Reply to Referee #2 (Dr. Dominé)

We would like to thank your helpful comments to improve our manuscript. We responded to the general and specific comments. All comment are addressed in the revised manuscript. The updated sentences by your comments indicates blue words.

Comment from Referee

The somewhat tedious point-by-point description of results should be completely replaced with a discussion focused on solving a few selected scientific questions such as for example: "how does the presence of FF affect aerosol composition?" or "what are the processes leading to halogen enrichment in FF and surface brine?". More aggressive attempts to make deductions from the observations are mandatory.

Reply from authors

Thank you for your suggestions. We focused on sea-salt cycles in seasonal sea-ice area including sea-salt fractionation, aging processes of frost flowers, the fractionated sea-salt aerosols, and release processes of sea-salt aerosols from sea-ice surface on basis of our field evidences and previous works.

Comment from Referee

The authors may cross their data with GOME2 BrO data to perhaps reach some interpretation on halogen activation.

Reply from authors

We agree with your suggestions. However, our measurement periods were too early to measure BrO and IO densities around northern areas of Greenland using satellite, because of lower solar angle. Thus, we did (could) not compare to BrO and IO data in the manuscript.

Comment from Referee

1- Select a couple of novel scientific questions to be addressed by the data set. and 2- Select the data to be presented to address the selected questions. A couple of case studies focused on a few events may be interesting.

Reply from authors

As mentioned above, we focused on sea-salt cycles in seasonal sea-ice area including sea-salt fractionation, aging processes of frost flowers, the fractionated sea-salt aerosols, and release processes of sea-salt aerosols from sea-ice surface on basis of our field evidences and previous works.

Comment from Referee

3- Separate results and discussion and write in a much more concise form to produce a much shorter paper. ,

4- Reach some strong and novel conclusion. For example, finding out that the presence of FF does not significantly affect aerosol composition would be quite interesting.
and

5- Place the data not used here but of potential interest to others in supplementary material or any other accessible place. The authors should feel free to adopt any other strategy, the objective being to make a good and concise use of the data to derive strong conclusions. At present, the manuscript is more a detailed preliminary campaign report than an actual scientific paper.

Reply from authors

We separated Results and Discussion in the revised manuscript. Also we made an effort to concise form. Actually, we removed the description of sea-salt modification to "Supplementary" and remove some repetition of the statements and discussion. Additionally, we added our proposal (hypothesis) on sea-salt cycles in seasonal sea-ice area in "Concluding remarks". Some data and descriptions (e.g., sea-salt concentrations in snow) were added into the section of Results and Discussion in the revised manuscript, because of comments from Referee#3.

Comment from Referee

Vapor is supplied TO the atmosphere, not FROM. See the references on the same line.
2, 31. Specific surface areas are now expressed in $m^2\ kg^{-1}$. *Please convert.*

Reply from authors

These editorial points were addressed in the revised manuscript.

Comment from Referee

Section 2.2. Was snow present in FF and brine samples? This should be mentioned as it dilutes the samples. Re. section 3.4.

Reply from authors

Yes. During the campaign, snowfall and blowing snow occurred. On some frost flowers at Site I and II, snow was present slightly present to extent to that fine structure of frost flower was identified clearly. This statement was wadded to “2.2 Sampling of frost flowers, brine, snow, and seawater”.

Comment from Referee

Please add a + sign: +1.8 °C, to avoid any ambiguity.

Throughout: replace liberated with released

Replace correlation with determination.

Reply from authors

We added “+” sign in +1.8 °C. Words of “liberated” and “liberation” replaced to “released” and “release”, respectively in the revised manuscript.

Comment from Referee

The structure of the paper is such that the mention of solar radiation here is a bit odd and unexpected, and maybe not readily understood by all. Separating results and discussion would have helped.

Reply from authors

With modification of structure in the manuscript, results and discussion were separated in the revised manuscript, as already noted.

Comment from Referee

A more in-depth discussion of the causes of Br and I enrichment is in order.

Reply from authors

All description was moved to section of “4-1. Sea-salt fractionation on sea-ice”. Also, we added plausible processes for enrichment of Br^- and I to the section. Then, we discussed each process, as follows.

Therefore, sea-salt fractionation by hydrohalite might be promoted in some samples of brine and frost flowers. Similar to Mg^{2+}, K^+, Ca^{2+}, and Cl^-, Br^- and I can be enriched in frost flowers and brine by precipitation of mirabilite and hydrohalite. The plausible processes for enrichment of Br^- and I can be listed as follows; (1) sea-salt fractionation by precipitation of salts containing Mg^{2+} or Cl^- and (2) surface enrichment of Br^- and I in liquid phase (e.g., brine). $MgCl_2$ and $MgSO_4$ were identified in aerosol particles as shown in Figs. 8 and S3. Considering presence of $MgCl_2$ and $MgSO_4$ in aerosol particles, Mg salts might be localized or precipitated in frost flowers and slush layer. According to previous laboratory and model studies (e.g., Mairon et al., 1999), $MgCl_2$ $6H_2O$ and KCl (sylvite) can be precipitated approximately at -36 °C and -34 °C, respectively. During the measurements, minimum air temperature (-34.1 °C) and temperature at surface of slush layer (T_{FF}) were higher than temperature at $MgCl_2$ $6H_2O$ precipitation. Therefore, $MgCl_2 \bullet 6H_2O$ precipitation might not occur during the measurements, although precipitation of mirabilite and hydrohalite can occur. If strong vertical gradient of temperature near surface engender that temperature at brine surface or around top of frost flower dropped to temperature at precipitation of sylvite and $MgCl_2$ $6H_2O$, these salts can be precipitated. However, temperature at brine surface or around top of frost flower might be higher than temperature for precipitation of sylvite and $MgCl_2$ $6H_2O$ during the measurements. Thus, precipitation of sylvite and $MgCl_2$ $6H_2O$ might not occur near surface of brine on sea-ice. Consequently, enrichment of Br^- and I in frost flowers might derive from sea-salt fractionation to a greater degree than precipitation of mirabilite and hydrohalite. In addition to sea-salt fractionation by precipitation of mirabilite, hydrohalite, $MgCl_2$ $6H_2O$ and sylvite, a molecular dynamics (MD) simulation conducted by Jungwirth and Tobias (2001) predicted considerable surface enhancement of Br^- and I in alkaline halide solutions. If Br^- and I prefer to be enhanced also at the brine surface, enrichment of Br^- and I might proceed in frost flower and brine by this surface enhancement and sea-salt fractionation.

These descriptions were added to “4-1. Sea-salt fractionation on sea-ice”.

Comment from Referee

Air T is a useful variable for many purposes, but the actual variable of interest here is surface T. All the speculation between air T and processes is really not useful, unless a surface T can be produced. Several lengthy discussions could just be removed.

Reply from authors

Although we showed air temperature measured by AWS (T_{AWS}), our discussion was based on T_{air} (temperature above 10 cm from sea-ice surface) and T_{FF} (temperature at base of frost flowers) in the revised manuscript. We updated description in discussion using air temperature in the sections of 4.1 and 4.2. Some sentences were removed from discussion in the revised manuscript.

Comment from Referee

Replace larger by greater. The sea ice thickness may be important for the relationship between air and surface T, but not for surface processes.

Reply from authors

We replaced “larger” to “greater” in the revised manuscript. Also, we rearrange the relation between T_{FF} and sea-ice thickness in “Concluding remarks”.

Comment from Referee

10, 4-5. This statement does not lead to any useful conclusion. Please delete.

Reply from authors

We removed this statement from the manuscript.

Comment from Referee

Since or until ?

Reply from authors

“Since” is correct.

Comment from Referee

What useful conclusion do we derive from these Mg-rich and K-rich particles? Data description just is not enough for a scientific paper.

Reply from authors

Presence of Mg-rich and K-rich sea-salt particles was direct evidence that the fractionated sea-salt particles were released from sea-ice area. We discussed more details about presence of these particles in the atmosphere, sea-salt fractionation, and release of the fractionated sea-salt particles to the atmosphere in the revised manuscript. The following discussion was added to “4-1. Sea-salt fractionation on sea-ice”.

Mg was enriched in sea-salt particles collected in this study. The following evidences are important to discuss the origins of Mg-rich sea-salt particles and Mg-rich salt particles in the atmosphere; (1) presence of highly Mg-rich particles (Mg-rich sea-salts, $MgCl_2$, and $MgSO_4$), (2)T_{FF} lower than temperature at precipitation of mirabilite and hydrohalite, (3) higher Mg/Na ratio in fine mode, and (4) small variability of Mg/Na ratio in strong winds and blowing snow. Because Mg-rich sea-salts and Mg-salts cannot be evaporated and vaporized under the ambient conditions, these particles must be released through physical processes. If sea-salt particles were fractured in the atmosphere, sea-salt fractionation can occur. However, direct evidence of fracture of sea-salt particles in the atmosphere has not been obtained (Lewis and Schwartz, 2004). With sea-salt fractionation in brine and frost flowers, sea-salt particles released from sea-ice had different sea-salt ratios from those of seawater, as discussed above. Actually, Mg^{2+}, K^+ and Ca^{2+} were enriched in frost flowers. Therefore, Mg-rich sea-salt particles (Fig.8b), K-rich sea-salt particles (Fig.8c), Mg-salt particles (Figs.8h-i), and K-salt particles (Fig.8j) might be originated from the sea-ice area and that they are associated with sea-salt fractionation.

Comment from Referee

Could you please discuss the presence of non-sea salt sulphate?

Reply from authors

Presence of nss-sulphate particles and some explanation were added to “3.7 Abundance of sea-salt particles and sea-salt-related particles”. Presence of nss-sulphates in sea-salt particles were already discussed in discussion of sea-salt modification, which we remove to Supplementary. Short discussion was added to “4-3. Fractionated sea-salt particles in the atmosphere”, because we focused on sea-salt cycles in the seasonal sea-ice areas in this study as follows.

Although the high aerosol number concentrations were observed occasionally at Siorapaluk under calm winds, the features might be caused by transport of (1) sea-salt particles released elsewhere by strong winds and (2) anthropogenic aerosols (i.e., sulphates and Arctic haze). Because of high abundance of sea-salt particles, most cases of higher aerosol number concentrations in calm winds were associated likely with release and transport of sea-salt particles. Similar phenomena were identified also in the Antarctic coasts (Hara et al., 2010).

Comment from Referee

This is where your impressive data set could be put to good use to address these points. "At the moment, release processes of mirabilite-like and ikaite-like particles from the sea-ice surface without frost flowers remain unknown". Sure, but is not this campaign supposed to contribute to solving this problem?

Reply from authors

We consider that presence of ikaite-like and mirabilite-like particles at Site III is an important evidence of sea-salt fractionation on sea-ice area and release of sea-salt particles from sea-ice. On basis of the evidence, we added more discussion about sea-salt fractionation near new sea-ice (Site III) to "4-3. Fractionated sea-salt particles in the atmosphere", as follows.

Mg was enriched in sea-salt particles collected in this study. The following evidences are important to discuss the origins of Mg-rich sea-salt particles and Mg-rich salt particles in the atmosphere; (1) presence of highly Mg-rich particles (Mg-rich sea-salts, $MgCl_2$, and $MgSO_4$), (2)T_{FF} lower than temperature at precipitation of mirabilite and hydrohalite, (3) higher Mg/Na ratio in fine mode, and (4) small variability of Mg/Na ratio in strong winds and blowing snow. Because Mg-rich sea-salts and Mg-salts cannot be evaporated and vaporized under the ambient conditions, these particles must be released through physical processes. If sea-salt particles were fractured in the atmosphere, sea-salt fractionation can occur. However, direct evidence of fracture of sea-salt particles in the atmosphere has not been obtained (Lewis and Schwartz, 2004). With sea-salt fractionation in brine and frost flowers, sea-salt particles released from sea-ice had different sea-salt ratios from those of seawater, as discussed above. Actually, Mg^{2+}, K^+ and Ca^{2+} were enriched in frost flowers. Therefore, Mg-rich sea-salt particles (Fig.8b), K-rich sea-salt particles (Fig.8c), Mg-salt particles (Figs.8h-i), and K-salt particles (Fig.8j) might be originated from the sea-ice area and that they are associated with sea-salt fractionation.

Comment from Referee

"Therefore, most of the aerosol particles around Na2SO4 ratio in fine mode might be the modified sea-salt particles by heterogeneous reactions with nss-SO42-." Can't you get to a stronger statement than just "might" by more in-depth examination of your data?

"Therefore, sea-salt modification (Cl loss) might be most likely to occur in fine mode." Sure, you may even use your data quantitatively and examine the role of aerosol

surface to volume ratio on reaction kinetics. Again, "might" is not sufficient here.

Reply from authors

We agree with your suggestions. We change "might" to "may" in the revised manuscript. Because we focused on sea-salt fractionation, description of sea-salt modification was moved to "Supplementary". Role of aerosol surface area to volume ratio in sea-salt modification was already discussed in our previous studies (Hara et al., 2003, 2005, 2013). Thus, we mentioned this in Supplementary of the revised manuscript.

Comment from Referee

Again, a more in-depth use of the data should allow useful conclusions, not vague suppositions.

These mentions of just observations, without any scientific deductions, are very disappointing.

Reply from authors

Using our field evidences and results by previous works, we proposed sea-salt cycles in the seasonal sea-ice area in "Concluding remarks". Because this proposal is hypothesis, this includes some speculation. We believe that our proposal (hypothesis) is useful and new set for sea-salt chemistry in the polar regions.