

Interactive comment on “Modeling particle nucleation and growth over northern California during the 2010 CARES campaign” by A. Lupascu et al.

Anonymous Referee #2

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Review of “Modeling particle nucleation and growth over northern California during the 2010 CARES campaign” by 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.

Evaluation of condensation and coagulation sink: Evaluating and comparing nucle-

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ation 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.

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).

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.

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).

P19733 L10-21: Why is there no discussion about how the Napari scheme correctly

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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.

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

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.

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).

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).

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 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

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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.

P19740 L8: Yli-Juuti should be Yli-Juuti

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).

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.

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.

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.

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.

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)

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 insen-

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sitive to reasonable changes in the nucleation rate. This occurs when the system is saturated with respect to nucleation (more nucleation = more competition for condensable 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.

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.

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?

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).

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

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responsible for all of these changes.

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)?

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 19729, 2015.

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