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Using HDO-HHO correlations to examine stratospheric chemistry and transport processes

Jörg Steinwagner (1), Thomas Röckmann (1), and Gabriele P. Stiller (2)

(1) Institute for Marine and Atmospheric Research Utrecht, Utrecht University, 3508 TA Utrecht, The Netherlands, (2) Karlsruhe Institute of Technology , Institute for Meteorology and Climate Research, 76021 Karlsruhe, Germany

The chemical processing of hydrogen in the stratosphere is to large degree a closed system where hydrogen is cycled between the main species H_2O, CH_4 and H_2. The analogue is true for deuterium D and the deuterated species HDO, CH_3D and HD. Strong correlations exist between the different species and their deuterated isotopologues. This makes it possible to examine the stratospheric hydrogen budget by examining the correlation of the global distributions of stratospheric deuterated (HDO) and non-deuterated water (HHO). Using satellite measurements made with the MIPAS instrument on Envisat provides a global view. Our findings support to a large extent a linear relationship between HDO and HHO that can be inferred from basic assumptions found in the literature (McCarthy et al., 2004). Deviations from this linear relationship exist at all latitudes. In the tropics we observe deviations that are linked to the tape recorder (Mote et al., 1996). At latitudes between +/-[10° 70°] seasonal deviations are localized in certain altitudes at around 25 km, pointing to the existence of a small maximum of roughly 5% in molecular hydrogen under summer conditions. Large deviations from the linear relationships are also observable in the winter months over Polar regions. During descent mesospheric air from altitudes above 55 km is transported downwards to altitudes around 40 km, where it partly mixes with upper stratospheric air. Such intrusions are clearly visible in correlations of deuterated and non-deuterated species because photolysis as the major sink process for water in the mesosphere differently impacts different water isotopologues, i.e. yields enrichment in HDO (Miller et al., 2000, Sandor et al., 2003). However, a close examination of the intrusions over the South Pole reveals a layer between the upper stratospheric and lower mesospheric air where HDO is being destroyed and at the same time the probed air mass gets depleted in deuterium.