Review Response: The complex response of Arctic CCN to sea-ice retreat
(Now: The complex response of Arctic aerosol to sea-ice retreat)
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We thank the reviewer for their comments. Our response is below, original review comments are shown in italic, while suggested alterations to the manuscript are highlighted in bold. Line and page numbers will reference our manuscript submitted in response to review.

Response to Anonymous Referee #2

The paper is difficult to read, or at least reading it is hard work. Specifically, one has to read the figures, e.g. Fig. 3, and the maps/color bars/details and Figure captions very carefully, while frequently going back and forth between the text (esp. that explaining the four core simulations on page 17093) and the figures, to follow the discussion.

Our paper addresses a complex collection of processes. However, we agree improvements could be made to improve the clarity of our study. Specific points (referred to by the reviewer) are addressed below, but in summary:

- Section 7.2 (Changes in CCN concentration) has been re-written, reducing references to run labels (i.e **in no-ice** changed to **after ice-loss**)
- Large CCN/accumulation+ mode particles now defined as **N100** throughout paper
- **Figures 4-5** have been updated with clearer labelling and improved colour bar
- Likewise**, Figure 9** has been improved
- Marine organic runs (PD-MOC, MOC-no-ice) have been removed from **figure 7** to improve clarity and clearer labels added. Line colours are now also consistent with figure 3
- **Table 1** has been simplified and its caption has been improved
- **Captions have been improved (Fig 1-9, table 1-2)**

The paper needs to discuss how well the model treats precipitation scavenging of aerosols in the Arctic, with appropriate references including discussion/recognition of uncertainties in this component of the model. An obvious issue is the very large increase in latent heat flux in a no-ice world, and how that would impact the results; would it represent a large increase in precipitation and precipitation scavenging, so that the surprising result here may be understated? A bit more discussion on this would be useful.

Discussion of the model stratocumulus scavenging treatment is now included at line 29, page 3:
The dominant sink for modelled summertime Arctic aerosol is stratocumulus scavenging (Browse et al., 2012). Stratocumulus precipitation (drizzle) rates are calculated from observations of droplet concentration and cloud height and scaled by the coverage fraction of low clouds, resulting in diagnosed drizzle rates between 0.1 and 0.3 mm/d north of 70N. In Browse et al. (2012), where we introduced scavenging of aerosol by drizzle, we showed that high latitude (north of 70N) sulphate mass concentrations increased by about a factor of 3 for a halving of drizzle rates. Thus, our baseline simulations are uncertain even before we consider cloud responses to changes in sea ice.

We also include greater discussion of the uncertainty in our results linked to scavenging at line 14, page 12:

In these simulations we have assumed that the change in aerosol emissions has no effect on cloud microphysics. However, complex microphysical responses of the Arctic shallow clouds cannot be excluded. We can project two scenarios; (1) enhanced aerosol and precursor emissions suppress precipitation, or (2) a large increase in latent heat flux increases precipitation.

As an extreme case we can quantify the response of CCN to sea-ice loss assuming that the extra aerosol suppresses all precipitation. In this scenario (1) the removal of ice leads to significant increases in central Arctic CCN concentrations (10-40%), while in some more southerly regions, CCN decreases (Fig. 9, top). Figure 9 shows that the aerosol response to ice loss combined with drizzle suppression is very similar to the response of CCN to drizzle suppression alone. The explanation lies in the role of drizzle in scavenging aerosol transported from lower latitudes (Browse et al., 2012). When drizzle is suppressed in the no-ice run, Arctic aerosol becomes dominated by N_{100} particles transported from low latitudes (Fig. 9,middle), which is the same aerosol that flows into the Arctic when drizzle is suppressed in the present day.

If precipitation were to increase (scenario 2), the negative response of CCN (and N_{100}) concentrations to enhanced sea-salt aerosol and DMS is unlikely to change with larger particles more quickly scavenged. In addition, we would expect a decrease in transport from lower-latitudes further depleting N_{100} concentrations in the high Arctic.

Both scenarios are plausible. However, given our poor understanding of Arctic clouds, any model response is speculative and our analysis points to the need for a much better understanding of aerosol-cloud interaction in the Arctic.

Page 17092: The paper makes no mention of the importance of secondary organic aerosol; do you think it is unimportant in the Arctic? Is there evidence that that is the case? The importance of dicarboxylic acids in the Arctic makes it seem that SOA is important, and that there should be some discussion about how comprehensive are the aerosol sources in GLOMAP. The sources do not include sea salt aerosol from wind blowing over saline surfaces, e.g. new ice, and frost flowers. Are these known to be unimportant?

These simulations do not include wind-blown sources of sea-salt aerosol from saline ice and snow surfaces as in summer when wind-speeds are generally below 7m/s sea-salt aerosol production from this source is negligible (Yang et al., 2008). We now discuss blowing snow at line 9, page 4:

Over sea-ice, sea-salt aerosol production from blowing snow could be significantly higher than from open ocean (Yang et al., 2008). However, the source is negligible at wind-speeds below 7 m s−1. Thus, for Arctic summertime conditions (Tjernstr¨om et al., 2012) neglect of this process in GLOMAP is unlikely to impact our results.

Secondary organic aerosol (SOA) from monoterpene emissions is included in GLOMAP (Scott et al., 2013). Discussion of secondary organics is now included from line 18, page 4:

The importance of secondary organic aerosol (SOA) in the Arctic is poorly quantified. Our simulations include SOA from monoterpene emissions (Scott et al., 2014) but neglect other biogenic volatile organic compounds (BVOCs). Measurements at lower Arctic latitudes (70-72°N) suggest that SOA derived from isoprene and α-pinene (excluded from these simulations) comprises 8% of the identified OC aerosol mass (Fu et al., 2013). However, (Fu et al., 2013) report a major portion (80%) of measured OC as unidentified (thought to derive from primary marine emission).

Page 17092 - do you mean to say H2O2, or is HO2 correct?

Should be **H2O2,** text has been amended (line 19, page 5)

Page 17094 - what is a "pollution controller"?

A pollution controller is a sensor which automatically shutdowns instruments during pollution events (i.e. contamination from ship mast). Text now reads **pollution sensor (line 5, page 7)**.

Page 17096, line 11 - the model-observation slope and intercept are not shown in Fig 2a. In Fig. 2a it is confusing, which is the observed ASCOS data - is it the black line? I think the legend in this figure should show coloured lines.

Legend in figure 2 has been amended. Discussion of model-observation slope in section 6 no longer refers to figure 2, from line 23 page 7:

We compare the model against the observations in terms of the size distribution and CCN frequency histogram (Fig. 3), and quantitatively compare the slope and intercept of modelled and observed integrated particle concentrations between 3 and 800nm diameter.

From line 33, page 7

Comparing the modelled and observed integrated particle concentrations (not shown) gives a slope of 0.96 in the PD run,

From line 14, page 8

Reproduction of the observed Aitken mode without BLN was not possible in GLOMAP without significant alterations to model processes or by reducing the size of the OC particles to less than 40 nm. Inclusion of the tuned 40 nm diameter marine OC emissions with BLN (run PD-MOC) improves the agreement with the observed accumulation mode particle number concentration (Fig 3a). It also improves the total particle concentration model-observation intercept (from 60

to 13 /cm^3). This ~5-fold reduction in intercept concentration is a result of the suppression of BLN caused by the higher primary OC surface area.

And from line 27, page 8

Suppressing stratocumulus cloud scavenging (PD-noDRIZZ) increases the positive bias in modelled CCN (Fig. 3b), results in a very poor agreement with the observed size distribution (Fig. 3a), and decreases the slope of modelled vs observed integrated particle concentrations to 0.38.

Regarding the discussion of H2SO4 nucleation - are there any H2SO4 measurements in the Arctic? It is being measured in various places with good sensitivity, I just don't know if it has been done in the Arctic.

Observations of nucleation events have been reported in the Arctic by Karl et al., (2012) and Rempillo et al., (2011) although direct observations of high-latitude nucleation events are rarer than from low-latitude sites. We now include discussion of the criteria used by Karl et al.,(2012) to characterize nucleation events (from line 5, page 8):

Observed nucleation events were characterised by the rapid (1h) enhancement of total particle number (typically <50 /cm^3) to high concentrations (200-1000 /cm^3), which persisted between 5-12h.

Last paragraph on page 17096, line 22 - really, your simulations indicate the (likely?) role of drizzle scavenging. I feel that to use the word "confirm", you need specific field observations of that process.

Our simulations confirm the role of drizzle scavenging in the model. Sentence now refers to modelled Arctic CCN (line 27, page 8).

Figure 3 is among those that takes some time to sort through. Line 8 of page 17098 says "larger than 100nm diameter", but the Figure 3 caption says 200nm. Fig. 3 should probably have a title line over the top, like, "Aerosol Impacts, No Sea Ice" or something appropriately descriptive. Similarly, Figure 4 could have a caption at the top that says "Aerosol Impacts of No Sea Ice, Drizzle Scavenging Suppressed". The Journal might not like that, but it would help with readability. And, it would be useful to have a bold label at the left of each row in Figure 3, like: (delta)CCN, (delta)particle # conc. (>200nm), (delta)particle # >3nm. At this page, around line 13, shouldn't the text also state that CCN increases in Arctic coastal regions, i.e. where Arctic people live? At line 24, it is hard to see from Figure 3 what you are referring to.

Figures 3 and 4 have been updated as suggested (see attached figures).

Page 17100, line 13 - do the model uncertainties justify 3 significant figures? Line 11 - it isn't clear what we are supposed to be looking at in Table 2.
Page 17101, line 17 - this isn't the number in Table 2, should it be 254? Maybe such numbers need to be rounded anyway, to maybe two sig figs?

The metric changes have been rounded to 2 significant figures in table 2 and at lines 7-20, page 11, **and line 4, page 12.**

Page 17103, line 4 - the decrease could well be greater than Voulgarakis et al. suggest, because sea ice loss also will most certainly decrease BrO, which reacts with DMS. At a typical sea-ice covered BL BrO concentration of 10 ppt, the DMS lifetime is 2.3 hrs. (cf. Brieder et al., GRL, 2010).

The modelling study Breider et al., (2010) report surface BrO concentrations at high-latitudes of ∼0.1ppt in January with a tropospheric column density of 0.5-0.75 x10^{13} molecules per cm ^{2}. However, Breider et al., (2010) underestimate tropospheric Arctic BrO because they do not include a sea-ice source of Bromine (Yang et al., 2008). Ridley et al. (2000) made a detailed analysis of the photochemical balance at Alert in April (a place known to have very rich halogen chemistry) and could constrain BrO to a maximum of 5 ppt (DMS lifetime in the BrO reaction between 4.5 - 5.4 hours).

Springtime Arctic BrO concentrations have been observed to be higher due to the occurrence of bromine explosions (Foster et al., 2001). Yang et al (2008) link these bromine explosions to blowing snow events at wind-speeds above 7m/s, a rare occurrence in the Arctic summertime boundary layer (Tjernstr¨om et al., 2012). Thus, summertime sea-ice loss is unlikely to deplete BrO derived from blowing snow. To the authors knowledge the response of Arctic BrO to sea-ice loss has not been quantified. However, we can not rule out the impact of ice-loss on other BrO sources such as frost flowers (Foster et al., 2001), or snow-pack chemistry (Pratt et al., 2013).
Discussion of the response of both OH and BrO to sea-ice loss has now been added from line 10, page 13:

However, these simulations do not include the reduction in photolysis rates driven by surface albedo change from sea-ice loss, which could decrease OH concentrations by 30-60% (Voulgarakis et al., 2009), or the effect of sea-ice retreat on Bromine chemistry.

Bromine has been shown to contribute ~20% (maximum 40%) to the DMS sink at high latitudes (Breider et al., 2010). High-latitude bromine radicals (Br, BrO) derive from photochemical reactions within the snow pack (Pratt et al., 2013), blowing snow events (Yang et al., 2008) and frost flowers (Foster et al., 2001). Thus, sea-ice retreat will likely decrease BrO abundance, although the impact of ice loss on Bromine sources is unquantified. The sensitivity of Arctic aerosol to either BrO or OH abundance has not been tested here. However, If oxidant concentrations decrease, then the aerosol response to summertime sea-ice retreat could be dominated by primary emissions (i.e the response of aerosol to no-ice[SS]) and thus, negligible.

Additions to Bibliography

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