

The authors assess the ability of the GEOS-Chem chemical transport model to simulate the average diel variability in PM_{2.5} over the United States in 2016. They perform sensitivity simulations to test improvements to the representation of anthropogenic emissions, dry deposition, boundary layer height, and vertical representation errors. Overall, the authors find that hourly anthropogenic emissions and correcting from the lowest model layer midpoint to the measurement altitude significantly improve the diel variation of simulated PM_{2.5}.

Major Comments

Overall, the authors should better discuss the reasons why nitrate aerosol drives the diurnal behavior in the model, and what chemical pathways produce this nitrate in GEOS-Chem. The authors do split their analysis into season and region. However, it could provide much more insight into PM_{2.5} diel behavior and model issues if the authors examined rural vs. urban behavior. With these revisions, this paper would be appropriate for publication in ACP.

Specific Comments

Line 63 – Which SOA scheme is being used? Simple, complex, VBS? Could the authors break down SOA and POA in their speciated PM_{2.5} plots?

Line 80 – NEI 2016 is available in GEOS-Chem and appears to be default according to the website. Why was this not used? http://wiki.seas.harvard.edu/geos-chem/index.php/EPA/NEI11_North_American_emissions

Line 84 – Are any anthropogenic or fire emissions emitted above the first model level?

Line 100 – The authors could highlight upfront that there has been a lot of recent developments in understanding particulate deposition and clarify for the reader the need to investigate how important these developments could be for understanding PM_{2.5} variability.

Section 3 – At what RH do these methods measure? Is the model PM_{2.5} adjusted for this?

Line 136 – It looks like the first peak is at 8am, that is not really ‘mid-morning’.

Line 142 – The authors should discuss which components of PM_{2.5} are driving the diurnal cycle. For example, recent work has described model nighttime overestimates in nitrate (Miao et al., 2020; Travis et al., 2022; Zhai et al., 2021). It might be better to describe the components driving the diurnal cycle behavior directly after Fig. 2 rather than later on.

Figure 2 – It would be very informative if Fig. 2 was split into urban and rural as there are likely large difference in dynamics, emissions, chemistry etc. between them.

Line 155 – It is very difficult to think about how emissions drive errors in PM_{2.5} without first seeing the speciated PM_{2.5} diurnal cycle.

Line 250 – The authors might consider that if the model nitrate is overestimated, hygroscopic growth might also be too larger.

Line 261 – Please provide references for this statement. “Vertical concentration gradients within 120 m of the surface have been widely observed for aerosol species in previous field campaigns.”

Line 265 – How different is the temperature and RH between 2m and the lowest model level, and how would that impact thermodynamic partitioning of sulfate-nitrate-ammonium aerosol?

Line 306 – Please briefly describe how the AMDAR PBLH is generated.

Line 313 – Please comment more on why the PBLH in GEOS-FP might be too high and what that could mean for other parts of the simulation (e.g., temperature).

Line 315 – Could you explain why it would be reasonable to have a PBLH maximum in spring?

Line 384 – See the additional two papers on nitrate overestimates (above Line 142). It would be worth commenting on the reasons described in those papers. The authors should comment that a large bias remains in their modeled nitrate. For example, there seems to be general consensus that NO₂ hydrolysis shouldn’t be very important in the model and that N₂O₅ hydrolysis should be the main pathway (one example <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019GL085498>). Which pathways dominate the authors’ results? The authors could consider turning NO₂ hydrolysis off if it seems unreasonably large.

Line 425 – Could you be quantitative about the impact of resolution?

Line 456 – What is this product and what will the increased resolution be in the PBL?