

Interactive comment on "Sea ice dynamics influence halogen deposition to Svalbard" by A. Spolaor et al.

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ANSWER REFEREE 2

Spolaor et al present a very interesting study of iodine and bromine contents in a glacier ice core from Spitzbergen. They find an intriguing positive correlation between bromine and seasonal sea-ice extent and a negative correlation for iodine and spring maximum sea ice extent which the authors argue is consistent with the likely sources for bromine and iodine. Overall this is a very interesting and well written paper. The main weakness is the lack of attempts to investigate the wind climatology as well as the seasonal changes in sea ice cover, extent and variability around Spitzbergen in order to substantiate a cause and effect relationship between sea ice extent and transport followed by deposition onto the glacier. I don't think it is appropriate to use total Arctic sea ice

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data especially given the very special location of Spitzbergen. The conclusions drawn are very plausible but there is no actual proof of the mentioned source mechanisms for bromine and iodine just using the presented data. This should be acknowledged more explicitly throughout the text and in the abstract which sounds rather definitive (esp. I. 8-12).

As suggested also by referee 1, we improved our interpretation regarding the wind climatology to better understand the provenance of the air masses (paragraph 3.2 and 3.3). In addition we evaluated the change of sea ice for the specific Arctic sectors especially for the Barents sea, Arctic Ocean, and Greenland sea. The main variability in the total seasonal sea ice area is due to changes in the Arctic Ocean that account for about 30% compared to the total Arctic. Six-day back trajectories show an influence of the sea ice in Baffin bay; however, the bromine explosions in this sector are likely much less important compared to explosions occurring in the sea ice around Spitzbergen. We report below our response for the similar concerns posed by referee 1.

Minor comments

p. 1078, l. 26: The idea of lofting aerosol from frost flowers has largely been discounted, see e.g. Roscoe et al., JGR, 2011

Chemical analysis of frost flowers show that Br/Na ratios are very similar to that of sea water (Rankin et al., 2000) indicating that no fractionation happens during frost flower formation. Frost flowers are considered to be a source of gas phase bromine by increasing the surface area available to the reaction between bromide and ozone. However a recent paper (Pratt et al., 2013) showed that frost flowers do not play a role in gas phase bromine emission, but instead it is the fresh snow above the first year sea ice that acts as a source for gas phase bromine. The direct transport of frost flowers from the sea ice surface up to the Holthedalfonna is likely insignificant but in any case would not affect the Br/Na ratio.

p. 1082, l. 18: Change unit at end of line from "mg L1" to "mg L-1"

Changed accordingly

section 3.2, 1st para: I'm not sure how useful the seawater I/Na ratio is in this regard as it has been shown since the 1960s that iodine is strongly (up to 1000x) enriched in airborne particles.

Our aim is only to demonstrate that iodine is mainly driven by other sources with respect to the sea-salt deposition. The enrichment that has been detected in airborne particles is most likely due to the uptake of inorganic iodine gas-phase species (HOI, INO3, HI etc, whose source is biological production in the ocean. Iodine concentrations in the snow also clearly demonstrate that sea salt aerosol plays a marginal role.

p. 1086, I. 5-7 vs. 9-12: Please explain better as currently this sounds like a contradiction. This is also linked to the main weakness that I mentioned above - I don't think this is a good justification to use Arctic basin rather than regional sea ice data.

We agree with the referee and we improved this section including the evaluation of regional sea ice extensions and wind patterns. The wind pattern during March-May, the period when halogen chemistry is most active and when the sea ice changes more dramatically, shows that the main sea ice source regions are located in the Arctic Ocean, Greenland sea and Barents sea. For bromine we choose to consider only these seas since the bromine atmospheric chemistry is more confined. For iodine we include also Baffin bay since 6-day back trajectories show a possible influence from Baffin bay. In addition the atmospheric lifetime of organic iodine compounds emitted by biological production are in the range of few minutes to few days (2-6 in case of CH3I), meaning that deposition in the Spitsbergen could be influenced by other region in the Arctic, such as the Bering sea. In conclusion we suggest that for bromine the Arctic Ocean and the Canadian Archipelago seem the main drivers, while for iodine we need to consider also Baffin bay, Greenland sea and the Bering sea. As explained earlier in our responses to the referees, the regions that seem to mainly influence Svalbard halogen deposition are the regions that also strongly influence the total Arctic seasonal sea ice

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and its extension during the March-May period.

p. 1087, l. 2-4: Please explain this correction in more detail. Don't you remove part of the signal by doing this?

Bromine and Sodium have a strong positive correlation (R=0.78) in our samples because of their similar, marine, sources. Since bromine is emitted from first year sea ice in the gas phase, while sodium is not, the most important thing is to isolate the bromine emitted from the bromine explosion, the enrichment. Our theory is that the more seasonal sea ice is present, the more bromine explosions can occur and the more bromine available to be enriched compared to sodium. For this statement only the enrichment of bromine is important. The bromine concentrations alone do not provide sufficient information. Removing the sea salt signal we extract in principle the seasonal sea ice signal.

p. 1087, l. 15: I don't think that based on your data you can draw any conclusions about bromine losses – simply because you can't measure it and the deposited amount of bromine might have been much higher then what is measured.

The referee is right and we have appropriately modified the paragraph. This is the main uncertainty in our work at the current stage. The recent paper by Pratt et al. 2013 is very useful in this regard, as they discovered that the bromine explosion is favoured in the fresh snow above first year sea ice and tundra. In Svalbard the influence of tundra compared to sea ice should be negligible. They show the activation of bromine is favoured by low pH (< 5) and a low Br/Cl ratio. Our samples show pH in the range of 5.3 and we could consider that sintered snow does not produce gas phase Br (the production is possible only with high ozone levels). However the fresh snow that is deposited on the glacier can suffer from post depositional processes and hence some bromine can be lost. The enrichment that we detect can be associated with sea ice. It is not possible to understand if post depositional processes can strongly modify the signal however data present in the literature suggest that without fresh snow (at least

a few centimetres on the surface), bromine can be conserved in the record. Referee 3 also mentions post-depositional processes, hence we would be pleased if the referee would also consider our response to referee 3 as a more complete evaluation.

USEFUL REFERENCES

Pratt, K. A., Custard, K. D., Shepson, P. B., Douglas, T. A., Pohler, D., General, S., Zielcke, J., Simpson, W. R., Platt, U., Tanner, D. J., Gregory Huey, L., Carlsen, M., and Stirm, B. H.: Photochemical production of molecular bromine in Arctic surface snowpacks, Nature Geosci, 6, 351-356, 10.1038/ngeo1779 http://www.nature.com/ngeo/journal/v6/n5/abs/ngeo1779.html#supplementary-information, 2013.

Rankin, A. M., Auld, V., and Wolff, E. W.: Frost flowers as a source of fractionated sea salt aerosol in the polar regions, Geophys. Res. Lett., 27, 3469-3472, 2000.

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