

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

### **Anonymous Referee #1**

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#### General comments:

The paper presents an estimation of the molecular hydrogen (H<sub>2</sub>) budget using a variational inversion system. The uncertainties of the H<sub>2</sub> budget are still large. In particular soil uptake, the major sink of H<sub>2</sub>, is still not well constraint. Therefore the paper is a useful contribution to further constrain the H<sub>2</sub> budget based on a combination of atmospheric mixing ratio measurements, information from sparse direct soil uptake measurements and emission inventories in combination with chemistry-transport modeling. The study is a direct follow-up of the paper by Bousquet et al. (2010) in which a synthesis inversion was used and the paper by Pison et al. (2009) which makes use

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of the same variational inversion system (PYVAR/LMDz-SACS). The authors of the present study introduced new developments in PYVAR/LMDz-SACS to further adapt the system for the inversion of H<sub>2</sub> and based their inversion of a specific new data set of continuous atmospheric H<sub>2</sub> mixing ratio measurements.

General aspects of the H<sub>2</sub> 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 from a more critical assessment (see specific comments). The authors make use of different priors for the H<sub>2</sub> 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<sub>2</sub> budget seems to be in general agreement with previous studies. 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.

The paper is well structured and well written. It presents a valuable contribution to a further assessment of the H<sub>2</sub> budget. However, the authors do not present a comprehensive analysis of all uncertainties in the inversion and the robustness of the resulting H<sub>2</sub> budget components. This needs to be improved to make this paper ready for publication in ACP.

Specific comments:

- p. 28966, l. 9: Does 6-11 Tg yr<sup>-1</sup> refer to the nitrogen fixation or to the complete sources? Reconsider the positioning of these numbers in the sentence.
- p. 28968, l. 5: The multi-species inversion of CH<sub>4</sub>, CO and H<sub>2</sub> by Pison et al. (2009) should also be listed here, even though the results for H<sub>2</sub> were rather inconclusive.
- p. 28968, l. 22-24: 'At all sites. . .' The meaning of this sentence is unclear. Was the H<sub>2</sub>

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analyser installed to monitor other greenhouse gases?

p. 28969, l. 17: Who measures at Bialystok, except MPI-BGC? Not obvious from Tables 1 and 2.

p. 28969, l. 19: Is Tver really a suburb of Moscow? The large 'scatter' in Fig. 2 at TVR, GRI and ORI. is obviously due to the fact that vertical profiles are plotted. This should at least be mentioned in the text or in the legend.

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.

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

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

p. 28975, l. 8-10: Is it reasonable to base the H<sub>2</sub> soil uptake (implicitly) on net primary production? Please comment.

p. 28976, l. 18-25: Please explain in more detail how H<sub>2</sub> soil uptake is estimated in LPJ and what the main assumptions/processes are? What is the relation between H<sub>2</sub> soil uptake and vegetation? Does the model simulate deposition velocity or H<sub>2</sub> soil uptake?

p. 28976, l. 26-29: What is the OSLO-CTM used for? In EUROHYDROS H<sub>2</sub> deposition velocity was provided from 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?

p. 28977, l. 16: Refer to the dep. velocity map for S4 as Eurohydros map net as 'Oslo map'.

p. 28977, l. 20: A 'yearly total deposition velocity' in  $\text{cm s}^{-1}$  is not a very common quantity. . . Better compare annual mean.

p. 20977, l. 25-28: Are these 'hotspots' realistic or just an artifact produced by the use of NPP? Are there any reasons for them? Please comment.

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

p. 28979, l. 2-4: Is this meant for all stations or only those with weak seasonal cycle? Please clarify.

p. 28979, l. 8-10: You find a decrease at a southern hemisphere background site although you expect that the H2 sink is too weak in S0. Please comment.

p. 28979, l. 13-14: Would be more interesting to have the correlation coefficients mentioned here directly instead of the vague statement of a 100% increase.

p. 28979 l. 23: It would be valuable information to include at least one of the prior fluxes (e.g. S5) in Fig.6 for comparison. This could already be the case but it is not explicitly mentioned and difficult to see in the figure.

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

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

p. 28982, l. 6-7: What is the maximum standard deviation of 15%? Standard deviation relative to what?

p. 28982, l. 8-10: Just state the absolute standard deviation and the relative std. dev.

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– whether it is adequate or not. . .

p. 28982, l. 14-15: Compare this directly to one of the recent studies, e.g. Ehhalt and Rohrer (2009)

p. 28983, l. 2-5: State more clearly that the comparison with Xiao and Bousquet is not fully consistent because regions are slightly different and the time period is different as well.

p. 28983, l. 2: Are you sure that you have sufficient constraints?

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.

p. 28984, l. 17: Is this really the correct name of the institute? Institut für Energiewirtschaft und Rationelle Energieanwendung (IER)

p. 28985, l. 3: What is meant by ‘the model’? the inversion?

p. 28985, l. 10: I cannot find the comparison with flux measurements? Table 3: Better refer to Eurohydros deposition velocity map instead of Oslo CTM.

Technical corrections:

p. 28970 l. 28: Grant et al. (2010) – there is only one reference. Change also in References.

p. 28977 l. 18: . . .right panel) . . .

p. 28979, l. 4: . . .fairly well represented. . .

p. 28980, l. 3: prior or a priori

p. 28980, l. 4: . . .with an error. . .

p. 28980, l. 8: . . .shifted to July. . . to August.

p. 28984, l. 6-7: Delete one ‘Table 5’

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p. 28984, l. 24: IER

p. 28988, l. 4: Ehhalt, D. H. and Rohrer, F. . . .

Table 4: 'mid 2006-mid 2009' only valid for the column 'This study'

Table 5: IER

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.

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

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