We thank both reviewers and the chief editor for their constructive comments on this article. We provide a point-by-point response to their comments below. In bold are the comments of the reviewers, followed by our replies.

Reviewer # 1

Line 6: remove comma after scheme

No change. The sentence reads correctly with the comma.

Line 14: 'by realistically simulating OH in the atmosphere' – after reading the paper, the support offered for this statement is a comparison of the tropospheric chemical lifetime of methane in EMAC (8.9 years) with an 'observational estimate' of 9.1 years from Holmes et al. (2013). Could the authors confirm that these two values are directly comparable (i.e. represent the same loss mechanisms)? The 9.1 yr value from Holmes et al. appears to come from Prather et al. 2012 – and is an estimate of the total atmospheric lifetime for methane (i.e. includes losses to the stratosphere and soil). The observational value for methane lifetime against OH based on methyl chloroform data is quoted as 11.2 years in Holmes et al. (2013) (and is also taken from Prather et al. 2012).

The abstract has been modified. The use of methane lifetime as an estimate for the OH radical is explicitly mentioned. In section 4.1, we have extended the discussion on CH_4 lifetime and explicitly explain the dominance of OH as methane sink as the reason why methane lifetime is regarded as a proxy for the global oxidising capacity of the atmosphere (Lelieveld et al., 2016; Saunois et al., 2025).

We also thank the reviewer for pointing us to Prather et al. (2012). The reference to Holmes et al. (2013) has been removed and replaced by Prather et al. (2012) for the total observational constrained methane lifetime of 9.1 ± 0.9 years. We also directly refer to the supporting information of Prather et al. (2012) for the tropospheric chemical lifetime: estimate of lifetime for reaction with tropospheric OH from methyl chloroform observations, 11.2 ± 1.3 years and reaction with tropospheric chlorine of 200 ± 100 years.

Line 49: 'We find that correctly representing the oxidising capacity of the atmosphere is critical for predicting H2 mixing ratios and their spatio-temporal variability'. I'm not sure that this has been done – you have captured observations of H2 mixing ratios using the model's OH and representations of the other sources/sinks, but you have not shown how sensitive H2 is to differences in OH? Spatially, and seasonally, H2 will likely be very sensitive to the soil sink – which is estimated to be significantly larger than the OH sink, as well as photochemical production of H2.

We have modified the text accordingly and explicitly present the global budget of the soil sink and atmospheric production of hydrogen, which are in fact quite comparable in our simulation.

Line 64: Just a note that diurnal variability is seen in longer-lived gases at the surface due to diurnal variations in boundary layer height and the resulting changes

to mixing (e.g. H2 measurements form Weybourne - Forster et al, Tellus B, 2012).

Thanks for making the authors aware of previous work undertaken by Forster et al. We find that the diurnal variations in H_2 measurement presented by Forster in Figure 5 are captured well within the uncertainty bounds of the measurements. Based on this finding, and due to computational constraints, we believe a daily time-step is appropriate for our model analysis.

Line 66: I think there should probably be an acknowledgement here that in 2020, methane was (and still is) not in steady-state in the atmosphere. Therefore a simulation run to steady-state using repeating 2020 emissions is not fully representative of 2020 atmospheric conditions.

We have changed the text accordingly.

Line 71: 'swamps or wetlands': these 2 words are not interchangeable - swamps are a type of wetland. Maybe change to 'wetlands other than bogs'?

We thank the reviewer for noticing this. Obviously, the category swamps contains all wetlands which are not bogs. We renamed accordingly.

Line 74: Change 'swamps' to 'wetlands' here (see comment above)?

We have made the requested change on line 74.

Line 86: Do the RETRO/GFED databases include estimates for H2 emissions? Usually, H2 emissions are obtained by using H2:CO emission factors to scale CO emission inventories – has the same been done here, and if so which emission factors are used?

Yes, we can confirm that both the RETRO and GFED databases provide direct estimates of H_2 emissions without relying upon an assumed H_2/CO emissions ratio. We added: 'Both the RETRO and GFED databases provide direct estimates of H_2 emissions without relying upon an assumed H_2/CO emissions ratio.'

Line 90: Could the authors clarify if the emissions for non-GHG species are transient or for a single year/climatology?

We added at the first paragraph of section 2.1: 'Therefore, emissions are based on the year 2020 or the closest year prior to 2020 and are repeated for each year, removing any interannual variability.'

Line 100, Section 2.2: It would be nice to see some analysis of the soil deposition velocities calculated within the model, including geographical variation etc. Have the authors compared model results to any deposition velocity measurements?

As there are only a few available measurements of deposition velocity, we validated the resulting H_2 mixing ratios instead. Paulot et al. (2021) have previously validated the deposition velocity parametrisation used here by comparing it with deposition measurements taken at a small number of stations.

Line 129: We are told the scaling factor 'A' is adjusted to obtain a global mean

deposition velocity of 0.033 following Yashiro et al. (2011). What was the reason for choosing this value to scale to? The Yashiro study tuned their global soil uptake to optimise the model agreement with observations (by adjusting their inactive layer thickness, delta). Also, what value of 'A' was required for the adjustment – i.e. was a large or small adjustment required?

Initially, the deposition velocity was calibrated by adjusting the parameter A (representing bacterial activity) to provide global dry deposition estimates that agreed well with existing estimates by (Paulot et al., 2021) and Ehhalt and Rohrer (2011). To improve the agreement with H_2 station data, the parameter A was further adjusted to obtain a small reduction in the globally averaged deposition velocity by about 5% to 0.033 $\mathrm{cm}s^{-1}$, consistent with Yashiro et al. (2011). The resulting value of the resolution dependent parameter A is 10.9 using ERA5 input data with 0.25 degree grid spacing.

Line 146: Does the soil uptake calculated vary with time during the simulation, or is the ERA5 data averaged over a set period?

The soil uptake varies with time according to the ERA5 input data which is synchronised with the ERA5 nudging data.

Figure 1: Much of the globe is a similar shade of green. You can see the IHG in the modelled data – but all observed data looks the same/very similar on this colour scale. Would it be possible to adjust the colour scale so that it is easier to discern differences in H2 between observational sites and see a bit more structure?

Agree – change made.

Figure 2: It would be helpful to have some further info on the station rather than just the station code in this figure, e.g. latitude/longitude. I think it would also help the reader if the plots were ordered by latitude going from north-south rather than alphabetical order. This would make it easier to discern latitudinal differences in seasonality and how the model is performing.

Agree. We have ordered the plots by latitude as requested.

Line 225-230: As mentioned above, it's not clear to me that the 8.9 and 9.1 year lifetimes quoted are comparable. The EMAC methane lifetime of 8.9 years is within the range of the other models referred to by Yang et al. 2024 that Yang argues underestimate the methane lifetime (8.3 to 9.5 years).

We thank the reviewer for this comment. The last paragraph of section 4.1 has been modified and we have consequently based our lifetime calculation on total atmospheric burden for methane and hydrogen. Therefore our estimated CH_4 lifetimes are directly comparable to Prather et al. (2012). We have also changed numbers in Table 1 accordingly and noted that all calculations are performed with respect to global burden.

Line 247: 'The small long-term trend in H2 captured by the model' – I thought this was a steady-state simulation with methane and hydrogen emissions held constant (I'm not sure if non GHG emissions are transient)? We know CH4 (which is a source of H2) is increasing in the atmosphere, and this will not be captured in a steady-state

simulation.

Agree. This sentence has been removed.

Line 254-5 (and elsewhere wherever bottom-up is mentioned): The authors refer to bottom-up and top-down estimates for the global soil sink (55-60 Tg/yr and 85-88 Tg/yr respectively). I think it is worth pointing out that many of the 'bottom-up' studies have scaled the global soil sink uniformly to capture H2 atmospheric observations (which makes them top-down global soil sink estimates from my perspective - although the geographical variability will be bottom-up). The only truly bottom-up global soil sink estimate I am aware of that did not scale the soil sink to match total sources is Sanderson et al. (2003). The 'top-down' studies that are referenced are HD and inversion studies in which the larger soil sink was balanced by larger estimates of the photochemical production of H2 to close the H2 and HD budgets. The impact of including a geological H2 source in these studies is not clear without further information on the spatial/temporal variability and isotopic composition of the geological source. The way this sentence is phrased – 'the unaccounted for source almost bridges the gap between bottom-up and top-down estimates' implies that the larger soil sink estimates are consistent with the existence of a geological source, which I don't think can be assumed.

We agree that, as it has been formulated, the geological hydrogen is highly speculative and that there might be a misinterpretation of bottom-up versus top-down estimates. We still think that a potential geological source, if it exists, would be beneficial for the top-down inversion studies. There would be no (lesser) need for an up-scaling of photochemical production, as mentioned by the reviewer. Nevertheless, since this H_2 source and its isotopic composition has not been confirmed, we removed the sentence and just mention this potentially unaccounted source term. We have rewritten: 'Ellis and Gelman (2024) estimate that global geological H_2 resources cause an additional global flux of 24 Tg yr^{-1} from the subsurface to the atmosphere. This is speculative and would add unaccounted hydrogen emissions almost of the strength of the current non-photochemical sources. Current knowledge concerning the budget of atmospheric H_2 does not exclude the existence of a large geological H_2 reservoir, and further emphasises the importance of the dry deposition for the global atmospheric H_2 budget.'

Line 280-1: 'the CH4 chemical lifetime in excellent agreement with observational estimates' – see my comments above about the CH4 lifetime.

We thank the reviewer for the very useful hint. We have improved the discussion on methane lifetime in section 4.1. We consequently refer the reader to the original paper of Prather et al. (2012) and have extended the comparison regarding tropospheric chemical lifetime.

Line 281: 'We conclude that correctly simulating the oxidising capacity is a key requirement for high accuracy simulation of H2' – I'm not sure why OH is emphasised here above other sources/sinks in the H2 budget.

We agree with the comments above about oxidising capacity that was not systematically explored and the major impact of the soil sink of H_2. But we believe that the good results concerning the meridional gradient of H_2 are supporting that the simulated interhemispheric differences in OH are adequate. The text has been changed accordingly. We have replaced the sentence: 'The H_2

soil sink, based on a two-layer soil model (Yonemura et al., 2000; Ehhalt and Rohrer, 2013; Paulot et al., 2021), in combination with monthly ERA5 reanalysis data for soil related parameters has been successfully used by the EMAC model. We conclude that atmosphere chemistry models with such features, capturing the most dominant terms of the atmospheric H_2 budget, should be able to generally simulate station observations of atmospheric hydrogen. This gives confidence that scenario simulations on future hydrogen economy provide reliable estimates of the atmospheric impact.'

Reviewer # 2

Title - 'long-term source sink budget simulation' is slightly misleading, given the input emissions for hydrogen are fixed to the year 2020 (but taken from the year 2000 in the RETRO dataset). I would suggest changing this title to reflect the actual simulation, or adding a comment discussing the trend in the deposition sink over the period.

We agree that the title might be slightly misleading. It has been changed to equilibrium simulation. We think that the wording long-term is still justified, since we are using flux boundary condition for H_2 and CH_4 emissions. The performance and long-term stability of such a setup for two closely coupled chemical species in the atmosphere is the topic of this paper. This is of special importance for applications of the EMAC model concerning scenarios regarding a future hydrogen economy.

Line 4: 'Long-term global simulations were performed, with a horizontal resolution of 1.9 degrees. The results of this simulation are compared with'. Same comment here regarding 'long-term'.

Please see, the comment above. We changed the text to: 'Long-term global equilibrium simulations'.

Line 6: Clarify that the soil sink is not the only sink present (as it currently reads), but that there is also a chemical sink for hydrogen in the model.

Agree. We changed the text to: 'We introduced H_2 sources and sinks, that latter which was inclusive of a soil uptake scheme that accounted for bacterial consumption'.

Line 38: State that all three species listed are radiatively active, not just stratospheric water vapour.

We have changed the text accordingly: '... increases in radiatively active species ...'

Line 39: Change 'can' to 'may'. The impact of hydrogen on aerosols is highly uncertain currently. In the next line 'Arising from these observations' should be changed to 'Arising from these modelled results' or similar.

Line 198: 'from' Zimmerman et al.

Agree – changes made.

Line 10: Here, and later in the paper, discussion of 'direct anthropogenic perturbation' could be expanded. Terminology such as 'unpolluted air masses' - see Derwent et al. 2023 (doi: 10.1016/j.atmosenv.2023.120029) may read better. I think the paper could benefit from a discussion (or an additional figure of the emissions input

such that sites near high emission regions can be clearly identified) of how it was determined which sites fit this criteria for being background/ undisturbed stations. This discussion regarding sites near high emission regions could also benefit from a statement on the long lifetime of hydrogen.

We thank the reviewer for the hint. Unpolluted has been added, and the term direct anthropogenic perturbation has been omitted. Air masses can be polluted without anthropogenic influences, e.g. naturally occurring wildfires etc.

Line 15: Remove 'realistically'.

This text has been changed according to the comment from Reviewer 1 at line 14.

Line 58: Link the sentences on spin-up to steady-state. In the intermediate sentence here, expand upon the issues which arise from prescribed boundary conditions. This is key to the benefits of this model simulation and should be highlighted.

We thank the reviewer for this comment. See our response to the paper title. Here we added additional information on the strong chemical coupling of H_2 and CH_4: 'H_2 and CH_4 are chemically coupled and have nearly the same chemical lifetime (Table 1). Both compete for the hydroxyl radical OH as a chemical sink term, with OH being by far the dominant sink for atmospheric methane (Saunois et al., 2025, see section 4.1 below). Furthermore, is the atmospheric oxidation of CH_4 which is the largest source for H_2 (Ehhalt and Rohrer, 2009). To adequately simulate such a coupled system, the EMAC model uses flux boundary conditions for sources and sinks of both species.'

Line 67: The COVID-19 pandemic in 2020 could first be mentioned here to clarify that emissions are estimates without lockdowns. It may also be useful to clarify here that although interannual variability for emissions is removed, there is still meteorological variability (and potentially other sources of variability?).

We have to apologise for a typographical error. The emissions of anthropogenic sources CAMS-GLOB-ANTv4.2 and CAMS-GLOB-AIRv1.1 are used with COVID-19 reductions. That's why Reifenberg et al. (2022) has been cited. The reduction in concentration of the hydroxyl radical OH is below 4% for most of the atmosphere with the exception of the uppermost troposphere and tropopause region, which is due to reduced flight activity. The small impact on OH is foremost confined to northern hemisphere mid latitudes. We added at line 91: '... and the emissions are estimated with reduction due to lockdowns during the COVID-19 pandemic (Reifenberg et al., 2022). The reduction in concentration of the hydroxyl radical OH is below 4% for most of the atmosphere with the exception of the uppermost troposphere and tropopause region, which is due to reduced flight activity. The small impact on OH is foremost confined to northern hemisphere mid latitudes.'

Line 70: Is this methane simulation published? If so, please cite this publication here. If not, a justification of this statement and further discussion on the methane representation is needed.

We refer to the publication of Zimmermann et al. (2020), which describes a transient simulation with methane emissions. The text has been changed to clarify this: "For the long-lived tracer CH_4, the a posteriori emissions and the best combination of the rising-methane scenario of Zimmermann et al. (2020) have been applied. In this work, Zimmermann et al. (2020) show that the EMAC model has been efficient in simulating interactive CH_4 mixing ratios over the last two decades. Therein, the model results compare quite well with NOAA and AGAGE stations and measurements from CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container) flight observations (Brenninkmeijer et al., 2007)."

Line 82: Based on these slightly older emissions, a more detailed discussion on the comparison between this dataset and those used in more recent literature would be beneficial. There is mention later on of Paulot et al. (2021), but there is also the slightly updated values in Paulot et al. (2024) (https://doi.org/10.5194/acp-24-4217-2024). This paper, alongside results presented in Sand et al. (2023), could also be added to Table 2.

Agree. The requested data from Sand et al. (2023) and Paulot et al. (2024) has been added to Table 2.

Line 85: There have been changes in hydrogen emissions in the last few decades (see Paulot et al. 2021 and 2024). Following on from above, a more direct comparison between the dataset used in this paper and in the literature is needed.

We have consulted both papers by Paulot et al. (2021, 2024) as recommended by the reviewer. In both cases, we respectfully disagree that the suggested papers change our claim regarding the global H_2 budget.

Line 90: Include a sentence on the aerosol set-up used in the model.

We omitted: 'The ONEMIS submodel (Kerkweg et al., 2006) calculates natural emission fluxes of sea salt (Guelle et al., 2001) and dust (Klingmüller et al., 2018; Astitha et al., 2012).' It is correct that the ONEMIS submodel provides this data in the simulation, but it is not used. It is replaced by: 'The simulation uses a climatology of the aerosol wet surface density to calculate heterogeneous reactions. It is based on the CMIP5 (Climate Model Intercomparison Project Phase 5) emissions climatology for the years 1996–2005 low S scenario (Righi et al., 2013). The aerosol distribution for radiative forcing calculation is the Tanre climatology (Jöckel et al., 2006)'.

Line 139: Rephrase the threshold moisture content to clarify that there is no uptake in the top, dry layer in the model.

We clarified the sentence; "The soil water content in the top, dry layer (i.e. θ_{wI}) is assumed to be the threshold moisture content below which the bacterial activity vanishes i.e. there is no H_2 uptake in this layer, and is given by: ...".

Figure 2: The line colours on this plot need to be more distinctive. A suggestion here is to split the stations into northern and southern hemisphere and to add lines to indicate January 1st of each cycle. This plot also demonstrates the changes in the observations in the period leading up to 2020. There has been an increase in background hydrogen concentrations in that decade, evident in the observations shown in sites such as SPO and PSA in Figure 2. The model does not capture this increase, due to emissions being fixed at one year. This is interesting, as it shows that this may not be related to any changes in the hydrogen soil sink over this period (I am assuming these interannual changes are simulated in this run, but this should be clarified).

We rearranged Figure 2 and Figures B1 and B2 so that the stations now appear from north to south as requested. Furthermore, we clarified that for the equilibrium simulation for the hydrogen soil sink, data for the year 2020 is repeated. The observed slight increase in atmospheric hydrogen, especially obvious in the Antarctic observations, coincides with the increase in atmospheric methane since 2010 after the hiatus during the previous decade. This is added to : 'Especially the Antarctic stations show an upward trend for atmospheric hydrogen. The model with its emissions and soil sink data repeating the year 2020, cannot capture this feature. This trend coincides with further increasing atmospheric methane concentrations after 2010 following the hiatus of the previous decade (Lan et al., 2024). Atmospheric CH_4 is the largest source for H_2 (see Table 2 in Ehhalt and Rohrer, 2009). To investigate this further in the future, a model simulation with flux boundary conditions for H_2 and CH_4 in transient mode is needed'.

Line 191: Or related to uncertainty in the soil sink? Broadly the soil sink is considered to be the most uncertain part of the hydrogen budget, however in Table 1, the uncertainty is very low. Comment on how this uncertainty was calculated and why it differs from other budgets.

We thank the reviewer for this comment. As described in the caption of Table 1, the uncertainty in Table 1 is calculated as the standard deviation of multi-year annual global means. Since the boundary conditions of emissions and deposition fluxes are repeated (i.e. an equilibrium simulation), the standard deviations are small. In our simulation, the uncertainties represent just a statistical measure, and do not represent those caused for example by limitations of the soil model or other parametrisations or ERA5 data etc. We have added that the uncertainty in the soil sink is still large and that large land masses have no observational stations: 'The coverage of many continental land masses by the observational stations is sparse. For South America, Australia, Africa, Central Asia, Siberia and India there are almost no measurements available. This is a problem, especially in validating the soil sink, which can be considered the most uncertain part of the hydrogen budget (Paulot et al., 2021).'

Line 218: Can a comparison be made to more up-to-date literature than Yashiro (2011)?

We confirm that more recent literature is available for comparison e.g. Pieterse et al. (2013); however, the paper by Yashiro et al. (2011) represented the most useful basis for comparison with our study since it provided multi-year time series model/data comparisons for a large number of observational stations with the same fitting metric we employed. In contrast, Pieterse et al. (2013) presents model/data time series comparisons for only two years for a much smaller number of sites and with a different fitting metric.

Reference:

G. Pieterse et al. (2013). Reassessing the variability in atmospheric H_2 using the two-way nested TM5 model. JGR Atmospheres, 118, 3764–3780

Line 227: Yang et al. (2025) is now published I believe as 10.1029/2024GL112445

We have updated the reference accordingly.

Line 229: The 9.1 years estimate from Prather et al. (2012) states that "The atmospheric lifetime for a process is defined properly as the total atmospheric burden divided by total losses". Therefore, comparing this estimate of the atmospheric life-

time to the tropospheric chemical one from this model is not accurate. Please clarify this difference and discuss how this model compares to the discussion on Yang et al. (2025) with the correct value. Yang also discusses the chemical lifetime, with their model giving this value to be 11.4 years, compared to an observed methane lifetime of 11.2 years (plus minus 1.3 years), based on Prather et al. (2012). To corroborate the statements in the paper regarding the accurate representation of hydroxyl in EMAC, a figure similar to Figure 1 in Yang et al. (2025) might be useful to compare the OH modelled output with measurements.

We thank the reviewer for this comment. Also in reply to comments of reviewer 1 on CH_4 lifetime, we have modified the last paragraph of section 4.1. The budget is always calculated with respect to the total atmospheric burden. Also Table 1 has been updated and the caption has been extended. We also took the data from the supporting information of (Prather et al., 2012), to calculate the tropospheric chemical lifetime. This value is directly compared to our model results.

Line 248: The trend in OH over the 2010's would be influenced by methane emissions which have been held constant.

This is correct. There is no trend in the model data. The sentence has been removed. The misleading sentence is an error which occurred during the final editing of the paper. There is a weak trend in atmospheric H_2 in the last 15 years, which might be caused by the further increase of atmospheric methane concentration after the hiatus, but not in the model.

Line 255: In the previous paragraph, it was stated that this simulation agreed well with bottom-up estimates. If the additional source of natural hydrogen is present, a large shift elsewhere in the budget is needed, however the very small uncertainties presented in the hydrogen budget for this work do not leave room for any shifts.

We thank the reviewer for this comment. Please see the comment of reviewer 1, lines 254–5, who has stated that for bottom-up estimates, usually the soil sink is scaled to capture H_2 atmospheric observations. Whereas, in the case of top-down studies, the estimated soil sink is balanced by a scaling of atmospheric chemical production. Therefore, we have removed the remark concerning bridging the gap between bottom-up and top-down estimates. Because in our simulation the total atmospheric oxidation capacity based on methane lifetime seems to be in line with estimates based on methylchloroform observations, we have to conclude that a higher soil sink is needed to compensate for this unaccounted source. Based on the top-down estimates for the soil-sink this is possible.

Figure B2: WIS appears to have the same number of observations (167) as the sites included in Figure 2. Is there a reason this is not included in that figure?

WIS has 167 observations, whereas Figure 2 includes sites with 168 observations. This was not apparent as the model data only covered 167 months. However, the simulation has now been extended to December 2023, providing 168 months of coverage. All 168 months are now shown in Fig. 2. A discussion of Figure B3 would be beneficial.

We have added to the caption of Figure B3: 'Model data is compared with detrended observational data for the years 2010-2023 (inclusive) to perform this calculations.' For a more extensive comparison see Zimmermann et al. (2020).

Chief Editor

Dear authors,

Regarding compliance with the journal's Code and Data policy in your submitted manuscript, could you please clarify the current situation regarding the publication of the EMAC model? We have now seen many papers submitted to the journal storing the code in private repositories. In the past, authors of papers from the development team claimed that steps were been taking to address this situation and make the EMAC code public and open. What is the current status of this issue? Is the situation that you can not obtain permission to publish the code from former developers?

Dear Juan,

Thank you very much for asking about progress in releasing the MESSy code (including EMAC) under an open source licence. As you might imagine, releasing such a code base, of which some legacy parts date three or more decades back in time, relate to times when nobody in science really thought about source-code licensing, is a major issue. A lot of legal issues need to be tackled to solve this matter. This is in particular difficult, if more than 10 institutions, each of which has its own legal department, are involved. We are on a good path to addressing this matter, but some more legal issues need to be resolved. It is important to understand that such an open source release needs to be "bullet proof" in terms of voidability from third parties. At present, none of our staff who are legally authorised to sign agreements will do that easily without being convinced about the agreement "being safe".

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