Reply to the comments of Reviewer #1

The paper by Heintzenberg et al. tries to uncover potential aerosol source regions by using clustering techniques. It is an informative paper capitalising on numerous research cruises into the Arctic ocean on board the icebreaker Oden. The Arctic Ocean is an important region both due to proximity of Northern Hemisphere land masses all of which contribute to aerosol pollution perturbing otherwise pristine marine environment.
At the same it is quite difficult to separate those contributing sources due to their diversity and intersecting air masses. The paper would be suitable for publication in this journal after addressing rather numerous comments. Last but not least, English of the manuscript makes following the text rather difficult.

Major comments
The main weakness of the paper is that it lacks clarity enormously throughout.
See our quotes from two other reviews below

Even the title is not very precise as the paper presents data not just of particles, but precursor gases as well (DMS).
The DMS data are only used to test the clustering algorithm. Because of that and not to over-freight the title we disagree with the reviewer on this point.

The whole paper is in-concise, lacking focus and containing weakly supported statements.
Here we would like to cite two quotes from the two other available review of our paper:
Reviewer #2: "*The analysis appears to be scientifically sound with no major errors. The paper is well structured and clearly written.*"
Reviewer Leaitch: "*Clearly a lot of effort has been put into the analyses making this a valuable piece of work.*"

Overall, the paper discusses source regions more than aerosol processes which are often implied or invoked.
This is true but not necessarily incriminating for want of further process information.

The third sentence of the abstract states about identification of five source regions and three aerosol types, but they are not listed/named. Instead there are long passages discussing *selected sources or processes. The main hypothesis at the end of the abstract is badly worded: the long travel time over pack ice and?? open water cannot control formation of ultrafine particles, but instead authors are probably arguing for fragmentation process taking place. Moreover, identifying a specific region with distinct aerosol size distribution does not necessarily mean a source region, but instead may indicate certain processes taking place in those specific regions or en-route to them: BL dynamics, aerosol activation and deposition, nucleation, fragmentation, primary , secondary production, so on and so forth.*

We apologize for the confusing abstract and hope that the revised version (with the addition requested by reviewer #2) will be clearer.

Introduction is supposed to be a brief summary of the latest findings and experimental techniques to be developed, but instead there are lengthy paragraphs arguing in favour of already published peer-reviewed papers. Many sections of the introduction should be moved to discussion while a short summary of relevant results should be mentioned in the introduction. No overview of clustering techniques is provided in the introduction despite the fact that clustering algorithms are numerous, results abound and they are quite central to the paper.

Following the suggestions of reviewer#1 we shortened the introduction to 60% of its original length including the addition that reviewer#2 requested. In order not to increase the length of the introduction further we added to the beginning of the section concerning the description of the clustering algorithm to "Many clustering approaches have been developed in exploratory data analysis (Jain et al., 1999). In atmospheric aerosol research they are used to find groups of similar aerosol data, particle origin or formation." For further background information we refer to Heintzenberg et al. (2013).

Line 93. What inconsistency the authors are talking about? Statistical interpretations arise from analysing direct observations, so that is one and the same. The derived result cannot contradict the original, otherwise something is wrong with the statistical technique.

The incriminated text has been eliminated while shortening the introduction.

Line 108. How can DMS directly condense on the particles? DMS derived products like SO_2 or H_2SO_4 can either directly or through cloud processing (aqueous reactions) become incorporated into droplets.

We did not make any such statement. Instead we wrote "Heterogeneous condensation and aerosol cloud processing occurs when the oxidation products of dimethyl sulfide (DMS) released by phytoplankton advected from open waters south of and along the marginal ice edge, (Leck and Persson, 1996a), condense on non-activated particles which then are incorporated into cloud droplets", which is correct.

Using 5day long trajectories is quite inaccurate when it comes to their origin. Typically, trajectory uncertainty can be anywhere between 15-30% of the travelled distance (http://www.arl.noaa.gov/faq_hg11.php) and consequently travel time over pack ice or open water highly uncertain too for trajectories of e.g. 1000km or longer. While the authors acknowledged the uncertainty (without reference, only by assumption) there is no discussion about the implications on the time trajectory spent over water or pack ice.

The original manuscript shows that we were quite aware of and explicit about trajectory uncertainties. In order to clarify our approach and intentions we revised the related paragraph to "We are aware of the limitations in trajectory accuracy. On one hand the data sparse Arctic region limits the validity of the meteorological fields on which the trajectory calculations are based. On the other hand, out to the nearest continental borders the meteorological setting, surface conditions and the resulting atmospheric fields in the central Arctic are relatively simple. Figure 9 in Leck and Persson (1996b) shows an example where the trajectories were able to resolve an influence of the settlements Barentsburg and Longyearbyen on Spitsbergen in the measurements onboard *Oden* which was located near the North Pole. If we assume some 30% position uncertainty relative to the trajectory length yielding on average 3000 km for a ten-day back trajectory (cf. Stohl, 1998) this will in general not allow us to differentiate between distant regions such as Beaufort Sea, Chukchi Sea, and Laptev Sea outside the pack ice. A distinction between these sees and Kara Seas is however possible. The meteorological information calculated along the trajectories was utilized in the analysis.

Instead of discussing paths of uncertain individual trajectories we plotted geographic results on maps of stereographic projection centered on the North Pole. These maps were covered with a coarse grid of 35 x 39 geocells, in which the passage of trajectories or the occurrence of other results of this study were counted. Fig. 2 shows that the geographical region covered by the back trajectories extends to and partly beyond the pack ice limits of the studied summers." Note that we added the review reference Stohl, 1998.

Section 4. The section title is missing clustering type (trajectory, I guess). This section demands that the title of the paper is modified to include "gaseous aerosol precursors".
The section title was changed to "Test of the trajectory clustering with DMS". With one exception (see below) the DMS data are only used to test the clustering algorithm. Because of that and not to over-freight the title we disagree with the reviewer on this point.

Despite obvious connection of DMS with aerosol particles there is no discussion of that relationship. Incidentally mentioned Tables 2&3 contain relevant information of particle size distribution clusters, but these are not discussed in connection to DMS while they should be.
With one exception, i.e. cluster experiment "marginal ice", the DMS data are only utilized in connection with the test of the clustering algorithm. In both cases we quantify and discuss cluster-median DMS-concentrations in the revised text but have eliminated DMS from the revised Table 3.

Section 5. The section starts with optimistic note that clustering worked, but missed to name them accordingly which makes the following text difficult to follow. There is little discussion of the observed differences between the clusters. For example, can the difference between the number concentrations of clusters 4&5 be at least partially attributed to anthropogenic activities? And many other similar questions: when the particles are called aged (line 487) are they biogenic or anthropogenic and which substances exactly became aged?
The "optimistic note" concerned the test of the algorithm with DMS in section 4 and not the clustering in section 5. In Fig. 5 the differences of the size distributions in clusters 4 and 5 are shown to be within the uncertainty limits of the two distributions. Here we only discuss strong differences with respect to the distributions in Fig 5a, and b. We have no arguments to differentiate between anthropogenic and biogenic at this stage of the paper.

Section 6. For comparing size distribution data between ice-breaker and e.g. Zeppelin station it is imperative to have a connected Lagrangian flow. Was the time lag applied considering the distance between the two sampling points? If not, spectral differences are difficult to interpret as to what was the cause and the outcome rendering any connection to aerosol processes. The whole section needs much more careful wording as to not overstate the findings.
Yes it was: "Size distributions measured on *Oden* at the time of minimal distance were compared to size distributions measured on Mt. Zeppelin at the time of trajectory arrival."

We checked the incriminated text according to the comment but could not find any wording that we were able to choose more carefully.

Conclusions and synopsis should not include lengthy discussions with references to the Figures and Tables as those sections belong to discussion. I suggest breaking section 7 into two: Discussion (7) and Conclusions (8). The latter will summarise the findings and will inform the abstract which is very loose at the moment.
The suggested change of section 7 seems to be based on the personal taste of the reviewer. We would like to leave it up to the editor to decide if our choice of structuring section 7 needs revision.

Minor comments
Line 62. I don't understand the sentence "A plume to be entrained: : :is brought down to its top".
This is a sentence in the initial manuscript that has been revised and clarified before publication in ACPD.

Line 65. The sentence belongs to discussion and above all is highly unclear. There are numerous papers demonstrating traces or a more significant pollution carried to the Arctic environment. Not measuring light-absorbing carbon particles was due to the lack of sensitivity of measurements? Even in a far more remote Antarctica there are measureable levels of light absorption.
We do not know which "numerous papers" the reviewer refers to but a) most papers dealing with pollution carried into the Arctic concern Arctic haze during winter, and b) there are no papers demonstrating pollution carried into the **Central Arctic** boundary layer because there are no other measurements besides our icebreaker data. Our light absorption measurements work down to nanograms per cubic meter (Heintzenberg, 1982).

Line 84. Please correct "presence of bubbles" to "bubbles generated by wave breaking/ air entrainment". Bubbles are not just present in water they appear there.
After they "appeared" they were present and were measured by us in number concentration (Norris et al., 2011).
Line 97. The sentence starts with "the same: : :" when biological processes were not discussed previously.

The incriminated text has been eliminated while shortening the introduction.

Line 118. The sentence refers to unspecified time period. New particle formation events do not occur as an increase in particle concentrations, but rather manifesting themselves as an increase.

Thank you for the suggestion. The sentence was changed to "However, these events often manifested themselves as a simultaneous increase of particle number concentrations…"

Section 2 Database should be renamed to "Sampling techniques on ice-breaker Oden" as there is no database mentioned here.

This comment seems to stem from an earlier version of the manuscript

Line 215. Please specify data cleaning procedures. Why was it necessary if sampling section refers to pollution controller?

A sector control is not enough to exclude emissions from the ship stack that may reach the inlets on tortuous paths. Signals from a fast Ultrafine particle counter and from a fast mass spectrometer were utilized as detailed in Heintzenberg and Leck (2012), which we quoted.

Line 445. The ice maps are called "controlling factor" without proving it first. Controlling factor may be ok in the conclusions, not at the start of discussion.

We added "potentially" to "controlling factor".

Line 630. How anything measured during different times can confirm? The observations may be indicative or supporting, but not confirming.

The length of a bar of steel measured at different times can confirm the stability of the bar.

Line 634. There is no inconsistency when the measurements do not agree with the mechanistic model, but rather point to knowledge gaps.

We agree and changed the sentence to "Conventional nucleation paradigms (Karl et al., 2012) fail to explain observations of small particle formation over the inner Arctic and those south of the pack ice."

Literature

J. Heintzenberg, Size-segregated measurements of particulate elemental carbon and aerosol light absorption at remote Arctic locations., *Atmos. Environ.* **16**(1982), 2461-2469.
J. Heintzenberg *et al.*, Mapping the aerosol over Eurasia from the Zotino Tall Tower (ZOTTO), *Tellus B* **65**(2013), doi:http://dx.doi.org/10.3402/tellusb.v3465i3400.20062.
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Reply to the comments of Reviewer #2

This manuscript analyses aerosol particle sources over the Arctic Ocean during summertime based on in situ measurements conducted during several cruises. The analysis appears to be scientifically sound with no major errors. The paper is well structured and clearly written.
Thank you for the encouraging review.

I have a few suggestions for improving the paper a bit further.
The last two paragraphs of "Introduction" mainly list the contents of the paper. Preferably, scientific goals of this paper should be listed as well. What is the main goal of the paper? Which questions the paper is searching for answers?
At the end of the revised and shortened introduction we added "With the combined data set and the clustering algorithm the main goal of the present study is to identify potential source regions of aerosol particles observed over the central summer Arctic. Specifically, we would like to differentiate between local sources within the pack ice region and distant sources. Extending our previous analyses discussed above with the locally measured parameters to different source regions we aim at identifying factors controlling the aerosol life cycle over the inner Arctic."

The authors test their clustering algorithm in section 4. Rather than just saying "lending confidence in" at the end, I would recommend adding a short (2-3 sentences) summary of the outcome of this testing exercise. Now the reader needs to make his/her own judgment of whether and how well the chosen approach performs.
Instead of the last sentence of section 4 we now state: "The test of the clustering algorithm with all available DMS(g) data has the following outcome: The potential source regions identified by the algorithm in the MIZ and adjacent open waters agree with previous DMS studies. Consequently, we expect the clustering algorithm to be able to identify other potential source regions of the surface aerosol over the Arctic summer pack ice."

The last section of the paper (synopsis and conclusions) would benefit of having a paragraph discussing the major implications that the findings made here might have in terms of the Arctic climate system. One line related to this issue could also be added to the "Abstract".
We gladly take up this suggestion and formulated: "What are the possible implications of our findings for the Arctic climate system? In the course of the ongoing reduction of the summer pack ice favorable biological conditions for new particle formation might increase over the Central Arctic with more frequent broken-ice or open water patches. More open water increases biological activity in surface water promoting the formation of biological particles. Consequently, number concentrations of small particles might increase over the inner Arctic. Provided that enough condensates are available, e.g., DMS oxidation products or emissions from increasing Arctic shipping, more cloud condensation nuclei might result, which would affect the prevalent low clouds and fogs in the summer Arctic. Changing clouds would affect the surface energy balance, which in turn would have effects on ice melt."
We added to the Abstract: "Future more frequent broken-ice or open water patches in summer will spur biological activity in surface water promoting the formation of biological particles. Thereby low clouds and fogs and subsequently the surface energy balance and ice melt may be affected."

Minor/technical issues:
Page 8449, line 21: should maybe be "derived".

Yes, thanks, changed.

Page 8454, lines 8-9: "one" and "two" are bit strange choices of words here. Maybe something like "The first case" and "The second case".
Yes, thanks. We changed the sentence to "The first case covers polluted North Atlantic air that had passed over Svalbard (cf. Fig. 7b). The second case covers free tropospheric air that had crossed Greenland before arriving at *Oden* (cf. Fig. 7c)."

Page 8455, line 1: "super-micrometer particles", particles missing?
Yes, thanks, "particles" added.

Reply to Richard Leaitch's comments

This paper examines aerosol size distributions measured in the high Arctic from the Swedish icebreaker Oden during four summers. Back trajectory cluster analyses are used in combination with the size distribution information and ice cover data to examine the regions associated with features of the size distributions up to 10 days prior to the distribution measurements. Before application to the aerosol particles, the cluster analysis is tested using measured DMS, which reasonably shows higher concentrations of DMS tracing back to known DMS source regions. Clearly a lot of effort has been put into the analyses making this a valuable piece of work.

Thank you for the encouraging review.

A few specific comments follow:

1) Concerning section 7

a. In section 7, you say that "Previously reported results from Alert in spring, (Leaitch et al., 2013), and on Mt. Zeppelin, Spitsbergen in early summer, (Engvall et al., 2008), showed nucleation events followed by subsequent prototypical "banana growth" (e.g., c.f. Kulmala et al., 2001), which the authors explained by solar radiation in concert with the presences of precursor gases and attendant low condensational sinks." The Alert data in the Leaitch et al paper referred to focused on the period June, July, August and September. It was summer not spring. Also, there was no mention of a banana growth in Leaitch et al because none was observed. Please correct.

Sorry about the seasonal confusion which we corrected. The text now reads: "Previously reported results from Alert in summer, (Leaitch et al., 2013), and on Mt. Zeppelin, Spitsbergen in spring, (Engvall et al., 2008), showed nucleation events. On Spitsbergen they were followed by prototypical "banana growth" (e.g., c.f. Kulmala et al., 2001). The nucleation events at both Alert and Zeppelin are explained by a conventional nucleation mechanism involving solar radiation in concert with the presences of precursor gases and attendant low condensational sinks."

b. Later in section 7, you say that "Possible reasons for the inconsistency with the data collected during the four icebreaker expeditions could be that the DMS source and photochemical sink generating the precursor gases for nucleation and early growth is both seasonal and temperature dependent (Leck and Persson, 1996a, b; Kerminen and Leck, 2001; Karl et al., 2007, 2012). Given that, perhaps the main difference between the studies concerns how efficiently nucleation and growth of particles resulting from DMS oxidation are predicted by the choice of model and lack of observations to constrain the model assumptions." You have not demonstrated an inconsistency among the datasets. It is quite the opposite. Your analysis shows that all measurements you have used are relatively consistent. Even the trajectory analysis for the Alert data included in Leaitch et al showed the central Arctic (as well as air off Greenland) to be potential source regions, and your results, including those associated with Zeppelin as well as with Alert, indicate the presence of smaller particles when the condensation sink is reduced. Where there is an inconsistency is in the interpretation of "Trajectories connected with high concentrations of newly formed small particles, however, experienced more open 15 water during the last four days before arrival in heavy ice conditions at Oden." There seem to be two possible explanations to the presence of the newly formed particles. Your interpretation is that it is due to the fragmentation of microgels connected with cloud processing. The other (more conventional) interpretation is that it is due to nucleation of new particles for situations of very low concentrations of precursor gases that is facilitated by a low condensation sink. However, at the moment the

real interpretation seems to be that we do not know which answer is correct, if either, and I hope the authors will consider adjusting section 7 to reflect that lack of knowledge.
We hope the reviewer can live with the following revised text: "A major difference between the two land stations and the inner Arctic lies in the different DMS levels. To our best knowledge (Karl et al., 2013) the extremely low DMS concentrations, (Leck and Persson, 1996a, b), in the inner Arctic are not sufficient for the conventional nucleation mechanism. Given that, perhaps the main difference between the studies concerns how efficiently nucleation and growth of particles resulting from DMS oxidation are predicted by the choice of model and lack of observations to constrain the model assumptions."

2) Page 8433, line 22 – "occasionally TO as few as. . ."
Yes, thanks

3) Page 8436, lines 5-7 – I do not understand this sentence: "As compared to the 1271 hourly DMS values. . .a total of 2035 h of DMS data were available. . ."
One, DMS measurements had been taken at times when the DMPS instrument was not operating. Two, because of the stringent contamination criteria applied to the DMPS data, some aerosol data were rejected while DMS data could be accepted.

4) Page 8443, line 21 – week.
Yes, of course, thanks.

5) Page 8447, lines 7-9 – I don't see how these number concentrations, which are really quite modest (130/cc) are an indication of polluted air. The sizes of these particles are mostly below 50 nm diameter and almost all smaller than 100 nm diameter, which means that the associated mass concentrations are very small. Why could this not be an indicator for new particle formation with modest growth over the Greenland ice cap?
Thanks for the suggestion. We corrected the sentence to "This monomodal distribution may be the result of very long aging of polluted air in the free troposphere (e.g., Leaitch and Isaac, 1991; Parungo et al., 1990) or may indicate new particle formation with modest growth over the Greenland ice cap."

6) Page 8447, line 22 – "strongly reminds of the. . ."
Changed to: "strongly reminds us of the"

7) Page 8455, line 15 (acknowledgements) – Richard not Robert.
We are sorry, Richard.

Literature

A.-C. Engvall *et al.*, Changes in aerosol properties during spring-summer period in the Arctic troposphere, *Atmos. Chem. Phys.* **8**(2008), 445-462.
M. Karl, C. Leck, E. Coz and J. Heintzenberg, Marine nanogels as a source of atmospheric nanoparticles in the high Arctic, *Geophys. Res. Lett.* **40**(2013), 3738–3743.
M. Kulmala *et al.*, On the formation, growth and composition of nucleation mode particles, *Tellus* **53B**(2001), 479-490.
W.R. Leaitch and G.A. Isaac, Tropospheric aerosol size distributions from 1982 to1988 over Eastern North America, *Atmos. Environ.* **25A**(1991), 601-619.
W.R. Leaitch *et al.*, Dimethyl sulfide control of the clean summertime Arctic aerosol and cloud, *Elem. Sci. Anth.* **1**(2013), 000017.

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