



## **The long-term trend in the fractionation of stratospheric water and its implications for the troposphere-stratosphere exchange**

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Our understanding of processes controlling water entering the stratosphere is incomplete. The unexplained long-term trend of stratospheric water may be due to trends in particulate water entering the stratosphere through the tropical tropopause. Measurements of the isotopic fractionation of water, the deuterium depletion, suggest an important role for convectively lofted ice in the tropical tropopause layer. We present balloon-borne remote sensing measurements of stratospheric H<sub>2</sub>O, HDO, CH<sub>4</sub> and CH<sub>3</sub>D, which allow us to derive the deuterium depletion of water at the entry point into the stratosphere. We find that the seasonal variations of the deuterium depletion are correlated with H<sub>2</sub>O, with a slope in good agreement with the temperature-dependent Rayleigh fractionation. Our data allow for the first time to constrain the magnitude of changes in particulate water entering the stratosphere for the measurement period 1991 – 2007. We find that the measured trends in the deuterium depletion over this period do not show significant deviation from a Rayleigh fractionation. This suggests that over this period changes in particulate water entering the stratosphere were small. As a consequence our measurements suggest that changes in particulate water following changes in aerosol from the increased industrial emissions in Asia and increased tropical biomass burning are unlikely to explain the increase in stratospheric water vapour.