

This study makes use of measurements of particle emission flux from Arctic leads to produce a parameterisation of the process that is suitable for models. The work is a novel contribution to a research question that is highly relevant to the scope of ACP, since this is a key polar aerosol process that is missing in current state-of-the-art climate models. The parameterisation is tested both using inputs from reanalysis to calculate emissions, and within the Weather Research and Forecasting model with chemistry (WRF-Chem). Results are compared to measurements where possible, and the relative importance of leads as an aerosol source compared to blowing snow and transported sea salt is assessed. I recommend that this article be published following some minor comments detailed below.

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General comments

Emissions from the marginal ice zone (MIZ) are largely excluded from the parameterisation. I acknowledge the justification for doing this in section 2.2.4, and analysis of the sensitivity of the results to this choice in Figure A1 and section 3.6. However, given the importance of the MIZ in the question of how Arctic aerosol will respond to future warming, some more discussion of modelling emissions from the MIZ would be welcome in the introduction and in 3.6 or 4. Specific recommendations about this are included below.

Regarding figure 7 and the associated discussion, the potential role of transport and wet removal has been neglected as a potential source of disagreement between the conceptual modelling and the measured concentrations. Please address this in the text (see below).

There is some confusion caused by terminology in figure labels and captions, for example mixing “monthly sum” and “daily average” in the same figure or using the word “flux” both for emissions per unit area per unit time and simply for emissions per unit time. Please address these so that there is consistency throughout.

Specific comments

Lines 21-22: sea spray has also been shown to have a key effect on the climate impact of secondary aerosol particles in the changing Arctic climate system, by changing the condensation sink (Browse et al. 2014) but this is by no means well established because results vary across models (Gilgen et al. 2018).

Line 33-35: it is important to highlight (here or elsewhere in introduction) why sources from leads are an important factor in establishing the sensitivity of Arctic aerosol to sea ice in the present day and in the future (or other differing climate states). Eg in the warming Arctic as sea ice extent/thickness changes and the marginal ice zone becomes wider at certain times of year (Strong and Rigor 2013), emissions from leads may be relatively more or less important than emissions from open ocean or pack ice. So without including this process in models, our knowledge of the changing natural aerosol baseline in the Arctic may be incomplete. (This point is made via the inclusion of the question in line 76, but the motivation could be made stronger).

Figure 1b: since the Salter et al emission parameterisation includes an SST dependence, it's not clear whether the size distribution shown here in the figure is for a particular value of SST. Please give this information in the figure or caption.

Line 128: "...only affect the sea salt emissions, but not..." surely it will also affect the organic emissions? Perhaps clearer to say "only affect the composition of the emitted particles, but not the absolute emissions flux of sea spray".

Line 195: the important conclusion that emissions from leads/blowing snow could be as important for the Arctic aerosol budget as transport relies on the comparison with MERRA-2 data. As such, please give some information here on how MERRA-2 sea salt aerosol is modelled (emission, removal processes) - has the representation of aerosols in MERRA-2 been previously evaluated for the Arctic? In particular I think it is important to highlight any uncertainties in the calculation of sea salt transport in MERRA-2 that could impact the comparison.

Lines 202-203 and figure 2: it's not clear which underlying emissions flux is used to calculate the fluxes presented in figure 2(d-f) (although the figure caption for figure 3 states Gong (2003) is used, which suggests the same is true for figure 2). Please give this information here in the text or in the caption for figure 2, or both.

Figure 3 and 5: the y-axis label gives these values as an "average rate" in #/s, while the figure caption states "monthly sum of sea salt number emissions". As such, I struggle to interpret whether the values represent total number of emitted particles calculated over each month (with units of particle number), or an average emission rate of particles for the month. Moreover, the y-axis label refers to a "daily average" which conflicts with the monthly aggregation here. I interpret this to mean that the aggregation is sampled from daily mean flux values, but I don't think it's necessary to write that here. The daily meaning is described in the text (lines 120, 163, 195) and I think that's sufficient. Alternatively if I have interpreted this wrong, please clarify the meaning of "daily average" as used here.

Figure 4: similar to previous comment on figures 3 and 5, same note here about the use of the phrase "daily average" to describe seasonal means.

Figures 7 and 9: values in maps are given units of [mass per unit time], whereas I would expect [mass per unit area per unit time] for a mass flux, and this would seem to be more consistent with your description of normalising the flux values by PBLH (in fig 7). Please double check these units.

Figure 7 and line 294-296: comparing emissions data to concentration measurements: "...which signals that our parameterization could be improved despite already showing better agreement..." The conceptual model is of local emissions only (with some accounting for vertical mixing via the use of PBLH) whereas measured concentrations have also been influenced by removal and transport. As such this conceptual model of the "usual approach" is not totally consistent with what a climate model would produce when including explicit representations of the vertical mixing, transport, and removal processes. Given the fluctuations in transport and removal throughout the year at these locations, it seems possible that the wintertime underestimation and summertime overestimation by "usual approach" could be influenced by significant contributions from long-range transport and wet deposition (respectively). This should be emphasised in the text to highlight the uncertainties in modelling polar sea salt aerosol other than just these missing sources. Also, it may be informative to calculate the sodium concentrations for these locations from the WRF-Chem output (where transport and removal are included) and add those values here as datapoints for the months available.

Figure A1: it would be instructive to see maps of the extent of the revised "high Arctic" areas using the different sea ice thresholds, to understand how the choice of threshold influences the spatial area under consideration. Moreover, one interesting application of the parameterisation introduced

in this study will be to study the effect of sea ice changes on the future Arctic aerosol budget – specifically, the changing extent of the MIZ relative to total ice extent and how this may change the relative importance of open ocean/transported sea spray particles vs particles from open water areas in sea ice regions. It would be good to add some discussion of this aspect of the research either here or in the conclusion section. Specifically, it is important to emphasise the need for a better understanding of aerosol sources in the MIZ (including sea spray) and a need for this to be translated into climate models via parameterisations such as the one presented in this study.

Line 444-445: it's shown in Figure 1 that it is not just the magnitude of the emissions that is uncertain, but even the sign of the response with increasing wind speed is unconstrained. This is an interesting result that could be stressed here.

Technical corrections

Line 92: “the total particle number aerosol flux over the open Arctic Ocean was...” open ocean measurements reported in Nilsson are from the Barents Sea region further south rather than the Arctic Ocean itself. Please double check this and amend your statement here.

Figure 6: y-label is not very intuitive, suggest to change to something like “Total sea salt source / Gg month⁻¹”

Figure 8: please explain exactly which areas are included in “open ocean”, i.e. what's the southerly extent.

Figure 9: the map of Utqiagvik here uses a different projection from that in figure 7 - suggest to use a consistent projection for both.

Line 427: “This difference mass and number” should read “difference in”?

Lines 638-end: should the references by Ólason et al. come under “O”?