Anthropogenic emissions at the global and regional scale during the past three decades

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Abstract

Accurate, timely, and accessible emissions information is critical for understanding and forecasting atmospheric composition at the global and regional scale. Over the past few years, several inventories providing the distributions of surface emissions of different chemical compounds were developed at both global and regional scales. We will review the most recent developments in surface emissions inventories for anthropogenic and biomass burning emissions, and provide details on the newest publicly available datasets.

The quality of emissions inventories is very difficult to assess, since the methodology, input data and assumptions vary strongly between the inventories. We will discuss an evaluation of emissions distributions for the 1970-2010 period, and focus on the emissions of the following compounds: methane (CH_4), carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2), black and organic carbon, volatile organic compounds (VOCs), and ammonia (NH_3). The consistency between global and regional datasets in different regions of the world will be discussed.

Anthropogenic emissions of VOCs are generally reported as total VOCs emissions; however, global and regional models require a good knowledge of individually speciated VOCs. First results of an evaluation of VOCs speciation used in current inventories will be discussed, where ratios of selected VOCs to CO concentrations from different observations are compared with ratios provided by emissions inventories.

Introduction

The spatial and temporal distributions of atmospheric compounds are in large part driven by the distribution of their emissions. An accurate knowledge of the surface emissions and of their evolution with time is therefore essential to support analyses and modeling of air quality. Up-to-date and consistent emissions are moreover required for the forecasting of atmospheric composition at different scales (Frost et al., 2012).

Gridded global, regional, and national emission estimates have been developed that take into account the main atmospheric compounds that are important for understanding and analyzing the distribution of ozone and its precursors as well as aerosols and their precursors. These inventories provide emissions for NO_x, CO, CH_4 and VOCs, as well as for SO_2, black and organic carbon, NH_3 and particulate matter (PM10 and PM2.5). Some of these inventories are publicly available, whereas others are developed by individual research groups or government agencies to study specific aspects of emissions or atmospheric processes and are not always easy to access. In this paper, we will focus only on publicly available datasets.

In the paper, we will provide the main characteristics of the most recent global and regional inventories; both anthropogenic and biomass burning emission datasets will be considered. We will then compare the emissions provided by the different datasets since the 1960s, and use these comparisons to estimate the uncertainties on the emissions for different regions of the world. We will also discuss the speciation of VOCs in the different inventories that have been recently developed.

The conclusions of this work will represent the basis for the development of a new community historical emission dataset, which will cover the 1750-2015 period. The comparisons of currently

available datasets will allow better quantification of the differences between datasets, and will provide more information on the uncertainties in anthropogenic and biomass burning emissions in different regions.

Description of a few recent global and regional emission inventories

The MACCity global anthropogenic and biomass burning emissions for 1960 – 2015

A community effort in 2008-2009 led to the development of the ACCMIP emissions, a dataset of decadal, monthly, sectoral, gridded anthropogenic and biomass burning emissions covering the historical period of 1850 – 2000 (Lamarque et al, 2010). The primary purpose of this work was to provide consistent gridded emissions of reactive gases and aerosols for use in chemistry model simulations in support of the Fifth Assessment Report of the IPCC (Intergovernmental Panel on Climate Change).

The year 2000 was chosen as the reference year for the ACCMIP emissions, since 2000 emissions represented a combination of the best and latest information available on existing regional and global inventories in the years 2008-2009 when the inventory was built.

An extension of the historical emissions dataset was then developed, called MACCity, which provides monthly averaged sectoral emissions for each year during the 1960-2010 period. This dataset was based on the decadal ACCMIP emissions for 1960-2000 and the 2005 and 2010 emissions provided by one of the future emissions scenarios, or representative concentration pathways (RCPs), i.e. RCP 8.5. This scenario was chosen since it included some information on recent emissions at the regional scale in Europe and North America. A seasonal cycle was first applied sector by sector and emissions were interpolated on a yearly basis between the base years. Details of the methodology are given in Granier et al. (2011).

Emissions of individual VOCs are obtained through a speciation of total VOCs. The data used for the speciation were originally developed for the RETRO inventory (Schultz et al., 2007) which was based on a speciation developed by the TNO group (Utrecht, The Netherlands).

The monthly mean emissions of trace species resulting from biomass burning provided by the MACCity inventory are derived from spatio-temporally modified RETRO carbon emission data (Schultz et al., 2008) for the years 1960 to 1996 and from GFED-v2 carbon emission data for the years 1997 to 2008 (van der Werf et al., 2006), by applying a single set of vegetation-type specific emission factors and the predominant vegetation map used in the GFED-v2 fire inventory. The fire emissions for a large set of atmospheric compounds are provided in MACCity, using emission factors from the compilation of Andreae and Merlet (2001).

The EDGARv4 and HTAPv2 global anthropogenic emission inventories

The EDGAR (Emission Database for Global Atmospheric Research) inventory, versions 4.2 and 4.3 (European Commission, 2011) are products developed by the Joint Research Center (Ispra, Italy) and the PBL Netherlands Assessment Agency. Emissions for greenhouse gases and air pollutants are provided.

In EDGARv4.2, emissions are calculated as totals by country for different sectors for the 1970-2008 period, and distributed on a 0.1x0.1 degree grid over the globe. Emissions in EDGARv4.2

are calculated for each compound as a product of country-specific activity data, the mix of technologies, the abatement percentage, and country-specific emission factors. Emissions by country are allocated on a spatial grid using proxy data, which makes them directly usable in model simulations. Point source are also considered, using maps with the locations of individual plants and industrial activity facilities.

The EDGARv4.3 global anthropogenic emission inventory (Crippa et al., in preparation) of several gaseous and particulate air pollutants has been used to develop retrospective emission scenarios for the years 1970-2010 to quantify the effectiveness of emission reduction measures, the effects of changes in fuel consumption and technological developments on air quality emissions, and their impacts on health, crops, and climate. Based on statistics and expert knowledge, EDGARv4.3 considers changes in activity data, fuel and air pollution abatement technology as they likely happened during the past 4 decades. Additionally, two retrospective scenarios are created, one simulating the complete stagnation of technology (STAG_TECH: lack of abatement measures and no improvement in emission standards), and one assuming constant fuel mixture and consumption as they were in 1970 (STAG_FUEL: no change in human activities).

The HTAPv2 dataset consists of 0.1x0.1 degree gridmaps of several atmospheric gaseous and particulate compounds for the years 2008 and 2010. HTAPv2 uses nationally reported catalogs combined with regional scientific inventories in the format of sector-specific gridmaps. The gridmaps are complemented with EDGARv4.3 data for those regions where regulatory or scientific data are absent. The global gridmaps are a joint effort of the US EPA, the MICS-Asia group, EMEP/TNO, the REAS group and the EDGAR group to serve primarily the scientific community studying hemispheric transport of air pollution. A speciation of total VOCs has been applied to the total VOCs emissions in HTAPv2, in order to provide emissions for a large set of individual VOCs. These data are available from the ECCAD website, described in the final section.

The TNO-MACC regional emission inventory for Europe

In Europe, surface emissions are reported by the different countries to EMEP (European Monitoring and Evaluation Program). The quality of these reported emissions varies across the different European countries. As an alternative to the official emissions inventory, a spatially high-resolution emission inventory (7x7 km) for Europe has been developed by Kuenen et al. (2014) for the 2003-2009 period. A new version of this inventory was released in March 2015 that provides emissions for the 2000-2011 period. The dataset includes emissions for the main pollutants, together with a speciation for coarse and particulate matter into elemental carbon, organic carbon, sulfates, sodium and other minerals.

The TNO-MACC emissions are spatially distributed consistently across the European countries by using proxy parameters, Point source emissions are spatially distributed using the specific location of each source and are year-specific.

The REASv2 regional emission inventory for Asia

Version 2 of REAS (Regional Emission inventory in Asia) provides emissions of most air pollutants and greenhouse gases for each year during the 2000-2008 period for different regions

in Asia. Monthly emissions are available on a 0.25x0.25 degree grid for the full period for different sectors. Detailed information on the methodology, activity data and emission factors for the estimation of emissions from stationary combustion, industrial processes, road transport, agricultural activities and solvents that were used to develop REASv2 are given in Kurokawa et al. (2013).

Other recent datasets considered in the evaluation

Several other emissions datasets are also considered in this evaluation that will not be described in detail in this paper, i.e.:

- The ECLIPSE global emission dataset provides emissions for the years 2005, 2010, 2030 and 2050. Emissions are calculated within the GAINS model framework (Amann et al., 2011). Version 4a and 5 of ECLIPSE will be used in the analysis
- The GFAS (Global Fire Assimilation System) calculates global biomass burning emissions by assimilating Fire Radiative Power (FRP) observations from the MODIS instruments onboard the Terra and Aqua satellites (Kaiser et al., 2012). Daily emissions are available, calculated on a global $0.5^{\circ} \times 0.5^{\circ}$ grid from 2003 to the present.
- The Fire INventory from NCAR (FINN) model provides high resolution, global emission estimates from open burning. The inventory framework produces daily emission estimates at a horizontal resolution of about 1 km^2. Emissions from FINN are available from 2002 to 2014.
- The regional MEIC (Multi-resolution Emission Inventory for China) provides emissions of several atmospheric compounds at a 0.5x0.5 degree resolution for China. More information can be found at: http://www.meicmodel.org/.

Evaluation of global and regional inventories

Most global anthropogenic emissions estimates are developed using the same generic method based on the product of estimates for activity data and emission factors. Activity data usually originate either with country records or with international organizations such as the United Nations or the International Energy Agency. Emissions from biomass burning are calculated on the basis of observations of burned areas, active fires or fire radiative power. However, since these data are not always consistent, many inventory developers adjust the data in a different way for use in their emission algorithms. Emission factors for most gaseous and all particulate species depend on the generating process. This dependence is represented either by specifying the type of technology or fire in each world region or by choosing emission factors that are thought to be representative of each region, especially with attention to development characteristics. All this information is generally not provided in detail in currently available inventories. Therefore, significant differences can be found between emission datasets, which cannot always be explained.

Comparisons of emissions inventories in different regions of the world allow the range of estimates provided by the different datasets to be quantified, which can give insights into the uncertainties of the inventories. Several such comparisons are shown in our oral presentation. The

focus is on anthropogenic emissions, but comparisons of different datasets providing emissions from fires will also be discussed. Figure 1 shows, for example, the MACCity global total emissions for carbon monoxide from 1960 to 2010, and the respective contribution of anthropogenic sources, fires and natural emissions. This figure shows the large contribution of both anthropogenic and fire emissions to the total CO emissions globally.

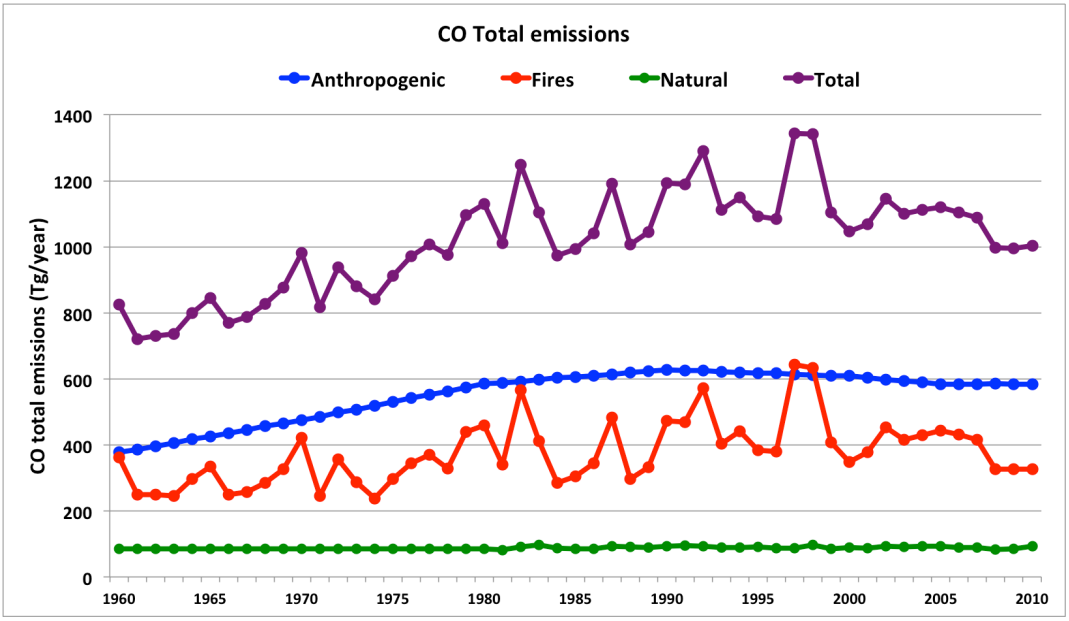

Figure 1: Anthropogenic, fire, natural and total CO emissions from 1960 to 2010, from the MACCity inventory.

Evaluation of VOCs speciation in anthropogenic emission inventories

The emissions of VOC are generally not very well detailed in emission inventories and their speciation has a high level of uncertainty. Usually anthropogenic emissions of volatile organic compounds are presented in emissions inventories as "total VOC", referring to a lumped quantity of different VOCs such as alkanes, alkenes, alkynes, alcohols, aldehydes, ketones and aromatic compounds.

We have evaluated the speciation used in the MACCity inventory through a comparison of the ratios between selected VOCs and CO, as obtained from different observations and for the model speciation. There are only a few long-term measurements of individual VOCs available, mostly in Los Angeles, London and Paris. We also included in our analysis VOCs observations obtained in canisters during field campaigns in Europe, Latin America, China and Pakistan.

The observed ethane/CO ratio shows rather good agreement with MACCity in China and southeast Asia (Hong Kong, Beijing, and Singapore), but the ratio is generally higher in the observations than in the inventory in other megacities globally. For the propene/CO ratio, good agreement is found in London and Karachi, but the inventory/observations ratio is generally higher for all other cities.

Availability of the emissions data: the ECCAD database

The ECCAD (Emissions of Atmospheric Compounds & Compilation of Ancillary Data) database was developed to provide to scientific and policy users a large set of data on the surface emissions of gaseous and particulate chemical species, as well as with ancillary data required to quantify and to analyze surface emissions. Currently, ECCAD provides user-friendly access to global and regional emission inventories, ancillary data, and tools for the analysis and evaluation of these data.

ECCAD aims to provide access to a large set of data, and to give detailed information on each dataset, in order to increase the visibility of each of the datasets and of the groups who developed them. Many of the datasets available within ECCAD have been developed, formatted or adapted through funding of different projects. Emissions available in ECCAD can be downloaded in ASCII and NetCDF-CF format. Several tools are available in ECCAD for the visualization and the analysis of emissions and ancillary data, such as the calculation of totals and averages for a certain period or for a specific region or country. Time series for a specific latitude/longitude can also be obtained. ECCAD is currently limited to gridded datasets at 0.5x0.5 or 1x1 degree resolution. A new version of ECCAD is under development (ECCAD-2), which will accommodate data at any spatial resolution, together with non-gridded emissions.

Acknowledgments

The authors greatly acknowledge the support of the European Union under the MACC-II, and PANDA projects of the 7th Framework Programme, and the MACC-III project of the Horizon-2020 Framework Programme for Research and Innovation.

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