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Synoptic-scale meteorological control on reactive bromine production and ozone depletion in the Arctic boundary layer: 3-D simulation with the GEM-AQ model

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Episodes of high bromine levels and surface ozone depletion in the springtime Arctic are simulated by an online air-quality model, GEM-AQ, with gas-phase and heterogeneous reactions of inorganic bromine species and a simple scheme of air-snowpack chemical interactions implemented for this study. Snowpack on sea ice is assumed to be the only source of bromine to the atmosphere and capable of converting relatively stable bromine species to photolabile $\mathrm{Br_2}$ via air-snowpack interactions. A "bromine explosion", by which $\mathrm{Br^-}$ retained in the snowpack is autocatalytically released to the atmosphere as a result of dry deposition of HOBr and $\mathrm{BrONO_2}$, is assumed to occur on young, first-year (FY) sea ice (or its overlying snowpack), whereas the snowpack on old, multi-year (MY) sea ice and over land is assumed only to recycle a part (but up to 100%) of bromine reservoirs lost via dry deposition back to $\mathrm{Br_2}$. Model runs are performed for April 2001 at a horizontal resolution of approximately 100 km x 100 km in the Arctic. The model simulates temporal variations in surface ozone mixing ratios as observed at stations in the high Arctic and the synoptic-scale evolution of enhanced BrO column amounts ("BrO clouds") as seen from satellite reasonably well. The results strongly suggest: (1) the ubiquitous source of reactive bromine exists on the FY sea ice during the Arctic springtime; and (2) the timing of bromine release to the atmosphere is largely controlled by meteorological forcing on the transport of ozone to the near-surface air. Also, if reactive bromine in the Arctic boundary layer is supplied predominantly from the surface snowpack, it should be capable of releasing reactive bromine at temperatures as high as $-10\ ^\circ\mathrm{C}$, particularly on the FY sea ice in the central and eastern Arctic Ocean. Dynamically-induced BrO column variability in the lowermost stratosphere appears to interfere with the use of satellite BrO column measurements for interpreting BrO variability in the lower troposphere but probably not to the extent of totally obscuring "BrO clouds" associated with the surface source of bromine in the high Arctic. Contrary to our original intention, the present air-snowpack interaction scheme yields a majority of atmospheric bromine input via $\mathrm{Br_2}$ release associated empirically with a dry deposition of ozone on the snow/ice surface under sunlight to represent a trigger of bromine explosion. This implies that the bromine explosion actually occurs in the interstitial air of snowpack and/or is accelerated by heterogeneous reactions on the surface of wind-blown snow in ambient air, both of which are missing in our model but could have been approximated by a parameter adjustment for the yield of $\mathrm{Br_2}$ from the trigger.