

Dear Editor Jason Williams,

Thanks again for taking your time to organize another round of review. I am submitting the amended version of the manuscript as well as our response to the comments for this minor revision. My co-authors and I would like to thank additional reviewers for their valuable suggestions. I think we have addressed all the concerns accordingly, which has helped us make a much better article that we hope you will publish. In the pages attached, you will see details on how we addressed each reviewer's comment, as well as the revised language that followed.

We appreciate your time.

Sincerely,

Qian Shu

## **Comparison of Ozone Formation Attribution Techniques in the Northeast United States**

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### **Response to Anonymous Referee #3**

First, I would like to thank you for the helpful comments. Each comment is addressed accordingly.

**One remaining issue needs to be addressed before the publication of this revised version of the manuscript: the explanation of why the bulk concentrations of O<sub>3</sub>, VOC etc. are found quite different between CMAQ and CAMx is not specific but more like a guess, and hence not convincing, though the concentration difference seems can explain the difference of apportionments between OSAT and ISAM. I list some of the related such weak explanations as below:**

**Line 402-407 Given that the difference is present in the total concentration, this is unlikely caused by different source apportionment formulation between CMAQ and CAMx. As CAMx only gives pre-lumped VOC, it is challenging to compare individual VOC species between CMAQ and CAMx to explain this difference at current stage. Another possible reasons to cause it could be that models have different internal treatments for advection and diffusion, which can impact surface-level concentrations and indirectly impact chemical reactions.**

**Line 424-428: In Fig. 6, OSAT exhibits the same spatial distribution of MDA8 O<sub>3</sub> total concentrations as other CMAQ-based simulations (OP1, OP2, OP3, OP4, OP5, and CMAQ-BF), with the exception of OSAT's relatively high marine and offshore total concentrations (> 5 ppbv), which could be explained by the difference in planetary boundary layer dynamics or different marine chemistry configuration between the two parent models.**

**Line 452-454: As CMAQ and CAMx both use the same BEIS inventory data, the difference in total VOC concentrations may result from other differences between two models, like chemistry or deposition, accordingly, leading to higher biogenic sources in CAMx (BIO).**

**The actual confusion to the audience is that on one hand the authors claim that “all base meteorological and emissions inputs for CAMx were identical to those for CMAQ”, and from the Table2, it shows that the inputs of ic and bc seem the same as well. The only known explicit difference is the halogen and DMS chemistry in the mechanisms, but as the authors stated that the resulted difference should be small over the land which has been demonstrated by other studies. On the other hand, significant differences in concentrations**

and hourly performances are found between the two models. For example, the Tables 5 and 6 (NMB and R of hourly NO<sub>2</sub> and O<sub>3</sub>), and the following statement:

**Line 359-361: In Fig. 3, CMAQ and CAMx predict 360 similar O<sub>3</sub> concentrations during the day, but differences appear at night, with a maximum difference of 5 ppb. This disparity was discussed in Section 4.1 and can be mitigated by employing the MDA8 O<sub>3</sub> metric.**

Such differences in performances and in the simulated hourly concentrations, plus some unexpected differences in spatial patterns of the results, it seems there are differences existing in the meteorology used in the simulations. Suggestions:

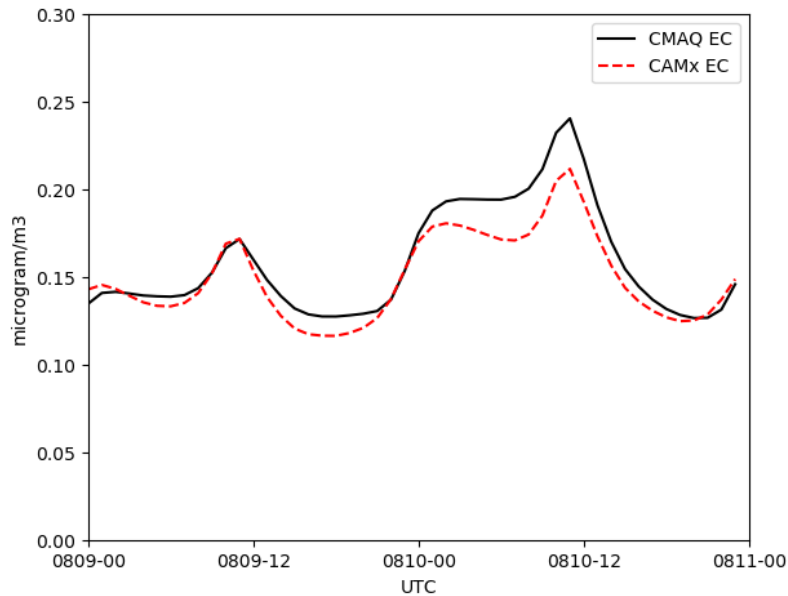
**(1) Compare the simulated spatial fields of hourly concentrations for a couple of inert species, such as EC (or even CO if no other inert species can be found), especially look for differences during the night. The purpose is to check or demonstrate if the input meteorological fields are really identical between the two models' simulations. If significantly different (most likely), then**

**(2) Compare the K<sub>zz</sub> values between the two models. In CMAQ, KZMIN is a default option for setting K<sub>zz</sub> cutoffs to limit K<sub>zz</sub> values over different landuse during the night. In CAMx, there is no such runtime choice, but it provides a KVPATCH program to implement such fixes to the WRF-CAMx outputs. Were KVPATCH outputs used as inputs to CAMx?**

First, it is impossible for the models to have the same bulk prediction or tagged contribution; thus, we want to be clear that identical results are not what we anticipate or want. The main goals of this paper are to 1) document how we recently made ISAM options for secondary species, especially ozone, and 2) evaluate how these ISAM options work. We used the brute force approach and CAMx-OSAT as references because there isn't a perfect reference for assessing our methods. There are a lot of uncertainties when comparing CMAQ-ISAM and CAMx-OSAT, including the ones we mentioned in the manuscript (Lines 285-290) and the ones the reviewer brought up in the comments. We don't think the two models will be the same when it comes to bulk prediction or tagged contribution because they are quite different in many ways, both internally and in their source apportionment methods. To show that our ISAM model is reasonably designed, we anticipate that the results of ISAM should approach OSAT or be reasonable in common sense. Therefore, in addition to OSAT, we also add brute force as another reference since our brute force simulations are running under the CMAQ parent model, in which case we do not need to worry about parent differences. However, there are still imperfections in the comparison between CMAQ-ISAM and CMAQ-BF for secondary species because CMAQ-BF also has its own limitations for nonlinear chemistry (Lines 56-68). To minimize the impact of uncertainties from parent models, we constrained all controllable elements by (1) providing identical inputs as stated in Table 2, (2) providing the same IC and BC, and (3) using the same or at least comparable model processes. One exception is the chemical mechanism because there is not the same option for the model versions we have tested in this study. Even if these variables are tightly controlled, there are structural and parametric differences between the models. The structural differences include different algorithms applied for chemistry and physical processes, which includes different numerical approximations during implementation of identical algorithms. The parametric differences include rate coefficients (JPL vs IUPAC vs and adjustments), physical properties (e.g., surrogate properties for lumped species), and different criteria for computational time-step adjustment. These differences manifest differently by species and cannot be universally explained without performing extensive additional research on each

species, property, and process representation. There are already several studies on model comparisons that show differences with the same inputs (Tesche et al., 2005; Koo et al., 2014; Dolwich et al., 2015; 2014; Shu et al., 2017). The explanation that the reviewer requests is outside the scope of this paper. There are several approaches that could be applied in the future to isolate the difference more perfectly. Future research should either implement source apportionment approaches, both ISAM and OSAT, to the same parent, CAMx or CMAQ, or use process analysis to quantify each process for each species, such as ozone and individual VOCs to comprehend the distinction between two parent models. It requires a significant amount of extra work and is beyond the scope of this work. As a result, we are unable to conclusively explain the bulk difference.

We plotted the EC for CMAQ and CAMx for the two-day case simulations as you asked. EC is an inert species in both models. The outcomes of the two models are anticipated to be somewhat similar but not the same. CMAQ EC is overall higher than CAMx most of the time in the two-day case simulation. This is an expected outcome since we have updated particle dry deposition for CMAQ v5.3 which tends to slow down particle deposition, leading to higher EC concentrations in the surface atmosphere (Shu et al., 2022). A similar EC analysis can also be found in Shu et al. (2017). This is also a good example of how other process uncertainties influence the concentration results, even for chemically inert species when identical inputs are given. Again, we can't draw the conclusion that this EC difference is only caused by deposition without performing process analysis to quantify the influence from other processes. To be clear, the dry deposition update in CMAQv5.3 only affects particles and should not influence gas dry deposition for ozone and VOCs in this study. However, differences in the representation of gas dry deposition exist between the two models (Shu et al., 2017), and this is just one example of how process representations can and often do differ between the two models. In this case, it takes extra efforts in process analysis for more complicated chemical species like ozone and VOC to understand what causes model differences. We do agree it is valuable to understand it and provide a good explanation for that, but this is beyond the scope of the current study. We think more work can be done in the future, as we previously discussed both in the manuscript and in our response here. We are not going to add this EC analysis to either our main manuscript or supplement but keep it here as a response to your comment.



**Other comments:**

**Table 2: It would be better to also list the model options in the table 2, such as advection, diffusion, and deposition schemes, chemistry solver, aero modules etc. In the addition, are the BC identical? It is only stated that the BC are from 12km simulations, but it doesn't say it is from the same CMAQ (or CAMx) 12km simulations or each from their own model's 12km simulations.**

We have added more configuration information in the Table 2 as below. BC and IC are the same. To be clearer, we added the sentence "Identical ICs and BCs were applied to the two models" In Lines 219-220.

Table 2. CMAQ and CAMx model configurations

Model option	CMAQ	CAMx
Model version	Version 5.3.2	Version 7.10
Horizontal resolution	4 km	4 km
Vertical layers	35	35
Meteorology	WRF3.8	WRF3.8
Anthropogenic emissions	2016 NEI version 1 <sup>a</sup>	2016 NEI version 1 <sup>b</sup>
Biogenic emissions	BEIS <sup>c</sup>	BEIS <sup>c</sup>
BC/IC	12km US CONUS	12km US CONUS
Gas phase chemistry	CB6R3	CB6R4
Chemistry solver	EBI	EBI
Aerosol dynamics and chemistry	AERO7/ISORROPIA	SOAP/ISORROPIA
Horizontal advection	PPM	PPM
Vertical advection	PPM	Emery et al. (2011) <sup>d</sup>
Horizontal diffusion	Implicit <sup>e</sup>	Explicit simultaneous 2-D solver

Vertical diffusion	ACM2 <sup>f</sup>	Based on ACM2 <sup>g</sup>
Gas deposition	Pleim and Ran (2011)	Zhang et al. (2003)
Particle deposition	Shu et al. (2022)	Zhang et al. (2001)
Source apportionment	ISAM	OSAT3

<sup>a</sup>EGU were based on continuous emissions monitoring data from 2018 where available. Onroad emissions were projected to 2018.

<sup>b</sup>CAMx EGU and Onroad were identically processed as CMAQ.

<sup>c</sup>BELD v4.1 vegetation data for biogenic emissions, BEIS version is 3.61.

<sup>d</sup>Backward-Euler (time) hybrid centered/upstream (space) solver.

<sup>e</sup>Horizontal diffusion fluxes for transported pollutants were parameterized using eddy diffusion theory.

The horizontal diffusivity coefficients were formulated using the approach of Smagorinsky (1963).

<sup>f</sup>KZMIN was turned on in CMAQ as default.

<sup>g</sup>Vertical diffusivity coefficients were calculated with Yonsei University (YSU) bulk boundary layer scheme (Hong et al., 2006) and were adjusted with the KVPATCH which is comparable to the KZMIN approach in CMAQ.

**Line 64-65: Further, to separate the contributions and interactions of “n” sources, Stein and Alpert (1993) showed that BF would require two to the power of the number of sources (2<sup>n</sup> 65 ).**

**... require two to the power of the number of sources of simulations?**

We have changed it to ‘the number of sources of simulations’

**Line 263-265: As for OTHR, there is no suitable way to retain an appropriate chemical state of the troposphere after subtracting necessary emission categories, initial and boundary conditions from an original CMAQ simulation.**

**What is exactly OTHR? Does it mean the apportionments of IC, BC and all emissions categories don’t add up to 100%? Why not?**

In Lines 258–260. "OTHR is used for all remaining untagged emission categories. In this study, all emissions sectors were tracked as previously mentioned above for OSAT and ISAM." When it comes to OTHR, the user can define how many sectors to track in ISAM. For example, when there are a total of ten emission streams but only five of them are tracked in ISAM, the remaining five emission streams will be defined as OTHR. We have also added the previous sentence in Lines 258–260 to make it clearer for readers. Which is to say, the apportionments of IC, BC, user-defined emission tags, and remaining untagged emission categories should add up to 100%. It may not be perfectly summed up to 100% of the bulk concentrations, but it should match the bulk concentrations very closely.

## References

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