

We thank Tonatiuh Guillermo Nuñez Ramirez for the thoughtful comment and respond to specific points below.

In the study, OH concentrations were reduced proportionally to the proportion of the sink that the tropospheric Cl represented. However, in reality the relationship between OH and Cl concentrations and the lifetime and thus the CH₄ mole fraction is much more complex and non-linear, and depends also on the geographic and seasonal distribution of the OH, Cl and CH₄ fields. I believe the fact that the SimStd and SimGC simulations underestimate and overestimate, respectively, the observed $\delta^{13}\text{C}_{\text{CH}_4}$ in 2004, despite the fact that both include only a relatively small CH₄ sink from Cl arise from this issue. This makes the scenarios less comparable.

We do not alter the OH field between the SimStd and SimGC simulations, so differences in OH do not drive the differences between those simulations. In general, multiple factors including complex differences between the models' chemical mechanisms (not just Cl) drive OH differences between models. Since the purpose of this study is to quantify the effect of Cl on the isotopic composition of methane, we keep the scaling of OH for the SimTom and SimMBL simulations as simple as possible since it would otherwise be more difficult to disentangle differences due to Cl from differences due to OH distribution.

This is a forward simulation and as stated by the authors, the isotopic composition is not in steady state, then why should the CH₄ be in steady state? I fear reducing OH may mask some of the impact of Cl on the CH₄ mole fraction. The fact that neither the CH₄ mole fraction nor the $\delta^{13}\text{C}_{\text{CH}_4}$ are in steady state could be dealt with by subtracting the measured value at SPO.

CH₄ in the real atmosphere is not in steady state for either total CH₄ or the isotopic ratio, but as described by Tans (1997), the isotopic ratio takes longer to adjust to a perturbation than total CH₄. The purpose of our statement on line 206, “the isotopic composition is not in steady state in 2004 and the results of the sensitivity simulations diverge further with additional years of simulation, with SimMBL becoming clearly inconsistent with observations”, is to indicate that the temporal evolution of $\delta^{13}\text{C}$ in the SimMBL simulation does not match the temporal evolution of the observations.

Furthermore, if one of the objectives of this study was to compare the modeled gradient with observations, why were the OH fields scaled to be higher in the Northern Hemisphere when observational evidence points towards a NH/SH ratio of 0.97 ± 0.12 (Patra et al., 2015)? In the same spirit, if the spatial variations of the $\delta^{13}\text{C}_{\text{CH}_4}$ signal of emissions has been proven to be important to reproduced the observed signals in the atmosphere (e.g. Schwietzke et al., 2016, Ganesan et al., 2018) why is the SimWet only a sensitivity case and not the standard?

We altered the NH/SH ratio of the OH field to better align with what is typically simulated by global chemistry models, since we are using Cl fields from global chemistry models. Since the isotopic methane budget is under-constrained, there are many possible combinations of sources, source isotopic signatures, sink distributions, etc. that we could have chosen for our standard simulation, and some combinations would likely match observations better than SimStd. However, our goal was not to optimize the fit to observations, but rather to quantify the strong sensitivity of $\delta^{13}\text{C}$ to the range of Cl

concentrations reported in the literature. The finding of strong sensitivity to CI does not depend on the specific NH/SH OH ratio or source signatures used in SimStd.

I would like to make you aware of a possible conceptual error. Your VPDB value (0.0112372) is the old Craig (1957) value. This reference material has long been depleted. The actually valid number is 0.0111802 (recognized by IUPAC) from Zhang (1990). Please note that the measured reference in the Zhang (1990) paper was NBS 19, which defines the VPDB scale with $\delta^{13}\text{C} = +1.95$ per mill (Berglund and Wieser, 2011).

It is true that we, like many others in the literature, used the old value both for partitioning the total methane source into ^{12}C and ^{13}C components, and for converting our simulated ^{13}C and ^{12}C concentrations to $\delta^{13}\text{C}$. We felt that using the old value for the source partitioning was most consistent with the isotopic source signatures in the literature, such as the compilations by Houweling et al (2000) and Lassey et al (2007), which both cite the Craig (1957) number. We then used the same number for calculating $\delta^{13}\text{C}$ for consistency. However, we tested the impact of this choice by conducting an additional simulation identical to SimStd using the Zhang (1990) value for both the source partitioning and the calculation of $\delta^{13}\text{C}$. The new simulation gives very similar results to SimStd, demonstrating that our results are robust to the choice of VPDB as long as the same value used to calculate the source partitioning is used to calculate $\delta^{13}\text{C}$. We elaborate on this important in the Model Description section of our revised manuscript.

Also, you may be treating the CH_4 mole fraction as $^{12}\text{CH}_4$. However, for the CH_4 mixing ratio the mass range covers all masses from 16 ($^{12}\text{CH}_4$) to 21 ($^{13}\text{CD}_4$). Gas chromatography does not discriminate against higher isotopologues. Laser-based instruments, in contrast, do actually measure $^{12}\text{CH}_4$, but they are calibrated with gases whose composition was determined by gas chromatography. Thus the calibration gases must have a realistic isotopic composition to correct for this issue. To obtain, $^{12}\text{CH}_4$ you have to subtract the contribution of all other isotopologues from the mixing ratio.

We treat CH_4 mol fraction as the sum of $^{12}\text{CH}_4$ and $^{13}\text{CH}_4$. Our simulation only includes $^{13}\text{CH}_4$ and $^{12}\text{CH}_4$, as there are currently not sufficient observations available globally to constrain simulations of e.g. $^{13}\text{CD}_4$. Given the very minor relative abundance of the other isotopologues, we believe this error is very small compared to other uncertainties in the methane budget.

I would like to point out some potential improvement for the transmission of information. I would find an analog to figure 5(c and d) for $\delta^{13}\text{CH}_4$ more useful than the maps.

Fig. 7 shows the isotopic information in a similar way (as a function of latitude). We also add an additional figure showing time series of the isotopic values at individual stations.

I would also recommend to use sine latitude in all the latitudinal profile plots to retain the proportion with respect to the Earth surface. The correlation coefficients of latitudinal gradients of the different scenarios in figure 7 as well a comparison with their latitudinal derivative might also provide additional information.

The spatial correlation is provided in Section 3.2.

With this respect I believe figures figure S2 and S3 should be in the main text. Finally, a good validation for the different scenarios would be the reproduction of phase ellipses from Allan et al. (2001).

Since we added additional figures to the main text in the revised manuscript, we prefer to keep S2 and S3 in the Supplemental section. We experimented with including the phase ellipses, but found that they were noisy and provided little additional insight compared to what was already shown in the season cycle plots.

Finally some details in the text: • In the methods section you describe simStd as • No description of simWet in methods • In the description of SimTom "very short lived substances" is repeated twice in the sentence

We added a description of SimWet to the methods section.

References:

- Craig, H.: Isotopic Standards for Carbon and Oxygen and Correction Factors for Mass-Spectrometric Analysis OF Carbon Dioxide, *Geochimica Et Cosmochimica Acta*, 12, 133-149, 10.1016/0016-7037(57)90024-8, 1957.**
- Houweling S, Dentener F, and Lelieveld J: Simulation of preindustrial atmospheric methane to constrain the global source strength of natural wetlands, *J. Geophys. Res.*, 105(D13), doi:[10.1029/2000JD900193](https://doi.org/10.1029/2000JD900193), 2000.**
- Lassey KR, Etheridge DM, Lowe DC, Smith AM, Ferretti DF: Centennial evolution of the atmospheric methane budget: what do the carbon isotopes tell us? *Atmos Chem Phys* 7(8): 2119-2139. doi:10.5194/acp-7-2119-2007, 2007.**
- Tans P. 1997. A note on isotopic ratios and the global atmospheric methane budget. *Global Biogeochemical Cycles* 11(1): 77-81. doi:10.1029/96GB03940.**
- Zhang, Q.-L. and Li, W.-J.: A Calibrated Measurement of the Atomic Weight of Carbon, *Chinese Science Bulletin* 35, 290, doi: 10.1360/sb1990-35-4-290, 1990.**