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Modeling of the Recurrence of Tropospheric Ozone Depletion Events in the Arctic Spring

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Ozone depletion events have been observed in the lower troposphere during spring and to a lesser extent during fall in both, the Arctic and the Antarctic. During an ozone depletion event, the mixing ratio of ozone in the atmospheric boundary layer drops from its background level of several 10 parts per billion (ppb) to small or even undetectable levels (< 1 ppb) on a timescale of hours to days. Ozone depletion events are driven by halogen chemistry. Reactive halogen species are released by both, gas phase and heterogeneous reactions, the latter occur on the surface of substrates such as snow, ice, or aerosols. Halogenides stored in substrates may be activated by strong oxidants like HOBr or O_3 and are initially released into the gas phase as Br_2 or BrCl. Under sunlight, the photolysis of Br_2 forms two Br atoms, which consume ozone and form BrO, which is converted to new HOBr. This autocatalytic process is referred to as "bromine explosion".

Frequently, several consecutive ozone depletion events (ODEs) are observed at the same location, e.g. thirteen ODEs were observed at Barrow, Alaska, during the spring of 2009.

This recurrence of ODEs was thought to be caused solely by horizontal transport of air from ozone-depleted regions to the measurement site. In the present numerical investigation, the chemical reactions in a one-dimensional model configuration are studied where the vertical turbulent diffusion acts as the sole transport mechanism in order to investigate possible different scenarios of the recurrence of ODEs. It is fond that ODEs are occurring periodically with periods of high ozone in between, the first recurrence seen after 20 days, and thereafter, ODEs recur approximately every 15 days. The first ozone depletion takes about two days and the following ozone depletions last for approximately one day each. The precondition for the recurrence in the model is the removal of the reactive halogen species from the boundary layer, either due to dry deposition on the ice/snow surface and the aerosols or through vertical mixing into the upper troposphere. It is found that a strong inversion layer is needed for the reactive halogen species to vanish and the ODEs to terminate. The ozone can then replenish through transfer from the upper troposphere to the boundary layer or through photolytic reactions of NO_x species. PAN appears to play an important role to regulate the amount of NO_x after the termination of an ODE.

Moreover, the Weather Research and Forecasting model coupled to Chemistry (WRF-Chem) is used to perform three-dimensional simulations of ozone depletion events. For this purpose, the MOZART reaction mechanism is extended to include bromine species, and the results are compared to satellite data of bromine VCDs north of Barrow, Alaska, in 2009. The results show good qualitative agreement with the observations.