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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A new estimation of the recent tropospheric molecular hydrogen budget using atmospheric observations and variational inversion" *by* C. Yver et al.

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We thank the reviewer for the useful and accurate comments. The comments are in italics and the answers in plain text. The comments about typo or legends will be taken into account for the revised version of the manuscript.

General comments: General aspects of the H_2 budget, the new data sets and the inversion system are well presented in the introduction and the first sections of the paper. The presentation of the deposition velocity maps (in 3.3 and 3.4) could profit form a more critical assessment (see specific comments).

We answer to this in the specific comments and we will detail the text in sections 3.3

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and 3.4.

The authors make use of different priors for the H_2 deposition velocity in order to investigate the sensitivity of the inversion results. However, a more systematic investigation of the robustness of the inversion results would be appreciated. The estimated H_2 budget seems to be in general agreement with previous studies.

The focus of the paper is on the soil uptake. We only test the sensitivity of the results to different spatio-temporal distributions of the soil uptake, the main H_2 sink. It is true that we do not test the sensitivity of our results to surface emissions or photochemical production. This is an important matter but beyond the scope of our focus here. We will rephrase some sentences in the abstract and introduction to highlight the fact that we concentrate on soil uptake and do not provide a full sensitivity study of our H_2 budget.

A detailed analysis is presented for Europe where most of the new continuous stations are located and hence the best constraint is expected. However, it remains unclear how robust this estimate is.

We produced national estimates for Western Europe. The assessment of the robustness of these results is not easy as no inventory exists and H_2 is not as well studied as CO_2 or CH_4. We compared the anthropogenic part of our S5 scenario with the independent estimates from IER, and the agreement is good.

The paper is well structured and well written. It presents a valuable contribution to a further assessment of the H2 budget. However, the authors do not present a comprehensive analysis of all uncertainties in the inversion and the robustness of the resulting H_2 budget components. This needs to be improved to make this paper ready for publication in ACP.

Specific comments:

p. 28968, l. 22-24:'At all sites. . .' The meaning of this sentence is unclear. Was the H_2 analyser installed to monitor other greenhouse gases?

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The H_2 analyser was installed in June 2006 at Gif-sur-Yvette to complete an analytical system which already measured CO_2, CH_4, N_2O and SF_6. That is why it was stated that most of the sampling began before this analyser was installed: other gases were already measured. This sentence will be erased as the analysis of others gases is of no interest in this paper.

p. 28971, l. 28: Is the soil uptake really strongest in autumn? Could you give a reference. In 4.2 you mention that the soil uptake measurements show the maximum end of August or beginning of September.

The soil uptake due to bacterial activity depends on soil moisture and temperature as detailed in Schmitt et al. Briefly, at the end of the summer (August, September), the soil is relatively dry and warm which ensures a high bacterial activity and a good diffusion of air in the soil. On the contrary, in spring, when the soil is cold and very humid, the bacterial activity and the diffusion are slowed. In the NHN, the atmospheric signal is minimum in autumn (defined as September, October and November), as the combination of the soil uptake (maximum in August-early September, and still significant in autumn) and of the rapidly decreasing photochemical production (after August). We will clarify the text on this point: 'In the HNH, the minimum is reached in the autumn when the soil uptake is still significant after its late summer maximum (see section 4.2), and the photochemical production is rapidly decreasing compared to the summer. The maximum occurs in the spring when the soil uptake is the weakest and the photochemical production is increasing fast.'

p. 28974, l. 7: What is meant by 'the equivalent of the observations'?

This sentence was unclear. We will rephrase it: 'H is the operator representing the chemistry-transport model and the simulated concentrations at the same time and location than the measurements.'

p. 28974, l. 25: What does it mean that the observation error matrix R is supposed to be diagonal? and filled with the standard deviation of the measurements? Please

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explain in more detail the assumptions concerning the observation, model and representativity errors in the inversion.

We agree that this sentence was too condensed. We will rephrase: 'The observation error matrix R should combine the measurement errors, the model errors (transport and chemistry) and the representation errors (e.g. only a large model cell to represent the fine space scales of the measurements). We neglect cross-correlation terms, which makes R diagonal. For the diagonal elements of R (variances), the variability of the double sampling measurement as proxy for the observation error, with a corresponding ceiling standard deviation of $\pm$5 ppb for H_2 and $\pm$1.2 ppt for MCF.'

p. 28975, l. 8-10: Is it reasonable to base the H_2 soil uptake (implicitly) on net primary production? Please comment.

Indeed, NPP is probably not the best proxy for H_2 uptake but as stated in the introduction, the uptake is still not well understood. At the time of the paper of Hauglustaine et al (2002), the measurements were very sparse. The authors have then decided to use a proxy instead of a parametrization. We use Hauglustaine et Ehalt as a first scenario but we present different alternative scenarios for soil uptake in section 3.3 and discuss them in the results. We will add two sentences : 'The H_2 prior emissions and monthly deposition velocity maps are taken from Hauglustaine and Ehhalt (2002) and constitute scenario S0.' and 'This leads to deposition velocities between zero and 0.1 cm s-1. Alternative scenarios for soils uptake are presented in section 3.3.'

p. 28976, l. 18-25: Please explain in more detail how H_2 soil uptake is estimated in LPJ and what the main assumptions/processes are? What is the relation between H_2 soil uptake and vegetation? Does the model simulate deposition velocity or H_2 soil uptake?

The H_2 soil uptake is estimated based on the assumption that it is mainly driven by molecular diffusion. The uptake is then expressed using Fick's law and depends on the mixing ratio at the surface, the diffusivity of H_2 in the soil and the oxidation constant rate. The diffusivity in the soil itself depends on the soil porosity and temperature whereas

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the oxidation rate depends on soil temperature, moisture and organic content. This submodel is integrated into the Dynamic Global Vegetation Model LPJ, LPJ-WHyMe. The soil properties are based on the Food and Agriculture Organization (FAO) data set overlain by soil organic carbon data from the IGBP-DIS data set [Global Soil Data Task Group, 2000]. Soil temperature, moisture and H_2 mixing ratio are given by LPJ-WHyMe. Zero values are applied when the snow layer is thicker than 50 cm or when the NPP is lower than 10 gC m^{-2} (Morfopoulos et al., 2010).

p. 28976, l. 26-29: What is the OSLO-CTM used for?

We will clarify this point in the text : 'For S4, the monthly map was produced by the Oslo CTM2, an eulerian chemical transport model (Søvde et al., 2008), which combined soil deposition velocities estimated within the European project EUROHYDROS with bottom-up and top-down methods compiled in Schillert (2010).'

In EUROHYDROS, H_2 deposition velocity was provided form the chamber measurements and in a first attempt all measurements were combined to construct a mean seasonal cycle for the latitude bands north of 30N and south of 30S and a constant value was proposed for 30S-30N. Reduced deposition velocities were suggested for wetlands and deserts. This is obviously not included in the map for S4. Why not?

The Oslo CTM2 coupled the ECMWF IFS meteorological data and the MODIS annual L3 global 0.05 Deg landcover map (REF), to Eurohydros deposition velocities to take into account the latitudinal repartition but also the effect of snow and wetlands. Reduced velocity in deserts is visible in the map but they are only set to zero if the desert fraction is 100%, which is not always the case in T42 resolution. In the MODIS dataset, permanent wetlands are also taken into account. However, the fraction of wetlands in T42 resolution (2.8x2.8 degrees) is very small. The maximum in a grid-cell is 19% (in Siberia). Otherwise, the values are less than 10%. Hence, the wetlands do not visibly contribute to reducing deposition velocities on the map of S4.

p. 20977, l. 25-28: Are these 'hotspots' realistic or just an artifact produced by the use

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of NPP? Are there any reasons for them? Please comment.

S0 presents important spatio-temporal variations with marked hotspots. These high values can be explained by meteorological conditions which drive a high NPP and then a high H_2 deposition velocity. In the Southern Hemisphere, these hotspots reach 0.1 cm s-1 while in the Northern Hemisphere, they reach up to 0.14 cm s-1. In Lallo et al. (2008), the highest values found in boreal forest was 0.07 cm s-1 which is about two times lower than the values here. These high deposition velocities are to be considered with caution as possible artifacts of the use of NPP as a proxy of H_2 deposition velocity. We will add the sentence : 'In summer, hotspots are observed mostly in North America and in the north of Russia. These high values are due to the direct link existing between NPP and deposition velocities in the assumptions of scenario S0: High NPP produced by favorable meteorological conditions may lead to too high deposition velocities.'

p. 28978, l. 20: Were all mixing ratios scaled or just the initial conditions? This needs to be stated more clearly.

The initial conditions, the mixing ratios and the deposition velocities are scaled. We will clarify the text: 'For the other scenarios, the initial mixing ratios have been scaled and the prior fluxes have been updated...'

p. 28979, l. 8-10: You find a decrease at a southern hemisphere background site although you expect that the H_2 sink is too weak in S0. Please comment.

The soil uptake is supposed to be too weak in the Northern Hemisphere during winter and spring. Hence, the decrease observed in a southern site is not correlated to this assumption.

p. 28980, l. 23: Could you speculate what other causes? Regional distribution of dep. vel.?

Other causes could indeed be a different regional distribution of the deposition velocity as it depends on the non homogeneous atmospheric H_2 . As stated before, in S0, the

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deposition velocities are inferred from the net primary production which could be a non-suitable proxy. Emissions in S0 might also be too large. We will add a sentence: '...has other causes. Errors in the regional distribution of deposition velocities or in emission intensities are possible explanations for such an offset.'

p. 28981, l. 23-28: A more extensive estimation and discussion of the uncertainties associated with the inversion is needed here.

We will rewrite this part: 'Estimating the uncertainties of the posterior fluxes can be done using the Monte-Carlo approach of Chevallier et al. (2007). However, due the large computational expense of this method, a simpler approach was preferred. The one-sigma uncertainties are estimated from the spread of the fluxes in scenarios S1 to S4 compared to reference scenario S5. We do not include S0 because, in this scenario, the prior HCHO flux is 5 Tg yr^{-1} lower than the prior flux in the other scenarios and, as explained previously, prescribed with small uncertainties. Moreover, the uncertainties of Table 4 do not include all sources of uncertainties. For instance, they do not account explicitly for transport model errors, for chemistry model errors, or for uncertainties in the inversion setup other than the distribution of deposition velocities. They should therefore be considered as lower estimates. Performing an analysis of the full uncertainties associated to values in Table 4 is an important and complex matter which lays beyond the scope of the work.'

p. 28983, l. 12: Are you sure that you have sufficient constraints?

We will rephrase this sentence: 'In this study, Europe contains the largest number of observation sites, therefore being the best constrained area of the world.'

p. 28983, l. 15-16: Why are the fluxes interpolated to a higher resolution? When only looking at the maps this simulates an unrealistically high resolution of the fluxes.

The sentence 'To observe the difference better, the data are interpolated on a higher resolution grid (1x 1).' is poorly formulated. The intention of the authors was to transfer

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the data from the global model onto a map zoomed on Europe. The higher resolution does not change the general pattern of the map but to be more consistent, the map will be modified to have the same resolution as the model output.

p. 28985, l. 10: I cannot find the comparison with flux measurements?

The comparison with flux measurement is shortly discussed in section 4.2. The authors did not compare values but the timing of the maximum and minimum of the soil uptake as well as the biomass burning maximum emissions.

Fig. 6: Explain the grey shaded area. What is S5 fwd? The prior flux for S5? The meaning of the two dotted green lines in the Emissions plots are not unambiguous.

The grey shaded area represents the spread between the different scenarios. S5 fwd is indeed the prior fluxes for S5. We will add these two sentences in the legend: 'The grey shaded area represents the spread between the different scenarios. The prior emissions for the S5 scenario are plotted in light green and labelled S5fwd.' Due to an error in the plot, there are two light green dotted line instead of a plain one for the total emissions and a dotted one for the biomass burning emissions. We will correct this in the revised version.

Fig. 8: The white color in the soil uptake maps is not part of the color bar? What does it stand for?

The white color corresponds to missing values. We will add it in the legend.

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