

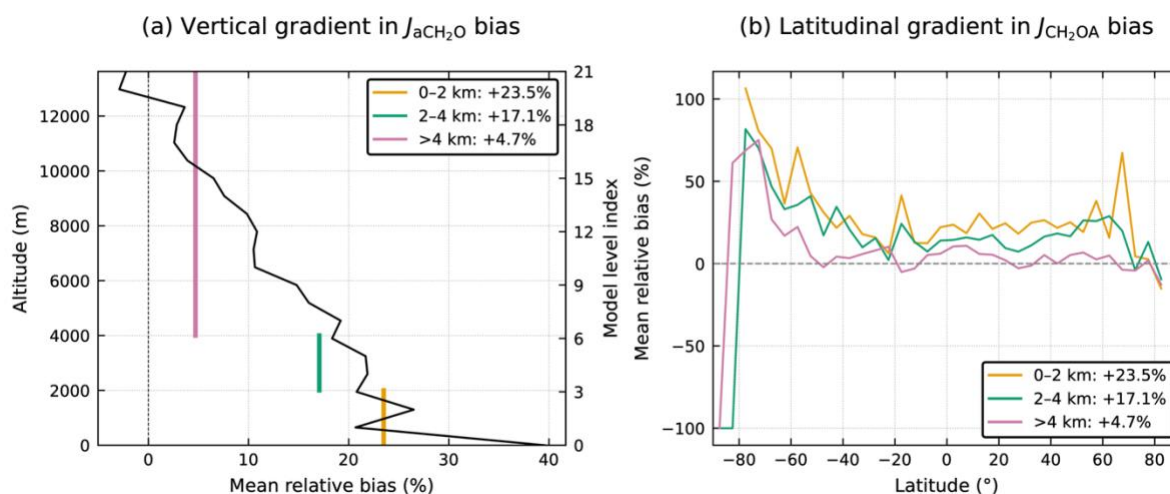
Dear Professor Archibald,

We thank you for taking the time to read our manuscript, and for your insightful comments. We would also like to thank you for directing us to the ATom-derived J values, which gave new insights into our results.

Regarding your first comment, our intention was indeed to discuss the photolysis rate constant in Section 4.2. This should be specified more precisely in the next iteration of the manuscript.

Regarding your second comment, thank you very much for this suggestion. We have performed additional simulations to make this comparison possible. It shows that our hourly simulated J values display a mean bias of +7.4% across all altitudes relative to the ATom observations. However, Fig. A (see below) shows that closer to the surface the bias increases; between 0-2km altitude it is +23.5%, and between 2-4km it is +17.1%. This confirms our hypothesis from Section 4.2 that J was overestimated in our simulations. Besides this vertical structure, the bias is relatively uniform across latitudes, except for 60S-90S. However, we note that the latter region has relatively little ATom measurements and is also relatively unimportant for H₂ production as both J and CH₂O are relatively low here.

Figure A: Vertical and latitudinal gradient in simulated bias in the photolysis rate constant for the photolysis of CH₂O through the molecular channel ($J_{\text{aCH}_2\text{O}}$).



Based on the found vertical bias structure, we calculated a correction factor for J for each model layer. Applying this bias-correction in the full-chemistry simulation reduced the annual mean photochemical H₂ production from 55.0 Tg yr⁻¹ to 50.3 Tg yr⁻¹ for the 2016 to 2017 period. This is still substantially larger than our posterior of 36.0 Tg yr⁻¹ for this period, which followed from the application of a global scaling factor of 0.7 in Section 3.1 and subsequent inversions in Section 3.2. We deem it highly likely that this difference can be attributed to an overestimation of CH₂O mole fractions in regions in our model where CH₂O mole fractions are largest, i.e. closer to the surface and above the continents, which contribute

relatively a lot to the photochemical H_2 production. In hindsight, this overestimation of CH_2O mole fractions in these regions was already visible in Appendix Fig. A1 for the flight above continental USA. We are aware that this contradicts our previously reported overall mean negative simulated bias (which still applies), but this contradiction follows from the fact that the ATom observations predominantly sampled oceanic and background conditions (which are relatively unimportant in terms of photochemical H_2 production). So, in summary, our inversions (global scaling factor of 0.7 + more advanced inversions) have likely compensated for an overestimation of CH_2O in our full-chemistry run derived prior, in conjunction with an overestimation of J . We plan to update Section 4.2 and Appendix A based on your comments and the additional analysis they motivated.

A natural question arising from these new insights is whether the prior photochemical H_2 production fields used in our inversions remain appropriate. We argue that they do. Although the full-chemistry simulation exhibits regional differences in CH_2O , this bias is predominantly systematic in nature, with a relatively uniform latitudinal distribution and vertical structure apart from an overestimation around the 1000 m level (Appendix Fig. A2). Consequently, the global scaling factor of 0.7 derived in Section 3.1, is expected to remain effective in producing a reasonable prior estimate of the photochemical H_2 source. Furthermore, reducing the CH_2O bias in the full-chemistry simulation would require substantial modifications to the underlying chemistry and emissions, which are beyond the scope of the present study. Finally, although informative, we note that the comparison with the ATom observations does not fully constrain the prior photochemical H_2 production on a global scale, as sampling above continental regions was limited to a few flights over North America.

Since the J -value bias correction results in a slightly different prior photochemical H_2 source, differing by approximately 3 Tg yr^{-1} after applying the global scaling factor of 0.7 derived in Section 3.1, we plan to assess its impact on our posterior fluxes. We will perform two additional inversion experiments for 2016–2018, using chemical sources derived with and without the bias correction. The uncorrected case also needs to be rerun, as this shorter simulation period will exclude observations outside 2016–2018 that influenced the original 2003–2023 inversion through the imposed temporal correlations described and motivated in Appendix Section B1. Given the relatively small difference between the two prior chemical production fields, and the more extreme prior perturbations explored in Appendix Section C, we expect only limited differences in the posterior fluxes. Nevertheless, we consider this sensitivity experiment important for quantifying the impact of the J -value bias correction.

We would be very interested to hear your thoughts on our analysis and would like to thank you once again for initiating this valuable discussion.