Analysis of nucleation events in the European boundary layer using the regional aerosol-climate model REMO-HAM with a solar radiation-driven OH-proxy

Author's Response

Dear Dr. Nemitz,

The Copernicus system wants all changes to be made to their LaTeXversion of the manuscript and unfortunately this does not work perfectly on my machine. This means that some styles of equations, variables etc. might not be in their final form (in the pdf that includes the modifications based on the referee comments). As this will be corrected in the final phase (assuming that the paper will be eventually accepted), I ask you to only check the text and updated figures (if needed; in most of my comments I included the text-changes).

A point-by-point responses to the reviews can be found at the end of this document. No other modifications was done.

Thank you for your help.

Best Regards,
Joni

Response to Anonymous Referee #1

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 re-

actants. 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."

Response to Anonymous Referee #2

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 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 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. 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 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".

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.

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 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 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↓ is often measured and SWF↓ 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 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 $\Delta_\mathrm{r} = -71\,\%$)"

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.

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).

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.