Dr. Rolf Weller

**ALFRED-WEGENER-INSTITUTE**
**FOR POLAR- AND MARINE RESEARCH**
**Am Handelshafen 12**
**D-27570 BREMERHAVEN**

☎ (xx471) 4831 - 1508
T-fax: (xx471) 4831 - 1425

email: rweller@awi-bremerhaven.de

-02 October 2015

**Copernicus Publications**

**Editorial Support**

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

Dear Natascha Töpfer,

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

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

Sincerely yours
-Rolf Weller

encls.

**Responses to referee #3**

We thank referee #3 for his constructive and detailed comments and suggestions, which added to improve and clarify our manuscript.

*1. Lines 21-22. I think that you mean that the particles did not grow to CCN within a day or two. How do you know these particles, after transferred to elsewhere, will not continue to grow to CCN sizes? Also the meaning of "Thus in the main," is not very clear and may need to be rephrased.*

We agree that our statement was misleading and we rephrased the sentence.

*2. Line 121. Could you provide an estimation of the "enhanced uncertainties"?*

All size distribution spectra presented are diffusion corrected by the TSI software. The equivalent pipe length, on which the diffusion correction is based, is around a factor of two higher for the DMA 3081 compared to the nano-DMA 3085. In general, DMAs can reach uncertainties of around 10% between 20 and 200 nm $D_p$ and significantly higher for smaller sizes (Wiedensohler et al., 2012). According to the manufacturer (personal communication), to be conservative one should anticipate in the size range between 10 nm and 6 nm uncertainties of about a factor of 1.5 to 2 higher for the DMA 3081 compared to the nano-DMA 3085. This is now mentioned in the text.

*3. Line 222. Appreciate the link to the data. I only found the SMPS data there. I encourage the authors to provide the time series of "concurrently measured total CP concentrations, meteorological parameters, and the ionic composition of the bulk aerosol" as well (in addition to the figures given in the supplementary material).*

We now provide respecting data sets as Excel-spreadsheets within our Supplementary Material. In addition, we just like to note at this point, that access to complete data sets from the Air Chemistry- and Meteorological Observatory can be found under:

http://www.pangaea.de/ (example: http://doi.pangaea.de/10.1594/PANGAEA.815285 )

http://www.awi.de/en/science/long-term-observations/atmosphere/antarctic-neumayer/meteorology/synoptic-observations.html

http://www.awi.de/en/science/long-term-observations/atmosphere/antarctic-neumayer/meteorology/radiation.html

*4. Lines 270-271. It appears to me that the conclusion of this sentence is not consistent with previous discussions (Lines 261-269). You mentioned that "During NPF events trajectories mainly stayed below 1500 m above ground for the last 48 hours before arrival at NM" and "Only for the NPF event at 16 March 2012 air masses clearly descended from the free troposphere". If the air masses for ALL the events EXCEPT ONE were from recent MBL, your observations did indicate "a pronounced impact of recent MBL air advection". Your conclusion should reflect this.*

We agree completely and rephrased the sentence accordingly.

*5. Equation (3). What is typical water content of nucleation mode particles? What's the contribution of water uptake on the growth rates?*

The water content is implicated by assuming a density of 1.6 g $cm^{-3}$ in equation (3). This corresponds to a 68% by weight $H_2SO_4$ solution in water (which is now mentioned in the text). Note that our calculations are just an order of magnitude estimate!

*6. Equ. (3) and Table 2. If I plug the numbers you gave into Equ. (3), I got growth rate of 0.44 nm/hr per 1E7/cm3 of H2SO4. Table 2 shows ~ 0.24-0.27 nm/hr per 1E7/cm3 of H2SO4. Did I miss something here?*

Thanks a lot for this important hint! Actually there is a typo in equation (3): There must be a factor "4" (instead of "2") in the denominator (see Yli-Juuti et al., 2011, eq. (8) therein)

*7. Lines 382-401. Ions have been well recognized to enhance nucleation (e.g., Yu and Turco, 2000). Based on the CLOUD chamber study (Almeida et al., 2013; Riccobono et al., 2014), ion-mediated nucleation is significant or dominant when nucleation rate is below ~ 1 cm-3 s-1 but neutral nucleation takes over when nucleation rate > ~ 1 cm-3 s-1. Since the observed nucleation rates at NM were well below 1 cm-3 s-1, I think that the possible role of ion mediated nucleation in the region (e.g., Yu and Luo, 2010) should also be discussed here.*

We agree that addressing the potential importance of ion mediated nucleation is beneficial and added a short paragraph at the end of chapter 4.4.

*8. Lines 430-431. This conclusion is not justified. To make such a conclusion, you need to calculate the ternary nucleation rate with reasonable values of H2SO4 and NH3 concentrations. Any information about the possible NH3 gas concentrations? Due to the cold temperature and lack of significant sources, I think that NH3 gas concentrations at NM are likely to be very low. The observed 5-9 ng/m3 of ammonium in the bulk mass is about two orders of magnitude smaller than that of MSA and nss-SO4, indicating that the particles were clearly acidic. The presence of small amount of ammonium in bulk mass doesn't necessarily imply that NH3 was involved in the nucleation as these mass might be a result of long range transport.*

In fact we can only speculate on the nucleation mechanism, which is now explicitly stated. We do not have a reliable value regarding $NH_3$ gas concentration at hand. We merely refer here to a recent article by Kürten et al. (2015) who found that $NH_3$ concentrations in the pptv range strongly enhance $H_2SO_4$ nucleation. Clearly the presence of small amount of ammonium in bulk aerosol mass doesn't necessarily imply that $NH_3$ was involved (what we did not claim), but it appears also not appropriate to exclude the impact of $NH_3$ per se.

Reference

Kürten, A., et al.: Thermodynamics of the formation of sulfuric acid dimers in the binary ($H_2SO_4$–$H_2O$) and ternary ($H_2SO_4$–$H_2O$–$NH_3$) system, Atmos. Chem. Phys., 15, 10701-10721, doi:10.5194/acp-15-10701-2015, 2015.

**Responses to referee #2:**

First of all we would like to thank again referee #2 for his effort in evaluating our revised manuscript!

*I have only one minor comment. When specifying measurement range for SMPS, please use integral nm. There is no instrument available with precision one hundredth of nm.*

This is now corrected in our most recent version.