

Author's Response to Reviewers

Dear Editor, Referee,

Please find enclosed our detailed answers to minor revisions suggested by the referee.

We thank for excellent work done for improving our manuscript and hope you find our answers satisfactory.

Our answers to Anonymous Referee #1

Once again, we're very grateful for careful review of our manuscript. Thank you very much for your comments. We've done our best to address each point as stated below.

1. a new version of the SI is not available at the moment. at least i could only find the old one which was rather sub-par. i hope the one that will eventually be published will be (much) better than the initial version.

REPLY: We hope so too. At least, now the supplement is all in one pdf-file.

2. p. 4 l. 8: in the Eurasian side -> on the ...

REPLY: Corrected.

3. p. 4 l. 18: fulfill -> fill

REPLY: Corrected.

4. p. 5 l. 18: "well above the treeline": of course the site is not above the treeline but north of the treeline. only a mountain site can be above the treeline. north may be up on most maps but that still doesn't mean that north and up or above can be used as synonyms in a scientific text.

REPLY: True, corrected.

5. p. 6 l. 3: isn't the inner diameter much more relevant information than the outer diameter?

REPLY: Yes it is. I.D.=12 mm was added in text.

6. p. 7 l. 27: the accuracy of the numbers given for the correlation coefficients seems a bit much. please also check the rest of the manuscript and make sure to provide only meaningfully rounded numbers.

REPLY: True. Last digit was removed from these values.

7. p. 9 l. 18: "zero-intercept forcing": why exactly? a non-zero intercept can give instrumental insight in the same way that a non-unity slope can. why force it? unless the measured size ranges are perfectly identical, a non-zero intercept is actually quite impossible and i wouldn't expect it at all. how is this forcing justified?

REPLY: In our opinion, the zero-intercept forcing is physically best justified, and looking at the data supported this linear correlation. Basically, we assume that the percentage difference in concentrations between CPC and DMPS remains constant, i.e. in this case DMPS shows typically 98% of the concentration of the CPC. This could be caused for example by higher losses in DMPS. To our view it is much more difficult to justify a situation where this intercept would be non-zero, assuming instruments were working properly.

8. p. 11 section 2.3.5: much along the lines of the cited beddows paper, it would be nice to have some information on the quality of the clustering. how uniform are distributions within each cluster? this and similar questions could be addressed. i still feel that this cluster business is more or less dropped on the reader without discussing underlying motivations and choices.

REPLY: We appreciate the reviewer view on this, however, considering the request to keep this cluster business as minor part of the manuscript as possible, we feel that further

explanations on inner-cluster variability would not help to meet this goal. Inner-cluster variability is partly addressed in Tables 2 and 3, by giving the standard deviations of particle number and mass within each of the clusters. We hope that referee understands this choice.

9. p. 12 l. 10f: "also extra-modal coagulation losses": i don't understand this phrase at all. modes are not involved when calculation the CoagS for a given particle size. what then is this supposed to mean?

REPLY: We calculated the formation and growth rate of nucleation mode particles. The coagulation sink was calculated using the mean diameter of this mode with all the particles with diameters larger than the upper diameter of the nucleation mode. So no intra-modal coagulation included (since this is believed to be very minor). To explain this terminology, we added: "losses of nucleation mode particles to bigger particle sizes".

10. the two clusterings and their analysis are still somewhat confusing. on page 15, the N-cluster 4 is related to sec. part. form. and has mostly marine influences. on the next page, the M-cluster 3 represents sec. part. form. and continental air masses. while i do understand that both those things can be true despite the superficial contradiction, this underlines what i have said in the first review: the benefits of the clustering business are somewhat nebulous. in the clusters, various factors are mixed together and then have to be de-entangled one way or another. compared to continental vs. marine split up by seasons and BB separated, what is it that we learn from this cluster analysis?

REPLY: While N-cluster 4 is very specific to fresh secondary particle formation (and marine), M-cluster 3 has very small nucleation mode concentration. Instead, M-cluster represents more aged secondary aerosol (continental). To us this is interesting to see how clustering the size distributions actually divide them not only by shape, but also indicates different properties and histories of particles as seen with other parameters. We find the

clustering gives more complete information on each of the cases, helps the reader to understand the variability in size distributions and the factors behind this variability, yet presenting this relatively compact with only one chapter. Making analysis on the impact of each of the variables separately would need much more space. It's also not meaningful because all of those factors affect size distributions simultaneously, which would in fact require highly developed multivariate analysis tools.

We also see that this clustering chapter kind of stands as an introduction for the remaining, more specific, analysis of the particle sources.

11. p. 17 l. 24: reassembles -> resembles

REPLY: Thank you, corrected.

12. p. 18 l. 1: "other arctic sites": how would a comparison to antarctic sites look like?

REPLY: Thank you, this is a good idea. We decided to limit the comparison of particle formation parameters to other Arctic sites because Antarctica is a completely different environment with different precursors and sinks. Also very few seasonal studies on secondary particle formation exist from Antarctica. However, what is common in most places around the world (and also in Antarctica) is the need of sunlight for any significant particle growth to occur. The slowly growing winter-time events in Tiksi, which have also been seen in Antarctica during polar night, are therefore an interesting observation. For this we added some text and two recent references from Antarctica (Järvinen et al., 2013 ACP and Weller et al., 2015, ACP).

13. p. 18 l. 8: "Average annual growth rates [...] were only seen in summer months": this doesn't seem to make much sense. annual growth rates in the summer months? what's annual growth rates anyways in this context?

REPLY: Thank you for this correction. Text was modified to say “average growth rates of.. were only seen in summer months..” instead of average annual, which really doesn’t make any sense here.

14. p. 18 l. 17: inter-annual -> annual

REPLY: Corrected.

15. p. 18 l. 20: "regional-scale": how do you know?

REPLY: By their characteristics, events don’t look like local particle formation (they appear for long-time with growing particle mode).

16. p. 19 l. 19: "time [s]": no, that can't be right. the unit, i mean. and the respective figure has [h] which makes more sense.

REPLY: True, we’re sorry for this mistake. Corrected.

17. p. 19 l. 24: "it must be bear in mind" -> probably "it must be kept in mind"

REPLY: Corrected.

18. p. 19 l. 25f: "data suggests rather exponential than linear dependence": then why not fit such a function and see how well it works out? would the R^2 be better?

REPLY: There was an additional fit in previous manuscript version, but having separated the BB cases and no-BB cases, the figure becomes too messy if plotted. But we did made the exponential fits anyway to answer this question and yes, R^2 values improved to 0.93 and 0.94 (for BB and no-BB cases). This is now added in manuscript text. For some

reason also the R2 in Figure 10 was too low as recalculated. We apologize for this mistake. The correct values are now added in figure caption: for BB 0.84 and for no-BB 0.88.

19. p. 20 l. 18f: "beta value of 0.126": since temperature has a unit, beta needs one, too, otherwise exp doesn't make any sense.

REPLY: Thank you, unit 1/C added.

20. p. 22 l. 15: "is Tiksi" -> in Tiksi

REPLY: Corrected.

21. p. 22 l. 17f: "Some sporadic events are also observed during dark winter months.": does that mean that you have nucleation without any sunlight? really? surely that would deserve a bit more discussion than a half-sentence in the conclusions, no?

REPLY: Yes, agree. We completed the sentence: "...providing evidence that the formation of secondary particles can take place even in the lack of photo-chemistry"

22. p. 23 l. 1: "all the way up to the Arctic": again, the Arctic is north, not up.

REPLY: Corrected.

Figure 1: the map is still kinda tiny

REPLY: Which map? The topographic or the underlying map? We made the underlying map larger based on previous comments, and added one regional topographic map to supplement.

Figure 2: fonts generally borderline-tiny. in the bottom two panels on the right far too small font. also: those two panels would probably be much easier to read if e.g. continental/marine went up/down from the same baseline. now the straight line is at the bottom, it might be better in between. finally, the left axis numbers overlap between panels. please fix.

REPLY: Font size was increased in legend and in y-axis. Y-axis overlapping numbers were corrected. I'm not quite sure what referee means with the comment on straight line?

Figure 5: why does the aps part of the lower panel have different crosses for the most common cluster?

REPLY: For no reason, this was corrected. Thank you.

Figure 6: could use some color. really hard to read in print.

REPLY: Colors added.

Figure 7: formation rate needs to be more specific. this is J₇, right? also in the text, this needs to be clear. and, btw, why not calculate the more relevant J_{1.5} or J₂? according to the kulmala protocol, this should be standard. and it also makes it easier to compare results from different sites/instruments with different cut-offs.

REPLY: This is not exactly J₇. Only Class 1 events are always J₇ (i.e. we see them starting at the lowest measurable diameter). In most cases this is close to J₇ (or even equal) but for clarity we added a note in table and in manuscript text: particle formation rates were calculated for the lowest observed particle size, which was not however in all cases the lowest measurable size of 7 nm. Typically the difference is not large (see table

4), also because we're dealing with FR7 and not with FR1.5. Calculating back to FR_1.5 would therefore be possible only for class 1 events, but I still find it a bit questionable (assuming we understand the growth of the particles between 1.5 and 7 nm, which is not true especially in the Arctic).

Figure 8: the text above the panels is ridiculously small. please enlarge.

REPLY: This was done.

Figure 10: font size is borderline, again. the color code on the left is hard to make out. please consider larger dots with thinner edges.

REPLY: We increased the font size and modified the dots.

Figure 11: all those numbers inside the plot on the left are impossible to read without lots of zooming. doesn't work in print.

REPLY: The numbers are not really that important, just additional information (in which case you have the zooming option). Or should I take the numbers completely out?