



Contribution ID: 36

Type: **not specified**

Reactive halogen release from the polar snowpack and the depletion of ozone and mercury in the air: Insights from 1-D (mechanistic) and 3-D (chemical transport) models

Wednesday, 8 June 2011 14:00 (15 minutes)

Reactive halogens, especially bromine, are known to play a major role in the rapid loss of both ozone and gaseous elemental mercury (GEM) in the polar boundary layer during the spring. Measurements of relevant chemical species in the field and from satellites have shown that reactive bromine is released to the high-latitude atmosphere extensively and most notably from the ice-covered ocean. However, there are a lot of open questions with regard to (micro-) physical and chemical mechanisms behind the occurrence of the high bromine content in the polar snow and the movement and transformation of bromine (and chlorine and iodine as well) between seawater, sea ice, snowpack and atmosphere.

In this talk, we first present the status of our development of a 1-D mechanistic model (PHANTAS, a model of PHotochemistry ANd Transport in Air and Snowpack), which describes the two-way transport of trace gases between the snowpack and the overlying atmosphere as well as their photochemical reactions in the gas phase and in the condensed phase of aerosols and snowpack. Vertical diffusivity in the lowest hundred meters of the atmosphere is diagnosed empirically from surface wind speed and heat flux. Gas transport in the snowpack interstitial air is represented by a combination of molecular diffusivity and wind-pumping advection. Model simulations show that strong surface winds, as well as promoting turbulent mixing in the boundary layer, also lead to the pumping of air in and out of the snowpack with the consequent release of reactive halogens. Ozone and GEM are both lost rapidly via reactions with bromine atoms in both the atmospheric boundary layer and the snowpack interstitial air while the level of the bromine activation in the snowpack is highly sensitive to the assumed fate of key radical species against gas-to-snow uptake. The effect of dissolved organic matters (DOMs), in reducing the oxidized mercury back into elemental form, is represented by a combination of one-step, first-order photolysis reactions of mercury-DOM complexes of varying stability. The sensitivity of model results to the initial concentration profiles of halides and DOMs in the snow as well as to the type of DOM-mercury interactions is explored in order to provide insights into the fate of mercury after its deposition to the polar snow.

We also present results from a 3-D model (GEM-AQ) with a simple representation of air-snow chemical interactions to study reactive bromine production and ozone depletion in the Arctic boundary layer (Toyota et al., 2011, ACP). A set of sensitivity experiments have been performed to explore the effects of temperature and the age of sea ice (first-year, FY, versus multi-year, MY) on the release of reactive bromine to the atmosphere. The model simulations capture much of the temporal variations in the surface ozone mixing ratios as observed at stations in the high Arctic and the synoptic-scale evolution of "BrO clouds" in the lower troposphere as estimated from satellite observations. In this work it was suggested that reactive bromine is released ubiquitously from the snow on the sea ice during the Arctic spring while the timing and location of the bromine release are largely controlled by meteorological factors but through the transport of ozone to the surface air over the snow/ice surface. The evaluation of the various simulations indicated, although moderately, the likelihood that physical and/or chemical conditions on the FY sea ice are more advantageous for bromine release relative to the conditions on the MY sea ice, as suggested previously by Simpson et al. (2007). Also, if indeed the surface snowpack does supply most of the reactive bromine in the Arctic boundary layer, it appears to be capable of releasing reactive bromine at temperatures as high as -10 degree Celsius, particularly on the sea ice in the central and eastern Arctic Ocean.

Please list some keywords

Ozone, bromine, mercury, snowpack, Arctic, Sea ice, GEM-AQ, PHANTAS

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Session Classification: Modeling Workshop