Frost flowers and sea-salt aerosols over seasonal sea-ice areas in north-western Greenland

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Sea-salts and halogens in aerosols, frost flowers and brine play an important role in atmospheric chemistry in polar regions. Sea-salt fractionation proceeds on new and young sea ice. For that reason, sea-salt ratios in sea-salt particles (or aerosols) released from sea-ice areas differ from those of bulk seawater ratio, because of sea-salt fractionation on frost flower and in brine. Sea-salt fractionation can engender modification of aerosol hygroscopicity, which is closely related to phase transformation, heterogeneous reactions, and abilities of cloud condensation nuclei and ice nuclei. To elucidate the atmospheric impact of fractionated sea-salt particles, and their relation between sea-salt particles in the atmosphere and frost flowers on sea ice, one must ascertain their (1) chemical properties (e.g., concentrations, ratios, and pH) of frost flowers and brine, and (2) the physical and chemical properties of aerosols (e.g., size distribution, constituents, and mixing states) above seasonal sea ice with frost flowers. In spite of the importance, simultaneous observations and measurements of aerosols and frost flowers over seasonal ice areas with frost flower appearance have not been reported for polar regions. Simultaneous sampling and observations of frost flowers, brine, and aerosol particles were conducted on several types (fresh - aged) of sea-ice and frost flowers around Siorapaluk in north-western Greenland during December 2013 –March 2014. Sea-salt constituents in frost flowers were determined with ion chromatograph (IC). Br- and iodine (I^- + IO_3^-) were analyzed with IC-MS and ICP-MS, respectively. Individual aerosol particles were observed and analyzed using SEM-EDX.
Results show that water-soluble frost flower and brine constituents were sea salt constituents (e.g., Na^+, Cl^-, Mg^{2+}, and Br^-). Concentration factors of sea-salt constituents of frost flowers and brine relative to seawater were 1.14–3.67. Sea-salt enrichments of Mg^{2+}, K^+, Ca^{2+}, and halogens (Cl^-, Br^-, and I) in frost flowers were associated with sea-salt fractionation by precipitation of mirabilite (Na_2SO_4 $10H_2O$), hydrohalite (NaCl $2H_2O$), and sylvite (KCl). Comparison between sea-salt ratios in brine and frost flower implied that precipitation of mirabilite and hydrohalite proceed in slush layer and then the residual brine were migrated vertically onto frost flowers in our research conditions. Molar ratios of sea-salts (Mg^{2+}/Cl^-, K^+/Cl^-, Ca^{2+}/Cl^-, and Br^-/Cl^-) changed gradually with aging and growth of frost flowers and sea-salt fractionation under colder conditions. Furthermore, sea-salt fractionation was associated with not only surface air temperature but also sea-ice thickness which related to heat conduction from seawater. In contrast to Br- enrichment in frost flower with the aging, changes of I/Cl^- ratio in frost flowers, however, were not clear. Iodine release from frost flowers might be more likely to proceed relative to Br^- release. Aerosol number concentrations, particularly in coarse mode, were increased considerably by release from the sea-ice surface under strong wind conditions. Sulphate depletion by sea-salt fractionation was found to be slight in sea-salt aerosols because of heterogeneous SO_4^{2-} formation on sea-salt particles. Mg was enriched in coarse and fine sea-salt particles collected on sea-ice area. Mg in sea-salt particles was in the form of $MgCl_2$ and $MgSO_4$. Strong Mg enrichment might be more likely to proceed in fine sea-salt particles. Mg enrichment in sea-salt particles was enhanced under colder conditions. In addition, ikaite-like and mirabilite-like particles identified in the atmosphere only near new sea ice are close to the sea-ice margin. Thus, Ikaite-like and mirabilite particles might be released from initial sea-ice before freezing over, and Mg-rich sea-salt particles might be released from sea-ice surface with frost flowers.

Keywords: Sea-salt aerosols, Halogens, Frost flower

