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The potential importance of frost flowers, recycling on snow, and open leads for ozone depletion events

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Abstract. We present model studies with the one-dimensional model MISTRA to investigate the potential role of frost flowers, recycling on snow, and open leads in the depletion of tropospheric ozone in the Arctic spring. In our model, we assumed frost flower aerosols to be the major source of bromine. We show that a major ozone depletion event can be satisfactorily reproduced only if the recycling on snow of deposited bromine into gas phase bromine is assumed. In the model, this cycling is more efficient than the bromine explosion process and maintains sufficiently high levels of bromine to deplete ozone down to few nmol mol^{-1} within four days. We assessed the influence of different surface combinations (open lead/frost flowers) on the chemistry in the model. Results showed noticeable modifications affecting the composition of aerosols and the deposition velocities. A model run with a series of coupled frost flower fields and open leads, separated by large areas of snow, showed results comparable with field observations. In addition, we studied the effects of modified temperature of either the frost flower field or the ambient airmass. A warmer frost flower field increases the relative humidity and the aerosol deposition rate. The deposition/re-emission process gains in importance, inducing more reactive bromine in the gas phase, and a stronger ozone depletion. A decrease of 1K in airmass temperature shows in our model that the aerosol uptake capacities of all gas phase species substantially increases, leading to enhanced uptake of acids from the gas phase. Consequently, the so-called bromine explosion accelerated and O_3 mixing ratios decreased. In our model representation, variations in wind speed affected the aerosol source function and influenced the amount of bromine in the atmosphere and thus the ozone depletion strength. Recent studies have suggested the important role of the precipitation of calcium carbonate ($CaCO_3$) out of the brine layer for the possible acidification of the liquid phase by acid uptake. Our investigation showed that this precipitation is a crucial process for the timing of the bromine explosion in aerosols. Nevertheless, model runs with either 50% precipitation or complete precipitation displayed a relatively weak difference in ozone mixing ratios after four simulated days. By considering conditions typical for "Arctic Haze" pollution events at the start of the run we obtained a low pH in frost flower aerosols due to a greater mixing ratio of SO_2, and a strong recycling efficiency via large aerosol number concentration. The aerosol acidification during a haze event most likely intensifies the ozone depletion strength and occurrence. The comparison between our modeled deposition on snow and sampled snow at Barrow (Alaska) shows that approximately 75% of

deposited bromine may be re-emitted into the gas phase as Br_2/BrCl. Among several non-halogen fluxes from the snow, model simulations showed that only HONO affects the chemistry. Finally, we investigated the release of Br_2 potentially produced by heterogeneous reactions directly on frost flowers. In this case, we obtained unrealistic results of aerosol compositions and deposition rates on snow compared to observations in the Arctic.

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