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Atmospheric Chemistry and Physics Discussions

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

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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 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.

We thank the reviewer for valuable comments for improving our manuscript. Throughout the text reviewers comments are marked with boldface and after each comment follows our reply.

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 NOx 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 NOx concentrations, and in general OH will be a 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 NOx and VOC dependencies of OH in the text. This could lead to potentially large differences in OH between Hyytiala, Melpitz and SPC. There is a parameterization in Stevens et al. (2012) (http://www.atmos-chemphys.net/12/189/2012/acp-12-189-2012.html) for OH as a function of radiation, NOx and high vs. low VOCs (equations A1 vs. A6) which the authors could implement in the future. It would require at minimum adding NOx to simulations, but the dominant loss of NOx is by reaction with OH, so this might not be too difficult

Thank you for this good point. As the aim was to modify the simple chemistry routine, we did not include analysis of other species. A better representation of the chemical species would be without a doubt an interesting modification to our model (actually some work has been done to couple an online chemistry model with REMO-HAM). The idea of including Stevens et al. (2012) parameterization is also interesting and will be considered in the future.

For the discussion, we have added new chapter to the end of section 3.2 “The simplified sulphate chemistry module can be one reason for the continuation of events. The OH-proxy is based on measurements from Hyytiälä, which means that the influences of other relevant chemical species to OH concentrations are based on Hyytiälä conditions. For example, nitrogen oxide (NOx) and volatile organic compound (VOCs) are two competing species for the reaction with OH producing eventually ozone (Seinfeld and Pandis, 1998). The VOC/NOx ratio tells which species is predominant in the reaction. As this is now implicitly included in the proxy through measurements from Hyytiälä, error may be caused in environments where typical VOC/NOx ratios differ from those in Hyytiälä. This will impact the H2SO4 concentrations and could partially explain why the J3nm values have different bias in Fig. 2 and why the length of events is not captured in Fig. 3.”

- With no SOA in the model, the condensation sinks will be biased low (and the authors mention in the text the lack of Dp $>$ 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 will 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-11-12109-2011.html) is added. Note the drop in H2SO4 concentrations going from biogeniconly

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).

This is a good point. Nevertheless, the condensation sink increases when the number of large particles increases, which means that the effect is not significant during the “main event time” (morning; measurement show low condensation sink during this time). This is why we believe that our nucleation rates would not drop that much. Of course, if the areas of nucleation event already has larger particles during the start of the event, then the situation changes as you mentioned. This is very rare, even in places like Po Valley (again, based on our measurements). So overall, including the SOA in our model would lead to higher growth rates, thus increasing the condensation sink and lead to shorter nucleation event (as we speculated), but would not affect the nucleation rates during the event(s) that much.

We have added the following text: “On the other hand, higher condensation sink would lead to lower H2SO4 concentrations and decrease the J3nm values. This effect, however, would not be very strong, because the nucleation event usually starts when the air is clean (measurements show low condensation sink) and during this time H2SO concentrations would stay almost as high as without the SOA growth. This leads back to the point that nucleation events would be shorter with SOA in the model due to increasing condensation sink and faster depletion of H2SO4 as the events progress.”

- 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.

We have used this approach in our previous study with REMO-HAM (http://www.geosci-model-dev.net/5/1323/2012/gmd-5-1323-2012.html). There, we showed how the high SO2 bias (with the missing SOA) lead to unrealistically high nucleation mode concentration. This was actually one of the "motivation points" for this study. Yes, we could have included similar analysis to this work, but the decision was to focus more on the nucleation (and related) results. In future, we will add SOA production in the model and study the evolution of the size distributions.

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."

Yes that is true, we changed it to troposphere.

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.

Also true, we changed it to "global and local" and added the reference of Merikanto 2009 also here.

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)

We added Yu's IMN to the list.

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

Not that we know of.

P8920 L8-9: What is the rational 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.

The aim was to include nucleation throughout the troposphere. It is true that this can be a source of error, but what are the mechanisms at higher altitudes? There is indication that classical sulphuric acid–water nucleation rates are in accord with available observations above 4 km. However, we do not think that very general conclusion can be made. Our approach may not be perfect, but as we are focusing on BL nucleation, we think it is adequate.

We added to the “Nucleation scheme” chapter (last paragraph) a sentence “As the nucleation mechanism(s) at higher altitudes are unknown, this approach may generate some error. However, our focus is on boundary layer nucleation, and therefore our conclusions are more or less independent of the assumed free tropospheric nucleation mechanism.”

P8922 L26-28: “The dependence of OH on reactants such as NOx, 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, NOx and VOCs. The new method accounts for instantaneous changes in cloud cover (radiation), but no changes in NOx and VOCs, so an improvement in one aspect (radiation) and a regression in another aspect (NOx and VOC effects).

True, but as the results show, our approach gives much better results than using the monthly mean values.

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

See our response above.

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).)

We added the unit, which is cm-3. From the time point of view, the checking is done once per our from instantaneous values (offline).

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 the there was no nucleation observed that the model correctly predicted no nucleation (e.g. true positives, false positives, true negatives and false negatives)?

No we have not done such an analysis. The simulation were not nudged towards meteorology (not forced to follow the re-analysis data), but forced from the lateral boundaries. This means that inside the domain, the model was in a freely-running mode and the meteorology was different than what the observed (not totally, but so far the model cannot reproduce perfect match for meteorological conditions). This is the reason why a direct day-to-day comparison of model results and measurement is not meaningful (the daily mean J3nm rates in Figure 2 are for overall analysis of the features and the mean values). In the analysis, we have concentrated more on the statistics of nucleation (most of the results are analyzed on a monthly scale).

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

We wanted to be consistent with the colors and maybe in this figure the choice of colors in not the best one. Using black for measurement would fix the problem, but does not work in Fig. 2.

P8928 L20 and y-axis in Fig 4: "fractions of nucleation days", what does this mean? Do you mean the fraction of days that have nucleation? Please make more clear.

This has been changed to "fraction of event days"

P8932 L15: "emissiona" should be "emissions"

Corrected as suggested.

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.

Yes it is the surface. We think it is useful to show how the model predicts the nucleation at the boundary layer. To test the vertical profiles, measurements, for example from the PEGASOS projects Zeppelin, should be used. In this work this is not in the main focus and such a comparison will be left for the future studies.

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).

A good point. This sentence has been removed.

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.

Yes, the transport of SO2 plays a bigger role, but as the convective transport does not include microphysical processes (condensation), H2SO4 can trigger nucleation after transport.

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.

We divided this section into two separate sections: "Mean nucleation rates in Europe" and “Spatial extent of events”. Also, the comparison part (Crippa 2013) is moved to the latter section.

Figure 11 and section 3.6. In the text and in the y-axis label, is 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?

Good point, it is the burden of nucleated particles. We have change the label to “burden”.

P8939 L8-10: This last sentence doesn’t make much sense. Are you saying that NPF plays an important role in nucleation events? Seems circular.

Modified to “The regional meteorological and chemical features play an important role in shaping the nucleation events.”

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