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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of aerosol number concentrations in NorESM with improved nucleation parameterisation" *by* R. Makkonen et al.

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We thank the anonymous referee for suggestions, which have improved the manuscript. We answer the specific questions below. The referee comments are in bold.

As a general comment, I wonder if trying to compare multiple simulations to such a wide range of observation "types", in sufficient detail, may be too much for one manuscript.

We agree with the reviewer that the evaluation is rather extensive. However, the aerosol

number concentrations of NorESM have not been thoroughly evaluated previously. With a globally rather sparse measurement network (60 stations in this manuscript), we did not feel that any station would bring insignificant information to the evaluation. Although we left a comprehensive evaluation of simulated vertical aerosol profiles out of the manuscript, we included a more qualitative comparison against flight data.

Section 2.4.2, p26397, line 12: just from ozonolysis? Or all oxidation products?

Only 50% monoterpene ozonolysis products are assumed to participate in nucleation and nuclei growth (dp$<$24 nm). The assumption is based on the work by Yli-Juuti et al. (2011), who showed a positive correlation between growth rates of 7-20 nm particles, monoterpene concentration and oxidation rate by ozone.

Section 2.4.2, p26397, line 13: can the oxidation products condense onto other particles? (i.e. larger than the nucleation mode?)

In our version of NorESM, condensation of organic vapours onto pre-existing particle population is not implemented. Rather, the SOA formation is formulated as in Kirkevåg et al. (2013): SOA is lumped in an internally mixed Aitken size OM/BC mode. We are aware that this implementation is not realistic in terms of atmospheric SOA formation, and there are ongoing efforts to improve the SOA scheme in NorESM. In this manuscript the focus is on total aerosol number concentration, and we show that the implemented organic vapor partitioning in the sub-24 nm range significantly improves the simulation of the effect of SOA on number concentration.

Section 5.1, p26403, lines 20-21: is this sentence referring specifically to the ActNuc_BC12 simulation? That seems to be more like five-fold, i.e., 2205 / 409 ?

In the revised manuscript, the sentence is revised to:

"The model simulates a strong land-ocean contrast with number concentrations ranging between 890 cm-3 and 3250 cm-3 over land areas and between 250 cm-3 and 660 cm-3 over ocean areas in the NoNuc_BC24 and ActNuc_BC24_Nuc10 simulations,

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respectively."

Section 5.1, p26403, lines 24 onwards: It's a bit confusing to have the figures like they are in Figure 2, i.e., no nucleation – nucleation in Figure 2a, which gives a general reduction in particle concentration, but then in the text discuss the sensitivity to nucleation as an increase. Would it make more sense to plot this the other way around? Or at least discuss the percentage decrease in terms of a lack of nucleation?

We agree about the confusion between text and figure. We have reversed the plots in Figure 2 to correspond to "nucleation minus no nucleation", "SOA formation minus no SOA formation", "small BC particles minus large BC particles" and "organic nucleation minus activation nucleation". This order is perhaps more intuitive for the reader.

Section 5.2.1, p26406, lines 10-12: April-May peak not in observations

We have revised the sentence to read: "In Barrow, the model simulates two distinct concentration peaks in April–May and July-August, while only the latter is found in the observations. "

Section 5.2.1, p26406, line 15: I'd rephrase "seems to work best" to something more scientific.

We have modified the sentence to read: "...simulation ActNuc_BC24_Online with different meteorology yields the highest correlation coefficient of R^2=0.62 and a bias of +22%..."

Section 5.2.2, p26406, line 25: It may aid the reader to clarify that these two locations are in different hemispheres and therefore have opposing seasonality. There also seems to be a large difference between the median and mean values on the plots at these two locations Section 5.2.2, p26407, lines 5-7: It is touched on briefly here but it would be useful to have some discussion of how representative the mean vs. median is at these locations and a sense of the interannual

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variability in the observations (e.g., standard deviation?).

In the revised manuscript, we have included a mention on different hemispheres and revised the text to include:

"As in Spracklen et al. (2010), the model has difficulties in simulating the high mean concentrations observed in Cape Grim during winter months: in Spracklen et al. (2010), the model results are generally outside one standard deviation of the observations during May-September, although it seems that increasing the primary sulfate emissions slightly improves the simulated winter concentrations. The winter concentrations in NorESM are fairly insensitive to the simulated experiments. However, the observed variability of aerosol concentration in Cape Grim is considerable, and simulations without nucleation can capture the median concentrations throughout the year."

Section 5.2.3, p26407, line 26: from the plot several of the simulations look pretty similar, why is ActNuc_BC24 chosen as reproducing the concentrations very well?

We have revised the sentence to read:

"Although the measurements at Niwot Ridge station were heavily influenced by local pollution from lower elevations, the simulations with nucleation (including NorESM1-M) reproduce the observed concentrations very well except for an overestimation during late summer."

Section 5.2.5, p26409, line 1: I'm not sure you can call this a summer "minimum", the concentrations are still higher than in the winter.

The minimum referred to a local minimum typically observed during summer at certain sites, although the concentrations during the local summer minimum are usually higher than during winter conditions. We have revised the text to:

"The local summer minimum, observed during July–August in..."

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Section 5.2.5, p26409, lines 4-5: Is this surprising? What year are the wildfire emissions from?

The wildfire emissions correspond to year 2000, while Tomsk and Listvyanka observations cover years 2005-2006. While the discrepancy between model and observation is certainly not surprising, we wanted to address the similarity and discrepancy of the nearby stations in model and observations, respectively.

Section 5.2.6, p26409, lines 23-25: could this also be affected by the lack of seasonal cycle in primary anthropogenic emissions?

The discrepancy in a location such as Po Valley or Ispra is certainly affected by both the incapability of the model to simulate subgrid-scale conditions and the timing of anthropogenic emissions. We have revised the text to read:

"NorESM can clearly not reproduce the high concentrations in Po Valley or Ispra during wintertime, possibly due to omitted seasonality of anthropogenic emissions, overestimated scavenging, or difficulties in simulating the boundary layer structure and topography with the coarse spatial resolution of NorESM."

Section 5.2.7, p26411, lines 1-3: as a more general comment, doesn't the way this model treats SOA (even with the improved representation) preclude any detailed analysis of its impact on total particle number concentration, since (beyond the nucleation mode) you are adding new particles at 40 nm rather than allowing the SOA to partition to existing particles (which would increase their size but not number).

As discussed in section 2.4.3, the implemented SOA formation model improves the description of SOA effect on total particle number by taking into account the increased survival probability of nucleated particles and the possibility of organic nucleation. Still, we acknowledge that the implemented SOA formation mechanism is far from realistic for the atmosphere. The current version of NorESM is not able to treat condensation

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of organics, but this issue is under development.

Section 5.4, p26412, lines 14-16: that is true for the mean, but the simulations appear able to capture the median value?

It is true that for TRACE-P and INTEX-A, the model is unable to capture the high mean concentrations below 2 km and 1 km, respectively. In ACE-Asia, the _BC12 simulations are able to produce also the high mean concentrations together with the vertical gradient. In all three regions the observed vertical median profile is well simulated. We have added the following to the revised manuscript:

"Although the high mean concentrations at low-altitudes in TRACE-P and INTEX-A are not captured, the median vertical profiles in the three regions including ACE-Asia are well simulated."

Section 5.4, p26412, lines 18-19: what is the 'uncertainty range of observations' that you refer to here?

The sentence was revised to say: "In TRACE-P and INTEX-A, the simulations are generally within one standard deviation of observations."

Section 5.4, p26414, line 20: 100% of what?

Text is revised to:

"... decreasing the BC particle size from 24 nm to 12 nm (ActNuc_BC12 vs. ActNuc_BC24) doubles the near-surface (0–1 km) particle number concentrations in ACE-Asia and TRACE-P."

Section 5.5: I am not sure that this section adds much to the previous analyses?

The poorly quantified aerosol properties over global oceans together with the potential for a strong climate effect substantiate the need to evaluate aerosol number concentration over the ocean. We feel that Fig. 12 casts light on the roles of different processes and emissions controlling the number concentration over ocean. While the analysis

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does not allow for a detailed point-by-point comparison, Fig. 12 indicates that the model could be overestimating oceanic aerosol number. This overestimation is similar to what was found in e.g. Spracklen et al. (2010).

Technical Suggestions:

As far as I can tell, the Supplement just contains the same figures as the paper?

Due to a technical error, the supplement was replaced by the manuscript document. This is corrected in the final version.

p26400, line 2: Should the Metzger reference be in brackets?

Yes, this is corrected in the revised manuscript.

p26402, line 8: insert "one" before "location" p26413, line 21: change "campaing" to "campaign"

The above corrections were implemented.

Figure 3: there is no scale for the background contours?

We have include a scale for the background in the revised version of the manuscript.

Figure 4: where was IMPEX? MIRAGE is on here twice

The IMPEX-campaign was held in West-US and was erroneously labeled as MIRAGE in the Figure 4. This is fixed in the revised manuscript.

Figure 5: it would be useful to define the components (in the pie chart) in the figure caption.

We have defined the components in the figure caption in the revised manuscript.

Figures 6 – 12: Depending on the layout of the final manuscript it might be worth reproducing the legend from Figure 5 on a couple of these to save the reader from referring back each time. Also, it might be better if they are shown in the

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same order that they are discussed: e.g., 5, 8, 7, 6, 9, 10, 11, 12.

We have duplicated the legend to figures 6-7 and synchronized the ordering of figures 5-10 to match the sections.

Figure 10: these plots are far too small

We will redistribute the panels for the ACP-version to ensure the readability of the plots.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 26389, 2013.