

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

Anonymous Referee #1

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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.

Major issue:

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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 k_{ACT} , k_{KIN} and k_{ORG} (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.

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.

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.

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

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PM1 and PM2.5. Also, is the model performance for CN100 similar to that for PM1?

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 simulate aerosol condensation growth, including concentrations of condensable vapors (sulphuric acid and low-volatile organic vapours).

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.

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.

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.

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

Technical issues:

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

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

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

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.

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

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

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