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Surface Snowpack Key to Bromine Activation in a Changing Arctic Environment

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Arctic sea ice is rapidly declining and transforming from a multiyear ice pack to thinner, more saline, seasonal ice, which has important implications for Arctic atmospheric composition. Following springtime polar sunrise, "ozone depletion events", attributed to bromine chemistry, lead to episodic decreases in lower tropospheric ozone concentrations to near zero, concurrent with mercury depletion and deposition. Despite our increasing understanding of the spatial variability of BrO and possible reaction pathways based on laboratory studies, important questions remain regarding the most efficient sources of and mechanisms for Arctic halogen activation. During the March-April 2012 BRomine, Ozone, and Mercury EXperiment (BROMEX) in Barrow, Alaska, outdoor chamber experiments with snow and ice samples were conducted. Ozone was added as the precursor oxidant, and the samples were investigated with and without ambient sunlight. Samples included first-year sea ice, brine icicles, several layers of snow above first-year sea ice, and seasonal snow above the tundra. Chemical ionization mass spectrometry was utilized to monitor Br2 production. Tundra snow and surface snow above sea ice produced the most Br2, with no production resulting from sea ice and basal snow directly above sea ice. Overall, the most efficient Br2 production was observed from snow samples characterized by lower pH and higher bromide/chloride ratios. Br2 was only observed in the presence of sunlight, indicating the role of snowpack photochemical reactions and the hydroxyl radical in its production. Br2 production via the surface snowpack explains previously-observed BrO enhancements above sea ice, as well as observations of inland tundra hotspots in measured BrO by aircraft-based nadir MAX-DOAS (Multi Axis-Differential Optical Absorption Spectroscopy) measurements, conducted during BROMEX. The findings indicate that atmospherically processed snow is likely a major source of Arctic bromine release, which impacts the distribution and occurrence of ozone depletion events and BrO. The implications of snowpack photochemical bromine production on tropospheric reactive bromine concentrations were explored using the one-dimensional halogen model MISTRA.