

Response to Anonymous Referee #2 comments on “Modeling particle nucleation and growth over northern California during the 2010 CARES campaign” by A. Lupascu et al.

In paper explores WRF-Chem modelling of nucleation and growth during the CARES campaign. The authors evaluate the model against observations using three different nucleation schemes and several different bin structures. The topic and overall approach fits with ACP; however, I feel that there are some areas where I feel the paper needs some improvement before being published in ACP.

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.

Evaluation of condensation and coagulation sink: Evaluating and comparing nucleation schemes is a convoluted task when one does not know if (1) the condensation/coagulation sinks are correct and (2) the chemical production of low-volatility vapors are correct. Statements about “adjusting empirical coefficients” of the nucleation schemes (e.g. P19761 L4) are baseless if these other factors impacting nucleation/growth are not evaluated. It seems like you do not have measurements of the gas-phase vapors involved in nucleation and growth, so only #1 is possible, but this needs to be done. One cannot evaluate a nucleation scheme with any confidence without knowing if the other factors that affect nucleation and growth are correct.

Taking into account the reviewer’s comment, we have added the following discussion at the end of Section 4.1: “New particle formation depends on the concentrations of low-volatility vapors involved in nucleation and the initial growth of the nucleated particles and the coagulation losses of new particles during their initial growth. The low-volatility vapor concentrations depend on their photochemical production and condensational loss to particles. When evaluating and comparing nucleation schemes, it is useful to compare measurement-based and simulated estimates of these sources and sinks. The concentrations and photochemical production of low-volatility vapors was not measured, but we can compare modeled and observed precursor gas (i.e., SO<sub>2</sub>) concentrations. For

the entire period, the modeled SO<sub>2</sub> concentrations at T0 were fairly close to observed, with NMBs of 30% , but there was poor agreement in temporal variability (R=0.30). The coagulation sinks for 1 nm particles and the condensation sinks for H<sub>2</sub>SO<sub>4</sub> and at T0 and T1 were calculated (offline) using observed and simulated size distributions of 10 nm – 1 μm particles. The temporal variability of the coagulation and condensation sinks is fairly well reproduced (R between 0.67 and 0.76). At T0, the simulated coagulation and condensation sinks were about twice those calculated using observations (NMBs of 94% and 106%), while at T1 the coagulation and condensation sinks had NMBs of 35% and 40%. These biases are generally consistent with the CN100 biases at the two sites. The high biases for the condensation and coagulation sinks would tend to give lower new particle formation under the simulation conditions compared to observed conditions, and thus cannot account for the higher simulated NPF (e.g., CN10) in the simulations compared to observations. Also, differences among the simulations with different nucleation parameterizations for SO<sub>2</sub> concentrations and the two sink terms were small, so biases in them compared to the observed (or observation-based) values should have had similar impacts on NPF in all three simulations. “

Literature review: The literature review seems (to me at least) to be selective both in terms of what papers are cited as well as what info is taken from the papers (or information being incorrect). I give specifics below.

Bin structure matters more than having nucleation turned on/off: Why do CCN change by more when changing the bin structure but keeping the nucleation scheme constant (see tables 7 and 8... CN100 going from NMB of 78% to -58% with the same nucleation scheme... a factor of 3 change in CN100) than by shutting nucleation off entirely (“20-30% changes”)? This seems odd to me, and I think it deserves to be explored and explained.

Specific comments:

P19730 L22-26: There is a mix a verb tenses here (were vs. are).

Corrected.

P19731 L4-7: This sentence is confusing without any figures and other discussion to add context. Could remove it or replace it with a simpler sentence about differences in N40-100 being minor between simulations with different nucleation schemes.

We have removed this sentence.

P19733 L1: Please specify that the Kirkby paper explored ternary nucleation where Ammonia was the ternary species. This distinguishes from ternary nucleation with other ternary species (e.g. dimethyl amine).

Done.

P19733 L10-21: Why is there no discussion about how the Napari scheme correctly predicts nucleation vs. no nucleation on all of the 10 days explored in Jung et al. (2008), outperforming all other schemes (at least in terms of predicting the occurrence of nucleation...not necessarily rates)? This discussion seems arbitrarily selective.

We have added the following discussion regarding the Napari scheme. “They showed that most of the nucleation schemes included in their study had difficulty reproducing the observed events, except for the ternary mechanism of Napari et al. (2002) that predicts the occurrence of the events during the analyzed period. The empirical particle formation mechanism (Sihto et al., 2006) performed well on 70% of the analyzed days, but still it predicted nucleation events on days when nucleation was not observed.”

P19734 L1-2: Jung et al. (2010) uses Napari (scaled down by 1E-5) rather than Merikanto.

Thank you for correcting our mistake, we replaced Merikanto to Napari.

P19733-19734: Westervelt et al. (2013) (ACP, 13, 7645-7663, doi:10.5194/acp-13-7645-2013, 2013) evaluates a model’s ability to predict the occurrence and rates of nucleation events at 5 different locations for 2 nucleation schemes and belongs in this discussion. Yu et al. (2015) (ACPD, 15, 21271-21298, doi:10.5194/acpd-15-21271-2015, 2015) performs a similar analysis and should also be discussed.

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-Mediation 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.”

P19739 L8-11: The Spracklen and Reddington papers (not sure about Matsui) did not use Wexler in the FT. Rather, they used the binary nucleation scheme of Kulmala et al. (1998) that explicitly predicts a nucleation rate (rather than Wexler that takes all excess

H<sub>2</sub>SO<sub>4</sub> and forms new particles with it). Wexler has the limitation that when predicting formation \*rates\* by taking all excess H<sub>2</sub>SO<sub>4</sub> into new particles, the rates can easily exceed the barrierless nucleation rate (i.e. the time it takes sulfuric acid molecules to find each other).

We have followed the approach of Matsui et al., (2011) and have used the Wexler scheme in the free troposphere. To avoid any confusion we have changed the sentence to “These parameterizations are used in the boundary layer, while the Wexler parameterization is applied in the free troposphere. This approach was tested in previous global and regional modeling studies (e.g. Spracklen et al, 2006, Matsui et al., 2011, Reddington et al., 2011)” to “These parameterizations are used in the boundary layer, while following Matsui et al. (2011) the Wexler parameterization is applied in the free troposphere.”

P19740 L3 and throughout: I’d recommend using LVOV (low-volatility organic vapors) or similar (maybe org\_nuc as used in some papers) rather than OV (which could lead people skimming the paper to think it refers to all organic vapors).

We agree this is a more appropriate abbreviation and have changed the OV to NucOrg, which is used in Metzger et al. (2010).

P19740 L6: This range of saturation vapor concentrations ( $C^* = 0.1 - 0.001 \text{ ug m}^{-3}$ ) is very likely too high to be contributing to nucleation (unless they undergo some reaction decreasing their volatility in the cluster). Pierce et al. (2011) (ACP, 11, 9019-9036, doi:10.5194/acp-11-9019-2011, 2011) showed that organics with vapor pressures higher than  $0.001 \text{ ug m}^{-3}$  should not even contribute to the growth of new particles, let alone stabilize the cluster. Further, the EL-VOCs found in recent experiments (Ehn et al., Nature, 506, 476–479 doi:10.1038/nature13032, 2014) thought to contribute to nucleation and early growth have volatilities less than  $C^*=0.001 \text{ ug m}^{-3}$ . Please add some discussion saying that you are using these species as a proxy for nucleating organics, and that it’s unclear if they could directly contribute to nucleation.

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<sub>3</sub>, OH, NO<sub>3</sub>) 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<sup>-3</sup>). 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<sup>3</sup> 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<sup>3</sup> 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<sup>8</sup> molecules cm<sup>-3</sup> 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<sup>3</sup> organic vapors, while the C\* = 0.1 μg/m<sup>3</sup> species contribute on average ~7 times more to the ORG nucleation rate compared to those species having C\* = 0.001 μg/m<sup>3</sup>. 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.”

P19740 L8: Yli-Juunti should be Yli-Juuti

Corrected.

P19748 L2-4: Shouldn't the lifetime of OV and H<sub>2</sub>SO<sub>4</sub> in the vapor phase be essentially the same (i.e. determined by the condensation sink).

Taking into account the reviewer's comment, we have revised the discussion of H<sub>2</sub>SO<sub>4</sub> and OV (now NucOrg) diurnal patterns as follows: "Although both H<sub>2</sub>SO<sub>4</sub> and NucOrg are formed by oxidation of precursor gases by OH radicals, and hence are linked to solar radiation that regulates the intensity of photochemical reactions, their different diurnal patterns can be related to the diurnal patterns of their respective precursor gases. The anthropogenic organic vapor with the C\* = 0.1 µg/m<sup>3</sup> has the highest contribution to the NucOrg vapors concentration at T0, and their concentrations start to increase at 1300 PST and reach a maximum around 1800 PST. Their aromatic precursor gas has a similar temporal pattern during the afternoon, while SO<sub>2</sub> is decreasing during this period (not shown)."

P19748 L16-19: Westervelt et al. (2014), referenced later in the manuscript, explores the relationship between how changing the nucleation rate affects the condensational growth and coagulation loss rates, and in turn how the CCN concentrations are strongly dampened to changes in the nucleation rate. It would be appropriate to compare/discuss this paper here.

We have added the following discussion: "This is in agreement with the Westervelt et al. (2014) study with the GEOS-Chem-TOMAS modeling system, which showed that CN10 concentrations are more sensitive to BL NPF parameterizations than CCN-sized particle concentrations, consistent with the loss of newly formed particles through coagulation. Moreover, they investigated the nucleation rate and gas condensation sinks impact growth rate, coagulation sink, and survival probability, and showed that the relatively small differences in the predicted BL CCNs concentration are due to a strong damping effect. This might also explain the relative insensitivity of CN40 and CN100 to choice of NPF mechanism in our study."

P19750 L11-13: Even in unstable boundary layers, the particles are still generally constricted to the boundary layer, it's just that the boundary layer is generally deeper.

To avoid any confusion, we replaced “constricted to the boundary layer” with “constricted near the surface” to emphasize that we are discussing the transition in the boundary layer structure around sunset.

P19750 L16: With lower windspeeds, fewer particles are also transported \*in\*, so how windspeed affects concentrations depends on what air mass is upwind relative to there being a net production/loss locally. If there air mass upwind is clean and there is a local net production of particles, then a lower windspeed would cause an increase in particles.

Following the reviewer recommendations, we have rephrased this as follows: “With lower wind speeds, we have a reduced effect of horizontal transport and vertical mixing on the 1-10 nm, and 10-40 nm range particles (Figs. 15b, 16b). “

P19754 L25-27: Westervelt et al. (2014) (already cited in paper) goes into details of the processes controlling CCN formation from new-particle formation events. Kuang et al., Determination of cloud condensation nuclei production from measured new particle formation events, GRL, (2009) is worth discussing here too.

Following the reviewer recommendations, we have added the following. “Kuang et al., (2009) quantified the role of self-coagulation loss (up to 20%), coagulation (up to 10%), condensation on pre-existing aerosols (up to 30%), and condensation on nucleated particles (up to 80%) to the CN100 particles concentration using field study data acquired at three North American locations for twenty CCN formation events. They showed an enhancement on average by a factor of 3.8 of the pre-existing CCN number due to NPF. Using the GEOS-Chem-TOMAS model, Westervelt et al., (2014) calculated particle growth rates, condensation sinks, coagulation sinks, survival probabilities, and CCN formation rates for eight different nucleation parameterization. Their study showed that increases in the nucleation rate led to decreased survival probability, so that the CCN number concentrations are relatively insensitive to the nucleation mechanism.”

P19755 L19-21: What Kappa (hygroscopicity) values do these correspond to (i.e. a critical diameter of 55 nm for 0.5% supersaturation corresponds to a kappa of XX)

We have changed the sentence to contain information about kappa as follows: “ 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 (kappas) of 0.24, 0.23, 0.22, and 0.20. There is no clear distinction in these diameters between the T0 and T1 sites. “

P19756 L21-23: This sentence isn't necessarily true. It is possible to have a system where 100% of the particles are created by nucleation and growth, but be very insensitive to reasonable changes in the nucleation rate. This occurs when the system is saturated with respect to nucleation (more nucleation = more competition for condensible vapors = slower growth = lower survival probability). See Westervelt et al., 2014. Of course what you suggest is also true: if most of the CCN are due to primary emissions, the system also won't be very sensitive to nucleation rates.

We rephrased the text, taking in account the reviewer's comment as follows: “However, the ACT, KIN, and ORG parameterizations give fairly similar results, especially for low supersaturations, suggesting that CCN at these supersaturations (and sizes) are less sensitive to the nucleation mechanism and rate, possibly due to a slow growth rate of freshly nucleated particles to these sizes and low survival probability (Westervelt et al., 2014), or that the concentrations of these are mainly driven by other processes (e.g., primary aerosol emissions).”

Section 4.7: Lee et al. (2013) (Representation of nucleation mode microphysics in a global aerosol model with sectional microphysics, *Geosci. Model Dev.*, 6, 1221-1232, doi:10.5194/gmd-6-1221-2013, 2013) explored the how model predictions change due to different start diameters for the size bins and the use of the Kerminen and Kulmala scheme, similar as to what was done here. It would be worth comparing with their results.

We have added the following discussion that compares our results with Lee et al. (2013). “In our simulations, the ACT-8BIN configuration (which does not explicitly treat initial growth and loss of new particles) gives higher CN40 concentrations compared to the

ACT-12BIN and ACT-20BIN configurations. This result is counter to the Lee et al. (2013) study that used the TOMAS model with lowest bin diameters of 1 (explicit treatment of initial growth and coagulation loss) and 3 and 10 nm (Kerminen and Kulmala (2002) parameterization) with 40, 36, and 30 size bins, respectively., 3, and 10 nm. They found that the CN10 particles were overpredicted when the Kerminen and Kulmala (2002) parameterization was used. Several factors may explain the differences between Lee et al. (2013) and our study: the nucleation parameterizations (binary/ternary vs. activation type mechanism), lower size bin (3/10 nm vs. 40 nm), analysis period (average over the spring season vs. 10 days in summer), horizontal resolution ( $4^{\circ}$  latitude x  $5^{\circ}$  longitude vs. 4 km), emissions inventory, distribution of primary aerosol particles, gas-phase photochemistry, and the estimation of growth time from 1 nm to 40 nm.”

P19757 L18-19: “We speculate that...more dependent on condensational growth than coagulation loss.” I’m not sure I understand. The survival probability to CCN sizes depends on both the condensational growth rate and the coagulation loss rate (see Kuang et al., GRL, 2009 and Pierce and Adams, Efficiency of cloud condensation nuclei formation from ultrafine particles, Atmospheric Chemistry and Physics, 7, 1367-1379, 2007). Changes in either will affect the survival probability, thus they both always matter. Did you mean to say that you suspect that the condensational growth is fast enough relative to the coagulation loss rates such that nearly all of the nucleated particles survive to become CCN?

This sentence was removed. The differences in CN40 but not CN100 between WEX-8BIN and ACT-8BIN could be due to several reasons, and a detailed analysis of the rather dated Wexler scheme vs. the newer scheme is outside the main objectives of the paper.

P19757 L20-24: If the simulations with the lower number of bins outperform the simulations with the higher number of bins, the lower number of bins must be getting things right for the wrong reasons (i.e. compensating for some other error in the model). Having a finer bin structure will more-accurately simulate the aerosol dynamics (assuming the same numerical technique for both).

We explain the observed differences through the following paragraph. “The reduced overestimation of CN40 particles by more than 40% could at least partially be explained by the differences in the bin boundaries: the simulated CN40 are calculated as particles larger than 39.8 and 46.4 nm for the 20 and 12 bin configurations respectively (i.e., no interpolation is used).. For CN100 particles the lower bin edge is at 100 nm for both ACT-12BIN and ACT-20BIN, so the lower CN100 (and CN40) with ACT-12BIN suggests that transfer of particles to larger sizes during condensational growth is somewhat slower with the coarser size resolution.”

P19758 L13: I don't think the word “producing” is the best word to use here since it implies that it is changes in production rather than loss (e.g. deposition) that is responsible for all of these changes.

We have changed the word “producing” to “leading”.

P19758 L17-20: I have a couple questions about this sentence... “enhancement of gas-particle partitioning”... did you mean to say “an enhancement of partitioning of vapors to the particle phase”? Is it really increased surface area (e.g. the condensation rates are faster) or is it increased mass (e.g. shifting equilibrium towards the particle phase)?

Indeed, it was the “an enhancement of partitioning of vapors to the particle phase”. And both factors mentioned are responsible, the increase of total mass together with the increase of surface area. We obtained an averaged CS of  $7.57e-4 \text{ s}^{-1}$  for ACT-20BIN simulation compared to  $5.09e-4$  for ACT-8BIN at the T1 site and of  $8.09 e-4$  for ACT-20BIN compared to  $5.34e-4$  for ACT-8BIN.

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