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Supplement of

Frost flowers and sea-salt aerosols over seasonal sea-ice areas in northwestern Greenland during winter–spring

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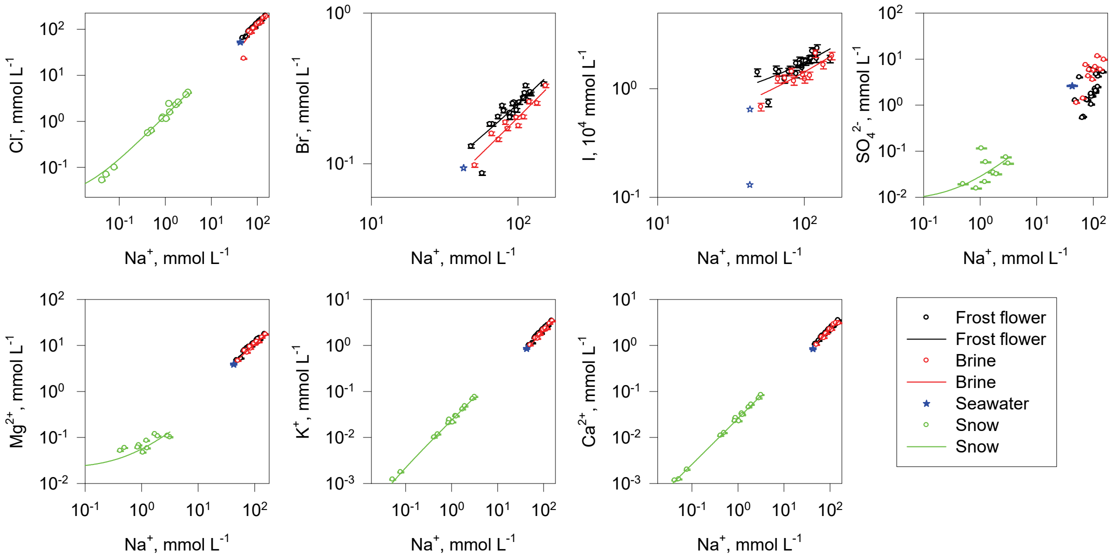

Figure S1 Logarithmic plots of Figure 4. Symbols and lines have the same meanings as those in Fig. 4.

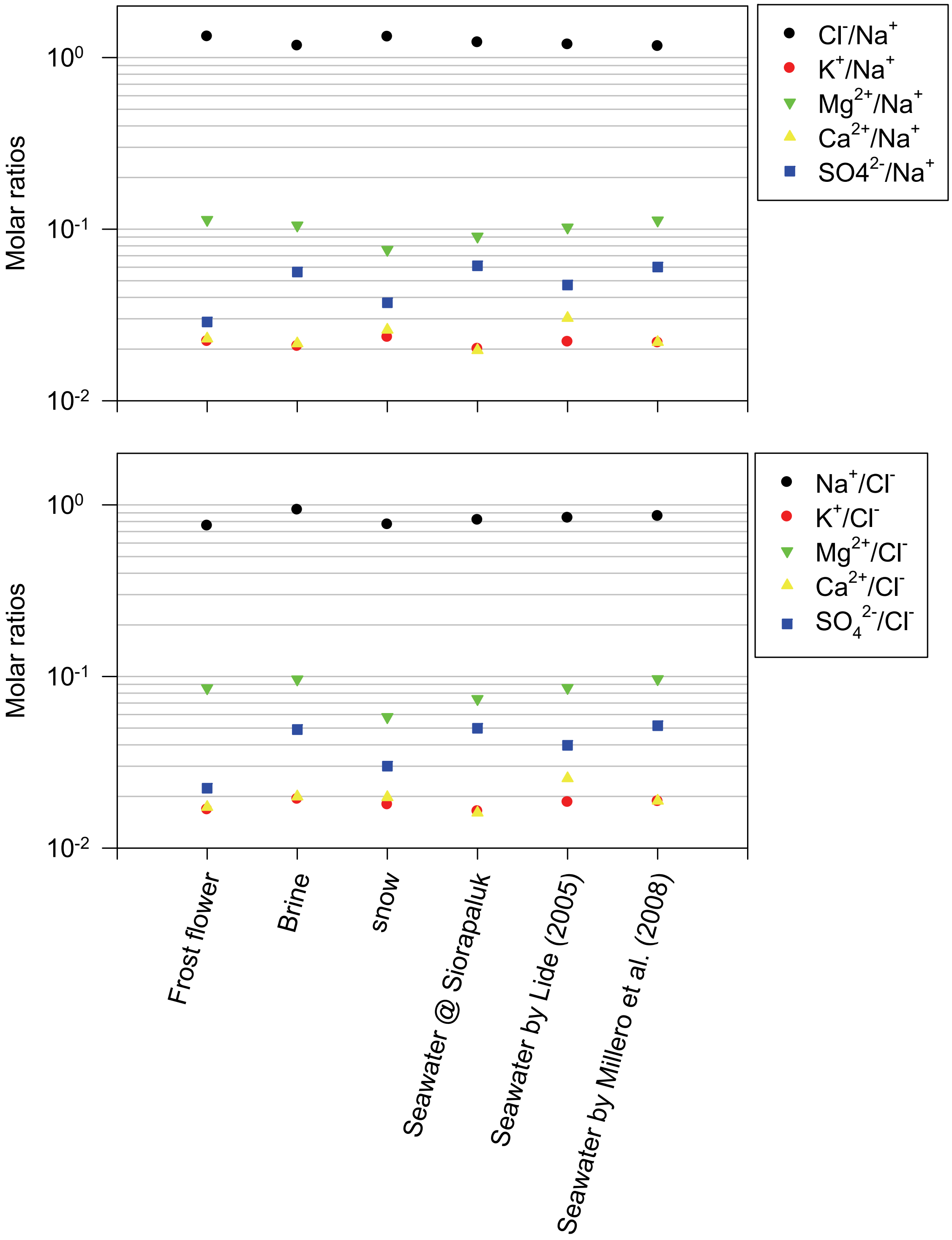

Figure S2 Comparison of molar sea-salt ratios among frost flowers, brines, snows, seawater collected at Siorapaluk, and literature values (Lide, 2005; Millero et al., 2008).

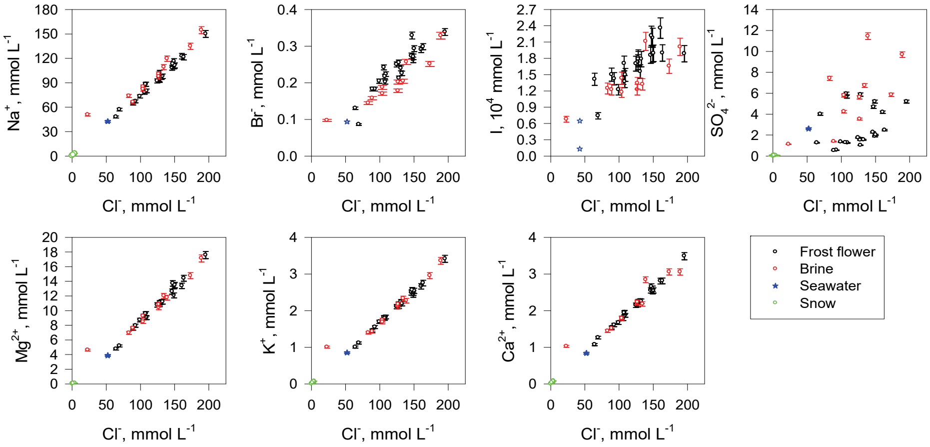

Figure S3 Relations among respective constituents to Cl$^-$ concentrations of frost flowers, brine, and seawater found in this study. Symbols and lines have the same meanings as those in Fig. 4.

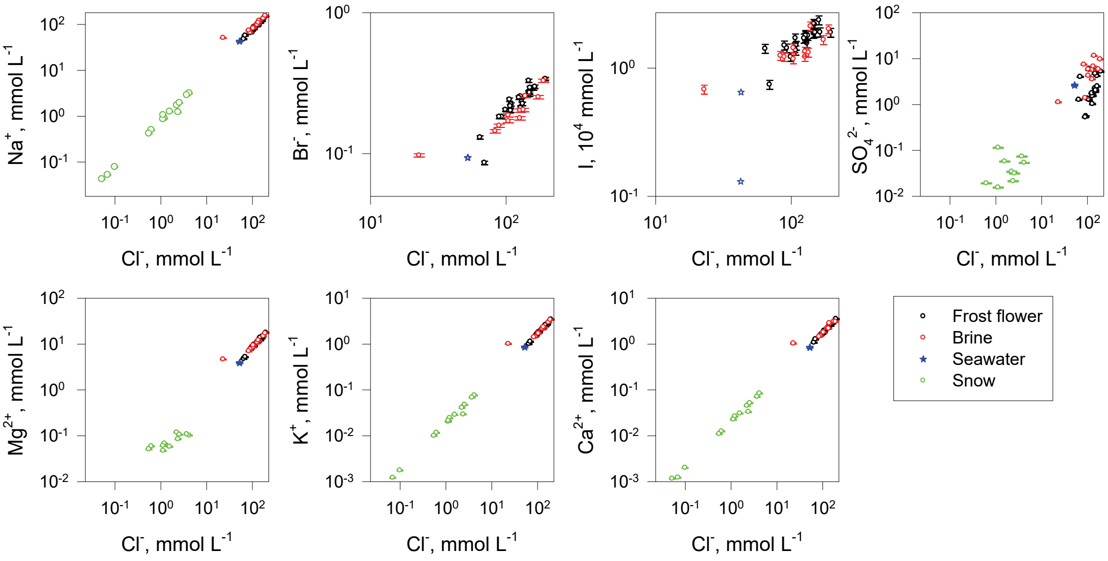

Figure S4 Logarithmic plots of Fig. S2. Symbols and lines have the same meanings as those in Fig. S2.

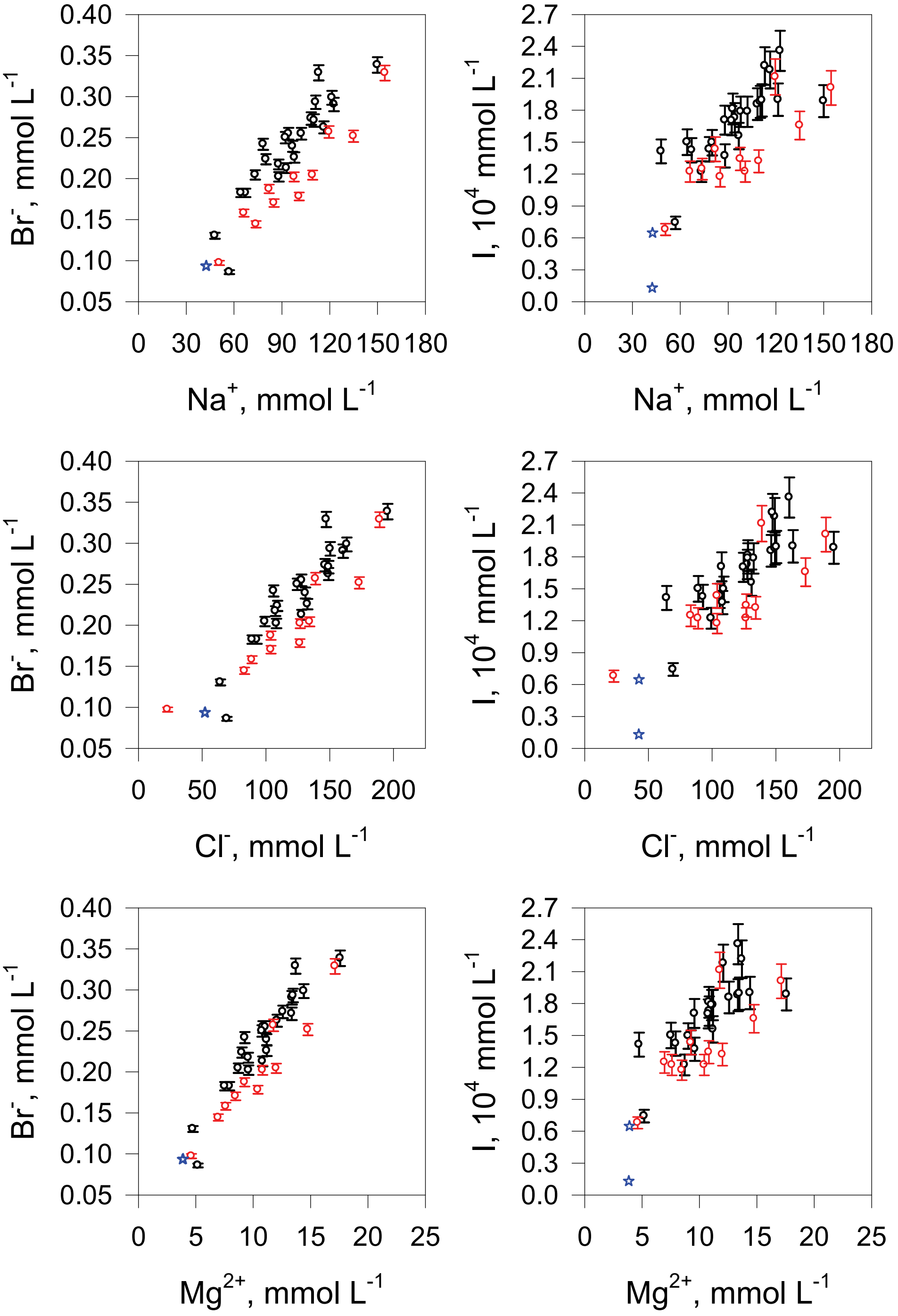

Figure S5 Relation among respective constituents to Mg^{2+} concentrations of frost flowers, brine, and seawater found in this study. Symbols and lines have the same meanings as those in Fig. 4.

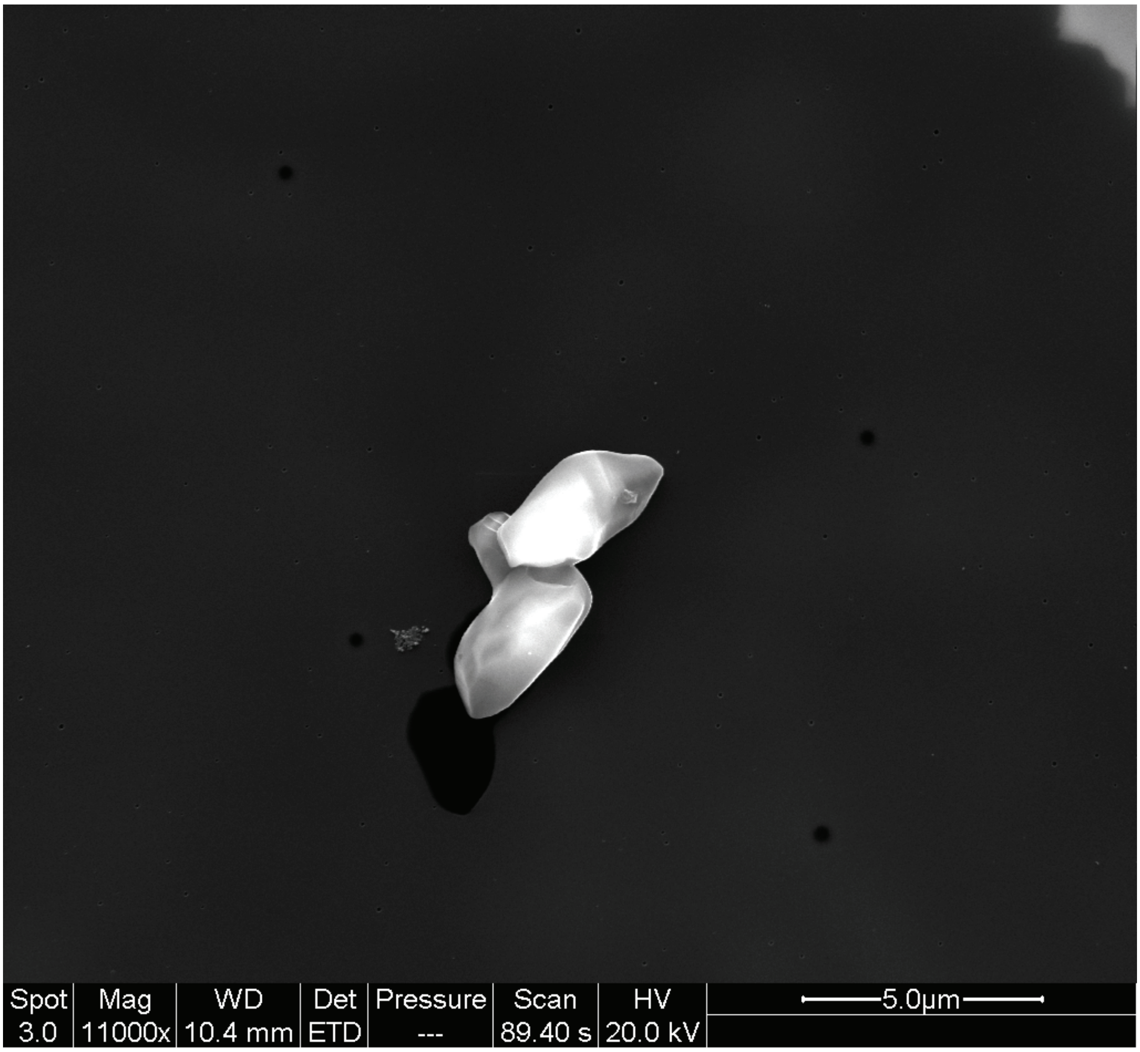

Figure S6 SEM image of mirabilite-like particle collected on 3 March.

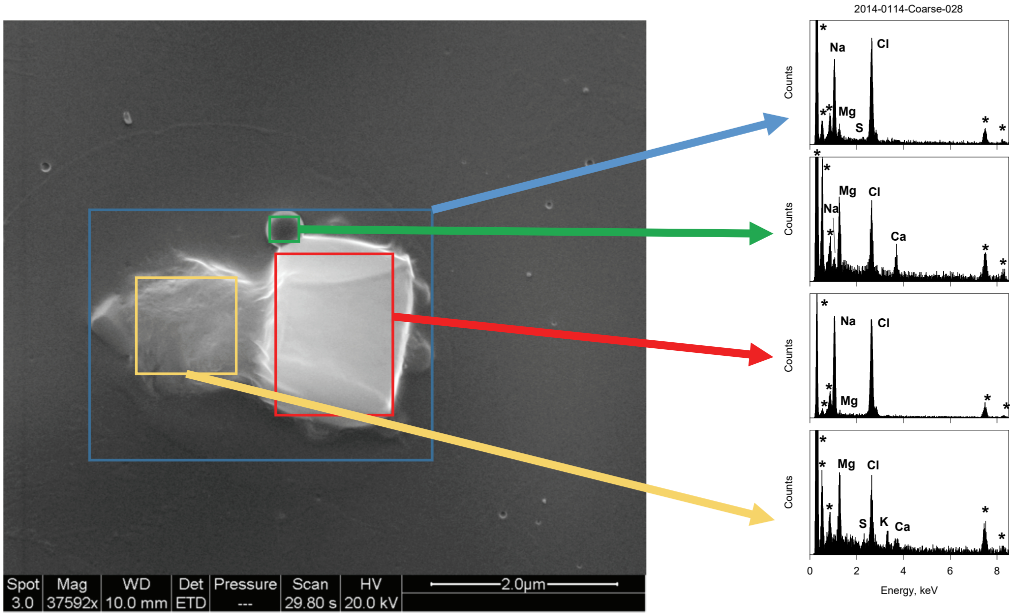

Figure S7 EDX spectra of sea-salt particle collected on 14 January, 2014.

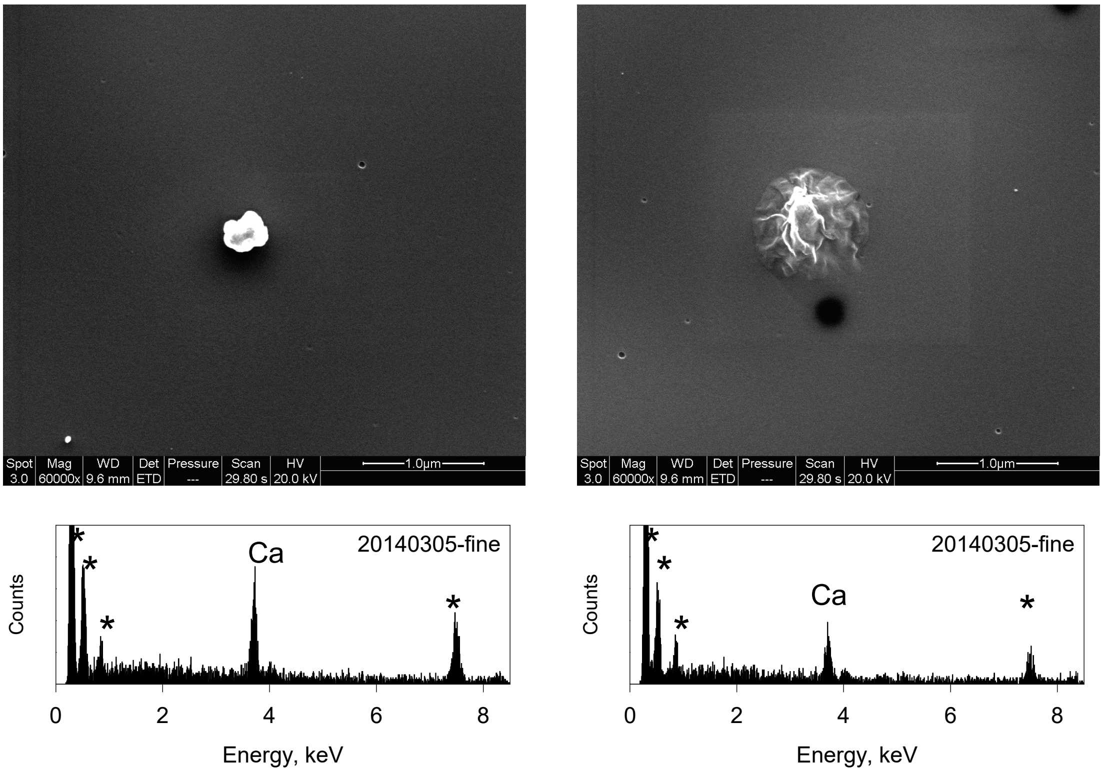

Figure S8 SEM images and EDX spectra of $CaCO_3$-like particle collected on 5 March.

S-1 Sea-salt modification of aerosol particles in coarse and fine modes

Figure S8 portrays ternary plots (Na–S–Cl) of sea-salt particles and the modified sea-salt particles. The internal mixture of sea salts and minerals were removed from the ternary plots and discussion in order to avoid misunderstanding of sea-salt modification. Labels of A and B respectively denote atomic ratios of bulk seawater ratios and wholly Cl depleted sea-salt particles by SO_4^{2-}. The black line represents the stoichiometric line. When Cl in sea-salt particles is released stoichiometrically by heterogeneous reactions (SO_4^{2-} formation), each sea salt particle is distributed along the stoichiometric line.

Coarse sea-salt particles collected near new sea ice (Site III) in DOY = 61.61 (3 March, 2014) were classified into (1) sea-salt particles distributed around bulk seawater ratio and (2) aerosol particles distributed around Na_2SO_4 ratio. Frost flowers had not been formed on a new sea-ice surface on 3 March. Although strong winds were observed in DOY = 58.9–60.3 (28 February – 1 March, 2014) and although they lead to the appearance of an open sea surface by break and flow of sea ice, the wind speed dropped to less than 2 m s^{-1} in DOY = 60.8 (end of 1 March). Nevertheless, Mg was not detected in the aerosol particles having ratios similar to Na_2SO_4, as presented in Fig. 8g and Fig. S4. Additionally, aerosol particles with atomic ratios similar to ikaite or $CaCO_3$ were observed in same aerosol samples (shown in *Supplementary Materials*, Fig. S7). These particles were observed only in aerosol samples collected near new sea ice in this study. In contrast, Na-S-Cl ratios in fine sea-salt particles were varied largely in fine mode on 3 March. Modifying sea-salt particles had S ratios lower than the stoichiometric line between bulk seawater and Na_2SO_4.

Coarse sea-salt particles collected over young sea-ice areas (near Site I) with frost flowers (e.g., on 14 January and 21 February as shown in Figs. S8b–S8c) were also distributed around the bulk seawater ratio. Some sea-salt particles had lower Na ratio and higher Cl ratio than the bulk seawater ratio. Mg was enriched considerably in the aerosol particles with lower Na ratios. Details of Mg enrichment are discussed in a later section herein. Similarly to the sample on 3 March, ratios of sea-salt particles in fine mode were varied largely. Sea-salt and modifying sea-salt particles in fine mode on 14 January were distributed around bulk seawater ratios and along with the stoichiometric line. Fine particles collected on 21 February were distributed around the stoichiometric line and Cl ratio = 0.

Under conditions with storms and blowing or drifting snow, most of the sea-salt particles in both coarse and fine modes were distributed around the bulk sea-water ratio in Na-Cl-S plot on 1 March (Fig. S8d).

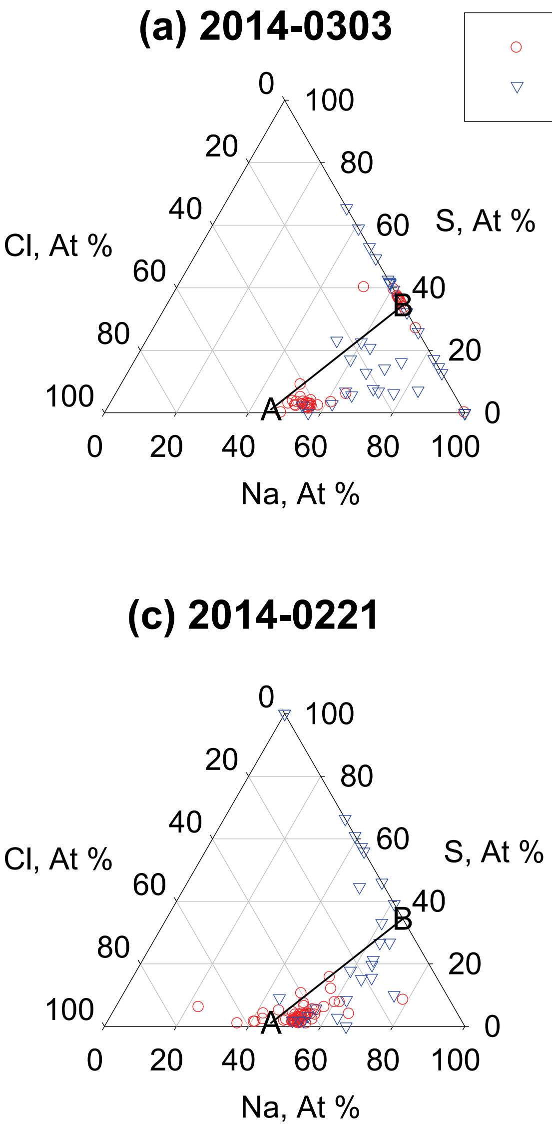

Figure S9 Ternary plots of Na–S–Cl of sea-salt particles and the modified sea-salt particles collected over the sea-ice area. A and B in this figure respectively denote ratios of seawater and fully modified with SO_4^{2-}. Red open circles and blue triangles respectively present ratios of each particle in coarse and fine modes.

3.9 Variations of sea-salt modification of aerosol particles during winter

Figure S9 depicts variations of Cl/Na and S/Na ratios in coarse and fine sea-salt particles. Sea-salt particles internally mixed with mineral particles were excluded, as the figure shows, to avoid misunderstanding of the sea-salt chemistry. The Cl/Na ratios in coarse mode were higher than those in fine mode. A similar tendency was observed and discussed in spring in the Arctic (Hara et al., 2003) and Antarctica (Hara et al., 2005, 2013). High Cl/Na ratios and small variance of Cl/Na ratios in both modes corresponded to samples collected under conditions with blowing or drifting snow and strong winds. Under calm wind conditions, Cl/Na ratios in sea-salt particles vary greatly, which implies that sea-salt particles immediately after release from the sea-ice surface were modified, but not significantly.

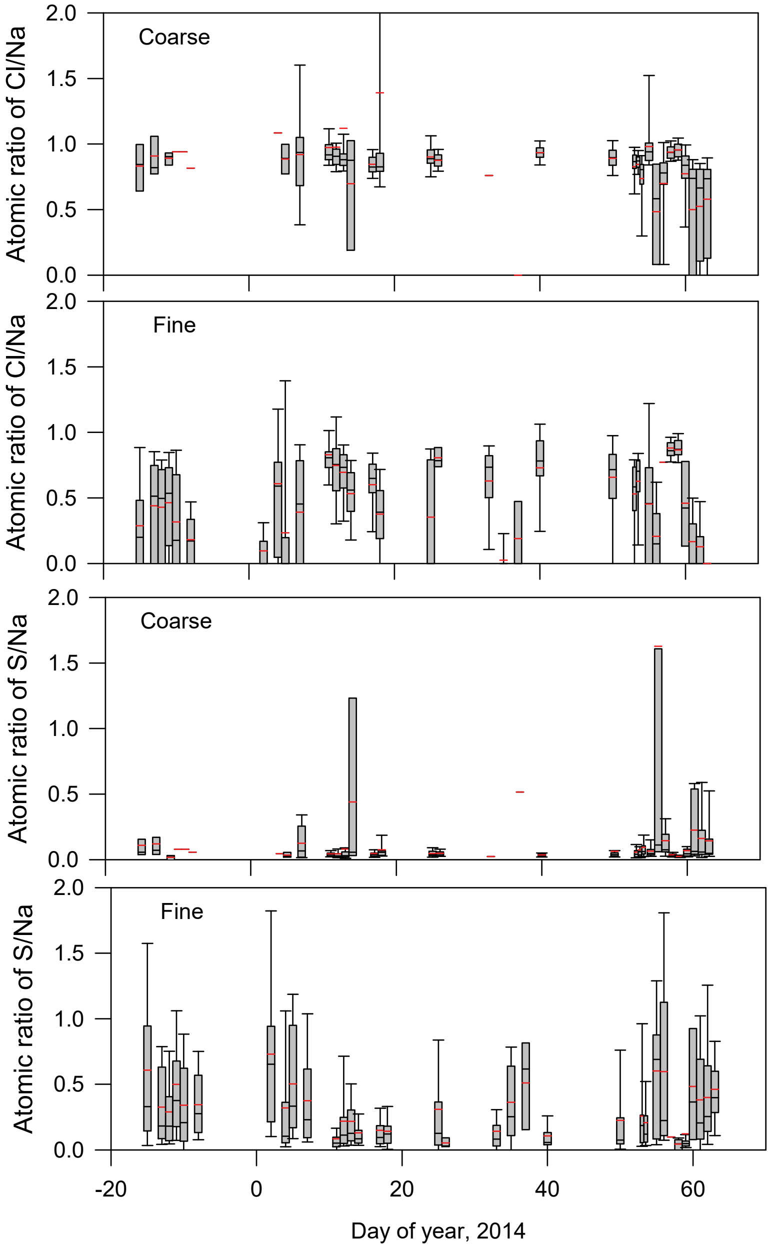

Figure S10 Variations of atomic ratios of Cl/Na and S/Na in sea-salt particles and the modified sea-salt particles collected over the sea-ice area. In box plots, the top bar, top box line, black middle box line, bottom box line, and bottom bar respectively show values of 90, 75, 50 (median), 25, and 10%. The red line shows mean values.