Geophysical Research Abstracts
Vol. 14, EGU2012-1887, 2012
EGU General Assembly 2012

The stable isotopic composition of molecular hydrogen in the tropopause region probed by the CARIBIC aircraft

A.M. Batenburg (1), T.J. Schuck (2), A.K. Baker (2), A. Zahn (3), C.A.M. Brenninkmeijer (2), and T. Röckmann (1)

(1) Utrecht University (UU), Institute for Marine and Atmospheric Research (IMAU), Physics & Astronomy, Utrecht, Netherlands (annekebatenburg@gmail.com), (2) Max Planck Institute for Chemistry, Air Chemistry Division, Mainz, Germany, (3) Karlsruhe Institute of Technology, Institute for Meteorology and Climate Research - Atmospheric Trace Gases and Remote Sensing, Karlsruhe, Germany

Atmospheric molecular hydrogen (H_2) has been little studied for some time, but has recently drawn more attention due to its expected future use as an energy carrier. Concerns have been raised that this use may lead to large-scale leakage of H_2 into the atmosphere, with implications for the atmosphere's oxidative capacity and stratospheric ozone chemistry. A thorough understanding of the global H_2 cycle is therefore needed, but at present, the uncertainties are still large.
Studying the stable isotopic composition of H_2 (δD(H_2)) is a promising way to gain more information about the H_2 cycle. Over the last decade, studies of the isotope effects in H_2 source and sink processes have appeared, δD(H_2) has been incorporated into global chemical transport models and many more environmental observations of δD(H_2) have been published. However, some knowledge gaps can be easily identified. Stratosphere-Troposphere Exchange (STE) has a strong influence on tropospheric δD(H_2), but very few δD(H_2) data are available from samples taken around the tropopause, where this exchange takes place. For large regions of the globe, no δD(H_2) data have been published.
In the CARIBIC project, air samples are collected in the Upper Troposphere-Lower Stratosphere (UTLS) region with a commercial passenger aircraft and routinely analysed for various gases. This sampling platform can potentially provide global coverage. More than 450 CARIBIC samples have been analysed for H_2 mixing ratios (m(H_2)) and δD(H_2).
More than 120 of these samples consisted of lowermost stratosphere (LMS) air. They show the lack of variation in m(H_2) and the δD(H_2) increase that is typical for the stratosphere, caused by the competing and deuterium-enriching source and sink processes of H_2 in the stratosphere. The deuterium-enrichment signal grows stronger with distance above the tropopause. As a result of the relatively long lifetimes of H_2, CH_4 and N_2O, strong negative correlations appear between δD(H_2) and m(CH_4) and between δD(H_2) and m(N_2O). These are similar to previously published results obtained from stratospheric balloon campaigns. The similarity between different campaigns indicates that these correlations likely hold globally and can be used for parameterizing the δD value of H_2 that is imported to the troposphere by STE.
This dataset also contains the first δD(H_2) data collected in or over India. A marked decrease in δD(H_2) is observed in the summer monsoon season, which correlates with the monsoon increase in m(CH_4), but is not reflected in a change in m(H_2). The correlation with m(CH_4) and the lack of change in m(H_2) lead to the hypothesis that the δD(H_2)-lowering is at least partly caused by the microbial production of H_2, which has a very deuterium-depleted source signature.