3 December 2014

109 T.W. Alexander Drive
Research Triangle Park, NC 27711

Natascha Töpfer
Editorial Support
Geoscientific Model Development

Dear Natascha Töpfer,

I am enclosing the revised manuscript for consideration for publication in *Geoscientific Model Development.* My co-authors and I would like to thank the reviewer and the editorial staff for time and effort in evaluating the suitability of our work for publication in this journal and improving its quality.

I believe that we have addressed all concerns raised by the reviewer, enabling us to prepare a much better manuscript, which we hope you will find suitable for publication. In the attached pages, you will find specifics on how we addressed each reviewer comment along with the relevant text that was changed as a result. We also made other minor editorial changes to the manuscript in order to increase clarity in the presentation.
Thank you for your time.

Sincerely,

Sergey L. Napelenok, Ph.D.
U.S. EPA

Anonymous Referee #1

Summary: The authors have implemented and applied ozone "source apportionment" in the Community Multi-scale Air Quality model (CMAQ). Implementation is a significant effort that provides important capabilities to CMAQ that are relevant to scientific exploration and regulatory application. Application and evaluation showed results that would be expected from implementation in similar models.

The paper would be improved by addressing three things. 1) The weaknesses inherent sensitivity metric application, particularly with respect to biogenic VOCs. 2) Discussion different ozone endpoint for results. 3) Minor comments.

We appreciate the suggestions from the reviewer on how to improve the presentation of our work. Below, we outline how we addressed each of the three areas suggested.

1 Sensitivity Metrics:

The source apportionment technique relies on the PH2O2/PHNO3 indicator that has been extensively used for Carbon Bond IV. The PH2O2/PHNO3 ratio, often referred to as the Sillman ratio, is a simplification of a more complex relationship (Sillman, 1995, A5 (PH2O2+PROOH)/PHNO3). The Carbon Bond IV mechanism lacks an organic peroxide (ROOH), which requires the simplification of based on the ratio of PROOH/PH2O2 in a 1995 Lack Michigan simulation. Though Dennis and Tonnesen showed its robustness, I am not aware of a published comparison to the robustness of (PH2O2+PROOH)/PHNO3. The poor performance of biogenic zero out could be related to ROOH production. The speciation of VOCs are important in determining the PH2O2/PROOH ratio in the presence/absence of NOx. The PH2O2/PROOH dependence could suggest that biogenic VOC sensitivity would be mischaracterized by PH2O2/PHNO3. Though the explanation of non-linearity is suggestive, it is not definitive. In fact, the first order sensitivity shows better performance for BIOG for both the slope and correlation.

At least some discussion of the metric is warranted, as well as more discussion of the fundamental issues with applying a binary metric.

It is correct that the apportionment technique relies on the PH2O2/PHNO3 indicator ratio in the current implementation. However, we had spent a substantial amount of time prior to settling on this ratio testing and comparing nine other ozone regime indicators including (PH2O2+PROOH)/PHNO3. At one point, the code even offered users a choice for selecting the indicator they wanted or an ensemble estimated from all of them. However, we moved away from this option as it offered little change in the performance of the attribution estimates and offered little additional information in the results. Specifically, the underestimation of biogenic VOC attribution was evident in the simulations using each ratio and was identical in the results using the Sillman ratio with and without the ROOH production term.

To clarify this point in the manuscript, we have added the following text to section 3.1:

> Ozone production is attributed to either VOC or NO_x emissions sources based on the ozone chemical formation regime that is estimated using the $PH_2O_2/PHNO_3$ indicator ratio, similar to the implementation in CAMx. Other indicator ratios were also explored including $H_2O_2/(O_3+NO_2)$, O_3/NO_Z, $HCHO/NO_Y$, and $(PH_2O_2+PROOH)/PHNO_3$, but these offered no additional gain in model performance.

The concern that biogenic VOC attribution is underestimated is certainly troublesome and we are currently investigating the issue in more detail. The nonlinearity in the interaction is the most likely cause, because performance of 2nd order DDM estimate is substantially better than 1st order DDM estimate. The fact 1st order DDM performance is better than ISAM for this metric is due to different model formulation and does not really offer any explanation for poor ISAM biogenic attribution.

2 Ozone Endpoint:

The authors show only daytime-average for most results and all-hour averages for ISAM/DDM. I suspect that the choice to average was based on the autocorrelation of hourly results, but this is not discussed. Averaging, however, removes variability. An alternative metric that would be more relevant, would be maximum daily 1-hour average or maximum daily 8-hour average. The 8-hour average would still have some reduction in variability, but would be more relevant to the regulatory application that is likely to use this tool. Thus, the brute-force evaluation of 1- or 8-hour average would be more interesting. In addition, why do some results use all-hour and others daytime only?

The choice for showing the results for daytime hours for ozone was made keeping in mind the typical use of the tool. Inline apportionment tools, like ISAM, by design, do not account for sensitivity to ozone titration, which often occurs outside of daylight hours. This is actually evident if figure 5 (zero-out comparison with DDM and ISAM), where ISAM is never less than zero, while DDM shows negative sensitivities that match with zero-out. Choosing all hours or daytime hours for other species depended on the goal of specific figures and metrics – either diagnostic or demonstrative.

The results are generally not further averaged. All the statistics and figures show all of the hours provided in the model output (aside from excluding nighttime hours for some species), so no further variability is lost. We had considered evaluating 1-hr max and 8-hr max ozone quantities, but decided against it due to the loss in variability that the reviewer notes.

To clarify these decision in the manuscript, the following text was added at first mention of excluding nighttime hours in section 4.1:

> The CMAQ model performed well this day with an O_3 bias of 4.7ppb and average error of 9ppb across the domain during daytime hours. Nighttime hours were excluded from ozone analysis, because ISAM was not designed to predict titration events that often occur at night in areas of large NO_x emissions.

3 Minor comments:

Page 5802, line 23-24 - add some more detail than just a hanging line.

We have added the following text to further describe the deposition processes:

> ISAM apportions CMAQ-calculated wet and dry deposition O3, NOx, and all VOC species into individual sources as done in the previous PM2.5 ISAM code. Both of these processes follow simple linear algorithms where the mass removed is a function of existing mass and a removal coefficient and thus did not require additional consideration for the ozone implementation.

*Page 5803, line 5 - is J = J*something?*

We appreciate the reviewer noticing the awkward phrasing and have changed the text to state:

> … **J** the Jacobian matrix calculated based on the average of bulk concentrations before and after any gas-phase solver for CB05 model species $\left(\frac{[C_{S,bulk}^{new}]+[C_{S,bulk}^{old}]}{2}\right)$ …

Page 5803, line 11 - The effect on accuracy will likely depend on the time of day, as sensitivity typically shifts with time. During the PH2O2/PHNO3 transition, this could be important if the synchronization time step is not controlled. Some discussion is warranted.

This implementation is similar to what is currently used in implementation of DDM in both CMAQ and CAMx. It was demonstrated that there is minimal loss of accuracy from solving during the middle of model synchronization time step during all hours including regime transitions. This was noted in more detail in the text and reference was provide for further information to the reader:

> The solution is obtained only once for every model synchronization time step Δt instead of incremental chemical time steps to increase computational efficiency at little expense to accuracy as was previously shown in the original implementation of the CMAQ decoupled direct method (DDM) (Hakami, 2004) .

Page 5803, 5804 - consider harmonizing subscripts in equation 7 with 8 and 9.

The species subscript for VOCs was changed in equation 7 to *s* in order to match equations 8 and 9.

Page 5812, line 2-5 - reword.

The awkward phrasing was changed to the following:

In general, correlations between ISAM/zero-out estimates are high for both ambient concentration and deposition of O_3 for the major emissions sectors. ISAM estimates of NO_x are higher than nitrogen-out in most sectors except boundary condition. And ISAM VOC estimates are similar when compared to VOC-out in most sectors except EGU and MARINE.

Page 5812, line 11 - consider splitting this paragraph to help distinguish between what I believe are very distinct points.

A paragraph break was added to better distinguish the discussion between the two points.

Anonymous Referee #2

GENERAL COMMENTS

This paper describes the implementation of the O3-VOC-NOx version of the Integrated Source Apportionment Method (ISAM) in the Community Multiscale Air Quality (CMAQ) model and then shows results from an example application and comparisons with results from a source-sensitivity method ("brute-force" method). This paper is a companion paper to an earlier paper by three of the authors on the implementation of the PM2.5 version of ISAM in CMAQ. The O3-VOC-NOx version of ISAM is similar in some respects to the Ozone Source Apportionment Technology (OSAT) method previously implemented in the CAMx air quality model, but the ISAM method is more detailed, particularly for VOC species, and it can be applied to deposition as well as to ambient concentrations.

This is a well-written paper that describes a significant new analysis method for air quality modeling. The method itself is clearly described as is the relationship of this source-apportionment method to other source-apportionment and source-sensitivity methods. The illustrative example that is provided shows the power and usefulness of the method, and the similarities and differences between the ISAM results and the brute-force results appear to be reasonable.

I have made a number of specific comments and suggestions below related to clarity and completeness that I would ask the authors to consider. I have also included a number of editorial comments and corrections.

We appreciate the support from the reviewer of our work and for the thoughtful and detailed suggests for how we can improve its presentation in the manuscript. Below, we outline how we addressed each comment.

SPECIFIC COMMENTS

1. The authors should consider expanding the title slightly to "Photochemical grid model implementation and application of VOC, NOx, and O3 source apportionment" (cf. line 9 of Abstract and Kwok et al. (2013) title).

The title was changed as the reviewer suggested. We hope that it still fits with the parameters of the journal short title formating requirements.

2. Line 14 (p. 5801) of Section 3.1 mentions lateral boundary conditions and initial conditions. Are contributions from the upper boundary also considered (this question was motivated in part by Supplemental Fig. S1j and lines 12-13 on p. 5807)?

Only lateral boundary conditions are tracked simply because standard CMAQ currently considers upper boundary as zero. However, flexibility exists in the code to track non-zero values from the upper boundaries. Boundary influence is typically high in areas of elevated terrain, because often these areas are impacted by long range transport such as from lateral boundary conditions.

3. Lines 23-24 on page 5802 of Section 3.1 provide a very brief mention of dry and wet deposition, although a number of figures in the Supplement show deposition results. It would help the reader to interpret these figures if it were stated in Section 3.1 which CB05 species dry deposit and which CB05 species wet deposit. For example, Supplement Fig. 7 shows O3 total deposition results (dry+wet). Does CMAQ consider O3 wet deposition? And Supplement Fig. 12 shows non-zero total deposition for only four VOC species; is this due to dry deposition only or to wet deposition only or to both, and are the other VOC species assumed not to dry deposit or wet deposit?

All of the tracked species including O3, NOx, and VOCs both dry and wet deposit in the base CMAQ model and therefore are tracked by ISAM through these processes. Only four species are shown in the supplemental figures, because their behavior and performance is the same as this is a highly linear process. A similar question was raised by Reviewer #1 and the text in Section 3.1 was changed to add more detail about this process:

> ISAM apportions CMAQ-calculated wet and dry deposition of O3, NOx, and all VOC species into individual sources as done in the previous PM2.5 ISAM code. Both of these processes follow simple linear algorithms where the mass removed is a function of existing mass and a removal coefficient and thus did not require additional consideration for the ozone implementation.

4. I was surprised that Section 3 did not include any discussion of the computational costs of ISAM-O3, including the actual and percentage increase in the number of tracer fields and the increase in simulation time. This is very relevant information to a user.

The reviewer is correct that CPU cost is definitely pertinent and useful information for the readers. We have added the following text at the end of Section 3 in order to describe CPU usage:

> 3.3 Computational Efficiency
>
> The additional computational burden of CMAQ-ISAM was also analyzed over the test domain. Additional time required to execute the model over the base simulation varied with the number of tagged parameters and also with the type of parameter with ozone tags requiring more CPU time than secondary PM tags, which, in turn, required more CPU time than primary PM time. CPU usage for ozone tags was approximately equal to base mode CPU time multiplied by the number of tracked tags. For example tracking ozone from emissions from five different geographical regions would require a CPU time investment of approximately five times that of the base model simulation. Work is currently underway to improve computational efficiency of CMAQ-ISAM.

5. There are a few details that could be added to the beginning of Section 4: Which CMAQ version was used? What was the model top and what were the elevations of the lowest few model levels? Which WRF version was used? What was the WRF horizontal grid spacing? Were the CMAQ and WRF grid map projections the same? What were the GEOS-Chem grid characteristics (e.g., horizontal grid spacing, number of vertical levels, model top)? Which gas-phase chemistry mechanism did GEOS-Chem use, and if VOC species mapping was required, what was it?

The reviewer is correct to point out the version number of CMAQ (version 5.0.2) should be included in the methods section. The methods section has been updated to include the version of CMAQ that ISAM was implemented and used for this application.

> The ISAM for O3 has been implemented in the CMAQ version 5.0.2 model, which was developed by the United States Environmental Protection Agency (EPA) and is used by EPA, other regulatory agencies, and academic institutions to characterize local to continental scale ozone formation and transport (Byun and Schere, 2006; Foley et al., 2010).

Additional information about the inputs to the CMAQ application used to illustrate the functionality of source apportionment has been added to the Supporting Information for readers interested in this detail. We provided additional information about the WRF simulation used to supply meteorology to CMAQ and the GEOS-CHEM simulation used to supply boundary conditions to the coarse 36 km CMAQ simulation that was then used to supply boundary conditions to the 12 km simulation used in this analysis. This information was added to the Supporting Information due to the current length of the manuscript and because choices made for WRF and GEOS-CHEM would not impact the comparison between CMAQ estimates with source apportionment and brute-force sensitivity. The following section has been added to the Supporting Information.

The CMAQ model was applied from June 28 to July 5, 2007 for a domain covering the State of California (CA) using 12 km sized grid cells (79 columns and 106 rows) and 24 vertical layers extending to the surface (layer 1 height ~20 meters) to the model top of 50 mb. Meteorological inputs to CMAQ were generated using the Weather Research and Forecasting model (WRF), Advanced Research WRF core (ARW) model version 3.1 (Skamarock et al., 2008). The WRF domain uses the exact same grid projection (Lambert Conformal), grid origins, and datum as CMAQ. However, the WRF 12 km domain covers the continental U.S. so data used for the CMAQ simulation is a sub-set of the WRF simulation. Additionally, WRF used 34 vertical layers to represent the atmosphere to 50 mb so some layer collapsing was done when generating CMAQ inputs. Layers are matched one-to-one nearest the surface to best represent diurnal changes in the height of the boundary layer. Selected physics options, input analysis, grid structure, and evaluation are provided in detail elsewhere (U.S. Environmental Protection Agency, 2011).

The 12 km model domain was nested in a 36 km continental domain, and boundary inflow to the 36 km domain were based on spatially and temporally variant concentration data from a 2007 year-specific annual GEOS-Chem version 8-03-02 simulation (Yantosca, 2004). The GEOS-CHEM simulation had a grid resolution of 2.0 by 2.5 degrees using a latitude-longitude grid projection. The troposphere and stratosphere were vertically resolved with 47 vertical layers. GEOS-CHEM output was used to generate space and time (3-hourly) variant boundary conditions and initial conditions for CMAQ. Evaluation included comparison to model performance plots generated using a similar version of the model also applied for 2007 (Lam et al, 2010). Grid and chemical species translation of GEOS-CHEM to CMAQ was based on methods described in Henderson et al. (2014).

6. The second paragraph of Section 4 describes the 11 tracer sectors that were considered in this example study. Figure 1 then shows spatial tiles for nine of these sectors for one time. For completeness would the authors consider expanding this figure by adding one more row of three tiles to show the OTHR, ICON, and bulk O3 fields?
7. Table 2 shows that the OTHR sector contributes 15.8

We assume that comment #7 is just a continuation of comment #6. Figure 2 currently shows contributions from all tracked sectors and initial conditions as well as the total bulk ozone concentration. There, it is evident that ICON contribution is irrelevant after 1-3 days of simulation. The OTHR sector does contribute 15.8% of the VOC emissions in the domain and its contributions is also a similar fraction of the bulk O3 concentrations. There may be some interest for the reader to see bulk O3 fields alongside the sector contributions, but we feel that including all these extra panels will detract from the main purpose of the figures in showing the important sectors.

8. I did not understand Supplement Figure S5 and how it relates to Figure 2. Does Figure S5 show time series for 6 different sites in the Riverside area plus time series for 6 different sites in the Sacramento area (the caption is unclear)? What is the spatial size of these subnetworks? Does Figure 2 show an average time series over multiple sites or a time series for an individual

site, and if the latter, are these different sites from those shown in Figure S5? Some expanded discussion in Section 4.1 could address these questions.

We agree with the reviewer that the presentation of these figures is confusing. The reviewer is correct that the supplemental figures show the 6 different sites that exist in each metropolitan area and that Figure 2 is showing the average of the 6 for each area. The spatial extant is simply the MSA for each location. Section 4.1 and the caption for Figure S5 were changed as follows to provide more clarity:

> This is demonstrated for two O3 monitor siteslocations, at Riverside in Figure 2-a and Sacramento in Figure 2-b, where monitor data are displayed with the model simulated ozone and ISAM-attributed data. Each location had data availability at six separate monitoring sites and Figure 2 shows the average measured and modeled quantities. Individual monitor results are shown in Supp Fig 5.
>
> Supp Figure 5: Hourly time series of O3 observations (crosses) at sites of Californian Air Resources Board monitoring network; and the corresponding CMAQ-ISAM sector breakdowns (stacking colored bars). Locations are six distinct Riverside sites (top six panels) and six distinct Sacramento sites (bottom six panels)….

9. Section 3.1 describes an "NOy" family of nine CB05 nitrogen species. In the "NOx" portion of Section 4.1, I was confused as to whether what is being analyzed in Figures 6 and 7 is NOx = NO + NO2 or NOy. If the former, then does the NOx deposition considered in Supplement Figures S8 and S9 only correspond to NO2 dry deposition or do any of the results presented in the manuscript relate to NOz species? I also noted that the maximum daily O3 total deposition is about 80 g/ha (Figure S7), that the maximum daily VOC total deposition is about 20 g/ha (Figure S11), but that the maximum daily NOx total deposition is only about 1 g/ha (Figure S9) – is this reasonable?

Section 3.1 presents the gaseous nitrogen species included in source apportionment tracking. Any discussion, Figures, or Tables that follow in the text only include a subset of these species as noted in the text or Figure caption. Figures 6 and 7 include NOx species NO and NO_2. Additional text has been added to the captions to be clear about which species constitute NOx in these Figures. Supplemental Figures 8 and 9 also include NO and NO_2 to represent NOx. Again, text has been added to these Figure captions to be clear about which species were summed to represent NOx. Unfortunately we do not have ideal constraints on total (wet and dry) deposition through measurements to provide an indication about whether these daily total values are reasonable. However, for the purposes of this comparative source apportionment approach study the similarity in estimates using multiple approaches is emphasized rather than how well model estimates reflect reality.

TECHNICAL AND TYPOGRAPHICAL CORRECTIONS

p. 5792, l. 4 Perhaps "Source-based apportionment techniques" (as a general comment on the manuscript, compound adjectives are sometimes hyphenated but more often they are not, and this is done inconsistently; consider the many uses of both "zero out" and "zero-out")

The manuscript was edited for consistency in hyphenating compound adjectives as suggested.

p. 5792, l. 8 Perhaps "track source emissions through ..."

The sentence was change to read:

> Photochemical model source apportionment has been used to estimate track source impacts of specific sources, groups of sources (sectors), sources in specific geographic areas, and stratospheric and lateral boundary inflow on O3.

p. 5792, l. 25 "... are either to be accounted for pollutant levels in a given scenario" – awkward phrase; same comment for p. 5811, l. 23

The first text location was changed to read:

> Low correlation coefficients occur for chemical regimes that have strong non-linearity in O3 sensitivity, which demonstrates different functionalities between source apportionment and zero-out approaches. Where appropriate use depends on whether source attribution or source sensitivity is desired.

The second was changed to:

> The choice of appropriate methodology depends on whether source sector/region attribution or sensitivity is of interest.

Hopefully, this clarifies the message we were trying to convey.

p. 5793, l. 28 Perhaps "... Zhang et al., 2009) and contributions from specific ..."

Edited as suggested.

p. 5794, l. 7 "... compared directly with source sensitivity apportionment approaches"?

Edited as suggested.

p. 5797, l. 4-6 Instead of N in all 3 equations, perhaps NO3, NV OC, and NNOx

We prefer for the notation to distinguish the sources of the species (O3, VOC, NOx) more explicitly. However, for better presentation, we moved *n* into the subscript:

$$C_{bulk,O3}^{i,j,k} = \sum_{n=1}^{N} C_{O3_n}^{i,j,k}$$

p. 5797, l. 20 "emissions sources that contributed"

Edited as suggested.

p. 5801, l. 13 "depostion"

Corrected.

p. 5802, l. 14, 18 Could mention the number of VOC and NOy tracers (i.e., "14 CB05 VOC species", "nine nitrogen compounds in CB05")

Edited as suggested.

p. 5803, l. 5 "is the Jacobian matrix"

Edited as suggested by both reviewers.

p. 5803, l. 14 The use of P to denote "production" was introduced on p. 5795 (l. 25), but this convention is not followed in this line or in a few other places (e.g., p. 5803, l. 24; p. 5804, l. 4)

The letter P to denote production of the species in questions is included consistently in the text.

p. 5804, l. 6 "Milford"

Corrected

p. 5804, l. 13-14 Perhaps "the total PO3 and DO3 terms"

Edited as suggested.

p. 5805, l. 7 Perhaps "Following the ozone production apportionment, subsequent apportionment ..."

Edited as suggested.

p. 5806, l. 1 Perhaps "... from a 2007 year-specific annual GEOS-Chem"

Edited as suggested.

p. 5806, l. 12 Perhaps "contributed" rather than "taken up"

Edited as suggested.

p. 5806, l. 13 Perhaps "... the leading NOx emissions sectors by mass were ..."

Edited as suggested.

p. 5806, l. 21 "at 16:00 LT"

Edited as suggested.

p. 5807, l. 20 "Together with Supplement Fig. S5, which shows ..., Fig. 2 demonstrates"

Changed the text to read:

> Results presented in Supplement Fig 5 and Figure 2 demonstrate the base model's ability to capture the temporal and regional variability in observed ozone.

p. 5807, l. 24 Perhaps "... was used to provide an alternate estimate of source ..."

Edited as suggested.

p. 5809, l. 2 Perhaps "... shows notable inflow of O3 from the boundaries, as shown more clearly in the stacked bar charts in Fig. 4"

Edited as suggested.

p. 5809, l. 14 Perhaps "contributes" rather than "attributes"

We kept the original wording here, because we believe we consider ISAM a tool that "attributes" amounts of "contributions."

p. 5811, l. 5, 9 Insert references to Supplement Figs. S13a,c and S13b,d as well; also, since the discussion refers to the BIOG, BCON, and ONRD sectors, you could swap Figs. S13 and S14 to maintain the same order.

Edited as suggested.

p. 5811, l. 24-25 Perhaps "Implementation of O3 tracking capability in CMAQ-ISAM for the CB05 gas-phase mechanism adopts ..."

Edited as suggested.

p. 5812, l. 14 Should be "complements"

Corrected.

p. 5813-5818 Missing references:
** Chameides et al. (1988)*
** Finlayson-Pitts and Pitts (1986) [tangled up with first Foley et al. entry]*
** Henderson et al. (2011)*

** Pfister et al. (2013)*
** Lu et al. (1998)*
** Milford et al. (1994)*
** Sillman et al. (2000)*
** Tonnesen (1999)*
** Tonnesen and Dennis (2000a,b)*

References Added

p. 5813-5818 References not cited in text:

** Andreani-Aksoyoglu et al. (2002)*
** Haagen-Smit and Fox (1954)*
** Kleinman et al. (1994)*

** Baker and Foley (2011)*
** Choi et al. (2014)*
** Guenther et al. (2006)*
** U.S. Environmental Protection Agency (2011a)*

The first 3 are used in the text are were kept; the remaining were removed.

p. 5813, l. 18 "Jama-J. Am."
p. 5813, l. 20 "Chameldes"
p. 5817, l. 23 "Naitonal"
p 2 of Supplement "June 10"

Corrected

Figures

** The captions for Figures 3, 6, and 8 do not give the averaging period; they could also indicate that the data points correspond to all 8374 surface grid cells.*

The captions were changed to read as follows:

> Figure 3. ISAM/both-out O3 comparison for each sector during daytime hours. The sectors are: (a) biogenic BIOG, (b) wild fires FIRE, (c) non-electricity generation units Non-EGU, (d) non-road mobile NNRD, (e) on-road mobile ONRD, (f) other point sources MEX, (g) electricity generation units EGU, (h) marine MARINE, and (j) boundary conditions BCON. Note different scales across the panels.
>
> Figure 6. ISAM/N-out NOx comparison for each sector and all simulated hours. The sectors are: (a) biogenic BIOG, (b) wild fires FIRE, (c) non-electricity generation units Non-EGU, (d) non-road mobile NNRD, (e) on-road mobile ONRD, (f) other point

sources MEX, (g) electricity generation units EGU, (h) marine MARINE, and (j) boundary conditions BCON. Note different scales across the panels.

Figure 8. ISAM/V-out VOC scatter plotscomparison for each sector, with all hours' samples used and all simulated hours. The sectors are: (a) biogenic BIOG, (b) wild fires FIRE, (c) non-electricity generation units Non-EGU, (d) non-road mobile NNRD, (e) on-road mobile ONRD, (f) other point sources MEX, (g) electricity generation units EGU, (h) marine MARINE, and (j) boundary conditions BCON. Note different scales across the panels.

** Figure 2 shows time series for the entire simulation period but Figures 4, 7, and 9 only consider the last 5 days and no explanation is given.*

As is evident on Figure 2, the first 3 days have large influences from initial conditions and, thus, were excluded from analysis. The other captions were updated to mention this decision.

(June 28-30 excluded due to large initial conditions influence)

** The yellow NNRD label in Figures 4, 7, and 9 does not show up very well; could a darker shade of yellow be used?*

The figure was reformatted in an attempt to improve contrast.

** The mention of bulk total deposition in the Figure 4 caption is confusing and unnecessary.*

The deposition phrasing was removed.

* Some figure captions refer to "biogenic BIOG" and some to "BIOG3 vegetation (BIOG)".

All references were changed to “biogenic” for consistency as suggested.

** In the upper labels for Figures S7, S9, and S11-S14, perhaps "Daily-total domainaverage".*

Figures were changed as suggested.