Response to Anonymous Referee #1

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:

$$GR = \frac{\Delta GMD}{\Delta t} \qquad (4)$$

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.

Response to Anonymous Referee #2

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_2) concentrations. For the entire period, the modeled SO_2 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_2SO_4 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_2 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

H2SO4 and forms new particles with it). Wexler has the limitation that when predicting formation *rates* by taking all excess H2SO4 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 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 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 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_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.”

P19740 L8: Yli-Juunti should be Yli-Juuti

Corrected.

P19748 L2-4: Shouldn't the lifetime of OV and H2SO4 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_2SO_4 and OV (now NucOrg) diurnal patterns as follows: "Although both H_2SO_4 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^3 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_2 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 the 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 airmass is upwind relative to there being a net production/less locally. If there airmass 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 (hygroscipicity) 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, respecitively., 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^O latitude x 5^O 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 coagulational 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 coagulational 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 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.

Response to Anonymous Referee #3

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 bellow. 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 SO2 at the ground sites or on-board the G1?

SO_2 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_2.

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_2SO_4 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_2SO_4 condensation only and due to H_2SO_4 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_2SO_4 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 H2SO4 compare with the H2SO4 proxy (calculated from measured SO2)?

Generally, the H_2SO_4 proxy is higher than modeled H_2SO_4. 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_2SO_4 proxy concentration. The average daytime of observed H_2SO_4 proxy is within a factor of 3 higher than the modeled H_2SO_4, it shows almost the same daily variability as the modeled H_2SO_4 concentration, with a peak of 2.77e8 molecules cm^{-3} at 1100 PST. Although the model overestimates the SO_2 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_2SO_4 vapors which might explain the differences between modeled and observed proxy concentration of H_2SO_4." We have also updated Figure 11 to include the H_2SO_4 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, l. 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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