

Interactive comment on “Analysis of nucleation events in the European boundary layer using the regional aerosol-climate model REMO-HAM with a solar radiation-driven OH-proxy” by J.-P. Pietikäinen

Anonymous Referee #1

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Review of “Analysis of nucleation events in the European boundary layer using the regional aerosol-climate model REMO-HAM with a solar radiation-driven OH proxy” by Pietikaninen et al.

This manuscript assesses the ability of the REMO-HAM regional climate model with online aerosol microphysics to predict nucleation events in Europe against measurements. Previously, the REMO-HAM model used fixed monthly OH fields (with a fixed diurnal profile) and the authors here make OH radiation-dependent, which leads to

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improved results in predicting nucleation event diagnostics.

Overall I feel that the paper could be published in ACP, but there are several areas where it should be improved first.

General comments:

- Many global and regional chemical transport and climate models with online aerosol microphysics include prognostic predictions of OH that depend not only on radiation but also on NO_x and VOC (e.g. monoterpenes, isoprene, propene) concentrations (e.g. WRF-Chem, PMCAMx-UF, GEOS-Chem-APM, GEOS-Chem-TOMAS). In particular, OH is strongly dependent on NO_x concentrations, and in general OH will be a factor of 2 or more higher in moderately polluted regions compared to clean regions. However for very polluted regions, OH will be generally lower than in moderately polluted regions. There is insufficient discussion of the NO_x and VOC dependencies of OH in the text. This could lead to potentially large differences in OH between Hyytiälä, Melpitz and SPC.

There is a parameterization in Stevens et al. (2012) (<http://www.atmos-chem-phys.net/12/189/2012/acp-12-189-2012.html>) for OH as a function of radiation, NO_x and high vs. low VOCs (equations A1 vs. A6) which the authors could implement in the future. It would require at minimum adding NO_x to simulations, but the dominant loss of NO_x is by reaction with OH, so this might not be too difficult.

- With no SOA in the model, the condensation sinks will be biased low (and the authors mention in the text the lack of D_p > 100 nm particles because of the lack of SOA). If nucleation rates depend only on H₂SO₄ concentrations, and H₂SO₄ concentrations are inversely proportional to the condensation sink, nucleation rates in this paper should be biased high compared to simulations with SOA. Yet, the improved REMO-OHP simulations were already biased low for J3s without adding the SOA. These J3s may be quite a bit (factor of 2?) lower if SOA is added, particularly if “anthropogenically enhanced” SOA (see <http://www.atmos-chem-phys.net/11/12109/2011/acp-11-12109->

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2011.html) is added. Note the drop in H₂SO₄ concentrations going from biogenic-only SOA to adding anthro-enhanced SOA in Figure 4 of <http://www.atmos-chem-phys.net/13/11519/2013/acp-13-11519-2013.html>... these drops in H₂SO₄ will be even more substantial when starting from no SOA.

The discussion of lack of SOA and condensation sink needs to be extended in the paper to be more than just a reason for why nucleation events continue longer than observed (there are many more implications than this).

- In general, the model evaluation in the paper could be stronger if a more holistic view of the aerosol size distribution are used. E.g. how do the different modes of the size distribution compare to measurements and how do these modes change due to changes to the model? There is a lot more that can be learned about the model predictions from this than focusing only on nucleation rates and duration (and as stated in the last point errors in the size distribution will feed back as errors in nucleation rates). It would be substantial work to change the focus of this paper to add evaluation of overall aspects of the size distribution, so I don't think it is necessary here for publication, but I ask the authors to strongly consider this approach for future model evaluation and papers.

Specific comments

P8917 L22: "everywhere in the atmosphere" do you mean to say troposphere here rather than atmosphere? I don't know much about nucleation in the upper atmosphere.

P8917 L24: "local CCN concentrations", nucleation can impact CCN concentrations far away from the place of nucleation. See Merikanto 2009 (cited in manuscript) where FT nucleation leads to a large fraction of the nucleation impact on BL CCN concentrations.

P8918 L3-14: It looks like this paragraph is a rather complete list of the nucleation schemes typically used in large-scale models. Yu's IMN is missing though (<http://onlinelibrary.wiley.com/doi/10.1029/2009JD012630/abstract>)

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P8919 L10-14: Has anyone quantified how sensitive the nucleation mode is to grid spacing?

P8920 L8-9: What is the rationale for having kinetic nucleation outside of the BL? My general understanding is that organics in the BL contribute to the kinetic functional dependence of continental BL nucleation and that the kinetic scheme might fail outside of the continental BL.

P8922 L26-28: "The dependence of OH on reactants such as NO_x, hydrocarbons... is condensed into the single per-exponential coefficient." "a" is a constant, so by definition there is *no* dependence on the reactants. You are stuck with whatever the mean reactant conditions were during the Mikkonen study, and you apply these everywhere. I assume that the old method of using monthly mean OH concentrations has different OH concentrations at different locations based on variance in monthly mean cloud cover, NO_x and VOCs. The new method accounts for instantaneous changes in cloud cover (radiation), but no changes in NO_x and VOCs, so an improvement in one aspect (radiation) and a regression in another aspect (NO_x and VOC effects).

P8924 L8-10: Again, is it justified to apply kinetic nucleation in the free troposphere?

P8925 L15-17: What are the units here? 2000 cm⁻³ s⁻¹? cm⁻³ hr⁻¹? cm⁻³ event⁻¹? dN/dlogDp usually has units of cm⁻³, but what are the time units (note, "event" isn't really a time, cm⁻³ event⁻¹ isn't really a rate).

Section 3: Have the authors done an analysis of what fraction of the days where nucleation was observed where the model correctly predicted nucleation, and what fraction of the days where there was no nucleation observed that the model correctly predicted no nucleation (e.g. true positives, false positives, true negatives and false negatives)?

Figure 3: The colors of the 2 simulations are hard to tell apart.

P8928 L20 and y-axis in Fig 4: "fractions of nucleation days", what does this mean?

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Do you mean the fraction of days that have nucleation? Please make more clear.

P8932 L15: "emissiona" should be "emissions"

Figure 7 and section 3.4: What vertical level is the size distribution panels taken from, the surface? Are the vertical profiles of aerosol number concentrations that useful? Is there any way to test how good the vertical predictions are? I especially find them concerning because of the use of kinetic nucleation outside of the BL.

P8935 L2: When I see the word "downdraft" I think of convection, are these convectively driven downdrafts? I don't think REMO-HAM would resolve them. It's more likely large-scale subsidence (or a change in aerosols w/ height due to horizontal convection over the measurement site).

P8935 L6-10: I highly doubt that the transport of H₂SO₄ vapor in convective clouds is causing much FT nucleation, it's condensational lifetime onto aerosols is generally on the order of minutes. I'd bet that SO₂ transport by convective clouds (and subsequent oxidation in the FT) is the main contributor to your FT nucleation.

Figures 8, 9 and section 3.5: Figures 8 and 9 don't show us anything about the spatial extent of nucleation events (i.e. how large an area do nucleation events occur across?). Because the authors are averaging across many nucleation events (that take place in different places with different spatial extents), Figures 8 and 9 do not show us how spatially large nucleation events generally are, they just show the mean nucleation rates over Europe. Figure 10 shows some representation of the spatial extent of events (though only from 6 cases). Please update the section name and text to be more consistent with what the figures show.

Figure 11 and section 3.6. In the text and in the y-axis label, it says this is a production, but the units are given as #/m², which is a column burden. The caption also says it is a 3 nm particle burden. Is this a column production rate or a column burden?

P8939 L8-10: This last sentence doesn't make much sense. Are you saying that NPF

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plays an important role in nucleation events? Seems circular.

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