Authors' response to comments made by anonymous reviewer #1:

Summary

The manuscript of Tsimpidi et al. "Aerosol Composition Trends during 2000 -2020: In depth insights from model predictions and multiple worldwide observation datasets" compares measured and modeled aerosol pollutants between 2000-2020 in a global scale. The paper includes a large experimental data set, and it examines extensively the pollutants trend for each area. This is an important study that should be published, after modifications.

We would like to thank the reviewer for his/her thoughtful review and positive response. Below is a point-by-point response (in black) to the major and minor comments (in blue).

Major Comments

1. *My main concern is that due to the large amount of the compared data (time and space) the paper is quite long (50 pages without including references), and the reader gets tired fast. From my point of view, it is difficult to digest all this information. So, it should be somehow more concentrated and shortened. In addition, there is a lot of statistical information but in general there is little connection between all these results. Moreover, the reasons for any discrepancies between measurements and simulations should be discussed in more detail and propose modification/addition in the model to capture more accurately the measurements.*

Thank you for your valuable feedback. We do understand the concern regarding the length of the manuscript. We have made efforts to reduce the size of the revised manuscript, particularly in the evaluation section, to make it more concise and coherent. Specifically, the evaluation section (Section 4) has been reduced and is now a subsection of Section 3 (Model Calculated Dataset). Furthermore, the scatterplots have been removed, and the evaluation metric tables have been moved to the supplement to reduce the number of figures and tables in the main manuscript, making it easier for the reader. At the same time, we aimed to enhance the discussion on aerosol trends, which is the main focus of our paper, by providing more detailed explanations for any discrepancies between measurements and simulations, including references to the emission trends used and the model's performance against observations. Additionally, we have discussed in more detail, where appropriate, possible modifications and additions to the model that could help improve its accuracy in capturing the observed trends.

2. *Section 4 (In depth model Evaluation) describes the comparison between measured and modeled mass concentrations. In each sub-section (4.1-4.4) the authors compare the average mass concentration (sulfate, nitrate, ammonium, OA, SOA and POA) from each campaign to the average mass concentration that the model predicts for the specific site and time. What is the time resolution of the EMAC? Hourly/daily/monthly? This should be explained in the text. The comparison between the model and the measurements should be made throughout the whole campaign and not taking on only one value from each campaign. This could be misleading for the model's performance as for example some days (if the model resolution is every 24 hours) could be very badly simulated, leading to a high overall discrepancy.*

We agree that a comprehensive evaluation over the entire campaign may reveal discrepancies in the model that may not be observed when comparing monthly averages. However, the AMS/ACSM data used in our study includes 744 datasets from different field campaigns conducted worldwide over the last 20 years. Therefore, it is not feasible to provide a detailed evaluation for each campaign. The primary purpose of this evaluation is to establish the ability of the model to capture long-term average trends in pollutants, which is the main focus of our paper. The model runs with a time resolution of about 20 minutes for chemistry and saves the output of pollutant concentrations as daily averages. The comparison with observations is made using monthly averages, or campaign averages if the campaign lasts less than a month. This information has been included in the revised text.

3. *On the contrary section 5 (Aerosol Trends) is much more meaningful for measurements and model comparison. The core of the results should be this part. Section 4 should be complementary to section 5, and I suggest that the authors should incorporate selected parts of section 4 to section 5 accordingly. The discussion should be done by area (i.e. Europe, N. America, E. Asia) so that the reader reads the "story" of each area.*

We fully agree with the proposed changes. We have shortened the discussion of the model evaluation (Section 4) and now focus more on the actual observed and simulated aerosol trends (Section 5). Selected parts of the model evaluation have now been incorporated into the discussion in Section 5 to explain discrepancies between the model and observations on the simulated trends. The evaluation section (Section 4) has been reduced and is now a subsection of Section 3 (Model Calculated Dataset). In addition, the discussion on aerosol trends is organized by region (i.e. Europe, North America, East Asia) to provide a clear and coherent description of aerosol trends for each region.

Minor Comments

1. *There are several mistakes using the words "best" and "worst" in the text. For example, lines, 618, 620, 649, 651, 810 etc. Please check the whole manuscript and make the appropriate changes. Also check the usage of the word "highest" (e.g., lines, 286, 297, 411 etc.).*

Thank you for pointing this out. We have corrected the text accordingly.

2. *Line 70: Please replace "form" with "formed".*

Corrected.

3. *Line 228: "high OA fractions with regional means" please rephrase.*

We have rephrased the sentence as "Campaign data from tropical and subtropical regions (e.g., Latin America and Southern/Southeast Asia) is strongly influenced by biomass burning and biogenic VOC emissions, resulting in notably large OA fractions in aerosol composition, with regional averages around 65% and a peak of 92% in the Amazon.

4. *Line 414: PM2.5 please use subscript for '2.5'.*

Done.

5. *Line 624: Please replace "underestimating" with "it underestimates".*

Done.

6. *Line 626: Please add "it" after therefore.*

Done.

7. *Line 637: Please replace "show" with "shows".*

Done.

8. *Line 653: Please replace "resolved" with "simulated".*

Done.

9. *Line 678: Please delete "the".*

Done.

10. *Line 681: Please replace "lie" with "lies".*

Done.

11. *Line 713-714: "Ammonium tends to be overestimated during autumn and underestimated during the rest of the seasons; especially during the summer" Is there any explanation for this tendency?*

Current emissions inventories offer reasonable estimates of total annual NH_3 emissions, but significant uncertainties remain regarding their seasonal distribution. Since animal husbandry and fertilizer application are the primary sources, seasonal variations are difficult to quantify (Paulot et al., 2014). Studies in the U.S. suggest NH_3 emissions may be underestimated in summer and overestimated in other seasons, while estimates for spring and fall remain uncertain due to biases in precipitation predictions (Gilliland et al., 2006; Paulot et al., 2014). This information has been added to the revised text.

12. *Line 719: "While the good model performance" please rephrase.*

We have rephrased the sentence as "The model's strong performance for ammonium over Europe indicates an accurate emission inventory for agricultural and livestock NH_3. However, the overprediction of nitrate and underprediction of sulfate suggest that the model overpredicts the fraction of ammonium that exists as ammonium nitrate rather than ammonium sulfate."

13. *Lines 727-728: "On the other hand, ammonium is overpredicted close to the deserts of Inland China (e.g., over Tibet) and over South Korea" Do you have any explanation about this behavior?*

These areas exhibit the most significant nitrate overpredictions (Figure 11a). As a result, errors in nitrate levels cause excessive NH_3 condensation into the aerosol phase, leading to unrealistic ammonium nitrate formation. This information has been added to the revised text.

14. *Line 750: "EMAC tends to overpredict some low OA concentrations measured by AMS" this sentence is not very clear, please rephrase.*

We have rephrased the sentence as "However, EMAC tends to overpredict certain low OA concentrations observed by AMS at a few rural locations during summertime (Figure 13a)."

15. *Line 785: ".. evaporation of organic compounds upon emission…" So, vaporization is not considered by the model? Please explain.*

The ORACLE module, which describes the phase partitioning of organic compounds, does not explicitly simulate evaporation or vaporization processes. Instead, it assumes instantaneous equilibrium between the gas and particle phases and determines the amount of material that evaporates or condenses based on the compound's volatility, total ambient concentration, and ambient temperature. Due to the model's coarse spatial resolution, it underestimates organic compound concentrations near emission sources, leading to an overestimation of their evaporation into the gas phase and, consequently, an underestimation of POA concentrations. More details on ORACLE's phase partitioning calculations can be found in Tsimpidi et al. (2014).

The sentence in the text has been revised to: *"Over urban locations, POA is more severely underestimated (NMB = -68%) due to the coarse spatial resolution of the model and the evaporation of organic compounds upon emission, as the model underestimates local organic compound concentrations near the source."*

16. *Line 803: Please replace "are in very good" with "are in a good".*

We have replaced "are in very good" with "are in a good".

17. *Figure 19: There are no g and h subplots, so please make the appropriate changes in the figure caption.*

Done.

18. *Figure 21: There are no g and h subplots, so please make the appropriate changes in the figure caption.*

Done.

19. *Line 1065: PM1 please use subscript for '1'.*

Done.

20. *Line 1080: Since there is little discussion about EC in the text, it should not be mentioned in the conclusions*

We have removed the reference to EC.

21. *Figures 17, 20 and 22 should be moved to the SI, they are just the average of Figures 16, 18, 19 and 21 and they don't add value to the paper.*

Figures 16, 18, 19 and 21 show the trends of the observed (and the corresponding simulated) concentrations for the $PM_{2.5}$ aerosol components measured by the filters. Figures 17, 20 and 22 show the trends of the observed (and the corresponding simulated) concentrations for the PM_1 aerosol components measured by the AMS field campaigns. For the $PM_{2.5}$ concentrations routinely measured by the filters, we were able to plot the temporal evolution of the observed concentrations. However, the AMS field campaigns do not provide consistent measurements of PM_1 components throughout the decade. Therefore, we only show the decadal averages for each region to allow a rough statistical comparison between the two decades and to provide insight into the overall tendency of the observed aerosol composition trends for each region. This information is provided at the beginning of Section 5. The captions of the figures have also been changed to make it clear that the first set of figures is calculated on the basis of filter observations for $PM_{2.5}$, while the second set refers to AMS observations for PM_1 components.

References

Gilliland, A. B., Wyat Appel, K., Pinder, R. W., and Dennis, R. L.: Seasonal NH3 emissions for the continental united states: Inverse model estimation and evaluation, Atmospheric Environment, 40, 4986-4998, https://doi.org/10.1016/j.atmosenv.2005.12.066, 2006.

Paulot, F., Jacob, D. J., Pinder, R. W., Bash, J. O., Travis, K., and Henze, D. K.: Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3), Journal of Geophysical Research: Atmospheres, 119, 4343-4364, https://doi.org/10.1002/2013JD021130, 2014.

Tsimpidi, A. P., Karydis, V. A., Pozzer, A., Pandis, S. N., and Lelieveld, J.: ORACLE (v1.0): module to simulate the organic aerosol composition and evolution in the atmosphere, Geoscientific Model Development, 7, 3153-3172, 10.5194/gmd-7-3153-2014, 2014.