Atmos. Chem. Phys. Discuss., 15, C9037–C9040, 2015
www.atmos-chem-phys-discuss.net/15/C9037/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source attribution of aerosol size distributions and model evaluation using Whistler Mountain measurements and GEOS-Chem-TOMAS simulations" *by* S. D. D'Andrea et al.

Anonymous Referee #2

Received and published: 6 November 2015

General comments:

In this study, the authors evaluated prediction ability of GEOS-Chem-TOMAS at 4x5 and 0.5x0.667 horizontal resolutions with online aerosol microphysics measurements from the Peak of Whistler Mountain. They found that using temperature as a proxy for BL influence can significantly improve the model measurement comparisons. The best threshold temperature was around 275K for the 4x5 simulations and around 279K for the 0.5x0.667 simulation. By running the cases without Asian anthropogenic emissions

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and without biomass burning emissions, they quantify the contribution of these sources to aerosols at the Peak of Whistler Mountain. There are very few observation and modeling studies on remote and free tropospheric aerosol microphysics like this work, therefore, this kind of study and exploration should be encouraged and worth to be published on ACP. However, I have two major questions would like to be addressed by the authors.

1. I think using temperature as a proxy for BL influence is useful for data analyzing of mountain measurement at Whistler Peak. But I do not think it is a good/reasonable method for improving model measurement comparison. In GEOS-Chem-TOMAS, it already considered vertical transport processes. It means the direct way for model validation is comparing simulated and observed aerosol at the same height/pressure level like the work done by Yu and Hallar (2014). If the authors doubt model ability to represent vertical transport processes, they'd better to give an alternative method trusted by them or assess the uncertainties due to vertical transport processes. In this study, GEOS-Chem-TOMAS simulated aerosol at surface is generally larger than observation, while the simulated aerosol at mountain peak layer is generally lower than observation. Therefore, the threshold temperature shown in this study is just a weighting factor to make simulation closer to observation.

2. Ternary homogeneous nucleation (Napari et al., 2002; Westervelt et al., 2014) is an old nucleation scheme which can hardly be supported by current laboratorial and field observations. One of the defects of the modified nucleation treatment in this work is that they predict too low nucleation rate within boundary layer. Yu et al. (2010) evaluated major nucleation schemes in GEOS-Chem. Their work indicated that different nucleation schemes do have significant impacts on aerosol number concentrations. New particle formation is the principle step of aerosol microphysics modeling. I am very interesting about how state-of-the-art nucleation schemes impact this work's summaries. In GEOS-Chem-TOMAS, it has some options for different nucleation schemes. The authors need to present some discussions and results on this issue.

ACPD
15, C9037–C9040, 2015

Special comments:

1. P24811, L2-3. Primary black carbon and organic carbon emissions in GEOS-Chem include anthropogenic source and open fire source. For anthropogenic source, the assumption of geometric mean diameter of 100 nm is OK. But for open fire source, this assumed size is too small. The impact on aerosol number around forest region could be remarkable.

2. P24811, L3-4. Please double check it.

3. P24811, L22. Is it 890 m?

4. P24811, L25-27. GEOS-Chem includes vertical transport processes.

5. P24813, L1-20. The authors do not point out that coarse simulation shows better performance of capturing observed aerosol number concentration comparing to nest simulation. Could the authors give some explanations why coarse simulation is better than nest simulation at both surface layer and mountain peak height layer?

6. P24814, L23-24. I agree with the authors to use threshold temperature to determine whether air mass is from boundary layer or free atmosphere. But I disagree with the authors to use this kind of threshold temperature to filter simulated aerosol number from surface layer and mountain peak layer. One of the reasonable ways to my opinion is the authors can divide observed and simulated samples into BL condition and FA condition and then discuss about the results. GEOS-Chem includes upward and downward vertical transport processes. However, the key question is whether GEOS-Chem can capture upslope/downslope flows measured at Whistler Peak.

7. P24814, L26-28. What are the physical meanings of 275K threshold temperature for the 4x5 simulations and 279K threshold temperature for the 0.5x0.667 simulation?

References

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ACPD

15, C9037–C9040, 2015

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