verification Procedures section in the introduction of the IPPU Chapter.

QA/QC and Verification

For Petrochemical Production, QA/QC activities were conducted consistent with the U.S. Inventory QA/QC plan, as described in the QA/QC and Verification Procedures section of the IPPU Chapter and Annex 8. Source-specific

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[220] See <https://www.epa.gov/ghgreporting/historical-rulemakings>.

quality control measures for this category included the QA/QC requirements and verification procedures of EPA's GHGRP.

For ethylene, ethylene dichloride, and ethylene oxide it is possible to compare CO_2 emissions calculated using the GHGRP data to the CO_2 emissions that would have been calculated using the Tier 1 approach if GHGRP data were not available. For ethylene, the GHGRP emissions typically are within 5 percent of the emissions calculated using the Tier 1 approach (except for 2010 when the difference was 11 percent). For ethylene dichloride, the GHGRP emissions are typically within 20 percent of the Tier 1 emissions (except in 2014 due to incorrect GHGRP emissions that were not corrected before the most recent publication of the data). For ethylene oxide, GHGRP emissions vary from 10 percent less than the Tier 1 emissions to 30 percent more than the Tier 1 emissions, depending on the year.

As part of a planned improvement effort, EPA has assessed the potential of using GHGRP data to estimate CH_4 emissions from ethylene production. As discussed in the Methodology section above, CO_2 emissions from ethylene production in this chapter are based on data reported under the GHGRP, and these emissions are calculated using a Tier 2 approach that assumes all of the carbon in the fuel (i.e., ethylene process off-gas) is converted to CO_2. Ethylene production facilities also calculate and report CH_4 emissions under the GHGRP when they use the optional combustion methodology. The facilities calculate CH_4 emissions from each combustion unit that burns off-gas from an ethylene production process unit using a Tier 1 approach based on the total quantity of fuel burned, a default higher heating value, and a default emission factor. Because multiple other types of fuel in addition to the ethylene process unit off-gas may be burned in these combustion units, the facilities also report an estimate of the fraction of emissions that is due to burning the ethylene process off-gas component of the total fuel. Multiplying the total emissions by the estimated fraction provides an estimate of the CH_4 emissions from the ethylene production process unit. These ethylene production facilities also calculate CH_4 emissions from flares that burn process vent emissions from ethylene processes. The emissions are calculated using either a Tier 2 approach based on measured gas volumes and measured carbon content or higher heating value, or a Tier 1 approach based on the measured gas flow and a default emission factor. Nearly all ethylene production facilities use the optional combustion methodology under the GHGRP, and the sum of reported emissions from combustion in stationary combustion units and flares at all of these facilities is on the same order of magnitude as the combined CH_4 emissions presented in this chapter from methanol and acrylonitrile production. The CH_4 emissions from ethylene production under the GHGRP have not been included in this chapter because this approach double counts carbon (i.e., all of the carbon in the CH_4 emissions is also included in the CO_2 emissions from the ethylene process units). EPA continues to assess the GHGRP data for ways to better disaggregate the data and incorporate it into the inventory.

Future QC efforts to validate the use of Tier 1 default EFs and report on the comparison of Tier 1 emissions estimates and GHGRP data are described below in the Planned Improvements section.

Recalculations Discussion

As previously noted above, GHGRP data are used to develop CO_2 emission factors for carbon black, ethylene, ethylene dichloride, and ethylene oxide production. These factors are used with production data to estimate CO_2 emissions from production of these petrochemicals in 1990 through 2009. In previous versions of the Inventory, average emission factors were developed from all years of available GHGRP data. However, in recent years, the emission factor for ethylene oxide has been steadily declining as a result of process efficiencies being implemented through the industry; thus, in an effort to better characterize the emissions from 1990 through 2009, the emissions factor for ethylene oxide in this year's Inventory is based on the GHGRP data only from 2010 through 2013. The emission factor calculated using only these 4 years of data is 11 percent higher than the emission factor using all data from 2010 through 2016. Thus, estimated CO_2 emissions from ethylene oxide production in 1990 through 2009 are about 11 percent higher in the current Inventory (i.e., 1990 through 2017) than the previous Inventory (i.e., 1990 through 2016).

Planned Improvements

Improvements include completing category-specific QC of activity data and emission factors, along with further assessment of CH_4 and N_2O emissions to enhance completeness in reporting of emissions from U.S. petrochemical production, pending resources, significance and time-series consistency considerations. For example, EPA is planning additional assessment of ways to use CH_4 data from the GHGRP in the inventory. One possible approach

EPA is assessing would be to adjust the CO_2 emissions from the GHGRP downward by subtracting the carbon that is also included in the reported CH_4 emissions, per the discussion in the Petrochemical Production QA/QC and Verification section, above. As of this current report, timing and resources have not allowed EPA to complete this analysis of activity data, emissions, and emission factors and remains a priority improvement within the IPPU chapter.

Pending resources, a secondary potential improvement for this source category would focus on continuing to analyze the fuel and feedstock data from EPA's GHGRP to better disaggregate energy-related emissions and allocate them more accurately between the Energy and IPPU sectors of the Inventory. Some degree of double counting may occur between CO_2 estimates of non-energy use of fuels in the energy sector and CO_2 process emissions from petrochemical production in this sector. As noted previously in the methodology section, data integration is not feasible at this time as feedstock data from the EIA used to estimate non-energy uses of fuels are aggregated by fuel type, rather than disaggregated by both fuel type and particular industries. As described in the methodology section of this source category, EPA is currently unable to use GHGRP reported data on quantities of fuel consumed as feedstocks by petrochemical producers, only feedstock type, due to the data failing GHGRP CBI aggregation criteria. Incorporating this data into future inventories will allow for easier data integration between the non-energy uses of fuels category and the petrochemicals category presented in this chapter. This planned improvement is still under development and has not been completed to report on progress in this current Inventory.

4.14 HCFC-22 Production (CRF Source Category 2B9a)

Trifluoromethane (HFC-23 or CHF_3) is generated as a byproduct during the manufacture of chlorodifluoromethane (HCFC-22), which is primarily employed in refrigeration and air conditioning systems and as a chemical feedstock for manufacturing synthetic polymers. Between 1990 and 2000, U.S. production of HCFC-22 increased significantly as HCFC-22 replaced chlorofluorocarbons (CFCs) in many applications. Between 2000 and 2007, U.S. production fluctuated but generally remained above 1990 levels. In 2008 and 2009, U.S. production declined markedly and has remained near 2009 levels since. Because HCFC-22 depletes stratospheric ozone, its production for non-feedstock uses is scheduled to be phased out by 2020 under the U.S. Clean Air Act.[221] Feedstock production, however, is permitted to continue indefinitely.

HCFC-22 is produced by the reaction of chloroform ($CHCl_3$) and hydrogen fluoride (HF) in the presence of a catalyst, $SbCl_5$. The reaction of the catalyst and HF produces $SbCl_xF_y$, (where $x + y = 5$), which reacts with chlorinated hydrocarbons to replace chlorine atoms with fluorine. The HF and chloroform are introduced by submerged piping into a continuous-flow reactor that contains the catalyst in a hydrocarbon mixture of chloroform and partially fluorinated intermediates. The vapors leaving the reactor contain HCFC-21 ($CHCl_2F$), HCFC-22 ($CHClF_2$), HFC-23 (CHF_3), HCl, chloroform, and HF. The under-fluorinated intermediates (HCFC-21) and chloroform are then condensed and returned to the reactor, along with residual catalyst, to undergo further fluorination. The final vapors leaving the condenser are primarily HCFC-22, HFC-23, HCl and residual HF. The HCl is recovered as a useful byproduct, and the HF is removed. Once separated from HCFC-22, the HFC-23 may be released to the atmosphere, recaptured for use in a limited number of applications, or destroyed.

Two facilities produced HCFC-22 in the United States in 2017. Emissions of HFC-23 from this activity in 2017 were estimated to be 5.2 MMT CO_2 Eq. (0.3 kt) (see Table 4-50). This quantity represents an 85 percent increase from 2015 emissions and an 89 percent decrease from 1990 emissions. The decrease from 1990 emissions was caused primarily by changes in the HFC-23 emission rate (kg HFC-23 emitted/kg HCFC-22 produced). An uptick in this rate, as well as in the quantity of HCFC-22 produced, was responsible for the increase in HFC-23 emissions between 2016 and 2017. The long-term decrease in the emission rate is primarily attributable to six factors: (a) five plants that did not capture and destroy the HFC-23 generated have ceased production of HCFC-22 since 1990; (b)

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[221] As construed, interpreted, and applied in the terms and conditions of the Montreal Protocol on Substances that Deplete the Ozone Layer. [42 U.S.C. §7671m(b), CAA §614]

one plant that captures and destroys the HFC-23 generated began to produce HCFC-22; (c) one plant implemented and documented a process change that reduced the amount of HFC-23 generated; (d) the same plant began recovering HFC-23, primarily for destruction and secondarily for sale; (e) another plant began destroying HFC-23; and (f) the same plant, whose emission factor was higher than that of the other two plants, ceased production of HCFC-22 in 2013.

Table 4-50: HFC-23 Emissions from HCFC-22 Production (MMT CO_2 Eq. and kt HFC-23)

Year	MMT CO_2 Eq.	kt HFC-23
1990	46.1	3
2005	20.0	1
2013	4.1	0.3
2014	5.0	0.3
2015	4.3	0.3
2016	2.8	0.2
2017	5.2	0.3

Methodology

To estimate HFC-23 emissions for five of the eight HCFC-22 plants that have operated in the United States since 1990, methods comparable to the Tier 3 methods in the *2006 IPCC Guidelines* (IPCC 2006) were used. Emissions for 2010 through 2017 were obtained through reports submitted by U.S. HCFC-22 production facilities to EPA's Greenhouse Gas Reporting Program (GHGRP). EPA's GHGRP mandates that all HCFC-22 production facilities report their annual emissions of HFC-23 from HCFC-22 production processes and HFC-23 destruction processes. Previously, data were obtained by EPA through collaboration with an industry association that received voluntarily reported HCFC-22 production and HFC-23 emissions annually from all U.S. HCFC-22 producers from 1990 through 2009. These emissions were aggregated and reported to EPA on an annual basis.

For the other three plants, the last of which closed in 1993, methods comparable to the Tier 1 method in the *2006 IPCC Guidelines* were used. Emissions from these three plants have been calculated using the recommended emission factor for unoptimized plants operating before 1995 (0.04 kg HCFC-23/kg HCFC-22 produced).

The five plants that have operated since 1994 measure (or, for the plants that have since closed, measured) concentrations of HFC-23 to estimate their emissions of HFC-23. Plants using thermal oxidation to abate their HFC-23 emissions monitor the performance of their oxidizers to verify that the HFC-23 is almost completely destroyed. Plants that release (or historically have released) some of their byproduct HFC-23 periodically measure HFC-23 concentrations in the output stream using gas chromatography. This information is combined with information on quantities of products (e.g., HCFC-22) to estimate HFC-23 emissions.

To estimate 1990 through 2009 emissions, reports from an industry association were used that aggregated HCFC-22 production and HFC-23 emissions from all U.S. HCFC-22 producers and reported them to EPA (ARAP 1997, 1999, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, and 2010). To estimate 2010 through 2017 emissions, facility-level data (including both HCFC-22 production and HFC-23 emissions) reported through EPA's GHGRP were analyzed. In 1997 and 2008, comprehensive reviews of plant-level estimates of HFC-23 emissions and HCFC-22 production were performed (RTI 1997; RTI 2008). The 1997 and 2008 reviews enabled U.S. totals to be reviewed, updated, and where necessary, corrected, and also for plant-level uncertainty analyses (Monte-Carlo simulations) to be performed for 1990, 1995, 2000, 2005, and 2006. Estimates of annual U.S. HCFC-22 production are presented in Table 4-51.

Table 4-51: HCFC-22 Production (kt)

Year	kt
1990	139
2005	156

2012	96
2013	C
2014	C
2015	C
2016	C
2017	C

C (CBI)
Note: HCFC-22 production in 2013 through 2017 is considered Confidential Business Information (CBI) as there were only two producers of HCFC-22 in those years.

Uncertainty and Time-Series Consistency

The uncertainty analysis presented in this section was based on a plant-level Monte Carlo Stochastic Simulation for 2006. The Monte Carlo analysis used estimates of the uncertainties in the individual variables in each plant's estimating procedure. This analysis was based on the generation of 10,000 random samples of model inputs from the probability density functions for each input. A normal probability density function was assumed for all measurements and biases except the equipment leak estimates for one plant; a log-normal probability density function was used for this plant's equipment leak estimates. The simulation for 2006 yielded a 95-percent confidence interval for U.S. emissions of 6.8 percent below to 9.6 percent above the reported total.

The relative errors yielded by the Monte Carlo Stochastic Simulation for 2006 were applied to the U.S. emission estimate for 2017. The resulting estimates of absolute uncertainty are likely to be reasonably accurate because (1) the methods used by the two remaining plants to estimate their emissions are not believed to have changed significantly since 2006, and (2) although the distribution of emissions among the plants has changed between 2006 and 2017 (because one plant has closed), the plant that currently accounts for most emissions had a relative uncertainty in its 2006 (as well as 2005) emissions estimate that was similar to the relative uncertainty for total U.S. emissions. Thus, the closure of one plant is not likely to have a large impact on the uncertainty of the national emission estimate.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-52. HFC-23 emissions from HCFC-22 production were estimated to be between 4.8 and 5.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below and 10 percent above the emission estimate of 5.2 MMT CO_2 Eq.

Table 4-52: Approach 2 Quantitative Uncertainty Estimates for HFC-23 Emissions from HCFC-22 Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
HCFC-22 Production	HFC-23	5.2	4.8	5.7	-7%	+10%

[a] Range of emissions reflects a 95 percent confidence interval.

QA/QC and Verification

General (Tier 1) and category-specific (Tier 2) QA/QC activities were conducted consistent with the U.S. Inventory QA/QC plan, as described in Annex 8. Source-specific quality control measures for the HCFC-22 Production category included the QA/QC requirements and verification procedures of EPA's GHGRP. Under EPA's GHGRP, HCFC-22 producers are required to (1) measure concentrations of HFC-23 and HCFC-22 in the product stream at least weekly using equipment and methods (e.g., gas chromatography) with an accuracy and precision of 5 percent or better at the concentrations of the process samples, (2) measure mass flows of HFC-23 and HCFC-22 at least weekly using measurement devices (e.g., flowmeters) with an accuracy and precision of 1 percent of full scale or better, (3) calibrate mass measurement devices at the frequency recommended by the manufacturer using traceable standards and suitable methods published by a consensus standards organization, (4) calibrate gas chromatographs at least monthly through analysis of certified standards, and (5) document these calibrations.

EPA verifies annual facility-level reports from HCFC-22 producers through a multi-step process (e.g., a combination of electronic checks and manual reviews by staff) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent. Based on the results of the verification process, EPA follows up with facilities to resolve mistakes that may have occurred.[222]

4.15 Carbon Dioxide Consumption (CRF Source Category 2B10)

Carbon dioxide (CO_2) is used for a variety of commercial applications, including food processing, chemical production, carbonated beverage production, and refrigeration, and is also used in petroleum production for enhanced oil recovery (EOR). Carbon dioxide used for EOR is injected underground to enable additional petroleum to be produced. For the purposes of this analysis, CO_2 used in commercial applications other than EOR is assumed to be emitted to the atmosphere. Carbon dioxide used in EOR applications is discussed in the Energy chapter under "Carbon Capture and Storage, including Enhanced Oil Recovery" and is not discussed in this section.

Carbon dioxide is produced from naturally-occurring CO_2 reservoirs, as a byproduct from the energy and industrial production processes (e.g., ammonia production, fossil fuel combustion, ethanol production), and as a byproduct from the production of crude oil and natural gas, which contain naturally occurring CO_2 as a component. Only CO_2 produced from naturally occurring CO_2 reservoirs, and as a byproduct from energy and industrial processes, and used in industrial applications other than EOR is included in this analysis. Carbon dioxide captured from biogenic sources (e.g., ethanol production plants) is not included in the Inventory. Carbon dioxide captured from crude oil and gas production is used in EOR applications and is therefore reported in the Energy chapter.

Carbon dioxide is produced as a byproduct of crude oil and natural gas production. This CO_2 is separated from the crude oil and natural gas using gas processing equipment, and may be emitted directly to the atmosphere, or captured and reinjected into underground formations, used for EOR, or sold for other commercial uses. A further discussion of CO_2 used in EOR is described in the Energy chapter in Box 3-7 titled "Carbon Dioxide Transport, Injection, and Geological Storage."

In 2017, the amount of CO_2 produced and captured for commercial applications and subsequently emitted to the atmosphere was 4.5 MMT CO_2 Eq. (4,471 kt) (see Table 4-53). This is consistent with 2014 through 2016 levels and is an increase of approximately 204 percent since 1990.

Table 4-53: CO_2 Emissions from CO_2 Consumption (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.5	1,472
2005	1.4	1,375
2013	4.2	4,188
2014	4.5	4,471
2015	4.5	4,471
2016	4.5	4,471
2017	4.5	4,471

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[222] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf>.

Methodology

Carbon dioxide emission estimates for 1990 through 2017 were based on the quantity of CO_2 extracted and transferred for industrial applications (i.e., non-EOR end-uses). Some of the CO_2 produced by these facilities is used for EOR and some is used in other commercial applications (e.g., chemical manufacturing, food production). It is assumed that 100 percent of the CO_2 production used in commercial applications other than EOR is eventually released into the atmosphere.

2010 through 2017

For 2010 through 2017, data from EPA's GHGRP (Subpart PP) were aggregated from facility-level reports to develop a national-level estimate for use in the Inventory (EPA 2019). However, for the years 2015 through 2017, GHGRP Subpart PP values did not pass GHGRP confidential business information (CBI) criteria for data aggregation. Facilities report CO_2 extracted or produced from natural reservoirs and industrial sites, and CO_2 captured from energy and industrial processes and transferred to various end-use applications to EPA's GHGRP. This analysis includes only reported CO_2 transferred to food and beverage end-uses. EPA is continuing to analyze and assess integration of CO_2 transferred to other end-uses to enhance the completeness of estimates under this source category. Other end-uses include industrial applications, such as metal fabrication. EPA is analyzing the information reported to ensure that other end-use data excludes non-emissive applications and publication will not reveal CBI. Reporters subject to EPA's GHGRP Subpart PP are also required to report the quantity of CO_2 that is imported and/or exported. Currently, these data are not publicly available through the GHGRP due to data confidentiality reasons and hence are excluded from this analysis.

Facilities subject to Subpart PP of EPA's GHGRP are required to measure CO_2 extracted or produced. More details on the calculation and monitoring methods applicable to extraction and production facilities can be found under Subpart PP: Suppliers of Carbon Dioxide of the regulation, Part 98.[223] The number of facilities that reported data to EPA's GHGRP Subpart PP (Suppliers of Carbon Dioxide) for 2010 through 2017 is much higher (ranging from 44 to 48) than the number of facilities included in the Inventory for the 1990 to 2009 time period prior to the availability of GHGRP data (4 facilities). The difference is largely due to the fact the 1990 to 2009 data includes only CO_2 transferred to end-use applications from naturally occurring CO_2 reservoirs and excludes industrial sites.

As previously mentioned, data from EPA's GHGRP (Subpart PP) was unavailable for use for the years 2015 through 2017 due to data confidentiality reasons. As a result, the emissions estimates for 2015 through 2017 have been held constant from 2014 levels to avoid disclosure of proprietary information. EPA continues to evaluate options for utilizing GHGRP data to update these values for future Inventories. Additional information on evaluating GHGRP Subpart PP data is included in the Planned Improvements section.

1990 through 2009

For 1990 through 2009, data from EPA's GHGRP are not available. For this time period, CO_2 production data from four naturally-occurring CO_2 reservoirs were used to estimate annual CO_2 emissions. These facilities were Jackson Dome in Mississippi, Brave and West Bravo Domes in New Mexico, and McCallum Dome in Colorado. The facilities in Mississippi and New Mexico produced CO_2 for use in both EOR and in other commercial applications (e.g., chemical manufacturing, food production). The fourth facility in Colorado (McCallum Dome**)** produced CO_2 for commercial applications only (New Mexico Bureau of Geology and Mineral Resources 2006).

Carbon dioxide production data and the percentage of production that was used for non-EOR applications for the Jackson Dome, Mississippi facility were obtained from Advanced Resources International (ARI 2006, 2007) for 1990 to 2000, and from the Annual Reports of Denbury Resources (Denbury Resources 2002 through 2010) for 2001 to 2009 (see Table 4-54). Denbury Resources reported the average CO_2 production in units of MMCF CO_2 per day for 2001 through 2009 and reported the percentage of the total average annual production that was used for EOR. Production from 1990 to 1999 was set equal to 2000 production, due to lack of publicly available production data for 1990 through 1999. Carbon dioxide production data for the Bravo Dome and West Bravo Dome were obtained from ARI for 1990 through 2009 (ARI 1990 to 2010). Data for the West Bravo Dome facility were only

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[223] See <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>.

available for 2009. The percentage of total production that was used for non-EOR applications for the Bravo Dome and West Bravo Dome facilities for 1990 through 2009 were obtained from New Mexico Bureau of Geology and Mineral Resources (Broadhead 2003; New Mexico Bureau of Geology and Mineral Resources 2006). Production data for the McCallum Dome (Jackson County), Colorado facility were obtained from the Colorado Oil and Gas Conservation Commission (COGCC) for 1999 through 2009 (COGCC 2014). Production data for 1990 to 1998 and percentage of production used for EOR were assumed to be the same as for 1999, due to lack of publicly-available data.

Table 4-54: CO_2 Production (kt CO_2) and the Percent Used for Non-EOR Applications

Year	Jackson Dome, MS CO_2 Production (kt) (% Non-EOR)	Bravo Dome, NM CO_2 Production (kt) (% Non-EOR)	West Bravo Dome, NM CO_2 Production (kt) (% Non-EOR)	McCallum Dome, CO CO_2 Production (kt) (% Non-EOR)	Total CO_2 Production from Extraction and Capture Facilities (kt)	% Non-EOR[a]
1990	1,344 (100%)	63 (1%)	+	65 (100%)	NA	NA
2005	1,254 (27%)	58 (1%)	+	63 (100%)	NA	NA
2013	NA	NA	NA	NA	68,435 [b]	6%
2014	NA	NA	NA	NA	72,000 [b]	6%
2015	NA	NA	NA	NA	72,000 [b]	6%
2016	NA	NA	NA	NA	72,000 [b]	6%
2017	NA	NA	NA	NA	72,000b	6%

+ Does not exceed 0.5 percent.

NA (Not Available)

[a] Includes only food & beverage applications.

[b] For 2010 through 2017, the publicly available GHGRP data were aggregated at the national level. From 2010 through 2014, those aggregated values based GHGRP CBI criteria. For 2015 through 2017, values were held constant with those from 2014. Facility-level data are not publicly available from EPA's GHGRP.

Uncertainty and Time-Series Consistency

There is uncertainty associated with the data reported through EPA's GHGRP. Specifically, there is uncertainty associated with the amount of CO_2 consumed for food and beverage applications given a threshold for reporting under GHGRP applicable to those reporting under Subpart PP, in addition to the exclusion of the amount of CO_2 transferred to all other end-use categories. This latter category might include CO_2 quantities that are being used for non-EOR industrial applications such as firefighting. Second, uncertainty is associated with the exclusion of imports/exports data for CO_2 suppliers. Currently these data are not publicly available through EPA's GHGRP and hence are excluded from this analysis. EPA verifies annual facility-level reports through a multi-step process (e.g., combination of electronic checks and manual reviews by staff) to identify potential errors and ensure that data submitted to EPA are accurate, complete, and consistent. Based on the results of the verification process, EPA follows up with facilities to resolve mistakes that may have occurred.[224]

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-55. Carbon dioxide consumption CO_2 emissions for 2017 were estimated to be between 4.2 and 4.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the emission estimate of 4.5 MMT CO_2 Eq.

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[224] See <https://www.epa.gov/sites/production/files/2015-07/documents/ghgrp_verification_factsheet.pdf >.

Table 4-55: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from CO_2 Consumption (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
CO_2 Consumption	CO_2	4.5	4.2	4.7	-5%	+5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

EPA will continue to evaluate the potential to include additional GHGRP data on other emissive end-uses to improve the accuracy and completeness of estimates for this source category. Particular attention will be made to ensuring time-series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[225] In addition, EPA is also investigating the possibility of utilizing only extraction facility Subpart PP data, while also updating the values for 2015 through 2017.

These improvements, in addition to updating the time series when new data is available, are still in process and will be incorporated into future Inventory reports. These are near- to medium-term improvements.

4.16 Phosphoric Acid Production (CRF Source Category 2B10)

Phosphoric acid (H_3PO_4) is a basic raw material used in the production of phosphate-based fertilizers. Phosphoric acid production from natural phosphate rock is a source of carbon dioxide (CO_2) emissions, due to the chemical reaction of the inorganic carbon (calcium carbonate) component of the phosphate rock.

Phosphate rock is mined in Florida and North Carolina, which account for more than 75 percent of total domestic output, as well as in Idaho and Utah and is used primarily as a raw material for wet-process phosphoric acid production (USGS 2018). The composition of natural phosphate rock varies depending upon the location where it is mined. Natural phosphate rock mined in the United States generally contains inorganic carbon in the form of calcium carbonate (limestone) and also may contain organic carbon. The calcium carbonate component of the phosphate rock is integral to the phosphate rock chemistry. Phosphate rock can also contain organic carbon that is physically incorporated into the mined rock but is not an integral component of the phosphate rock chemistry.

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[225] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

The phosphoric acid production process involves chemical reaction of the calcium phosphate ($Ca_3(PO_4)_2$) component of the phosphate rock with sulfuric acid (H_2SO_4) and recirculated phosphoric acid (H_3PO_4) (EFMA 2000). However, the generation of CO_2 is due to the associated limestone-sulfuric acid reaction, as shown below:

$$CaCO_3 + H_2SO_4 + H_2O \rightarrow CaSO_4 \bullet 2H_2O + CO_2$$

Total U.S. phosphate rock production used in 2017 was an estimated 26.7 million metric tons (USGS 2018). Total imports of phosphate rock to the United States in 2017 were estimated to be approximately 2.1 million metric tons (USGS 2018). Between 2013 and 2016, most of the imported phosphate rock (67 percent) came from Peru, with 32 percent being from Morocco and 1 percent from other sources (USGS 2018). All phosphate rock mining companies are vertically integrated with fertilizer plants that produce phosphoric acid located near the mines. Some additional phosphoric acid production facilities are located in Texas, Louisiana, and Mississippi that used imported phosphate rock.

Over the 1990 to 2017 period, domestic production has decreased by nearly 46 percent. Total CO_2 emissions from phosphoric acid production were 1.0 MMT CO_2 Eq. (1,023 kt CO_2) in 2017 (see Table 4-56). Domestic consumption of phosphate rock in 2017 was estimated to have increased 2 percent over 2016 levels (USGS 2018).

Table 4-56: CO_2 Emissions from Phosphoric Acid Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.5	1,529
2005	1.3	1,342
2013	1.1	1,149
2014	1.0	1,038
2015	1.0	999
2016	1.0	998
2017	1.0	1,023

Methodology

Carbon dioxide emissions from production of phosphoric acid from phosphate rock are estimated by multiplying the average amount of inorganic carbon (expressed as CO_2) contained in the natural phosphate rock as calcium carbonate by the amount of phosphate rock that is used annually to produce phosphoric acid, accounting for domestic production and net imports for consumption. The estimation methodology is as follows:

$$E_{pa} = C_{pr} \times Q_{pr}$$

where,

E_{pa} = CO_2 emissions from phosphoric acid production, metric tons
C_{pr} = Average amount of carbon (expressed as CO_2) in natural phosphate rock, metric ton CO_2/ metric ton phosphate rock
Q_{pr} = Quantity of phosphate rock used to produce phosphoric acid

The CO_2 emissions calculation methodology is based on the assumption that all of the inorganic C (calcium carbonate) content of the phosphate rock reacts to produce CO_2 in the phosphoric acid production process and is emitted with the stack gas. The methodology also assumes that none of the organic C content of the phosphate rock is converted to CO_2 and that all of the organic C content remains in the phosphoric acid product. The United States uses a country-specific methodology to calculate emissions from production of phosphoric acid from phosphate rock.

From 1993 to 2004, the U.S. Geological Survey (USGS) *Mineral Yearbook: Phosphate Rock* disaggregated phosphate rock mined annually in Florida and North Carolina from phosphate rock mined annually in Idaho and Utah, and reported the annual amounts of phosphate rock exported and imported for consumption (see Table 4-57). For the years 1990 through 1992, and 2005 through 2017, only nationally aggregated mining data was reported by USGS. For the years 1990, 1991, and 1992, the breakdown of phosphate rock mined in Florida and North Carolina, and the amount mined in Idaho and Utah, are approximated using average share of U.S. production in those states from 1993 to 2004 data. For the years 2005 through 2017, the same approximation method is used, but the share of U.S. production in those states data were obtained from the USGS commodity specialist for phosphate rock (USGS 2012). Data for domestic sales or consumption of phosphate rock, exports of phosphate rock (primarily from Florida and North Carolina), and imports of phosphate rock for consumption for 1990 through 2017 were obtained from USGS *Minerals Yearbook: Phosphate Rock* (USGS 1994 through 2015b), and from USGS *Minerals Commodity Summaries: Phosphate Rock* (USGS 2016, 2017, 2018). From 2004 through 2017, the USGS reported no exports of phosphate rock from U.S. producers (USGS 2005 through 2015b).

The carbonate content of phosphate rock varies depending upon where the material is mined. Composition data for domestically mined and imported phosphate rock were provided by the Florida Institute of Phosphate Research (FIPR 2003a). Phosphate rock mined in Florida contains approximately 1 percent inorganic C, and phosphate rock imported from Morocco contains approximately 1.46 percent inorganic carbon. Calcined phosphate rock mined in North Carolina and Idaho contains approximately 0.41 percent and 0.27 percent inorganic C, respectively (see Table 4-58).

Carbonate content data for phosphate rock mined in Florida are used to calculate the CO_2 emissions from consumption of phosphate rock mined in Florida and North Carolina (80 percent of domestic production) and carbonate content data for phosphate rock mined in Morocco are used to calculate CO_2 emissions from consumption of imported phosphate rock. The CO_2 emissions calculation is based on the assumption that all of the domestic production of phosphate rock is used in uncalcined form. As of 2006, the USGS noted that one phosphate rock producer in Idaho produces calcined phosphate rock; however, no production data were available for this single producer (USGS 2006). The USGS confirmed that no significant quantity of domestic production of phosphate rock is in the calcined form (USGS 2012).

Table 4-57: Phosphate Rock Domestic Consumption, Exports, and Imports (kt)

Location/Year	1990	2005	2013	2014	2015	2016	2017
U.S. Domestic Consumption	49,800	35,200	28,800	26,700	26,200	26,700	26,700
FL and NC	42,494	28,160	23,040	21,360	20,960	21,360	21,360
ID and UT	7,306	7,040	5,760	5,340	5,240	5,340	5,340
Exports—FL and NC	6,240	0	0	0	0	0	0
Imports	451	2,630	3,170	2,390	1,960	1,590	2,100
Total U.S. Consumption	**44,011**	**37,830**	**31,970**	**29,090**	**28,160**	**28,290**	**28,800**

Table 4-58: Chemical Composition of Phosphate Rock (Percent by Weight)

Composition	Central Florida	North Florida	North Carolina (calcined)	Idaho (calcined)	Morocco
Total Carbon (as C)	1.60	1.76	0.76	0.60	1.56
Inorganic Carbon (as C)	1.00	0.93	0.41	0.27	1.46
Organic Carbon (as C)	0.60	0.83	0.35	0.00	0.10
Inorganic Carbon (as CO_2)	3.67	3.43	1.50	1.00	5.00

Source: FIPR (2003a).

Uncertainty and Time-Series Consistency

Phosphate rock production data used in the emission calculations were developed by the USGS through monthly and semiannual voluntary surveys of the active phosphate rock mines during 2017. For previous years in the time series, USGS provided the data disaggregated regionally; however, beginning in 2006, only total U.S. phosphate rock production was reported. Regional production for 2017 was estimated based on regional production data from previous years and multiplied by regionally-specific emission factors. There is uncertainty associated with the degree to which the estimated 2017 regional production data represents actual production in those regions. Total U.S. phosphate rock production data are not considered to be a significant source of uncertainty because all the domestic phosphate rock producers report their annual production to the USGS. Data for exports of phosphate rock used in the emission calculation are reported by phosphate rock producers and are not considered to be a significant source of uncertainty. Data for imports for consumption are based on international trade data collected by the U.S. Census Bureau. These U.S. government economic data are not considered to be a significant source of uncertainty.

An additional source of uncertainty in the calculation of CO_2 emissions from phosphoric acid production is the carbonate composition of phosphate rock; the composition of phosphate rock varies depending upon where the material is mined, and may also vary over time. The Inventory relies on one study (FIPR 2003a) of chemical composition of the phosphate rock; limited data are available beyond this study. Another source of uncertainty is the disposition of the organic carbon content of the phosphate rock. A representative of FIPR indicated that in the phosphoric acid production process, the organic C content of the mined phosphate rock generally remains in the phosphoric acid product, which is what produces the color of the phosphoric acid product (FIPR 2003b). Organic carbon is therefore not included in the calculation of CO_2 emissions from phosphoric acid production.

A third source of uncertainty is the assumption that all domestically-produced phosphate rock is used in phosphoric acid production and used without first being calcined. Calcination of the phosphate rock would result in conversion of some of the organic C in the phosphate rock into CO_2. However, according to air permit information available to the public, at least one facility has calcining units permitted for operation (NCDENR 2013).

Finally, USGS indicated that approximately 7 percent of domestically-produced phosphate rock is used to manufacture elemental phosphorus and other phosphorus-based chemicals, rather than phosphoric acid (USGS 2006). According to USGS, there is only one domestic producer of elemental phosphorus, in Idaho, and no data were available concerning the annual production of this single producer. Elemental phosphorus is produced by reducing phosphate rock with coal coke, and it is therefore assumed that 100 percent of the carbonate content of the phosphate rock will be converted to CO_2 in the elemental phosphorus production process. The calculation for CO_2 emissions is based on the assumption that phosphate rock consumption, for purposes other than phosphoric acid production, results in CO_2 emissions from 100 percent of the inorganic carbon content in phosphate rock, but none from the organic carbon content.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-59. 2017 phosphoric acid production CO_2 emissions were estimated to be between 0.9 and 1.3 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 19 percent below and 21 percent above the emission estimate of 1.0 MMT CO_2 Eq.

Table 4-59: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Phosphoric Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Phosphoric Acid Production	CO_2	1.0	0.9	1.3	-19%	+21%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

EPA continues to evaluate potential improvements to the Inventory estimates for this source category, which include direct integration of EPA's GHGRP data for 2010 through 2017 and the use of reported GHGRP data to update the inorganic C content of phosphate rock for prior years. Confidentiality of data continues to be assessed, in addition to the applicability of GHGRP data for the averaged inorganic C content data (by region) from 2010 through 2017 to inform estimates in prior years in the required time series (i.e., 1990 through 2009). In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[226] This long-term planned improvement is still in development by EPA and have not been implemented into the current Inventory report.

4.17 Iron and Steel Production (CRF Source Category 2C1) and Metallurgical Coke Production

Iron and steel production is a multi-step process that generates process-related emissions of carbon dioxide (CO_2) and methane (CH_4) as raw materials are refined into iron and then transformed into crude steel. Emissions from conventional fuels (e.g., natural gas, fuel oil) consumed for energy purposes during the production of iron and steel are accounted for in the Energy chapter.

Iron and steel production includes six distinct production processes: coke production, sinter production, direct reduced iron (DRI) production, pig iron[227] production, electric arc furnace (EAF) steel production, and basic oxygen furnace (BOF) steel production. The number of production processes at a particular plant is dependent upon the specific plant configuration. Most process CO_2 generated from the iron and steel industry is a result of the production of crude iron.

In addition to the production processes mentioned above, CO_2 is also generated at iron and steel mills through the consumption of process byproducts (e.g., blast furnace gas, coke oven gas) used for various purposes including heating, annealing, and electricity generation. Process byproducts sold for use as synthetic natural gas are deducted and reported in the Energy chapter. In general, CO_2 emissions are generated in these production processes through the reduction and consumption of various carbon-containing inputs (e.g., ore, scrap, flux, coke byproducts). In addition, fugitive CH_4 emissions can also be generated from these processes, as well as from sinter, direct iron and pellet production.

Currently, there are approximately nine integrated iron and steel steelmaking facilities that utilize BOFs to refine and produce steel from iron. These facilities have 21 active blast furnaces between them as of 2015. Almost 100 steelmaking facilities utilize EAFs to produce steel primarily from recycled ferrous scrap (USGS 2018). The trend in the United States for integrated facilities has been a shift towards fewer BOFs and more EAFs. EAFs use scrap steel as their main input and use significantly less energy than BOFs. In addition, there are 16 cokemaking facilities, of which 3 facilities are co-located with integrated iron and steel facilities (ACCCI 2016). In the United States, four states – Indiana, Ohio, Michigan, and Pennsylvania – count for roughly 51 percent of total raw steel production (USGS 2018).

Total annual production of crude steel in the United States was fairly constant between 2000 and 2008 ranged from a low of 99,320,000 tons to a high of 109,880,000 tons (2001 and 2004, respectively). Due to the decrease in demand caused by the global economic downturn (particularly from the automotive industry), crude steel production in the United States sharply decreased to 65,459,000 tons in 2009. Crude steel production was fairly constant from 2011 through 2014, and after a dip in production from 2014 to 2015, crude steel production has slowly and steadily increased for the past few years. The United States was the fourth largest producer of raw steel in the world, behind China, Japan and India, accounting for approximately 4.8 percent of world production in 2017 (AISI 2004 through 2018).

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[226] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

[227] Pig iron is the common industry term to describe what should technically be called crude iron. Pig iron is a subset of crude iron that has lost popularity over time as industry trends have shifted. Throughout this report pig iron will be used interchangeably with crude iron, but it should be noted that in other data sets or reports pig iron and crude iron may not be used interchangeably and may provide different values.

The majority of CO_2 emissions from the iron and steel production process come from the use of coke in the production of pig iron and from the consumption of other process byproducts, with lesser amounts emitted from the use of flux and from the removal of carbon from pig iron used to produce steel.

According to the *2006 IPCC Guidelines*, the production of metallurgical coke from coking coal is considered to be an energy use of fossil fuel and the use of coke in iron and steel production is considered to be an industrial process source. Therefore, the *2006 IPCC Guidelines* suggest that emissions from the production of metallurgical coke should be reported separately in the Energy sector, while emissions from coke consumption in iron and steel production should be reported in the Industrial Processes and Product Use sector. However, the approaches and emission estimates for both metallurgical coke production and iron and steel production are presented here because much of the relevant activity data is used to estimate emissions from both metallurgical coke production and iron and steel production. For example, some byproducts (e.g., coke oven gas) of the metallurgical coke production process are consumed during iron and steel production, and some byproducts of the iron and steel production process (e.g., blast furnace gas) are consumed during metallurgical coke production. Emissions associated with the consumption of these byproducts are attributed at the point of consumption. Emissions associated with the use of conventional fuels (e.g., natural gas, fuel oil) for electricity generation, heating and annealing, or other miscellaneous purposes downstream of the iron and steelmaking furnaces are reported in the Energy chapter.

Metallurgical Coke Production

Emissions of CO_2 from metallurgical coke production in 2017 were 0.6 MMT CO_2 Eq. (578 kt CO_2) (see Table 4-60 and Table 4-61). Emissions decreased significantly in 2017 by 56 percent from 2016 levels and have decreased by 77 percent (1.9 MMT CO_2 Eq.) since 1990. Coke production in 2017 was 38 percent lower than in 2000 and 53 percent below 1990.

Table 4-60: CO_2 Emissions from Metallurgical Coke Production (MMT CO_2 Eq.)

Gas	1990	2005	2013	2014	2015	2016	2017
CO_2	2.5	2.1	1.8	2.0	2.8	1.3	0.6
Total	**2.5**	**2.1**	**1.8**	**2.0**	**2.8**	**1.3**	**0.6**

Table 4-61: CO_2 Emissions from Metallurgical Coke Production (kt)

Gas	1990	2005	2013	2014	2015	2016	2017
CO_2	2,504	2,050	1,830	2,020	2,843	1,327	578
Total	**2,504**	**2,050**	**1,830**	**2,020**	**2,843**	**1,327**	**578**

Iron and Steel Production

Emissions of CO_2 and CH_4 from iron and steel production in 2017 were 41.2 MMT CO_2 Eq. (41,201 kt) and 0.0073 MMT CO_2 Eq. (0.3 kt CH_4), respectively (see Table 4-62 through Table 4-65), totaling approximately 41.2 MMT CO_2 Eq. Emissions slightly increased in 2017 from 2016 but have decreased overall since 1990 due to restructuring of the industry, technological improvements, and increased scrap steel utilization. Carbon dioxide emission estimates include emissions from the consumption of carbonaceous materials in the blast furnace, EAF, and BOF, as well as blast furnace gas and coke oven gas consumption for other activities at the steel mill.

In 2017, domestic production of pig iron did not change from 2016 levels. Overall, domestic pig iron production has declined since the 1990s. Pig iron production in 2017 was 53 percent lower than in 2000 and 55 percent below 1990. Carbon dioxide emissions from iron production have decreased by 77 percent since 1990. Carbon dioxide emissions from steel production have decreased by 16 percent (1.3 MMT CO_2 Eq.) since 1990, while overall CO_2 emissions from iron and steel production have declined by 58 percent (57.9 MMT CO_2 Eq.) from 1990 to 2017.

Table 4-62: CO_2 Emissions from Iron and Steel Production (MMT CO_2 Eq.)

Source/Activity Data	1990	2005	2013	2014	2015	2016	2017
Sinter Production	2.4	1.7	1.1	1.1	1.0	0.9	0.9

Iron Production	45.7	17.7	12.0	18.7	11.8	9.9	10.4
Pellet Production	1.8	1.5	1.1	1.1	1.0	0.9	0.9
Steel Production	8.0	9.4	8.6	7.5	6.9	6.9	6.7
Other Activities[a]	41.2	35.9	28.7	27.9	24.3	22.5	22.4
Total	**99.1**	**66.2**	**51.6**	**56.3**	**45.0**	**41.0**	**41.2**

[a] Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Note: Totals may not sum due to independent rounding.

Table 4-63: CO_2 Emissions from Iron and Steel Production (kt)

Source/Activity Data	**1990**	**2005**	**2013**	**2014**	**2015**	**2016**	**2017**
Sinter Production	2,448	1,663	1,117	1,104	1,016	877	869
Iron Production	45,704	17,664	12,031	18,722	11,780	9,928	10,386
Pellet Production	1,817	1,503	1,146	1,126	964	869	867
Steel Production	7,965	9,396	8,638	7,469	6,941	6,858	6,691
Other Activities[a]	41,193	35,934	28,709	27,911	24,280	22,451	22,390
Total	**99,126**	**66,160**	**51,641**	**56,332**	**44,981**	**40,983**	**41,204**

[a] Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Note: Totals may not sum due to independent rounding.

Table 4-64: CH_4 Emissions from Iron and Steel Production (MMT CO_2 Eq.)

Source/Activity Data	**1990**	**2005**	**2013**	**2014**	**2015**	**2016**	**2017**
Sinter Production	+	+	+	+	+	+	+
Total	**+**	**+**	**+**	**+**	**+**	**+**	**+**

\+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-65: CH_4 Emissions from Iron and Steel Production (kt)

Source/Activity Data	**1990**	**2005**	**2013**	**2014**	**2015**	**2016**	**2017**
Sinter Production	0.9	0.6	0.4	0.4	0.3	0.3	0.3
Total	**0.9**	**0.6**	**0.4**	**0.4**	**0.3**	**0.3**	**0.3**

Methodology

Emission estimates presented in this chapter utilize a country-specific approach based on Tier 2 methodologies provided by the *2006 IPCC Guidelines*. These Tier 2 methodologies call for a mass balance accounting of the carbonaceous inputs and outputs during the iron and steel production process and the metallurgical coke production process. Tier 1 methods are used for certain iron and steel production processes (i.e., sinter production, pellet production and DRI production) for which available data are insufficient to apply a Tier 2 method.

The Tier 2 methodology equation is as follows:

$$E_{CO_2} = \left[\sum_a (Q_a \times C_a) - \sum_b (Q_b \times C_b)\right] \times \frac{44}{12}$$

where,

E_{CO2} = Emissions from coke, pig iron, EAF steel, or BOF steel production, metric tons
a = Input material *a*
b = Output material *b*
Q_a = Quantity of input material *a*, metric tons
C_a = Carbon content of input material *a*, metric tons C/metric ton material
Q_b = Quantity of output material *b*, metric tons

C_b = Carbon content of output material *b*, metric tons C/metric ton material
44/12 = Stoichiometric ratio of CO_2 to C

The Tier 1 methodology equations are as follows:

$$E_{s,p} = Q_s \times EF_{s,p}$$

$$E_{d,CO2} = Q_d \times EF_{d,CO2}$$

$$E_{p,CO2} = Q_p \times EF_{p,CO2}$$

where,

$E_{s,p}$ = Emissions from sinter production process for pollutant *p* (CO_2 or CH_4), metric ton
Q_s = Quantity of sinter produced, metric tons
$EF_{s,p}$ = Emission factor for pollutant *p* (CO_2 or CH_4), metric ton *p*/metric ton sinter
$E_{d,CO2}$ = Emissions from DRI production process for CO_2, metric ton
Q_d = Quantity of DRI produced, metric tons
$EF_{d,CO2}$ = Emission factor for CO_2, metric ton CO_2/metric ton DRI
Q_p = Quantity of pellets produced, metric tons
$EF_{p,CO2}$ = Emission factor for CO_2, metric ton CO_2/metric ton pellets produced

Metallurgical Coke Production

Coking coal is used to manufacture metallurgical coke that is used primarily as a reducing agent in the production of iron and steel, but is also used in the production of other metals including zinc and lead (see Zinc Production and Lead Production sections of this chapter). Emissions associated with producing metallurgical coke from coking coal are estimated and reported separately from emissions that result from the iron and steel production process. To estimate emissions from metallurgical coke production, a Tier 2 method provided by the *2006 IPCC Guidelines* was utilized. The amount of carbon contained in materials produced during the metallurgical coke production process (i.e., coke, coke breeze and coke oven gas) is deducted from the amount of carbon contained in materials consumed during the metallurgical coke production process (i.e., natural gas, blast furnace gas, and coking coal). Light oil, which is produced during the metallurgical coke production process, is excluded from the deductions due to data limitations. The amount of carbon contained in these materials is calculated by multiplying the material-specific carbon content by the amount of material consumed or produced (see Table 4-66). The amount of coal tar produced was approximated using a production factor of 0.03 tons of coal tar per ton of coking coal consumed. The amount of coke breeze produced was approximated using a production factor of 0.075 tons of coke breeze per ton of coking coal consumed (AISI 2008; DOE 2000). Data on the consumption of carbonaceous materials (other than coking coal) as well as coke oven gas production were available for integrated steel mills only (i.e., steel mills with co-located coke plants). Therefore, carbonaceous material (other than coking coal) consumption and coke oven gas production were excluded from emission estimates for merchant coke plants. Carbon contained in coke oven gas used for coke-oven underfiring was not included in the deductions to avoid double-counting.

Table 4-66: Material Carbon Contents for Metallurgical Coke Production

Material	kg C/kg
Coal Tar	0.62
Coke	0.83
Coke Breeze	0.83
Coking Coal	0.73
Material	**kg C/GJ**
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC (2006), Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

Although the *2006 IPCC Guidelines* provide a Tier 1 CH_4 emission factor for metallurgical coke production (i.e., 0.1 g CH_4 per metric ton of coke production), it is not appropriate to use because CO_2 emissions were estimated using the Tier 2 mass balance methodology. The mass balance methodology makes a basic assumption that all carbon that enters the metallurgical coke production process either exits the process as part of a carbon-containing output or as CO_2 emissions. This is consistent with a preliminary assessment of aggregated facility-level greenhouse gas CH_4 emissions reported by coke production facilities under EPA's GHGRP. The assessment indicates that CH_4 emissions from coke production are insignificant and below 500 kt or 0.05 percent of total national emissions. Pending resources and significance, EPA continues to assess the possibility of including these emissions in future Inventories to enhance completeness but has not incorporated these emissions into this report.

Data relating to the mass of coking coal consumed at metallurgical coke plants and the mass of metallurgical coke produced at coke plants were taken from the Energy Information Administration (EIA) *Quarterly Coal Report: October through December* (EIA 1998 through 2018) (see Table 4-67). Data on the volume of natural gas consumption, blast furnace gas consumption, and coke oven gas production for metallurgical coke production at integrated steel mills were obtained from the American Iron and Steel Institute (AISI) *Annual Statistical Report* (AISI 2004 through 2018) and through personal communications with AISI (AISI 2008) (see Table 4-68). The factor for the quantity of coal tar produced per ton of coking coal consumed was provided by AISI (AISI 2008). The factor for the quantity of coke breeze produced per ton of coking coal consumed was obtained through Table 2-1 of the report *Energy and Environmental Profile of the U.S. Iron and Steel Industry* (DOE 2000). Currently, data on natural gas consumption and coke oven gas production at merchant coke plants were not available and were excluded from the emission estimate. Carbon contents for coking coal, metallurgical coke, coal tar, coke oven gas, and blast furnace gas were provided by the *2006 IPCC Guidelines*. The C content for coke breeze was assumed to equal the C content of coke.

Table 4-67: Production and Consumption Data for the Calculation of CO_2 Emissions from Metallurgical Coke Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2013	2014	2015	2016	2017
Metallurgical Coke Production							
Coking Coal Consumption at Coke Plants	35,269	21,259	19,481	19,321	17,879	14,955	15,910
Coke Production at Coke Plants	25,054	15,167	13,898	13,748	12,479	10,755	11,746
Coal Breeze Production	2,645	1,594	1,461	1,449	1,341	1,122	1,193
Coal Tar Production	1,058	638	584	580	536	449	477

Table 4-68: Production and Consumption Data for the Calculation of CO_2 Emissions from Metallurgical Coke Production (Million ft^3)

Source/Activity Data	1990	2005	2013	2014	2015	2016	2017
Metallurgical Coke Production							
Coke Oven Gas Production	250,767	114,213	108,162	102,899	84,336	74,807	74,997
Natural Gas Consumption	599	2,996	3,247	3,039	2,338	2,077	2,103
Blast Furnace Gas Consumption	24,602	4,460	4,255	4,346	4,185	3,741	3,683

Iron and Steel Production

To estimate emissions from pig iron production in the blast furnace, the amount of carbon contained in the produced pig iron and blast furnace gas were deducted from the amount of carbon contained in inputs (i.e., metallurgical coke, sinter, natural ore, pellets, natural gas, fuel oil, coke oven gas, carbonate fluxes or slagging materials, and direct coal injection). The carbon contained in the pig iron, blast furnace gas, and blast furnace inputs was estimated by multiplying the material-specific C content by each material type (see Table 4-69). Carbon in blast furnace gas used to pre-heat the blast furnace air is combusted to form CO_2 during this process. Carbon contained in blast furnace gas used as a blast furnace input was not included in the deductions to avoid double-counting.

Emissions from steel production in EAFs were estimated by deducting the carbon contained in the steel produced from the carbon contained in the EAF anode, charge carbon, and scrap steel added to the EAF. Small amounts of carbon from DRI and pig iron to the EAFs were also included in the EAF calculation. For BOFs, estimates of carbon contained in BOF steel were deducted from C contained in inputs such as natural gas, coke oven gas, fluxes (e.g. burnt lime or dolomite), and pig iron. In each case, the carbon was calculated by multiplying material-specific carbon contents by each material type (see Table 4-69). For EAFs, the amount of EAF anode consumed was approximated by multiplying total EAF steel production by the amount of EAF anode consumed per metric ton of steel produced (0.002 metric tons EAF anode per metric ton steel produced [AISI 2008]). The amount of flux (e.g., burnt lime or dolomite) used in pig iron production was deducted from the "Other Process Uses of Carbonates" source category (CRF Source Category 2A4) to avoid double-counting.

Carbon dioxide emissions from the consumption of blast furnace gas and coke oven gas for other activities occurring at the steel mill were estimated by multiplying the amount of these materials consumed for these purposes by the material-specific carbon content (see Table 4-69).

Carbon dioxide emissions associated with the sinter production, direct reduced iron production, pig iron production, steel production, and other steel mill activities were summed to calculate the total CO_2 emissions from iron and steel production (see Table 4-62 and Table 4-63).

Table 4-69: Material Carbon Contents for Iron and Steel Production

Material	kg C/kg
Coke	0.83
Direct Reduced Iron	0.02
Dolomite	0.13
EAF Carbon Electrodes	0.82
EAF Charge Carbon	0.83
Limestone	0.12
Pig Iron	0.04
Steel	0.01
Material	**kg C/GJ**
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC (2006), Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production process for sinter results in fugitive emissions of CH_4, which are emitted via leaks in the production equipment, rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors taken from the *2006 IPCC Guidelines* for sinter production (see Table 4-70). Although the *1995 IPCC Guidelines* (IPCC/UNEP/OECD/IEA 1995) provide a Tier 1 CH_4 emission factor for pig iron production, it is not appropriate to use because CO_2 emissions were estimated using the Tier 2 mass balance methodology. The mass balance methodology makes a basic assumption that all carbon that enters the pig iron production process either exits the process as part of a carbon-containing output or as CO_2 emissions; the estimation of CH_4 emissions is precluded. A preliminary analysis of facility-level emissions reported during iron production further supports this assumption and indicates that CH_4 emissions are below 500 kt CO_2 Eq. and well below 0.05 percent of total national emissions. The production of direct reduced iron also results in emissions of CH_4 through the consumption of fossil fuels (e.g., natural gas, etc.); however, these emission estimates are excluded due to data limitations. Pending further analysis and resources, EPA may include these emissions in future reports to enhance completeness. EPA is still assessing the possibility of including these emissions in future reports and have not included this data in the current report.

Table 4-70: CH_4 Emission Factors for Sinter and Pig Iron Production

Material Produced	Factor	Unit
Sinter	0.07	kg CH_4/metric ton

Source: IPCC (2006), Table 4.2.

Emissions of CO_2 from sinter production, direct reduced iron production and pellet production were estimated by multiplying total national sinter production and the total national direct reduced iron production by Tier 1 CO_2 emission factors (see Table 4-71). Because estimates of sinter production, direct reduced iron production and pellet production were not available, production was assumed to equal consumption.

Table 4-71: CO_2 Emission Factors for Sinter Production, Direct Reduced Iron Production and Pellet Production

Material Produced	Metric Ton CO_2/Metric Ton
Sinter	0.2
Direct Reduced Iron	0.7
Pellet Production	0.03

Source: IPCC (2006), Table 4.1.

The consumption of coking coal, natural gas, distillate fuel, and coal used in iron and steel production are adjusted for within the Energy chapter to avoid double-counting of emissions reported within the IPPU chapter as these fuels were consumed during non-energy related activities. More information on this methodology and examples of adjustments made between the IPPU and Energy chapters are described in Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

Sinter consumption and pellet consumption data for 1990 through 2017 were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2018) and through personal communications with AISI (AISI 2008) (see Table 4-72). In general, direct reduced iron (DRI) consumption data were obtained from the U.S. Geological Survey (USGS) *Minerals Yearbook – Iron and Steel Scrap* (USGS 1991 through 2016) and personal communication with the USGS Iron and Steel Commodity Specialist (Fenton 2015 through 2018). However, data for DRI consumed in EAFs were not available for the years 1990 and 1991. EAF DRI consumption in 1990 and 1991 was calculated by multiplying the total DRI consumption for all furnaces by the EAF share of total DRI consumption in 1992. Also, data for DRI consumed in BOFs were not available for the years 1990 through 1993. BOF DRI consumption in 1990 through 1993 was calculated by multiplying the total DRI consumption for all furnaces (excluding EAFs and cupola) by the BOF share of total DRI consumption (excluding EAFs and cupola) in 1994.

The Tier 1 CO_2 emission factors for sinter production, direct reduced iron production and pellet production were obtained through the *2006 IPCC Guidelines* (IPCC 2006). Time-series data for pig iron production, coke, natural gas, fuel oil, sinter, and pellets consumed in the blast furnace; pig iron production; and blast furnace gas produced at the iron and steel mill and used in the metallurgical coke ovens and other steel mill activities were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2018) and through personal communications with AISI (AISI 2008) (see Table 4-72 and Table 4-73).

Data for EAF steel production, flux, EAF charge carbon, and natural gas consumption were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2018) and through personal communications with AISI (AISI 2006 through 2016 and AISI 2008). The factor for the quantity of EAF anode consumed per ton of EAF steel produced was provided by AISI (AISI 2008). Data for BOF steel production, flux, natural gas, natural ore, pellet, sinter consumption as well as BOF steel production were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2018) and through personal communications with AISI (AISI 2008). Data for EAF and BOF scrap steel, pig iron, and DRI consumption were obtained from the USGS *Minerals Yearbook – Iron and Steel Scrap* (USGS 1991 through 2016). Data on coke oven gas and blast furnace gas consumed at the iron and steel mill (other than in the EAF, BOF, or blast furnace) were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2018) and through personal communications with AISI (AISI 2008).

Data on blast furnace gas and coke oven gas sold for use as synthetic natural gas were obtained from EIA's *Natural Gas Annual* (EIA 2016b). Carbon contents for direct reduced iron, EAF carbon electrodes, EAF charge carbon, limestone, dolomite, pig iron, and steel were provided by the *2006 IPCC Guidelines*. The carbon contents for natural gas, fuel oil, and direct injection coal were obtained from EIA (EIA 2017c) and EPA (EPA 2010). Heat contents for fuel oil and direct injection coal were obtained from EIA (EIA 1992, 2011); natural gas heat content was obtained from Table 37 of AISI's *Annual Statistical Report* (AISI 2004 through 2018). Heat contents for coke oven gas and blast furnace gas were provided in Table 37 of AISI's *Annual Statistical Report* (AISI 2004 through 2018) and confirmed by AISI staff (Carroll 2016).

Table 4-72: Production and Consumption Data for the Calculation of CO_2 and CH_4 Emissions from Iron and Steel Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2013	2014	2015	2016	2017
Sinter Production							

Sinter Production	12,239	8,315	5,583	5,521	5,079	4,385	4,347
Direct Reduced Iron Production							
Direct Reduced Iron Production	516	1,303	3,350	4,790	4,790	C	C
Pellet Production							
Pellet Production	60,563	50,096	38,198	37,538	32,146	28,967	28,916
Pig Iron Production							
Coke Consumption	24,946	13,832	9,308	11,136	7,969	7,124	7,101
Pig Iron Production	49,669	37,222	30,309	29,375	25,436	22,293	22,395
Direct Injection Coal Consumption	1,485	2,573	2,675	2,425	2,275	1,935	2,125
EAF Steel Production							
EAF Anode and Charge Carbon Consumption	67	1,127	1,122	1,062	1,072	1,120	1,127
Scrap Steel Consumption	42,691	46,600	47,300	48,873	44,000	C	C
Flux Consumption	319	695	771	771	998	998	998
EAF Steel Production	33,511	52,194	52,641	55,174	49,451	52,589	55,825
BOF Steel Production							
Pig Iron Consumption	47,307	34,400	29,600	23,755	20,349	C	C
Scrap Steel Consumption	14,713	11,400	7,890	5,917	4,526	C	C
Flux Consumption	576	582	454	454	454	408	408
BOF Steel Production	43,973	42,705	34,238	33,000	29,396	25,888	25,788

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Table 4-73: Production and Consumption Data for the Calculation of CO_2 Emissions from Iron and Steel Production (Million ft^3 unless otherwise specified)

Source/Activity Data	1990	2005	2013	2014	2015	2016	2017
Pig Iron Production							
Natural Gas Consumption	56,273	59,844	48,812	47,734	43,294	38,396	38,142
Fuel Oil Consumption (thousand gallons)	163,397	16,170	17,468	16,674	9,326	6,124	4,352
Coke Oven Gas Consumption	22,033	16,557	17,710	16,896	13,921	12,404	12,459
Blast Furnace Gas Production	1,439,380	1,299,980	1,026,973	1,000,536	874,670	811,005	808,499
EAF Steel Production							
Natural Gas Consumption	15,905	19,985	10,514	9,622	8,751	3,915	8,105
BOF Steel Production							
Coke Oven Gas Consumption	3,851	524	568	524	386	367	374
Other Activities							
Coke Oven Gas Consumption	224,883	97,132	89,884	85,479	70,029	62,036	62,164
Blast Furnace Gas Consumption	1,414,778	1,295,520	1,022,718	996,190	870,485	807,264	804,816

Uncertainty and Time-Series Consistency

The estimates of CO_2 emissions from metallurgical coke production are based on material production and consumption data and average carbon contents. Uncertainty is associated with the total U.S. coking coal consumption, total U.S. coke production and materials consumed during this process. Data for coking coal consumption and metallurgical coke production are from different data sources (EIA) than data for other carbonaceous materials consumed at coke plants (AISI), which does not include data for merchant coke plants.

There is uncertainty associated with the fact that coal tar and coke breeze production were estimated based on coke production because coal tar and coke breeze production data were not available. Since merchant coke plant data is not included in the estimate of other carbonaceous materials consumed at coke plants, the mass balance equation for CO_2 from metallurgical coke production cannot be reasonably completed. Therefore, for the purpose of this analysis, uncertainty parameters are applied to primary data inputs to the calculation (i.e., coking coal consumption and metallurgical coke production) only.

The estimates of CO_2 emissions from iron and steel production are based on material production and consumption data and average C contents. There is uncertainty associated with the assumption that pellet production, direct reduced iron and sinter consumption are equal to production. There is uncertainty with the representativeness of the associated IPCC default emission factors. There is uncertainty associated with the assumption that all coal used for purposes other than coking coal is for direct injection coal. There is also uncertainty associated with the C contents for pellets, sinter, and natural ore, which are assumed to equal the C contents of direct reduced iron, when consumed in the blast furnace. There is uncertainty associated with the consumption of natural ore under current industry practices. For EAF steel production, there is uncertainty associated with the amount of EAF anode and charge carbon consumed due to inconsistent data throughout the time series. Also for EAF steel production, there is uncertainty associated with the assumption that 100 percent of the natural gas attributed to "steelmaking furnaces" by AISI is process-related and nothing is combusted for energy purposes. Uncertainty is also associated with the use of process gases such as blast furnace gas and coke oven gas. Data are not available to differentiate between the use of these gases for processes at the steel mill versus for energy generation (i.e., electricity and steam generation); therefore, all consumption is attributed to iron and steel production. These data and carbon contents produce a relatively accurate estimate of CO_2 emissions. However, there are uncertainties associated with each.

For calculating the emissions estimates from iron and steel and metallurgical coke production, EPA utilizes a number of data points taken from the AISI *Annual Statistical Report* (ASR). This report serves as a benchmark for information on steel companies in United States, regardless if they are a member of AISI, which represents integrated producers (i.e., blast furnace and EAF). During the compilation of the 1990 through 2016 Inventory report EPA initiated conversation with AISI to better understand and update the qualitative and quantitative uncertainty metrics associated with AISI data elements. AISI estimates their data collection response rate to range from 75 to 90 percent, with certain sectors of the iron and steel industry not being covered by the ASR. Therefore, there is some inherent uncertainty in the values provided in the AISI ASR, including material production and consumption data. There is also some uncertainty to which materials produced are exported to Canada. As indicated in the introduction to this section, the trend for integrated facilities has moved to more use of EAFs and fewer BOFs. This trend may not be completely captured in the current data which also increases uncertainty. EPA currently uses an uncertainty range of ±10 percent for the primary data inputs to calculate overall uncertainty from iron and steel production, consistent with *2006 IPCC Guidelines*. During EPA's discussion with AISI, AISI noted that an uncertainty range of ±5 percent would be a more appropriate approximation to reflect their coverage of integrated steel producers in the United States. EPA will continue to assess the best range of uncertainty for these values.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-74 for metallurgical coke production and iron and steel production. Total CO_2 emissions from metallurgical coke production and iron and steel production for 2017 were estimated to be between 34.4 and 49.2 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 18 percent below and 18 percent above the emission estimate of 41.8 MMT CO_2 Eq. Total CH_4 emissions from metallurgical coke production and iron and steel production for 2017 were estimated to be between 0.006 and 0.009 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 19 percent below and 19 percent above the emission estimate of 0.007 MMT CO_2 Eq.

Table 4-74: Approach 2 Quantitative Uncertainty Estimates for CO_2 and CH_4 Emissions from Iron and Steel Production and Metallurgical Coke Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Metallurgical Coke & Iron and Steel Production	CO_2	41.8	34.4	49.2	-18%	+18%
Metallurgical Coke & Iron and Steel Production	CH_4	+	+	+	-19%	+19%

+ Does not exceed 0.05 MMT CO_2 Eq.
[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Future improvements involve improving activity data and emission factor sources for estimating CO_2 and CH_4 emissions from pellet production. EPA will also evaluate and analyze data reported under EPA's GHGRP to improve the emission estimates for this and other Iron and Steel Production process categories. Particular attention will be made to ensure time-series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[228] This is a medium-term improvement and EPA estimates that earliest this improvement could be incorporated is the 2020 Inventory submission.

Additional improvements include accounting for emission estimates for the production of metallurgical coke to the Energy chapter as well as identifying the amount of carbonaceous materials, other than coking coal, consumed at merchant coke plants. Other potential improvements include identifying the amount of coal used for direct injection and the amount of coke breeze, coal tar, and light oil produced during coke production. Efforts will also be made to identify information to better characterize emissions from the use of process gases and fuels within the Energy and IPPU chapters. Additional efforts will be made to improve the reporting between the IPPU and Energy chapters, particularly the inclusion of a quantitative summary of the carbon balance in the United States. This planned improvement is a medium-term improvement and is still in development; therefore, it is not included in this current Inventory report and is not expected until the 2021 Inventory submission.

EPA also received comments during the Expert Review cycle of the previous (i.e., 1990 through 2016) Inventory on recommendations to improve the description of the iron and steel industry and emissive processes. EPA began incorporating some of these recommendations into the previous Inventory (i.e., 1990 through 2016) and will require some additional time to implement other substantive changes. In addition, the EPA is currently developing an iron and steel carbon balance diagram to include in future inventory reports that will aid in the discussion of iron and steel processes. This improvement is expected for the 2020 Inventory submission.

4.18 Ferroalloy Production (CRF Source Category 2C2)

Carbon dioxide (CO_2) and methane (CH_4) are emitted from the production of several ferroalloys. Ferroalloys are composites of iron (Fe) and other elements such as silicon (Si), manganese (Mn), and chromium (Cr). Emissions from fuels consumed for energy purposes during the production of ferroalloys are accounted for in the Energy chapter. Emissions from the production of two types of ferrosilicon (25 to 55 percent and 56 to 95 percent silicon), silicon metal (96 to 99 percent silicon), and miscellaneous alloys (32 to 65 percent silicon) have been calculated. Emissions from the production of ferrochromium and ferromanganese are not included here because of the small

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[228] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

number of manufacturers of these materials in the United States, and therefore, government information disclosure rules prevent the publication of production data for these production facilities.

Similar to emissions from the production of iron and steel, CO_2 is emitted when metallurgical coke is oxidized during a high-temperature reaction with iron and the selected alloying element. Due to the strong reducing environment, CO is initially produced, and eventually oxidized to CO_2. A representative reaction equation for the production of 50 percent ferrosilicon (FeSi) is given below:

$$\mathrm{Fe_2O_3 + 2SiO_2 + 7C \rightarrow 2FeSi + 7CO}$$

While most of the carbon contained in the process materials is released to the atmosphere as CO_2, a percentage is also released as CH_4 and other volatiles. The amount of CH_4 that is released is dependent on furnace efficiency, operation technique, and control technology.

When incorporated in alloy steels, ferroalloys are used to alter the material properties of the steel. Ferroalloys are used primarily by the iron and steel industry, and production trends closely follow that of the iron and steel industry. As of 2017, ten companies in the United States produce ferroalloys (USGS 2016a).

Emissions of CO_2 from ferroalloy production in 2017 were 2.0 MMT CO_2 Eq. (1,975 kt CO_2) (see Table 4-75 and Table 4-76), which is an 8 percent reduction since 1990. Emissions of CH_4 from ferroalloy production in 2017 were 0.01 MMT CO_2 Eq. (0.6 kt CH_4), which is an 18 percent decrease since 1990.

Table 4-75: CO_2 and CH_4 Emissions from Ferroalloy Production (MMT CO_2 Eq.)

Gas	1990	2005	2013	2014	2015	2016	2017
CO_2	2.2	1.4	1.8	1.9	2.0	1.8	2.0
CH_4	+	+	+	+	+	+	+
Total	**2.2**	**1.4**	**1.8**	**1.9**	**2.0**	**1.8**	**2.0**

+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-76: CO_2 and CH_4 Emissions from Ferroalloy Production (kt)

Gas	1990	2005	2013	2014	2015	2016	2017
CO_2	2,152	1,392	1,785	1,914	1,960	1,796	1,975
CH_4	1	+	1	1	1	1	1

+ Does not exceed 0.5 kt.

Methodology

Emissions of CO_2 and CH_4 from ferroalloy production were calculated[229] using a Tier 1 method from the *2006 IPCC Guidelines* by multiplying annual ferroalloy production by material-specific default emission factors provided by IPCC (IPCC 2006). The Tier 1 equations for CO_2 and CH_4 emissions are as follows:

$$E_{CO_2} = \sum_i (MP_i \times EF_i)$$

where,

E_{CO2} = CO_2 emissions, metric tons
MP_i = Production of ferroalloy type *i*, metric tons
EF_i = Generic emission factor for ferroalloy type *i*, metric tons CO_2/metric ton specific ferroalloy product

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[229] EPA has not integrated aggregated facility-level GHGRP information to inform these estimates. The aggregated information (e.g., activity data and emissions) associated with production of ferroalloys did not meet criteria to shield underlying confidential business information (CBI) from public disclosure.

$$E_{CH_4} = \sum_{i} (MP_i \times EF_i)$$

where,

E_{CH4} = CH_4 emissions, kg
MP_i = Production of ferroalloy type i, metric tons
EF_i = Generic emission factor for ferroalloy type i, kg CH_4/metric ton specific ferroalloy product

Default emission factors were used because country-specific emission factors are not currently available. The following emission factors were used to develop annual CO_2 and CH_4 estimates:

- Ferrosilicon, 25 to 55 percent Si and Miscellaneous Alloys, 32 to 65 percent Si – 2.5 metric tons CO_2/metric ton of alloy produced; 1.0 kg CH_4/metric ton of alloy produced.
- Ferrosilicon, 56 to 95 percent Si – 4.0 metric tons CO_2/metric ton alloy produced; 1.0 kg CH_4/metric ton of alloy produced.
- Silicon Metal – 5.0 metric tons CO_2/metric ton metal produced; 1.2 kg CH_4/metric ton metal produced.

It was assumed that 100 percent of the ferroalloy production was produced using petroleum coke in an electric arc furnace process (IPCC 2006), although some ferroalloys may have been produced with coking coal, wood, other biomass, or graphite carbon inputs. The amount of petroleum coke consumed in ferroalloy production was calculated assuming that the petroleum coke used is 90 percent carbon (C) and 10 percent inert material (Onder and Bagdoyan 1993).

The use of petroleum coke for ferroalloy production is adjusted for within the Energy chapter as this fuel was consumed during non-energy related activities. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

Ferroalloy production data for 1990 through 2017 (see Table 4-77) were obtained from the U.S. Geological Survey (USGS) through the *Minerals Yearbook: Silicon* (USGS 1996 through 2013) and the *Mineral Industry Surveys: Silicon* (USGS 2014, 2015b, 2016b, 2017). The following data were available from the USGS publications for the time series:

- Ferrosilicon, 25 to 55 percent Si: Annual production data were available from 1990 through 2010.
- Ferrosilicon, 56 to 95 percent Si: Annual production data were available from 1990 through 2010.
- Silicon Metal: Annual production data were available from 1990 through 2005. The production data for 2005 were used as proxy for 2006 through 2010.
- Miscellaneous Alloys, 32 to 65 percent Si: Annual production data were available from 1990 through 1998. Starting 1999, USGS reported miscellaneous alloys and ferrosilicon containing 25 to 55 percent silicon as a single category.

Starting with the 2011 publication, USGS ceased publication of production quantity by ferroalloy product and began reporting all the ferroalloy production data as a single category (i.e., Total Silicon Materials Production). This is due to the small number of ferroalloy manufacturers in the United States and government information disclosure rules. Ferroalloy product shares developed from the 2010 production data (i.e., ferroalloy product production/total ferroalloy production) were used with the total silicon materials production quantity to estimate the production quantity by ferroalloy product type for 2011 through 2017 (USGS 2013, 2014, 2015b, 2016b, 2017, 2018).

Table 4-77: Production of Ferroalloys (Metric Tons)

Year	Ferrosilicon 25%-55%	Ferrosilicon 56%-95%	Silicon Metal	Misc. Alloys 32-65%
1990	321,385	109,566	145,744	72,442
2005	123,000	86,100	148,000	NA
2013	164,229	144,908	158,862	NA

2014	176,161	155,436	170,404	NA
2015	180,372	159,151	174,477	NA
2016	165,282	145,837	159,881	NA
2017	181,775	160,390	175,835	NA

NA - Not Available for product type, aggregated along with ferrosilicon (25-55% Si)

Uncertainty and Time-Series Consistency

Annual ferroalloy production was reported by the USGS in three broad categories until the 2010 publication: ferroalloys containing 25 to 55 percent silicon (including miscellaneous alloys), ferroalloys containing 56 to 95 percent silicon, and silicon metal (through 2005 only, 2005 value used as proxy for 2005 through 2010). Starting with the *2011 Minerals Yearbook*, USGS started reporting all the ferroalloy production under a single category: total silicon materials production. The total silicon materials quantity was allocated across the three categories based on the 2010 production shares for the three categories. Refer to the Methodology section for further details. Additionally, production data for silvery pig iron (alloys containing less than 25 percent silicon) are not reported by the USGS to avoid disclosing proprietary company data. Emissions from this production category, therefore, were not estimated.

Also, some ferroalloys may be produced using wood or other biomass as a primary or secondary carbon source (carbonaceous reductants), however information and data regarding these practices were not available. Emissions from ferroalloys produced with wood or other biomass would not be counted under this source because wood-based carbon is of biogenic origin.[230] Even though emissions from ferroalloys produced with coking coal or graphite inputs would be counted in national trends, they may be generated with varying amounts of CO_2 per unit of ferroalloy produced. The most accurate method for these estimates would be to base calculations on the amount of reducing agent used in the process, rather than the amount of ferroalloys produced. These data, however, were not available, and are also often considered confidential business information.

Emissions of CH_4 from ferroalloy production will vary depending on furnace specifics, such as type, operation technique, and control technology. Higher heating temperatures and techniques such as sprinkle charging will reduce CH_4 emissions; however, specific furnace information was not available or included in the CH_4 emission estimates.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-78. Ferroalloy production CO_2 emissions from 2017 were estimated to be between 1.7 and 2.2 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below and 12 percent above the emission estimate of 2.0 MMT CO_2 Eq. Ferroalloy production CH_4 emissions were estimated to be between a range of approximately 12 percent below and 12 percent above the emission estimate of 0.01 MMT CO_2 Eq.

Table 4-78: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Ferroalloy Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Ferroalloy Production	CO_2	2.0	1.7	2.2	-12%	+12%
Ferroalloy Production	CH_4	+	+	+	-12%	+12%

+ Does not exceed 0.05 MMT CO_2 Eq.

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

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[230] Emissions and sinks of biogenic carbon are accounted for in the Land Use, Land-Use Change, and Forestry chapter.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Pending available resources and prioritization of improvements for more significant sources, EPA will continue to evaluate and analyze data reported under EPA's GHGRP that would be useful to improve the emission estimates and category-specific QC procedures for the Ferroalloy Production source category. Given the small number of facilities, particular attention will be made to ensure time-series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[231] This is a long-term planned improvement and EPA is still assessing the possibility of incorporating this improvement into the national Inventory report. This improvement has not been included in the current Inventory report.

4.19 Aluminum Production (CRF Source Category 2C3)

Aluminum is a light-weight, malleable, and corrosion-resistant metal that is used in many manufactured products, including aircraft, automobiles, bicycles, and kitchen utensils. As of recent reporting, the United States was the twelfth largest producer of primary aluminum, with approximately 1 percent of the world total production (USGS 2018). The United States was also a major importer of primary aluminum. The production of primary aluminum—in addition to consuming large quantities of electricity—results in process-related emissions of carbon dioxide (CO_2) and two perfluorocarbons (PFCs): perfluoromethane (CF_4) and perfluoroethane (C_2F_6).

Carbon dioxide is emitted during the aluminum smelting process when alumina (aluminum oxide, Al_2O_3) is reduced to aluminum using the Hall-Heroult reduction process. The reduction of the alumina occurs through electrolysis in a molten bath of natural or synthetic cryolite (Na_3AlF_6). The reduction cells contain a carbon (C) lining that serves as the cathode. Carbon is also contained in the anode, which can be a C mass of paste, coke briquettes, or prebaked C blocks from petroleum coke. During reduction, most of this C is oxidized and released to the atmosphere as CO_2.

Process emissions of CO_2 from aluminum production were estimated to be 1.2 MMT CO_2 Eq. (1,205 kt) in 2017 (see Table 4-79). The C anodes consumed during aluminum production consist of petroleum coke and, to a minor extent, coal tar pitch. The petroleum coke portion of the total CO_2 process emissions from aluminum production is considered to be a non-energy use of petroleum coke, and is accounted for here and not under the CO_2 from Fossil Fuel Combustion source category of the Energy sector. Similarly, the coal tar pitch portion of these CO_2 process emissions is accounted for here.

Table 4-79: CO_2 Emissions from Aluminum Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	6.8	6,831
2005	4.1	4,142
2013	3.3	3,255
2014	2.8	2,833

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[231] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf >.

2015	2.8	2,767
2016	1.3	1,334
2017	1.2	1,205

In addition to CO_2 emissions, the aluminum production industry is also a source of PFC emissions. During the smelting process, when the alumina ore content of the electrolytic bath falls below critical levels required for electrolysis, rapid voltage increases occur, which are termed “anode effects.” These anode effects cause C from the anode and fluorine from the dissociated molten cryolite bath to combine, thereby producing fugitive emissions of CF_4 and C_2F_6. In general, the magnitude of emissions for a given smelter and level of production depends on the frequency and duration of these anode effects. As the frequency and duration of the anode effects increase, emissions increase.

Since 1990, emissions of CF_4 and C_2F_6 have declined by 96 percent and 89 percent, respectively, to 0.7 MMT CO_2 Eq. of CF_4 (0.1 kt) and 0.4 MMT CO_2 Eq. of C_2F_6 (0.03 kt) in 2017, as shown in Table 4-80 and Table 4-81. This decline is due both to reductions in domestic aluminum production and to actions taken by aluminum smelting companies to reduce the frequency and duration of anode effects. These actions include technology and operational changes such as employee training, use of computer monitoring, and changes in alumina feeding techniques. Since 1990, aluminum production has declined by 82 percent, while the combined CF_4 and C_2F_6 emission rate (per metric ton of aluminum produced) has been reduced by 72 percent. Emissions decreased by approximately 18 percent between 2016 and 2017 due to decreases in aluminum production. CF_4 and C_2F_6 emissions per metric ton of aluminum produced decreased between 2016 and 2017.

Table 4-80: PFC Emissions from Aluminum Production (MMT CO_2 Eq.)

Year	CF_4	C_2F_6	Total
1990	17.9	3.5	21.5
2005	2.9	0.6	3.4
2013	2.3	0.7	3.0
2014	1.9	0.6	2.5
2015	1.5	0.5	2.0
2016	0.9	0.4	1.4
2017	0.7	0.4	1.1

Note: Totals may not sum due to independent rounding.

Table 4-81: PFC Emissions from Aluminum Production (kt)

Year	CF_4	C_2F_6
1990	2.4	0.3
2005	0.4	+
2013	0.3	0.1
2014	0.3	0.1
2015	0.2	+
2016	0.1	+
2017	0.1	+

+ Does not exceed 0.05 kt.

In 2017, U.S. primary aluminum production totaled approximately 0.7 million metric tons, a 9 percent decrease from 2016 production levels (USAA 2018). In 2017, two companies managed production at five operational primary aluminum smelters. One smelter that previously announced a permanent shutdown changed its status to temporarily shut down, and plans to start production again in 2018. Three smelters remained on standby throughout 2017 (USGS 2018). During 2017, monthly U.S. primary aluminum production was lower for every month in 2016 except August, October, and December when compared to the corresponding months in 2016 (USAA 2018, 2017).

For 2018, total production for the January to August period was approximately 0.55 million metric tons compared to 0.49 million metric tons for the same period in 2017, a 10 percent increase (USAA 2018). Based on the increase in production, process CO_2 and PFC emissions are likely to be higher in 2018 compared to 2017 if there are no significant changes in process controls at operational facilities.

Methodology

Process CO_2 and PFC (i.e., CF_4 and C_2F_6) emission estimates from primary aluminum production for 2010 through 2017 are available from EPA's GHGRP—Subpart F (Aluminum Production) (EPA 2018). Under EPA's GHGRP, facilities began reporting primary aluminum production process emissions (for 2010) in 2011; as a result, GHGRP data (for 2010 through 2017) are available to be incorporated into the Inventory. EPA's GHGRP mandates that all facilities that contain an aluminum production process must report: CF_4 and C_2F_6 emissions from anode effects in all prebake and Søderberg electrolysis cells, CO_2 emissions from anode consumption during electrolysis in all prebake and Søderberg cells, and all CO_2 emissions from onsite anode baking. To estimate the process emissions, EPA's GHGRP uses the process-specific equations detailed in subpart F (aluminum production).[232] These equations are based on the Tier 2/Tier 3 IPCC (2006) methods for primary aluminum production, and Tier 1 methods when estimating missing data elements. It should be noted that the same methods (i.e., *2006 IPCC Guidelines*) were used for estimating the emissions prior to the availability of the reported GHGRP data in the Inventory. Prior to 2010, aluminum production data were provided through EPA's Voluntary Aluminum Industrial Partnership (VAIP).

As previously noted, the use of petroleum coke for aluminum production is adjusted for within the Energy chapter as this fuel was consumed during non-energy related activities. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

Process CO_2 Emissions from Anode Consumption and Anode Baking

Carbon dioxide emission estimates for the years prior to the introduction of EPA's GHGRP in 2010 were estimated *2006 IPCC Guidelines* methods, but individual facility reported data were combined with process-specific emissions modeling. These estimates were based on information previously gathered from EPA's Voluntary Aluminum Industrial Partnership (VAIP) program, U.S. Geological Survey (USGS) Mineral Commodity reviews, and The Aluminum Association (USAA) statistics, among other sources. Since pre- and post-GHGRP estimates use the same methodology, emission estimates are comparable across the time series.

Most of the CO_2 emissions released during aluminum production occur during the electrolysis reaction of the C anode, as described by the following reaction:

$$2Al_2O_3 + 3C \rightarrow 4Al + 3CO_2$$

For prebake smelter technologies, CO_2 is also emitted during the anode baking process. These emissions can account for approximately 10 percent of total process CO_2 emissions from prebake smelters.

Depending on the availability of smelter-specific data, the CO_2 emitted from electrolysis at each smelter was estimated from: (1) the smelter's annual anode consumption, (2) the smelter's annual aluminum production and rate of anode consumption (per ton of aluminum produced) for previous and/or following years, or (3) the smelter's annual aluminum production and IPCC default CO_2 emission factors. The first approach tracks the consumption and carbon content of the anode, assuming that all C in the anode is converted to CO_2. Sulfur, ash, and other impurities in the anode are subtracted from the anode consumption to arrive at a C consumption figure. This approach corresponds to either the IPCC Tier 2 or Tier 3 method, depending on whether smelter-specific data on anode impurities are used. The second approach interpolates smelter-specific anode consumption rates to estimate emissions during years for which anode consumption data are not available. This approach avoids substantial errors and discontinuities that could be introduced by reverting to Tier 1 methods for those years. The last approach

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[232] Code of Federal Regulations, Title 40: Protection of Environment, Part 98: Mandatory Greenhouse Gas Reporting, Subpart F—Aluminum Production. See <www.epa.gov/ghgreporting/documents/pdf/infosheets/aluminumproduction.pdf>.

corresponds to the IPCC Tier 1 method (IPCC 2006), and is used in the absence of present or historic anode consumption data.

The equations used to estimate CO_2 emissions in the Tier 2 and 3 methods vary depending on smelter type (IPCC 2006). For Prebake cells, the process formula accounts for various parameters, including net anode consumption, and the sulfur, ash, and impurity content of the baked anode. For anode baking emissions, the formula accounts for packing coke consumption, the sulfur and ash content of the packing coke, as well as the pitch content and weight of baked anodes produced. For Søderberg cells, the process formula accounts for the weight of paste consumed per metric ton of aluminum produced, and pitch properties, including sulfur, hydrogen, and ash content.

Through the VAIP, anode consumption (and some anode impurity) data have been reported for 1990, 2000, 2003, 2004, 2005, 2006, 2007, 2008, and 2009. Where available, smelter-specific process data reported under the VAIP were used; however, if the data were incomplete or unavailable, information was supplemented using industry average values recommended by IPCC (2006). Smelter-specific CO_2 process data were provided by 18 of the 23 operating smelters in 1990 and 2000, by 14 out of 16 operating smelters in 2003 and 2004, 14 out of 15 operating smelters in 2005, 13 out of 14 operating smelters in 2006, 5 out of 14 operating smelters in 2007 and 2008, and 3 out of 13 operating smelters in 2009. For years where CO_2 emissions data or CO_2 process data were not reported by these companies, estimates were developed through linear interpolation, and/or assuming representative (e.g., previously reported or industry default) values.

In the absence of any previous historical smelter-specific process data (i.e., 1 out of 13 smelters in 2009; 1 out of 14 smelters in 2006, 2007, and 2008; 1 out of 15 smelters in 2005; and 5 out of 23 smelters between 1990 and 2003), CO_2 emission estimates were estimated using Tier 1 Søderberg and/or Prebake emission factors (metric ton of CO_2 per metric ton of aluminum produced) from IPCC (2006).

Process PFC Emissions from Anode Effects

Smelter-specific PFC emissions from aluminum production for 2010 through 2017 were reported to EPA under its GHGRP. To estimate their PFC emissions and report them under EPA's GHGRP, smelters use an approach identical to the Tier 3 approach in the *2006 IPCC Guidelines* (IPCC 2006). Specifically, they use a smelter-specific slope coefficient as well as smelter-specific operating data to estimate an emission factor using the following equation:

$$PFC = S \times AE$$

$$AE = F \times D$$

where,

PFC = CF_4 or C_2F_6, kg/MT aluminum
S = Slope coefficient, PFC/AE
AE = Anode effect, minutes/cell-day
F = Anode effect frequency per cell-day
D = Anode effect duration, minutes

They then multiply this emission factor by aluminum production to estimate PFC emissions. All U.S. aluminum smelters are required to report their emissions under EPA's GHGRP.

Perfluorocarbon emissions for the years prior to 2010 were estimated using the same equation, but the slope-factor used for some smelters was technology-specific rather than smelter-specific, making the method a Tier 2 rather than a Tier 3 approach for those smelters. Emissions and background data were reported to EPA under the VAIP. For 1990 through 2009, smelter-specific slope coefficients were available and were used for smelters representing between 30 and 94 percent of U.S. primary aluminum production. The percentage changed from year to year as some smelters closed or changed hands and as the production at remaining smelters fluctuated. For smelters that did not report smelter-specific slope coefficients, IPCC technology-specific slope coefficients were applied (IPCC 2006). The slope coefficients were combined with smelter-specific anode effect data collected by aluminum companies and reported under the VAIP to estimate emission factors over time. For 1990 through 2009, smelter-specific anode effect data were available for smelters representing between 80 and 100 percent of U.S. primary aluminum production. Where smelter-specific anode effect data were not available, representative values (e.g., previously reported or industry averages) were used.

For all smelters, emission factors were multiplied by annual production to estimate annual emissions at the smelter level. For 1990 through 2009, smelter-specific production data were available for smelters representing between 30 and 100 percent of U.S. primary aluminum production. (For the years after 2000, this percentage was near the high end of the range.) Production at non-reporting smelters was estimated by calculating the difference between the production reported under VAIP and the total U.S. production supplied by USGS or USAA, and then allocating this difference to non-reporting smelters in proportion to their production capacity. Emissions were then aggregated across smelters to estimate national emissions.

Between 1990 and 2009, production data were provided under the VAIP by 21 of the 23 U.S. smelters that operated during at least part of that period. For the non-reporting smelters, production was estimated based on the difference between reporting smelters and national aluminum production levels (USGS and USAA 1990 through 2009), with allocation to specific smelters based on reported production capacities (USGS 1990 through 2009).

National primary aluminum production data for 2017 were obtained via USAA (USAA 2018). For 1990 through 2001, and 2006 (see Table 4-82) data were obtained from USGS *Mineral Industry Surveys: Aluminum Annual Report* (USGS 1995, 1998, 2000, 2001, 2002, 2007). For 2002 through 2005, and 2007 through 2016, national aluminum production data were obtained from the USAA's *Primary Aluminum Statistics* (USAA 2004 through 2006, 2008 through 2017).

Table 4-82: Production of Primary Aluminum (kt)

Year	kt
1990	4,048
2005	2,478
2013	1,948
2014	1,710
2015	1,587
2016	818
2017	741

Uncertainty and Time-Series Consistency

Uncertainty was assigned to the CO_2, CF_4, and C_2F_6 emission values reported by each individual facility to EPA's GHGRP. As previously mentioned, the methods for estimating emissions for EPA's GHGRP and this report are the same, and follow the *2006 IPCC Guidelines* methodology. As a result, it was possible to assign uncertainty bounds (and distributions) based on an analysis of the uncertainty associated with the facility-specific emissions estimated for previous Inventory years. Uncertainty surrounding the reported CO_2, CF_4, and C_2F_6 emission values were determined to have a normal distribution with uncertainty ranges of ±6, ±16, and ±20 percent, respectively. A Monte Carlo analysis was applied to estimate the overall uncertainty of the CO_2, CF_4, and C_2F_6 emission estimates for the U.S. aluminum industry as a whole, and the results are provided below.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-83. Aluminum production-related CO_2 emissions were estimated to be between 1.17 and 1.24 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 3 percent below to 3 percent above the emission estimate of 1.2 MMT CO_2 Eq. Also, production-related CF_4 emissions were estimated to be between 0.7 and 0.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 10 percent below to 10 percent above the emission estimate of 0.7 MMT CO_2 Eq. Finally, aluminum production-related C_2F_6 emissions were estimated to be between 0.3 and 0.4 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 16 percent below to 17 percent above the emission estimate of 0.4 MMT CO_2 Eq.

Table 4-83: Approach 2 Quantitative Uncertainty Estimates for CO_2 and PFC Emissions from Aluminum Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Aluminum Production	CO_2	1.2	1.2	1.2	−3%	+3%
Aluminum Production	CF_4	0.7	0.7	0.8	−10%	+10%
Aluminum Production	C_2F_6	0.4	0.3	0.4	−16%	+17%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time-series to ensure time-series consistency from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

4.20 Magnesium Production and Processing (CRF Source Category 2C4)

The magnesium metal production and casting industry uses sulfur hexafluoride (SF_6) as a cover gas to prevent the rapid oxidation of molten magnesium in the presence of air. Sulfur hexafluoride has been used in this application around the world for more than thirty years. A dilute gaseous mixture of SF_6 with dry air and/or carbon dioxide (CO_2) is blown over molten magnesium metal to induce and stabilize the formation of a protective crust. A small portion of the SF_6 reacts with the magnesium to form a thin molecular film of mostly magnesium oxide and magnesium fluoride. The amount of SF_6 reacting in magnesium production and processing is considered to be negligible and thus all SF_6 used is assumed to be emitted into the atmosphere. Alternative cover gases, such as AM-cover™ (containing HFC-134a), Novec™ 612 (FK-5-1-12) and dilute sulfur dioxide (SO_2) systems can, and are being used by some facilities in the United States. However, many facilities in the United States are still using traditional SF_6 cover gas systems.

The magnesium industry emitted 1.1 MMT CO_2 Eq. (0.05 kt) of SF_6, 0.1 MMT CO_2 Eq. (0.1 kt) of HFC-134a, and 0.003 MMT CO_2 Eq. (3.1 kt) of CO_2 in 2017. This represents a decrease of approximately 4 percent from total 2016 emissions (see Table 4-84) and a decrease in SF_6 emissions by 5 percent. The decrease can be attributed to decrease in secondary production SF_6 emissions between 2016 and 2017 as reported through the GHGRP. In 2017, total HFC-134a emissions increased from 0.096 MMT CO_2 Eq. to 0.098 MMT CO_2 Eq., or a 2 percent increase as compared to 2016 emissions. This is mainly attributable to the increased use of this alternative for secondary production. FK 5-1-12 emissions did not change substantially from 2016 levels. The emissions of the carrier gas, CO_2, increased from 2.7 kt in 2016 to 3.1 kt in 2017, or 14 percent.

Table 4-84: SF_6, HFC-134a, FK 5-1-12 and CO_2 Emissions from Magnesium Production and Processing (MMT CO_2 Eq.)

Year	1990	2005	2013	2014	2015	2016	2017
SF_6	5.2	2.7	1.3	0.9	1.0	1.1	1.1
HFC-134a	0.0	0.0	0.1	0.1	0.1	0.1	0.1
CO_2	+	+	+	+	+	+	+
FK 5-1-12[a]	*0.0*	*0.0*	+	+	+	+	+
Total	**5.2**	**2.7**	**1.4**	**1.0**	**1.1**	**1.2**	**1.2**

+ Does not exceed 0.05 MMT CO_2 Eq.

[a] Emissions of FK 5-1-12 are not included in totals.

Table 4-85: SF_6, HFC-134a, FK 5-1-12 and CO_2 Emissions from Magnesium Production and Processing (kt)

Year	1990	2005	2013	2014	2015	2016	2017
SF_6	0.2	0.1	0.1	+	+	+	+
HFC-134a	0.0	0.0	0.1	0.1	0.1	0.1	0.1
CO_2	1.4	2.9	2.1	2.3	2.6	2.7	3.1
FK 5-1-12 [a]	*0.0*	*0.0*	+	+	+	+	+

+ Does not exceed 0.05 kt

[a] Emissions of FK 5-1-12 are not included in totals.

Methodology

Emission estimates for the magnesium industry incorporate information provided by industry participants in EPA's SF_6 Emission Reduction Partnership for the Magnesium Industry as well as emissions data reported through subpart T (Magnesium Production and Processing) of EPA's GHGRP. The Partnership started in 1999 and, in 2010, participating companies represented 100 percent of U.S. primary and secondary production and 16 percent of the casting sector production (i.e., die, sand, permanent mold, wrought, and anode casting). SF_6 emissions for 1999 through 2010 from primary production, secondary production (i.e., recycling), and die casting were generally reported by Partnership participants. Partners reported their SF_6 consumption, which is assumed to be equivalent to emissions. Along with SF_6, some Partners also reported their HFC-134a and FK 5-1-12 usage, which is also assumed to be equal to emissions. The last reporting year was 2010 under the Partnership. Emissions data for 2011 through 2017 are obtained through EPA's GHGRP. Under the program, owners or operators of facilities that have a magnesium production or casting process must report emissions from use of cover or carrier gases, which include SF_6, HFC-134a, FK 5-1-12 and CO_2. Consequently, cover and carrier gas emissions from magnesium production and processing were estimated for three time periods, depending on the source of the emissions data: 1990 through 1998 (pre-EPA Partnership), 1999 through 2010 (EPA Partnership), and 2011 through 2017 (EPA GHGRP). The methodologies described below also make use of magnesium production data published by the U.S. Geological Survey (USGS).

1990 through 1998

To estimate emissions for 1990 through 1998, industry SF_6 emission factors were multiplied by the corresponding metal production and consumption (casting) statistics from USGS. For this period, it was assumed that there was no use of HFC-134a or FK 5-1-12 cover gases and hence emissions were not estimated for these alternatives.

Sulfur hexafluoride emission factors from 1990 through 1998 were based on a number of sources and assumptions. Emission factors for primary production were available from U.S. primary producers for 1994 and 1995. The primary production emission factors were 1.2 kg SF_6 per metric ton for 1990 through 1993, and 1.1 kg SF_6 per metric ton for 1994 through 1997. The emission factor for secondary production from 1990 through 1998 was assumed to be constant at the 1999 average Partner value. An emission factor for die casting of 4.1 kg SF_6 per metric ton, which was available for the mid-1990s from an international survey (Gjestland and Magers 1996), was used for years 1990 through 1996. For 1996 through 1998, the emission factor for die casting was assumed to decline linearly to the level estimated based on Partner reports in 1999. This assumption is consistent with the trend in SF_6 sales to the magnesium sector that is reported in the RAND survey of major SF_6 manufacturers, which shows a decline of 70 percent from 1996 to 1999 (RAND 2002). Sand casting emission factors for 1990 through 2001 were assumed to be the same as the 2002 emission factor. The emission factors for the other processes (i.e., permanent mold, wrought, and anode casting), about which less is known, were assumed to remain constant at levels defined in Table 4-84. These emission factors for the other processes (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives.

The quantities of CO_2 carrier gas used for each production type have been estimated using the 1999 estimated CO_2 emissions data and the annual calculated rate of change of SF_6 use in the 1990 through 1999 time period. For each year and production type, the rate of change of SF_6 use between the current year and the subsequent year was first estimated. This rate of change is then applied to the CO_2 emissions of the subsequent year to determine the CO_2 emission of the current year. The emissions of carrier gases for permanent mold, wrought and anode processes are not estimated in this Inventory.

1999 through 2010

The 1999 through 2010 emissions from primary and secondary production are based on information provided by EPA's industry Partners. In some instances, there were years of missing Partner data, including SF_6 consumption and metal processed. For these situations, emissions were estimated through interpolation where possible, or by holding company-reported emissions (as well as production) constant from the previous year. For alternative cover gases, including HFC-134a and FK 5-1-12, mainly reported data was relied upon. That is, unless a Partner reported using an alternative cover gas, it was not assumed it was used. Emissions of alternate gases were also estimated through linear interpolation where possible.

The die casting emission estimates for 1999 through 2010 are also based on information supplied by industry Partners. When a Partner was determined to be no longer in production, its metal production and usage rates were set to zero. Missing data on emissions or metal input was either interpolated or held constant at the last available reported value. In 1999 through 2008 through 2010, Partners did not account for all die casting tracked by USGS, and, therefore, it was necessary to estimate the emissions of die casters who were not Partners. For 1999, die casters who were not Partners were assumed to be similar to Partners who cast small parts. Due to process requirements, these casters consume larger quantities of SF_6 per metric ton of processed magnesium than casters that process large parts. Consequently, emission estimates from this group of die casters were developed using an average emission factor of 5.2 kg SF_6 per metric ton of magnesium. This emission factor was developed using magnesium production and SF_6 usage data for the year 1999.

The emissions from other casting operations were estimated by multiplying emission factors (kg SF_6 per metric ton of metal produced or processed) by the amount of metal produced or consumed from USGS, with the exception of some years for which Partner sand casting emissions data are available. The emission factors for sand casting activities were acquired through the data reported by the Partnership for 2002 to 2006. For 1999-2001, the sand casting emission factor was held constant at the 2002 Partner-reported level. For 2007 through 2010, the sand casting Partner did not report and the reported emission factor from 2005 was applied to the Partner and to all other sand casters. Activity data for 2005 was obtained from USGS (USGS 2005b).

The emission factors for primary production, secondary production and sand casting for the 1999 to 2010 are not published to protect company-specific production information. However, the emission factor for primary production has not risen above the average 1995 Partner value of 1.1 kg SF_6 per metric ton. The emission factors for the other industry sectors (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives. The emission factors for casting activities are provided below in Table 4-86.

The emissions of HFC-134a and FK-5-1-12 were included in the estimates for only instances where Partners reported that information to the Partnership. Emissions of these alternative cover gases were not estimated for instances where emissions were not reported.

Carbon dioxide carrier gas emissions were estimated using the emission factors developed based on GHGRP-reported carrier gas and cover gas data, by production type. It was assumed that the use of carrier gas, by production type, is proportional to the use of cover gases. Therefore, an emission factor, in kg CO_2 per kg cover gas and weighted by the cover gases used, was developed for each of the production types. GHGRP data on which these emissions factors are based was available for primary, secondary, die casting and sand casting. The emission factors were applied to the total quantity of all cover gases used (SF_6, HFC-134a, and FK-5-1-12) by production type in this time period. Carrier gas emissions for the 1999 through 2010 time period were only estimated for those Partner companies that reported using CO_2 as a carrier gas through the GHGRP. Using this approach helped ensure time-series consistency. The emissions of carrier gases for permanent mold, wrought and anode processes are not estimated in this Inventory.

Table 4-86: SF_6 Emission Factors (kg SF_6 per metric ton of magnesium)

Year	Die Casting[a]	Permanent Mold	Wrought	Anodes
1999	1.75[b]	2	1	1
2000	0.72	2	1	1
2001	0.72	2	1	1
2002	0.71	2	1	1
2003	0.81	2	1	1
2004	0.79	2	1	1
2005	0.77	2	1	1
2006	0.88	2	1	1
2007	0.64	2	1	1
2008	0.97	2	1	1
2009	1.41	2	1	1
2010	2.71	2	1	1

[a] Weighted average includes all die casters, Partners and non-Partners. For the majority of the time series (2000 through 2007), Partners made up 100 percent of die casters in the United States.

[b] Weighted average that includes an estimated emission factor of 5.2 kg SF_6 per metric ton of magnesium for die casters that do not participate in the Partnership.

2011 through 2017

For 2011 through 2017, for the primary and secondary producers and die casting, GHGRP-reported cover and carrier gases emissions data were used. For sand casting, some emissions data was obtained through EPA's GHGRP. The balance of the emissions for this industry segment was estimated based on previous Partner reporting (i.e., for Partners that did not report emissions through EPA's GHGRP) or were estimated by multiplying emission factors by the amount of metal produced or consumed. Partners who did not report through EPA's GHGRP were assumed to have continued to emit SF_6 at the last reported level, which was from 2010 in most cases, unless publicly available sources indicated that these facilities have closed or otherwise eliminated SF_6 emissions from magnesium production (ARB 2015). All Partners were assumed to have continued to consume magnesium at the last reported level. Where the total metal consumption estimated for the Partners fell below the U.S. total reported by USGS, the difference was multiplied by the emission factors discussed in the section above, i.e. non-partner emission factors. For the other types of production and processing (i.e., permanent mold, wrought, and anode casting), emissions were estimated by multiplying the industry emission factors with the metal production or consumption statistics obtained from USGS (USGS 2018). USGS data for 2017 was not yet available at the time of the analysis, so the 2016 values were held constant through 2017 as a proxy.

Due to some GHGRP facilities originally submitting their GHGRP reports with errors, reporting their data late, or not submitting data for 2017, some values were held constant at 2016 levels, affecting the overall calculations.

Uncertainty and Time-Series Consistency

Uncertainty surrounding the total estimated emissions in 2017 is attributed to the uncertainties around SF_6, HFC-134a, and CO_2 emission estimates. To estimate the uncertainty surrounding the estimated 2017 SF_6 emissions from magnesium production and processing, the uncertainties associated with three variables were estimated: (1) emissions reported by magnesium producers and processors for 2017 through EPA's GHGRP, (2) emissions estimated for magnesium producers and processors that reported via the Partnership in prior years but did not report 2017 emissions through EPA's GHGRP, and (3) emissions estimated for magnesium producers and processors that did not participate in the Partnership or report through EPA's GHGRP. An uncertainty of 5 percent was assigned to the emissions (usage) data reported by each GHGRP reporter for all the cover and carrier gases (per the *2006 IPCC Guidelines*). If facilities did not report emissions data during the current reporting year through EPA's GHGRP, SF_6 emissions data were held constant at the most recent available value reported through the Partnership. The uncertainty associated with these values was estimated to be 30 percent for each year of extrapolation. In 2017, a higher proportion of emissions were estimated by holding values constant at the previous year's emissions as compared to 2016, so the uncertainty of the 2017 total inventory estimate is relatively higher than it was in 2016.

Alternate cover gas and carrier gases data was set equal to zero if the facilities did not report via the GHGRP. For those industry processes that are not represented in the Partnership, such as permanent mold and wrought casting, SF_6 emissions were estimated using production and consumption statistics reported by USGS and estimated process-specific emission factors (see Table 4-87). The uncertainties associated with the emission factors and USGS-reported statistics were assumed to be 75 percent and 25 percent, respectively. Emissions associated with die casting and sand casting activities utilized emission factors based on Partner reported data with an uncertainty of 75 percent. In general, where precise quantitative information was not available on the uncertainty of a parameter, a conservative (upper-bound) value was used.

Additional uncertainties exist in these estimates that are not addressed in this methodology, such as the basic assumption that SF_6 neither reacts nor decomposes during use. The melt surface reactions and high temperatures associated with molten magnesium could potentially cause some gas degradation. Previous measurement studies have identified SF_6 cover gas degradation in die casting applications on the order of 20 percent (Bartos et al. 2007). Sulfur hexafluoride may also be used as a cover gas for the casting of molten aluminum with high magnesium content; however, the extent to which this technique is used in the United States is unknown.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-87. Total emissions associated with magnesium production and processing were estimated to be between 1.05 and 1.21 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below to 7 percent above the 2017 emission estimate of 1.1 MMT CO_2 Eq. The uncertainty estimates for 2017 are larger relative to the uncertainty reported for 2016 in the previous Inventory. This is because, as discussed above, a larger proportion of emissions from GHGRP reporters in 2017 were set equal to 2016 reported emissions due to late or non-verified GHGRP reports.

Table 4-87: Approach 2 Quantitative Uncertainty Estimates for SF_6, HFC-134a and CO_2 Emissions from Magnesium Production and Processing (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	**Upper Bound**	**Lower Bound**	**Upper Bound**
Magnesium Production	SF_6, HFC-134a, CO_2	1.1	1.1	1.2	-7%	+7%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure time-series consistency from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Recalculations Discussion

One GHGRP-reported value for 2016 was revised due to a data verification issue. Additionally, the USGS revised some of its production numbers for 2015, resulting in changes in SF_6 emissions for die casting, sand casting, and permanent mold. Lastly, based upon a review of historical activity data from various sources, EPA revised estimates of non-Partner or non-GHGRP reporter die casting activity data to be zero metal produced from 2008 through 2017.

Planned Improvements

Cover gas research conducted over the last decade has found that SF_6 used for magnesium melt protection can have degradation rates on the order of 20 percent in die casting applications (Bartos et al. 2007). Current emission estimates assume (per the *2006 IPCC Guidelines*) that all SF_6 utilized is emitted to the atmosphere. Additional research may lead to a revision of the *2006 IPCC Guidelines* to reflect this phenomenon and until such time, developments in this sector will be monitored for possible application to the Inventory methodology.

Usage and emission details of carrier gases in permanent mold, wrought and anode processes will be researched as part of a future Inventory. Based on this research, it will be determined if CO_2 carrier gas emissions are to be estimated.

Additional emissions are generated as byproducts from the use of alternate cover gases, which are not currently accounted for. Research on this topic is developing, and as reliable emission factors become available, these emissions will be incorporated into the Inventory.

4.21 Lead Production (CRF Source Category 2C5)

In 2017, lead was produced in the United States only using secondary production processes. Until 2014, both lead production in the United States involved both primary and secondary processes—both of which emit carbon dioxide (CO_2) (Sjardin 2003). Emissions from fuels consumed for energy purposes during the production of lead are accounted for in the Energy chapter.

Primary production of lead through the direct smelting of lead concentrate produces CO_2 emissions as the lead concentrates are reduced in a furnace using metallurgical coke (Sjardin 2003). Primary lead production, in the form of direct smelting, previously occurred at a single smelter in Missouri. This primary lead smelter was closed at the end of 2013. In 2014, the smelter processed a small amount of residual lead during demolition of the site (USGS 2015) and in 2017 the smelter processed no lead (USGS 2016, 2017).

Similar to primary lead production, CO_2 emissions from secondary lead production result when a reducing agent, usually metallurgical coke, is added to the smelter to aid in the reduction process. Carbon dioxide emissions from secondary production also occur through the treatment of secondary raw materials (Sjardin 2003). Secondary production primarily involves the recycling of lead acid batteries and post-consumer scrap at secondary smelters. Of all the domestic secondary smelters operational in 2017, 11 smelters had capacities of 30,000 tons or more and were collectively responsible for more than 95 percent of secondary lead production in 2017 (USGS 2017). Secondary lead production has increased in the United States over the past decade while primary lead production has decreased to production levels of zero. In 2017, secondary lead production accounted for 100 percent of total lead production. The lead-acid battery industry accounted for more than 85 percent of the reported U.S. lead consumption in 2017 (USGS 2017).

In 2017, total secondary lead production in the United States was slightly higher than that in 2016. A new secondary lead refinery, located in Nevada, was completed in 2016 and production was expected to begin by the end of the year. The plant was expected to produce about 80 tons per day of high-purity refined lead for use in advanced lead-acid batteries using an electromechanical battery recycling technology system. The United States has become more reliant on imported refined lead in recent years owing to the closure of the last primary lead smelter in 2013, and to an increase in exports of spent starting-lighting-ignition lead-acid batteries that reduced the availability of scrap for secondary smelters (USGS 2017).

As in 2016, U.S. primary lead production remained at production levels of zero for 2017, and has also decreased by 100 percent since 1990. This is due to the closure of the only domestic primary lead smelter in 2013 (year-end), as stated previously. In 2017, U.S. secondary lead production increased from 2016 levels (increase of 7 percent), and has increased by 16 percent since 1990 (USGS 1995 through 2017).

In 2017, U.S. lead production totaled 1,010,000 metric tons (USGS 2018). The resulting emissions of CO_2 from 2017 lead production were estimated to be 0.5 MMT CO_2 Eq. (455 kt) (see Table 4-88). At last reporting, the United States was the third largest mine producer of lead in the world, behind China and Australia, accounting for approximately 7 percent of world production in 2017 (USGS 2017).

Table 4-88: CO_2 Emissions from Lead Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	0.5	516
2005	0.6	553
2013	0.5	546
2014	0.5	459
2015	0.5	473
2016	0.5	450
2017	0.5	455

After a steady increase in total emissions from 1995 to 2000, total emissions have gradually decreased since 2000 and are currently 7 percent lower than 1990 levels.

Methodology

The methods used to estimate emissions for lead production[233] are based on Sjardin's work (Sjardin 2003) for lead production emissions and Tier 1 methods from the *2006 IPCC Guidelines*. The Tier 1 equation is as follows:

$$CO_2\ Emissions = (DS \times EF_{DS}) + (S \times EF_S)$$

where,

DS = Lead produced by direct smelting, metric ton
S = Lead produced from secondary materials
EF_{DS} = Emission factor for direct Smelting, metric tons CO_2/metric ton lead product
EF_S = Emission factor for secondary materials, metric tons CO_2/metric ton lead product

For primary lead production using direct smelting, Sjardin (2003) and the IPCC (2006) provide an emission factor of 0.25 metric tons CO_2/metric ton lead. For secondary lead production, Sjardin (2003) and IPCC (2006) provide an emission factor of 0.25 metric tons CO_2/metric ton lead for direct smelting, as well as an emission factor of 0.2 metric tons CO_2/metric ton lead produced for the treatment of secondary raw materials (i.e., pretreatment of lead acid batteries). Since the secondary production of lead involves both the use of the direct smelting process and the treatment of secondary raw materials, Sjardin recommends an additive emission factor to be used in conjunction with the secondary lead production quantity. The direct smelting factor (0.25) and the sum of the direct smelting and pretreatment emission factors (0.45) are multiplied by total U.S. primary and secondary lead production, respectively, to estimate CO_2 emissions.

The production and use of coking coal for lead production is adjusted for within the Energy chapter as this fuel was consumed during non-energy related activities. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

The 1990 through 2017 activity data for primary and secondary lead production (see Table 4-89) were obtained from the U.S. Geological Survey (USGS 1995 through 2018). The 2016 lead production value was also updated and is summarized in Table 4-89 (USGS 2018).

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[233] EPA has not integrated aggregated facility-level Greenhouse Gas Reporting Program (GHGRP) information to inform these estimates. The aggregated information (e.g., activity data and emissions) associated with Lead Production did not meet criteria to shield underlying confidential business information (CBI) from public disclosure.

Table 4-89: Lead Production (Metric Tons)

Year	Primary	Secondary
1990	404,000	922,000
2005	143,000	1,150,000
2013	114,000	1,150,000
2014	1,000	1,020,000
2015	0	1,050,000
2016	0	1,000,000
2017	0	1,010,000

Uncertainty and Time-Series Consistency

Uncertainty associated with lead production relates to the emission factors and activity data used. The direct smelting emission factor used in primary production is taken from Sjardin (2003) who averaged the values provided by three other studies (Dutrizac et al. 2000; Morris et al. 1983; Ullman 1997). For secondary production, Sjardin (2003) added a CO_2 emission factor associated with battery treatment. The applicability of these emission factors to plants in the United States is uncertain. There is also a smaller level of uncertainty associated with the accuracy of primary and secondary production data provided by the USGS which is collected via voluntary surveys; the uncertainty of the activity data is a function of the reliability of reported plant-level production data and the completeness of the survey response.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-90. Lead production CO_2 emissions in 2017 were estimated to be between 0.4 and 0.5 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 15 percent below and 15 percent above the emission estimate of 0.5 MMT CO_2 Eq.

Table 4-90: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Lead Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		Uncertainty Range Relative to Emission Estimate[a] (%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Lead Production	CO_2	0.5	0.4	0.5	-15%	+15%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches discussed below were applied to applicable years to ensure time-series consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Pending resources and prioritization of improvements for more significant sources, EPA will continue to evaluate and analyze data reported under EPA's GHGRP that would be useful to improve the emission estimates and category-specific QC for the Lead Production source category, in particular considering completeness of reported lead production given the reporting threshold. Particular attention will be made to ensuring time-series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[234]

Initial review of activity data show that EPA's GHGRP Subpart R lead production data differ from those reported by USGS by between 2 percent and 18 percent across the 2012 through 2017 time-series. Preliminary emissions estimates differ by roughly the same percentages (ranging from 2 to 18 percent) across the same time period. EPA is still reviewing available GHGRP data, differences in data reporting, and assessing the possibility of including this planned improvement in future Inventory reports. Currently, GHGRP data is used for QA purposes and EPA expects the earliest to begin incorporating GHGRP data would be the 2020 Inventory submission.

4.22 Zinc Production (CRF Source Category 2C6)

Zinc production in the United States consists of both primary and secondary processes. Of the primary and secondary processes used in the United States, only the electrothermic and Waelz kiln secondary processes result in non-energy carbon dioxide (CO_2) emissions (Viklund-White 2000). Emissions from fuels consumed for energy purposes during the production of zinc are accounted for in the Energy chapter.

The majority of zinc produced in the United States is used for galvanizing. Galvanizing is a process where zinc coating is applied to steel in order to prevent corrosion. Zinc is used extensively for galvanizing operations in the automotive and construction industry. Zinc is also used in the production of zinc alloys and brass and bronze alloys (e.g., brass mills, copper foundries, and copper ingot manufacturing). Zinc compounds and dust are also used, to a lesser extent, by the agriculture, chemicals, paint, and rubber industries.

Primary production in the United States is conducted through the electrolytic process, while secondary techniques include the electrothermic and Waelz kiln processes, as well as a range of other metallurgical, hydrometallurgical, and pyrometallurgical processes. Worldwide primary zinc production also employs a pyrometallurgical process using the Imperial Smelting Furnace process; however, this process is not used in the United States (Sjardin 2003).

In the electrothermic process, roasted zinc concentrate and secondary zinc products enter a sinter feed where they are burned to remove impurities before entering an electric retort furnace. Metallurgical coke is added to the electric retort furnace as a carbon-containing reductant. This concentration step, using metallurgical coke and high temperatures, reduces the zinc oxides and produces vaporized zinc, which is then captured in a vacuum condenser. This reduction process also generates non-energy CO_2 emissions.

$$ZnO + C \rightarrow Zn(gas) + CO_2 \qquad \text{(Reaction 1)}$$

$$ZnO + CO \rightarrow Zn(gas) + CO_2 \qquad \text{(Reaction 2)}$$

In the Waelz kiln process, electric arc furnace (EAF) dust, which is captured during the recycling of galvanized steel, enters a kiln along with a reducing agent (typically carbon-containing metallurgical coke). When kiln temperatures reach approximately 1,100 to 1,200 degrees Celsius, zinc fumes are produced, which are combusted with air entering the kiln. This combustion forms zinc oxide, which is collected in a baghouse or electrostatic precipitator, and is then leached to remove chloride and fluoride. The use of carbon-containing metallurgical coke in a high-temperature fuming process results in non-energy CO_2 emissions. Through this process, approximately 0.33 metric tons of zinc is produced for every metric ton of EAF dust treated (Viklund-White 2000).

The only companies in the United States that use emissive technology to produce secondary zinc products are American Zinc Recycling (AZR) (formerly "Horsehead Corporation"), PIZO, and Steel Dust Recycling (SDR). For AZR, EAF dust is recycled in Waelz kilns at their Calumet, IL; Palmerton, PA; Rockwood, TN; and Barnwell, SC facilities. These Waelz kiln facilities produce intermediate zinc products (crude zinc oxide or calcine), most of

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[234] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

which was transported to their Monaca, PA facility where the products were smelted into refined zinc using electrothermic technology. In April 2014, AZR permanently shut down their Monaca smelter. This was replaced by their new facility in Mooresboro, NC. The new Mooresboro facility uses a hydrometallurgical process (i.e., solvent extraction with electrowinning technology) to produce zinc products. The current capacity of the new facility is 155,000 short tons, with plans to expand to 170,000 short tons per year. Direct consumption of coal, coke, and natural gas have been replaced with electricity consumption at the new Mooresboro facility. The new facility is reported to have a significantly lower greenhouse gas and other air emissions than the Monaca smelter (Horsehead 2012b).

The Mooresboro facility uses leaching and solvent extraction (SX) technology combined with electrowinning, melting, and casting technology. In this process, Waelz Oxide (WOX) is first washed in water to remove soluble elements such as chlorine, potassium, and sodium, and then is leached in a sulfuric acid solution to dissolve the contained zinc creating a pregnant liquor solution (PLS). The PLS is then processed in a solvent extraction step in which zinc is selectively extracted from the PLS using an organic solvent creating a purified zinc-loaded electrolyte solution. The loaded electrolyte solution is then fed into the electrowinning process in which electrical energy is applied across a series of anodes and cathodes submerged in the electrolyte solution causing the zinc to deposit on the surfaces of the cathodes. As the zinc metal builds up on these surfaces, the cathodes are periodically harvested in order to strip the zinc from their surfaces (Horsehead 2015). Hydrometallurgical production processes are assumed to be non-emissive since no carbon is used in these processes (Sjardin 2003).

PIZO and SDR recycle EAF dust into intermediate zinc products using Waelz kilns, and then sell the intermediate products to companies who smelt it into refined products.

Emissions of CO_2 from zinc production in 2017 were estimated to be 1.0 MMT CO_2 Eq. (1,009 kt CO_2) (see Table 4-91). All 2017 CO_2 emissions resulted from secondary zinc production processes. Emissions from zinc production in the United States have increased overall since 1990 due to a gradual shift from non-emissive primary production to emissive secondary production. In 2017, emissions were estimated to be 60 percent higher than they were in 1990.

Table 4-91: CO_2 Emissions from Zinc Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	0.6	632
2005	1.0	1,030
2013	1.4	1,429
2014	1.0	956
2015	0.9	933
2016	0.9	925
2017	1.0	1,009

In 2017, United States primary and secondary refined zinc production were estimated to total 130,000 metric tons (USGS 2018) (see Table 4-92). Domestic zinc mine production decreased by 9 percent in 2017, owing mostly to the ongoing strike at the Lucky Friday Mine in Idaho and decreased output at the Red Dog Mine in Alaska (USGS 2018). Refined zinc production increased by 6 percent as a result of production resuming at the Middle Tennessee Mines and increased production at the Clarksville, TN smelter (USGS 2018). Primary zinc production (primary slab zinc) increased by five percent in 2017, while secondary zinc production in 2017 decreased by 13 percent relative to 2016.

Table 4-92: Zinc Production (Metric Tons)

Year	Primary	Secondary	Total
1990	262,704	95,708	358,412
2005	191,120	156,000	347,120
2013	106,000	127,000	233,000
2014	110,000	70,000	180,000
2015	125,000	50,000	175,000

2016	111,000	15,000	126,000
2017	117,000	13,000	130,000

Methodology

The methods used to estimate non-energy CO_2 emissions from zinc production[235] using the electrothermic primary production and Waelz kiln secondary production processes are based on Tier 1 methods from the *2006 IPCC Guidelines* (IPCC 2006). The Tier 1 equation used to estimate emissions from zinc production is as follows:

$$E_{CO2} = Zn \times EF_{default}$$

where,

E_{CO2} = CO_2 emissions from zinc production, metric tons
Zn = Quantity of zinc produced, metric tons
$EF_{default}$ = Default emission factor, metric tons CO_2/metric ton zinc produced

The Tier 1 emission factors provided by IPCC for Waelz kiln-based secondary production were derived from coke consumption factors and other data presented in Vikland-White (2000). These coke consumption factors as well as other inputs used to develop the Waelz kiln emission factors are shown below. IPCC does not provide an emission factor for electrothermic processes due to limited information; therefore, the Waelz kiln-specific emission factors were also applied to zinc produced from electrothermic processes. Starting in 2014, refined zinc produced in the United States used hydrometallurgical processes and is assumed to be non-emissive.

For Waelz kiln-based production, IPCC recommends the use of emission factors based on EAF dust consumption, if possible, rather than the amount of zinc produced since the amount of reduction materials used is more directly dependent on the amount of EAF dust consumed. Since only a portion of emissive zinc production facilities consume EAF dust, the emission factor based on zinc production is applied to the non-EAF dust consuming facilities while the emission factor based on EAF dust consumption is applied to EAF dust consuming facilities.

The Waelz kiln emission factor based on the amount of zinc produced was developed based on the amount of metallurgical coke consumed for non-energy purposes per ton of zinc produced (i.e., 1.19 metric tons coke/metric ton zinc produced) (Viklund-White 2000), and the following equation:

$$EF_{Waelz\ Kiln} = \frac{1.19\ metric\ tons\ coke}{metric\ tons\ zinc} \times \frac{0.85\ metric\ tons\ C}{metric\ tons\ coke} \times \frac{3.67\ metric\ tons\ CO_2}{metric\ tons\ C} = \frac{3.70\ metric\ tons\ CO_2}{metric\ tons\ zinc}$$

The Waelz kiln emission factor based on the amount of EAF dust consumed was developed based on the amount of metallurgical coke consumed per ton of EAF dust consumed (i.e., 0.4 metric tons coke/metric ton EAF dust consumed) (Viklund-White 2000), and the following equation:

$$EF_{EAF\ Dust} = \frac{0.4\ metric\ tons\ coke}{metric\ tons\ EAF\ Dust} \times \frac{0.85\ metric\ tons\ C}{metric\ tons\ coke} \times \frac{3.67\ metric\ tons\ CO_2}{metric\ tons\ C} = \frac{1.24\ metric\ tons\ CO_2}{metric\ tons\ EAF\ Dust}$$

The total amount of EAF dust consumed by AZR at their Waelz kilns was available from AZR (formerly "Horsehead Corporation") financial reports for years 2006 through 2015 (Horsehead 2007, 2008, 2010a, 2011, 2012a, 2013, 2014, 2015, and 2016). Total EAF dust consumed by AZR at their Waelz kilns was not available for 2017 so 2015 data was used as proxy. Consumption levels for 1990 through 2005 were extrapolated using the percentage change in annual refined zinc production at secondary smelters in the United States as provided by the U.S. Geological Survey (USGS) *Minerals Yearbook: Zinc* (USGS 1995 through 2006). The EAF dust consumption

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[235] EPA has not integrated aggregated facility-level Greenhouse Gas Reporting Program (GHGRP) information to inform these estimates. The aggregated information (e.g., activity data and emissions) associated with Zinc Production did not meet criteria to shield underlying confidential business information (CBI) from public disclosure.

values for each year were then multiplied by the 1.24 metric tons CO_2/metric ton EAF dust consumed emission factor to develop CO_2 emission estimates for AZR's Waelz kiln facilities.

The amount of EAF dust consumed by SDR and their total production capacity were obtained from SDR's facility in Alabama for the years 2011 through 2017 (SDR 2012, 2014, 2015, and 2017). SDR's facility in Alabama underwent expansion in 2011 to include a second unit (operational since early- to mid-2012). SDR's facility has been operational since 2008. Annual consumption data for SDR was not publicly available for the years 2008, 2009, and 2010. These data were estimated using data for AZR's Waelz kilns for 2008 through 2010 (Horsehead 2007, 2008, 2010a, 2010b, and 2011). Annual capacity utilization ratios were calculated using AZR's annual consumption and total capacity for the years 2008 through 2010. AZR's annual capacity utilization ratios were multiplied with SDR's total capacity to estimate SDR's consumption for each of the years, 2008 through 2010 (SDR 2013).

PIZO Technologies Worldwide LLC's facility in Arkansas has been operational since 2009. The amount of EAF dust consumed by PIZO's facility for 2009 through 2017 was not publicly available. EAF dust consumption for PIZO's facility for 2009 and 2010 were estimated by calculating annual capacity utilization of AZR's Waelz kilns and multiplying this utilization ratio by PIZO's total capacity (PIZO 2012). EAF dust consumption for PIZO's facility for 2011 through 2017 were estimated by applying the average annual capacity utilization rates for AZR and SDR (Grupo PROMAX) to PIZO's annual capacity (Horsehead 2012, 2013, 2014, 2015, and 2016; SDR 2012, 2014 and 2017; PIZO 2012, 2014 and 2017). The 1.24 metric tons CO_2/metric ton EAF dust consumed emission factor was then applied to PIZO's and SDR's estimated EAF dust consumption to develop CO_2 emission estimates for those Waelz kiln facilities.

Refined zinc production levels for AZR's Monaca, PA facility (utilizing electrothermic technology) were available from the company for years 2005 through 2013 (Horsehead 2008, 2011, 2012, 2013, and 2014). The Monaca facility was permanently shut down in April 2014 and was replaced by AZR's new facility in Mooresboro, NC. The new facility uses hydrometallurgical process to produce refined zinc products. This process is assumed to be non-emissive. Production levels for 1990 through 2004 were extrapolated using the percentage changes in annual refined zinc production at secondary smelters in the United States as provided by USGS *Minerals Yearbook: Zinc* (USGS 1995 through 2005). The 3.70 metric tons CO_2/metric ton zinc emission factor was then applied to the Monaca facility's production levels to estimate CO_2 emissions for the facility. The Waelz kiln production emission factor was applied in this case rather than the EAF dust consumption emission factor since AZR's Monaca facility did not consume EAF dust.

The production and use of coking coal for zinc production is adjusted for within the Energy chapter as this fuel was consumed during non-energy related activities. Additional information on the adjustments made within the Energy sector for Non-Energy Use of Fuels is described in both the Methodology section of CO_2 from Fossil Fuel Combustion (3.1 Fossil Fuel Combustion (CRF Source Category 1A)) and Annex 2.1, Methodology for Estimating Emissions of CO_2 from Fossil Fuel Combustion.

Beginning with the 2017 USGS *Minerals Commodity Summary: Zinc*, United States primary and secondary refined zinc production were reported as one value, total refined zinc production. Prior to this publication, primary and secondary refined zinc production statistics were reported separately. For the current Inventory report, EPA sought expert judgement from the USGS mineral commodity expert to assess approaches for splitting total production into primary and secondary values. For 2016 and 2017, only one facility produced primary zinc. Primary zinc produced from this facility was subtracted from the USGS 2016/2017 total zinc production statistic to estimate secondary zinc production for these two years.

Uncertainty and Time-Series Consistency

The uncertainty associated with these estimates is two-fold, relating to activity data and emission factors used.

First, there is uncertainty associated with the amount of EAF dust consumed in the United States to produce secondary zinc using emission-intensive Waelz kilns. The estimate for the total amount of EAF dust consumed in Waelz kilns is based on (1) an EAF dust consumption value reported annually by AZR/Horsehead Corporation as part of its financial reporting to the Securities and Exchange Commission (SEC), and (2) an EAF dust consumption value obtained from the Waelz kiln facility operated in Alabama by Steel Dust Recycling LLC. Since actual EAF dust consumption information is not available for PIZO's facility (2009 through 2010) and SDR's facility (2008 through 2010), the amount is estimated by multiplying the EAF dust recycling capacity of the facility (available from the company's website) by the capacity utilization factor for AZR (which is available from Horsehead Corporation financial reports). Also, the EAF dust consumption for PIZO's facility for 2011 through 2016 was estimated by multiplying the average capacity utilization factor developed from AZR and SDR's annual capacity utilization rates by PIZO's EAF dust recycling capacity. Therefore, there is uncertainty associated with the assumption used to estimate PIZO and SDR's annual EAF dust consumption values (except SDR's EAF dust consumption for 2011 through 2017, which were obtained from SDR's recycling facility in Alabama).

Second, there is uncertainty associated with the emission factors used to estimate CO_2 emissions from secondary zinc production processes. The Waelz kiln emission factors are based on materials balances for metallurgical coke and EAF dust consumed as provided by Viklund-White (2000). Therefore, the accuracy of these emission factors depend upon the accuracy of these materials balances. Data limitations prevented the development of emission factors for the electrothermic process. Therefore, emission factors for the Waelz kiln process were applied to both electrothermic and Waelz kiln production processes.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-93. Zinc production CO_2 emissions from 2017 were estimated to be between 0.8 and 1.2 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 16 percent below and 16 percent above the emission estimate of 1.0 MMT CO_2 Eq.

Table 4-93: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Zinc Production (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Zinc Production	CO_2	1.0	0.8	1.2	-16%	+16%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

Pending resources and prioritization of improvements for more significant sources, EPA will continue to evaluate and analyze data reported under EPA's GHGRP that would be useful to improve the emission estimates and category-specific QC for the Zinc Production source category, in particular considering completeness of reported zinc production given the reporting threshold. Given the small number of facilities in the United States, particular attention will be made to risks for disclosing CBI and ensuring time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[236] This is a long-term planned improvement and EPA is still assessing the possibility of including this improvement in future Inventory reports.

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[236] See <http://www.ipcc-nggip.iges.or.jp/public/tb/TFI_Technical_Bulletin_1.pdf>.

4.23 Semiconductor Manufacture (CRF Source Category 2E1)

The semiconductor industry uses multiple greenhouse gases in its manufacturing processes. These include long-lived fluorinated greenhouse gases used for plasma etching and chamber cleaning, fluorinated heat transfer fluids used for temperature control and other applications, and nitrous oxide (N_2O) used to produce thin films through chemical vapor deposition.

The gases most commonly employed in plasma etching and chamber cleaning are trifluoromethane (HFC-23 or CHF_3), perfluoromethane (CF_4), perfluoroethane (C_2F_6), nitrogen trifluoride (NF_3), and sulfur hexafluoride (SF_6), although other fluorinated compounds such as perfluoropropane (C_3F_8) and perfluorocyclobutane (c-C_4F_8) are also used. The exact combination of compounds is specific to the process employed.

A single 300 mm silicon wafer that yields between 400 to 600 semiconductor products (devices or chips) may require more than 100 distinct fluorinated-gas-using process steps, principally to deposit and pattern dielectric films. Plasma etching (or patterning) of dielectric films, such as silicon dioxide and silicon nitride, is performed to provide pathways for conducting material to connect individual circuit components in each device. The patterning process uses plasma-generated fluorine atoms, which chemically react with exposed dielectric film to selectively remove the desired portions of the film. The material removed as well as undissociated fluorinated gases flow into waste streams and, unless emission abatement systems are employed, into the atmosphere. Plasma enhanced chemical vapor deposition (PECVD) chambers, used for depositing dielectric films, are cleaned periodically using fluorinated and other gases. During the cleaning cycle the gas is converted to fluorine atoms in plasma, which etches away residual material from chamber walls, electrodes, and chamber hardware. Undissociated fluorinated gases and other products pass from the chamber to waste streams and, unless abatement systems are employed, into the atmosphere.

In addition to emissions of unreacted gases, some fluorinated compounds can also be transformed in the plasma processes into different fluorinated compounds which are then exhausted, unless abated, into the atmosphere. For example, when C_2F_6 is used in cleaning or etching, CF_4 is generated and emitted as a process byproduct. In some cases, emissions of the byproduct gas can rival or even exceed emissions of the input gas, as is the case for NF_3 used in remote plasma chamber cleaning, which generates CF_4 as a byproduct.

Besides dielectric film etching and PECVD chamber cleaning, much smaller quantities of fluorinated gases are used to etch polysilicon films and refractory metal films like tungsten.

Nitrous oxide is used in manufacturing semiconductor devices to produce thin films by CVD and nitridation processes as well as for N-doping of compound semiconductors and reaction chamber conditioning (Doering 2000).

Liquid perfluorinated compounds are also used as heat transfer fluids (F-HTFs) for temperature control, device testing, cleaning substrate surfaces and other parts, and soldering in certain types of semiconductor manufacturing production processes. Leakage and evaporation of these fluids during use is a source of fluorinated gas emissions (EPA 2006). Unweighted F-HTF emissions consist primarily of perfluorinated amines, hydrofluoroethers, perfluoropolyethers, and perfluoroalkylmorpholines. One percent or less consist of HFC, PFC and SF_6 (where PFCs are defined as compounds including only carbon and fluorine). With the exceptions of the hydrofluoroethers and most of the HFCs, all of these compounds are very long-lived in the atmosphere and have global warming potentials (GWPs) near 10,000.[237]

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[237] The GWP of PFPMIE, a perfluoropolyether used as an F-HTF, is included in the *IPCC Fourth Assessment Report* with a value of 10,300. The GWPs of the perfluorinated amines and perfluoroalkylmorpholines that are used as F-HTFs have not been evaluated in the peer-reviewed literature. However, evaluations by the manufacturer indicate that their GWPs are near 10,000 (78 FR 20632), which is expected given that these compounds are both saturated and fully fluorinated. EPA assigns a default GWP of 10,000 to compounds that are both saturated and fully fluorinated and that do not have chemical-specific GWPs in either the Fourth or the Fifth Assessment Reports.

For 2017, total GWP-weighted emissions of all fluorinated greenhouse gases and N_2O from deposition, etching, and chamber cleaning processes in the U.S. semiconductor industry were estimated to be 5.0 MMT CO_2 Eq. These emissions are presented in Table 4-94 and Table 4-95 below for the years 1990, 2005, and the period 2013 to 2017. (Emissions of F-HTFs that are HFCs, PFCs or SF_6 are presented in Table 4-94 and Table 4-95. Emissions of F-HTFs that are not HFCs, PFCs or SF_6 are presented in Table 4-95, Table 4-96, and Table 4-97 but are not included in Inventory totals.) The rapid growth of this industry and the increasing complexity (growing number of layers)[238] of semiconductor products led to an increase in emissions of 153 percent between 1990 and 1999, when emissions peaked at 9.1 MMTCO_2 Eq. Emissions began to decline after 1999, reaching a low point in 2009 before rebounding slightly and plateauing at the current level, which represents a 44 percent decline from 1999 levels. Together, industrial growth, adoption of emissions reduction technologies (including but not limited to abatement technologies), and shifts in gas usages resulted in a net increase in emissions of 41 percent between 1990 and 2017.

Total emissions from semiconductor manufacture in 2017 were similar to 2016 emissions, decreasing by 1 percent.

Only F-HTF emissions that consist of HFC, PFC and SF_6 are included in the Inventory totals; emissions of other F-HTFs, which account for the vast majority of F-HTF emissions, are provided for informational purposes and are not included in the Inventory totals. Since reporting of F-HTF emissions began under EPA's GHGRP in 2011, total F-HTF emissions (reported and estimated non-reported) have fluctuated between 0.6 MMT CO_2 Eq. and 1.1 MMT CO_2 Eq., with an overall declining trend. An analysis of the data reported to EPA's GHGRP indicates that F-HTF emissions account for anywhere between 11 percent and 18 percent of total annual emissions (F-GHG, N_2O and F-HTFs) from semiconductor manufacturing.[239] Table 4-97 shows the emissions of the F-HTF compounds with the highest emissions in tons based on reporting to EPA's GHGRP during years 2011 through 2017.[240]

Table 4-94: PFC, HFC, SF_6, NF_3, and N_2O Emissions from Semiconductor Manufacture[241] (MMT CO_2 Eq.)

Year	1990	2005	2013	2014	2015	2016	2017
CF_4	0.8	1.1	1.3	1.5	1.5	1.5	1.6
C_2F_6	2.0	2.0	1.5	1.4	1.3	1.2	1.2
C_3F_8	+	0.1	0.1	0.1	0.1	0.1	0.1
C_4F_8	0.0	0.1	0.1	0.1	0.1	0.1	0.1
HFC-23	0.2	0.2	0.3	0.3	0.3	0.3	0.4
SF_6	0.5	0.7	0.7	0.7	0.7	0.8	0.7
NF_3	+	0.5	0.5	0.5	0.6	0.6	0.6
Total F-GHGs	**3.6**	**4.6**	**4.4**	**4.6**	**4.7**	**4.7**	**4.7**
N_2O	+	0.1	0.2	0.2	0.2	0.2	0.2
HFC, PFC and SF_6 F-HTFs	0.0	+	+	+	+	+	+
Total	**3.6**	**4.7**	**4.6**	**4.8**	**4.9**	**5.0**	**5.0**

+ Does not exceed 0.05 MMT CO_2 Eq.

Note: Totals may not sum due to independent rounding.

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[238] Complexity is a term denoting the circuit required to connect the active circuit elements (transistors) on a chip. Increasing miniaturization, for the same chip size, leads to increasing transistor density, which, in turn, requires more complex interconnections between those transistors. This increasing complexity is manifested by increasing the levels (i.e., layers) of wiring, with each wiring layer requiring fluorinated gas usage for its manufacture.

[239] Emissions data for HTFs (in tons of gas) from the semiconductor industry from 2011 through 2017 were obtained from the EPA GHGRP annual facility emissions reports.

[240] Many fluorinated heat transfer fluids consist of perfluoropolymethylisopropyl ethers (PFPMIEs) of different molecular weights and boiling points that are distilled from a mixture. "BP 200 °C" (and similar terms below) indicate the boiling point of the fluid in degrees Celsius. For more information, see <https://www.regulations.gov/document?D=EPA-HQ-OAR-2009-0927-0276>.

[241] An extremely small portion of emissions from Semiconductor Manufacture are from the manufacturing of MEMs and photovoltaic cells.

Table 4-95: PFC, HFC, SF_6, NF_3, and N_2O Emissions from Semiconductor Manufacture (kt)

Year	1990	2005	2013	2014	2015	2016	2017
CF_4	0.11	0.15	0.17	0.20	0.21	0.21	0.22
C_2F_6	0.16	0.16	0.13	0.11	0.11	0.10	0.10
C_3F_8	+	+	+	+	+	+	+
C_4F_8	0.0	+	+	+	+	+	+
HFC-23	+	+	+	+	+	+	+
SF_6	+	+	+	+	+	+	+
NF_3	+	+	+	+	+	+	+
N_2O	0.12	0.41	0.59	0.65	0.71	0.71	0.84
HFC, PFC and SF_6 F-HTFs	0.00	+	+	+	+	+	+
Total	**0.43**	**0.81**	**0.98**	**1.07**	**1.14**	**1.14**	**1.27**

+ Does not exceed 0.05 kt.

Table 4-96: F-HTF Emissions Based on GHGRP Reporting (MMT CO_2 Eq.)

Year	2011	2012	2013	2014	2015	2016	2017
HFCs	0.000	0.000	0.000	0.003	0.003	0.004	0.003
PFCs	0.002	0.002	0.000	0.003	0.003	0.004	0.003
SF_6	0.000	0.000	0.000	0.021	0.013	0.012	0.017
Other F-HTFs	0.877	1.097	0.676	0.790	0.735	0.654	0.586
Total F-HTFs	**0.879**	**1.099**	**0.676**	**0.816**	**0.754**	**0.673**	**0.609**

Table 4-97: Top 10 F-HTF Compounds with Largest Emissions Based on GHGRP Reporting (tons)

Fluorinated Heat Transfer Fluid[242]	GWP	GHGRP-Reported Emissions (tons)						
		2011	2012	2013	2014	2015	2016	2017
Perfluorotripropylamine (3M™ Fluorinert™ FC-3283/FC-8270)	10,000	24.36	35.86	22.72	17.03	10.22	20.57	12.47
Perfluoroisopropylmorpholine (3M™ Fluorinert™ FC-770)	10,000	12.27	9.27	10.09	7.16	3.13	7.35	5.11
PFPMIE fraction, BP 200 °C (Solvay Galden™ HT-200)	10,000	5.81	20.71	9.49	2.21	1.58	6.41	2.20
3-ethoxy-1,1,1,2,3,4,4,5,5,6,6,6-dodecafluoro-2-trifluoromethyl-hexane (3M™ HFE-7500)	270	8.57	7.21	13.85	2.68	2.92	2.23	7.09
HFE-569sf2, (3M™ HFE-7200)	59	8.17	10.53	5.78	4.27	2.92	3.17	6.86
HFE-449s1 (3M™ HFE-7100)	297	10.63	2.94	4.53	0.37	0.35	0.70	0.75
Perfluorotributylamine (PTBA, 3M™ Fluorinert™ FC40/FC-43)	10,000	10.52	3.77	1.45	0.80	0.25	1.35	1.38
PFPMIE fraction, BP 170 °C (Solvay Galden™ HT-170)	10,000	3.37	6.93	0.57	0.55	0.93	2.22	1.95
PFPMIE fraction, BP 165 °C (Solvay Galden™ DO2-TS)	10,000	2.61	2.45	4.89	0.88	0.00	1.46	1.35
PFPMIE fraction, BP 110 °C (Solvay Galden™ HT-110)	10,000	1.90	1.52	0.83	0.49	0.60	0.98	0.63

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[242] Many fluorinated heat transfer fluids consist of perfluoropolymethylisopropyl ethers (PFPMIEs) of different molecular weights and boiling points that are distilled from a mixture. “BP 200 °C” (and similar terms below) indicate the boiling point of the fluid in degrees Celsius. For more information, see <https://www.regulations.gov/document?D=EPA-HQ-OAR-2009-0927-0276>

Additional Emissions from MEMS and PV

Similar to semiconductor manufacturing, the manufacturing of micro-electro-mechanical devices (MEMs) and photovoltaic cells requires the use of multiple long-lived fluorinated greenhouse gases for various processes. GHGRP-reported emissions from the manufacturing of MEMs and photovoltaic cells are available for the years 2011 to 2017. They are not included in the semiconductor manufacturing totals reported above. The emissions reported by facilities manufacturing MEMs included emissions of C_2F_6, C_3F_8, C_4F_8, CF_4, HFC-23, NF_3, and SF_6, and were equivalent to only 0.08 percent to 0.40 percent of the total reported emissions from semiconductor manufacturing in 2011 to 2017. These emissions ranged from 0.0038 to 0.0171 MMT CO_2 Eq. from 2011 to 2017. Similarly, emissions from manufacturing of photovoltaic cells were equivalent to only 0.23 percent and 0.15 percent of the total reported emissions from semiconductor manufacturing in 2015 and 2016 respectively. Reported emissions from photovoltaic cell manufacturing consisted of CF_4, C_2F_6, C_4F_8, and CHF_3.

Based upon information in the World Fab Forecast (WFF), it appears that some GHGRP reporters that manufacture both semiconductors and MEMS are reporting their emissions as only from semiconductor manufacturing; however, the emissions from MEMS manufacturing are likely being included in semiconductor totals.

Methodology

Emissions are based on data reported through Subpart I, Electronics Manufacture, of EPA's GHGRP, Partner reported emissions data received through EPA's PFC[243] Reduction/Climate Partnership, EPA's PFC Emissions Vintage Model (PEVM)—a model that estimates industry emissions from etching and chamber cleaning processes in the absence of emission control strategies (Burton and Beizaie 2001),[244] and estimates of industry activity (i.e., total manufactured layer area). The availability and applicability of reported emissions data from the EPA Partnership and EPA's GHGRP and activity data differ across the 1990 through 2017 time series. Consequently, fluorinated greenhouse gas (F-GHG) emissions from etching and chamber cleaning processes were estimated using seven distinct methods, one each for the periods 1990 through 1994, 1995 through 1999, 2000 through 2006, 2007 through 2010, 2011 and 2012, 2013 and 2014, and 2015 through 2017. Nitrous oxide emissions were estimated using five distinct methods, one each for the period 1990 through 1994, 1995 through 2010, 2011 and 2012, 2013 and 2014, and 2015 through 2017.

Facility emissions of F-HTFs from semiconductor manufacturing are reported to EPA under its GHGRP and are available for the years 2011 through 2017. EPA estimates the emissions of F-HTFs from non-reporting facilities by calculating the ratio of GHGRP-reported fluorinated HTF emissions to GHGRP reported F-GHG emissions from etching and chamber cleaning processes, and then multiplying this ratio by the F-GHG emissions from etching and chamber cleaning processes estimated for non-reporting facilities. Fluorinated HTF use in semiconductor manufacturing is assumed to have begun in the early 2000s and to have gradually displaced other HTFs (e.g., de-ionized water and glycol) in electronics manufacturing (EPA 2006). For time-series consistency, EPA interpolated the share of F-HTF emissions to F-GHG emissions between 2000 (at 0 percent) and 2011 (at 22 percent) and applied these shares to the unadjusted F-GHG emissions during those years to estimate the fluorinated HTF emissions.

1990 through 1994

From 1990 through 1994, Partnership data were unavailable and emissions were modeled using PEVM (Burton and Beizaie 2001).[245] The 1990 to 1994 emissions are assumed to be uncontrolled, since reduction strategies such as chemical substitution and abatement were yet to be developed.

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[243] In the context of the EPA Partnership and PEVM, PFC refers to perfluorocompounds, not perfluorocarbons.

[244] A Partner refers to a participant in the U.S. EPA PFC Reduction/Climate Partnership for the Semiconductor Industry. Through a Memorandum of Understanding (MoU) with the EPA, Partners voluntarily reported their PFC emissions to the EPA by way of a third party, which aggregated the emissions through 2010.

[245] Various versions of the PEVM exist to reflect changing industrial practices. From 1990 to 1994 emissions estimates are from PEVM v1.0, completed in September 1998. The emission factor used to estimate 1990 to 1994 emissions is an average of the 1995 and 1996 emissions factors, which were derived from Partner reported data for those years.

PEVM is based on the recognition that fluorinated greenhouse gas emissions from semiconductor manufacturing vary with: (1) the number of layers that comprise different kinds of semiconductor devices, including both silicon wafer and metal interconnect layers, and (2) silicon consumption (i.e., the area of semiconductors produced) for each kind of device. The product of these two quantities, Total Manufactured Layer Area (TMLA), constitutes the activity data for semiconductor manufacturing. PEVM also incorporates an emission factor that expresses emissions per unit of manufactured layer-area. Emissions are estimated by multiplying TMLA by this emission factor.

PEVM incorporates information on the two attributes of semiconductor devices that affect the number of layers: (1) linewidth technology (the smallest manufactured feature size),[246] and (2) product type (discrete, memory or logic).[247] For each linewidth technology, a weighted average number of layers is estimated using VLSI product-specific worldwide silicon demand data in conjunction with complexity factors (i.e., the number of layers per Integrated Circuit (IC) specific to product type (Burton and Beizaie 2001; ITRS 2007). PEVM derives historical consumption of silicon (i.e., square inches) by linewidth technology from published data on annual wafer starts and average wafer size (VLSI Research, Inc. 2012).

The emission factor in PEVM is the average of four historical emission factors, each derived by dividing the total annual emissions reported by the Partners for each of the four years between 1996 and 1999 by the total TMLA estimated for the Partners in each of those years. Over this period, the emission factors varied relatively little (i.e., the relative standard deviation for the average was 5 percent). Since Partners are believed not to have applied significant emission reduction measures before 2000, the resulting average emission factor reflects uncontrolled emissions. The emission factor is used to estimate world uncontrolled emissions using publicly-available data on world silicon consumption.

As it was assumed for this time period that there was no consequential adoption of fluorinated-gas-reducing measures, a fixed distribution of fluorinated-gas use was assumed to apply to the entire U.S. industry to estimate gas-specific emissions. This distribution was based upon the average fluorinated-gas purchases made by semiconductor manufacturers during this period and the application of IPCC default emission factors for each gas (Burton and Beizaie 2001).

To estimate N_2O emissions, it is assumed the proportion of N_2O emissions estimated for 1995 (discussed below) remained constant for the period of 1990 through 1994.

1995 through 1999

For 1995 through 1999, total U.S. emissions were extrapolated from the total annual emissions reported by the Partners (1995 through 1999). Partner-reported emissions are considered more representative (e.g., in terms of capacity utilization in a given year) than PEVM-estimated emissions, and are used to generate total U.S. emissions when applicable. The emissions reported by the Partners were divided by the ratio of the total capacity of the plants operated by the Partners and the total capacity of all of the semiconductor plants in the United States; this ratio represents the share of capacity attributable to the Partnership. This method assumes that Partners and non-Partners have identical capacity utilizations and distributions of manufacturing technologies. Plant capacity data is contained in the World Fab Forecast (WFF) database and its predecessors, which is updated quarterly. Gas-specific emissions were estimated using the same method as for 1990 through 1994.

For this time period, the N_2O emissions were estimated using an emission factor that was applied to the annual, total U.S. TMLA manufactured. The emission factor was developed using a regression-through-the-origin (RTO) model: GHGRP reported N_2O emissions were regressed against the corresponding TMLA of facilities that reported no use

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[246] By decreasing features of Integrated Circuit components, more components can be manufactured per device, which increases its functionality. However, as those individual components shrink it requires more layers to interconnect them to achieve the functionality. For example, a microprocessor manufactured with 65 nm feature sizes might contain as many as 1 billion transistors and require as many as 11 layers of component interconnects to achieve functionality, while a device manufactured with 130 nm feature size might contain a few hundred million transistors and require 8 layers of component interconnects (ITRS 2007).

[247] Memory devices manufactured with the same feature sizes as microprocessors (a logic device) require approximately one-half the number of interconnect layers, whereas discrete devices require only a silicon base layer and no interconnect layers (ITRS 2007). Since discrete devices did not start using PFCs appreciably until 2004, they are only accounted for in the PEVM emissions estimates from 2004 onwards.

of abatement systems. Details on EPA's GHGRP reported emissions and development of emission factor using the RTO model are presented in the 2011 through 2012 section. The total U.S. TMLA was estimated using PEVM.

2000 through 2006

Emissions for the years 2000 through 2006—the period during which Partners began the consequential application of fluorinated greenhouse gas-reduction measures—were estimated using a combination of Partner-reported emissions and adjusted PEVM modeled emissions. The emissions reported by Partners for each year were accepted as the quantity emitted from the share of the industry represented by those Partners. Remaining emissions, those from non-Partners, were estimated using PEVM, with one change. To ensure time-series consistency and to reflect the increasing use of remote clean technology (which increases the efficiency of the production process while lowering emissions of fluorinated greenhouse gases), the average non-Partner emission factor (PEVM emission factor) was assumed to begin declining gradually during this period. Specifically, the non-Partner emission factor for each year was determined by linear interpolation, using the end points of 1999 (the original PEVM emission factor) and 2011 (a new emission factor determined for the non-Partner population based on GHGRP-reported data, described below).

The portion of the U.S. total emissions attributed to non-Partners is obtained by multiplying PEVM's total U.S. emissions figure by the non-Partner share of U.S. total silicon capacity for each year as described above.[248] Gas-specific emissions from non-Partners were estimated using linear interpolation of gas-specific emission distribution of 1999 (assumed same as total U.S. Industry in 1994) and 2011 (calculated from a subset of non-Partner facilities from GHGRP reported emissions data). Annual updates to PEVM reflect published figures for actual silicon consumption from VLSI Research, Inc., revisions and additions to the world population of semiconductor manufacturing plants, and changes in IC fabrication practices within the semiconductor industry (see ITRS 2008 and Semiconductor Equipment and Materials Industry 2011).[249,250,251]

Nitrous oxide emissions were estimated using the same methodology as the 1995 through 1999 methodology.

2007 through 2010

For the years 2007 through 2010, emissions were also estimated using a combination of Partner reported emissions and adjusted PEVM modeled emissions to provide estimates for non-Partners; however, two improvements were made to the estimation method employed for the previous years in the time series. First, the 2007 through 2010

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[248] This approach assumes that the distribution of linewidth technologies is the same between Partners and non-Partners. As discussed in the description of the method used to estimate 2007 emissions, this is not always the case.

[249] Special attention was given to the manufacturing capacity of plants that use wafers with 300 mm diameters because the actual capacity of these plants is ramped up to design capacity, typically over a 2–3 year period. To prevent overstating estimates of partner-capacity shares from plants using 300 mm wafers, *design* capacities contained in WFF were replaced with estimates of *actual installed* capacities for 2004 published by Citigroup Smith Barney (2005). Without this correction, the partner share of capacity would be overstated, by approximately 5 percent. For perspective, approximately 95 percent of all new capacity additions in 2004 used 300 mm wafers, and by year-end those plants, on average, could operate at approximately 70 percent of the design capacity. For 2005, actual installed capacities were estimated using an entry in the World Fab Watch database (April 2006 Edition) called "wafers/month, 8-inch equivalent," which denoted the actual installed capacity instead of the fully-ramped capacity. For 2006, actual installed capacities of new fabs were estimated using an average monthly ramp rate of 1100 wafer starts per month (wspm) derived from various sources such as semiconductor fabtech, industry analysts, and articles in the trade press. The monthly ramp rate was applied from the first-quarter of silicon volume (FQSV) to determine the average design capacity over the 2006 period.

[250] In 2006, the industry trend in co-ownership of manufacturing facilities continued. Several manufacturers, who are Partners, now operate fabs with other manufacturers, who in some cases are also Partners and in other cases are not Partners. Special attention was given to this occurrence when estimating the Partner and non-Partner shares of U.S. manufacturing capacity.

[251] Two versions of PEVM are used to model non-Partner emissions during this period. For the years 2000 to 2003 PEVM v3.2.0506.0507 was used to estimate non-Partner emissions. During this time, discrete devices did not use PFCs during manufacturing and therefore only memory and logic devices were modeled in the PEVM v3.2.0506.0507. From 2004 onwards, discrete device fabrication started to use PFCs, hence PEVM v4.0.0701.0701, the first version of PEVM to account for PFC emissions from discrete devices, was used to estimate non-Partner emissions for this time period.

emission estimates account for the fact that Partners and non-Partners employ different distributions of manufacturing technologies, with the Partners using manufacturing technologies with greater transistor densities and therefore greater numbers of layers.[252] Second, the scope of the 2007 through 2010 estimates was expanded relative to the estimates for the years 2000 through 2006 to include emissions from research and development (R&D) fabs. This additional enhancement was feasible through the use of more detailed data published in the WFF. PEVM databases were updated annually as described above. The published world average capacity utilization for 2007 through 2010 was used for production fabs, while for R&D fabs a 20 percent figure was assumed (SIA 2009).

In addition, publicly-available actual utilization data was used to account for differences in fab utilization for manufacturers of discrete and IC products for 2010 emissions for non-Partners. The Semiconductor Capacity Utilization (SICAS) Reports from SIA provides the global semiconductor industry capacity and utilization, differentiated by discrete and IC products (SIA 2009 through 2011). PEVM estimates were adjusted using technology-weighted capacity shares that reflect the relative influence of different utilization. Gas-specific emissions for non-Partners were estimated using the same method as for 2000 through 2006.

Nitrous oxide emissions were estimated using the same methodology as the 1995 through 1999 methodology.

2011 through 2012

The fifth method for estimating emissions from semiconductor manufacturing covers the period 2011 through 2012. This methodology differs from previous years because the EPA's Partnership with the semiconductor industry ended (in 2010) and reporting under EPA's GHGRP began. Manufacturers whose estimated uncontrolled emissions equal or exceed 25,000 MT CO_2 Eq. per year (based on default F-GHG-specific emission factors and total capacity in terms of substrate area) are required to report their emissions to EPA. This population of reporters to EPA's GHGRP included both historical Partners of EPA's PFC Reduction/Climate Partnership as well as non-Partners some of which use GaAs technology in addition to Si technology.[253] Emissions from the population of manufacturers that were below the reporting threshold were also estimated for this time period using EPA-developed emission factors and estimates of facility-specific production obtained from WFF. Inventory totals reflect the emissions from both reporting and non-reporting populations.

Under EPA's GHGRP, semiconductor manufacturing facilities report emissions of F-GHGs used in etch and clean processes as well as emissions of fluorinated heat transfer fluids. (Fluorinated heat transfer fluids are used to control process temperatures, thermally test devices, and clean substrate surfaces, among other applications.) They also report N_2O emissions from CVD and other processes. The F-GHGs and N_2O were aggregated, by gas, across all semiconductor manufacturing GHGRP reporters to calculate gas-specific emissions for the GHGRP-reporting segment of the U.S. industry. At this time, emissions that result from heat transfer fluid use that are HFC, PFC and SF_6 are included in the total emission estimates from semiconductor manufacturing, and these GHGRP-reported emissions have been compiled and presented in Table 4-94. F-HTF emissions resulting from other types of gases (e.g., HFEs) are not presented in semiconductor manufacturing totals in Table 4-94 and Table 4-95 but are shown in Table 4-96 and Table 4-97 for informational purposes.

Changes to the default emission factors and default destruction or removal efficiencies (DREs) used for GHGRP reporting affected the emissions trend between 2013 and 2014. These changes did not reflect actual emission rate changes but data improvements. Therefore, for the current Inventory, EPA adjusted the time series of GHGRP-reported data for 2011 through 2013 to ensure time-series consistency using a series of calculations that took into account the characteristics of a facility (e.g., wafer size and abatement use). To adjust emissions for facilities that did not report abatement in 2011 through 2013, EPA simply applied the revised emission factors to each facility's estimated gas consumption by gas, process type and wafer size. In 2014, EPA also started collecting information on

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[252] EPA considered applying this change to years before 2007, but found that it would be difficult due to the large amount of data (i.e., technology-specific global and non-Partner TMLA) that would have to be examined and manipulated for each year. This effort did not appear to be justified given the relatively small impact of the improvement on the total estimate for 2007 and the fact that the impact of the improvement would likely be lower for earlier years because the estimated share of emissions accounted for by non-Partners is growing as Partners continue to implement emission-reduction efforts.

[253] GaAs and Si technologies refer to the wafer on which devices are manufactured, which use the same PFCs but in different ways.

fab-wide DREs and the gases abated by process type, which were used in calculations for adjusting emissions from facilities that abated F-GHGs in 2011 through 2013.

- To adjust emissions for facilities that abated emissions in 2011 through 2013, EPA first calculated the quantity of gas abated in 2014 using reported F-GHG emissions, the revised default DREs (or the estimated site-specific DRE,[254] if a site-specific DRE was indicated), and the fab-wide DREs reported in 2014.[255] To adjust emissions for facilities that abated emissions in 2011 through 2013, EPA first estimated the percentage of gas passing through abatement systems for remote plasma clean in 2014 using the ratio of emissions reported for CF_4 and NF_3.
- EPA then estimated the quantity of NF_3 abated for remote plasma clean in 2014 using the ratio of emissions reported for CF_4 (which is not abated) and NF_3. This abated quantity was then subtracted from the total abated quantity calculated as described in the bullet above.
- To account for the resulting remaining abated quantity, EPA assumed that the percentage of gas passing through abatement systems was the same across all remaining gas and process type combinations where abatement was reported for 2014.
- The percentage of gas abated was then assumed to be the same in 2011 through 2013 (if the facility claimed abatement that year) as in 2014 for each gas abated in 2014.

The revised emission factors and DREs were then applied to the estimated gas consumption for each facility by gas, process type and wafer size.[256]

For the segment of the semiconductor industry that is below EPA's GHGRP reporting threshold, and for R&D facilities, which are not covered by EPA's GHGRP, emission estimates are based on EPA-developed emission factors for the F-GHGs and N_2O and estimates of manufacturing activity. The new emission factors (in units of mass of CO_2 Eq./TMLA [MSI]) are based on the emissions reported under EPA's GHGRP by facilities without abatement and on the TMLA estimates for these facilities based on the WFF (SEMI 2012, 2013).[257] In a refinement of the method used to estimate emissions for the non-Partner population for prior years, different emission factors were developed for different subpopulations of fabs, disaggregated by wafer size (200 mm or less and 300 mm). For each of these groups, a subpopulation-specific emission factor was obtained using a regression-through-the-origin (RTO) model: facility-reported aggregate emissions of seven F-GHGs (CF_4, C_2F_6, C_3F_8, C_4F_8, CHF_3, SF_6 and NF_3)[258] were regressed against the corresponding TMLA to estimate an aggregate F-GHG emissions factor (CO_2 Eq./MSI TMLA), and facility-reported N_2O emissions were regressed against the corresponding TMLA to estimate a N_2O emissions factor (CO_2 Eq./MSI TMLA). For each subpopulation, the slope of the RTO model is the emission factor for that subpopulation. Information on the use of point-of-use abatement by non-reporting fabs was not available; thus, EPA conservatively assumed that non-reporting facilities did not use point-of-use abatement.

For 2011 and 2012, estimates of TMLA relied on the capacity utilization of the fabs published by the U.S. Census Bureau's Historical Data Quarterly Survey of Plant Capacity Utilization (USCB 2011, 2012). Similar to the assumption for 2007 through 2010, facilities with only R&D activities were assumed to utilize only 20 percent of their manufacturing capacity. All other facilities in the United States are assumed to utilize the average percent of the manufacturing capacity without distinguishing whether fabs produce discrete products or logic products.

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[254] EPA generally assumed site-specific DREs were as follows: CF_4, Etch (90 percent); all other gases, Etch (98 percent); NF_3, Clean (95 percent); CF_4, Clean (80 percent), and all other gases, Clean (80 percent). There were a few exceptions where a higher DRE was assumed to ensure the calculations operated correctly when there was 100 percent abatement.

[255] If abatement information was not available for 2014 or the reported incorrectly in 2014, data from 2015 or 2016 was substituted.

[256] Since facilities did not report by fab before 2014, fab-wide DREs were averaged if a facility had more than one fab. For facilities that reported more than one wafer size per facility, the percentages of a facility's emissions per wafer size were estimated in 2014 and applied to earlier years, if possible. If the percentage of emissions per wafer size were unknown, a 50/50 split was used.

[257] EPA does not have information on fab-wide DREs for this time period, so it is not possible to estimate uncontrolled emissions from fabs that reported POU abatement. These fabs were therefore excluded from the regression analysis. (They are still included in the national totals.)

[258] Only seven gases were aggregated because inclusion of F-GHGs that are not reported in the Inventory results in overestimation of emission factor that is applied to the various non-reporting subpopulations.

Non-reporting fabs were then broken out into similar subpopulations by wafer size using information available through the WFF. The appropriate emission factor was applied to the total TMLA of each subpopulation of non-reporting facilities to estimate the GWP-weighted emissions of that subpopulation.

Gas-specific, GWP-weighted emissions for each subpopulation of non-reporting facilities were estimated using the corresponding reported distribution of gas-specific, GWP-weighted emissions from which the aggregate emission factors, based on GHGRP-reported data, were developed. Estimated in this manner, the non-reporting population accounted for 4.9 and 5.0 percent of U.S. emissions in 2011 and 2012, respectively. The GHGRP-reported emissions and the calculated non-reporting population emissions are summed to estimate the total emissions from semiconductor manufacturing.

2013 and 2014

For 2013 and 2014, as for 2011 and 2012, F-GHG and N_2O emissions data received through EPA's GHGRP were aggregated, by gas, across all semiconductor-manufacturing GHGRP reporters to calculate gas-specific emissions for the GHGRP-reporting segment of the U.S. industry. However, for these years WFF data was not available. Therefore, an updated methodology that does not depend on the WFF derived activity data was used to estimate emissions for the segment of the industry that are not covered by EPA's GHGRP. For the facilities that did not report to the GHGRP (i.e., which are below EPA's GHGRP reporting threshold or are R&D facilities), emissions were estimated based on the proportion of total U.S. emissions attributed to non-reporters for 2011 and 2012. EPA used a simple averaging method by first estimating this proportion for both F-GHGs and N_2O for 2011, 2012, and 2015 through 2017, resulting in one set of proportions for F-GHGs and one set for N_2O, and then applied the average of each set to the 2013 and 2014 GHGRP reported emissions to estimate the non-reporters' emissions. Fluorinated gas-specific, GWP-weighted emissions for non-reporters were estimated using the corresponding reported distribution of gas-specific, GWP-weighted emissions reported through EPA's GHGRP for 2013 and 2014.

GHGRP-reported emissions in 2013 were adjusted to capture changes to the default emission factors and default destruction or removal efficiencies used for GHGRP reporting affected the emissions trend between 2013 and 2014. EPA used the same method to make these adjustments as described above for 2011 and 2012 GHGRP data.

2015 through 2017

Similar to the methods described above for 2011 and 2012, and 2013 and 2014, EPA relied upon emissions data reported directly through the GHGRP. For 2015 through 2017, EPA took an approach similar to the one used for 2011 and 2012 to estimate emissions for the segment of the semiconductor industry that is below EPA's GHGRP reporting threshold, and for R&D facilities, which are not covered by EPA's GHGRP. However, in a change from previous years, EPA was able to develop new annual emission factors for 2015 through 2017 using TMLA from WFF and a more comprehensive set of emissions, i.e., fabs with as well as without abatement control, as new information about the use of abatement in GHGRP fabs and fab-wide were available. Fab-wide DREs represent total fab CO_2 Eq.-weighted controlled F-GHG and N_2O emissions (emissions after the use of abatement) divided by total fab CO_2 Eq.-weighted uncontrolled F-GHG and N_2O emissions (emission prior to the use of abatement).

Using information about reported emissions and the use of abatement and fab-wide DREs, EPA was able to calculate uncontrolled emissions (each total F-GHG and N_2O) for every GHGRP reporting fab. Using this, coupled with TMLA estimated using methods described above (see 2011 through 2012), EPA derived emission factors by year, gas type (F-GHG or N_2O), and wafer size (200 mm or 300 mm) by dividing the total annual emissions reported by GHGRP reporters by the total TMLA estimated for those reporters. These emission factors were multiplied by estimates of non-reporter TMLA to arrive at estimates of total F-GHG and N_2O emissions for non-reporters for each year. For each wafer size, the total F-GHG emissions were disaggregated into individual gases using the shares of total emissions represented by those gases in the emissions reported to the GHGRP by unabated fabs producing that wafer size.

Data Sources

GHGRP reporters, which consist of former EPA Partners and non-Partners, estimated their emissions using a default emission factor method established by EPA. Like the Tier 2b Method in the *2006 IPCC Guidelines*, this method uses different emission and byproduct generation factors for different F-GHGs and process types, but it goes beyond the Tier 2b Method by requiring use of updated factors for different wafer sizes (i.e., 300mm vs. 150 and 200mm) and CVD clean subtypes (in situ thermal, in situ thermal, and remote plasma). Starting with 2014 reported emissions, EPA's GHGRP required semiconductor manufacturers to apply updated emission factors to estimate their F-GHG emissions (40 CFR Part 98). For the years 2011 through 2013 reported emissions, semiconductor manufacturers used older emission factors to estimate their F-GHG emissions (Federal Register / Vol. 75, No. 230 /December 1, 2010, 74829). Subpart I emission factors were updated for 2014 by EPA as a result of a larger set of emission factor data becoming available as part of the Subpart I petition process, which took place from 2011 through 2013.

Historically, partners estimated and reported their emissions using a range of methods and uneven documentation. It is assumed that most Partners used a method at least as accurate as the IPCC's Tier 2a Methodology, recommended in the *2006 IPCC Guidelines*. Partners are estimated to have accounted for between 56 and 79 percent of F-GHG emissions from U.S. semiconductor manufacturing between 1995 and 2010, with the percentage declining in recent years as Partners increasingly implemented abatement measures.

Estimates of operating plant capacities and characteristics for Partners and non-Partners were derived from the Semiconductor Equipment and Materials Industry (SEMI) WFF (formerly World Fab Watch) database (1996 through 2012 and 2015) (e.g., Semiconductor Materials and Equipment Industry 2017). Actual worldwide capacity utilizations for 2008 through 2010 were obtained from Semiconductor International Capacity Statistics (SICAS) (SIA 2009 through 2011). Estimates of the number of layers for each linewidth was obtained from International Technology Roadmap for Semiconductors: 2013 Edition (Burton and Beizaie 2001; ITRS 2007; ITRS 2008; ITRS 2011; ITRS 2013). PEVM utilized the WFF, SICAS, and ITRS, as well as historical silicon consumption estimates published by VLSI. Actual quarterly U.S. capacity utilizations for 2011, 2012 and 2015 were obtained from the U.S. Census Bureau's Historical Data Quarterly Survey of Plant Capacity Utilization (USCB 2011, 2012, 2015, and 2016).

Uncertainty and Time-Series Consistency

A quantitative uncertainty analysis of this source category was performed using the IPCC-recommended Approach 2 uncertainty estimation methodology, the Monte Carlo Stochastic Simulation technique. The equation used to estimate uncertainty is:

$$\text{Total Emissions } (E_T) = \text{GHGRP Reported F-GHG Emissions } (E_{R,F\text{-}GHG}) + \text{Non-Reporters' Estimated F-GHG Emissions } (E_{NR,F\text{-}GHG}) + \text{GHGRP Reported } N_2O \text{ Emissions } (E_{R,N_2O}) + \text{Non-Reporters' Estimated } N_2O \text{ Emissions } (E_{NR,N_2O})$$

where E_R and E_{NR} denote totals for the indicated subcategories of emissions for F-GHG and N_2O, respectively.

The uncertainty in E_T presented in Table 4-98 below results from the convolution of four distributions of emissions, each reflecting separate estimates of possible values of $E_{R,F\text{-}GHG}$, $E_{R,N2O}$, $E_{NR,F\text{-}GHG}$, and $E_{NR,N2O}$. The approach and methods for estimating each distribution and combining them to arrive at the reported 95 percent confidence interval (CI) are described in the remainder of this section.

The uncertainty estimate of $E_{R,\ F\text{-}GHG}$, or GHGRP-reported F-GHG emissions, is developed based on gas-specific uncertainty estimates of emissions for two industry segments, one processing 200 mm wafers and one processing 300 mm wafers. Uncertainties in emissions for each gas and industry segment were developed during the assessment of emission estimation methods for the subpart I GHGRP rulemaking in 2012 (see Technical Support for Modifications to the Fluorinated Greenhouse Gas Emission Estimation Method Option for Semiconductor Facilities under Subpart I, docket EPA–HQ–OAR–2011–0028).[259] The 2012 analysis did not take into account the use of

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[259] On November 13, 2013, EPA published a final rule revising subpart I (Electronics Manufacturing) of the GHGRP (78 FR 68162). The revised rule includes updated default emission factors and updated default destruction and removal efficiencies that are slightly different from those that semiconductor manufacturers were required to use to report their 2012 emissions. The uncertainty analyses that were performed during the development of the revised rule focused on these updated defaults, but are expected to be reasonably representative of the uncertainties associated with the older defaults, particularly for estimates at the country level. (They may somewhat underestimate the uncertainties associated with the older defaults at the facility level.) For simplicity, the 2012 estimates are assumed to be unbiased although in some cases, the updated (and therefore more

abatement. For the industry segment that processed 200 mm wafers, estimates of uncertainties at a 95 percent CI ranged from ±29 percent for C_3F_8 to ±10 percent for CF_4. For the corresponding 300 mm industry segment, estimates of the 95 percent CI ranged from ±36 percent for C_4F_8 to ±16 percent for CF_4. These gas and wafer-specific uncertainty estimates are applied to the total emissions of the facilities that did not abate emissions as reported under EPA's GHGRP.

For those facilities reporting abatement of emissions under EPA's GHGRP, estimates of uncertainties for the no abatement industry segments are modified to reflect the use of full abatement (abatement of all gases from all cleaning and etching equipment) and partial abatement. These assumptions used to develop uncertainties for the partial and full abatement facilities are identical for 200 mm and 300 mm wafer processing facilities. For all facilities reporting gas abatement, a triangular distribution of destruction or removal efficiency is assumed for each gas. The triangular distributions range from an asymmetric and highly uncertain distribution of zero percent minimum to 90 percent maximum with 70 percent most likely value for CF_4 to a symmetric and less uncertain distribution of 85 percent minimum to 95 percent maximum with 90 percent most likely value for C_4F_8, NF_3, and SF_6. For facilities reporting partial abatement, the distribution of fraction of the gas fed through the abatement device, for each gas, is assumed to be triangularly distributed as well. It is assumed that no more than 50 percent of the gases are abated (i.e., the maximum value) and that 50 percent is the most likely value and the minimum is zero percent. Consideration of abatement then resulted in four additional industry segments, two 200-mm wafer-processing segments (one fully and one partially abating each gas) and two 300-mm wafer-processing segment (one fully and the other partially abating each gas). Gas-specific emission uncertainties were estimated by convolving the distributions of unabated emissions with the appropriate distribution of abatement efficiency for fully and partially abated facilities using a Monte Carlo simulation.

The uncertainty in $E_{R,F\text{-}GHG}$ is obtained by allocating the estimates of uncertainties to the total GHGRP-reported emissions from each of the six industry segments, and then running a Monte Carlo simulation which results in the 95 percent CI for emissions from GHGRP reporting facilities ($E_{R,F\text{-}GHG}$).

The uncertainty in $E_{R,N2O}$ is obtained by assuming that the uncertainty in the emissions reported by each of the GHGRP reporting facilities results from the uncertainty in quantity of N_2O consumed and the N_2O emission factor (or utilization). Similar to analyses completed for subpart I (see Technical Support for Modifications to the Fluorinated Greenhouse Gas Emission Estimation Method Option for Semiconductor Facilities under Subpart I, docket EPA–HQ–OAR–2011–0028), the uncertainty of N_2O consumed was assumed to be 20 percent. Consumption of N_2O for GHGRP reporting facilities was estimated by back-calculating from emissions reported and assuming no abatement. The quantity of N_2O utilized (the complement of the emission factor) was assumed to have a triangular distribution with a minimum value of zero percent, mode of 20 percent and maximum value of 84 percent. The minimum was selected based on physical limitations, the mode was set equivalent to the subpart I default N_2O utilization rate for chemical vapor deposition, and the maximum was set equal to the maximum utilization rate found in ISMI Analysis of Nitrous Oxide Survey Data (ISMI 2009). The inputs were used to simulate emissions for each of the GHGRP reporting, N_2O-emitting facilities. The uncertainty for the total reported N_2O emissions was then estimated by combining the uncertainties of each of the facilities reported emissions using Monte Carlo simulation.

The estimate of uncertainty in $E_{NR,\ F\text{-}GHG}$ and $E_{NR,\ N2O}$ entailed developing estimates of uncertainties for the emissions factors and the corresponding estimates of TMLA.

The uncertainty in TMLA depends on the uncertainty of two variables—an estimate of the uncertainty in the average annual capacity utilization for each level of production of fabs (e.g., full scale or R&D production) and a corresponding estimate of the uncertainty in the number of layers manufactured. For both variables, the distributions of capacity utilizations and number of manufactured layers are assumed triangular for all categories of non-reporting fabs. The most probable utilization is assumed to be 82 percent, with the highest and lowest utilization assumed to be 89 percent, and 70 percent, respectively. For the triangular distributions that govern the number of possible layers manufactured, it is assumed the most probable value is one layer less than reported in the ITRS; the smallest number

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representative) defaults are higher or lower than the older defaults. Multiple models and sensitivity scenarios were run for the subpart I analysis. The uncertainty analysis presented here made use of the Input gas and wafer size model (Model 1) under the following conditions: Year = 2010, f = 20, n = SIA3.

varied by technology generation between one and two layers less than given in the ITRS and largest number of layers corresponded to the figure given in the ITRS.

The uncertainty bounds for the average capacity utilization and the number of layers manufactured are used as inputs in a separate Monte Carlo simulation to estimate the uncertainty around the TMLA of both individual facilities as well as the total non-reporting TMLA of each sub-population.

The uncertainty around the emission factors for non-reporting facilities is dependent on the uncertainty of the total emissions (MMT CO_2 Eq. units) and the TMLA of each reporting facility in that category. For each wafer size for reporting facilities, total emissions were regressed on TMLA (with an intercept forced to zero) for 10,000 emission and 10,000 TMLA values in a Monte Carlo simulation, which results in 10,000 total regression coefficients (emission factors). The 2.5th and the 97.5th percentile of these emission factors are determined and the bounds are assigned as the percent difference from the estimated emission factor.

For simplicity, the results of the Monte Carlo simulations on the bounds of the gas- and wafer size-specific emissions as well as the TMLA and emission factors are assumed to be normally distributed and the uncertainty bounds are assigned at 1.96 standard deviations around the estimated mean. The departures from normality were observed to be small.

The final step in estimating the uncertainty in emissions of non-reporting facilities is convolving the distribution of emission factors with the distribution of TMLA using Monte Carlo simulation.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-98, which is also obtained by convolving—using Monte Carlo simulation—the distributions of emissions for each reporting and non-reporting facility. The emissions estimate for total U.S. F-GHG and N_2O emissions from semiconductor manufacturing were estimated to be between 4.7 and 5.2 MMT CO_2 Eq. at a 95 percent confidence level. This range represents 5 percent below to 5 percent above the 2017 emission estimate of 5.0 MMT CO_2 Eq. This range and the associated percentages apply to the estimate of total emissions rather than those of individual gases. Uncertainties associated with individual gases will be somewhat higher than the aggregate, but were not explicitly modeled.

Table 4-98: Approach 2 Quantitative Uncertainty Estimates for HFC, PFC, SF_6, NF_3 and N_2O Emissions from Semiconductor Manufacture (MMT CO_2 Eq. and Percent)[a]

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[b] (MMT CO_2 Eq.)		(%)	
			Lower Bound[c]	Upper Bound[c]	Lower Bound	Upper Bound
Semiconductor Manufacture	HFC, PFC, SF_6, NF_3, and N_2O	5.0	4.7	5.2	-5%	+5%

[a] This uncertainty analysis does not include quantification of the uncertainty of emissions from heat transfer fluids.

[b] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

[c] Absolute lower and upper bounds were calculated using the corresponding lower and upper bounds in percentages.

It should be noted that the uncertainty analysis for this source category does not quantify the uncertainty of HFC, PFC, and SF_6 emissions from the use of heat transfer fluids. While these emissions are included in the semiconductor manufacturing F-GHG total emissions, they make up a considerably small portion of total emissions from the source category (less than 1 percent). Any uncertainty of these emissions would have minimal impact on the overall uncertainty estimates, and therefore the uncertainties associated for HTF HFC, PFC and SF_6 emissions was not included in this analysis for this Inventory year. In an effort to improve the uncertainty analysis for this source category, HFC, PFC and SF_6 emissions from the use of heat transfer fluids may be added in future inventory years (see Planned Improvements section below). The emissions reported under EPA's GHGRP for 2014, 2015, 2016, and 2017, which are included in the overall emissions estimates, were based on an updated set of default emission factors. This may have affected the trend seen between 2013 and 2014 (a 24-percent increase), which reversed the trend seen between 2011 and 2013. As discussed in the Planned Improvements section, EPA is planning to conduct analysis to determine how much of the 2013 to 2014 trend may be attributable to the updated factors and to improve time-series consistency.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Recalculations Discussion

Emissions from 2011 through 2017 were updated to reflect updated emissions reporting in EPA's GHGRP, relative to the previous Inventory. Additionally, as discussed above, GHGRP-reported emissions for 2011, 2012, and 2013 were adjusted to capture changes to the default emission factors and default destruction or removal efficiencies used for GHGRP reporting affected the emissions trend between 2013 and 2014. Lastly, SF_6 emissions from the use of heat transfer fluids were separated from the "Other HTF" category and were estimated and reported independently.

Planned Improvements

This Inventory contains emissions estimates for N_2O and for seven fluorinated gases emitted from etching and chamber cleaning processes. However, other fluorinated gases (e.g., C_5F_8) are also emitted from etching and chamber cleaning processes in much smaller amounts, accounting for less than 0.02 percent of emissions from these processes. Previously, emissions data for these other fluorinated gases was not reported through the EPA Partnership. However, through EPA's GHGRP, these data are available. Therefore, a point of consideration for future Inventory reports is the inclusion of other fluorinated gases from etching and chamber cleaning processes.

In addition, EPA's GHGRP requires the reporting of emissions from other types of electronics manufacturing, including MEMs, flat panel displays, and photovoltaic cells. There currently are seven MEMs manufacturers (most of which report emissions for semiconductor and MEMs manufacturing separately), and no flat panel displays manufacturing facilities reporting to EPA's GHGRP; one photovoltaic cell manufacturer previously reported to the GHGRP.[260] Emissions from MEMs and photovoltaic cell manufacturing could be included in totals in future Inventory reports–currently they are not represented in Inventory emissions totals for electronics manufacturing. These emissions could be estimated for the full time series (including prior to the GHGRP) and for MEMS and photovoltaic cell manufacturers that are not reporting to the GHGRP; however, at this time the contribution to total emissions is not significant enough to warrant the development of the methodologies that would be necessary to back-cast these emissions to 1990 and estimate emissions for non-reporters for 2011 through 2017.

The Inventory methodology uses data reported through the EPA Partnership (for earlier years) and EPA's GHGRP (for later years) to extrapolate the emissions of the non-reporting population. While these techniques are well developed, the understanding of the relationship between the reporting and non-reporting populations is limited. Further analysis of the reporting and non-reporting populations could aid in the accuracy of the non-reporting population extrapolation in future years. In addition, the accuracy of the emissions estimates for the non-reporting population could be further increased through EPA's further investigation of and improvement upon the accuracy of estimated activity in the form of TMLA.

The Inventory uses utilization from two different sources for various time periods–SEMI to develop PEVM and to estimate non-Partner emissions for the period 1995 to 2010 and U.S. Census Bureau for 2011 through 2014. SEMI reported global capacity utilization for manufacturers through 2011. U.S. Census Bureau capacity utilization include U.S. semiconductor manufacturers as well as assemblers. Further analysis on the impacts of using a new and different source of utilization data could prove to be useful in better understanding of industry trends and impacts of utilization data sources on historical emission estimates.

The current Inventory now includes HFC, PFC and SF_6 emissions resulting the use of heat transfer fluids in the total estimates of F-GHG emissions from semiconductor manufacturing. A point of consideration for future Inventory

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[260] Based upon information in the WFF, it appears that a small portion of GHGRP semiconductor reporters are manufacturing both semiconductors and MEMS; however, these reporters are only reporting semiconductor emissions.

reports is the inclusion of the uncertainty surrounding these emissions in the source category uncertainty analysis (see also uncertainty and time-series consistency).

4.24 Substitution of Ozone Depleting Substances (CRF Source Category 2F)

Hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) are used as alternatives to several classes of ozone-depleting substances (ODSs) that are being phased out under the terms of the *Montreal Protocol* and the Clean Air Act Amendments of 1990.[261] Ozone depleting substances—chlorofluorocarbons (CFCs), halons, carbon tetrachloride, methyl chloroform, and hydrochlorofluorocarbons (HCFCs)—are used in a variety of industrial applications including refrigeration and air conditioning equipment, solvent cleaning, foam production, sterilization, fire extinguishing, and aerosols. Although HFCs and PFCs are not harmful to the stratospheric ozone layer, they are potent greenhouse gases. Emission estimates for HFCs and PFCs used as substitutes for ODSs are provided in Table 4-99 and Table 4-100.[262]

Table 4-99: Emissions of HFCs and PFCs from ODS Substitutes (MMT CO_2 Eq.)

Gas	1990	2005	2013	2014	2015	2016	2017
HFC-23	0.0	+	+	+	+	+	+
HFC-32	0.0	0.3	2.8	3.4	3.9	4.6	5.3
HFC-125	+	9.0	36.5	40.0	43.4	47.0	50.0
HFC-134a	+	75.8	65.3	63.1	61.1	57.7	54.1
HFC-143a	+	9.4	25.7	26.9	27.6	28.3	28.0
HFC-236fa	0.0	1.2	1.4	1.4	1.3	1.2	1.2
CF_4	0.0	+	+	+	+	+	+
Others[a]	0.3	6.5	10.0	10.4	11.7	12.9	14.0
Total	**0.3**	**102.1**	**141.7**	**145.3**	**149.2**	**151.8**	**152.7**

+ Does not exceed 0.05 MMT CO_2 Eq.

[a] Others represent an unspecified mix of HFCs and PFCs, which includes HFC-152a, HFC-227ea, HFC-245fa, HFC-43-10mee, HCFO-1233zd(E), HFO-1234yf, HFO-1234ze(E), HFO-1336mzz(Z), C_4F_{10}, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications. For estimating purposes, the GWP value used for PFC/PFPEs was based upon C_6F_{14}.

Note: Totals may not sum due to independent rounding.

Table 4-100: Emissions of HFCs and PFCs from ODS Substitution (Metric Tons)

Gas	1990	2005	2013	2014	2015	2016	2017
HFC-23	0	1	2	2	2	2	2
HFC-32	0	397	4,190	5,001	5,841	6,799	7,799
HFC-125	+	2,583	10,415	11,439	12,403	13,416	14,291
HFC-134a	+	52,974	45,644	44,095	42,735	40,358	37,846
HFC-143a	+	2,096	5,749	6,011	6,183	6,326	6,272
HFC-236fa	0	118	147	145	134	127	119
CF_4	0	2	5	5	5	6	6
Others[a]	M	M	M	M	M	M	M

+ Does not exceed 0.5 MT.

M (Mixture of Gases)

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[261] [42 U.S.C § 7671, CAA Title VI]

[262] Emissions of ODS are not included here consistent with UNFCCC reporting guidelines for national inventories noted in Box 4-1. See Annex 6.2 for more details on emissions of ODS.

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[a] Others represent an unspecified mix of HFCs and PFCs, which includes HFC-152a, HFC-227ea, HFC-245fa, HFC-43-10mee, HCFO-1233zd(E), HFO-1234yf, HFO-1234ze(E), HFO-1336mzz(Z), C_4F_{10}, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications.

In 1990 and 1991, the only significant emissions of HFCs and PFCs as substitutes to ODSs were relatively small amounts of HFC-152a—used as an aerosol propellant and also a component of the refrigerant blend R-500 used in chillers. Beginning in 1992, HFC-134a was used in growing amounts as a refrigerant in motor vehicle air-conditioners and in refrigerant blends such as R-404A.[263] In 1993, the use of HFCs in foam production began, and in 1994 ODS substitutes for halons entered widespread use in the United States as halon production was phased out. In 1995, these compounds also found applications as solvents.

The use and subsequent emissions of HFCs and PFCs as ODS substitutes has been increasing from small amounts in 1990 to 152.7 MMT CO_2 Eq. emitted in 2017. This increase was in large part the result of efforts to phase out CFCs and other ODSs in the United States. In the short term, this trend is expected to continue, and will likely continue over the next decade as HCFCs, which are interim substitutes in many applications, are themselves phased-out under the provisions of the Copenhagen Amendments to the *Montreal Protocol*. Improvements in the technologies associated with the use of these gases and the introduction of alternative gases and technologies, however, may help to offset this anticipated increase in emissions.

Table 4-101 presents emissions of HFCs and PFCs as ODS substitutes by end-use sector for 1990 through 2017. The end-use sectors that contributed the most toward emissions of HFCs and PFCs as ODS substitutes in 2017 include refrigeration and air-conditioning (126.8 MMT CO_2 Eq., or approximately 83 percent), aerosols (10.3 MMT CO_2 Eq., or approximately 7 percent), and foams (11.2 MMT CO_2 Eq., or approximately 7 percent). Within the refrigeration and air-conditioning end-use sector, large retail food was the highest emitting end-use (34.3 MMT CO_2 Eq.), followed by motor vehicle air-conditioning. Each of the end-use sectors is described in more detail below.

Table 4-101: Emissions of HFCs and PFCs from ODS Substitutes (MMT CO_2 Eq.) by Sector

Sector	1990	2005	2013	2014	2015	2016	2017
Refrigeration/Air Conditioning	+	89.7	120.0	122.5	124.8	126.5	126.8
Aerosols	0.3	7.6	10.5	10.8	11.0	10.7	10.3
Foams	+	2.1	7.4	7.9	9.3	10.3	11.2
Solvents	+	1.7	1.8	1.8	1.8	1.9	1.9
Fire Protection	+	1.1	2.1	2.2	2.3	2.4	2.5
Total	**0.3**	**102.1**	**141.7**	**145.3**	**149.2**	**151.8**	**152.7**

+ Does not exceed 0.05 MMT CO_2 Eq.
Note: Totals may not sum due to independent rounding.

Refrigeration/Air Conditioning

The refrigeration and air-conditioning sector includes a wide variety of equipment types that have historically used CFCs or HCFCs. End-uses within this sector include motor vehicle air-conditioning, retail food refrigeration, refrigerated transport (e.g., ship holds, truck trailers, railway freight cars), household refrigeration, residential and small commercial air-conditioning and heat pumps, chillers (large comfort cooling), cold storage facilities, and industrial process refrigeration (e.g., systems used in food processing, chemical, petrochemical, pharmaceutical, oil and gas, and metallurgical industries). As the ODS phaseout has taken effect, most equipment has been retrofitted or replaced to use HFC-based substitutes. Common HFCs in use today in refrigeration/air-conditioning equipment are HFC-134a, R-410A,[264] R-404A, and R-507A.[265] Lower-GWP options such as hydrofluoroolefin (HFO)-1234yf in motor vehicle air-conditioning, R-717 (ammonia) in cold storage and industrial applications, and R-744 (carbon dioxide) and HFC/HFO blends in retail food refrigeration, are also being used. These refrigerants are emitted to the

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[263] R-404A contains HFC-125, HFC-143a, and HFC-134a.

[264] R-410A contains HFC-32 and HFC-125.

[265] R-507A, also called R-507, contains HFC-125 and HFC-143a.

atmosphere during equipment manufacture and operation (as a result of component failure, leaks, and purges), as well as at servicing and disposal events.

Aerosols

Aerosol propellants are used in metered dose inhalers (MDIs) and a variety of personal care products and technical/specialty products (e.g., duster sprays and safety horns). Many pharmaceutical companies that produce MDIs—a type of inhaled therapy used to treat asthma and chronic obstructive pulmonary disease—have replaced the use of CFCs with HFC-propellant alternatives. The earliest ozone-friendly MDIs were produced with HFC-134a, but the industry has started to use HFC-227ea as well. Conversely, since the use of CFC propellants was banned in 1978, most non-medical consumer aerosol products have not transitioned to HFCs, but to “not-in-kind” technologies, such as solid or roll-on deodorants and finger-pump sprays. The transition away from ODS in specialty aerosol products has also led to the introduction of non-fluorocarbon alternatives (e.g., hydrocarbon propellants) in certain applications, in addition to HFC-134a or HFC-152a. Other low-GWP options such as HFO-1234ze(E) are being used as well. These propellants are released into the atmosphere as the aerosol products are used.

Foams

Chlorofluorocarbons and HCFCs have traditionally been used as foam blowing agents to produce polyurethane (PU), polystyrene, polyolefin, and phenolic foams, which are used in a wide variety of products and applications. Since the *Montreal Protocol*, flexible PU foams as well as other types of foam, such as polystyrene sheet, polyolefin, and phenolic foam, have transitioned almost completely away from fluorocompounds, into alternatives such as CO_2 and hydrocarbons. The majority of rigid PU foams have transitioned to HFCs—primarily HFC-134a and HFC-245fa. Today, these HFCs are used to produce PU appliance, PU commercial refrigeration, PU spray, and PU panel foams—used in refrigerators, vending machines, roofing, wall insulation, garage doors, and cold storage applications. In addition, HFC-152a, HFC-134a and CO_2 are used to produce polystyrene sheet/board foam, which is used in food packaging and building insulation. Low-GWP fluorinated foam blowing agents in use include HFO-1234ze(E) and HCFO-1233zd(E). Emissions of blowing agents occur when the foam is manufactured as well as during the foam lifetime and at foam disposal, depending on the particular foam type.

Solvents

Chlorofluorocarbons, methyl chloroform (1,1,1-trichloroethane or TCA), and to a lesser extent carbon tetrachloride (CCl_4) were historically used as solvents in a wide range of cleaning applications, including precision, electronics, and metal cleaning. Since their phaseout, metal cleaning end-use applications have primarily transitioned to non-fluorocarbon solvents and not-in-kind processes. The precision and electronics cleaning end-uses have transitioned in part to high-GWP gases, due to their high reliability, excellent compatibility, good stability, low toxicity, and selective solvency. These applications rely on HFC-43-10mee, HFC-365mfc, HFC-245fa, and to a lesser extent, PFCs. Electronics cleaning involves removing flux residue that remains after a soldering operation for printed circuit boards and other contamination-sensitive electronics applications. Precision cleaning may apply to either electronic components or to metal surfaces, and is characterized by products, such as disk drives, gyroscopes, and optical components, that require a high level of cleanliness and generally have complex shapes, small clearances, and other cleaning challenges. The use of solvents yields fugitive emissions of these HFCs and PFCs.

Fire Protection

Fire protection applications include portable fire extinguishers (“streaming” applications) that originally used halon 1211, and total flooding applications that originally used halon 1301, as well as some halon 2402. Since the production and import of virgin halons were banned in the United States in 1994, the halon replacement agent of choice in the streaming sector has been dry chemical, although HFC-236fa is also used to a limited extent. In the total flooding sector, HFC-227ea has emerged as the primary replacement for halon 1301 in applications that require clean agents. Other HFCs, such as HFC-23 and HFC-125, are used in smaller amounts. The majority of HFC-227ea in total flooding systems is used to protect essential electronics, as well as in civil aviation, military mobile weapons systems, oil/gas/other process industries, and merchant shipping. Fluoroketone FK-5-1-12 is also used as a low-

GWP option and 2-BTP is being considered. As fire protection equipment is tested or deployed, emissions of HFCs occur.

Methodology

A detailed Vintaging Model of ODS-containing equipment and products was used to estimate the actual—versus potential—emissions of various ODS substitutes, including HFCs and PFCs. The name of the model refers to the fact that it tracks the use and emissions of various compounds for the annual "vintages" of new equipment that enter service in each end-use. The Vintaging Model predicts ODS and ODS substitute use in the United States based on modeled estimates of the quantity of equipment or products sold each year containing these chemicals and the amount of the chemical required to manufacture and/or maintain equipment and products over time. Emissions for each end-use were estimated by applying annual leak rates and release profiles, which account for the lag in emissions from equipment as they leak over time. By aggregating the data for 67 different end-uses, the model produces estimates of annual use and emissions of each compound. Further information on the Vintaging Model is contained in Annex 3.9.

Uncertainty and Time-Series Consistency

Given that emissions of ODS substitutes occur from thousands of different kinds of equipment and from millions of point and mobile sources throughout the United States, emission estimates must be made using analytical tools such as the Vintaging Model or the methods outlined in IPCC (2006). Though the model is more comprehensive than the IPCC default methodology, significant uncertainties still exist with regard to the levels of equipment sales, equipment characteristics, and end-use emissions profiles that were used to estimate annual emissions for the various compounds.

The uncertainty analysis quantifies the level of uncertainty associated with the aggregate emissions across the 67 end-uses in the Vintaging Model. In order to calculate uncertainty, functional forms were developed to simplify some of the complex "vintaging" aspects of some end-use sectors, especially with respect to refrigeration and air-conditioning, and to a lesser degree, fire extinguishing. These sectors calculate emissions based on the entire lifetime of equipment, not just equipment put into commission in the current year, thereby necessitating simplifying equations. The functional forms used variables that included growth rates, emission factors, transition from ODSs, change in charge size as a result of the transition, disposal quantities, disposal emission rates, and either stock for the current year or original ODS consumption. Uncertainty was estimated around each variable within the functional forms based on expert judgment, and a Monte Carlo analysis was performed. The most significant sources of uncertainty for this source category include the total stock of refrigerant installed in industrial process refrigeration and cold storage equipment, as well as the percent of non-MDI aerosol propellant that is HFC-152a.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-102. Substitution of ozone depleting substances HFC and PFC emissions were estimated to be between 152.6 and 171.5 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 0.04 percent below to 12.3 percent above the emission estimate of 152.7 MMT CO_2 Eq.

Table 4-102: Approach 2 Quantitative Uncertainty Estimates for HFC and PFC Emissions from ODS Substitutes (MMT CO_2 Eq. and Percent)

Source	Gases	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Substitution of Ozone Depleting Substances	HFCs and PFCs	152.7	152.6	171.5	-0.04%	+12.3%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure time-series consistency from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Comparison of Reported Consumption to Modeled Consumption of HFCs

Data from EPA's Greenhouse Gas Reporting Program (GHGRP) was also used to perform quality control as a reference scenario check on the modeled emissions from this source category as specified in *2006 IPCC Guidelines for National Greenhouse Gas Inventories*. To do so, consumption patterns demonstrated through data reported under GHGRP Subpart OO—Suppliers of Industrial Greenhouse Gases and Subpart QQ—Importers and Exporters of Fluorinated Greenhouse Gases Contained in Pre-Charged Equipment or Closed-Cell Foams were compared to the modeled demand for new saturated HFCs (excluding HFC-23) used as ODS substitutes from the Vintaging Model. The collection of data from suppliers of HFCs enables EPA to calculate the reporters' aggregated net supply–the sum of the quantities of chemical produced or imported into the United States less the sum of the quantities of chemical transformed (used as a feedstock in the production of other chemicals), destroyed, or exported from the United States.[266] This allows for a quality control check on emissions from this source because the Vintaging Model uses modeled demand for new chemical as a proxy for total amount supplied, which is similar to net supply, as an input to the emission calculations in the model.

Reported Net Supply (GHGRP Top-Down Estimate)

Under EPA's GHGRP, suppliers (i.e., producers, importers, and exporters) of HFCs under Subpart OO began annually reporting their production, transformation, destruction, imports, and exports to EPA in 2011 (for supply that occurred in 2010) and suppliers of HFCs under Subpart QQ began annually reporting their imports and exports to EPA in 2012 (for supply that occurred in 2011). Beginning in 2015, bulk consumption data for aggregated HFCs reported under Subpart OO were made publicly available under EPA's GHGRP. Data include all saturated HFCs (except HFC-23) reported to EPA across the GHGRP-reporting time series (2010 through 2016). The data include all 26 such saturated HFCs listed in Table A-1 of 40 CFR Part 98, where regulations for EPA's GHGRP are promulgated, though not all species were reported in each reporting year. For the first time in 2016, net imports of HFCs contained in pre-charged equipment or closed-cell foams reported under Subpart QQ were made publicly available under EPA's GHGRP.

Modeled Consumption (Vintaging Model Bottom-Up Estimate)

The Vintaging Model, used to estimate emissions from this source category, calculates chemical demand based on the quantity of equipment and products sold, serviced and retired each year, and the amount of the chemical required to manufacture and/or maintain the equipment and products.[267] It is assumed that the total demand equals the amount supplied by either new production, chemical import, or quantities recovered (usually reclaimed) and placed back on the market. In the Vintaging Model, demand for new chemical, as a proxy for consumption, is calculated as any chemical demand (either for new equipment or for servicing existing equipment) that cannot be met through recycled or recovered material. No distinction is made in the Vintaging Model between whether that need is met through domestic production or imports. To calculate emissions, the Vintaging Model estimates the quantity

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[266] Chemical that is exported, transformed, or destroyed—unless otherwise imported back to the United States—will never be emitted in the United States.

[267] The model builds an inventory of the in-use stock of equipment and products and ODSs and HFCs in each of the sub-applications. Emissions are subsequently estimated by applying annual and disposal emission rates to each population of equipment and products.

released from equipment over time. Thus, verifying the Vintaging Model's calculated consumption against GHGRP reported data is one way to check the Vintaging Model's emission estimates.

There are ten saturated HFC species modeled in the Vintaging Model: HFC-23, HFC-32, HFC-125, HFC-134a, HFC-143a, HFC-152a, HFC-227ea, HFC-236fa, HFC-245fa, and HFC-43-10mee. For the purposes of this comparison, only nine HFC species are included (HFC-23 is excluded), to more closely align with the aggregated total reported under EPA's GHGRP. While some amounts of less-used saturated HFCs, including isomers of those included in the Vintaging Model, are reportable under EPA's GHGRP, the data are believed to represent an amount comparable to the modeled estimates as a quality control check.

Comparison Results and Discussion

Comparing the estimates of consumption from these two approaches (i.e., reported and modeled) ultimately supports and improves estimates of emissions, as noted in the *2006 IPCC Guidelines* (which refer to fluorinated greenhouse gas consumption based on supplies as "potential emissions"):

> [W]hen considered along with estimates of actual emissions, the potential emissions approach can assist in validation of completeness of sources covered and as a QC check by comparing total domestic consumption as calculated in this 'potential emissions approach' per compound with the sum of all activity data of the various uses (IPCC 2006).

Table 4-103 and Figure 4-2 compare the published net supply of saturated HFCs (excluding HFC-23) in MMT CO_2 Eq. as determined from Subpart OO (supply of HFCs in bulk) and Subpart QQ (supply of HFCs in products and foams) of EPA's GHGRP for the years 2010 through 2017 (U.S. EPA 2019a) and the chemical demand as calculated by the Vintaging Model for the same time series. 2017 Subpart QQ GHGRP values are not yet publicly available and are proxied to the average of 2010 through 2016 estimates.

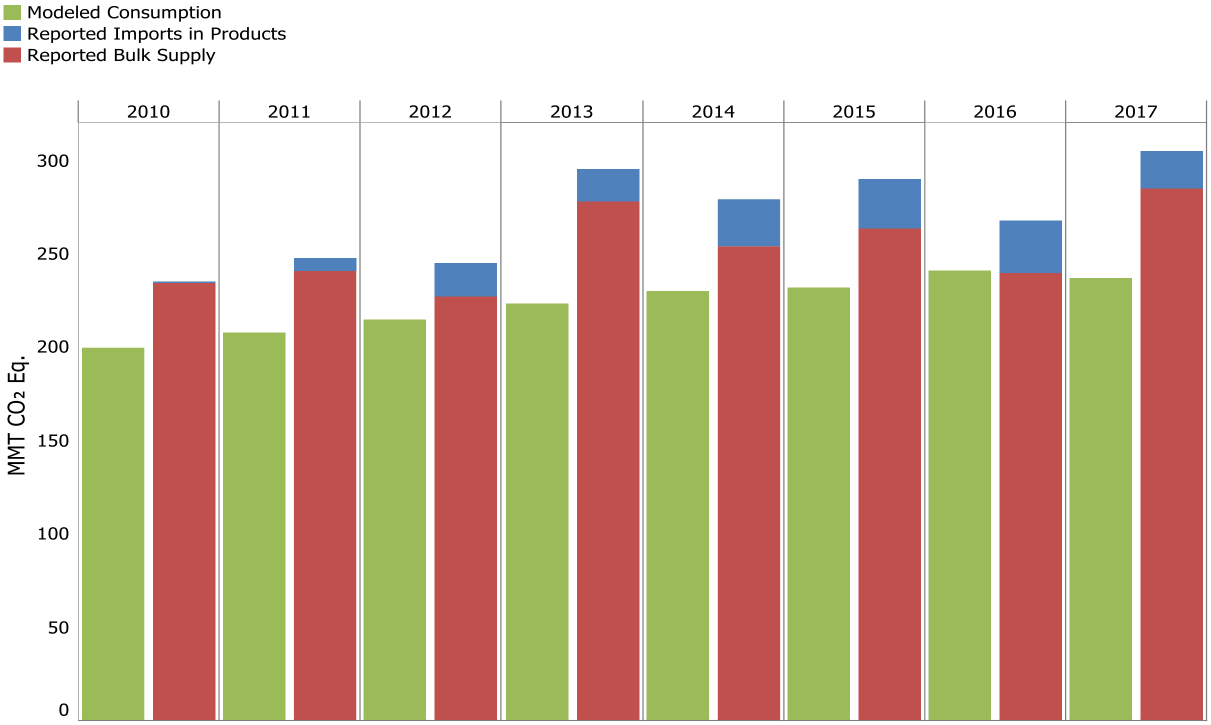
Table 4-103: U.S. HFC Supply (MMT CO_2 Eq.)

	2010	2011	2012	2013	2014	2015	2016	2017
Reported Net Supply (GHGRP)	235	248	245	295	279	290	268	305
Industrial GHG Suppliers	235	241	227	278	254	264	240	285
HFCs in Products and Foams	NA[a]	7	18	17	25	26	28	20
Modeled Supply (Vintaging Model)	200	207	214	223	230	232	241	237
Percent Difference	-15%	-16%	-13%	-24%	-18%	-20%	-10%	-22%

NA (Not Available)

[a] Importers and exporters of fluorinated gases in products were not required to report 2010 data.

Figure 4-2: U.S. HFC Consumption (MMT CO_2 Eq.)



As shown, the estimates from the Vintaging Model are lower than the GHGRP estimates by an average of 17 percent across the time series (i.e., 2010 through 2017). This difference is significantly greater than that reported in the previous Inventory, due to a lower model estimate of consumption. The lower model estimates stem primarily from changes made during a peer review of the Vintaging Model (see Recalculations Discussion below), calling into question the accuracy and thoroughness of the changes made. Irrespective of these changes, potential reasons for the differences between the reported and modeled data, include:

- The Vintaging Model includes fewer saturated HFCs than are reported to EPA's GHGRP. However, the additional reported HFCs represent a small fraction of total HFC use for this source category, both in GWP-weighted and unweighted terms, and as such, it is not expected that the additional HFCs reported to EPA are a major driver for the difference between the two sets of estimates. To the extent lower-GWP isomers were used in lieu of the modeled chemicals (e.g., HFC-134 instead of HFC-134a), lower CO_2 Eq. amounts in the GHGRP data compared to the modeled estimates would be expected.
- Because the top-down data are reported at the time of actual production or import, and the bottom-up data are calculated at the time of actual placement on the market, there could be a temporal discrepancy when comparing data. Because the GHGRP data generally increases over time (although some year-to-year variations exist) and the Vintaging Model estimates also increase (through 2016), EPA would expect the modeled estimates to be slightly lower than the corresponding GHGRP data due to this temporal effect.
- An additional temporal effect can result from the stockpiling of chemicals by suppliers and distributors. Suppliers might decide to produce or import additional quantities of HFCs for various reasons such as expectations that prices may increase or supplies may decrease in the future. Such stockpiling behavior was seen during ODS phasedowns, but it is unclear if such behavior exists amongst HFC suppliers in anticipation of potential future controls on HFCs. Any such activity would increase the GHGRP data as compared to the modeled data.
- Under EPA's GHGRP, all facilities that produce HFCs are required to report their quantities, whereas importers or exporters of HFCs or pre-charged equipment and closed-cell foams that contain HFCs are only

required to report if either their total imports or their total exports of greenhouse gases are greater than or equal to 25,000 metric tons of CO_2 Eq. per year. Thus, some imports may not be accounted for in the GHGRP data. On the other hand, some exports might also not be accounted for in this data.

- In some years, imports and exports may be greater than consumption because the excess is being used to increase chemical or equipment stockpiles as discussed above; in other years, the opposite may hold true. Similarly, relocation of manufacturing facilities or recovery from the recession could contribute to variability in imports or exports. Averaging imports and exports over multiple years can minimize the impact of such fluctuations. For example, when the 2012 and 2013 net additions to the supply are averaged, as shown in Table 4-104, the percent difference between the consumption estimates decreases compared to the 2013-only estimates.

Table 4-104: Averaged U.S. HFC Demand (MMT CO_2 Eq.)

	2010-2011 Avg.	2011-2012 Avg.	2012-2013 Avg.	2013-2014 Avg.	2014-2015 Avg.	2015-2016 Avg.	2016-2017 Avg.
Reported Net Supply (GHGRP)	242	247	270	287	285	279	287
Modeled Demand (Vintaging Model)	204	211	219	227	231	236	239
Percent Difference	-16%	-14%	-19%	-21%	-19%	-15%	-17%

- The Vintaging Model does not reflect the dynamic nature of reported HFC consumption, with significant differences seen in each year. Whereas the Vintaging Model projects a slowly increasing overall demand through 2016, and a slight lowering after that, actual consumption for specific chemicals or equipment may vary over time and could even switch from positive to negative (indicating more chemical exported, transformed, or destroyed than produced or imported in a given year). Furthermore, consumption as calculated in the Vintaging Model is a function of demand not met by disposal recovery. If, in any given year, a significant number of units are disposed, there will be a large amount of additional recovery in that year that can cause an unexpected and not modeled decrease in demand and thus a decrease in consumption. On the other hand, if market, economic, or other factors cause less than expected disposal and recovery, actual supply would decrease, and hence consumption would increase to meet that demand not satisfied by recovered quantities, increasing the GHGRP amounts.
- The Vintaging Model is used to estimate the emissions that occur in the United States. As such, all equipment or products that contain ODS or alternatives, including saturated HFCs, are assumed to consume and emit chemicals equally as like equipment or products originally produced in the United States. The GHGRP data from Subpart OO (industrial greenhouse gas suppliers) includes HFCs produced or imported and used to fill or manufacture products that are then exported from the United States. The Vintaging Model estimates of demand and supply are not meant to incorporate such chemical. Likewise, chemicals may be used outside the United States to create products or charge equipment that is then imported to and used in the United States. The Vintaging Model estimates of demand and supply are meant to capture this chemical, as it will lead to emissions inside the United States. The GHGRP data from Subpart QQ (supply of HFCs in products) accounts for some of these differences; however, the scope of Subpart QQ does not cover all such equipment or products and the chemical contained therein. Depending on whether the United States is a net importer or net exporter of such chemical, this factor may account for some of the difference shown above or might lead to a further discrepancy.

One factor, however, would only lead to modeled estimates to be even higher than the estimates shown and hence for some years possibly higher than GHGRP data:

- Saturated HFCs are also known to be used as a cover gas in the production of magnesium. The Vintaging Model estimates here do not include the amount of HFCs for this use, but rather only the amount for uses that traditionally were served by ODS. Nonetheless, EPA expects this supply not included in the Vintaging Model estimates to be very small compared to the ODS substitute use for the years analyzed. An indication of the different magnitudes of these categories is seen in the fact that the 2017 emissions from that non-modeled source (0.1 MMT CO_2 Eq.) are much smaller than those for the ODS substitute sector (152.7 MMT CO_2 Eq.).

Using a Tier 2 bottom-up modeling methodology to estimate emissions requires assumptions and expert judgment. Comparing the Vintaging Model's estimates to GHGRP-reported estimates, particularly for more widely used chemicals, can help validate the model but it is expected that the model will have limitations. This comparison shows that Vintaging Model consumption estimates are well within the same order of magnitude as the actual consumption data as reported to EPA's GHGRP although the differences in reported net supply and modeled demand are still significant. Although it can be difficult to capture the observed market variability, the Vintaging Model is periodically reviewed and updated to ensure that the model reflects the current and future trajectory of ODS and ODS substitutes across all end-uses and the Vintaging Model will continue to be compared to available top-down estimates in order to ensure the model accurately estimates HFC consumption and emissions.

Recalculations Discussion

For the current Inventory, updates to the Vintaging Model were included in response to a peer review conducted on end-uses within the Refrigeration/Air Conditioning and Fire Protection sectors. (EPA 2018).

In the Refrigeration/Air Conditioning sector, updates included revisions to servicing leak rate assumptions for light-duty vehicle and light-duty truck air conditioners and updates to the annual leak rate for road transport refrigeration systems containing HFC refrigerant.

For the unitary air conditioning end-uses, charge sizes were adjusted for residential unitary systems, annual loss rates were reduced for small and large commercial unitary AC systems, and disposal loss rates were reduced for residential and small and large commercial unitary systems. In addition, HCFC-22 dry-shipped condensing units were added to the residential unitary air conditioning end-use.

Within the Fire Protection sector, replacement ratios, growth rates, and annual loss rates for total flooding agents and market transitions and lifetimes for total flooding and streaming agents were updated in response to the peer review and comments received during the Public Review comment period for the 2017 Inventory (i.e., 1990 through 2015 report) for the Fire Protection sector.

Further improvements to the Vintaging Model were included in the current Inventory to reflect findings from stakeholder outreach and research on the integral skin foam end-use. The integral skin foam blowing agent transitions were revised to include HFC-245fa, hydrocarbons, oxygenated hydrocarbons, and HFOs. Current assumptions in the domestic refrigeration foam end-use, including HFC and alternative blowing agent market penetration and historical, current, and projected shipments of appliances, were also reviewed against recent data from the Association of Home Appliance Manufacturers (AHAM) and EPA's Responsible Appliance Disposal (RAD) program, but it was determined that current assumptions in the Vintaging Model were accurate and did not require updates (EPA 2019b).

In response to comments from the UNFCCC Expert Review Team, initial (i.e., first-fill) emissions that occur during manufacture or installation of Ref/AC equipment were modeled. Per IPCC (2006) guidance, first-fill emissions were considered for all Ref/AC equipment that are charged with refrigerant within the United States, including those which are produced for export, and excluding those that are imported pre-charged. (EPA 2019c)

Together, these updates decreased greenhouse gas emissions on average by 1.1 percent between 1990 and 2016.

Planned Improvements

Future improvements to the Vintaging Model are planned for the Foam Blowing and Aerosols sectors. A review of blowing agent transition assumptions for Commercial Refrigeration Foam and the disaggregation of the rigid polyurethane (PU): spray foam end-use into low-pressure, two-component spray foam and high-pressure, two-component spray foam are anticipated to be completed by the 2020 submission.

The non-metered dose inhaler (non-MDI) aerosol end-use may be renamed to consumer aerosol and stock and emission estimates will be updated to align with a recent national market characterization. In addition, a technical aerosol end-use may be added to the aerosols sector, in order to capture a portion of the market that may not be adequately encompassed by the current non-MDI aerosol end-use. These updates are anticipated to be completed by the 2020 submission.

4.25 Electrical Transmission and Distribution (CRF Source Category 2G1)

The largest use of sulfur hexafluoride (SF_6), both in the United States and internationally, is as an electrical insulator and interrupter in equipment that transmits and distributes electricity (RAND 2004). The gas has been employed by the electric power industry in the United States since the 1950s because of its dielectric strength and arc-quenching characteristics. It is used in gas-insulated substations, circuit breakers, and other switchgear. SF_6 has replaced flammable insulating oils in many applications and allows for more compact substations in dense urban areas.

Fugitive emissions of SF_6 can escape from gas-insulated substations and switchgear through seals, especially from older equipment. The gas can also be released during equipment manufacturing, installation, servicing, and disposal. Emissions of SF_6 from equipment manufacturing and from electrical transmission and distribution systems were estimated to be 4.3 MMT CO_2 Eq. (0.2 kt) in 2017. This quantity represents an 81 percent decrease from the estimate for 1990 (see Table 4-105 and Table 4-106). There are two potential causes for this decrease: a sharp increase in the price of SF_6 during the 1990s and a growing awareness of the magnitude and environmental impact of SF_6 emissions through programs such as EPA's voluntary SF_6 Emission Reduction Partnership for Electric Power Systems (Partnership) and EPA's GHGRP. Utilities participating in the Partnership have lowered their emission factor (kg SF_6 emitted per kg of nameplate capacity) by more than 86 percent since the Partnership began in 1999. A recent examination of the SF_6 emissions reported by electric power systems to EPA's GHGRP revealed that SF_6 emissions from reporters have decreased by 31 percent from 2011 to 2017,[268] with much of the reduction seen from utilities that are not participants in the Partnership. These utilities may be making relatively large reductions in emissions as they take advantage of relatively large and/or inexpensive emission reduction opportunities (i.e., "low hanging fruit," such as replacing major leaking circuit breakers) that Partners have already taken advantage of under the voluntary program (Ottinger et al. 2014).

Table 4-105: SF_6 Emissions from Electric Power Systems and Electrical Equipment Manufacturers (MMT CO_2 Eq.)

Year	Electric Power Systems	Electrical Equipment Manufacturers	Total
1990	22.8	0.3	23.1
2005	7.7	0.7	8.3
2013	4.0	0.4	4.4
2014	4.2	0.4	4.6
2015	3.8	0.3	4.1
2016	4.1	0.3	4.4
2017	4.0	0.3	4.3

Note: Totals may not sum due to independent rounding.

Table 4-106: SF_6 Emissions from Electric Power Systems and Electrical Equipment Manufacturers (kt)

Year	Emissions
1990	1.0
2005	0.4

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[268] Analysis of emission trends from the GHGRP is imperfect due to an inconsistent group of reporters year to year.

2013	0.2
2014	0.2
2015	0.2
2016	0.2
2017	0.2

Methodology

The estimates of emissions from Electrical Transmission and Distribution are comprised of emissions from electric power systems and emissions from the manufacture of electrical equipment. The methodologies for estimating both sets of emissions are described below.

1990 through 1998 Emissions from Electric Power Systems

Emissions from electric power systems from 1990 through 1998 were estimated based on (1) the emissions estimated for this source category in 1999, which, as discussed in the next section, were based on the emissions reported during the first year of EPA's SF_6 Emission Reduction Partnership for Electric Power Systems (Partnership), and (2) the RAND survey of global SF_6 emissions. Because most utilities participating in the Partnership reported emissions only for 1999 through 2011, modeling was used to estimate SF_6 emissions from electric power systems for the years 1990 through 1998. To perform this modeling, U.S. emissions were assumed to follow the same trajectory as global emissions from this source during the 1990 to 1999 period. To estimate global emissions, the RAND survey of global SF_6 sales were used, together with the following equation for estimating emissions, which is derived from the mass-balance equation for chemical emissions (Volume 3, Equation 7.3) in the *2006 IPCC Guidelines*.[269] (Although Equation 7.3 of the *2006 IPCC Guidelines* appears in the discussion of substitutes for ozone-depleting substances, it is applicable to emissions from any long-lived pressurized equipment that is periodically serviced during its lifetime.)

Emissions (kilograms SF_6) = SF_6 purchased to refill existing equipment (kilograms) + nameplate capacity of retiring equipment (kilograms) [270]

Note that the above equation holds whether the gas from retiring equipment is released or recaptured; if the gas is recaptured, it is used to refill existing equipment, thereby lowering the amount of SF_6 purchased by utilities for this purpose.

Gas purchases by utilities and equipment manufacturers from 1961 through 2003 are available from the RAND (2004) survey. To estimate the quantity of SF_6 released or recovered from retiring equipment, the nameplate capacity of retiring equipment in a given year was assumed to equal 81.2 percent of the amount of gas purchased by electrical equipment manufacturers 40 years previous (e.g., in 2000, the nameplate capacity of retiring equipment was assumed to equal 81.2 percent of the gas purchased in 1960). The remaining 18.8 percent was assumed to have been emitted at the time of manufacture. The 18.8 percent emission factor is an average of IPCC default SF_6 emission rates for Europe and Japan for 1995 (IPCC 2006). The 40-year lifetime for electrical equipment is also based on IPCC (2006). The results of the two components of the above equation were then summed to yield estimates of global SF_6 emissions from 1990 through 1999.

U.S. emissions between 1990 and 1999 are assumed to follow the same trajectory as global emissions during this period. To estimate U.S. emissions, global emissions for each year from 1990 through 1998 were divided by the estimated global emissions from 1999. The result was a time series of factors that express each year's global emissions as a multiple of 1999 global emissions. Historical U.S. emissions were estimated by multiplying the factor for each respective year by the estimated U.S. emissions of SF_6 from electric power systems in 1999 (estimated to be 14.3 MMT CO_2 Eq.).

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[269] Ideally, sales to utilities in the United States between 1990 and 1999 would be used as a model. However, this information was not available. There were only two U.S. manufacturers of SF_6 during this time period, so it would not have been possible to conceal sensitive sales information by aggregation.

[270] Nameplate capacity is defined as the amount of SF_6 within fully charged electrical equipment.

Two factors may affect the relationship between the RAND sales trends and actual global emission trends. One is utilities' inventories of SF_6 in storage containers. When SF_6 prices rise, utilities are likely to deplete internal inventories before purchasing new SF_6 at the higher price, in which case SF_6 sales will fall more quickly than emissions. On the other hand, when SF_6 prices fall, utilities are likely to purchase more SF_6 to rebuild inventories, in which case sales will rise more quickly than emissions. This effect was accounted for by applying 3-year smoothing to utility SF_6 sales data. The other factor that may affect the relationship between the RAND sales trends and actual global emissions is the level of imports from and exports to Russia and China. SF_6 production in these countries is not included in the RAND survey and is not accounted for in any another manner by RAND. However, atmospheric studies confirm that the downward trend in estimated global emissions between 1995 and 1998 was real (see the Uncertainty discussion below).

1999 through 2017 Emissions from Electric Power Systems

Emissions from electric power systems from 1999 to 2017 were estimated based on: (1) reporting from utilities participating in EPA's SF_6 Emission Reduction Partnership for Electric Power Systems (Partners), which began in 1999; (2) reporting from utilities covered by EPA's GHGRP, which began in 2012 for emissions occurring in 2011 (GHGRP-Only Reporters); and (3) the relationship between utilities' reported emissions and their transmission miles as reported in the 2001, 2004, 2007, 2010, 2013, and 2017 Utility Data Institute (UDI) Directories of Electric Power Producers and Distributors (UDI 2001, 2004, 2007, 2010, 2013, and 2017), which was applied to the electric power systems that do not report to EPA (Non-Reporters). (Transmission miles are defined as the miles of lines carrying voltages above 34.5 kV).

Partners

Over the period from 1999 to 2017, Partner utilities, which for inventory purposes are defined as utilities that either currently are or previously have been part of the Partnership,[271] represented 50 percent, on average, of total U.S. transmission miles. Partner utilities estimated their emissions using a Tier 3 utility-level mass balance approach (IPCC 2006). If a Partner utility did not provide data for a particular year, emissions were interpolated between years for which data were available or extrapolated based on Partner-specific transmission mile growth rates. In 2012, many Partners began reporting their emissions (for 2011 and later years) through EPA's GHGRP (discussed further below) rather than through the Partnership. In 2017, approximately 0.5 percent of the total emissions attributed to Partner utilities were reported through Partnership reports. Approximately 88 percent of the total emissions attributed to Partner utilities were reported and verified through EPA's GHGRP. Partners without verified 2017 data accounted for approximately 11 percent of the total emissions attributed to Partner utilities.[272]

The GHGRP program has an "offramp" provision (40 CFR Part 98.2(i)) that exempts facilities from reporting under certain conditions. If reported total greenhouse gas emissions are below 15,000 metric tons of carbon dioxide equivalent (MT CO_2 Eq.) for three consecutive years or below 25,000 MT CO_2 Eq. for five consecutive years, the facility may elect to discontinue reporting. Partners that are GHGRP reporters and have off-ramped (i.e., non-reporting Partners), are still treated as Partners, and estimates are gap-filled based on the methodology as described in this section.

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[271] Starting in the 1990 to 2015 Inventory, partners who had reported three years or less of data prior to 2006 were removed. Most of these Partners had been removed from the list of current Partners, but remained in the Inventory due to the extrapolation methodology for non-reporting partners.

[272] Only data reported as of August 20, 2018 are used in the emission estimates for the prior year of reporting. For Partners that did not report to the GHGRP, emissions were extrapolated based upon historical Partner-specific transmission mile growth rates, and those Partners are included in the 'non-reporting Partners' category.

It should be noted that data reported through EPA's GHGRP must go through a verification process. For electric power systems, verification involved a series of electronic range, completeness, and algorithm checks for each report submitted. In addition, EPA manually reviewed the reported data and compared each facility's reported transmission miles with the corresponding quantity in the UDI 2017 database (UDI 2017). In the first year of GHGRP reporting, EPA followed up with reporters where the discrepancy between the reported miles and the miles published by UDI was greater than 10 percent, with a goal to improve data quality.

GHGRP-Only Reporters

EPA's GHGRP requires users of SF_6 in electric power systems to report emissions if the facility has a total SF_6 nameplate capacity that exceeds 17,820 pounds. (This quantity is the nameplate capacity that would result in annual SF_6 emissions equal to 25,000 metric tons of CO_2 equivalent at the historical emission rate reported under the Partnership.) As under the Partnership, electric power systems that report their SF_6 emissions under EPA's GHGRP are required to use the Tier 3 utility-level mass-balance approach. Many Partners began reporting their emissions through EPA's GHGRP in 2012 (reporting emissions for 2011 and later years) because their nameplate capacity exceeded the reporting threshold. Some Partners who did not report through EPA's GHGRP continued to report through the Partnership.

In addition, many non-Partners began reporting to EPA for the first time through its GHGRP in 2012. Non-Partner emissions reported and verified under EPA's GHGRP were compiled to form a new category of reported data (GHGRP-Only Reporters). GHGRP-Only Reporters accounted for 17 percent of U.S. transmission miles and 21 percent of estimated U.S. emissions from electric power system in 2017.[273]

GHGRP-only reporters that no longer report due to off-ramping are treated as non-reporters, and emissions are subsequently estimated based on the methodology described below.

Non-Reporters

Emissions from Non-Reporters (i.e., utilities other than Partners and GHGRP-Only Reporters) in every year since 1999 were estimated using the results of a regression analysis that correlated emissions from reporting utilities (using verified data from both Partners and GHGRP-Only Reporters) with their transmission miles.[274] As noted above, non-Partner emissions were reported to the EPA for the first time through its GHGRP in 2012 (representing 2011 emissions). This set of reported data was of particular interest because it provided insight into the emission rate of non-Partners, which previously was assumed to be equal to the historical (1999) emission rate of Partners. Specifically, emissions were estimated for Non-Reporters as follows:

- <u>Non-Reporters, 1999 to 2011:</u> First, the 2011 emission rates (per kg nameplate capacity and per transmission mile) reported by Partners and GHGRP-Only Reporters were reviewed to determine whether there was a statistically significant difference between these two groups. Transmission mileage data for 2011 was reported through GHGRP, with the exception of transmission mileage data for Partners that did not report through GHGRP, which was obtained from UDI. It was determined that there is no statistically significant difference between the emission rates of Partners and GHGRP-Only reporters; therefore, Partner and GHGRP-Only reported data for 2011 were combined to develop regression equations to estimate the emissions of Non-Reporters. Historical emissions from Non-Reporters were estimated by linearly interpolating between the 1999 regression coefficient (based on 1999 Partner data) and the 2011 regression coefficient.

- <u>Non-Reporters, 2012 to Present:</u> It was determined that there continued to be no statistically significant difference between the emission rates reported by Partners and by GHGRP-Only Reporters. Therefore, the emissions data from both groups were combined to develop regression equations for 2012. This was repeated for 2013 through 2017 using Partner and GHGRP-Only Reporter data for each year.

    - The 2017 regression equation for reporters was developed based on the emissions reported by a subset of Partner utilities and GHGRP-Only utilities who reported non-zero emissions and non-zero transmission miles (representing approximately 66 percent of total U.S. transmission miles). The regression equation for 2017 is:

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[273] GHGRP-reported and Partner transmission miles from a number of facilities were equal to zero with non-zero emissions. These facilities emissions were added to the emissions totals for their respective parent companies when identifiable and not included in the regression equation when not identifiable or applicable. Other facilities reported non-zero transmission miles with zero emissions, or zero transmission miles and zero emissions. These facilities were not included in the development of the regression equations (discussed further below). These emissions are already implicitly accounted for in the relationship between transmission miles and emissions.

[274] In the United States, SF_6 is contained primarily in transmission equipment rated above 34.5 kV.

$$\text{Emissions (kg)} = 0.226 \times \text{Transmission Miles}$$

Table 4-107 below shows the percentage of transmission miles covered by reporters (i.e., associated with reported data) and the regression coefficient for 1999 (the first year data was reported), and for 2011 through present (the years with GHGRP reported data). The coefficient increased between 2015 and 2017.

Table 4-107: Transmission Mile Coverage (Percent) and Regression Coefficients (kg per mile)

	1999		2005		2013	2014	2015	2016	2017
Percentage of Miles Covered by Reporters	50%		51%		74%	75%	73%	68%	67%
Regression Coefficient[a]	0.71		0.35		0.23	0.23	0.21	0.21	0.23

[a] Regression coefficient for emissions is calculated utilizing transmission miles as the explanatory variable and emissions as the response variable. The equation utilizes a constant intercept of zero. When calculating the regression coefficient, outliers are also removed from the analysis when the standard residual for that reporter exceeds the value 3.0. In 2017, one reporter was removed with abnormally high emissions as compared to the last several years.

Data on transmission miles for each Non-Reporter for the years 2000, 2003, 2006, and 2009, 2012, 2016 and 2017 were obtained from the 2001, 2004, 2007, 2010, 2013, and 2017 UDI Directories of Electric Power Producers and Distributors, respectively (UDI 2001, 2004, 2007, 2010, 2013, and 2017). The following trends in transmission miles have been observed over the time series:

- The U.S. transmission system grew by over 22,000 miles between 2000 and 2003 yet declined by almost 4,000 miles between 2003 and 2006. Given these fluctuations, periodic increases are assumed to occur gradually. Therefore, transmission mileage was assumed to increase at an annual rate of 1.2 percent between 2000 and 2003 and decrease by 0.20 percent between 2003 and 2006.
- The U.S. transmission system's growth rate grew to 1.7 percent from 2006 to 2009 as transmission miles increased by more than 33,000 miles.
- The growth rate for 2009 through 2012 was calculated to be 1.2 percent as transmission miles grew yet again by approximately 24,000 during this time period.
- The annual transmission mile growth rate for 2012 through 2017 was calculated to be 0.9 percent, as transmission miles increased by approximately 26,000 miles.

Total Industry Emissions

As a final step, total electric power system emissions from 1999 through 2017 were determined for each year by summing the Partner reported and estimated emissions (reported data was available through the EPA's SF_6 Emission Reduction Partnership for Electric Power Systems), the GHGRP-Only reported emissions, and the non-reporting utilities' emissions (determined using the regression equations).

1990 through 2017 Emissions from Manufacture of Electrical Equipment

Three different methods were used to estimate 1990 to 2017 emissions from original electrical equipment manufacturers (OEMs).

- OEM emissions from 1990 through 2000 were derived by assuming that manufacturing emissions equaled 10 percent of the quantity of SF_6 provided with new equipment. The 10 percent emission rate is the average of the "ideal" and "realistic" manufacturing emission rates (4 percent and 17 percent, respectively) identified in a paper prepared under the auspices of the International Council on Large Electric Systems (CIGRE) in February 2002 (O'Connell et al. 2002). The quantity of SF_6 provided with new equipment was estimated based on statistics compiled by the National Electrical Manufacturers Association (NEMA). These statistics were provided for 1990 to 2000.
- OEM emissions from 2000 through 2010 were estimated by (1) interpolating between the emission rate estimated for 2000 (10 percent) and an emission rate estimated for 2011 based on reporting by OEMs through the GHGRP (5.8 percent), and (2) estimating the quantities of SF_6 provided with new equipment for 2001 to 2010. The quantities of SF_6 provided with new equipment were estimated using Partner reported data and the total industry SF_6 nameplate capacity estimate (155.48 MMT CO_2 Eq. in 2010). Specifically, the ratio of new nameplate capacity to total nameplate capacity of a subset of Partners for

which new nameplate capacity data was available from 1999 to 2010 was calculated. These ratios were then multiplied by the total industry nameplate capacity estimate for each year to derive the amount of SF_6 provided with new equipment for the entire industry. Additionally, to obtain the 2011 emission rate (necessary for estimating 2001 through 2010 emissions), the estimated 2011 emissions (estimated using the third methodology listed below) were divided by the estimated total quantity of SF_6 provided with new equipment in 2011. The 2011 quantity of SF_6 provided with new equipment was estimated in the same way as the 2001 through 2010 quantities.

- OEM emissions from 2011 through 2017 were estimated using the SF_6 emissions from OEMs reporting to the GHGRP, and an assumption that these reported emissions account for a conservative estimate of 50 percent of the total emissions from all U.S. OEMs.

Uncertainty and Time-Series Consistency

To estimate the uncertainty associated with emissions of SF_6 from Electrical Transmission and Distribution, uncertainties associated with four quantities were estimated: (1) emissions from Partners, (2) emissions from GHGRP-Only Reporters, (3) emissions from Non-Reporters, and (4) emissions from manufacturers of electrical equipment. A Monte Carlo analysis was then applied to estimate the overall uncertainty of the emissions estimate.

Total emissions from the SF_6 Emission Reduction Partnership include emissions from both reporting (through the Partnership or EPA's GHGRP) and non-reporting Partners. For reporting Partners, individual Partner-reported SF_6 data was assumed to have an uncertainty of 10 percent. Based on a Monte Carlo analysis, the cumulative uncertainty of all Partner-reported data was estimated to be 5.2 percent. The uncertainty associated with extrapolated or interpolated emissions from non-reporting Partners was assumed to be 20 percent.

For GHGRP-Only Reporters, reported SF_6 data was assumed to have an uncertainty of 20 percent.[275] Based on a Monte Carlo analysis, the cumulative uncertainty of all GHGRP-Only reported data was estimated to be 8.8 percent.

There are two sources of uncertainty associated with the regression equations used to estimate emissions in 2016 from Non-Reporters: (1) uncertainty in the coefficients (as defined by the regression standard error estimate), and (2) the uncertainty in total transmission miles for Non-Reporters. Uncertainties were also estimated regarding (1) estimates of SF_6 emissions from OEMs reporting to EPA's GHGRP, and (2) the assumption on the percent share of OEM emissions from OEMs reporting to EPA's GHGRP.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-108. Electrical Transmission and Distribution SF_6 emissions were estimated to be between 3.7 and 5.0 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 14 percent below and 17 percent above the emission estimate of 4.3 MMT CO_2 Eq.

Table 4-108: Approach 2 Quantitative Uncertainty Estimates for SF_6 Emissions from Electrical Transmission and Distribution (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to 2017 Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Electrical Transmission and Distribution	SF_6	4.3	3.7	5.0	-14%	+17%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

In addition to the uncertainty quantified above, there is uncertainty associated with using global SF_6 sales data to estimate U.S. emission trends from 1990 through 1999. However, the trend in global emissions implied by sales of SF_6 appears to reflect the trend in global emissions implied by changing SF_6 concentrations in the atmosphere. That

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[275] Uncertainty is assumed to be higher for the GHGRP-Only category, because 2011 is the first year that those utilities have reported to EPA.

is, emissions based on global sales declined by 29 percent between 1995 and 1998 (RAND 2004), and emissions based on atmospheric measurements declined by 17 percent over the same period (Levin et al. 2010).

Several pieces of evidence indicate that U.S. SF_6 emissions were reduced as global emissions were reduced. First, the decreases in sales and emissions coincided with a sharp increase in the price of SF_6 that occurred in the mid-1990s and that affected the United States as well as the rest of the world. A representative from DILO, a major manufacturer of SF_6 recycling equipment, stated that most U.S. utilities began recycling rather than venting SF_6 within two years of the price rise. Finally, the emissions reported by the one U.S. utility that reported its emissions for all the years from 1990 through 1999 under the Partnership showed a downward trend beginning in the mid-1990s.

Methodological approaches were applied to the entire time series to ensure time-series consistency from 1990 through 2016. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Recalculations Discussion

The historical emissions estimated for this source category have undergone some revisions. SF_6 emission estimates for the period 1990 through 2016 were updated relative to the previous report based on revisions to interpolated and extrapolated non-reported Partner data.[276] For the current Inventory, historical estimates for the period 2011 through 2016 were also updated relative to the previous report based on revisions to reported historical data in EPA's GHGRP.

In previous inventory years, non-reporter nameplate capacity was estimated by dividing the non-reporter emissions by the average reporter leak rate. This reliance on calculated emission values to estimate nameplate capacity often results in similar trends between the values. EPA reevaluated this methodology and developed a new approach that relates nameplate capacity directly to transmission miles. Non-reporter nameplate capacity estimates were recalculated by regressing reporter nameplate capacity and reporter transmission miles; the resulting coefficient was applied to non-reporter transmission miles to determine non-reporter nameplate capacity.

Also in previous inventory years, a utility specific transmission miles growth rate was applied to determine transmission miles for instances when a Partner utility did not report for a given year. However, when calculating total transmission miles, a national annual growth rate, based on UDI data, was applied to extrapolate for a Partner that did not report for a given year. These two separate approaches created an inconsistency with transmission mile values used to arrive at a national total estimate and a utility-specific value. To ensure that these values did match, EPA chose to apply the annual growth rate for all utilities to extrapolate for Partners who had not reported for a given year.

As a result of the recalculations, SF_6 emissions from electrical transmission and distribution increased by 0.66 percent for 2016 relative to the previous report, and SF_6 nameplate capacity increased by 5.9 percent for 2016 relative to the previous report. On average, SF_6 emission estimates for the entire time series decreased by approximately 0.45 percent per year.

Planned Improvements

EPA is continuing research to improve the methodology for estimating non-reporter nameplate capacity, specifically the distinction of the nameplate capacity of hermetically-sealed and non-hermetically sealed equipment. The current methodology determines the end of year nameplate capacity by summing the Beginning of Year Nameplate Capacity and the Net Increase in Nameplate Capacity for the GHGRP reporters, which aggregates a small portion of

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[276] The earlier year estimates within the time series (i.e., 1990 through 1998) were updated based on revisions to the 1999 U.S. emission estimate because emissions for 1990 through 1998 are estimated by multiplying a series of annual factors by the estimated U.S. emissions of SF_6 from electric power systems in 1999 (see Methodology section).

hermetically sealed equipment and high-voltage equipment. This calculation is necessary for time-series consistency as the partner-reported data from partnership in the prior years represents the end of year nameplate capacity. Beginning in the 2017 reporting year, EPA's GHGRP required that reporters distinguish between the nameplate capacity of non-hermetically sealed equipment from equipment that is hermetically sealed. EPA is planning to leverage this new reported data to apply an adjustment factor for the GHGRP-reported nameplate capacity totals for 2011 through 2016 to remove the nameplate capacity values attributed to the hermetically-sealed equipment. Reported nameplate capacity totals prior to 2011 can be left as is, since it can be assumed that no hermetically sealed equipment was reported in these totals by partners. This planned improvement will ensure better consistency of the type of equipment nameplate capacity included in the time-series. Additionally, information on the type of new and retiring equipment is expected to provide insight into the relative importance of the two types of equipment as potential emission sources. Historically, hermetically sealed pressure equipment has been considered to be a relatively small source of SF_6 in the United States; however, better estimating its potential source of emissions upon end-of-life (i.e., disposal emissions) is an area for further analysis.

Due to the GHGRP policy that allows reporters to "off-ramp" from the reporting program when their emissions remain below certain levels for certain periods of time (e.g., below 25,000 MT CO_2 Eq. for five years), the number of electric power systems whose reports are used to develop regression coefficients and country-wide emissions estimates is decreasing. While EPA continues to account for emissions from these electric power systems using the estimation method for non-reporters, it is possible that their cessation of reporting could influence the value and/or stability of the emission factors (per transmission mile) that are applied to non-reporters. EPA is planning to explore whether this is the case. If so, EPA is planning to evaluate whether the current methodology for scaling emissions is the best option.

Finally, EPA is exploring the possibility of discontinuing extrapolating emissions for Partners for which reported estimates are not provided for a given length of time, e.g., for more than three or five consecutive years. Emissions from these electric power systems would instead be estimated using the non-reporter methodology.

4.26 Nitrous Oxide from Product Uses (CRF Source Category 2G3)

Nitrous oxide (N_2O) is a clear, colorless, oxidizing liquefied gas with a slightly sweet odor which is used in a wide variety of specialized product uses and applications. The amount of N_2O that is actually emitted depends upon the specific product use or application.

There are a total of three N_2O production facilities currently operating in the United States (Ottinger 2014). Nitrous oxide is primarily used in carrier gases with oxygen to administer more potent inhalation anesthetics for general anesthesia, and as an anesthetic in various dental and veterinary applications. The second main use of N_2O is as a propellant in pressure and aerosol products, the largest application being pressure-packaged whipped cream. Small quantities of N_2O also are used in the following applications:

- Oxidizing agent and etchant used in semiconductor manufacturing;
- Oxidizing agent used, with acetylene, in atomic absorption spectrometry;
- Production of sodium azide, which is used to inflate airbags;
- Fuel oxidant in auto racing; and
- Oxidizing agent in blowtorches used by jewelers and others (Heydorn 1997).

Production of N_2O in 2017 was approximately 15 kt (see Table 4-109).

Table 4-109: N_2O Production (kt)

Year	kt
1990	16
2005	15

2013	15
2014	15
2015	15
2016	15
2017	15

Nitrous oxide emissions were 4.2 MMT CO_2 Eq. (14 kt N_2O) in 2017 (see Table 4-110). Production of N_2O stabilized during the 1990s because medical markets had found other substitutes for anesthetics, and more medical procedures were being performed on an outpatient basis using local anesthetics that do not require N_2O. The use of N_2O as a propellant for whipped cream has also stabilized due to the increased popularity of cream products packaged in reusable plastic tubs (Heydorn 1997).

Table 4-110: N_2O Emissions from N_2O Product Usage (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	4.2	14
2005	4.2	14
2013	4.2	14
2014	4.2	14
2015	4.2	14
2016	4.2	14
2017	4.2	14

Methodology

Emissions from N_2O product uses were estimated using the following equation:

$$E_{pu} = \sum_{a} (P \times S_a \times ER_a)$$

where,

E_{pu} = N_2O emissions from product uses, metric tons
P = Total U.S. production of N_2O, metric tons
a = specific application
S_a = Share of N_2O usage by application *a*
ER_a = Emission rate for application *a*, percent

The share of total quantity of N_2O usage by end-use represents the share of national N_2O produced that is used by the specific subcategory (e.g., anesthesia, food processing). In 2017, the medical/dental industry used an estimated 86.5 percent of total N_2O produced, followed by food processing propellants at 6.5 percent. All other categories combined used the remainder of the N_2O produced. This subcategory breakdown has changed only slightly over the past decade. For instance, the small share of N_2O usage in the production of sodium azide has declined significantly during the 1990s. Due to the lack of information on the specific time period of the phase-out in this market subcategory, most of the N_2O usage for sodium azide production is assumed to have ceased after 1996, with the majority of its small share of the market assigned to the larger medical/dental consumption subcategory (Heydorn 1997). The N_2O was allocated across the following categories: medical applications, food processing propellant, and sodium azide production (pre-1996). A usage emissions rate was then applied for each sector to estimate the amount of N_2O emitted.

Only the medical/dental and food propellant subcategories were estimated to release emissions into the atmosphere, and therefore these subcategories were the only usage subcategories with emission rates. For the medical/dental subcategory, due to the poor solubility of N_2O in blood and other tissues, none of the N_2O is assumed to be metabolized during anesthesia and quickly leaves the body in exhaled breath. Therefore, an emission factor of 100 percent was used for this subcategory (IPCC 2006). For N_2O used as a propellant in pressurized and aerosol food products, none of the N_2O is reacted during the process and all of the N_2O is emitted to the atmosphere, resulting in an emission factor of 100 percent for this subcategory (IPCC 2006). For the remaining subcategories, all of the N_2O is consumed/reacted during the process, and therefore the emission rate was considered to be zero percent (Tupman 2003).

The 1990 through 1992 N_2O production data were obtained from SRI Consulting's *Nitrous Oxide, North America* report (Heydorn 1997). Nitrous oxide production data for 1993 through 1995 were not available. Production data for 1996 was specified as a range in two data sources (Heydorn 1997; Tupman 2003). In particular, for 1996, Heydorn (1997) estimates N_2O production to range between 13.6 and 18.1 thousand metric tons. Tupman (2003) provided a narrower range (15.9 to 18.1 thousand metric tons) for 1996 that falls within the production bounds described by Heydorn (1997). Tupman (2003) data are considered more industry-specific and current. Therefore, the midpoint of the narrower production range was used to estimate N_2O emissions for years 1993 through 2001 (Tupman 2003). The 2002 and 2003 N_2O production data were obtained from the Compressed Gas Association Nitrous Oxide Fact Sheet and Nitrous Oxide Abuse Hotline (CGA 2002, 2003). These data were also provided as a range. For example, in 2003, CGA (2003) estimates N_2O production to range between 13.6 and 15.9 thousand metric tons. Due to the unavailability of data, production estimates for years 2004 through 2017 were held constant at the 2003 value.

The 1996 share of the total quantity of N_2O used by each subcategory was obtained from SRI Consulting's *Nitrous Oxide, North America* report (Heydorn 1997). The 1990 through 1995 share of total quantity of N_2O used by each subcategory was kept the same as the 1996 number provided by SRI Consulting. The 1997 through 2001 share of total quantity of N_2O usage by sector was obtained from communication with a N_2O industry expert (Tupman 2003). The 2002 and 2003 share of total quantity of N_2O usage by sector was obtained from CGA (2002, 2003). Due to the unavailability of data, the share of total quantity of N_2O usage data for years 2004 through 2017 was assumed to equal the 2003 value. The emissions rate for the food processing propellant industry was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997), and confirmed by a N_2O industry expert (Tupman 2003). The emissions rate for all other subcategories was obtained from communication with a N_2O industry expert (Tupman 2003). The emissions rate for the medical/dental subcategory was obtained from the *2006 IPCC Guidelines*.

Uncertainty and Time-Series Consistency

The overall uncertainty associated with the 2017 N_2O emission estimate from N_2O product usage was calculated using the *2006 IPCC Guidelines* (2006) Approach 2 methodology. Uncertainty associated with the parameters used to estimate N_2O emissions include production data, total market share of each end use, and the emission factors applied to each end use, respectively.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-111. Nitrous oxide emissions from N_2O product usage were estimated to be between 3.2 and 5.2 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 24 percent below to 24 percent above the emission estimate of 4.2 MMT CO_2 Eq.

Table 4-111: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from N_2O Product Usage (MMT CO_2 Eq. and Percent)

Source	Gas	2017 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
N_2O from Product Uses	N_2O	4.2	3.2	5.2	-24%	+24%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological approaches were applied to the entire time series to ensure consistency in emissions from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.

Planned Improvements

EPA has recently initiated an evaluation of alternative production statistics for cross-verification and updating time-series activity data, emission factors, assumptions, etc., and a reassessment of N_2O product use subcategories that accurately represent trends. This evaluation includes conducting a literature review of publications and research that may provide additional details on the industry. This work is currently ongoing and thus the results have not been incorporated into the current Inventory report.

Pending additional resources and planned improvement prioritization, EPA may also evaluate production and use cycles, and the potential need to incorporate a time lag between production and ultimate product use and resulting release of N_2O. Additionally, planned improvements include considering imports and exports of N_2O for product uses.

Finally, for future Inventories, EPA will examine data from EPA's GHGRP to improve the emission estimates for the N_2O product use subcategory. Particular attention will be made to ensure aggregated information can be published without disclosing CBI and time-series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as required in this Inventory. EPA is still assessing the possibility of incorporating aggregated GHGRP CBI data to estimate emissions; therefore, this planned improvement is still in development and not incorporated in the current Inventory report.

4.27 Industrial Processes and Product Use Sources of Precursor Gases

In addition to the main greenhouse gases addressed above, many industrial processes can result in emissions of various ozone precursors. The reporting requirements of the UNFCCC[277] request that information be provided on precursor greenhouse gases, which include carbon monoxide (CO), nitrogen oxides (NO_x), non-CH_4 volatile organic compounds (NMVOCs), and sulfur dioxide (SO_2). These gases are not direct greenhouse gases, but indirectly affect terrestrial radiation absorption by influencing the formation and destruction of tropospheric and stratospheric ozone, or, in the case of SO_2, by affecting the absorptive characteristics of the atmosphere. Additionally, some of these gases may react with other chemical compounds in the atmosphere to form compounds that are greenhouse gases. As some of industrial applications also employ thermal incineration as a control technology, combustion byproducts, such as CO and NO_x, are also reported with this source category. NMVOCs, commonly referred to as "hydrocarbons," are the primary gases emitted from most processes employing organic or petroleum based products, and can also result from the product storage and handling.

Accidental releases of greenhouse gases associated with product use and handling can constitute major emissions in this category. In the United States, emissions from product use are primarily the result of solvent evaporation, whereby the lighter hydrocarbon molecules in the solvents escape into the atmosphere. The major categories of product uses include: degreasing, graphic arts, surface coating, other industrial uses of solvents (e.g., electronics), dry cleaning, and non-industrial uses (e.g., uses of paint thinner). Product usage in the United States also results in the emission of small amounts of hydrofluorocarbons (HFCs) and hydrofluoroethers (HFEs), which are included under Substitution of Ozone Depleting Substances in this chapter.

Total emissions of NO_x, CO, and NMVOCs from non-energy industrial processes and product use from 1990 to 2017 are reported in Table 4-112. Sulfur dioxide emissions are presented in Section 2.3 of the Trends chapter and Annex 6.3.

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[277] See <http://unfccc.int/resource/docs/2013/cop19/eng/10a03.pdf>.

Table 4-112: NO$_x$, CO, and NMVOC Emissions from Industrial Processes and Product Use (kt)

Gas/Source	1990	2005	2013	2014	2015	2016	2017
NO$_x$	**592**	**572**	**427**	**414**	**414**	**414**	**414**
Industrial Processes							
Other Industrial Processes[a]	343	437	307	300	300	300	300
Metals Processing	88	60	64	63	63	63	63
Chemical and Allied Product Manufacturing	152	55	44	43	43	43	43
Storage and Transport	3	15	10	5	5	5	5
Miscellaneous[b]	5	2	3	2	2	2	2
Product Uses							
Surface Coating	1	3	1	1	1	1	1
Graphic Arts	+	0	0	0	0	0	0
Degreasing	+	0	0	0	0	0	0
Dry Cleaning	+	0	0	0	0	0	0
Other Industrial Processes[a]	+	0	0	0	0	0	0
Non-Industrial Processes[c]	+	0	0	0	0	0	0
Other	NA	0	0	0	0	0	0
CO	**4,129**	**1,557**	**1,247**	**1,251**	**1,251**	**1,251**	**1,251**
Industrial Processes							
Metals Processing	2,395	752	600	553	553	553	553
Other Industrial Processes[a]	487	484	455	530	530	530	530
Chemical and Allied Product Manufacturing	1,073	189	129	117	117	117	117
Miscellaneous[b]	101	32	48	42	42	42	42
Storage and Transport	69	97	13	7	7	7	7
Product Uses							
Surface Coating	+	2	2	1	1	1	1
Other Industrial Processes[a]	4	0	0	0	0	0	0
Dry Cleaning	+	0	0	0	0	0	0
Degreasing	+	0	0	0	0	0	0
Graphic Arts	+	0	0	0	0	0	0
Non-Industrial Processes[c]	+	0	0	0	0	0	0
Other	NA	0	0	0	0	0	0
NMVOCs	**7,638**	**5,849**	**3,855**	**3,816**	**3,816**	**3,816**	**3,816**
Industrial Processes							
Storage and Transport	1,352	1,308	724	613	613	613	613
Other Industrial Processes[a]	364	414	309	314	314	314	314
Chemical and Allied Product Manufacturing	575	213	72	70	70	70	70
Metals Processing	111	45	28	26	26	26	26
Miscellaneous[b]	20	17	27	24	24	24	24
Product Uses							
Surface Coating	2,289	1,578	1,104	1,134	1,134	1,134	1,134
Non-Industrial Processes[c]	1,724	1,446	1,012	1,039	1,039	1,039	1,039
Degreasing	675	280	196	202	202	202	202
Dry Cleaning	195	230	161	165	165	165	165
Graphic Arts	249	194	136	139	139	139	139
Other Industrial Processes[a]	85	88	61	63	63	63	63
Other	+	36	25	26	26	26	26

+ Does not exceed 0.5 kt

NA (Not Available)

[a] Includes rubber and plastics manufacturing, and other miscellaneous applications.

[b] Miscellaneous includes the following categories: catastrophic/accidental release, other combustion, health services, cooling towers, and fugitive dust. It does not include agricultural fires or slash/prescribed burning, which are accounted for under the Field Burning of Agricultural Residues source.

[c] Includes cutback asphalt, pesticide application adhesives, consumer solvents, and other miscellaneous applications.

Note: Totals may not sum due to independent rounding.

Methodology

Emission estimates for 1990 through 2017 were obtained from data published on the National Emission Inventory (NEI) Air Pollutant Emission Trends web site (EPA 2018), and disaggregated based on EPA (2003). Data were collected for emissions of CO, NO_x, volatile organic compounds (VOCs), and SO_2 from metals processing, chemical manufacturing, other industrial processes, transport and storage, and miscellaneous sources. Emissions were calculated either for individual source categories or for many categories combined, using basic activity data (e.g., the amount of raw material processed or the amount of solvent purchased) as an indicator of emissions. National activity data were collected for individual categories from various agencies. Depending on the category, these basic activity data may include data on production, fuel deliveries, raw material processed, etc.

Emissions for product use were calculated by aggregating product use data based on information relating to product uses from different applications such as degreasing, graphic arts, etc. Emission factors for each consumption category were then applied to the data to estimate emissions. For example, emissions from surface coatings were mostly due to solvent evaporation as the coatings solidify. By applying the appropriate product-specific emission factors to the amount of products used for surface coatings, an estimate of NMVOC emissions was obtained. Emissions of CO and NO_x under product use result primarily from thermal and catalytic incineration of solvent-laden gas streams from painting booths, printing operations, and oven exhaust.

Activity data were used in conjunction with emission factors, which together relate the quantity of emissions to the activity. Emission factors are generally available from the EPA's *Compilation of Air Pollutant Emission Factors*, AP-42 (EPA 1997). The EPA currently derives the overall emission control efficiency of a source category from a variety of information sources, including published reports, the 1985 National Acid Precipitation and Assessment Program emissions inventory, and other EPA databases.

Uncertainty and Time-Series Consistency

Uncertainties in these estimates are partly due to the accuracy of the emission factors and activity data used. A quantitative uncertainty analysis was not performed.

Methodological approaches were applied to the entire time series to ensure time-series consistency from 1990 through 2017. Details on the emission trends through time are described in more detail in the Methodology section, above.

For more information on the general QA/QC process applied to this source category, consistent with Volume 1, Chapter 6 of the *2006 IPCC Guidelines*, see the QA/QC and Verification Procedures section in the introduction of the IPPU chapter.