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Comment

Interactive comment on “Iodine observed in new particle formation events in the Arctic atmosphere during ACCACIA” by J. D. Allan et al.

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Reviewer's comments in **bold**

Have the authors considered photolysis of iodocarbons as a source of the observed iodine? Can this be excluded? In addition, it is said that halocarbons were measured from air and water but no iodocarbon data are presented (only CH₂Br₂). Why is this so? This data could potentially strengthen the presented results.

No significant fluxes or concentrations of iodocarbons were detected on this cruise. These data are now included in the supplement, which has also necessitated the inclusion of two new co-authors (S. J. Andrews and S. C. Hackenberg). While it has been

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Interactive Discussion

Discussion Paper



shown that iodocarbons can act as a source of nucleating IO in the laboratory (e.g. Jimenez et al., 2003), as discussed in the paper, they have shown to be insignificant compared to molecular iodine in other environments.

Iodine emissions have been quite conclusively shown to lead to NPF, but often the halogen chemistries are coupled to each other. Was bromine or its oxidation products sought from the AMS spectra? Same (or similar) algae that produce iodinated species are known to produce even bigger amounts of bromine species.

There was no evidence of bromine compounds being present in the AMS data. Also, in contrast to iodine, bromine does not form stable oxides in the particle phase. This is pointed out in the revised version.

The time period of the case-study highlighted in Figure S1.2 shows a very large portion of organic matter. Is it, or can it be excluded that some larger (and/or heavier) halogenated organic compounds contribute directly to the phenomena, or is it necessarily I₂ and its further oxidation products? I suggest this should be discussed in more length in the text.

As already discussed in the manuscript, the organics present include misattributed sea salt, MSA and OOA. Given that the AMS destroys much chemical information through its vaporisation and ionisation processes, it is impossible to rule out the possibility that the OOA fraction contains halogenated organics. However, one might expect ions such as Br⁺ to be present, which they are not. Furthermore, OOA is usually associated with heavily functionalised, higher molecular weight species than what would be expected from the gas-phase oxidation of biogenic halocarbons.

in Page 28956, Line 15 it is said that: “The I⁺ signal was not represented in any of the factors derived, only manifested in the residual data, which implies that the particulate iodine had a source that was distinct from the processes controlling the formation of particulate organic matter”. Maybe it would be worth to mention here about the size limitation of AMS, and on the timescales the particulate

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matter has needed to grow to be detectable by the AMS. In addition, I would like to see a sentence about the PMF analysis in the main text describing how the different sources are decided based on the data and the PMF analysis.

The sentence regarding PMF is added as suggested. The size limitation is explained in the paragraph above and further explained in the revised version.

I would suggest adding a Figure showing some of the basic meteorological parameters during the case study. I guess these were also routinely measured during the campaign? For example, temperature and humidity could be useful. Even better if solar UV flux measurements are available (as comments above show), or, even just the indication of ‘cloudy or sunny’ would make the estimation of the importance of photolytic processes easier. Iodine species are known to be especially photochemically active and the study was performed during the Arctic summer, when the sun never sets. Thus it would help to know about these parameters.

Temperature, humidity and total incident radiation added to the new figure 2.

Figure 2. The ^{127}Th signal seems to have a time lag with respect to observed NPF event. If iodine is the initiating compound, then how come its observation has a time lag? Comments?

The AMS does not detect iodine until significant amounts of mass reside in particle greater than 30 nm. This will not occur until significant amounts of growth have occurred after the initial nucleation. This is reiterated in the revised version.

Page 28950, Line 8: What is a persistent event in this context? Suggest adding an explanation. (cf. Page 28952, Line 21: “a particularly strong and persistent case study..”?)

By persistent, we mean that the phenomenon was evident in the data for a protracted period. This is clarified in the revised version.

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Interactive Discussion

Discussion Paper



Page 2891, Lines 22-24: This sentence is hard to understand. Suggest rewriting it.

Revised as suggested.

Page 28952, Line 1: I would add few words explaining the concept of “organic biogel”.

This is explained in the revised version as aggregations of biological macromolecules.

Page 28955, Line 14: Suggest giving a short description already here on what is meant by “ice diatom” (this is explained later in page 28958). For people familiar with the topic, this is trivial, but for interested non-specialized reader, especially the word diatom can be confusing.

The term is replaced with ‘microalgal’ here for the sake of brevity. Later in the manuscript, the following is added: "these are prominent members of microalgal blooms occurring at the receding ice edge, and also in communities growing within the ice itself"

Page 28955, Line 23: To me the presented Figures show quite banana type events.

The following behaviours noted in these events distinguish these from the ‘banana’ events noted at Hyytiällä and other sites: 1) There are breaks in the growth observed (e.g. 25 Jul 00:00) 2) The apparent growth around 26 Jul 20:00 is exponential rather than linear in diameter space 3) The growth apparently reverses at 26 Jul 00:30 and 27 Jul 09:00. These are pointed out in the revised text.

What is meant by saying (Line 25): “result from diurnal modulation of boundary layer dynamics and photochemistry, which is missing under the continuous insolation of the Arctic summer.”? If the meteorological situation is so different, maybe an explanation is required to explain why the observed events look so similar.

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Interactive Discussion

Discussion Paper



The text is rephrased as follows for brevity: “. . .the previously reported behaviours result from the diurnal modulation of boundary layer dynamics and photochemistry caused by the local day-night cycle in conjunction with a stable source footprint. This is not the case here, due to continuous insolation and reduced dynamics of the marine boundary layer, combined with a varying source footprint (due to the movement of the ship and varying wind direction). Therefore, analogies with the temporal behaviour at other locations cannot necessarily be drawn.” This statement is intended not to specifically contrast the two cases, but to explain why the apparent growth around 26 Jul 20:00 cannot be assumed to be indicative of in situ growth, as is done at other sites.

Page 28958, Line 24: “There are also sources of atmospheric iodine in the form of iodocarbons, but we do not consider these likely to be responsible for the observations, as it has been shown that these have a much lower NPF potential compared to I₂ (McFiggans et al., 2004).” This relates to comment presented already above: Iodine bearing hydrocarbons are very photolabile and thus some information on available radiation would be useful to estimate the importance of photochemistry of the iodocarbon species.

See response to point above. Note that McFiggans (2004) reference took account of the photolysis yields of iodocarbons relative to iodine and found them to be orders of magnitude different.

Figure 1 The Figure size should be increased to enable better readability.

The size of this figure is dictated by how the online PDF was generated, which we had little control over. We envisaged this occupying the full width of the final ‘printer friendly’ version and will naturally ensure that it is clear when the proofs are generated.

Figure 1b Include arrows to indicate wind direction for the trajectories.

We feel that the wind direction, i.e. towards the ship, is unambiguous and arrows would only serve to provide additional clutter. Note that it is not standard practice to use such

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arrows when plotting back trajectories, such as those generated by the commonly-used NOAA READY online HYSPLIT service.

Figure S2.2. More details on the mass-axis would benefit the reader. At the moment it's not possible to read the masses of the observed peaks.

These have been made clearer in the revised version.

Many of the supplemental figures are not mentioned in the text.

The S1.x figures are referred to in the main article and all of the S2.x figures are referred to within section S2. The latter are included to document the process performed to generate the PMF outputs and take the form of standard diagnostic outputs from the PET toolkit. They do not have a direct bearing on the text, but are still important to document in the interests of full disclosure and replicability. This is standard practice in PMF papers.

Technical corrections:

Page 28951, Line 15: Should it be “atmosphere”?

Corrected

Page 28957, Line 9: Should it be “Aitken”?

Corrected

Figure 3 is apparently mislabelled as Figure 4 (mentioned in Page 28957, Line 1).

Corrected

Supporting Figure S1.4 caption: case study time period is not found on the reported Figure S1.3.

Removed reference to S1.3, as the case study period is specified anyway.

Add spaces between supplemental references to enable “better browsing”.

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Interactive Discussion

Discussion Paper



Corrected

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Interactive
Comment

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Interactive Discussion

Discussion Paper

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