



On the discrepancy of chlorine activation in the dark polar vortices

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More than three decades after the discovery of the ozone hole, the processes involved in its formation are believed to be understood in great detail. Current state-of-the-art models are able to reproduce the distribution of the chemical compositions in the polar stratosphere, especially regarding the quantification of halogen-catalyzed ozone loss. However, here we report on a discrepancy between simulations and observations during the less-well studied period of the onset of chlorine activation. During this period, in the Antarctic between May and July, the simulations significantly over-estimate HCl, one of the key chemical species, inside the polar vortex during polar night. This HCl-discrepancy is present in the polar winter of both hemispheres in three independent models, the Lagrangian chemistry transport model CLaMS and the Eulerian models WACCM and TOMCAT/SLIMCAT. The HCl-discrepancy points to some unknown process in the formulation of stratospheric chemistry that is currently not represented in the models.

The HCl-discrepancy is characterized in space and time for the Lagrangian Chemistry Transport Model CLaMS. The discrepancy in the models WACCM and TOMCAT/SLIMCAT is developed to a lesser extent. Numerical diffusion in the Eulerian models is identified to be a likely cause for the inter-model differences. Although the missing process has not yet been identified, we investigate different hypotheses on the basis of the characteristics of the discrepancy. The ionisation caused by Galactic Cosmic Rays provides an additional NO_x and HO_x source that can explain a fraction of the discrepancy. An under-estimated uptake of HCl into the particles cannot explain the discrepancy due to the temperature correlation of the discrepancy. Also, a direct photolysis of particulate HNO_3 does not explain the discrepancy since it would also cause changes in the later course of the winter that have not been observed. A hypothetical decomposition of particulate HNO_3 by some other process not dependent on the solar elevation, e.g. involving Galactic Cosmic Rays may be a possible mechanism to resolve the HCl-discrepancy. Since the discrepancy is mainly seen during the beginning of the chlorine activation period where the ozone loss rates are slow, there is only minor impact on the overall ozone loss over the course of the winter and spring.