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3-D modeling of bromine chemistry and the boundary-layer depletion of ozone and mercury across the springtime Arctic: model evaluation using field and satellite data

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Gas-phase bromine radical chemistry is the main driver for the frequent and concurrent depletion of ozone and mercury (Hg) in the polar boundary layer during the spring. Sea ice and its overlying snow cover are broadly understood as the key elements in the production of reactive bromine in polar spring. However, a full characterization remains unsettled on how physicochemical states of snow and ice influence the release of bromine into the atmosphere. Uncertainties in the kinetics and reaction mechanisms of Hg redox chemistry add further complexity to accurately assessing the behavior of Hg during its depletion from air. Three-dimensional (3-D) models, developed to simulate the impact of bromine chemistry on Hg oxidation at both global and arctic-basin regional scales, have generally relied upon indirect representations of the sources, sinks and photochemical transformation of bromine radical species in the polar atmosphere. Within Environment and Climate Change Canada's air-quality model, GEM-MACH, we have developed a process-oriented representation for the coupled bromine-ozone-Hg chemistry and the exchange of bromine, ozone and Hg species between air and snow/ice surface. The model is run at 15-km horizontal resolution in a limited-area domain of the Arctic and is capable of capturing the evolution of high BrO column densities associated with synoptic weather disturbances during polar sunrise as can be seen from satellite. The concurrent depletion of ozone and Hg is simulated by consistent model formulations, where the release of reactive bromine from the frozen surfaces is facilitated by the presence of ozone in air. The concentrations of ozone and speciated Hg measured in the surface air and the vertical column densities of BrO retrieved from ground stations and buoys floating on the ice-covered ocean also allow us to evaluate the model simulations at fine temporal scales even though limited in spatial coverage. Our model framework for simulating the reactive bromine release from the snow/ice cover is found to work reasonably well for the representation of ozone and Hg depletion events across the Arctic region over synoptic and seasonal time scales. The deposition of oxidized Hg from the atmosphere is found to be enhanced particularly under the disturbed weather conditions, as a result of the vertical and horizontal inflow of ozone and gaseous elemental Hg otherwise persistently depleted in the polar boundary layer.