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Abiotic sources of reactive organic halogens in the Arctic atmosphere

L. J. Carpenter (1), J. R. Hopkins (1), C. E. Jones (1), A.C. Lewis (1), R. Parthipan (1), D. J. Wevill (1), Laurier Poissant (2)

(1) Department of Chemistry, University of York, York, (2) Service metéorologique du Canada, Montréal, Canada (ljc4@york.ac.uk / Fax: +44 1904 432516)

Sudden ozone and mercury depletion events in the spring-time polar boundary layer are a widespread and well-known phenomena that are apparently caused by autocatalytic release of inorganic bromine from the sea-ice surface, particularly frost flowers, and/or from sea salt particles on the snow pack. The role played by bromine has been reported in numerous studies and the suggested mechanism involves uptake of HOBr into a slightly acidic and concentrated brine (containing enhanced Br^- and Cl^-) on the sea-ice surface resulting in release of photolysable Br₂ and BrCl gas that rapidly enters ozone-depleting cycles. Little is known about the role of gaseous iodine in polar ozone chemistry despite iodine containing aerosol being associated with lower tropospheric ozone depletion at polar sunrise. Recent theoretical studies indicate that CH_2I_2 would be an extremely effective agent for tropospheric Arctic ozone depletion and that iodine compounds added to a Br₂/BrCl mixture have a significantly greater ozone (and mercury) depletion effect than additional Br_2 and BrCl molecules. Here we report the first observations of CH₂I₂, CH₂IBr, CH₂ICl, CHBr₃ and other reactive halocarbons during polar sunrise at Hudson Bay. The occurrence of such species was associated with northwesterly winds from the frozen bay, and, in the case of CHBr₃, anticorrelated with ozone and total gaseous mercury (TGM), suggesting a common source of inorganic and organic halogens. The absence of local ice leads coupled with the extremely short atmospheric lifetime of CH₂I₂indicates that production occurs in the surface of the snowpack over the bay. We propose an abiotic mechanism for the production of polyhalogenated iodo- and bromocarbons, via reaction of HOI and/or HOBr with organic material on the quasi-liquid layer above sea-ice/snow pack, and report laboratory data to support this mechanism.