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Correlations between the deuterium content of molecular hydrogen and other trace gases in the UTLS region obtained from CARIBIC samples

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Hydrogen (H_2) is present in the atmosphere at levels of ∼0.5 ppm. These levels are expected to rise due to use of H_2 as an energy carrier, which may affect methane (CH_4) lifetimes and stratospheric ozone depletion. Unfortunately, large uncertainties still exist in the global H_2budget.

The different sources and sinks of H_2 have very distinct isotopic signatures and fractionation coefficients, respectively. For this reason, measurements of the isotopic composition of H_2 are a promising tool to gain insight into H_2 source and sink processes and to constrain the terms in the global budget.

The CARIBIC project uses an automated instrument container on board of a commercial passenger aircraft to carry out in situ atmospheric measurements and to collect air samples, mostly in the Upper Troposphere Lower Stratosphere (UTLS) region. The CARIBIC samples are routinely analyzed for various gases. At present, more than 480 air samples of 19 CARIBIC return flights have been analysed for molecular hydrogen mixing ratio (m_{H2}) and H_2 deuterium content (δD-H_2).

More than 80 of the analysed samples contain air from the lower stratosphere. In the few previous experiments in the stratosphere it was found that production (from CH_4 oxidation) and destruction of H_2 (by hydroxyl radicals) balance out, but that H_2 gets progressively more enriched in deuterium. Indeed, for the stratospheric CARIBIC samples, m_{H2} does not vary with height above the tropopause, but δD-H_2 increases with height. A compact correlation is found between δD-H_2 and CH_4 mixing ratios; as the air is photochemically processed, the H_2 becomes more enriched as the CH_4 is destroyed. The found slope of this relation is very similar to those found in previous experiments, but the number of measurements is now increased threefold.

Samples from three flights to India during the summer monsoon season have also been analyzed. *Schuck et al.* [2010] found increased levels of CH_4 here, due to larger upward convection of boundary layer air, and increased microbial activity. Remarkably, in the samples with enhanced CH_4 the H_2 is depleted, and δD-H_2 and CH_4 are correlated. The deuterium depletion of H_2 in the 'Monsoon plume' is likely also caused by the increased upward convection, which brings deuterium-depleted hydrogen from the boundary layer to cruising altitude. The absence of a clear increase in H_2 concentration indicates that the source must be isotopically very depleted, maybe even more than what is expected from combustion sources. A possible explanation is that we observe increased microbial production of H_2 during the summer monsoon season, as biological production is the most depleted source of H_2.

References

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http://www.caribic-atmospheric.com