General Comments

This paper looks at the performance of the GEOS-Chem chemical transport model across a range of field campaigns for secondary inorganic aerosols. This is an important topic as models generally struggle to simulate nitrate and ammonium in particular. The authors point out that this issue prevents successful policy-relevant studies for air quality and climate. Overall, the paper analysis is thorough, within the scope of ACP, and will hopefully help push the field to address this longstanding issue. The comments below seek to broaden the discussion and analysis in just a few ways. For example, the paper could be strengthened by adding evaluation of the sulfate and nitrate oxidation ratios for each campaign in a similar manner as Figure 4. As the paper already presents SO_2 and HNO_3 data, these ratios should be straightforward to include. A second general comment is that throughout the paper, statistics (R^2, NMB) are presented but it is unclear to which underlying data they apply – point by point aircraft vs. model data? Vertical profile averages? More clarity on this would help the reader understand whether model issues are temporal, spatial, or both. Finally, to best guide the community, the paper should very clearly lay out exactly what types of measurements are needed to improve these long-standing model biases, related to both production and loss processes.

Specific Comments

Line 42 – Why is nitrate partitioning difficult to assess due to limited ammonia? Do you mean total NHx? Can you also add a sentence to the abstract clarifying what measurements are needed to constrain nitrate in models?

Line 63 – Consider citing also (Zhai et al., 2021) here.

Line 67 – Not necessarily true in urban areas, non-agricultural contribution can be large, see these citations: (Chang et al., 2019; Lim et al., 2022; Link et al., 2017; Phan et al., 2013; Song et al., 2009; Sun et al., 2017)

Line 89 – You could also cite (Chen et al., 2019), they found that models tend to overly partition HNO_3 into nitrate.

Line 178 – There are many new developments related to aerosols in GEOS-Chem since 13.3.4. You comment on some of them later. It could help the reader to mention that you discuss recent developments such as nitrate photolysis in Section 5.5.

Figure 2 – Can you explain the longer lifetime for nitrate compared to ammonium or sulfate?

Line 265 – Does this poor R^2 reflect spatial and/or temporal issues? Is the diurnal cycle well captured but the campaign-to-campaign variability poor? Or is it both? It is unclear if the R^2 in Figure S1 is for the vertical variability?

Figure 4 – Can you put a unit on NMB? In the text on line 259 there is a unit of %, but it seems like the campaign-to-campaign biases for nitrate should be larger than a few %.

Line 327 – Why not run with E-AIM to compare to ISORROPIAII as has been done to compare against models by one of your co-authors (Nault et al., 2021)?

Line 370 – Is there a figure for the NH_3 improvement? Can you make a strong statement then that observations of NH_3 are key to future field campaigns?

Line 376 – Can you determine for which campaigns nitrate formation is limited by NH_3 availability (rather than NOx)? Is there a low bias in campaigns that are limited by NH_3?

Line 384 – Can you show model (not just offline ISORROPIA II) nitrate oxidation ratio just for the campaigns where you have HNO_3?

Section 5.1.2 – Figure 11 is very interesting. It might be sufficient to move Figure 10 to the supplement along with its associated discussion and just focus on Figure 11 here.

Line 483 – Again it would be useful to know whether observationally, ammonium nitrate for each campaign is limited by HNO_3 or NH_3.

Line 495 – Previously you stated that the main source of NH_3 was agriculture? If this is true in the model, shouldn't you cut agricultural sources not anthropogenic sources?

Line 534 – If Heald et al., 2012 saw a weak response from a doubling of HNO_3 dry deposition velocity, why would you expect a different response here in your similar sensitivity test? You cite a factor of 5 from Travis et al., 2022, but do not explore this possibility?

Line 535 – If 800 mb is roughly 1.5 km, that could be at the top of the boundary layer or higher for some campaigns. (like EMERGE-EU or WINTER). Maybe provide the value for ~900 mb as a better indicator of the boundary layer for all campaigns?

Line 552 – It would be clearer to say that this update to H* actually reduced wet and dry deposition.

Line 563 – Can you put your model sensitivity with the wet deposition changes on Figure 6 in a different color? Could you also put these model sensitivities on Fig. 5, Fig. S1, Fig. S2, and Fig. S3?

Line 600 – Why would FIREX-AQ have an EF of 0.71? Doesn't this mean that there was a high fraction of sea salt aerosol which doesn't make sense for an inland wildfire campaign?

Line 606 – Could you provide a budget for the other 59% of surface nitrate production? Is there any other pathway worth considering for a sensitivity test?

Line 621 – Models typically overestimate OH (e.g., (Prather and Zhu, 2024)). Do you think that model HNO_3 would be well simulated if model OH was correct? Or is it clear that the lifetime of HNO_3 is too long for some unknown reason?

Line 636 – Can you give the % of these observations in these extremes across all field campaigns? How much is a better thermodynamic model needed? Is the new HETP model expected to help with these extremes? (Miller et al., 2024).

Line 660 – What about wet and dry deposition? What measurements are needed to better understand those processes?

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