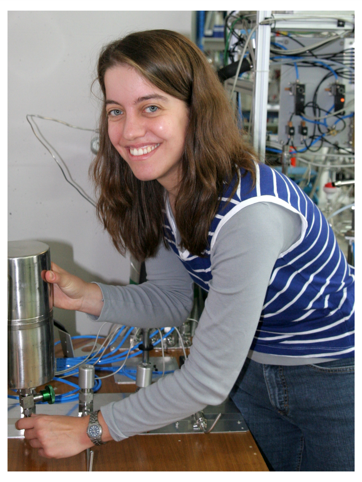

Temporal and spatial variability of δD(H_2) from six EUROHYDROS stations

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Worldwide χ(H_2) and δD(H_2) observations

Present atmospheric molecular hydrogen (H_2) mixing ratios (χ(H_2)) are around ~0.5 ppm. In the coming decades, H_2 levels are expected to rise due to use of hydrogen as an energy carrier. This may affect greenhouse gas lifetimes and stratospheric ozone depletion. Unfortunately, large uncertainties still exist in the global H_2 budget. The different sources and sinks of H_2 have very distinct isotopic signatures and fractionation coefficients, respectively. Therefore, measurements of isotopic composition (δD(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. Weekly to monthly air samples from six locations in the EUROHYDROS network have been analysed for δD(H_2) with a GC-IRMS system. The time series thus obtained now stretch over at least a year for all stations. This is the largest set of ground station observations of δD(H_2) so far.

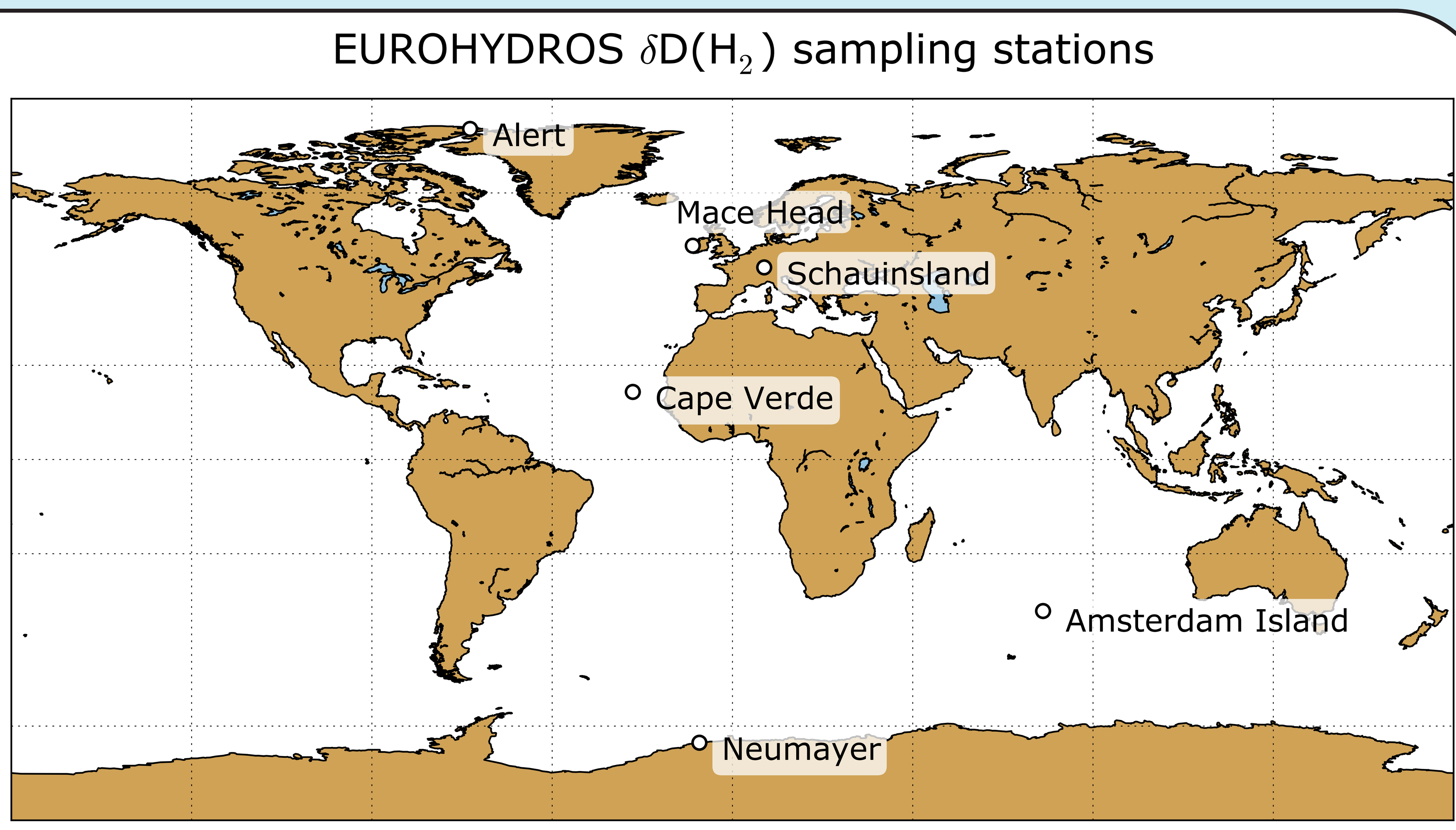
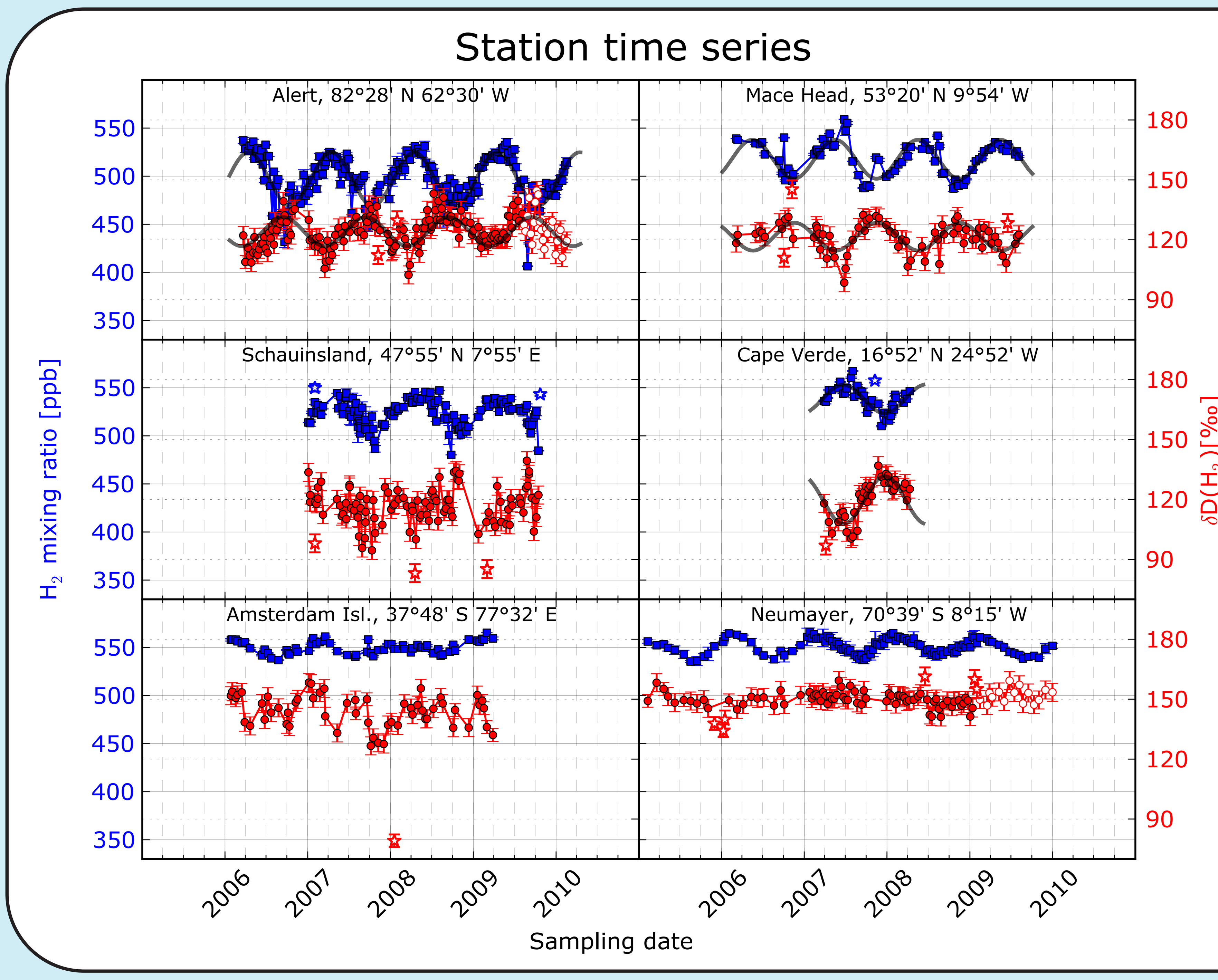


Fig 1: Locations of the EUROHYDROS flask sampling stations discussed here.



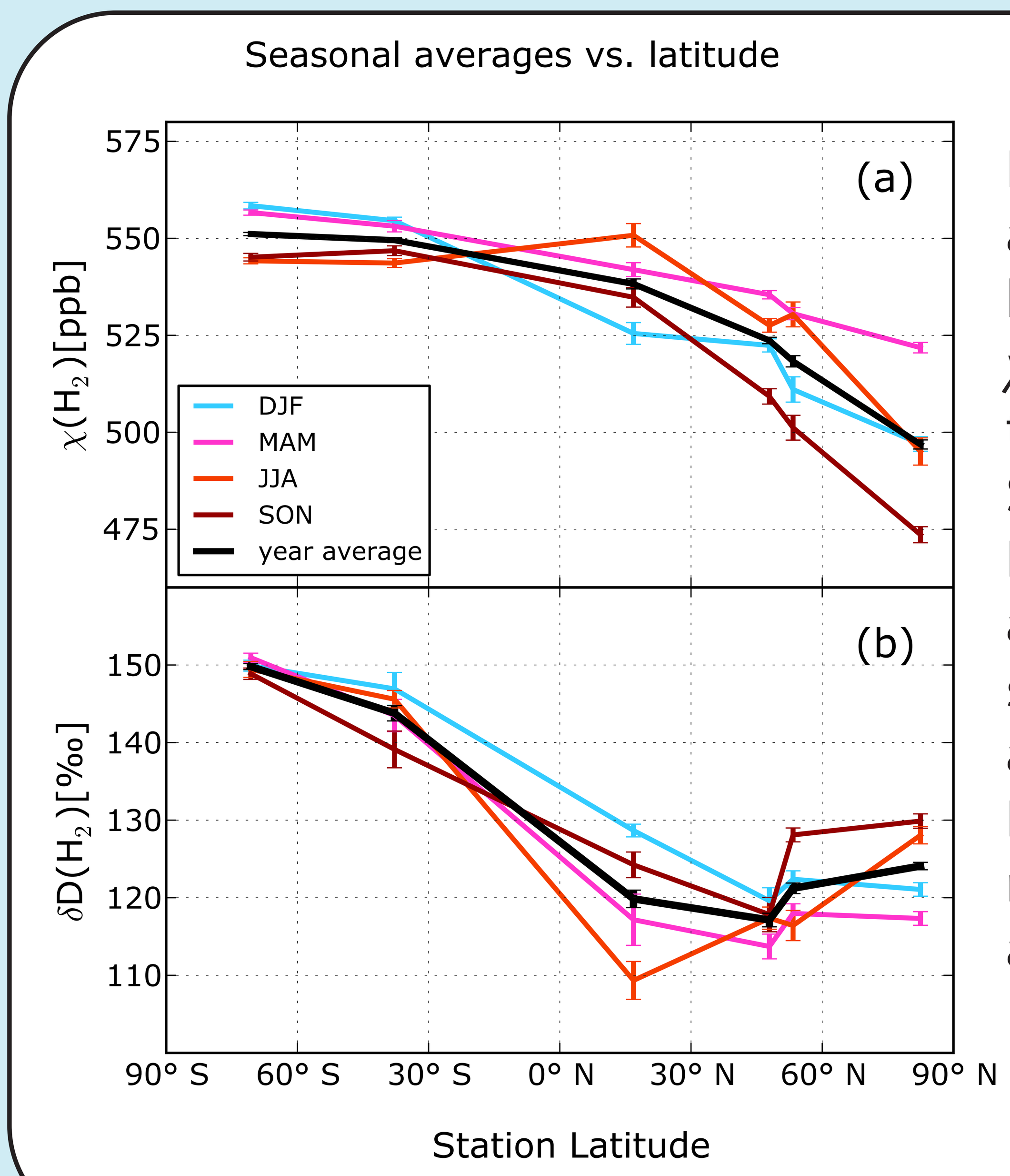
Time series

Fig. 2 shows the time series of χ(H_2) and δD(H_2) for the different stations. These data clearly show that the χ(H_2) and δD(H_2) variability is much larger in the Northern Hemisphere (NH) than in the Southern Hemisphere (SH).

The NH stations **Alert**, **Mace Head** and **Cape Verde** show clear cycles in both χ(H_2) and δD(H_2) that are 5-6 months out-of-phase. This phase difference is due to accumulation of H_2 from D-depleted (combustion) sources in winter and strong sinks (soil uptake and OH oxidation) in summer that preferentially remove the light hydrogen.

For **Schauinsland** and **Neumayer**, cycles are observed in χ(H_2) but not in δD(H_2). There is large scatter in the Schauinsland δD(H_2) data, possibly due to the continental location close to source regions. The **Amsterdam Island** χ(H_2) cycle is weak and no cycle is observed in δD(H_2).

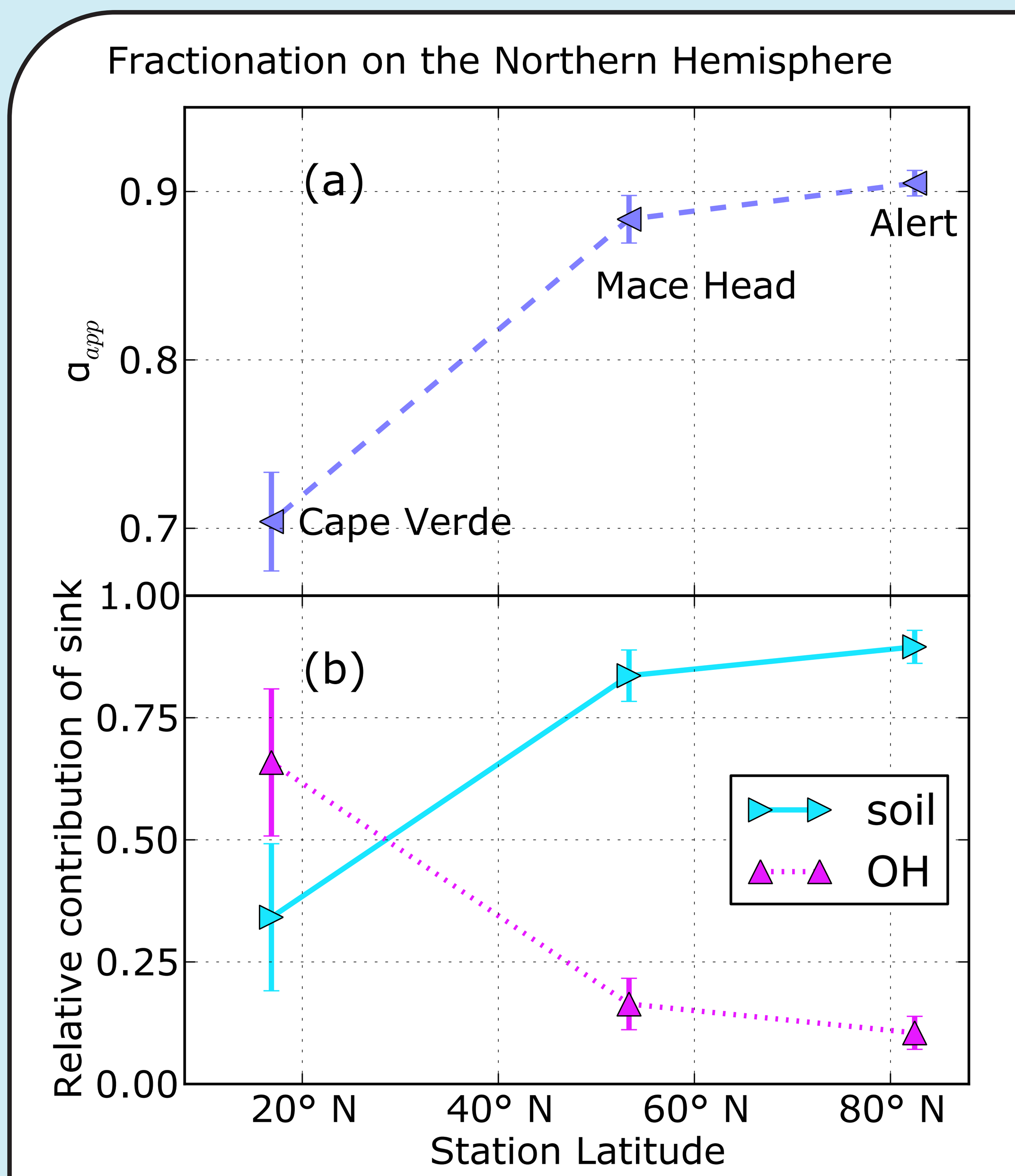
Fig 2: χ(H_2) (blue squares, by UHEI-IUP, LSCE and MPI-BGC) and δD(H_2) (red circles, by IMAU) measured on samples from the six stations. Solid lines represent harmonic best fits, error bars represent one standard error, open circles represent data that were affected by a system bias, open stars represent other outliers (some in Amsterdam Island are off the scale). Data denoted with open symbols are not used in the calculations.



Latitude gradient

In Fig. 3, the seasonal averages are plotted against station latitude. In all seasons, both χ(H_2) and δD(H_2) are higher in the SH than in the NH. Surprisingly, the minimum in δD(H_2) is not found in Alert, but at one of the lower-latitude stations. As H_2 from anthropogenic sources is D-depleted, this minimum may be a result of anthropogenic influence.

Fig 3: Seasonal averages of χ(H_2) and δD(H_2), plotted against station latitude. Error bars indicate one standard deviation



Sinks

If a seasonal cycle is assumed to be driven mainly by sinks, an apparent fractionation factor (α_{app}) can be calculated from a Rayleigh fractionation plot. Good fits were obtained for three stations only (Fig. 4(a)). From α_{app}, the relative contribution of the two H_2 sinks can be estimated (Fig. 4(b)). This shows that the relative importance of the uptake by soil increases with latitude (i.e. with larger land mass and lower OH levels).

Fig 4: (a) Apparent fractionation factors (α_{app}) for the NH plotted against station latitude. (b) Relative contribution of soil uptake and OH oxidation to the total sinks, assuming α_{app} is a mass-weighted average of α's of the two sinks.

Conclusions/Outlook

These regular observations of χ(H_2) and δD(H_2) provide insight into the seasonal and latitudinal distribution of H_2 and its isotopic composition. Tentative conclusions can be drawn about the geographical variations in its sources and sinks. These data have been used with the global chemical transport model TM5 and help to distinguish between different source/sink scenarios.

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Read more

These data were published in

- A. M. Batenburg et al., Temporal and spatial variability of the stable isotopic composition of atmospheric molecular hydrogen, ACP, 11, 6985-6999, 2011

TM5 model results were published in

- G. Pieterse et al., Global modelling of H_2 mixing ratios and isotopic compositions with the TM5 model, ACP, 11, 7001-7026, 2011