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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

Anonymous Referee #2

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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

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?

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)

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.

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”.

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. 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.

6) Introduction, page 8919, line 9 “each author having his/her own nucleation param-

eterization 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”.

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).

8) Introduction, page 8919, line 15 – suggest to replace “seem to be more appropriate 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”.

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.

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”.

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

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

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

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European sites were improved when a nucleation parameterization involving organics was used.

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 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 fromthe 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 OH-proxy method might be useful for other types of model where nucleation is important to resolve adequately but for whom a tropospheric chemistry scheme would be prohibitively expensive.

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".

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

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

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and use it in the equation stating in the text what it stands for.

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.

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.

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.

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.

24) Methods, page 8925 – line 4 – spelling “compairing” -> “comparing”.

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.

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.

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.

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

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

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).

31) Comparison with measurements, page 8927, line 2 – reword the phrase "REMO-OHP had some problems" – not scientific language – be specific about the bias you're

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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.

References

Metzger, A., B. Verheggen, J. Dommen, J. Duplissy, A. S. H. Prevota, E. Weingartner, I. Riipinen, M. Kulmala, D. V. Spracklen, K. S. Carslaw, and U. Baltensperger, Evidence for the role of organics in aerosol particle formation under atmospheric conditions, Proc. Nat. Acad. Sci., vol. 107, no. 15, pp. 6646-6651, 2011.

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