

Response to Anonymous Referee #3 comments on “Modeling particle nucleation and growth over northern California during the 2010 CARES campaign“ by A. Lupascu et al.

Summary:

In this manuscript, the authors present a modeling study using the WRF-Chem model v.3.5 to model the formation and subsequent growth of freshly nucleated particles observed during the CARES field campaign using various nucleation parameterizations. Notable model results include the observation that while nucleation and growth can account for up to 20 – 30% of CCN concentrations, the magnitude of the modeled CCN concentration is not very sensitive to the particular nucleation parameterization. Notable model developments include the extension of the aerosol microphysical model (MOSAIC) to include a nucleation mode at 1 nm and the use of budget diagnostic terms to investigate the impact of nucleation/growth on various processes (sources/sinks) that impact the aerosol number concentration. This manuscript is recommended for publication after the authors respond to the following minor questions/comments.

We thank the reviewer for reading our manuscript and providing constructive and valuable comments. Following the Reviewer’s suggestion, we revised the paper and have addressed all the concerns raised by providing response to individual comments below. Our responses are in blue.

Comments:

P. 19733, l. 7: Please include a citation to the work of (Ehn et al. 2014) in which the role of low volatility oxidized organic vapors in the condensational growth of nucleation mode aerosol in the ambient.

Following the reviewer’s suggestion we updated the sentence as it follows “Recent studies studies (e.g. Ehn et al., 2014, Jokinen et al., 2015) further demonstrate that the organic vapors with extremely low and low-volatility can enhance, or even dominate, the formation and growth of aerosol particles. Still, the role of organic vapors in particle nucleation and subsequent growth processes is quantitatively very uncertain.”

P. 19735, l. 10: Were there any measurements of SO<sub>2</sub> at the ground sites or on-board the G1?

SO<sub>2</sub> measurements were acquired at the T0 site and on-board of the G1 aircraft. The ability of the WRF-Chem model to reproduce this parameter was assessed in Fast et al. (2012) paper. A sentence was added to section 4.1 summarizing the model performance for SO<sub>2</sub>.

P. 19735, l. 26: Were any size distribution measurements made by the G1 FIMS (fast integrated mobility spectrometer) used in this study? To what extent do the FIMS measurements indicate the growth of nucleation model aerosol aloft?

We did not use the G1 FIMS data in this study. Since the aircraft rapidly passes through aerosol plumes and does not sample the same air parcel twice, the FIMS data would provide “snapshots” of the evolving size distributions, and it would be difficult to draw conclusions regarding aerosol growth over time from them. Therefore, we decided to focus on the aerosol distribution data from the ground sites in this study.

P. 19736, l. 2: While a CPC does measure particle concentrations through optical techniques, the cut-sizes for the CPCs mentioned refer to the particle Kelvin diameter, not the optical diameter. Please revise.

We deleted optical, thank you.

P. 19737, l. 7: To what extent do these modeled initial/boundary conditions for the aerosol size distribution (specifically the aerosol surface area that controls aerosol scavenging) agree with their observed ground/aloft counterparts?

The initial and boundary conditions for the aerosol size distribution are derived from monthly average aerosol mass mixing ratios simulated by the MOZART global model and the assumed bulk aerosol size parameters in Table 6 of Emmons et al., 2010. The MOZART global model was run at a coarse spatial resolution so it is unlikely to represent the spatial and temporal variability of the aerosols associated with the ground and aircraft measurements. However, local emissions and secondary aerosol processes substantially alter the aerosol size distribution as air moves from the boundary to the interior of the domain, and the T0 and T1 sites were about 500 km and 420 km from the model’s western and northern boundaries where low-level inflow normally occurred. We have not attempted to quantitatively assess the impact of the boundary conditions on the simulated size distribution in the boundary layer near the T0 and T1 sites, but we expect it to be fairly small.

P. 19739, l. 25: Please revise “. Metzger et al” to “, Metzger et al”.

Done.

P. 19741, l. 2 – 8: Please provide more detail regarding this "linear regression method" and why/how it was used to estimate growth times. Also, with a growth time estimate of 0.74 hours from 1 to 40 nm, does that indicate a growth rate of 52 nm/hr? Also, how do you account for the impact of time and size-dependent growth rate/scavenging in the Kerminen/Kulmala parameterization that itself assumes a constant growth rate?

The description of the KK2002 modifications was revised to provide more detail and be clearer: “In order to take account for coagulation losses during nuclei condensational growth from 1 nm to 40 nm in the 8 bin version, we applied the Kerminen and Kulmala (2002) parameterization (KK2002 hereafter). KK2002 estimates the growth time from the initial nuclei size to a larger size by assuming that the growth is due to H<sub>2</sub>SO<sub>4</sub> condensation only, and this was modified as follows to also account for condensation of organic vapors. In the 20 bin simulations at individual grid points and times, the growth time from 1 to 40 nm due to H<sub>2</sub>SO<sub>4</sub> condensation only and due to H<sub>2</sub>SO<sub>4</sub> plus organics condensation were estimated using the condensed masses from the MOSAIC aerosol chemistry module, and the ratio of these two growth times gave an organics enhancement factor for 1 to 40 nm growth (Y). The same calculations were done for growth from 40 to 63 nm giving another organics enhancement factor (X), and both X and Y were output. After the simulation, a zero-intercept linear regression of Y vs. X was performed ( $Y = aX$ ), using the entire X and Y data. In the 8 bin simulations, we calculated an organics enhancement factor for growth from 39 to 78 nm (X'), which is the width of first bin. We then estimated the organics enhancement factor for 1 to 40 nm growth as  $Y' = aX'$ , and applied this enhancement factor to the H<sub>2</sub>SO<sub>4</sub> only condensation growth time used in KK2002.

P. 19744, l. 8 – 12: To what extent is the lack of nucleation model aerosol at the smallest size bins in the SMPS due to transport of freshly formed aerosol from aloft down to the ground level? If nucleation is happening aloft, that could contribute to the result that the modeled number concentrations using the explicit nucleation schemes tend to over-estimate the measured number concentrations.

The CPC measurements suggest that the nucleation occurs throughout the PBL. However, we cannot quantify the impact of vertical mixing on the aerosol at ground level due to the fact that CPC measurements were acquired along the G-1 aircraft path, while the T0 and T1 sites are at a specific location. One would need continuous vertical profile measurements of aerosol size distribution to fully address this question. Yet, the analysis of the nucleation budget term (see Fig. 5 and the discussion related to it) suggests that freshly formed particles are transported to the surface by vertical mixing. Moreover, the highest concentration of particles detected at 13.6 nm at the T0 site and at 16 nm diameter at the T1 site suggests that also the horizontal transport from T0 to T1 might play a role on the model overestimation at the T1 site.

P. 19747, l. 25 – 28: How does the simulated H<sub>2</sub>SO<sub>4</sub> compare with the H<sub>2</sub>SO<sub>4</sub> proxy (calculated from measured SO<sub>2</sub>)?

Generally, the H<sub>2</sub>SO<sub>4</sub> proxy is higher than modeled H<sub>2</sub>SO<sub>4</sub>. Following the reviewer's question we have added the following paragraph in section 4.2: "Using Mikkonen et al. (2011) method, we calculated the H<sub>2</sub>SO<sub>4</sub> proxy concentration. The average daytime of observed H<sub>2</sub>SO<sub>4</sub> proxy is within a factor of 3 higher than the modeled H<sub>2</sub>SO<sub>4</sub>, it shows almost the same daily variability as the modeled H<sub>2</sub>SO<sub>4</sub> concentration, with a peak of 2.77e8 molecules cm<sup>-3</sup> at 1100 PST. Although the model overestimates the SO<sub>2</sub> concentration (NMB = 112%), the overestimation of modeled condensation sink by a NMB of ~40% in the same time suggests a large loss term for H<sub>2</sub>SO<sub>4</sub> vapors which might explain the differences between modeled and observed proxy concentration of H<sub>2</sub>SO<sub>4</sub>." We have also updated Figure 11 to include the H<sub>2</sub>SO<sub>4</sub> proxy.

P. 19755, l. 8 – 11: How do the observed/simulated CCN number concentrations compare with the measurements made in the study of (Mei et al. 2013) in which CCN activity of organic aerosol at the T1 site was studied?

Following the reviewer's suggestion we added the sentence as it follows: "Mei et al. (2013) performed size-resolved CCN measurements of 100-170 nm diameter particles at the T1 site, and they found that 90% or more of the size-selected particles were CCN active and had hygroscopicities between 0.10 and 0.21 (mean of 0.15). This suggests that the simulated low biases for 0.2% supersaturation CCN are not due to differences in simulated vs. observed mixing state and/or hygroscopicity."

P. 19760, I. 21: Do the authors mean “39 nm – 10 μm” for the size range of the default 8 size mode in MOSAIC?

[We corrected it, thank you](#)

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