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The potential impact of ClO_x radical complexes on polar stratospheric ozone loss processes

B. Vogel¹, W. Feng², M. Streibel^{3,4}, and R. Müller¹¹Research Centre Jülich, Institute for Stratospheric Research (ICG-I), Jülich, Germany²School of Earth and Environment, University of Leeds, Leeds, UK³Alfred Wegener Institute for Polar and Marine Research (AWI), Potsdam, Germany⁴European Ozone Research Coordinating Unit, University of Cambridge, Cambridge, UK

Abstract. The importance of radical-molecule complexes for atmospheric chemistry has been discussed in recent years. In particular, the existence of a ClO·O₂ and ClO_x water radical complexes like ClO·H₂O, OCIO·H₂O, OCIO·(H₂O)₂, and ClOO·H₂O could play a role in enhancing the ClO dimer (Cl₂O₂) formation and therefore may constitute an important intermediate in polar stratospheric ozone loss cycles. Model simulations performed with the Chemical Lagrangian Model of the Stratosphere (CLaMS) will be presented to study the role of radical complexes on polar stratospheric ozone loss processes. The model simulations are performed for the Arctic winter 2002/2003 at a level of 500 K potential temperature and the results are compared to observed ozone loss rates determined by the Match technique. Moreover, recently reported values for the equilibrium constant of the ClO dimer formation are used to restrict the number of possible model results caused by large uncertainties about radical complex chemistry. Our model simulations show that the potential impact of ClO·O₂ on polar ozone loss processes is small ($dO_3/dt \ll 0.5$ ppb/sunlight h) provided that the ClO·O₂ complex is only weakly stable. Assuming that the binding energies of the ClO_x water complexes are much higher than theoretically predicted an enhancement of the ozone loss rate by up to ≈ 0.5 ppb/sunlight h is simulated. Because it is unlikely that the ClO_x water complexes are much more stable than predicted we conclude that these complexes have no impact on polar stratospheric ozone loss processes. Although large uncertainties about radical complex chemistry exist, our findings show that the potential impact of ClO_x radical molecule complexes on polar stratospheric ozone loss processes is very small considering pure gas-phase chemistry. However the existence of ClO_x radical-molecule complexes could possibly explain discrepancies for the equilibrium constant of the ClO dimer formation found between recent laboratory and stratospheric measurements.

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