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Modeling chemistry in and above snow at Summit, Greenland: Impact of snowpack chemistry on the oxidation capacity of the boundary layer

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The chemical composition of the boundary layer in snow covered regions is impacted by chemistry in the snowpack via uptake, processing, and emission of atmospheric trace gases. We use the coupled one-dimensional snow chemistry and atmospheric boundary layer model MISTRA-SNOW to study the impact of snowpack chemistry on the oxidation capacity of the boundary layer. The model includes gas phase photochemistry and chemical reactions both in the 5 interstitial air and the atmosphere. Chemistry on snow grains is simulated assuming a liquid-like layer (LLL), treated as an aqueous layer on the snow grain surface. The model has been recently compared with BrO and NO data taken on June 10 - June 13, 2008 as part of the Greenland Summit Halogen-HO_x experiment (GSHOX). In the present study, we use the same focus period to investigate the influence 10 of snowpack derived chemistry on OH and HO_x+RO_2 in the boundary layer. We compare model results with chemical ionization mass spectrometry (CIMS) measurements of the hydroxyl radical (OH) and of the hydroperoxyl radical (HO_2) plus the sum of all organic peroxy radicals (RO_2) taken at Summit during summer 2008. Using sensitivity runs we show that snowpack influenced nitrogen cycling and bromine chemistry both increase the oxidation capacity of the boundary layer and that 15 together they increase the mid-day OH concentrations by a factor of $\sim$2. We show for the first time, using an unconstrained coupled one-dimensional snowpack-boundary layer model, that air-snow interactions impact the oxidation capacity of the boundary layer and that it is not possible to match measured OH levels without snowpack NO_x and halogen emissions. Model predicted HONO compared with mistchamber measurements suggests there is a large unknown HONO source at Summit. Other model predicted HO_x precursors, H_2O_2 and HCHO, compare well with 20 measurements taken in summer 2000. Snow sourced NO_x contributes an additional 2 ppb of boundary layer ozone over 3 days, while snow sourced bromine has the opposite effect and contributes 1 ppb of boundary layer ozone loss over 3 days.