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Analysis of the southern hemispheric H_2 Cycle using D/H measurements of H_2 and CH_4 in Cape Grim air archive

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Understanding the hydrogen cycle has gained additional importance because of the acknowledgement of the potential role of hydrogen as a major energy carrier in the future. We investigate the southern hemispheric H_2 cycle using D/H stable isotope analyses of H_2 and CH_4 in 29 samples of air collected from 1992 and 2004 at the Australian Cape Grim baseline air monitoring station and stored in the unique air archive in Aspendale. The D/H ratios were determined using the fairly new technique of continuous-flow isotope ratio mass spectrometry. Quality control tests for the mixing ratios and isotope ratios of H_2 in archived air confirm the absence of artifacts that could occur during the collection and subsequent storage. A harmonic-polynomial function was applied to isolate seasonal variation and long term trends during the period of observation. Mean seasonal variations of the D/H ratio of H_2 reveal a similar in-phase variation as observed in the southern hemispheric free troposphere (CARIBIC flights). The long-term trends for the mixing ratios and the D/H ratios of H_2 and CH_4 show a remarkable feature of the SH H_2 cycle, namely, the rate of increase of CH_4 is about 3.8 times faster than that of H_2, whereas the D/H ratio of CH_4 increases about 1.8 times slower than that for H_2. This paradox cannot be explained by the photochemical oxidation of CH_4 and H_2 in the southern hemisphere as the major process for these long-term trends. We suspect a strong external influences acting like source or sink for H_2 in the form of inter-hemispheric exchange of air masses.