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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 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. More references will be provided as well as the requested clarifications. Most of the other comments answered here will also be answered to in the revised version.

General Comments: This paper addresses an interesting question on the global budget of H_2, especially for its soil uptake which has the greatest magnitude and uncertainty. This reviewer is not familiar with the variational inversion system, but inverting so many

processes (soil uptake, emissions from fossil fuel combustion, biomass burning, and N_2 fixation, and photochemical production of HCHO) in each model grid cell seems challenging, considering the sparseness of the observations over regions outside Europe. The impact of the spatial coverage and resolution of the observations on the variational inversion results at model grid cell resolution needs to be discussed. Only the posterior flux maps for Europe are shown because of the relatively sufficient constraints for this region. How about the global posterior flux maps? How did the spatial distributions of the fluxes change between posterior and prior globally?

We decided to separately invert the different sources and sinks, at the model resolution, in order to limit the aggregation error (Kaminski et al., 2000; if grouped before inversion, an error in the spatio-temporal distribution of H_2 flux cannot be corrected properly by the inversion). Bousquet et al. (2011) showed that the 2000s flask network for H_2 alone can constrain neither the different components of the H_2 cycle, nor the regional fluxes. Therefore, here, we only present the results for large latitudinal bands. However, as the density of observations in time and space is much higher in Europe than the network used in Bousquet et al (2011), we specifically discuss the European sources and sinks. This text will be added in the paper: 'Bousquet et al. (2011) showed that the 2000s flask network for H_2 alone could constrain neither the different component of the H_2 cycle, nor the regional fluxes. Nevertheless, we invert separately the different sources and sinks, at model resolution, in order to limit the so called aggregation error (Kaminski et al., 2000); if grouped before inversion, an error in the spatio-temporal distribution of H_2 flux cannot be corrected properly by the inversion. Performing an analysis of the full uncertainties associated to every term of the budget laid however beyond the scope of this study. Therefore, here, we only present the results for large latitudinal bands with a focus on soil uptake. As the density in time and space is much higher in Europe than in Bousquet et al. (2011) for the late 2000s, we also discuss specifically the European sources and sinks.' It is also important to keep in mind that observations are not the only information pieces used by the inversion. The prior information on the fluxes which are globally relatively well constrained plays a major role. Performing the inversion is

indeed challenging and that is why the photochemical production was not optimised in this study, but only the deposition velocity and the emissions. The robustness of the inversion can be estimated by comparing the different scenarios.

Specific Comments: P. 28969: For both RAMCES and EUROHYDROS networks, did you treat sites sampling clean or polluted air masses differently? How?

Data from continuous stations were daily averaged. Data above 800ppb were filtered. This is the only filter applied to the data. This will be added in the text: 'For the continuous stations, the daily means are plotted and mixing ratios above 800 ppb, which correspond to strong local pollution events, are excluded. No other filters are applied to the data.'

p.28971, l.14-20: How about the seasonal cycle in the Southern Tropics?

The authors do not comment on the seasonal cycle in the Southern Tropics as there is no EUROHYDROS atmospheric site available there for 2006-2009.

p. 28971, l. 28: Please explain (or cite from literature) why the soil uptake is strongest in autumn in the HNH.

The soil uptake due to bacterial activity depends on soil moisture and temperature as detailed in Schmitt et al. (2010). 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. 28971: What is the time resolution of the observations used in the variational inversion?

We assimilate daily-mean observations for the continuous data and flask observations when they are sampled (typically every one or two weeks).

p. 28974, l. 9: What measurements are used to constrain the HCHO flux? Is HCHO production in each cell in the 3-D space inverted?

In this work, there is no observation to directly constrain the HCHO flux or the HCHO production. They are prescribed in the inversion with a small uncertainty and therefore stay close to their prior value. Nevertheless, for the scenarios after S0, we have prescribed the optimised HCHO 3D production (given by Bousquet et al. (2011)). This 3D production has been constrained with satellite data from OMI for several 3D large regions (one scaling factor per 3D region and per year). The sites in our study are indeed mostly surface sites but there are however several high altitude, tall tower or flight sites.

If you only use the surface measurements of H_2, are they sufficient to constrain so many variables in the state vector including soil uptake, emissions related to biomass burning, fossil fuel combustion, and N_2 fixation, and HCHO production in each grid cell? This needs to be discussed. See answer to the general comment.

p. 28974, l. 9: Why eight-day frequency of the state vector?

The eight-day frequency of the state vector is chosen as a compromise between a high temporal resolution and a reasonable computational time. At this frequency, to invert the data with a reduction of the gradient of 95%, around 20 iterations are necessary, corresponding to two to three weeks real-time. Moreover, with too high a frequency, we would need to assign temporal correlations, that are neglected here. Lastly, as stated

before, the observations are either at a daily frequency for the continuous stations or at a bimonthly frequency for the flask sites. The consistency between the time resolution for inverted fluxes and the time resolution of the observations is still a matter of debate in the atmospheric inversion community. We chose 8 days as a compromise between the time resolution of flasks and of daily means calculated from continuous observations.

p. 28974, l. 25: The diagonal elements of R should be the squares of the standard deviations (variances) of the measurements.

We agree that the diagonal of R is filled with variances, that are squares of some standard deviations. However, as detailed for rewiever 1, these variances should reflect measurement errors, model errors and representation errors, and not only measurement errors. This paragraph will be updated: '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), we use 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.21-22: Is soil uptake or vdep in the state vector? Since the soil uptake rate also depends on the mixing ratio of H_2, is the H_2 field also optimized in the framework and how?

The lack of clarity between soil uptake and deposition velocity will be corrected in the revised version. It is the deposition velocity that is added in the state vector, inverted and optimised. At each time step, the soil uptake is calculated as the product of the deposition velocity and the H_2 mixing ratio. This flux, as the production, loss and emissions of H_2, is inverted leading to a optimised deposition velocity and an optimised mixing ratio.

p. 28975, l. 21-27: Resolving the soil uptake, fossil fuel emissions, biomass burning emissions within each model grid seems difficult, given the sparseness of the observations. As you mentioned, different processes cannot be completely independently inverted. Are the inversion results robust regarding different settings of scenarios and prior conditions?

We did not test the robustness to all causes of uncertainties (transport model, chemistry model, inversion setup for other fluxes than the soil uptake). In this paper we focus on the sensitivity of the soil uptake to its prior spatio-temporal distribution. To do so, the inversion has been evaluated using various scenarios with different distribution of the soil uptake in particular. The authors have chosen to estimate the residual uncertainties as the maximum spread of the scenarios compared to the reference scenario S5. This is only part of the full uncertainty which estimation, although interesting, is far beyond the scope of this work. Clarification about what the provided uncertainties cover will be given in the text: '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 cost 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 the values in Table 4 is an important and complex matter which lays beyond the scope of this work.'

p. 28976, l. 14: how is the value 1.28 derived?

The value 1.28 is derived as explained in the following sentence, which will be added in

the revised paper: 'The first-guess modelling leads to a strong offset with a simulated mean mixing ratio 115 ppb higher than observed. Hauglustaine and Ehhalt (2002) attributed this mismatch between model and data to the underestimation of the soil sink in the northern hemisphere during winter and spring. Moreover, using the same scenario, Pison et al. (2009) found an urrealistic accumulation of H_2 in the atmosphere attributed partly to the same cause. The deposition velocity map has been scaled by a ratio of 1.28 to take into account the hypothesised underestimation and produce a better balanced budget assuming that the other terms (production, emission and OH loss) are known and fixed.'

p. 28977, last paragraph: Please comment on the hotspots in S0.

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. 28980, l. 1-2: It seems that the difference in photochemical production between S0 and other scenarios is greater than 5 Tg / yr.

If one looks at figure 7, the difference between S0 and the other scenarios is indeed of 5Tg yr^{-1}.

p. 28980, l. 22-23: Please comment on other causes.

Other causes could indeed be a different regional repartition of the deposition velocity as it depends on the non homogeneous atmospheric H_2 . As stated before, in S0, the 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. 28983, l. 26-27: Please comment on the reason for the smaller spring soil uptake than the autumn uptake.

The smaller spring soil uptake than the autumn uptake is explained in answer to comment p. 28971, l. 28

p. 28984, last paragraph: It would be helpful to find a way to add those percentage contribution values to Table 5.

The authors have chosen not to add these uncertainties to IER estimates because they are given for another inventory (EDGAR).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 28963, 2010.