January 13, 2010

The “Peer Review of Source Apportionment Tools in CAMx and CMAQ” contains the synthesis of a 5 panel review committee’s comments on the source apportionment implementations in CAMx and CMAQ. In addition, clarifications from the source apportionment developers are also included. The main document contains notations that refer to either the CAMx (cited as Response ENVIRON Page #) or CMAQ developer (cited as Response ICF Page #) response letter when a clarification or further discussion on a topic is available.

The panel of reviewers consisted of 5 members:

Dr. Harvey Jefferies, University of North Carolina
Dr. Talat Odman, Georgia Institute of Technology
Dr. Che-Jen (Jerry) Lin, Lamar University
Dr. Zion Wang, University of California at Riverside
Dr. Daewon Byun, University of Houston

Comments from the developer for the CAMx implementation were submitted by Greg Yarwood of ENVIRON and by Sharon Douglas of ICF International for the CMAQ implementation.

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Peer Review of Source Apportionment Tools in CAMx and CMAQ

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1. Introduction

Grid-based air quality models (AQMs) are powerful tools used in air quality management. Models are usually used to predict ambient concentrations of pollutants, such as ozone and fine particulate matter ($PM_{2.5}$), at multiple spatial and temporal scales. It is useful to understand specific sources or regions that contribute to the concentration levels estimated by AQMs. Understanding the contributions to specific geographic receptor locations from particular emission sources, specific model processes, or individual chemical pathways helps regulators develop effective emissions control strategies. Probing tools that help with this kind of assessment are broadly categorized into two types: mass balance analysis (MBA) techniques and sensitivity analysis (SA) techniques (Zhang et al., 2005). MBA provides quantitative information on the contribution of the various processes (e.g., transport and chemical reactions) to the modeled ambient concentrations, whereas SA provides quantitative information on the response of these concentrations to changes in the air pollution system. MBA techniques are appropriate for diagnostic evaluations of AQMs (i.e., to identify which chemical transformation pathways and which physical transport processes govern pollutant concentrations). MBA techniques are also useful for identifying which sources contribute to pollutant concentrations, while SA techniques provide information regarding how changes in the emissions affect the simulation results. This report focuses on source apportionment techniques for ozone, PM2.5, and mercury techniques that have been implemented in photochemical grid models. Source apportionment is an approach that is used to efficiently assess complete source contribution. The approach is evaluated for scientific credibility and briefly compared to other instrumented photochemical modeling approaches that also have the potential to estimate total source culpability: zero-out (brute force) and Decoupled Direct Method.

Source apportionment is an alternative approach to traditional brute-force (also called "zero-out") modeling, and it has the advantage of being much more efficient with computational resources. For instance, to estimate the contribution from 20 source regions, a total of 20 individual zero-out simulations would be needed, compared with only one source apportionment simulation. The incremental run time associated with the additional source region tracking performed in one source apportionment simulation is far less than the time required to run numerous iterative brute-force simulations. There are multiple source apportionment methodologies that can be developed. For a particular application, developers can choose a method by deciding the most appropriate balance of computation speed, thoroughness in model formulation, and ease of use.

Ozone source apportionment tracks the contribution to model ozone estimates from precursor emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs). Particulate source apportionment tracks contributions to particulate species from precursor emissions. Emissions of NO_x are tracked through all intermediate nitrogen species to particulate nitrate ion. Sulfur dioxide and sulfuric acid emissions are tracked to particulate sulfate ion. Ammonia emissions are tracked to particulate ammonium ion. Even though ammonium nitrate is chemically coupled, the apportionment schemes typically do not attempt to determine which species is limiting the formation, and directly attributes precursor gases to specific particulate ions. Ozone, $PM_{2.5}$, and mercury source apportionment are implemented in state-of-the-science photochemical grid models: the Community Multiscale Air Quality (CMAQ) model and the Comprehensive Air Quality Model with extensions (CAMx). These models are summarized in Sections 1.1 and 1.2.

Example uses of photochemical model source apportionment include:

- Quantifying the contribution of precursor emissions from source sectors (e.g., points, on-road mobile, non-road, fires) or specific types of facilities (e.g., power plants, pulp and paper mills, chemical manufacturing facilities) to estimated concentrations at selected locations throughout the modeling domain.
- Quantifying the contribution of emissions from selected geographical regions (e.g., one or more states) to regional ozone, mercury, or $PM_{2.5}$ concentrations.
- Quantifying the contribution from estimated boundary conditions to ozone, mercury, or $PM_{2.5}$ concentrations at selected locations.
- Tracking the fate of emissions from specific facilities to estimate their range of influence and contribution to ozone or $PM_{2.5}$ concentrations in nonattainment areas throughout the modeling domain.
- Quantifying the contribution of emissions from selected sources or source categories to visibility impairment in Class I areas (National Parks and Monuments).

In this report, we summarize the comments provided by external peer reviewers of the two source apportionment techniques that are instrumented in CMAQ and CAMx. These comments were provided during a peer review process sponsored by the Air Quality Modeling Group of the U.S. Environmental Protection Agency's (EPA) Office of Air Quality Planning and Standards. More information on the peer review process is given in Appendices A and B, while Appendix C provides the résumés of the peer reviewers.

To summarize the four approaches that are the focus of this review, the features of CMAQ/PPTM, CMAQ/OPTM, CAMx/PSAT and CAMx/OSAT are quite similar, although CAMx/PSAT and CAMx/OSAT seem to be more scientifically sound for the following reasons:

(a) careful consideration of mass conservation issues,

(b) explicit treatment of dry deposition of the tagged (reactive tracers) species, and

(c) separation of ozone production scenarios for NO_x-limited and VOC-limited conditions.

1.1 Source apportionment (ozone, PM, Hg) implementation in CMAQ

The CMAQ model (Byun and Schere, 1999, 2006) numerically simulates the physical processes that determine the magnitude, temporal variation, and spatial distribution of the concentrations of ozone and particulate species in the atmosphere and the amount, timing, and distribution of their deposition to the earth's surface. The simulation processes include advection, dispersion (or turbulent mixing), chemical transformation, cloud processes, and wet and dry deposition.

ICF International has implemented the Ozone Precursor Tagging Methodology (OPTM) and the Particulate Precursor Tagging Methodology (PPTM) in CMAQ version 4.6. Within the model, tagging is accomplished by the addition of duplicate model species variables for each source, source category, or source region that is to be tagged. For OPTM, the duplicated modeled species are ozone, NO_x, and VOCs. For PPTM, the duplicated species include all PM-related sulfur, nitrogen, and secondary organic compounds, as well as primary organic carbon, elemental carbon, and other inorganic particulates. PPTM also tracks the contribution from all precursor gas and particulate phase mercury species to model estimates. The tagged species have the same properties and are subjected to the same processes (e.g., advection, chemical transformation, deposition) as the actual (or base) species.

Because the tagged species are separate from the base species, tagging does not alter or affect the base simulation results. At each time step in the simulation, the effects of linear or species-independent processes, such as dry deposition and advection, are calculated directly for all tagged species. Potentially nonlinear processes, such as gas-phase chemistry, aqueous chemistry, and particle dynamics, are calculated for the overall (or base) species and apportioned to the tagged species. The results for the tagged species are not normalized to ensure that the sum of the tagged species equals the total that would be estimated by a regular model simulation. In fact, standard bulk species are not standard output from OPTM and PPTM; total bulk species such as ozone or $PM_{2.5}$ sulfate ion are the sum of all tagged species. Thus, the difference between the sum of all tags and the overall concentration gives an estimate of the numerical uncertainty in the contribution estimates. The tagged species are included as additional species in the model output files.

CMAQ OPTM requires an additional emissions processing step using a utility program called "ems ox". This program sums NO and NO_2 emissions for each grid cell and each hour for each tagged group to form an additional emissions species called NO_X. The program also estimates a new emissions species called VOC, which is the sum of the hydrocarbon species weighted by the number of carbons in the compound (ICF International, 2007). Technical details of the tagging methodologies are provided in Douglas et al. (2007).

1.2 Source apportionment (ozone, PM, Hg) implementation in CAMx

CAMx, like CMAQ, is an Eulerian photochemical dispersion model that allows for an integrated “one-atmosphere” treatment of gaseous and particulate air pollution, over many spatial scales from urban to continental. CAMx uses state of the science routines to model ozone and particulate matter formation and removal processes (ENVIRON, 2008).

CAMx contains a variety of ozone source apportionment tools, including the original ozone source apportionment tool (OSAT) and the anthropogenic pre-cursor culpability assessment (APCA) tool (ENVIRON, 2008). Ozone source apportionment in CAMx tracks the contributions to each grid cell from emissions source groups, emissions source regions, initial conditions, and boundary conditions with reactive tracer species. Source apportionment tracers are treated using the standard model algorithms for vertical advection, vertical diffusion, and horizontal diffusion. Horizontal advection fluxes for each of the regular model species that make up NO_X and VOC are combined and normalized by a concentration based weighted mean. The estimated normalized fluxes are used to advect the tracer species rather than solving them with the standard model formulation to improve consistency between tracer and regular model concentrations (ENVIRON, 2008). The deposition velocities for NO_X tracers are the concentration weighted average of the deposition velocities for NO and NO_2. The deposition velocities for VOC tracers are the concentration weighted average (done on a ppmC basis) of the deposition velocities for each of the CB05 species. Ozone tracers use the same deposition velocities as ozone (ENVIRON, 2008).

The APCA tool assesses regional and emission sector contribution to ozone formation and provides information that is most policy relevant. APCA is designed to provide more control strategy relevant information and recognizes that there are source categories such as biogenic emissions that can not be controlled so the model only attributes ozone to biogenic sources when it is due to the interaction of biogenic VOC + biogenic NO_x. In the case where ozone formed due to biogenic VOC + anthropogenic NO_x under VOC-limited conditions, OSAT attributes it to biogenic VOC, but APCA redirects the attribution to anthropogenic NO_x. In NO_x -limited conditions both OSAT and APCA attribute the ozone to anthropogenic NO_x (ENVIRON, 2008).

Similar to OSAT, the Particle Source Apportionment Technology (PSAT) has been instrumented in CAMx to provide PM source apportionment among specific geographic regions and source categories. Like the OSAT methodology, the PSAT technique uses reactive tracers, and can apportion both primary PM (elemental carbon, primary organic aerosol, crustal fine, other fine, crustal coarse, other coarse) and secondary PM (ammonium, sulfate, nitrate, secondary organic aerosol, and particulate mercury). In general, while a single tracer can track a primary PM species, tracking a secondary PM species requires several tracers to track the relationship between the gaseous precursors and the resultant secondary PM. If one wishes to apply CAMx with PSAT for all PM species, 32 tracers are needed for each source group.

Besides ozone reaction tracers, CAMx also uses a separate family of “reaction timing tracers” to allow source-receptor transport times to be estimated. It does this by releasing unique timing tracers from each geographic area selected for source apportionment. This feature provides a way to investigate temporal features of the source-receptor relationships, and perhaps to develop temporally targeted emissions control strategies.

Technical details of the formulation and implementation of the OSAT and PSAT algorithms are provided in the CAMx user’s guide (ENVIRON, 2008).

1.3 Outline of this report

The rest of this report focuses on the peer review comments provided by the external reviewers of the two source apportionment techniques, and contains the following sections:

- **Section 2:** Summary of the strengths and weaknesses of CAMx/OSAT and CMAQ/OPTM, and then of CAMx/PSAT and CMAQ/PPTM
- **Section 3:** Comparison of source apportionment to other methods such as DDM, zero-out, and the CMAQ Tagged Species Source Apportionment (CMAQ-TSSA)
- **Section 4:** Strengths and weaknesses in the documentation for these techniques
- **Section 5:** Strengths and weaknesses in preprocessing and postprocessing tools for these techniques
- **Section 6:** Reviewer comments on possible future work needed for these techniques
- **Appendix A** describes the peer review process that was adopted in the development of this report
- **Appendix B** contains the charge questions provided to the reviewers by EPA
- **Appendix C** contains the CVs of the peer reviewers

2. Source Apportionment as Related to Common/Intended Applications

2.1 Ozone: OSAT and OPTM implementation strengths and weaknesses

2.1.1 CAMx/OSAT

All reviewers agreed that the Ozone Source Apportionment Tool (OSAT) in CAMx is a very mature and well-evaluated methodology. It is well documented, fully integrated into the operation of CAMx, and relatively easy to use. The methodology is designed so that all ozone and precursor concentrations are attributed among the selected source groupings at all times. Thus, for all receptor locations and times, the ozone (or ozone precursor concentrations) predicted by CAMx is attributed amongst the source groupings selected for OSAT. An important feature of the ozone reaction tracer approach used in OSAT is that the normal CAMx calculations are not perturbed; thus, OSAT estimates the same ozone and precursor concentrations as CAMx.

The CAMx source apportionment methodology also permits "timing tracers" to be emitted from emission source regions. While several reviewers agreed that this is an interesting feature that does not exist in other methods, the degree of its reliability is questionable. Two tracers are emitted simultaneously for each tagged source: one that does not decay and one that does undergo decay at a fixed rate, thus allowing the two to be used to calculate the time of origin of the VOCs or NO_x from that region. The process for instrumenting CAMx with these timing tracers is fully described in the manual, and the setup is fairly simple.

One reviewer observed that once a tracer was emitted into the model, it never disappeared. Because at least two tracers are emitted each day for each source region specified, the file size keeps growing and quickly reaches an unusable large size. Further, the "restart" will remember timing tracers, but there is no way to run OSAT for just one day in the middle of a longer (e.g., 12-day) simulation, without having run from the first day, even if the tracers from the previous days have decayed to meaningless concentrations. Thus, the reviewer felt that an extremely useful feature needs additional implementation improvements.

Below are additional strengths and weaknesses of CAMx/OSAT that were identified by the reviewers.

OSAT Strengths

(a) The explicit tracking of O_3 production and O_3 destruction separately, using integrated reaction rates, instead of as a single change in O_3.

(b) The use of Sillman's indicator species to distinguish VOC- and NO_x-limited regimes.

(c) OSAT accounts for different VOC reactivities.

(d) Mass is conserved by normalizing total advective fluxes by the "weighted mean" concentrations of the tracers. This is generally seen as a plus because advection is a linear process that becomes nonlinear in the model due to numerical issues. One reviewer believed that the

OSAT approach would restore the linearity by assuring that the sum of the components is equal to the bulk.

(e) Use of multiple tracer species to account for ozone precursors from a given source region at a given receptor location. OSAT can also estimate the cumulative contribution to ozone production of these precursors while they were en route to the receptor.

(f) Tracers can provide information on the relative importance of transport and chemistry. OSAT is also capable of ranking the relative importance of sources as well as ranking the contributors to ozone.

(g) The methodology and model results have been peer-reviewed.

(h) Ability to directly relate ozone concentration to the emission sources and background precursor concentrations (ICs/BCs), allowing policy makers to evaluate the model results easily.

(i) The incorporation of the inert and decaying timing tracers to track the precursor transport pathways.

(j) Masses of the reactive tracer species (NO_x, VOCs, and O_3) are kept consistent with the standard CAMx species.

(k) Capable of tracking the range of precursor transport for both timing and ozone reaction tracers.

(l) Deposition of the tracer species and ozone destruction reactions are treated more explicitly compared to CMAQ/OPTM.

(m) Relatively computationally inexpensive compared to the traditional sensitivity techniques such as brute force (zero-out) methods.

(n) CAMx/APCA is a nice added feature complementing CAMx/OSAT to provide “adjusted” apportionment results related to controllable (i.e., anthropogenic) emission sources.

(o) Option to use a gridded “mask” to define source regions for tracking, which means the input files need no modification

<u>*OSAT Weaknesses*</u>

(a) OSAT uses only the H_2O_2/HNO_3 production ratio to determine VOC-limited or NO_x-limited regime. Other indicators ($HCHO/NO_y$, O_3/NO_z) are not used to assist in the determination.

(b) Uses a single value (1/3) instead of a threshold range (e.g., 0.3-0.5) to determine whether a region is NO_x or VOC limited. This may lead to errors in attribution in transitional ozone formation regimes.

(c) Requires additional effort (compared to emissions processing for standard model input) in processing the emissions inventory of reactive tracer species when the emissions “mask” feature is not used.

(d) May not represent the nonlinear response of atmospheric processes to emissions changes. Therefore, it can estimate the contribution of the tagged species only under the base-case emission scenarios; it cannot estimate the effects of emission changes on ozone concentrations.

(e) As shipped, CAMx outputs the tracer information only for layer 1. If the focus is to characterize sources of NO_x or non-surface O_3 this is problematic.

(f) Although species naming is straightforward, it needs some metadata description in the file (thus, users should be encouraged to provide metadata information for each tag).

2.1.2 CMAQ/OPTM

The Ozone Precursor Tagging Methodology (OPTM) elicited a mixed response from the reviewers. One reviewer felt strongly that the OPTM cannot be fully evaluated in its current state because the documentation is insufficient to configure and run a simulation. The documentation clearly indicates that OPTM requires complex configuration even for a simple case. Further, the documentation’s technical formulation section raises many questions that were not answerable with the code supplied. Details on this reviewer’s comments are given in the paragraphs that follow.

The “User Guide” (Douglas et al, 2007) and “Memo,” (Myers and Douglas, 2006) hereafter “documentation,” are inconsistent with the code provided. The documentation references utilities for tagged input creation (i.e., emis_optm, bcon_optm, icon_optm) that are either not supplied or are obscure. One reviewer felt there were inconsistencies between the documentation and the OPTM version of the CMAQ code given by the peer review panel team. The reviewer felt that without the entirety of tools and code, the documentation could not be evaluated as a guide for operation.

The simplest source apportionment case—a spatial source apportionment—requires too many steps for preparation and configuration. Based on the instructions, any implementation requires editing INCLUDE files and adding tracer species to the model emissions, boundary conditions, and initial conditions input files. Other methods, specifically CAMx/OSAT, allow for the creation of a spatial mask to use existing emission inventories. The mask-style implementation would be particularly useful when only the CMAQ-ready inputs are available.

The technical formulation raises several questions that need additional clarification:

(a) Physical process magnitudes for tracers are inconsistent with corresponding model species. Other methodologies have solved this problem by utilizing process analysis data for component species and aggregating process magnitudes for the tagged tracer species. These data already exist in the models. [Response ICF Page 1]

(b) Calculation of total oxidant (O_x) is incorrect. When two O_x species combine, as with NO_3 (NO_2+O) and N_2O_5 (NO_2+NO_3), the species should represent multiple units of O_x based on the number of “odd oxygen,” not the nitrogen. [Response ICF Page 1]

(c) Based on the documentation, ratios of initial precursors are used to apportion net chemical change of tagged VOCs, NO_x, and O_x. There are several false assumptions in the code that, if confirmed, should be addressed: [Response ICF Page 1]

    (1) Change in tagged species is solved algebraically, which inappropriately conserves initial tagged species. A differential solution would apply losses to the existing distribution of tags and to the distribution of chemically produced tags.

(2) Change in each region's NO_x and VOCs is proportional to the total local NO_x and VOCs. This assumes that each region's NO_x and VOC reactivity (or deposition velocity) profile is equal to that of the local NO_x and VOCs. In plumes or near sources, the locally contributed NO_x and VOCs are likely to have a different reactivity profile. [Response ICF Page 2]

(3) NO_x production is proportional to existing NO_x. It assumes that NO_x precursors (i.e., HNO_3, PAN, or NTR) are proportional to local NO_x concentrations. These longer-lived species may be transported and so not reflect local emissions of NO_x. [Response ICF Page 2]

(4) Total oxidant chemistry is apportioned based on precursor concentration distributions. While O_x production is likely to be proportional to precursors (excluding situations noted in [1] and [3] above), O_x loss reactions should be apportioned based on current O_x distribution. [Response ICF Page 2]

(d) Artificial "offset" to O_x potentially alters processes and requires further review of code. [Response ICF Page 3]

The documentation raised questions that could not be answered. Simple case implementations should be simplified with internal emission apportionment routines. The reviewer suggested that the comments above be addressed first, and that OPTM then be re-reviewed with updated documentation and code.

OPTM does not track ozone; instead, it tracks "the amount of oxidant defined as $NO_2 + NO_3 + 2*N_2O_5 + O_3$" from tagged sources. This is in an attempt to track simultaneously the titration by NO_2 of ozone and its reformation from NO. The reviewer felt unsure whether this was sufficient to attribute the right fraction of ozone to a given NO_x or VOC source. There are other species (e.g., odd hydrogen) involved in ozone formation, which can be important sources of nonlinearity.

Other major strengths and weaknesses of OPTM identified by the group of reviewers are given below.

<u>*OPTM Strengths*</u>

(a) OPTM accounts for the production of O_x ($O_x = O_3 + NO_2 + NO_3 + 2*N_2O_5$) instead of ozone for each emissions component.

(b) Use of NO_x and VOC internal tagged species for tracking chemical production of O_x. It should be noted, however, that one reviewer felt that this could be considered a weakness, because these species could easily be estimated internally, saving users time and avoiding potential introduction of errors into the inputs.

(c) Implementation utilizes basic CMAQ modeling methods and tools that CMAQ users are familiar with, such as the use of CMAQ standard I/O API protocols for the tagged species.

(d) Straightforward implementation of forward computation for the "counter species," which does not interfere with base CMAQ results.

(e) An utility exists to edit the model code include (*.EXT) files to add new tags

(f) Ability to handle multiple ozone precursors in a single simulation.

(g) Implemented into CMAQ version 4.6 (a fairly recent release)

(h) Ability to directly relate ozone concentration to the emission sources and background precursor concentration (ICs/BCs), allowing policy makers to evaluate the model results easily.

(i) Capable of tagging species from multiple source categories, in different geographical locations, and in initial and boundary conditions.

(j) Capable of tracking the range of precursor transport.

(k) Relatively computationally inexpensive compared to the traditional sensitivity techniques such as brute force (zero-out) methods.

OPTM Weaknesses

(a) Mass conservation of the tagged species is not checked in the formulation. [Response ICF Page 3]

(b) OPTM does not distinguish between NO_x-limited and VOC-limited regimes. OPTM also assumes that all VOC species have the same reactivity. Therefore, ozone may be attributed to the wrong source. [Response ICF Page 3]

(c) Assumption of the fractional change in the total oxidant (sum of NO_2, NO_3, N_2O_5, and O_3) concentration due to dry deposition is questionable. [Response ICF Page 3]

(d) The contribution to O_3 by tagged NO_x and VOCs is counted separately, which is somewhat misleading because O_3 is formed through photochemical reactions of both NO_x and VOCs *jointly*. [Response ICF Page 3]

(e) It appears that OPTM is developed only for CB05 chemistry.

(f) Although species naming is straightforward, it needs some metadata description in the file (thus, users should be encouraged to provide metadata information for each tag)

(g) Requires additional effort to prepare the emission inventory of the tagged species compared to emissions processing for standard emissions inputs.

(h) OSAT may not represent the nonlinear response of atmospheric processes to emissions changes. Therefore, it can estimate the contribution of the tagged species only under the base-case emission scenarios; it cannot estimate the effects of emission changes on ozone concentrations.

(i) Current OPTM setup requires the code to be recompiled for each new set of tags. This approach places an unnecessary burden on model users and increases the chances of making mistakes. [Response ICF Page 3]

2.2 $PM_{2.5}$: PSAT and PPTM implementation strengths and weaknesses

The reviewers identified that both CAMx Particle Source Apportionment Technology (PSAT) and CMAQ Particle Precursor Tagging Methodology (PPTM) assume that all secondary PM is linked to a specific precursor: sulfate to SO_x, nitrate to NO_x, ammonium to NH_3, and secondary organic aerosol (SOA) to VOC emissions. For this reason, the reviewers agreed that a common weakness of both CAMx/PSAT and CMAQ/PPTM is that they cannot capture any indirect effects, such as an increase in nitrate concentrations resulting from a reduction in SO_2 emissions.

We present below additional comments on the specific strengths and weaknesses identified by the reviewers for each of CAMx/PSAT and CMAQ/PPTM.

2.2.1 CAMx/PSAT:

The other strengths and weaknesses of CAMx/PSAT identified by the reviewers are given below.

PSAT Strengths

(a) PSAT accounts for different VOC reactivities (with OH). It also accounts for different aerosol yields in SOA production from condensable VOCs.

(b) Can perform culpability assessment based on the generalized source apportionment scheme that can be applied to the nonlinear processes.

(c) Allows source apportionment study for sulfate (SO_4), particulate nitrate (NO_3), ammonium (NH_4), particulate mercury (Hg(p)), SOA, and six primary PM categories (EC, primary OC, crustal fine, other fine, crustal coarse, other coarse).

(d) Single tracer can track the primary PM species, whereas secondary PM species require several tracers to track the relationship between gaseous precursors and the resulting PM concentrations.

(e) PSAT uses tagged tracers to identify selected sources, source categories, and/or source regions contributing to simulated $PM_{2.5}$ concentrations. Thus, it can assist regulators in designing the most effective and cost-effective PM control strategies.

(f) PSAT source apportionment technique has been extensively tested and evaluated against other source apportionment techniques.

(g) Ability to directly relate PM concentration to the emission sources and background PM concentration (ICs/BCs), allowing policy makers to evaluate the model results easily.

(h) Capable of tracking species from multiple source categories, in different geographical locations, and in initial and boundary conditions.

(i) Capable of tracking the range of PM transport

(j) Can handle both primary and secondary particulate species in a single simulation

(k) Relatively computationally inexpensive compared to the traditional sensitivity techniques such as brute force (zero-out) methods.

(l) Option to use a gridded "mask" to define source regions for tracking, which means the input files need no modification

PSAT Weaknesses

(a) Mass conservation of the reactive species is not checked in the formulation. [Response ENVIRON page 1]

(b) No negative production is allowed for each source category. Therefore, if adding a certain precursor emission results in negative production, the model would assume the contribution to be zero instead. This could result in inaccurate apportioning of source impacts on secondary aerosol production. [Response ENVIRON page 1]

(c) PSAT is implemented for parallel processing using OpenMP but not with Message Passing Interface (MPI). [Response ENVIRON page 1]

(d) Requires additional effort to prepare the emission inventory of reactive tracers when not using the emissions mask feature. [Response ENVIRON page 2]

(e) May not represent the nonlinear response of atmospheric processes to emission changes (even though one of the design objectives of the technique is to account for nonlinearity in model response), particularly in the formation of secondary PM. Therefore, CAMx/PSAT can estimate the contribution of the tagged species only under the base-case emission scenarios; it cannot estimate the effects of emission changes on PM concentrations.

2.2.2 CMAQ/PPTM

The reviewers felt that the implementation of the CMAQ Particulate Precursor Tagging Methodology (PPTM) is mostly ready for use. The documentation provides usable instructions, although they need some improvement. However, the reviewers indicated that the current implementation, like the OPTM implementation, has too complex a configuration for even the simplest case (see Section 2.1.2). Model input file preparation is tedious, and the provided utilities require further development.

The documentation for PPTM is comprehensible, but could benefit from more illustrative examples. The examples in the appendix would be more useful if provided in-line with the instructions and if better commented, especially with respect to numbered options. The documentation is also missing examples of editing the INCLUDE files (i.e., *.EXT). Editing INCLUDE files should be well documented because it is a particularly tedious process.

Model code preparation utilities are not usable as provided. The "set_tag" utility gives a run-time error when editing AE_EMIS.EXT. Initial efforts to debug found that the AE_EMIS.EXT file's species names could be modified to make the utility work. Following that modification, however, the utilities "nitrogen tag" section of code fails. Debugging found another error regarding string indexing. The commenting in the utility codes is lacking, which makes debugging difficult. The code modification is dispersed and tedious; therefore, preprocessing utilities are critical to making PPTM usable for community members. Nonreactive species implementation is ready, but gas-phase chemistry requires further review.

The PPTM documentation indicates a more detailed representation of gas-phase chemistry than in OPTM, although this difference must be confirmed. For example, the gas-phase nitrogen chemistry in PPTM uses different assumptions compared to OPTM's—that is, in OPTM NO_x is the tracer for ozone, while in PPTM the $PM_{2.5}$ tracers are NO, NO_2, HONO, HNO_3, and PNO_3. Unlike in OPTM, the PPTM documentation suggests that longer-lived species are tracked individually. This improvement over OPTM should be confirmed in the code, and if confirmed, ported to the OPTM implementation. The revised code should then be reviewed for consistency with comments (a) and (c)(1) through (c)(4) from Section 2.1.2 (the OPTM review).

According to the PPTM documentation, "*the results for the tagged species are not normalized to ensure that the sum of the tagged species equals the total.*" This is of course not applicable for instances where the change in tagged species is apportioned from the change in the total (e.g., aqueous chemistry or cloud scavenging). Also, it is not true in all cases, as stated later in the documentation: "*At the end of the gas phase chemistry step, tags are normalized such that they add up to the correct overall concentration.*" When it is true (e.g., advection and diffusion), it can be a strength. "*The difference between the sum of all tags and the overall concentration gives an estimate of the numerical uncertainty*." The difference may be due to hidden nonlinearities, such as those in advection. Although advection is a linear process in theory, its numerical solution necessitates nonlinear flux corrections. The difference after advection can be used as a measure of this nonlinearity.

The apportionment is done at several points in the aerosol and cloud processes. This is much better than treating the entire module as a black box. It seems that this approach is similar to fitting several line segments between two points (input and output) instead of one long straight line. This way a curved line (nonlinearity) can be better approximated. Of course, this may not necessarily be what happens when different aerosol or cloud processes are apportioned separately. Some theoretical arguments or a numerical example would be useful to demonstrate that this is indeed a strength of the method, if it really is. The computational overhead seems to be high, especially for nitrogen tagging (50% increase for three nitrogen tags).

Other major strengths and weaknesses of PPTM identified by the reviewers are given below.

PPTM Strengths (related to code implementation)

(a) Distinction between species that go through linear processes and nonlinear processes, and appropriate "special treatment" based on the initial fractional distribution of species.

(b) Distinct identification of contributions of ICs/BCs.

(c) Use of CMAQ standard I/O API protocols for the tagged species.

(d) Use of averaged concentrations (between layers BLEV and ELEV) for the output.

(e) Sulfur tagging in aerosol phase utilizing intermediate process checkpoint concentrations.

(f) SOA tagging, including tracking of toluene and cresol for each tag; distinction of anthropogenic and biogenic contributions.

(g) CMAQ/PPTM can assist regulators in designing effective PM control strategies.

(h) Implemented into CMAQ version 4.6 (a fairly recent release).

(i) Can handle both primary and secondary particulate species in a single simulation.

(j) Ability to directly relate PM concentration to the emission sources and background PM concentration (ICs/BCs), allowing policy makers to evaluate the model results easily.

(k) Capable of tagging species from multiple source categories, in different geographical locations, and in initial and boundary conditions.

(l) Capable of tracking the range of PM transport.

(m) Relatively computationally inexpensive compared to the traditional sensitivity techniques such as brute force (zero-out) methods.

PPTM Weaknesses

(a) Mass conservation of the tagged species is not checked in the formulation. [Response ICF page 4]

(b) PPTM assumes that all secondary PM is linked to a specific precursor: for example, sulfate to SO_x, nitrate to NO_x, ammonium to NH_3, and SOA to VOC emissions. For this reason, PPTM cannot capture any indirect effects, such as an increase in nitrate concentrations due to a reduction in SO_2 emissions. [Note: also a weakness of CAMx/PSAT] [Response ICF page 4]

(c) Sulfur tagging in aerosol lacks methods to account for individual chemical production pathways that are the main causes of nonlinearity in the sulfate production. [Note: also a weakness of CAMx/PSAT] [Response ICF page 4]

(d) Although species naming is straightforward, it needs some metadata description in the file (thus, users should be encouraged to provide metadata information for each tag).

(e) The code design is intrusive. It is not apparent in the user's guide why the developers chose the current design of CMAQ/PPTM, as opposed to the approach used by CAMx/PSAT and CMAQ/TSSA.

(f) Current CMAQ/PPTM design requires the CMAQ source code to be modified and recompiled for each set of PPTM simulations. Even though the "set_tag" utility program is provided to create the *.EXT files from templates, the PPTM approach not only increases the complexity of the source code (multiple versions of executables), it also increases the chances of making mistakes. [Response ICF page 4]

(g) Current PPTM approach also requires emissions tracers to be added to CMAQ emissions inputs. Again, this intrusive approach to emissions processing requires extra work and is prone to processing/QA errors. [Response ICF page 4]

(h) PPTM outputs are written into standard CMAQ output files. This would lead to large output CMAQ files, increasing the potential for file corruption and access problems. A better approach is to create additional output files specifically for CMAQ/PPTM results. [Response ICF page 4]

(i) May not truly represent the nonlinear response of atmospheric processes to emission changes (even though one of the design objectives of the technique is to account for nonlinearity in model response), particularly in the formation of secondary PM. Therefore, PPTM can estimate the contribution of the tagged species only under the base-case emission scenarios; it cannot estimate the effects of emission changes on PM concentrations.

3. Comparison to Other Methods Such as DDM, Zero-out, and TSSA

In this section, source apportionment techniques are compared with alternative approaches to estimating source contribution, such as the CMAQ Tagged Species Source Apportionment (CMAQ-TSSA) (Tonnessen et al, 2007, Wang et al, 2009) the Decoupled Direct Method (DDM) (Dunker et al, 2002a) and the traditional brute-force ("zero-out") methods for source sensitivities.

DDM is conceptually different from source apportionment because this technique estimates source sensitivity rather than total contribution. Sensitivity is something that can be translated into a response to an emission control strategy. The first-order sensitivity is defined as the change in concentration resulting from an infinitesimal emission perturbation (or first derivative of concentration with respect to emissions). The second-order sensitivities are the derivatives of the first-order sensitivity with respect to the same or other emissions (mixed derivatives). Third-order sensitivities are the derivatives of the second-order sensitivities, and so on. The response to an emission change can be approximated using Taylor series expansion. Typically, only first- and second-order sensitivities are calculated by DDM (second-order sensitivities are not yet developed for aerosol or cloud processes). The first-order sensitivities are sufficient to represent a linear response. Second- and higher-order sensitivities are needed to capture the nonlinearities of the response. When only (first- and) second-order sensitivities are used, an error is introduced due to the absence of higher-order terms. This error becomes larger for larger emission changes. Hence the (approximate) response is relatively more accurate for small changes in emissions. As an example, the response calculated from DDM sensitivities would be much more accurate for a 30% reduction of the emissions than for a 70% reduction of the emissions.

Source apportionment, on the other hand, is a concept designed for efficiently estimating the total contribution of emissions from a certain source. It is most suited for judging which source is the cause of conditions present in the model. Theoretically, the contribution of a source is equivalent to the response that should be observed when that source is taken out completely.[1] Zeroing out a source is the maximum possible perturbation of the base case, which includes 100% of that source's emissions. DDM sensitivities are the least reliable for 100% emission reductions. So, if the emission control decision to be made involves shutting down or completely turning off a source, then source apportionment may be the more appropriate application.

The tools reviewed here (OPTM, OSAT, PPTM, PSAT) utilize the same counter-species forward integration method, in which a set of counter species are individually (if possible) or as a group (if the responses are expected to be nonlinear) computed utilizing the identical algorithms of the base model to provide the source-receptor relationships of the inputs (ICs, BCs, and emissions of precursor species). For the species that can be linearly superimposed, the method is quite useful. On the other hand, for the species or inputs that go through nonlinear processes or depend on other species, the total mass budget of each processing step is used to prorate the incremental

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[1] In practice, when the same source is zeroed out, a different response can be obtained from the models, as is pointed out both in OPTM/PPTM and OSAT/PSAT documents.

contributions. Because of the positive-definiteness requirements of the concentrations in AQMs, the source apportionment results for the nonlinear processes or that involve multiple species (e.g., species interactions such as NO_x reductions changing sulfate or SOA) may not correctly apportion individual contributions of the tagged inputs. Because the tagging process does not interfere with the base model simulation, the source apportionment results are expected to be closer to those from the DDM- and Higher Order DDM (HDDM)-based tools (Hakami et al, 2003, 2004) if the “zero-out” of the inputs is accounted for with the sensitivity (gradient) information.

The brute-force “zero-out” simulation changes the base state when multiple “zero-outs” of inputs are realized. It may be closer to the way the real atmosphere would respond if sufficient inputs are changed, but computing the response to each of the inputs could be very tedious. On the other hand, if the delta (control) is large, we may not obtain a proper understanding of the shape of the response surface subject to the various magnitudes of input controls. Often entire states, emissions sectors, or metropolitan areas are zeroed out, which may lead to a drastic change in regional chemistry that is unrealistic. Therefore, a large number of simulations with incremental changes would be required to cover the whole range of control possibilities. Accomplishing this across multiple changes could be very costly and time consuming in terms of both computer and human time.

For DDM and HDDM, the base state does not change when estimating the gradient surface (response surface). Then the effects of control, including the zero-out, could be estimated using the gradient during the postprocessing. The magnitude of delta changes that can be applied depends on the nonlinearity of the system to the input changes. Usually, the higher-order DDM scheme can extend the range of control deltas, although it will still be difficult to justify the total zero-out of certain inputs.

For the counter-species-based tools reviewed here, as for DDM or HDDM, the base state is not altered during the computation. The effects of each tag can be assessed only at the 100% control level (i.e., zero-out). However, note that “zero-out” postprocessing of DDM and HDDM are actually not recommended (i.e., the computed sensitivity may not be as valid). In this regard, it would not be reasonable to select the tagged species to represent only small perturbations in the inputs. The counter-species tools reviewed here are more comparable to results from brute-force zero-out simulations.

CMAQ-TSSA is a particulate source apportionment approach implemented in an earlier version of the CMAQ model. It has not been implemented in recent versions of CMAQ. The CMAQ-TSSA approach makes simplifying assumptions, just as PPTM (or OPTM) and PSAT (or OSAT). The reviewers felt that TSSA may be more accurate than other methods because it employs the integrated reaction rates for the tagged species in gas-phase chemistry. However, for aerosols, it splits the change in the bulk species proportionally to the tagged species. Similar to PSAT, only one version/copy of CMAQ/TSSA executable is needed to perform multiple sets of source apportionment runs that vary in the species/source categories. CMAQ/TSSA creates new source apportionment output files and does not “perturb” the standard CMAQ outputs, which again is an approach more comparable to PSAT. It is important to note that TSSA was not specifically reviewed as part of this peer review process so a comprehensive list of strengths and weaknesses are not presented.

The most accurate approach would be a method that splits each pollutant into different source-specific species and tracks these source-specific species separately through the model, and makes no assumptions of linearity (except perhaps that all PM species can be traced back to a specific precursor gas-phase species or primary PM component; note that this would still be a significant weakness of source apportionment). An example of this approach can be found in Kleeman and Cass (2001). Such a method would be extremely computationally demanding, but it would be the most accurate. If implemented in CMAQ and CAMx, it could be used as a benchmark for the more computationally efficient methods discussed here.

To summarize:

(a) A zero-out run is a sensitivity simulation, and sensitivity experiments are affected by nonlinearity in chemical reactions, so they are not definitive in characterizing source contributions. Because PSAT and PPTM track mass contributions to PM formation, they identify source contributions in a way that accounts for chemical nonlinearity.

(b) DDM in an air quality model is an efficient and accurate way of performing sensitivity analysis to model inputs. However, while DDM gives information on the atmospheric response to emissions changes, and finds the uncertainty in the response, it does not attribute concentration among all sources regions and categories. DDM also may or may not be applicable for large perturbations.

(c) Compared with other source apportionment methods, TSSA is similar to CAMx/PSAT and CAMx/OSAT, but slightly different in the treatment of chemistry and dry deposition.

The source apportionment approaches are fundamentally different from the DDM and zero-out approaches. DDM calculates first- and higher-order sensitivity coefficients with a greater computational efficiency, although the implementation of DDM may be somewhat technically challenging, and it may not effectively captures the nonlinear response of models. Zero-out (or certain percentage of emission reduction) simulations might be computationally expensive, but the method is direct, intuitively easy to understand, and does not require additional modeling skills as compared to standard simulations.

We further provide a comparison to other source sensitivity/source attribution methods:

(a) Data analysis methods, such as chemical mass balance (CMB) and positive matrix factorization (PMF), are limited by the availability of ambient monitoring data, and can be used only to evaluate historical episodes.

(b) Back-trajectory analyses cannot adequately treat the complex chemistry of secondary PM formation.

(c) The source-oriented external mixture (SOEM) method is potentially the most accurate approach for tracking source contributions. However, this method is computationally demanding, as it solves an increasing number of stiff, coupled differential equations for each time step in the gas and aerosol chemistry modules as well as increasing the number of species in the aerosol calculation.

(d) Carbon tracking was implemented into CMAQ to track primary organic and elemental carbon contributions. However, this approach does not address a number of other gases and aerosol species in CMAQ.

4. Documentation Strengths and Weaknesses

In this section, we provide a composite summary of the reviewers' responses to a series of charge questions relating to documentation.

a. Is the current documentation for CMAQ/PPTM and CMAQ/OPTM adequate?

The reviewers felt that the documentation was barely adequate, and suggested that it could be more detailed for a typical modeler who wants to use the tools for routine model applications. The documentation provides very nice descriptions on how the technologies are implemented. However, the operational aspects to implement the technology, including tagged emission preparation, building the tools with the CMAQ modeling system, modifying the extension files, and execution of the tools, could be more explicit and organized. The documentation's indication of the estimated increase in the computational cost is helpful, giving users an idea of the elapsed time when using the tools.

Ideally, there are two types of documentation needed for this type of application. The first type is a user's guide. The second type consists of the online comments inserted into the CMAQ source code. The shortcomings of each of these are discussed below.

The shortcomings in the CMAQ/PPTM and CMAQ/OPTM user's guide identified by the reviewers include the following:

(a) Does not provide adequate discussions comparing PPTM/OPTM to other sensitivity approaches (zero-out run, DDM, etc.) available to model users.

(b) Some parts of the user's guide need more work. One example is the aerosol dynamics section for secondary organic aerosols. The technical discussions in this subsection seem to suggest that PPTM uses the kinetic approach, but it is not clear from the documentation.

(c) Does not explain what modules OPTM/PPTM were implemented for. For example, for which CMAQ chemistry solver(s) has OPTM/PPTM been implemented?

(d) Does not cover whether the OPTM/PPTM is incorporated into the CMAQ CVS repository.

(e) Does not adequately address how the "tags" are assigned.

(f) Does not include sample CMAQ run scripts with the OPTM/PPTM option.

(g) Does not cover the performance of OPTM/PPTM in terms of efficiency (speed) and accuracy (error) as compared to the traditional zero-out method.

(h) Lacking in formal statistical comparison. It seems that results presented were simply summaries from OPTM/PPTM runs and lack rigorous evaluation of these results compared with those from other methods (e.g., zero-out comparison).

(i) Details for species-specific approach (1) for each reactive species (sulfate, nitrogen, and secondary organics) and (2) for nonreactive species (primary organic carbon, elemental carbon, and other inorganic particles) are missing.

(j) Does not provide insights as to why model users should choose CMAQ/PPTM and CMAQ/OPTM over other source apportionment methods such as CAMx/PSAT, CAMx/OSAT, and CMAQ/TSSA.

(k) On page 6 (last paragraph) of the technical description and user's guide, the discussion regarding the offset in the oxidant tracers for ozone titration by NO seems a bit arbitrary and should be clarified.

(l) In the documentation of CMAQ/PPTM, the discussion on how the aerosol dynamics and cloud process calculation is formulated could be further elaborated. How does the tool assign I and J modes for each tagged PM species?

(m) The rationale behind the nitrate apportionment should be explained in the CMAQ/PPTM documentation. Currently, formulae under nitrogen tagging are difficult to follow.

The shortcomings for online documentation in the CMAQ/PPTM and CMAQ/OPTM source code include the following:

(a) As EPA developed CMAQ, extra efforts were expended to make the code "readable." However, the CMAQ/PPTM and CMAQ/OPTM implementation in CMAQ does not fully follow the "rules of coding."

(b) Although the CMAQ/PPTM code starts with the line "cicfpptm", the comments do not explain where the new code ends and how it differs from the standard CMAQ release.

(c) The comments do not always provide sufficient information describing the purpose of the new code in each OPTM/PPTM section.

(d) The OPTM/PPTM code fails to explain what each variable means at the beginning of each subroutine. The code also does not document the unit used for each variable.

b. Is the current documentation for CAMx/PSAT and CAMx/OSAT adequate?

The reviewers felt that the documentation for PSAT and OSAT is well written, but could also be improved. Operationally, it could be more detailed for a typical modeler who wants to use the tools for routine model applications. The documentation provides excellent technical discussion on the methodology and model formulation of OSAT and PSAT. The CAMx user's guide also provides sufficient references on other source apportionment methodologies. The paper by Dunker et al. (2002b) complements the OSAT documentation. The online documentation and comments in the CAMx code are also better than those in CMAQ/OPTM and CMAQ/PPTM. The operational aspects regarding how to implement the technology, including reactive tracers emission preparation, how to build the source apportionment tools, and the step-by-step procedure to run CAMx/PSAT and CAMx/OSAT, could be more detailed. A few other short-comings in the CAMx OSAT and PSAT documentation identified by the reviewers include the following:

(a) In the documentation of CAMx/PSAT, only the concentration change of the reactive tracers is discussed. Some description on how the aerosol dynamics and cloud processes are treated in the formulation would be helpful for readers.

(b) On page 6-8 of the OSAT documentation, it states that IC/BC ozone is split equally between the VOC- and NO_x-limited categories. This seems to contradict Figure 6-2.

(c) On page 6-9, advective flux normalization could be better explained by using equations. There is probably a difference between the weighting of tracers N and O3N because the first one combines NO and NO_2 but the second one splits O_3. Some equations (e.g., on pages 6-9 and 6-10) are written like FORTRAN code. The same quantities appear on both sides of the equation without distinction of their position in time.

Two typographical errors are pointed out:

(a) On page 6-13 “based on response the of the peak ozone”

(b) On page 6-34, “you will may be asked,”

c. Are the above model formulations sufficiently documented for technical critique and understanding?

The reviewers generally felt more satisfied with the documentation for CAMx/PSAT and CAMx/OSAT than with the documentation for CMAQ/OPTM and CMAQ/PPTM. The documentation of CAMx/PSAT and CAMx/OSAT is particularly well written. It provides detailed scientific assessments of the technologies and their relationship to other source apportionment technologies. The discussion on the rationales and design goals of the source apportionment tools is also very well described in the documentation. However, for a few nonlinear processes, detailed presentations of intermediate tagging species would be useful. The CMAQ OPTM/PPTM documentation could use a better overview of the source apportionment methodology. One reviewer felt that only after reading documentations for both CAMx and CMAQ together does one get a complete overview of the source apportionment procedures.

d. Is the discussion and presentation of the above models and their features clear? Please note any specific sections of the documentation that were unclear or confusing.

The responses to this question are combined in the responses to questions 4a and 4b above.

e. Is the documentation for each of the above models sufficient to guide a typical user in the use of the model and its preprocessors?

This is the area in which the reviewers felt that the documentation could be more explicit and detailed. Considering the additional efforts required to implement the technologies in modeling exercises, step-by-step instructions (especially in the preparation of tagged emissions), information on modifications needed for the model runs, and explanations of potential problems that may be encountered in the application of the technology should be included that are aimed at a typical user who is implementing the technologies. Again, the reviewers felt that this portion of the documentation was satisfactory for CAMx/OSAT and CAMx/PSAT, but not for CMAQ/OPTM and CMAQ/PPTM.

Because the reviewers were not asked to go through a complete modeling exercise using these tools, they felt that additional deficiencies might be uncovered as one goes through the entire process, from preprocessing through postprocessing.

f. What improvements are desirable for the documentation of each of the above models?

As noted in the responses to question 4e above, the reviewers felt that additional details on the operational aspects of the models would be very helpful. The application of CMAQ/OPTM and CMAQ/PPTM is easier to follow, although the execution of CAMx source apportionment tools seems to be more straightforward. A step-by-step procedure for running CAMx/PSAT and CAMx/OSAT would be very helpful.

In PPTM/OPTM documentation, a more detailed explanation of the Emissions and IC/BC Utility programs is recommended. The description of the examples (Douglas et al, 2007 Section 2.2 and 3.2) for OPTM and PPTM application procedures could be expanded. The PSAT documentation includes a comparison of source apportionment with source sensitivity. One reviewer urged that the documentation authors add a discussion of the practical value of source apportionment and source sensitivity in a regulatory context, placing that new text before the somewhat theoretical comparison already in the document. In other words, **in a real-world context, why do we need source apportionment?** Including examples of how source apportionment is (or could be) used by regulators would be extremely useful. Moreover, the reviewer suggested that a similar discussion is needed in the documentation for all of the tools.

g. What changes in organization of the documentation of each of the above models are desirable?

The organization of the PPTM/OPTM documentation could be revised such that the processes common to sulfur, nitrogen, and organics tagging are covered in a general tagging section. This way some of the repetition could be avoided. Further, for a few nonlinear processes, detailed presentations of intermediate tagging species would be useful.

h. Does the documentation contain useful and appropriate evaluations of source apportionment for ozone and $PM_{2.5}$ species compared to other approaches?

The PPTM/OPTM documentation does not contain any comparative evaluation. There are no comparisons or other types of evaluations for OSAT either. The PSAT documentation contains a comparison to a sulfate zero-out run, which is quite useful. There is also a comparison of reactive nitrogen (NO_y) apportionment by PSAT to the SOEM algorithm of Kleeman and Cass (2001). It is not clear, however, whether this one-dimensional (box-model) problem is relevant or robust enough to evaluate PSAT. There is a more realistic comparison of PSAT to a method similar to SOEM in Wagstrom et al. (2008).

The reviewers indicated that there are some data and visualizations showing that all the source apportionment tools agree reasonably well with the model results based on zero-out methods. However, they would have liked to see information on how much the source apportionment results would differ if the same emission tagging were applied for different grid resolutions and for emissions with very different reactivity. Some specific additional comments are given below.

CAMx-OSAT/PSAT:

(a) The CAMx user's guide contains more useful and appropriate evaluations than does the CMAQ/PPTM user's guide.

(b) The CAMx user's guide has a good comparison of the OSAT/PSAT methodologies with other approaches.

(c) The CAMx user's guide also provides some comparison of OSAT/PSAT results with results from other approaches (such as from zero-out and SOEM runs).

(d) The CAMx user's guide's references section is much better produced than that of the CMAQ-OPTM/PPTM user's guide.

CMAQ-OPTM/PPTM:

(a) The CMAQ-OPTM/PPTM user's guide has very little information on the comparison of OPTM/PPTM to other approaches. A well thought-out methodology should be able to withstand a rigorous comparison with other approaches. However, this discussion is notably lacking in the CMAQ-OPTM/PPTM user's guide.

(b) There is no information on the performance of CMAQ-OPTM/PPTM versus zero-out runs or other approaches (DDM, SOEM, etc.).

(c) Other than the developers' work (i.e., references to CMAQ and CMAQ's tools (I/O API and SMOKE), the user's guide does not refer to or quote any other source apportionment methodologies.

5. Preprocessing and Postprocessing Strengths and Weaknesses

In this section, we provide a composite summary of the reviewers' responses to a series of charge questions relating to preprocessing and postprocessing tasks/tools.

a. In general, how manageable do you consider the task of preprocessing/model setup to run source apportionment for CMAQ/PPTM, CMAQ/OPTM, CAMx/OSAT, and CAMx/PSAT?

The reviewers agreed that the task of preprocessing/model setup to run CMAQ/PPTM, CMAQ/OPTM, CAMx/OSAT, and CAMx/PSAT is manageable for experienced modelers but may not be as easily adapted for routine operation. The preparation required for the additional tagged or reactive tracers species can be massive, especially when many emission sources/locations are tagged (which is the primary strength of the source apportionment tools). Regarding the model setup to run source apportionment tools, more detailed and explicit descriptions seem necessary. It would be very helpful if detailed operational procedures for building and running the tools could be included in the documentation. Some specific comments for each of the two sets of tools are given below.

CMAQ/PPTM and CMAQ/OPTM

It seems generally manageable to apply these, especially utilizing the general tools such as "set_tag" for updating the tagged species list in the model, and scripts based on m3xtract for the preparation of emissions file. However, the following features need to be addressed:

(a) A method to check the completeness of code modifications (i.e., whether the modifications of the INCLUDE files are correct).

(b) The number of “tagged” species allowed in one I/O API file is generally the default, so the I/O API libraries will usually need to be recompiled by the user to include many more species.

(c) Setting up CMAQ-OPTM/PPTM requires more work than a regular CMAQ run. The biggest problem is having to maintain the different copies of CMAQ-OPTM/PPTM executables and input files. If a proper naming convention were not agreed upon at the initiation of a project, the housekeeping for these would become a nightmare. This problem would be exacerbated if model/data transfer occurs among different agencies/groups.

CAMx/OSAT and CAMx/PSAT

Considering that several different groups have applied CAMx/OSAT and CAMx/PSAT for source apportionment study over many different geographical areas, the preprocessing and postprocessing of these tools seem manageable.

(a) Setting up a CAMx-OSAT/PSAT source apportionment run is quite manageable for a modeler with previous experience running CAMx.

(b) The file structure of CAMx is not as portable as netCDF and requires additional modeling skills to deal with the model datasets.

b. Are postprocessing tools easily available and easy to use for CMAQ/PPTM, CMAQ/OPTM, CAMx/OSAT, and CAMx/PSAT?

There is no postprocessing tool provided for CMAQ-PPTM/OPTM. However, since the output of CMAQ-PPTM/OPTM is directly written as additional species in the standard CMAQ output files, the users can simply follow the general routines provided by I/O API utility tools to extract the needed output for postprocessing. On the other hand, if postprocessing run scripts or utility programs were to be provided, that would certainly be helpful for the users, who could then obtain the source apportionment results more directly.

The reliance of PPTM and OPTM on standard CMAQ postprocessors is an advantage, but at the same time it also limits the analyses to only those that can be performed with CMAQ post-processors. The analyses desirable for source apportionment results may be somewhat different than the analyses typically conducted with standard CMAQ results. For example, pie charts at certain receptor locations on a map showing the fractions of contributions may be more desirable than several maps of individual contributions. It is not clear how the various contribution charts in the documentation were prepared. It would be helpful if there were a postprocessor to generate these types of charts from standard model outputs. For example, formulae needed to calculate the amount of ozone attributed to each tag could be provided as a PAVE script. Scripts to interface with other charting options (e.g., MS Excel) could also be provided to the user.

For CAMx/OSAT, additional output files for instantaneous concentration, surface average concentration, and reactive tracer concentration at a given receptor location are written in either FORTRAN binary or ASCII format. This is a plus in terms of simplifying the postprocessing effort, since it is not necessary to deal with the large output files of the standard simulation. The reactive tracer concentration at a user-specified location output to an ASCII file format can be visualized easily using a rudimentary knowledge of Excel. For CAMx/PSAT, it is not clear from the documentation whether any postprocessing tools are provided.

6. Supplemental Questions

(c) What do you consider the general advantages and disadvantages of zero-out modeling when a 100% emissions perturbation is necessary for an application such as source culpability?

Zero-out modeling implies 100% reduction of emissions in the model and source culpability implies that the contribution of a source that is emitting at full capacity is being sought. If these are the intended implications, then one of the reviewers opined that zero-out modeling would yield the wrong contribution of a source to a pollutant that is a non-linear function of precursor emissions (e.g., ozone, secondary organic aerosol). Source apportionment methods such as those reviewed here (not considering their weaknesses in tracking non-linear relationships) are better suited for this purpose. However, if the goal is to see what would happen if a source were eliminated, then zero-out modeling is the better approach.

In addition to the above, the reviewers identified the following advantages and disadvantages:

<u>*Advantages*</u>:

- Zeroing-out directly changes the chemical composition, which should reflect the chemistry better than any of the source apportionments techniques.
- Intuitively easy to understand and to interpret the model results.

<u>*Disadvantages*</u>:

- Multiple runs are required if there are multiple sources that need to be evaluated
- Computationally expensive
- Although the zero-out modeling is useful in assessing the source culpability, if the area and amount of subtraction are substantial, then the base state may be changed and thus the response to the perturbation may be different from the base state. Further, sum of culpability may not add up to be balanced to the large changes in the state because the system would become more nonlinear.

(d) Do you feel that the implementation of mercury tagging in CMAQ/PPTM and CAMx/PSAT has any important weaknesses that may need to be addressed or strengths as a tool for culpability assessment?

One reviewer felt that the mercury tagging in CMAQ (and CAMx/PSAT, too, if the implementation schemes are similar) is a good tool for culpability assessment in terms of concentration and dry deposition of mercury because the inconsistent chemistry between the tracer and base mercury species is much less of an issue due to two reasons:

(1) Mercury emission is many orders of magnitude smaller compared to VOC and NOx emission. As a result, the presence of mercury in the air composition does not affect the photochemistry of oxidant formation, which is an important driver for mercury deposition.

(2) Air concentration of mercury is very small and existing model data indicate that the response of mercury dry deposition to air concentration is very linear. This agreed with the linear assumption in the source apportionment tool much better compared to the case of O_3. The response of mercury wet deposition to concentration is not as linear because of wet scavenging and aqueous chemistry, though.

Other reviewers however felt that the strengths and weaknesses that were listed for these models were general so they would also apply to mercury tagging. However, they could not answer the question of whether there are weaknesses (or strengths) particular to the mercury chemistry without further review of the models. Specifically, if the aqueous-phase and thus the response of the system to the input changes were significantly nonlinear, then similar difficulties would exist as the culpability assessment of the SOA, and hence further review was warranted.

7. Suggestions for Improvements to or Additional Evaluation of Source Apportionment

It is not clear whether either CMAQ-PPTM/OPTM or CAMx-PSAT/OSAT can perform source apportionment simulations for ozone and particulate matter *simultaneously*. In light of the one-atmosphere modeling in the CMAQ framework and CAMx modeling system, the capability of simultaneous source apportionment calculation for O_3 and PM should be considered [Response ENVIRON page 1]. Because there are complex interactions among the ozone precursors, the radical species, and various particulate species (including SO_4, NO_3, and SOA), how the tagged species (or the reactive tracers in CAMx-PSAT/OSAT) interact with each other, and how consistent these interactions are compared to those in the standard simulation, need to be carefully examined.

To encourage the application of these very useful tools, the reviewers recommended that additional emission processing tools and utility programs that facilitate the use of the source apportionment tools be developed and provided. The implementation of source apportionment tools into a modeling exercise requires extensive preprocessing work in the preparation of tagged emissions data (including IC and BC species) the addition of new model parameters in CMAQ or CAMx, modification of multiple run scripts, and dealing with the increased number of input and output parameters. These tasks can be cumbersome, so it would be very useful if additional utility programs were available to reduce the level of effort required.

It would be beneficial if the source apportionment tools could be more modular and portable. The codes seem to be closely tied to each science processor in their respective AQM. With the current pace of model advancement and mechanism updates, the source apportionment tools may not immediately work with the newer releases of the models and so may require additional code modifications by the users or by the developers. Such portability and code maintenance issues may present a challenge to a typical modeler.

The way that the four tools (CMAQ/PPTM, CMAQ/OPTM, CAMx/PSAT, and CAMX/OSAT) calculate the concentration change of the tagged species (or reactive tracers) is similar: the concentration change of a tagged species at each time step is calculated according to the concentration change of the corresponding species in the standard model weighted over its fraction in the total tagged species. This brings about some potential concerns that require further examination.

- First, the reactivity of the gas mixture in the standard model may not represent the reactivity of a tagged species, but the concentration change of the tagged species is calculated based on the concentration change of the gas mixture in the standard model. This may overestimate the concentration change for a tagged species having low reactivity and underestimate the concentration change for a tagged species having high reactivity. For example, for two emission sources (A and B) located in the same grid cell of the model domain, if Source A emits only the low-reactivity VOCs (VOCA) and Source B emits only high-reactivity VOCs (VOCB), the VOC mixture in that grid cell should have a reactivity somewhere between VOCA and VOCB. In CMAQ/OPTM, because ozone attributed to a tagged source is directly proportional to the oxidant production contributed by the tagged species, this can produce a bias in the source apportionment. In CAMx/OSAT and CAMx/PSAT, the concentration change of the reactive VOC tracers is also weighted according to their respective OH reactivity, which should produce a smaller bias. However, this needs to be verified with additional model data. [Response ENVIRON page 1]

- Similarly, the tools may produce different source apportionment results at different grid resolutions because of the different degrees of instantaneous dilution upon the emission of the tagged and standard species and/or the inclusion of a different number of emission sources at different grid sizes. Testing the tools at two different grid resolutions would be helpful to assess whether the apportionment results are sensitive to grid resolution. [Response ENVIRON page 2]

- For all these methods, it is necessary to track the effects of nonlinearities in all processes, but most urgently in chemistry. The treatment of chemistry in TSSA could be used as an example. Basically, the production and loss of each tracer (or tagged component) should be tracked independently. Processes that are treated as black boxes, where the change in the overall mass (or flux) is split proportionally to the components (tags), are essentially assumed to be linear. It is desirable to have more explicit tracking of the tags through these processes. [Response ENVIRON pages 2-3]

In the CAMx user's guide, there is no detailed information on the treatment of the nonlinear processes for CAMx/PSAT and CAMx/OSAT, other than the general approach of treating the nonlinear chemical processes based on the mass budget redistribution according to the initial concentrations of each step. In the CMAQ documents, at least there are statements that internal checkpointing is utilized to assure mass balancing. However, in one reviewer's opinion, if at all possible the direct computation of counter species would be preferable, to identify the degree of nonlinearity of each processing step, then reconcile these with the results of the base simulation.

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Appendix A: Peer Review Process Narrative

In an effort to provide for independent, credible peer reviews of source apportionment techniques that are available in air quality models, the Air Quality Modeling Group of the U.S. Environmental Protection Agency's Office of Air Quality Planning and Standards issued a work assignment (WA) under a general contract (EP-D-07-102) with the University of North Carolina at Chapel Hill's Institute for the Environment to manage and coordinate the peer review process.

Once the work assignment was issued (October 2008), UNC prepared a detailed work plan for the approval of the EPA Work Assignment Manager (WAM), Dr. Kirk Baker. The work plan provided a description of the tasks to be completed, the estimated time frame, and the estimated person-hours and cost requirements. The description below describes the process by which the peer review for the source apportionment tools in CAMx and CMAQ was conducted.

The information provided to UNC under the WA Statement of Work included:

- a "Charge to Reviewers," formulated by EPA, that outlines the specific direction and technical scope of the task for the peer review team;
- a list of qualified candidates known to EPA from which to choose the peer reviewers (UNC then added a few more candidates to the list); and
- a list of materials to be provided to the peer reviewers by UNC and EPA

UNC contacted people on the qualified candidates list, described the project to them, and requested their participation based on their interest and availability. Five candidates were retained: Dr. Daewon Byun (University of Houston), Dr. Harvey Jeffries (University of North Carolina at Chapel Hill), Dr. Jerry Lin (Lamar University), Dr. Talat Odman (Georgia Institute of Technology), and Dr. Gail Tonnesen (University of California, Riverside). However, Dr. Tonnesen was subsequently unable to complete the peer review due to schedule conflicts, so we identified one of her colleagues, Dr. Zion Wang, to replace her. The chosen peer review team was approved by the WAM. Dr. Sarav Arunachalam was designated the chairperson, in that he would compile a report that would summarize all of the peer reviewers' comments and opinions into one concise document.

UNC provided a subcontract mechanism through which the peer reviewers could be compensated for their time. UNC then reproduced and distributed the review materials to all reviewers. These materials were provided to UNC by EPA, and consisted of the following:

(a) Source code for CAMx with OSAT and PSAT.

(b) Source code for CMAQ with OPTM and PPTM.

(c) Inputs and outputs for test case CAMx application with OSAT and PSAT.

(d) Inputs and outputs for test case CMAQ application with OPTM and PPTM.

(e) Douglas, S., T. Myers, and Y. Wei (2006). Implementation of Mercury Tagging Methodologies in the Community Multiscale Air Quality (CMAQ) Model, Technical Description and User's Guide, prepared for the U.S. EPA OAQPS, July 2006.

(f) Myers, T., and S. Douglas (2007). Implementation of Ozone and Particle Precursor Tagging Methodologies in the Community Multiscale Air Quality (CMAQ) Model, Technical Memorandum, prepared for the U.S. EPA OAQPS, March 2007.

(g) Douglas, S., T. Myers, and Y. Wei (2007). Implementation of Ozone and Particle Precursor Tagging Methodologies in the Community Multiscale Air Quality (CMAQ) Model, Technical Description and User's Guide, prepared for the U.S. EPA OAQPS, November 2007.

(h) ENVIRON, Inc. (2008). Comprehensive Air Quality Model with Extensions Version 4.5, User's Guide, May 2008.

(i) Tonnessen, G., and Z. Wang (2007). Development of a Tagged Species Source Apportionment Algorithm (TSSA) to Characterize 3-Dimensional Transport and Transformation of Precursors and Secondary Pollutants, Final Report, University of California, Riverside, May 2007.

Once these materials were distributed, UNC scheduled a teleconference call that involved the peer reviewers, Sarav Arunachalam of UNC, and Kirk Baker of EPA. The conference call was conducted on 9/30/2008 to discuss the charge to the reviewers and establish commonality in the peer reviewers' efforts, and to initiate the peer review. As part of this discussion, the peer reviewers identified additional pieces of documentation that might be helpful or relevant for the review. Dr. Arunachalam distributed any additional information among the peer review team.

The peer reviewers were instructed to perform their review according to instructions in the "Charge to Reviewers" (Appendix B in this report). It was agreed that contact among them was permissible and encouraged.

After the reviewers provided their individual comments, UNC and EPA reviewed the comments, and generated a few additional questions for the reviewers. The reviewers were asked to respond to these questions, which were mostly requests for clarification of their original comments. UNC then compiled all the comments into a draft report and distributed this via e-mail on August 21, 2009 to all parties for their review.

The final version of the report was compiled, reproduced, and submitted to EPA by UNC. This compilation included the addition of other information, such as this overview of the process, and the résumés of the reviewers (Appendix C).

Appendix B: Charge to Reviewers

1. Model formulation

(a) What are the strengths and weaknesses of CMAQ/PPTM?

(b) What are the strengths and weaknesses of CAMx/PSAT?

(c) What are the strengths and weaknesses of CMAQ/OPTM?

(d) What are the strengths and weaknesses of CAMx/OSAT? (OSAT includes both the OSAT and APCA ozone source apportionment approaches)

(e) Are there any modules or features of CMAQ/PPTM, CMAQ/OPTM, CAMx/PSAT or CAMX/OSAT in which an improved formulation or treatment is necessary? If so, please discuss what is needed.

(f) In general, how do the above source apportionment approaches compare with other source apportionment methods like TSSA and source sensitivity approaches like DDM and zero-out simulations?

2. Documentation

(a) Is the current documentation for CMAQ/PPTM and CMAQ/OPTM adequate?

(b) Is the current documentation for CAMx/PSAT and CAMx/OSAT adequate?

(c) Are the above model formulations sufficiently documented for technical critique and understanding?

(d) Is the discussion and presentation of the above models and their features clear? Please note any specific sections of the documentation that were unclear or confusing.

(e) Is the documentation for each of the above models sufficient to guide a typical user in the use of the model and its preprocessors?

(f) What improvements are desirable for the documentation of each of the above models?

(g) What changes in organization of the documentation of each of the above models are desirable?

(h) Does the documentation contain useful and appropriate evaluations of source apportionment for ozone and $PM_{2.5}$ species compared to other approaches?

3. Evaluation

(a) In general, how manageable do you consider the task of preprocessing/model setup to run source apportionment for CMAQ/PPTM, CMAQ/OPTM, CAMx/OSAT, CAMx/PSAT?

(b) Are postprocessing tools easily available and easy to use for CMAQ/PPTM, CMAQ/OPTM, CAMx/OSAT, CAMx/PSAT?

4. Supplemental Questions

(a) What do you consider the general advantages and disadvantages of zero-out modeling when a 100% emissions perturbation is necessary for an application such as source culpability?

(b) Do you feel that the implementation of mercury tagging in CMAQ/PPTM and CAMx/PSAT has any important weaknesses that may need to be addressed or strengths as a tool for culpability assessment?

"Response to Comments on OPTM and PPTM in *Peer Review of Source Apportionment Tools in CAMx and CMAQ*"

Received from Sharon Douglas on October 30, 2009

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Response to Comments on OPTM and PPTM in “*Peer Review of Source Apportionment Tools in CAMx and CMAQ”*

The document “Peer Review of Source Apportionment Tools in CAMx and CMAQ” critiques the methodologies used in the CAMx model and the CMAQ model for assessing contributions to pollutant concentrations. ICF would like to take this opportunity to respond to some of the comments on the PPTM and OPTM methods implemented in CMAQ.

The review seems predisposed to favor the CAMx methods over the OPTM/PPTM methods in CMAQ, which is perhaps understandable since the former method has been available for some time and is more familiar to the modeling community. This is especially apparent in the lists of strengths and weakness of the two methods. For instance, on p. 9 the reviewers state that “a common weakness of both CAMx/PSAT and CMAQ/PPTM is that they cannot capture any indirect effects, such as an increase in nitrate concentrations resulting from a reduction in SO2 emissions.” The reviewers therefore view this as a common weakness of both methodologies. In the summary of weaknesses, this is listed in item “b” on p. 13 as a specific weakness of the PPTM implementation. Although common to both methodologies, this is not listed as a specific weakness of PSAT.

Below, we provide responses to some of the criticisms of OPTM/PPTM in CMAQ. Reviewer comments are in italics, and our response is in standard type face.

Responses to Selected Comments on CMAQ/OPTM

p. 7: (a) Physical process magnitudes for tracers are inconsistent with corresponding model species. Other methodologies have solved this problem by utilizing process analysis data for component species and aggregating process magnitudes for the tagged tracer species. These data already exist in the models.

The approach appears to have been misunderstood. The OPTM methodology is very much consistent with the CMAQ processes. While using process analysis may be one approach to getting some of the information that is required to track the species through the model processes, OPTM does this explicitly for each process. The OPTM approach maintains consistency between the overall modeled NOx, VOC, and oxidant with the total tagged NOx, VOC and oxidant at each process step.

p. 7: (b) Calculation of total oxidant (Ox) is incorrect. When two Ox species combine, as with NO3 (NO2+O) and N2O5 (NO2+NO3), the species should represent multiple units of Ox based on the number of “odd oxygen,” not the nitrogen.

We agree that the reviewer was correct. This issue has subsequently been fixed and our tests showed that it had minimal effects on the results.

p. 7 – 8: (c) Based on the documentation, ratios of initial precursors are used to apportion net chemical change of tagged VOCs, NOx, and Ox. There are several false assumptions in the code that, if confirmed, should be addressed:

(1) Change in tagged species is solved algebraically, which inappropriately conserves initial tagged species. A differential solution would apply losses to the existing

distribution of tags and to the distribution of chemically produced tags.

The reviewer is incorrect in his or her assumption of how the tagging is implemented. Losses of NOx and VOC are attributed to the precursor tags in each process. That includes the gas phase chemistry.

(2) Change in each region's NOx and VOCs is proportional to the total local NOx and VOCs. This assumes that each region's NOx and VOC reactivity (or deposition velocity) profile is equal to that of the local NOx and VOCs. In plumes or near sources, the locally contributed NOx and VOCs are likely to have a different reactivity profile.

Although the OPTM implementation does not make any explicit differentiation between VOCs of differing reactivity, the calculation of chemical changes made by the chemical mechanism would still respond to any overall changes in VOC reactivity. In addition, the assumption that the various tagged sources or categories have roughly similar reactivity is reasonably accurate, for typical situations. The current implementation of OPTM allows it to handle many tagged sources (either regions or categories of sources) simultaneously. The situations where the assumption of similar reactivity could result in inaccuracies in the OPTM attribution are unusual, such as tagging of a single source that emitted highly reactive compounds. Addition of more detailed tracers to the OPTM implementation could improve the accuracy of the representation of these unusual cases. The cost would be a dramatic increase in the number of tag species required in the model.

The reasoning of the above response is based consideration of differences in VOC reactivity. The question, however, brings up an example of a plume which is more a case of low hydrocarbon to NOx ratio than a VOC reactivity issue. This is more the province of a plume-in-grid treatment, and, at the resolution of modeling grids currently in use, neither the model nor OPTM can represent the very low hydrocarbon to NOx ratio found in plumes.

(3) NOx production is proportional to existing NOx. It assumes that NOx precursors (i.e., HNO3, PAN, or NTR) are proportional to local NOx concentrations. These longerlived species may be transported and so not reflect local emissions of NOx.

This is similar to the comment regarding reactivity. Additional tags could keep track of other species besides NOx and VOC. We're assuming, however, that for the most part NOx and VOC gradually decay via chemistry and deposition, for example. There can be some short term production of NOx from species such as PAN, but to keep number of tags to a manageable level, we've assumed that this production will be small compared to fresh emissions. This could be tracked using additional tags, but it would also require additions to the code to track the changes during chemistry and additional complexity.

(4) Total oxidant chemistry is apportioned based on precursor concentration distributions. While Ox production is likely to be proportional to precursors (excluding situationsnoted in [1] and [3] above), Ox loss reactions should be apportioned based on current Ox distribution.

Actually OPTM does apportion Ox loss based on current Ox distribution. So again this appears to be a misunderstanding. Although the changes are not tracked separately for production and destruction terms through the chemistry (again, additional tags and complexity could do that), when the net change in oxidant is negative, the change is apportioned to the tags according to the proportions of oxidant tags, not according to the precursor tags.

p. 8: (d) Artificial "offset" to Ox potentially alters processes and requires further review of code.

The offset is only included directly in the oxidant tracers for the advection step – and really only to overcome a limitation of the advection scheme. Other processes that depend directly on the magnitude of the species, such as deposition, are apportioned to the tags based on the change in the model species. The same change is therefore attributed to the tag regardless of whether there is an offset or not.

p. 9: (a) Mass conservation of the tagged species is not checked in the formulation.

For CMAQ/OPTM, mass is allocated to the oxidant, NOx, and VOC tracers at each time step and the sums of all the oxidant, NOx, and VOC tracers are required to be the same as the overall oxidant, NOx, and VOC concentrations. Therefore, mass consistency and consistency of the tracer species and the standard species is ensured.

p. 9: (b) OPTM does not distinguish between NOx-limited and VOC-limited regimes. OPTM also assumes that all VOC species have the same reactivity. Therefore, ozone may be attributed to the wrong source.

See our response above.

p. 9: (c) Assumption of the fractional change in the total oxidant (sum of NO2, NO3, N2O5, and O3) concentration due to dry deposition is questionable.

Dry deposition is a linear process and the changes in separate species add algebraically.

p. 9: (d) The contribution to O3 by tagged NOx and VOCs is counted separately, which is somewhat misleading because O3 is formed through photochemical reactions of both NOx and VOCs jointly.

The fact that ozone is the result of the interaction of VOC and NOx is why OPTM attributes ozone formed to both VOC and NOx, not to one or the other as some other methods do.

p.9: (i) Current OPTM setup requires the code to be recompiled for each new set of tags. This approach places an unnecessary burden on model users and increases the chances of making mistakes.

The set_tag program makes the changes automatically and it has been fixed to avoid the problems encountered by reviewers during testing. OPTM/PPTM implementation in CMAQ followed the existing structure of the code which requires modifying include files to add species (even inert tracer species). Although many of these include files are actually unnecessary in CMAQ (since the information could be generated within the code), we did not wish to restructure the code which would make it more difficult to move the OPTM/PPTM implementation to later versions of CMAQ.

Responses to Selected Comments on CMAQ/PPTM

p. 13: (a) Mass conservation of the tagged species is not checked in the formulation.

In the implementation of CMAQ/PPTM, the overall species are not simulated concurrently with the tagged species. The mass consistency of the implementation is therefore dependent on the mass consistency of the CMAQ algorithms that handle the various processes. A separate CMAQ run without tagging can be compared to a CMAQ PPTM run to assess the uncertainties in the simulations due to strictly numerical effects.

(b) PPTM assumes that all secondary PM is linked to a specific precursor: for example, sulfate to SOx, nitrate to NOx, ammonium to NH3, and SOA to VOC emissions. For this reason, PPTM cannot capture any indirect effects, such as an increase in nitrate concentrations due to a reduction in SO2 emissions.

This is an issue comment to both methodologies, but is only listed as a weakness for PPTM.

p. 13: (c) Sulfur tagging in aerosol lacks methods to account for individual chemical production pathways that are the main causes of nonlinearity in the sulfate production.

PPTM accounts for the net change in aerosol sulfur due to the several separate processes in CMAQ. It assumes, of course, that each of these processes acts on aerosol sulfate in proportion to the separate tag concentrations.

Why is this issue not also listed for PSAT, which takes the overall change in sulfate due to each major process and does some apportionment?

*p. 13: (f) Current CMAQ/PPTM design requires the CMAQ source code to be modified and recompiled for each set of PPTM simulations. Even though the "set_tag" utility program is provided to create the *.EXT files from templates, the PPTM approach not only increases the complexity of the source code (multiple versions of executables), it also increases the chances of making mistakes.*

See response to OPTM comment "j" above.

p. 13: (g) Current PPTM approach also requires emissions tracers to be added to CMAQ emissions inputs. Again, this intrusive approach to emissions processing requires extra work and is prone to processing/QA errors.

In order to correctly identify emissions by source category or by political boundary, it is necessary to prepare tags in the emissions step. A simple grid cell masking technique can also be used with PPTM, but that cannot differentiate source categories, nor can it correctly identify emissions from different states or counties since grid cells can overlap several of these entities.

(h) PPTM outputs are written into standard CMAQ output files. This would lead to large output CMAQ files, increasing the potential for file corruption and access problems. A

better approach is to create additional output files specifically for CMAQ/PPTM results.

Files are large, but, in all but the most extreme cases, no special handling is required except the use of an IOAPI version compiled for a larger number of variables.

ENVIRON

October 15, 2009

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Re: Peer review of Source Apportionment Tools in CAMx and CMAQ

Dear Dr. Baker:

The CAMx development team at ENVIRON appreciates this opportunity to provide comments on the draft report "Peer review of Source Apportionment Tools in CAMx and CMAQ" prepared for EPA. We hope that our comments will improve a report that is already a useful contribution to the photochemical modeling community.

Clarifications

- Mass conservation between tracers and model species is checked by PSAT (comment on page 10). PSAT conserves mass by (a) employing algorithms that inherently keep tracer mass consistent with their corresponding model species (e.g., sum of PSO4 tracers consistent with model species PSO4), and (b) enforcing mass consistency once per time step to prevent numerical noise from causing drift in mass conservation over time.

- OSAT and APCA can be run on multiple processors using both MPI and OMP (comment on page 11). The MPI capability was introduced with CAMx version 5 which was the first CAMx release with MPI. The reviewers evaluated version 4.5 which had OMP but not MPI.

- CAMx can perform source apportionment for ozone and PM simultaneously (comment on page 24). This is accomplished by running PSAT and choosing O3 as one of the constituents to be apportioned. The resulting ozone apportionment will be identical to running OSAT.

- On page 24, it was theorized that tagged species methods could not distinguish the reactivity difference between two VOC sources (called A and B) with differing VOC composition when the sources are located in the same grid cell. OSAT and PSAT can distinguish the reactivity of these sources if you specify them as separate source categories for apportionment (rather than combining them).

Points of Discussion

- The comment on Page 10 that "*it would be reasonable to select the tagged species to represent only small perturbations in the inputs to compare the counter-species tools reviewed here with those from the brute-force zero-out simulations*" bears discussion. A brute-force simulation that introduces only a small perturbation closely approximates a first-order sensitivity and, therefore, is least likely to be comparable to a tagged-species source apportionment. Tagged-species source apportionments tend to be more similar to brute force "zero-out" perturbations. This concept is discussed and illustrated using simulation results by Koo et al. (2009).1

- The comment on page 11 that no negative production is allowed by PSAT is not strictly correct but merits discussion. In cases where source apportioned species are subject to chemical destruction (e.g., evaporation of PM nitrate, photochemical destruction of ozone) such negative production is explicitly applied to the corresponding PSAT/OSAT tracers. However, OSAT/PSAT tracers never go negative because they are intended to provide mass tracking and negative mass contributions would be non-physical. In this regard, tagged-species methods stand in contrast to sensitivity methods, such as the DDM, where negative sensitivities can occur and are physically meaningful. See Koo et al. (2009) for further discussion.

- The reviewers comment that extra effort is required to prepare the emission inventory of reactive tracers for OSAT and PSAT (page 11). The amount of effort required is frequently minimal because suitable "pre-merged" emission files are available. For example, with separate files for biogenic, mobile, elevated point and other area sources (these files generally are available) it is possible to obtain source apportionment to four major inventory sectors. Extra effort can be minimized if source apportionment analyses are considered when setting up emissions processing.

- Sensitivity of source apportionment results to grid resolution was discussed on page 25. Since grid model results can be sensitive to grid resolution it must be anticipated that source apportionment results will also be sensitive to grid resolution. We expect that any differences in source apportionments at different grid resolutions will be a consequence of changes in the grid model result rather than OSAT or PSAT because there is nothing in the OSAT/PSAT formulation that depends directly on grid resolution.

- Approaches to apportioning pollutant production in nonlinear chemistry processes are discussed on page 25. Oxidation of SO2 to sulfate (SO4) provide a clear example for discussion. The rate of SO2 oxidation may be non-linear across a model time-step, but this nonlinearity does not enter into the apportionment of the SO4 produced. Because all SO2 molecules react with oxidants at the same rate, the SO4 produced inherits its source

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1 Koo, B., G. M. Wilson, R. E. Morris, A. M. Dunker and G. Yarwood. 2009. "Comparison of Source Apportionment and Sensitivity Analysis in a Particulate Matter Air Quality Model." Environ. Sci. Technol., 43 (17), pp 6669-6675. DOI: 10.1021/es9008129

apportionment from the SO2 that reacted. Therefore, the PSAT approach wherein "*the change in overall mass (or flux) is split proportionally to the components (tags)*" is accurate, mass conservative, and efficient. PSAT relies upon the fact that the SO4 source apportionment behaves linearly (proportionately) without assuming that the underlying chemical reactions of SO2 are linear. The same methodology applies strictly for ammonium and secondary organic aerosol but less strictly for nitrate and ozone where families of precursors are tracked by a single tracer (e.g., VOC precursors to ozone are tracked by a single reactivity weighted tracer).

Potential Modifications

Some potential enhancements to OSAT/PSAT were suggested that could be implemented:

- Gridded output of average tracer concentrations currently is provided for the surface layer only. CAMx could be modified to (optionally) output this information for layers aloft at the cost of greater memory (RAM) and disk space requirements. However, we note that tracer concentrations for layers aloft can be obtained now via the OSAT/PSAT receptor file by specifying an aloft receptor.

- OSAT distinguishes when ozone production is VOC or NOx-limited based on the production ratio H_2O_2/HNO_3 with a threshold of 0.35. Suggested alternatives were using a threshold range (e.g., 0.3 to 0.5) and using several different indicator ratios. Since there is no clear rationale for how to implement multiple criteria such as these, testing and evaluation would be required. Early on with OSAT we tested sensitivity to changing the H_2O_2/HNO_3 threshold and found little sensitivity to choosing values between 0.15 and 0.5 because the H_2O_2/HNO_3 ratio changes rapidly within this range, i.e., the chemistry moves quickly through the threshold. Therefore, we retained the originally proposed threshold of 0.35.

- Several constructive comments on the CAMx User's Guide will be considered in the next update.

Sincerely

Greg Yarwood
Principal

cc: Ralph Morris
Gary Wilson
Chris Emery
Bonyoung Koo