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Comment

## ***Interactive comment on “CESM/CAM5 improvement and application: comparison and evaluation of updated CB05\_GE and MOZART-4 gas-phase mechanisms and associated impacts on global air quality and climate” by J. He et al.***

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Reply to Comments by Reviewer #2

General comments:

The authors implemented two different gas-phase chemical mechanisms (CB05\_GE and MOZART-4x) into the CESM/CAM5 model and performed model simulations for three years. Model predictions obtained with one mechanism are compared to those obtained with the other mechanism and also to measurements from a large number

Full Screen / Esc

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Discussion Paper



of observational datasets. The article will be useful to air quality scientists and merits publication. However, several issues need to be addressed. Specific comments are provided below:

Reply:

We thank the reviewer for the positive review. We have addressed all the comments, please see below our point-by-point reply. The page and section numbers correspond to those in the manuscript with revision in track mode.

Specific comments:

Grid issue Large horizontal grids are employed in the simulation since a global model is used in the study. Surface measurements are generally done at fixed locations. Large spatial variations exist in pollutant concentrations (especially between urban, semi-urban and rural areas). A global model utilizing coarse horizontal grids is unable to capture such spatial variation. Presumably model comparisons with observed data from the Air Quality System (AQS) in the US are not performed for such reason. Comparison of model predictions employing large grids to observed data from fixed surface monitors contain inherent uncertainty. The readers will benefit from a general discussion on the ability of such models to capture spatial gradients of pollutants (especially near urban areas) and comparison with observed data.

Reply:

We agree with the reviewer that there may be large uncertainties associated with comparison of grid averaged model output with pointwise observations. The horizontal grid used in this work is  $0.9 \times 1.25 \text{ o}$ . For model evaluation, there may be multiple observational sites located in one grid cell, so all the observations within one grid cell are averaged and compared to the simulated results in that grid cell. While using grid averaged observations helps reduce the uncertainties to some extent, this approach cannot address the inherent uncertainties associated with the evaluation of the model

Full Screen / Esc

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Discussion Paper



results obtained at a coarse grid resolution.

To address the reviewer's comments, we have provided more information regarding the evaluation and also indicated the inherent uncertainties associated with the approach we used in Section 3.3.

NO<sub>x</sub> issue Column (Table 3) and zonal NO<sub>x</sub> (Figure 3) are over-predicted. In contrast, NO<sub>x</sub> from surface-based monitors (Table 3) and aircraft based monitors (Figure 4) are under-predicted. Despite under-prediction of NO<sub>x</sub> compared to observations from surface-based and aircraft based monitors, model over-predicts NO<sub>x</sub> compared to satellite data. Can the authors discuss some reasons for such behavior? A presentation on the comparison of model and satellite NO<sub>2</sub> is available at: ([https://www.cmascenter.org/conference/2012/slides/yarwood\\_evaluating\\_nox\\_2012.pdf](https://www.cmascenter.org/conference/2012/slides/yarwood_evaluating_nox_2012.pdf))

Reply:

The underpredictions of NO<sub>x</sub> against surface-based observations can be attributed to the uncertainties in the anthropogenic NO<sub>x</sub> emissions as well as vertical transport. It is likely that more NO<sub>x</sub> are transported into upper layers, resulting in the underpredictions in surface NO<sub>x</sub> predictions. The underpredictions of NO<sub>x</sub> against aircraft based observations may be due in part to the uncertainties in the measurements. Some field campaigns (e.g., ARCPAC) focused on the polluted regions with a significant contribution from biomass burning and local sources (Tilmes et al., 2015). The underestimations of emissions from these sources and uncertainties in the vertical mixing scheme can result in the underpredictions of their profiles.

The comparison of tropospheric NO<sub>2</sub> column against satellite data (e.g., SCIAMACHY) can be attributed to the uncertainties in NO<sub>x</sub> emissions and the satellite retrievals. As indicated in Yarwood et al. (2012), errors in satellite NO<sub>2</sub> retrievals are dominated by atmospheric mass factor, which has a large uncertainty due to errors in specification of clouds, surface albedo, a priori NO<sub>2</sub> profile shape, and aerosols. Boersma et al. (2004) found there is an error of 35-60% in the tropospheric NO<sub>2</sub> retrievals, especially

Full Screen / Esc

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Interactive Discussion

Discussion Paper



over polluted areas.

To address the reviewer's comments, we have included the above points along with relevant references in the revised paper, Section 4.2.

Ozone issue The model over-predicts ozone for both mechanisms compared to the observed data (Table 3). The over-predictions has been linked to less titration resulting from the under-prediction of NO<sub>x</sub>, coarse resolution, as well as dilution of NO<sub>x</sub>. It is well-known that the addition of more NO<sub>x</sub> reduces ozone only in NO<sub>x</sub> rich areas. As the additional NO<sub>x</sub> is transported to outside the NO<sub>x</sub> rich areas, it increases ozone in those areas. Thus, the addition of NO<sub>x</sub> may not necessarily reduce overall ozone. I think the use of coarse resolution is diluting NO<sub>x</sub>; thus coarse resolution and dilution of NO<sub>x</sub> are not independent reasons. In addition, model under-predicts VOC. If the model is revised to add corrected amount of VOC emissions, then it will produce more ozone which will further deteriorate the model performance.

Reply:

We agree with the reviewer that O<sub>3</sub> titration is more important over NO<sub>x</sub> rich areas and diluting NO<sub>x</sub> associated with coarse resolution can be one of the reasons for O<sub>3</sub> over-predictions. VOCs are underpredicted in the current model, so it cannot explain the O<sub>3</sub> overpredictions. Another possible reason for O<sub>3</sub> overpredictions may be underestimation in dry deposition. For example, Martin et al. (2014) reported the uncertainties in O<sub>3</sub> dry deposition associated with vegetation phenology in CAM-chem, which were responsible for the mean positive biases of 16 ppb in summertime surface O<sub>3</sub> mixing ratios over eastern U.S. and 8 ppb over Europe, respectively. Therefore, uncertainties in O<sub>3</sub> dry deposition can also partly explain the O<sub>3</sub> overpredictions.

To address the reviewer's comments, we have included the above discussion in the revised paper.

While the model under-predicts NO<sub>x</sub>, its NO<sub>y</sub> predictions agree closer to observed

Full Screen / Esc

Printer-friendly Version

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Discussion Paper



data (Figure 4). If the model is revised to use corrected NO<sub>x</sub>, then it is likely to over-predict NO<sub>y</sub>. Predictions with the CB05-GE mechanism agree better with observed NO<sub>y</sub>. Column NO<sub>y</sub> obtained with MOZART-4x is 46% lower than that that with CB05-GE (section 4.2) which suggests that NO<sub>x</sub> is processed quite differently in the two mechanisms. Which specific chemical reactions are causing such a large difference in NO<sub>y</sub> predictions and how are they different in the two mechanisms? What are the largest 2 chemical species in NO<sub>y</sub> and how they differ between the two mechanisms?

Reply:

To address the reviewer's comments, we have performed additional analyses and added a new Table (Table S2) and two new figures, i.e., Figure S1, to show the dominant species in NO<sub>y</sub> for both MOZART-4x and CB05\_GE, and Figure S2 to show the absolute and relative differences for major NO<sub>y</sub> species between MOZART-4x and CB05\_GE. As shown in Figure S1, NO<sub>x</sub>, HNO<sub>3</sub>, and TPAN (PAN+PANX for CB05\_GE and PAN+MPAN for MOZART-4x) are the major components for NO<sub>y</sub> concentrations, with ratios of 90.5% and 91.7%, respectively, for the sum of the mixing ratios of the top three species to that of NO<sub>y</sub>. NO<sub>x</sub> dominates over East Asia, eastern U.S., and western Europe, whereas TPAN dominates over most oceanic area. As shown in Figure S2, MOZART-4x predicts lower TPAN by  $2.9 \times 10^{19}$  molecules m<sup>-2</sup> (or by 63.4%), which dominates the differences in NO<sub>y</sub> predictions between the two simulations. The differences in TPAN predictions can be attributed to the differences in the kinetic reactions. Table S2 lists the reactions involving TPAN. As shown in Table S2, besides the differences in the reaction rate calculation, MOZART-4x includes one additional reaction for PAN destruction by OH, which is not included in CB05\_GE. In addition, OH levels are higher in MOZART-4x than CB05\_GE, which could result in more TPAN loss through oxidation by OH. These differences can explain the lower TPAN in MOZART-4x than in CB05\_GE. We have included the above information in the revised paper, see pages 35-36.

Some of the nitrogen species partition into aerosol nitrate. Does the difference in

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

NOY between two mechanisms decrease if aerosol nitrate is accounted in the NOY definition? How does the model predictions compare to observed data (Figure 4) if aerosol nitrate is accounted in the NOY definition?

Reply:

Table S3 lists the NOy species used in the calculation for Figure 4 and other NOy related comparisons. Note that Figure 8b includes the column comparison of aerosol nitrate. To address the reviewer's questions, we have included aerosol nitrate in the NOy calculation and replace the NOy plots in Figures 4 and 8a by those plots accounting for aerosol nitrate in NOy. Figure S3 shows the absolute differences in NOY (with and without inclusion of aerosol nitrate) between MOZART-4x and CB05\_GE. If aerosol nitrate is accounted for in the NOy definition, the differences in NOy between two mechanisms decrease over East Asia, eastern U.S., Europe, and middle Africa as aerosol nitrate is higher in MOZART-4x over these regions (see Figure 6b). For the rest of areas, the differences in NOY between two mechanisms increase if aerosol nitrate is accounted for in the NOY definition.

To address the reviewer's comments, we added the above discussions in page 36.

NOY definition includes BrONO2 which suggests bromine chemistry is being used in the model. Which bromine emissions are used in the model?

Reply:

We have bromine chemistry included in both MOZART-4x and CB05\_GE. For bromine/chlorine species (e.g., CF2CLBR, CF3BR, CFC11, CFC12, CH3BR, and CH3CL), their surface concentrations are specified using the historical reconstruction from Meinshausen et al. (2011). No bromine emissions were included.

The above point was added in Section 3.1.

Also, need to clarify that section 4.2 contains only model-to-model comparison.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Reply:

This has been clarified in Section 4.2.

HNO<sub>3</sub> issue The model over-predicts HNO<sub>3</sub> over CONUS while under-predicting it over Europe. Under-prediction of HNO<sub>3</sub> over Europe is linked to under-prediction on NO<sub>x</sub>. Surface NO<sub>2</sub> comparison for CONUS is not shown in Table 3. Does the over-prediction of HNO<sub>3</sub> over CONUS occur due to over-prediction on NO<sub>x</sub>?

Reply:

To address the reviewer's comment, we have included NO<sub>2</sub> evaluation against AQS sites over CONUS. The results show that both CB05\_GE and MOZART-4x underpredict surface NO<sub>2</sub> concentrations, with NMBs of -52.2% and -51.4%, respectively. The overpredictions of HNO<sub>3</sub> over CONUS are mainly due to more total nitrate partitioned into HNO<sub>3</sub> (which is reflected by the underpredictions of NO<sub>3</sub><sup>-</sup> over CONUS) resulted from the overpredictions of SO<sub>4</sub><sup>2-</sup> over CONUS. Compared to NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> can more easily combine with NH<sub>4</sub><sup>+</sup> to stay in aerosol phase. There are not enough NH<sub>4</sub><sup>+</sup> to neutralize NO<sub>3</sub><sup>-</sup>, driving NO<sub>3</sub><sup>-</sup> to HNO<sub>3</sub> in the gas-phase resulting in overpredictions of HNO<sub>3</sub> over CONUS. We have included this explanation in the revised paper, Section 4.1.1.

Cl<sup>-</sup> issue The model under-predicts Cl<sup>-</sup> over CONUS while over-predicting it over Europe. Over-prediction of Cl<sup>-</sup> over Europe has been linked to gas/particle partitioning. Can the authors suggest any reasons for under-prediction of Cl<sup>-</sup> over CONUS? Reff et al. (2009) suggest many sources can emit Cl<sup>-</sup>; are emissions from these sources included in the study? Reff, et al.: Emissions inventory of PM<sub>2.5</sub> trace elements across the United States, Environmental Science & Technology, 43, 5790–5796, 2009.

Reply:

We did not include any anthropogenic Cl<sup>-</sup> emissions in the model except from sea-salt emissions, which is calculated online in CESM/CAM5. The missing sources can

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



contribute to the underpredictions of  $\text{Cl}^-$ . On the other hand, due to the overpredictions of  $\text{SO}_4^{2-}$ , there are less  $\text{NH}_4^+$  available to neutralize  $\text{Cl}^-$ , driving  $\text{Cl}^-$  to  $\text{HCl}$  in the gas-phase, resulting in underpredictions of  $\text{Cl}^-$ . In addition, the performance of  $\text{Cl}^-$  over CONUS is only for fine  $\text{Cl}^-$  (Aitken, accumulation, fine sea-salt, and fine dust modes), whereas the performance of  $\text{Cl}^-$  over Europe is for fine and coarse  $\text{Cl}^-$  (all seven modes). As the thermodynamic equilibrium is not treated for coarse particles (the irreversible condensation of  $\text{HCl}$  is assumed to occur on the surface of coarse particles), it is likely that the model overpredicts coarse  $\text{Cl}^-$ , but underpredicts fine  $\text{Cl}^-$  due to the missing sources.

To address the reviewer's comments, we have clarified the above issues in page 23, Section 4.1.1.

**$\text{SO}_2/\text{SO}_4^{2-}$  issue** The model over-predicts surface  $\text{SO}_2/\text{SO}_4^{2-}$  compared to the observed data (Table 3). The over-estimation of  $\text{SO}_2$  has been explained with  $\text{SO}_2$  emissions, injection height uncertainty, and vertical mixing issue while the over-prediction of  $\text{SO}_2$  has been suggested to be the reason for over-prediction of  $\text{SO}_4^{2-}$ . Most models over-predict surface  $\text{SO}_2$  and tend to under-predict surface  $\text{SO}_4^{2-}$  compared to observed data. Here, the model over-predicts both surface  $\text{SO}_2/\text{SO}_4^{2-}$ . Interestingly column  $\text{SO}_2$  is underpredicted which suggests possible problem with vertical mixing in the model that may have contributed to the over-prediction of surface  $\text{SO}_2$ .

Reply:

As we explained, the overpredictions of surface  $\text{SO}_2$  concentrations may be due to the uncertainties in the  $\text{SO}_2$  emissions, injection height uncertainty, and vertical mixing. The underpredictions of column  $\text{SO}_2$  can also be attributed to the uncertainties in the vertical mixing as well as uncertainties in the satellite retrievals. For example, Lee et al. (2009) found that there is an overall error in the annual  $\text{SO}_2$  retrievals of 45-80% over polluted regions, especially over eastern China. Therefore, the uncertainties in the satellite  $\text{SO}_2$  retrievals can affect the model evaluation. The overpredictions of

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)



SO<sub>4</sub>2- can be due to the overpredictions of SO<sub>2</sub> as well as uncertainties in the SO<sub>4</sub>2- emissions. CESM/CAM5 also reads the default vertical SO<sub>4</sub>2- emission profiles for the simulations. The uncertainties in the SO<sub>4</sub>2- emissions can contribute to the inaccurate predictions of SO<sub>4</sub>2- as well.

To address the reviewer's comments, we have included the above explanation in the revised paper in Section 6.

Other factors may also affect SO<sub>4</sub>2- predictions. Aqueous-phase reaction with H<sub>2</sub>O<sub>2</sub> and gas-phase reaction with OH tend to be the most important pathways for the conversion into SO<sub>4</sub>2-. The article does contain any discussion of predicted oxidant levels (H<sub>2</sub>O<sub>2</sub> and OH). If the predicted oxidant levels are too high, SO<sub>4</sub>2- predictions will also be high. I am not suggesting to perform any detailed comparison of predicted H<sub>2</sub>O<sub>2</sub> and OH with observed data but some discussion of predicted oxidants levels and typical observed values will be helpful to readers.

Reply:

We agree with the reviewer that aqueous phase reaction with H<sub>2</sub>O<sub>2</sub> and gas-phase reaction with OH are important pathways for SO<sub>4</sub>2- formation. We have added the comparisons of OH and H<sub>2</sub>O<sub>2</sub> profiles with aircraft measurements in Figure 4 in the revised paper. As shown in Figure 4, both OH and H<sub>2</sub>O<sub>2</sub> are underpredicted, but MOZART-4x predicts slightly higher H<sub>2</sub>O<sub>2</sub> within 4-km above the surface compared to CB05\_GE. However, the performance here only represents the local condition, instead of global condition.

To address the reviewer's comment, we also compared the global mean tropospheric OH concentrations with other studies. The simulated air-mass weighted tropospheric mean OH concentrations predicted by MOZART-4x and CB05\_GE are both  $13.1 \times 10^5$  molec cm<sup>-3</sup>, which is slightly higher than Naik et al. (2013) with present-day tropospheric mean OH levels of  $11.1 \pm 1.6$  molec cm<sup>-3</sup>. So both CB05\_GE and MOZART-4x tend to predict higher OH levels, which may partly explain the overpredictions of

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

SO42-. We have included this explanation in the revised paper, Section 4.1.1.

The model under-predicts cloud liquid water path compared to MODIS data (Table 5). If the model is revised to include the correct amount of cloud liquid water, then it will produce more S<sub>ol</sub>CS<sub>42</sub>—and the model performance for S<sub>ol</sub>CS<sub>42</sub>—will further deteriorate and the model performance for SO<sub>2</sub> will improve. It is also possible that the model produces more S<sub>ol</sub>CS<sub>42</sub>— by placing clouds in incorrect vertical layers. Inadequate precipitation in the model may also lead to higher than observed S<sub>ol</sub>CS<sub>42</sub>—in the model.

Reply:

Cloud liquid water path (LWP) is moderately underpredicted by both MOZART-4x and CB05\_GE against MODIS data, but it is relatively well predicted against Bennartz (2007), which filtered out large uncertainties associated with MODIS retrievals. Therefore, the model predicts LWP relatively well against Bennartz (2007). But we agree with the reviewer that the model may inaccurately predict clouds vertically as the simulations with both MOZART-4x and CB05\_GE show moderate biases for COT and CDNC.

We also agree with the reviewer that inadequate precipitation can contribute to the overpredictions of SO<sub>42</sub>-. Both simulations are driven by prescribed meteorology, so we did not compare meteorology from the two simulations. To address the reviewer's comment, we evaluated precipitation and the results show that precipitation is well predicted by both MOZART-4x and CB05\_GE, with NMBs of 1.9% and 1.6%, respectively. Therefore, the overpredictions of SO<sub>42</sub>- are not due to the insufficient precipitation.

SOA issue The model is able to capture observed SOA (Table 3 and Figure 2). However, VOCs are under-predicted. If the model is revised to use corrected amount of VOC emissions, then SOA predictions will be higher. Does the model capture SOA for the correct reason? Again, discussion of predicted oxidant levels with typical observed values will also be helpful for discussing SOA predictions?

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

Reply:

We agree with the reviewer that using correct VOCs emissions can increase the SOA concentrations. As we discussed above, our model tends to predict higher OH levels on a global scale, which can partly explain the well predicted SOA despite the under-predictions of VOCs. Also, the SOA statistics are calculated using only four pairs of seasonal mean values at four sites in the U.S. where the observed SOA data are available during 2008-2010; they, therefore, are not be representative of the entire CONUS because of limited data used for calculation. We have indicated this limitation in the paper.

Minor editorial suggestions The objectives of the study are to examine the differences in the SOA predictions . . . , and study the sensitivity of air quality and climate predictions to different gas-phase chemical mechanisms (introduction section). However, the title does not reflect that SOA predictions are being examined. Perhaps the authors can reconcile the apparent difference.

Reply:

SOA is part of the global air quality, so the title does include examination of SOA predictions from both simulations.

Most chemical species in the article have been defined. However, some have not been defined. For example, CO, HO<sub>x</sub>, and NO<sub>x</sub> in line 24 (page 7191) are not defined. I suggest that the authors check the entire article and define the chemical species when it is used first time. In the description, two names for  $\alpha$ -pinene (alpha-pinene and  $\alpha$ -pinene) and  $\beta$ -pinene are used. One name should be used throughout the article (Chemical mechanism - 2.1)

Reply:

We have checked the paper thoroughly and defined all the species and we also use consistent names for the species in the revised paper.

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Full Screen / Esc

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Figure 1 also contains a comparison of SOA which is not mentioned in the caption

Reply:

SOA has been added in the Figure 1 caption in the revised paper.

Conclusion section is long and can be shortened

Reply:

Conclusion has been shortened in the revised paper.

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Full Screen / Esc

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Interactive Discussion

Discussion Paper

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