

Interactive
Comment

Interactive comment on “Isotopic signatures of production and uptake of H₂ by soil” by Q. Chen et al.

Q. Chen et al.

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We thank the editor and reviewers for their valuable comments. The analytical solution for the flask sampling processes uploaded by the editor as a supplement is very well written and it would be useful for sampling involving large volume sample collection from the chamber. Our evaluations have been double-checked with this analytical solution. Reviewer 1 did not request a change to the manuscript and our response to Reviewer 2 is shown below.

Technical comment: The term “mixing ratio” has been exchanged with the term “mole fraction” in the text. However, the axes of the graphs in Fig. 6 still use “mixing

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ratio". This may be confusing (P.23479, L.8).

Response: We will change all the "mixing ratio" into "mole fraction" for the axes of the graphs in Fig. 6.

Specific comment: The δD of the emitted H_2 was negative but somewhat larger than the value expected from isotopic equilibrium between H_2 and water (about -700 permil). The authors argue that this may be a characteristic of the nitrogenase reaction (assuming that H_2 was produced by the clover nodules at these field sites). They also discuss the possibility that deuterium was enriched when the freshly produced H_2 was partially consumed in the soil. I think that this may be the most parsimonious explanation. The authors argue that a large fraction of H_2 needs to be removed in the soil to explain the enrichment of D. I suggest calculating and reporting how large exactly this fraction would need to be.

Response: The estimate of the fraction of H_2 removal within the soil before entering atmosphere (f_{in}) can be very uncertain due to the lack of information about the initial δD of H_2 produced biogenically during N_2 fixation (δD_0) and the fractionation constant during uptake within the soil (α_{in}). f_{in} can be calculated as following.

Assuming initially produced H_2 by N_2 fixers will be consumed within the soil before entering atmosphere and the fractionation constant is α_{in} . Then the Rayleigh equation yields: $(c/c_0)^{\alpha_{in}-1} = (\delta D + 1)/(\delta D_0 + 1)$

where c and c_0 are the remaining and initial H_2 mole fractions, δD and δD_0 are for the remaining and initial H_2 . The fraction of H_2 removed within the soil before entering atmosphere would be $f_{in} = 1 - c/c_0$. It should be noted that both δD_0 and α_{in} are unknown. δD ($=\delta D_{soil}$) is the isotopic signature of H_2 emitted from the soil, which is measured in our experiments and shown in the manuscript.

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By assuming $\alpha_{in}=0.945$ (overall fractionation factor as determined in our deposition experiments), $\delta D=-530\text{ ‰}$ (averaged δD_{soil} of Cabauw net-emission experiments) and $\delta D_0=-611\text{ ‰}$ (averaged of $\delta D(H_2)$ derived from laboratory experiments in Luo et al. (1991) and Walter et al. (2012), see Section 4.4 in manuscript), we calculate $f_{in} = 1 - c/c_0 = 0.97$. Thus, 97% of H_2 produced by N_2 fixation is removed within soil before entering atmosphere. This is higher than the estimate from Conrad and Seiler (1979), which was from 30% to 90%. By using the lower limit of α_{in} in our experiment (0.911) and the upper limit of δD_0 in Luo et al. (1991) and Walter et al. (2012) (-569 ‰), we calculate a lower limit of f_{in} to be 0.62. The upper limit of f_{in} is 1.00 when α_{in} approaches 1.

For these calculations we have used a δD of -530 ‰ but it varies from -629 ‰ to -451 ‰ in our experiments. In addition, both δD_0 and α_{in} are unknown. We want to mention that we had such calculations in but removed them because all of the parameters (δD_0 and α_{in}) are uncertain and we do not think that we can constrain the consumed fraction in a meaningful way (other than it has to be a lot). Thus, we suggest not reporting how large exactly the fraction of H_2 removed within soil.

Changes in the manuscript: No changes in the text. We will change all “mixing ratio” into “mole fraction” for the axes of the graphs in Fig. 6.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 23457, 2015.

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