Comments to ‘Constraining the atmospheric hydrogen oxidation and soil sinks using HFC-152a’

Summary

This work looks to constrain the variation in the H2 budget by using 3 box models to represent northern, tropics and southern regions. They compare observations and a forward box model to a new Bayesian inversion model, which optimises OH, HFC-152a emissions, H2 emissions, H2 chemical production, and the H2 soil sink. HCF-152a observations are used alongside H2 observations to constrain the seasonality of OH, and therefore improve the OH estimate, which feeds into the H2 budget. Further to this, they use the optimised OH to estimate HCHO production, which in turn is used to calculate H2 chemical production. They run four scenarios with different uncertainty ranges for the H2 emission and chemical production. In the final uncertainty scenario, they estimate HCHO photolysis from the retrieved OH using a pseudo-linear relationship.

The authors found that the retrieved model performed better in the northern hemisphere and were able to create a seasonal variability of soil deposition which was in agreement with other GCMs with no prior assumptions for the soil sink seasonality. They found a larger uncertainty of soil deposition in the tropical region with reduced seasonality, and there was a minimal impact on soil deposition in southern region as expected due to limited land.

While they are unable to provide absolute values for H2 sources and sinks due to the high uncertainty in HFC-152a emissions, they are able to determine the seasonal cycle and amplitude of these processes, which can be used to compare against other interactive hydrogen schemes.

General Comments

This work is a novel approach to constraining OH by using both H2 and HFC-152a observations. The retrieved seasonality of the soil sink, particularly in the north box, is of great interest, as no physical assumptions of the soil uptake are made.

As the authors point out, they are not able to retrieve absolute values due to not being able to constrain absolute values of OH. As a result of this, I think the title of the paper could be misleading as this isn’t an absolute constraining of the hydrogen sinks, but rather their seasonality and amplitude.

I think some further analysis could be drawn out from these results. I’ve given some more details in the comments below, but

can the authors e.g. provide an intercomparison between boxes to establish percentage contributions of different sinks relative to other boxes? This would give a useful weight to how important sources/sinks of H2 are in different regions. Also, I think there could be more of a comparison with other literature, and authors could expand on what is meant when there results are in agreement with other studies.

It would help if the authors made more use of their figures and referenced them throughout the text for clarity (see technical comments)

Technical Comments

68 : Can you provide more information on the destruction of HFC-152a (e.g. a percentage estimate of OH destruction)

70-74 : It's difficult to follow why the authors cannot retrieve absolute values. They later state in line 117 that they scale the OH tropospheric concentration to 1e6 mole cm-3. Please can this sentence be rephrased/broken up to describe this clearer.

71 : Could you briefly explain the impact of HFC-152a emissions on constraining first order loss via OH (i.e. [HFC-152a] = Production / k[OH]) to make the reasoning clearer.

72 : In general, could you provide more information on HFC-152a? Is there expected to be a N-S hemisphere gradient given the short lifetime? What's the global surface average condition and is much of a seasonal variation?

Section 2.1 : It would be easier to follow if the authors introduced here they were running both a forward model and an inversion model (and that the latter is new) or made this more clear in lines 76-78.

83-4 : I know the refers to Chen et al. 2024 for more description on the box model, but I think the paper would greatly benefit from a more detail on the box model e.g. stating which species are included, how OH is parameterised in the forward model.

86 : What modifications were made to the box model boundaries?

89 : Authors say they use monthly temperatures and are running on a daily timestep. From S1, it looks like these are static values, but I think it would help to state this in the main text too if this is the case.

100 : It is not clear to me why an offset of 20% was applied to SMO, given that the observations from RPB were obscured by extra-tropical air. Could the authors expand on this?

108 : Please could the authors show a standard deviation or range for the x3 H2 time series in Fig S1b

116 : It's a little confusing what is being derived from HFC-152a for OH and what is being taken from the literature. In line 116, the authors state that the prior information for the seasonal amplitude of OH is taken from Spivakovsky et al, but in line 94, the authors state the OH seasonal cycle is derived from HFC-152a measurements. Do they mean that only the amplitude is taken from literature and the monthly cycle (i.e. when in the year this amplitude occurs) is derived from HFC-152a? Or simply that the prior OH seasonality is taken from the literature and then optimised by HFC-152a?

139 : Can you give a (very) brief justification for using up to 300hPa in the N and S box, and 200hPa in the tropics

147 : It is unclear what the authors mean by "method 1" here

150 : "A constant prior soil sink was used in each box" I assume the authors are referring to H2 here, but it sounds like they are referring to HCHO from wording of the previous sentence

Section 2.3 : Could the authors give more information about the Bayesian Inversion model? Do they assume any prior distributions for their inputs, or are these all based on monthly averaged time series? Are the prior uncertainties inputted as a normal distribution (where e.g. 35% is the standard deviation) or are they the range of inputs?

172-3 : Can authors clarify what is meant when HCHO photolysis rates are "retrieved with cross-correlation between OH"?

200 : Bring forward the reference to Fig 2b on line 201 to the last sentence of line 200 for clarity

201 : Do the authors mean to reference Fig 3c here (instead of 2b) which shows the double OH peak in the tropical region?

209 : Reference Fig 2a. It might be worth noting that the posterior standard deviation of these discrepancies is also within the observations.

212 : It is not immediately clear why the forward model would be 9 months out of phase but still simulate a strong seasonal cycle. Can the authors offer an explanation for this?

220 : Please refer to the relevant figure here

227 : This is a confusing way of describing the plots. Why done write “North box (3a), Tropics box (3b), South box (3e)” etc.?

234 : Could the authors expand on the comparison with Bousquet et al? How ‘good’ is the agreement? Did they have the same peak? Similar variation/standard deviations?

236 : What is the OH range in the tropics? Perhaps give the standard deviation as a percentage of this, so it is easier to compare with the North and South boxes (also see next comment)

231-243 : The reader is only given an OH range and STD for the north, a range for the tropics, and a range for the south box. Given that the authors have specifically chosen not to plot the absolute values in Figure 2, I think it is important to state consistently the OH values and standard deviations in all three regions.

260-261 : Need to state that there is reasonable agreement in the soil-sink *variation* between the two methods. Can the authors also state a percentage contribution from the soil sink from the different regions?

266 : Please state relevant figure

269-270 : I’m not convinced by this statement of seasonality. The authors say that the range of H2 soil-sink is also the same as the standard deviation, which implies there is no significant seasonality at all. If there is an agreement of a lack of seasonality, they should state this

273-275 : Similarly here, can the authors comment on the significance of the peak and trough sink soil in the south given that the posterior standard deviation is similar to the range

282 : Is it possible to say how much each box contributes to the overall the global soil sink (I imagine this is weighted heavily towards the north, but % contribution would help clarify this)

297 : I think the authors could expand on the reasons why there is more uncertainty for the soil sink phase in the tropics. There’s a greater abundance of OH in this region, so it would make sense for this area to be more sensitive to OH. We also wouldn’t expect to see as much seasonality in the tropics as in the other two boxes (averaging across ~20 years with various ENSO indices). Is it also possible that there is more uncertainty in the H2 observations for this region (the H2 observations were averaged between several sites)?

297 : Are authors referring to all cases here? (And again in line 300)

301 : Can the authors expand more on the H2 chemical production. I would also consider moving Fig S7 to the main text and add a short section about this.

Fig 4 + S7 : only three error bars are given in these plots (compared to Fig 3). Is there a specific reason why the authors cannot show the other error bars for the remaining months? For Fig S7, please also show error bars for the HCHO derived OH case

Table S1 : Do the authors mean days^{-1}? The values in the final two columns seem to show the lifetime transport? Also, are the same transport mixing timescales used in reverse e.g. Tropics → North and Tropics → South?

Fig S2 : What starting conditions are used for the slow and fast OH transport?

Technical Comments

44: "*on* the impact of OH on…"
87 : comma missing after SF_6
91 : remove brackets from citation
129 : Missing a bracket
146 : P0 → P_0
165 : m → *m*
170 : "the prior uncertainties values minimized retrieval uncertainties" → "the prior uncertainty values minimized retrieval uncertainties"
172 : section 2.2 → Section 2.2
229 : 2 → two
Table S1 : Difference hyphen type in North-Tropics to South-Tropics
300 : remove "is"
316 : remove comma after 'production'