

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 Leitch: *“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 SO₂ or H₂SO₄ 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 seas 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.
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