Atmos. Chem. Phys. Discuss., 14, C4872–C4885, 2014
www.atmos-chem-phys-discuss.net/14/C4872/2014/

Atmospheric Chemistry and Physics Discussions

ACPD

14, C4872–C4885, 2014

Interactive Comment

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

J.-P. Pietikäinen

joni-pekka.pietikainen@fmi.fi

Received and published: 15 July 2014

This paper presents an analysis of new particle formation events across Europe in a regional aerosol microphysics model compared to those derived from observational data.
The analysis examines the frequency, duration and spatial distribution of new particle formation events generated by two configurations of the model with different approaches to simulating OH.

The analysis will be of interest to the scientific community since new particle formation events have been shown to contribute a large proportion of cloud condensation nuclei in continental regions.
The modelling approach to represent OH via a solar radiation proxy is novel and provides another reason why the paper is within the scope of ACP.
I also consider that the paper represents a substantial achievement in bringing together observations from 13 European measurement sites to evaluate simulated nucleation events in the model.
The paper is reasonably well written and includes quite a detailed description of the methodology which, although a little lengthy at times, helps the reader understand the rationale and approach.

I recommend that the paper be published after several minor amendments are carried out

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.

1) Abstract, page 8917, lines 14-16: the last sentence of the abstract is rather vague. The authors should change this sentence to be more specific. In the text the authors refer to the fact that SOA is not included in the model. Are the authors here referring to the likelihood that organics may exert an important influence on nucleation rates not included in the present configuration?

Actually, the last sentence was removed, because it had information that is more meaningful in the conclusions. The SOA would lead to higher growth rates and

ACPD
14, C4872–C4885, 2014

Interactive Comment

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

eventually to bigger condensation sink, which would improve the simulated length of the events (please see also responses to Referee 1). The influence on the nucleation rates was not discussed earlier, but based on your comment 3 this was added.

2) Introduction, page 8917, line 23: Please reword "The atmospheric relevance of the nucleation is undisputed". First suggest to replace "of the nucleation" with "of new particle formation". Second perhaps better to refer to the "climate relevance" rather than the more general "atmospheric relevance". Third "undisputed" is a peculiar choice of word " suggest to replace with "has been demonstrated by several studies" and include at least 1 reference for the first papers which showed the importance for global CCN (e.g. Spracklen et al., 2006)

Corrected as suggested.

3) Introduction, page 8918, line 14: The authors should mention the studies which have demonstrated that organics plays an important role in new particle formation and/or initial nuclei growth. For example Metzger et al. (2011) showed that using a nucleation rate parameterized as proportional to the product of the gas phase concentrations of sulphuric acid and an oxidised organic species gave improved comparison against observations.

This has been now added. Also, when the underestimation of nucleation rates with REMO-OHP is discussed, we added “The underestimation can come from the chemistry part, but also from the nucleation parameterization. For example, better representation of organics and their influence to the nucleation rates could lead to more realistic J3nm values. Currently, the influence of organics comes indirectly from the kinetic coefficient K in Eq. (3), which is based on measurement and includes the

ACPD

14, C4872–C4885, 2014

Interactive Comment

effect of organics (if any). We chose this approach as the model does not have an SOA module."

4) Introduction, page 8918, lines 15-30: The paper gives the names of the models used for the different studies, but I find that distracting to the text and instead recommend those acronyms to be removed with a more general description of the type of model given. For example on line 16 replace "used a global aerosol microphysics model, GLOMAP, to. . ." with "used a global chemistry transport model with aerosol microphysics to. . ." Similarly on lines 20-21 replace "modified the global climate model ECHAM5-HAM with. . ." to "modified a global aerosol-climate model with. . .". On lines 26-27 please delete "in ECHAM5-HAM" as the implication is presumably that this is a general result. On line 30 please replace "in the global aerosol climate model ECHAM5-HAM" with "in a global aerosol-climate model".

Corrected as suggested.

5) Introduction, page 8919, lines 2-3: The sentence "The nucleation via cluster activation, which requires the presence of organics, was used only in the forested boundary layer" is confusing and is too detailed for discussion here. The text "was used only in the forested boundary layer" suggests the authors are discussing their model"s existing implementation of the combination of ion-induced nucleation (or is binary nucleation) and cluster activation parameterization " in which case the text ought to be in the model 2.3. But it is even more confusing because there (page 8923 lines 18-20) the authors explain that organics are not considered in the model. And in any case the cluster activation parameterizations mentioned (Kulmala et al., 2006; Sihto et al. 2006) are based on being proportional to sulphuric acid only without influence from organics.

ACPD
14, C4872–C4885, 2014

Please reword to clarify and move to section 2.3. Also the next sentence seems to be describing the model used rather than being a review of relevant literature. And which observations are these? At which type of site? Please move and reword to clarify.

The sentence indeed was too detailed and was removed. We have modified the end of this chapter to "...such a nucleation mechanism is a good candidate to explain NPF over the oceans and free troposphere. The combination of this and nucleation via cluster activation seemed to better explain the observations of ultrafine aerosol concentrations over Pacific Ocean than the cluster activation alone." Discussion about the influence of organics was done in the answer to the comment 3.

6) Introduction, page 8919, line 9 "each author having his/her own nucleation parameterization of choice" " this is rather non-scientific language " better to reword to say something like "each study assessing which parameterization leads to best comparison to observations in their model".

Corrected as suggested.

7) Introduction, page 8919, line 10 " delete the words "However, as.." and instead start the sentence "Global models. . .." " then replace "predicting the changes" with "hence prediction changes" (better English).

Corrected as suggested.

8) Introduction, page 8919, line 15 " suggest to replace "seem to be more appro-

ACPD

14, C4872–C4885, 2014

priate for this mission" with some text explicitly stating what you mean by "the mission". How about "have resolution of a few tens of km (?) and hence resolve much greater variability in emissions and processing, and provide a better framework to calibrate potential nucleation mechanisms against observations".

Changed to "Regional climate models, on the other hand, have resolution varying from kilometers to tens of kilometers and hence resolve much greater variability in emissions and processing, and provide a better framework to calibrate potential nucleation mechanisms against observations."

9) Introduction, page 8919, lines 15-30 " as with my point 4) above, I suggest to remove the acronyms for each model ("UAM-AERO" on line 16, "WRF-chem" on line 19 and "PMCAMx-UF" on line 25). Instead just mention the type of model with a reference and make the point be a general one for that model type. Note also that WRF-chem is not a regional climate model but a regional weather forecasting model.

Corrected as suggested.

10) Introduction, page 8919, line 23 " reword the text "because NPF tends to cancel out the effect of reductions" " maybe replace with "because NPF generates a stronger source of CCN in conditions with lower condensation sink".

Corrected as suggested.

ACPD

14, C4872–C4885, 2014

11) Introduction, page 8919, line 26 " suggest to replace "regionally" with "in some regions" (or explicitly state the regions where this is the case).

Corrected as suggested (in some regions).

12) Introduction, page 8919, line 30 " explicitly state which observations and/or in which environments this parameterization "performs better".

Changed to "Based on their results, a semi-empirical ternary sulphuric acid–ammonia–water parameterization shows better agreement with measurements of particles larger than 10 nm than kinetic or activation parameterization."

13) Introduction, page 8919, line 30 " I would recommend the authors add one more relevant study to their overview " the recent study by Scott et al. (2014) which showed that the seasonal cycle and magnitude of simulated particle concentrations at three European sites were improved when a nucleation parameterization involving organics was used.

Although the study by Scott et al. (2014) is very interesting, in this chapter we only show studies from regional (or similar limited area) models. Since the suggested paper describes global modelling study, we have included it in the list of studies done by global models (in introduction).

14) Introduction, page 8920, line 4 " I would recommend to strengthen your motivation for the study to say that by comparing to a full year"s measurements at these 13 sites you are able to test the nucleation in the model against the

ACPD

14, C4872–C4885, 2014

observations covering a range of seasons and environments.

Changed to "The results are compared with measurements from 13 European sites covering years 2003–2004 and 2008–2009, which allows us to test the nucleation in the model against the observations covering a range of seasons and environments."

15) Introduction, page 8920, line 9, You could say that your study is (to my knowledge) the first to compare nucleation rates from the model to those from observations. All the other studies you mention compare simulated particle concentrations. Comparing the model nucleation rate against that derived from the observations is a stronger constraint than comparing particle concentrations to observed particle concentrations because the latter has greater possibility for compensating errors (for example via biases in number sink due to coagulation or too rapid growth).

We modified the ending of this chapter to "...particles via H2SO4 condensation. This study is (to our knowledge) the first to compare nucleation rates from the model to those from observations. In the previous studies, the focus has been to compare simulated particle concentrations. Comparing the model nucleation rate against that derived from the observations is a stronger constraint than comparing particle concentrations to observed particle concentrations, because the latter has greater possibility for compensating errors (for example via biases in number sink due to coagulation or too rapid growth)."

16) Introduction, page 8920, line 9, You could also consider mentioning that your OHproxy method might be useful for other types of model where nucleation is important to resolve adequately but for whom a tropospheric chemistry scheme

ACPD

14, C4872–C4885, 2014

Interactive Comment

would be prohibitively expensive.

We modified the middle part of this sentence to "...linked to the OH concentrations, thus taking into account the effects of clouds. The method shown here can be very useful for other types of models where nucleation is important to resolve adequately, but for whom a tropospheric chemistry scheme would be prohibitively expensive. In addition, the particle..."

17) Methods, page 8922, lines 18 and 19 " You use the term "global radiation" twice here but you need to be more specific than that " presumably you"re using the incoming short-wave flux from the model " if so please say so and change to "downward SW flux".

Corrected as suggested.

18) Methods, page 8922, line 19 " what do you mean be "global radiation is more commonly available in different datasets" " do you mean available in the aerosol-climate model? And more commonly than what? Please reword accordingly

Modified to "The reasons for this are that the correlation between these two variables is evident, SWF$\downarrow$ is often measured and SWF$\downarrow$ is available in the climate models."

19) Methods, page 8922, line 24 " as in point 17 above, suggest to change "Radiation" for something more precise " is it "downward SW flux" " come up with a symbol for this and use it in the equation stating in the text what it stands

ACPD
14, C4872–C4885, 2014

for.

Corrected as suggested (SWF↓ now used).

20) Methods, page 8923, line 11 " replace "for the forested boundary layer" with "restricted to the forested boundary layer" as I think this better represents what you are describing here, which is the implementation into the model.

Corrected as suggested

21) Methods, page 8924, lines 5-6 " you have the text "is based on a comparison of the model results and measurements conducted within this work (not shown)". You mean "best comparison to the measurements"? As this is the basis for your comparisons the paper needs to be clear how this value was arrived at. Please state the specific observations where you got best agreement with this rate.

Added “We compared measured H2SO4 concentrations agains different K values from Hyytiälä, Melpitz and San Pietro Capofiume, and derived the best fit.”

22) Methods, page 8924, lines 10-11 " you explain that you follow the same approach as Makkonen et al. (2009) and that they only allow sulphuric acid to condense onto the aerosol. But I don"t understand, don"t Makkonen also have some SOA condensing too? Or was that only to particles larger than 3nm. Please clarify.

We removed the sentence altogether as the growth to 3 nm is not explicitly calculated in the model. However, we added the following sentence to the end of the previous paragraph: “The 3 nm particles are assumed to consist of sulphuric acid only (and thus a corresponding amount of H2SO4 is removed from the gas phase as the particles are formed)”

23) Methods, page 8924, lines 12-14: I don"t understand this " it seems too detailed here. I"d suggest to just briefly say that new particle formation is assumed not to occur in the cloudy part of the gridbox.

Corrected as suggested.

24) Methods, page 8925 " line 4 " spelling "compairing" → "comparing".

Corrected as suggested.

25) Methods, page 8925 " line 4 " suggest to change "the model results" to "simulated nucleation events" so that it is more specific about what you are comparing.

Corrected as suggested.

26) Methods, page 8925 " line 5 " where you say "observation data" from the 3 sites is used, I"d suggest to say what the instruments are. It"s good to refer the reader to the papers for full details but it is also good to say the type of instrument used in the text here.

We added “The aerosol size distributions, from which the event statistic were calculated, were measured using twin Differential Mobility Particle Sizer (DMPS) on all sites. “

27) Comparison with measurements, page 8926, section 3.1 " I"d suggest to start this subsection by first describing the observed seasonal cycle at each of the 3 sites and how they differ. For example Hyytiala has peak nucleation rate in the spring whereas Melpitz and San Pietro Capofiume peak in summer. Then go on to compare the model in each case.

We added a chapter “Overall, the measurement show that Hyytiälä and Melpitz has peak nucleation rates in the spring and autumn, whereas San Pietro Capofiume has peaks in spring, summer and autumn. Both model versions show similar features, although REMO-OHP can not reproduce the autumn peak in Hyytiälä. On the ohter hand, REMO-NCH shows much higher values at all locations and thus the peaks are not as clear as with REMO-OHP.”

28) Comparison with measurements, page 8926, line 12 " the capital Delta symbol with subscript r is not defined in the text. I'm assuming this is normalized mean bias. Please define the symbol before first use

We changed the first sentence to be more clear “(the relative change of 2-year mean Δ_r, calculated by first substracting the measured mean from the model mean, then dividing this by the measured mean and finally multiplying this by 100%, is $\Delta_r = -71\,\%$)”

29) Comparison with measurements, page 8926, line 13 " you explain that

ACPD
14, C4872–C4885, 2014

at Hyytiala the summer values are well reproduced by the model " yes the OHP model does. But the NCH model is a factor 10 too high in Figure 2. You should state in the text that the comparison improves from the NCH to the OHP. However you should also say that the diamonds for the NCH look closer to the observations during spring than the OHP so in that case season switching to the OHP mechanism has degraded the model skill against the observations.

We divided this chapter into two parts and the end of first chapter is now “During summer, the values are over 10 times higher in REMO-NCH than in the measurements, but in the spring, REMO-NCH reproduces the measured rates more realistically than REMO-OHP, which underestimates the values by a factor of 5-10. This shows that seasonally the new model version still has deficiencies.”

30) Comparison with measurements, page 8926, lines 18-21. It is noticeable to me that REMO-OHP is low-biased through much of the year compared to the observations. In fact from looking at Figure 2 I expected the average bias to be worse for OHP than for NCH. I suspect the reason it doesn’t is because you are using the normalised mean bias which weights towards the larger values. It would be interesting to see whether one found the NCH was closer to the observation is instead the "mean normalised bias" is used. This metric gives an average of the normalised bias " so if one is a factor two too high for one half of the period and a factor two too low in the other half then one gets an average bias of zero. By contrast one would get a normalised mean bias greater than zero because of the weighting to larger values. Although including both metrics of bias may be too much, the authors should at least include reference to the occasions where the OHP is too low against the observations (sometimes MCH compares better).

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

The calculation method is explained in answer to the question 28. Indeed, we did not feel necessary to include both metrics, but we added more discussion about the underestimations.

31) Comparison with measurements, page 8927, line 2 " reword the phrase "REMOOHP had some problems" " not scientific language " be specific about the bias you're talking about here. In general I think you should consider reworking the text in this section. To my mind, the first order thing from Figure 3 is the duration of the nucleation event " the start and end times are useful to interpret difference in the length of the episode, but the main results I would think should consider the length of the episode. Please try to improve the wording of this section to make it easier for the reader to take in the information.

We improved the language and moved the length analysis to the beginning of each chapter.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 8915, 2014.

ACPD

14, C4872–C4885, 2014

Interactive Comment