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Editorial Support

Manuscript Number: acp-2015-242
Title: "Natural new particle formation at the coastal Antarctic site Neumayer" by R. Weller et al.

Dear Natascha Töpfer,

Please find below our reply to the comments of the reviewers and how we revised our manuscript, including a list of relevant changes. We thank all reviewers for their constructive comments and suggestions. All comments have been taken in account in our revised version. Note that all relevant changes in the revised manuscript are marked in yellow.

I state that my co-authors concur with submission in its revised form.

Sincerely yours
-Rolf Weller

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encls.

Responses to Interactive comment by Referee #1

We thank referee #1 for his constructive and detailed comments and suggestions, which added to improve and clarify our manuscript (ms).

• 5 day trajectories. The decision on the length of trajectories used should be discussed here in terms of the uncertainty. Authors have mentioned this in passing but a more thorough discussion of the topic should be performed given the high uncertainties present in the input meteorological datasets in this region.

We agree that the limitations of our backward trajectory approach should be stated more clearly. However, it is inherently difficult to provide a reliable uncertainty assessment. We carefully used trajectory analyses in our evaluation to avoid any over-interpretation. In summary trajectories did neither indicate a pronounced impact of marine nor of descending air masses from the free troposphere. Hence in our case, trajectory analyses appeared equivocal in evaluating a rather local process like NPF, probably because of their inherent spatial uncertainty particularly in regions sparsely supported by meteorological data (**this conclusion is added in chapter 3.2**).

Uncertainties of trajectories are now embraced in chapter 2.1. We highlighted the somewhat equivocal outcome of trajectory analyses in chapter 3.2 as well as in chapter 4.1. We added Fig. 5 showing trajectory results in more detail regarding the case study presented in chapter 4.1.

• Detailed analysis/interpretation of the particle formation event isn't presented and would be useful, particularly for the single particle formation event that the authors pick out as a case study.

We added now a detailed case study (new chapter 4.1 in our revised ms).

• Discussion about precursors and conditions leading to NPF events is minimal, and given the other measurements available at Neumayer, could be significantly strengthened. This would significantly strengthen the precursor discussion presented in the paper.

Referee #2 raised the same point and in this regard we revised our ms, especially by the inserted **new chapter 4.1.**

• Discussion of iodine oxide nucleation requires a consideration of the seasonality of the IO concentrations. It should also be described what concentrations are required for nucleation to occur so that the reader is able to determine for themselves if the Antarctic concentrations are high and/or sufficient.

In fact IO concentrations required for observed NPF at NM cannot be assessed on the base of our measurements. Considering the available laboratory-, field- and model results, it appears difficult to estimate IO concentrations needed to provoke significant particle nucleation but it seems that several pptv IO or OIO would be necessary.

We provided more information on IO measurements at NM in chapter 4.4.

Specific minor concerns:

• *Page 15657, Line 1 - the sentence starting with "One focus of interest. . ." should begin a new paragraph*

Corrected.

• *Page 15657, Line 7 – sentence beginning "Concerning the marine troposphere. . ." should be revised, this currently does not flow nicely.*

We revised the sentence.

• *Page 15659, Line 22 – please give a reason as to why 4 consecutive spectra were averaged.*

It is just a reasonable compromise between time resolution and noise level.

• *Page 15660, Line 3 – change "referred to Dal Maso . . ." to, "As in Dal Maso. . ."*

Changed.

• *Page 15660 Line 8 – why are ionic composition measurements introduced in the methods section? They are not utilised at all throughout the study. These should be removed.*

The ionic composition of the aerosol is now involved in the discussion (new chapter 4.1).

• Page 15661, Line 15-19 – please revise this sentence, currently it does not make sense. It may also be worth defining what the particle growth criterion is that you are getting rid of and why the spatial distribution of the event is relevant here.

Originally Dal Maso et al. (2005) defined a NPF event exclusively when particle growth could be detected. But as already mentioned in Dal Maso et al. (2005) and O'Dowd et al. (2002a), this criterion seems not to be appropriate in case of more local sources.

We deleted this passage, because it does not add much to the case.

• *Page 15661, Line 23-25 – revise sentence grammar.*

Corrected.

• *Page 15663, Line 2 – please define the units of cvapour and γ*

Units are now defined; γ is dimensionless.

• *Page 15663, Line 15 – "striking NPF event happened in 27 January, where a simultaneous" should be changed to "striking NPF event that happened in on 27 January, where a simultaneous"*

Changed.

• *Page 15664, Line 14 – please define "bright". Does this mean "cloud-free"? What were the solar radiation levels?*

These terms are now specified.

• *Page 15664, Line 18 – "5 days" should be "5 day"*

Corrected.

• *Page 15666, Line 4 – total particle number concentration increased up to 3000 cm-3 from a background of what?? What was your average?*

This chapter is now completely rewritten.

• *Page 15666, Line 10 – Notwithstanding should have a comma after it, so it should become "Notwithstanding, some . . ."*

Corrected.

• Page 15667, Line 27 – define a scale for NH4+, and whether 10 ng/m3 is high enough to be involved and

• Page 15668, Line 2 – as for previous comment, but for WSOC

The role of NH_3 and WSOC in nucleation is highly complicated and to assess their importance in our specific case is virtually impossible due to the lack of appropriate data. Apart from the fact, that we just measured ammonia (NH_4^+) and not (gaseous) NH_3, one has to know the amount and preferably also the nature of low volatile organic compounds (LVOC) involved.

• Page 15669, Line 5 – please rephrase this to include the idea that this conclusion is achieved primarily through ancillary data, rather that online measurements.

This point is now added.

• Figure 1 – labelling the x axis and the color bar. Color bar should be relabelled in linear, rather than logarithmic units.

As for the contour plot a linear size distribution scale is not appropriate (we checked this), so we persist on the logarithmic scale.

• Figures in general – it may be useful to include legends, or axis color coding in the figures to enable quick interpretations of the figure (e.g. in Figure 1c, the right axis would be blue).

Changed.

Finally, because of a scaling error we **redrew Fig. S1d in the Supplementary Material**.

Responses to Interactive comment by referee #2:

First of all we would like to thank referee #2 for his effort in evaluating our manuscript (ms)! According to his comments, we reconsidered and rectified our ms. In a nutshell: We added a detailed case study and considered in more breadth auxiliary data from Neumayer.

1) Instrumentation used during 2012 is not directly comparable to instrumentation used in 2014. When focusing solely on qualitative definition of presence and absence of NPF, it should not play a major role, however, for comparison of growth rates and size distribution dynamics, direct intercomparison of both systems is necessary and should be presented. It is not uncommon that aerosol size spectrometers vary from each other significantly [Wiedensohler et al., 2012] as well as cut off characteristics of CPCs. Also using GR calculation and size ranges with two decimal precision has no realistic meaning.

Unfortunately a surely desirable intercomparison was not possible, because we merely have had one classifier available. In order to check the consistency of particle growth rates (GR) between both set-ups contemporaneous measurements would have been indispensable but was not feasible. The CPCs have been checked for consistency as described in Weller et al. 2011a and both instruments were calibrated by the manufacturer recently before the campaign. We are confident that the compulsory change of our set-up did not significantly restrict our analysis and our main conclusions. GRs are intrinsically difficult to compare, because they are usually determined within different size ranges (see table 1). We cannot anticipate that GRs are independent of particle size, even though we could not detect such a dependency due to the large uncertainty of the derived GR (table 1). Apart from this, GRs derived from 11 out of 13 NPF events were measured with an identical experimental set-up!

We agree, that presenting GR in two decimal precision is meaningless and corrected the values accordingly.

2) I understand that it is very demanding on resources and logistics to carry out measurements at such a remote place and it is difficult to run extensive instrumentation set up there. But authors did not explore even which they have available. Data analysis will be more robust if local meteorology and other aerosol and trace gas observations at Neumayer will be better linked to NPF observations. How different are conditions between class I and class II event? How different are conditions between NPF and non NPF days? Can importance of marine air on NPF be better assessed? Authors have available data from local meteorology, radiation, cloud cover, BC and scattering levels, OPC and two CPC data which are part of core program. Trace gases: Rn222 and O3 are observed with good temporal resolution, daily data about reactive trace gases. Can authors link air mass origin using trajectories with other observations to assess time spent over the sea/coastal Antarctica for NPF and nonNPF cases? Authors can also try to use water vapor as an air mass tracer of marine and continental air masses. Neumayer is a GAW station and potential of observations conducted there was not explored sufficiently in this manuscript.

Our ms seems to leave a mark of not having appropriately considered available secondary data. In our revised ms, we tried to clarify this important point, e.g. **presenting a detailed case study (new chapter 4.1), summarizing so far as possible and meaningful, typical meteorological and trace**

compound characteristics during NPF event and non-event days (new Table 2; we could not found significant differences of auxiliary parameter between case I and case II events).

It should be mentioned here, that in terms of meteorology we particularly relied on 2 m and 10 m data from the meteorological mast, i.e. temperature, relative and (calculated) absolute humidity, wind velocity vector as well as on the BSRN radiation data (focusing here on actinic uv-radiation). As mentioned in chapter 4.3 (now 4.4), rH tended to be lower during NPF, but this was simply due to the fact that in those cases bright weather prevailed. On the other hand, there were plenty of days with meteorological conditions typical for NPF days exhibiting no particle nucleation. Apart from the shifted diurnal variations between NPF and uv, no correlation between uv radiation and GR (or particle concentrations within the nucleation mode) was given.

Surface ozone, black carbon (BC) and ^{222}Rn: Again, we found no remarkable relationship between particle concentrations within the nucleation mode or periods with NPF and the concentration of continuously measured surface ozone, BC and ^{222}Rn concentrations. Particularly ^{222}Rn data are difficult to interpret at coastal Antarctica and there is no unequivocal link between ^{222}Rn concentrations and the characteristics of the air masses (continental or marine) as discussed in detail in Weller et al. 2014. On the other hand BC concentrations were in the lower ng m^{-3} range throughout, except spikes during very rare contamination events. As for surface ozone, our now more than 30 yearlong continuous record showed some cases where the typical ozone depletion events (ODE, usually sporadically occurring between August through October each year) were accompanied with enhanced CP concentrations, but this was not the case within the relevant measuring period. Apart from this, a discussion of this finding is another story and clearly beyond the scope of the present ms. Finally, apart from the CPC employed by the SMPS, only one independent CPC was running (and not two, as assumed by the referee).

Optical aerosol properties of the aerosol were measured by an integrating nephelometer and are considered in the revised version of our ms (chapters 2.2 and 4.1, new Tab. 2) as well as some more words on IO measurements at NM (chapter 4.4).

Our experience in interpreting the Neumayer ^{222}Rn time series (Weller et al., 2014) indicated, that assessing the time air masses spent over the sea/coastal Antarctica would need sophisticated Langrangian model calculations considering sea ice coverage, which is beyond the scope of our present ms. **In our revised ms we highlighted the somewhat equivocal outcome of trajectory analyses in chapter 3.2 as well as in chapter 4.1. We added Fig. 5 showing trajectory results in more detail regarding the case study presented in chapter 4.1.** The dependence of rH from air mass history on the other hand is quite complicated and again (highly) equivocal. If at all it is possible to distinguish between periods of cyclonic and katabatic impact by rH values.

In summary, the most relevant changes in the revised ms are (all marked in yellow):

1. We added a detailed case study (new chapter 4.1).

2. We considered in more breadth auxiliary data from Neumayer (throughout the Results and Discussion chapters, especially chapter 3.2 and 4.1).

3. We redraw and upgraded Fig. 1 and added Fig.5 showing trajectory results in more detail regarding the case study presented in chapter 4.1.

Minor comment: In Introduction on page 15656 authors present a picture of aerosol having decisive role in radiative forcing. GHG have decisive role, aerosols have largest uncertainty and we do not know how decisive role they actually play.

We agree that our statement is somewhat misleading: the decisive role of aerosols in radiative forcing is mainly due to their role in acting as cloud condensation nuclei. **This point is now clarified**.

List of relevant changes

1**.** We added a detailed case study (new chapter 4.1).

2. We considered in more breadth auxiliary data from Neumayer (throughout the Results and Discussion chapters, especially chapter 3.2 and 4.1).

3. We redraw and upgraded Fig. 1 and added Fig.5 showing trajectory results in more detail regarding the case study presented in chapter 4.1.