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The importance of methane oxidation for chlorine activation and ozone depletion in Antarctic spring

R. Müller (1), A. M. Zafar (1), J.-U. Grooß (1), and R. Lehmann (2)

(1) Forschungszentrum Jülich GmbH, IEK-7, Jülich, Germany (ro.mueller@fz-juelich.de), (2) Alfred Wegner Institut, Potsdam, Germany

The Antarctic ozone hole arises from ozone destruction driven by elevated levels of ozone destroying ("active") chlorine species in Antarctic spring. The established picture of the development of the ozone hole is that high levels of active chlorine are maintained in Antarctic spring by a competition between chlorine deactivation through gas-phase formation of $ClONO_2$ and HCl and activation of $ClONO_2$ and HCl by heterogeneous reactions. Here we show that in the heart of the ozone hole (16-18 km or 100-70 hPa, in the core of the vortex) formation of $ClONO_2$ is only of minor importance and that formation of HCl by reaction of Cl with CH_4 and CH_2O cannot lead to deactivation because it is balanced by immediate reactivation in effective reaction cycles involving the heterogeneous reaction HCl + HOCl. Further, for the (observed) almost complete activation of stratospheric chlorine the production of HOCl via HO_2 + ClO, is the dominating process. In this case, the necessary HO_2 results from CH_2O photolysis. These results are key for assessing the impact of changes of the future stratospheric composition on the recovery of the ozone hole. Based on our simulations we conclude that future increased methane concentrations will not lead to enhanced chlorine deactivation and that extreme ozone destruction to levels below ≈ 0.1 ppm will occur until mid-century.