

We would like to thank the anonymous reviewers for the overall positive and insightful comments on the manuscript. The original comments are in italics and the response to each comment is directly below the comment. We will submit a revised version of the manuscript and figures with the changes outlined below.

Response to review 2:

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.

While GEOS-Chem has vertical transport process, it cannot resolve sub-grid topographic effects, such as upslope winds. The air-mass characteristics at a mountain top do not always match the air-mass characteristics of the nearby region (but away from the peak) at the same altitude. This limitation is addressed in Yu and Hallar (2014), “One major issue is that the global model, with a horizontal resolution of $2^\circ \times 2.5^\circ$, is unable to resolve the subgrid-scale topography and processes. Actually, the model is unable to resolve Mount Werner on which SPL is located. The pressure of model surface layer in the SPL region is ~ 750 mbar, corresponding to that of Yampa Valley. The pressure of SPL is ~ 680 – 700 mbar, corresponding to the seventh model layer from the surface. In our comparison shown below, the modeled values in the seventh model layer are used to compare with the SPL measurements.”, although they do not attempt to parameterize topographic effects as we do here. However, these sub-grid topographic effects at SPL in GEOS-Chem may be different than Whistler in GEOS-Chem. SPL is 50–70 mbar above the modelled surface pressure in their simulations (7th model layer), while Whistler is ~ 150 – 160 mbar above the modelled surface pressure in our coarse simulations (12th model layer). SPL may simply be more frequently in the resolved modelled boundary layer relative to Whistler, and thus estimating upslope flow may be less necessary for capturing model-top aerosols at SPL.

Regarding, “*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.*”, we are not only trying to find the best annual size distribution as in Figure 5, we are trying to get the size distribution more correct at the *correct times* as in Figure 3. Our choice for the threshold temperatures were based on the time correlations (Table 3) based on Figure 3, and not on simply getting the best annual mean size distribution in Figure 5. It does work out, however, that the time-based best fit also greatly improves the annual-mean size distribution in Figure 5. Our work here is motivated by resolving the sources of the particles at Whistler (the second half of the paper). In order to do this, we needed to know at what times to sample the boundary layer vs. the free troposphere.

We have added a reference to Yu and Hallar (2014) in the introduction of our revised version: “While global models have been used to understand the processes shaping aerosols at mountain-top sites (e.g.

Yu and Hallar, 2014), these models have resolution too coarse to explicitly resolve topographic meteorology effects of many mountain peaks.”

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.

It is correct that the scaled Napari parameterization cannot be getting nucleation rates correct for the correct reason, and this should have been addressed in the paper. We have added the following text to the paper, “While these classical nucleation schemes do not get nucleation rates correct for the right reason, the scaled Napari scheme estimated nucleation rates within a factor of 5 and the annual number of nucleation days within 20% at 5 measurement sites in Westervelt et al. (2013).” We are unsure of the basis for the reviewer’s comment of “*One of the defects of the modified nucleation treatment in this work is that they predict too low nucleation rate within boundary layer.*”, and we have addressed it in our comment in the text.

We have added reference to Yu et al., 2010, “The choice of nucleation scheme impacts aerosol number concentrations, particularly at smaller sizes (Yu et al., 2010), and thus our choice nucleation scheme has some bearing on our results here.”

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.

We have modified the sentence to say, “The primary black carbon and organic carbon emission size distribution is assumed to be a lognormal distribution with a geometric mean diameter of 100 nm, consistent with field measurements of biomass-burning smoke within the first hour since emission (Janhäll et al., 2010), although larger diameters may be more appropriate for some fires once sub-grid aging has occurred (Sakamoto et al., 2015).”

We are currently working on parameterizations of aged biomass-burning size distributions that depend on fire and meteorological parameters, and we will update our biomass-burning size assumptions in GEOS-Chem once we have developed these schemes.

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

We reviewed the densities for black carbon and organic carbon in the model.

3. P24811, L22. Is it 890 m?

Good catch, yes it is 890 m.

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

We have added a sentence that says, “While GEOS-Chem does have vertical mixing for the resolved BL and synoptic/convective mixing between the BL and FT, it does not resolve sub-grid vertical transport due to topographic and upslope flows.”

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?*

We acknowledge that some of the metrics in the simulations without the filters applied are slightly better for the coarse simulations than those of the nested simulations. However, in all of these cases, the metrics are all extremely poor, and we felt that it was unnecessary to compare these simulations. We have included the following text to the manuscript: “Even though the metrics for the coarse simulations are slightly better in some cases than the nested simulations, all of the metrics were overall very poor.”

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.*

See our response to the reviewer’s first major point, above.

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?*

We have added a sentence to address this, “We note, however, that due to uncertainty in these best threshold temperatures from (1) different metrics giving best values at different temperatures and (2) model errors, there is likely no significance in the different threshold temperatures for the two different resolutions.”

Reference:

Janhäll, S., Andreae, M. O., and Pöschl, U.: Biomass burning aerosol emissions from vegetation fires: particle number and mass emission factors and size distributions, *Atmos. Chem. Phys.*, 10, 1427-1439, doi:10.5194/acp-10-1427-2010, 2010.