Response to Anonymous Referee #1 comments on “Modeling particle nucleation and growth over northern California during the 2010 CARES campaign” by A. Lupascu et al.

This manuscript simulates atmospheric new particle formation (NPF) and growth using a numerical model and, based on comparisons between model simulations and observations, aims to get information about the dominant nucleation mechanisms in the study region. While the scientific problem tackled by the authors is extremely challenging, they do quite a good job in comparison with the few earlier attempts aiming to model regional NPF and growth. Therefore, I am in favor of accepting this work for publication in ACP after the authors have addressed the issues summarized below.

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

Major issue

While several nucleation parameterizations are tested, the paper lacks a proper sensitivity study on how their results depend on the nucleation rate and subsequent nuclei growth rate.

Concerning the nucleation rate, the authors used only single values for the empirical nucleation coefficients kACT, kKIN and kORG (those based on recommendation by Reddington et al. 2011), even though these coefficients have found to vary by 4-5 orders of magnitude between different studies at different sites. The authors should definitely make a couple of additional simulations to investigate the influence of varying the values of empirical nucleation coefficients.

We performed additional simulations in which the values of empirical coefficients k_{ACT}, k_{KIN}, and k_{ORG} were by decreased by one and two orders of magnitude. However, due to the expensive computational cost for the entire analysis period, these sensitivity tests were done only for the June 8, 2012 case. The results are presented in a new section, "4.8 Sensitivity to the empirical coefficients of the BL nucleation parameterizations."

Concerning both nucleation and nuclei growth rate, the treatment of contributing organic vapors (here OV) is far from clear, as different studies have defined "OV" in equation 3 (page 19740) in different ways. In the papers by Metzger et al. (2010) and Paasonen et al. (2010), OV was derived from the "unexplained growth" of nucleated particles, whereas Reddington et al. (2011) define OV as a certain fraction of the first-stage oxidation products of volatile organic vapours. The most recent study by Jokinen et al. (2015, PNAS 112, p 7123-7128) goes a bit further by tying OV to the concentration of ELVOC derived from laboratory experiments. The way OV is defined in this work differs from all the other prior studies relying on empirical nucleation parameterization. This issue should be discussed shortly in the paper and the challenges, or ambiguity, of defining OV should be explicitly brought up.

Following the comments from both Reviewer 1 and 2, we have added the following paragraphs in section 3. "Previous studies have defined NucOrg in different ways, and there is considerable uncertainty involving these low-volatility organic vapors. Metzger et al., (2010) assumed that the organic vapors involved in nucleation were the same as those involved in the initial condensational growth of the nuclei, and they derived NucOrg concentrations from laboratory experimental data and the initial growth rates. Redington et al., (2011) assumed that the organic vapors involved in NPF were the first stage oxidation (with O_3, OH, NO_3) products of monoterpenes (with a 13% molar yield) and treated them as non-volatile, The studies made Riipinen et al., (2011), Yli-Juuti et al., (2011) assumed that the organic vapors have very low vapor pressures. Using the TOMAS model, Pierce et al., (2011) performed several sensitivity studies to analyze the impact of organic vapors saturation pressure on the growth of nanometer particles and showed that ultrafine mode particle composition is dominated by low-volatility SOA species (those with C* less than 0.001-0.01 μg m^{-3}). Recently, Schobesberger et al., (2013), Ehn et al., (2014), Jokinen et al., (2015) used extremely low volatility organic compounds (ELVOC) formed as first stage oxidation products of monoterpene with different yields (1-17 %) to account for the role of organic vapors in the early stage of new particle formation.

In our model treatment, NucOrg consists of the organic vapors with the lowest C* of the VBS approach. This includes the C* = 0.001 µg/m^3 species from the non-traditional SOA precursors (semi-volatile and intermediate-volatility organics associated with fossil and biofuel combustion and biomass burning) and the C* = 0.1 µg/m^3 species from the traditional SOA precursors (isoprene, terpenes and aromatics) included in this study. Following Yli-Juuti et al. (2013), an upper limit of 10^8 molecules cm^{-3} for the NucOrg is used in Eq. (3). In our simulations, we find that during initial particle formation periods, 96-99% of the growth involves the C* = 0.001 µg/m^3 organic vapors, while the C* = 0.1 µg/m^3 species contribute on average ~7 times more to the ORG nucleation rate compared to those species having C* = 0.001 µg/m^3. The volatilities of some of these species may in fact be too high to actually participate in nucleation, in which case they can be viewed as proxies for the even lower volatility species that do participate." Note that OV has been replaced by NucOrg as suggested by Reviewer 2.

Other scientific issues:

The authors have quite a comprehensive introduction to the research topic, yet they miss several essential papers on atmospheric NPF and growth published during the past couple of years. Adding citations to at least a few of them would make this paper stronger than at present.

Following the comments from both Reviewer 1 and 2, we have added the following paragraph in section 1. "Westervelt et al. (2013) have the Goddard Earth Observing System global chemical transport model (GEOS-Chem) coupled to the TwO-Moment Aerosol Sectional (TOMAS) scheme to evaluate the performance of a ternary nucleation parameterization (Napari et al., 2002, with an added 10^{-5} nucleation tuning factor) and the ACT nucleation parameterization (Sihto et al., 2006). Using metrics such as nucleation rate, growth rate, condensation and coagulation sink, survival probability, and CCN formation they investigated the limitation of nucleation and SOA parameterizations at five locations in the various location and environments showing that, although the model gave reasonable results on average, the largest discrepancies between model and measurements were obtained using the ACT parameterization at the urban sites (up to a factor of 5 for the formation rate of 3 nm particles). Yu (2011), Riipinen et al., (2011), Pierce et al., (2011), and Patoulias et al., (2015) studied the impact of secondary organic vapor condensation and the average saturation concentration of these vapors on formation of new particles and their growth to larger sizes. They found that the condensation of these vapors can contribute to new particle formation as well to the growth of these ultrafine particles. A recent study of Yu et al. (2015) compared the Ion-Mediated Nucleation (IMN) mechanism and the organics mediated mechanism derived from the Cosmics Leaving Outdoor Droplets (CLOUD) chamber experiment (Riccobono et al., 2014) for several locations in North-America. It was shown that the frequency of nucleation and the intensity of NPF predicted by the organics-mediated mechanism was too high, while IMN parameterization was closer to the observed values, especially during the spring. That study suggested that the spatial and temporal differences in the behavior of the two nucleation parameterizations could be related to differences in the predicted aerosol first indirect radiative forcing, a lower concentration of organic compounds in the atmosphere compared to those used in chamber studies and the temperature influence on atmospheric nucleation rate compared to the derived empirical coefficient at a 278 K temperature and 39% relative humidity."

Model performance (section 4.1). While analyzed in a prior study, the authors could briefly summarize (with 1-2 sentences) how well the used model performs in simulating PM1 and PM2.5. Also, is the model performance for CN100 similar to that for PM1?

Following the reviewer suggestion we have added the following sentences :"Model performance in simulating PM_1 and $PM_{2.5}$ concentrations using the default WRF-Chem 8 size-bin and Wexler nucleation parameterization configuration (WEX-8BIN) for the entire CARES domain, including the T0 and T1 sites, has been presented in Fast et al. (2014). In general, simulated PM1 is fairly close to observed during 7 – 16 June 2010 both in terms of mass concentrations (NMB=15% at T0 and NMB=-18% at T1) and temporal evolution (R=0.56 at T0 and R=0.64 at T1). However, the WEX-8BIN simulation does not reproduce as well the CN100 concentration (NMB=-55% at T0 and NMB=-65% at T1) nor the temporal evolution (R=0.32 at both sites). ”

Nuclei growth (sections 4.2 and 4.3). How do the simulated nuclei growth compare with the measured one? This information would give some information on how well the model is able to simulation aerosol condensation growth, including concentrations of condensable vapors (sulphuric acid and low-volatile organic vapours).

Following the reviewer suggestion we have added the following paragraph in section 4.2 “We calculated the observed and modeled growth rate of 10-40 nm particles as follows. Following Jeong et al. (2010), the geometric mean diameters (GMD) of 10-40 nm particles were calculated from the size distributions during the period when growth after formation was observed/modeled. The growth rate was obtained by fitting the GMD trend during the growth period:

$$GR = \frac{\Delta GMD}{\Delta t} \tag{4}$$

Thus, we obtained at T0 a GR_{OBS}=2.57 nm h^{-1}, GR_{ACT}=1.43 nm h^{-1}, GR_{KIN}=1.14 nm h^{-1}, GR_{ORG}=1.71 nm h^{-1}, and at T1 a GR_{OBS}=3.69 nm h^{-1}, and GR_{ACT}=1.78 nm h^{-1}, GR_{KIN}=1.51 nm h^{-1}, GR_{ORG}=1.92 nm h^{-1}. This indicates that the simulated growth rate was always slower than observed, which could be due to an underprediction in the concentration of condensable vapors compared to those in the ambient air or to the current treatment of SOA that does not include changes to viscosity and/or effective volatility by particle-phase aging processes (Shrivastava et al., 2013; Zaveri et al., 2014).”

The budget terms for the aerosol number concentrations (section 4.4. and table 3) should be explained better in the text. The terms “condensation tendencies” or “combined condensation and coagulation tendencies” are misleading. For example, the second column in Table 3 seems to describe loss of particles due to condensation out of the size range 1-10 nm, the last column seems represent the source of 10-100 nm due to growth by coagulation and condensation from smaller sizes. All these terms should be properly explained in the paper.

We added the following sentences to explain these terms "These terms show how the particle number concentrations are affected by the different processes. The four terms for CN1-10 particles comprise a complete budget: gain due to nucleation, loss due to coagulation, loss due to condensational growth to sizes > 10 nm, and net transport plus deposition (advection, vertical mixing, and dry deposition). The single term for CN10-100 particles shows the net gain by condensational growth of smaller particles into this size range, modulated by coagulation loss of CN10-100 particles (which is significant on some days)" and we have also replaced the term "tendency" to "budget term".

Role of coagulation (table 3 and the text referring to this table). The authors discuss the relative roles of self-coagulation (coagulation between nucleation mode particles) and coagulation scavenging (coagulation of growing nuclei with larger pre-existing particles) in their cases. It seems to me that self-coagulation plays an important, or even dominant, role in the simulations involving organic nucleation (due to high nuclei concentrations), whereas in most other cases coagulation scavenging is probably more important.

This may be the case. However, all of the coagulation loss rates in Figs. 8-10 and 15-17 and Table 3 show combined loss from self-coagulation and coagulation with larger sized particles, and we did not diagnose self-coagulation separately. Thus we prefer to not comment on this point in the paper.

CCN production (section 4.6). The simulations underpredict CCN concentrations, especially at low supersaturations, despite overpredicting the nuclei number concentrations. Can the authors provide a reason for this feature? Could this underprediction be caused by too weak simulated growth of nucleated particles, or is it due to some other factor like problems in primary particle emission inventories? Finally, the authors could cite Sihto et al. (2011, ACP 11, p 13269-13285) somewhere in this section, since that is the longest observational study where NPF and growth has been linked with CCN measurements.

We have cited Sihto et al. (2011), and we added the following discussion of the underprediction of CCN concentrations at lower supersaturations. “Using the simulated size-dependent particle composition, we calculated particle sizes having critical supersaturations corresponding to the CCN measurement supersaturations. For supersaturations of 0.5%, 0.35%, 0.2%, and 0.1%, the diameters are 56, 78, 125, and 168 nm, respectively, with corresponding average hygroscopicities of 0.24, 0.23, 0.22, and 0.20. There is no clear distinction in these diameters between the T0 and T1 sites. The simulated CCN concentrations at 0.2% and 0.1% supersaturation thus correspond roughly to CN125 and CN168 concentrations. The simulated CN168 particle concentrations are biased high at T0 but low at T1 (NMBs of about 44% and -16%, respectively), which can explain the underestimations of 0.1% SS CCN at T1 but not at T0. The simulated CN125 have high biases at both T0 and T1 (NMBs of about 72% and 21%, respectively), so the cause of the simulations’ underestimations of 0.2% SS CCN is not clear. 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”

The uncertainties associated with the presented analysis should briefly be summarized also at the end of section 5.

Done

Technical issues:

Page 19736, lines 6-7: What is the surrounding region of California, i.e. how large is the model domain?

We explain this by the following sentence: “The model domain covers the area between 32.2 to 42.7^{O} N, and 127.5 to 113.4^{O} W, which encompassed all of California and Nevada and extended about 400 km into the Pacific (west of San Francisco), using a 4-km grid spacing and 65 vertically-stretched layers from the ground up to 50 hPa.”

Page 19743, lines 5-6: should it be "in" or "By" Figs. 1 and 2.?

We corrected, thank you.

Page 19756, line 27: "Kuata 2008" is missing from the reference list. Should it be "Kuwata 2008"?

Thank you for your observation. Indeed, it is “Kuwata” and we added the reference.

The manuscript contains a very large number of figures. Some of them, for example all the figures showing time evolution of the aerosol number budget terms (figures 8-10 and 15-17) could be moved into an appendix.

While we understand there are added a large number of figures to support the discussion in the text, we would prefer to leave them in the main text. The discussion and references to the figures would not change if we adopted an appendix, and if we did, readers would have to go back and forth from the main text to the appendix.

Figures 13 and 14: Is the something wrong in the scale of the observed BLH in panels a? I do not see any values of this quantity in these figures.

We thank the reviewer for pointing this out. The observed boundary layer heights for this day were accidently left out and now have included them in the revised manuscript.

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