Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2022-340-RC2, 2022

Comment on acp-2022-340

Anonymous Referee #1

Referee comment on "Observation-based constraints on modeled aerosol surface area: implications for heterogeneous chemistry" by Rachel A. Bergin et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-340-RC2, 2022

The authors compared CMAQ simulated particle surface areas (Sa) with those observed during the 2011 DISCOVER-AQ Baltimore-Washington and showed that the model under-predicted the Sa by a factor of ~ 2. Possible reasons for the difference were analyzed and discussed. The authors also studied the impact of the model simulated Sa on heterogeneous chemistry. Overall, the manuscript provided a useful assessment of CMAQ predicted Sa. However, the following comments need to be properly addressed before I can recommend it to be published in ACP.

- Particle surface area and number concentrations depend strongly on the size distributions of primary particles emitted (especially in the urban region like this study). It is unclear in the manuscript how primary particles of different sources (industry, residential, traffic, etc.) are parameterized in the model. This information is necessary and a table summarizing these will be helpful.
- Particle size distributions (and thus Sa) are also affected by new particle formation (or nucleation) and growth, especially at higher altitudes and in areas away from the urban center. The authors shall discuss how new particle formation was treated in the model and how the process might have affected the results.
- The results presented in this paper are based on CMAQ simulations with 12 × 12km latitude and longitude horizontal resolution, and hourly temporal resolution. As the authors themselves acknowledged: "The 12 km ×12 km spatial resolution and 1-hour temporal resolution of CMAQ is significantly larger and longer than the spatial and temporal resolution of the aircraft data, resulting in an inherent contrast in resolution between model and measurement that may play a role in the variance in $Sa,$mod $/Sa,$meas". Any reasons why you did not use a nested domain with higher resolution? Also, why not use a higher temporal resolution? 1-hour temporal resolution is too coarse for this kind of simulation (12 km x 12 km).
- Figure 1. Is the figure for an average of values at all altitudes? It will be helpful to separate those in the boundary layer from those above the boundary layer. I expect a large difference in particle size distributions at different altitudes. Also maybe you can use the same y-axis scale for observed and model values for easier comparison?
- The manuscript does not have a summary session. I would suggest that the authors add one.

- L79-81: Are the median diameters for the three modes fixed in the model? How the size range for each mode is decided? How was the value of geometric standard deviation determined?
- L100: ">99%": What is the exact value?
- L104: How condensation of water is treated?
- Table 1: Q1, Q3 values from Jaegle et al. 2018 appear to be incorrect.
- Why did two simulations (2011 &2015) use two different versions of the CMAQ model and chemistry/aerosol schemes (CB5 vs CB6, AER06 vs AER07)?
- L222: Please explain why the measured values are dry Sa.
- L145: relative to?
- L283: Please give some details on how this was measured. Completely dry or at some fixed RH?
- L305-309: What are the possible reasons? Can the difference be reduced by increasing resolution and/or modifying assumed mode sizes?
- L310: This is inconsistent with what is stated in the figure caption. Please add more labels to the x-axis to make it clearer.
- L388: This is speculation here. Why not use a nested domain with 4 km resolution to look into the effect of resolution?
- L446: Does the condensation change the sizes of particles represented in the model?
- L481: Please explain how the reactive uptake coefficient was measured.
- L485: Why? Winter and summer particle size distribution (and thus Sa) could be quite different.