DEUTERIUM, OXYGEN-18, AND TRITIUM AS TRACERS FOR WATER VAPOUR TRANSPORT IN THE LOWER STRATOSPHERE AND TROPOPAUSE REGION

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Abstract. Simultaneous measurements of the three rare isotopes Deuterium (D), Tritium (T), and Oxygen-18 ($^{18}$O) in water vapour were made for the first time in the vicinity of the northern hemisphere tropopause. In contrast to expectation, high D/H and $^{18}$O/$^{16}$O ratios, but relatively low T/H ratios, were found within the lowermost stratosphere. Since water vapour in the low-latitude upper troposphere shows a similar isotopic signature, we conclude that in the mid-latitudes considerable amounts of tropospheric water vapour are injected into the lowermost stratosphere, probably resulting in a hydration of the lower stratosphere. In addition, T can serve as tracer for precipitation of water containing stratospheric aerosol particles, because the T/H ratio in stratospheric water vapour is orders of magnitude higher than in the upper troposphere. Thus, even a small contribution of water of stratospheric origin should be detectable in the tropopause region. In our measurements performed in the Arctic we did not find isotopic evidence for sedimentation of PSC particles down to the tropopause. This may be caused by the low spatial and temporal coverage of our observations; however, it may also be due to the much weaker wintertime dehydration of the Arctic vortex compared to the Antarctic.

Keywords: Isotopic composition of H$_2$O, water vapour transport, stratospheric-tropospheric exchange
1. Introduction

Water is one of the most important atmospheric trace constituents: it plays a key role (a) in the radiation balance of the atmosphere and (b) as reactant or reaction substrate (in liquid or solid form) for numerous homogenous and heterogenous chemical processes in the troposphere and stratosphere [IPCC 1995, Moortgat et al. 1995]. Although water vapour transport is an old topic of atmospheric research, in the recent years much effort has been devoted to its understanding, in particular within the stratosphere. For instance, the seasonal change of the H$_2$O mixing ratio in the tropical lower stratosphere ('tape recorder effect') clearly reflects the upward movement of air masses and the seasonal change in temperature there [Mote et al. 1996].

The main scope of this study is to obtain information about water vapour transport in the atmosphere from spatial variations in the isotopic composition of the water vapour. This technique is based on the fact that each phase transition of water vapour (evaporation, condensation) changes its isotopic composition [Raye 1987]. Thus, isotopic data for H$_2$O should trace the transport history of water vapour samples and accordingly should give information which cannot be derived by customary H$_2$O concentration measurements alone.

Our HDO, H$_2$O$^{18}$O, and HTO data collected in the vicinity of the northern extratropical tropopause are interpreted with regard to two transport processes of water vapour in the atmosphere: (a) Isentropic troposphereto-stratosphere transport across the extratropical tropopause leading to hydration of the lowermost stratosphere, and (b) sedimentation of PSC particles within the Arctic polar stratosphere.

(a) Already in the late 1960s first indications were reported that around tropopause foldings associated with upper-level cyclogenesis tropospheric air can irreversibly be mixed upward into the stratosphere (see [Reiter et al. 1966]). More recent experimental and theoretical studies confirmed the presence of this transport phenomenon [Ebel et al. 1991, Hoerling et al. 1993, Chen 1995, Dessler et al. 1995, Holton et al. 1995, Appenzeller et al. 1996]. Owing to the higher temperature and accordingly higher water vapour pressure encountered near the mid-latitude tropopause compared to the tropical tropopause, such an extratropical cross-tropopause flux should lead to hydration of the lower stratosphere.

(b) It is widely accepted that the extent of dehydration and denitrification of the polar winter stratospheres caused by PSC sedimentation is a crucial parameter determining the photochemical ozone destruction in early spring [Ondra and Turco 1991, Hofmann and Oltmans 1992, Beyer et al. 1997]. Because of the lower temperatures inside the southern polar vortex compared to the northern polar vortex, the formation of large, fast sedi-