Geophysical Research Abstracts
Vol. 19, EGU2017-8503, 2017
EGU General Assembly 2017

Observations of molecular hydrogen (H_2) mixing ratio and stable isotopic composition at the Cabauw tall tower; very depleted source signature suggests microbial H_2 production in Dutch pasture soil.

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Molecular hydrogen (H_2), though not toxic or a greenhouse gas itself, may influence air quality and climate indirectly by affecting the atmosphere's oxidative capacity. So as increased use of hydrogen fuel is expected, a better understanding of the global, regional and local atmospheric H_2 cycles is needed. Studying the stable isotopic composition of H_2 ($\delta D(H_2)$) is a promising way to achieve this. Since the start of this century, the isotope effects in H_2 source and sink processes have been estimated, $\delta D(H_2)$ has been incorporated into chemical transport models, and larger sets of environmental observations of $\delta D(H_2)$ have appeared. The latter, however, were mostly obtained from samples collected in remote regions of the atmosphere, which is not sufficient to fully characterize the H_2 cycle or to assess the possible environmental effects of H_2 leakage in urbanized regions. To address this gap, flask samples were collected at the Cabauw tall tower at the CESAR site in the Netherlands. The air was sampled from inlets at 20, 60, 120, and 200 meter altitude for the analysis of H_2 mixing ratio ($\chi(H_2)$) and $\delta D(H_2)$. More than 250 samples were collected and analysed over a period of four years.

The H_2 mixing ratios in the samples show frequent excursions to high values above the background. Previously published continuous $\chi(H_2)$ observations at Cabauw and other (sub)urban sites showed a similar pattern. With the isotope observations, we can now see that these high $\chi(H_2)$ excursions are accompanied by very low $\delta D(H_2)$ values; probably at least partly a result of anthropogenic emissions of deuterium(D)-depleted H_2.

However, with a simple "Keeling plot" analysis, we obtained an apparent source signature (-515 $\pm$ 26 ‰) that was much below the range of published values for H_2 emissions from the combustion of fossil fuels. Since the result of the fit depended markedly on the quality selection of the samples that were included, we applied a bootstrap method to this fit to obtain a realistic picture of the uncertainty of the result. This showed a wide distribution with more than 99 % of the values below -400 ‰, suggesting that the H_2 cycle at Cabauw is under the influence of a source mix that is much more D-depleted than currently accepted values for fossil fuel combustion. Since microbial production of very D-depleted H_2 has been observed previously at Cabauw, we consider it likely that this contributes to the low apparent source signature.

A comparison of the samples from different sampling heights shows that there is a significant shift to lower $\delta D(H_2)$ values at the lower sampling levels. This shows that the uptake of H_2 by the soil, which preferentially removes "light" H_2, is relatively weak at the site. It also points again to local to regional microbial production of H_2, and possibly to differences between national vehicle fleets.