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First remote sensing measurements of ClOOCl along with ClO and $ClONO_2$ in activated and deactivated Arctic vortex conditions using new ClOOCl IR absorption cross sections

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Abstract. Active chlorine species play a dominant role in the catalytic destruction of stratospheric ozone in the polar vortices during the late winter and early spring seasons. Recently, the correct understanding of the ClO dimer cycle was challenged by the release of new laboratory absorption cross sections (Pope et al., 2007) yielding significant model underestimates of observed ClO and ozone loss (von Hobe et al., 2007). Under this aspect, nocturnal Arctic stratospheric limb emission measurements carried out by the balloon version of the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS-B) from Kiruna (Sweden) on 11 January 2001 and 20/21 March 2003 have been reanalyzed with regard to the chlorine reservoir species $ClONO_2$ and the active species, ClO and ClOOCl (Cl_2O_2). New laboratory measurements of IR absorption cross sections of ClOOCl for various temperatures and pressures allowed for the first time the retrieval of ClOOCl mixing ratios from remote sensing measurements. High values of active chlorine (ClO_x) of roughly 2.3 ppbv at 20 km were observed by MIPAS-B in the cold mid-winter Arctic vortex on 11 January 2001. While nighttime ClOOCl shows enhanced values of nearly 1.1 ppbv at 20 km, $ClONO_2$ mixing ratios are less than 0.1 ppbv at this altitude. In contrast, high $ClONO_2$ mixing ratios of nearly 2.4 ppbv at 20 km have been observed in the late winter Arctic vortex on 20 March 2003. No significant ClO_x amounts are detectable on this date since most of the active chlorine has already recovered to its main reservoir species $ClONO_2$. The observed values of ClO_x and $ClONO_2$ are in line with the established polar chlorine chemistry. The thermal equilibrium constants between the dimer formation and its dissociation, as derived from the balloon measurements, are on the lower side of reported data and in good agreement with values recommended by von Hobe et al. (2007). Calculations with the ECHAM/MESSy Atmospheric Chemistry model (EMAC) using established kinetics show similar chlorine activation and deactivation, compared to the measurements in January 2001 and March 2003, respectively.

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