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Comment on acp-2022-317

Martin Manning (Referee)

Referee comment on "Estimating emissions of methane consistent with atmospheric measurements of methane and $\delta^{13}C$ of methane" by Sourish Basu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-317-RC2, 2022

Summary.

This paper is an important extension of earlier work by the same group (Lan et al 2021). It now provides a detailed analysis of inverse modelling using CH_4 mole fraction and $\delta^{13}C$ data that shows the latter can significantly modify estimates of the methane sources. It also provides a careful comparison with a similar but different analysis of the methane budget that included $\delta^{13}C$ data (Thanwerdas et al, 2022). Problems with this type of inverse modelling for CH_4 were noted by Houweling, et al, 2017 (Global inverse modeling of CH_4 sources and sinks: An overview of methods. ACP, 17, 235-256) as requiring more careful approaches to prior estimates of source parameters and to model spin-up because of much longer isotopic equilibration times. More generally it is known that different approaches to inverse modelling can affect some forms of bias in determining the source – e.g. Biegler, et al. 2011. Large-Scale Inverse Problems and Quantification of Uncertainty, Wiley.

While this paper has not resolved all the issues with inverse modelling to determine CH_4 sources, it has made substantial progress. Concerns that most estimates of the CH_4 fossil fuel fraction were too low were raised by Schwietzke, et al, 2016 (Upward revision of global fossil fuel methane emissions based on isotope database. Nature, 538, 88-91). Furthermore, long term records of $^{14}CH_4$ in the southern hemisphere (Lassey et al, 2007, The atmospheric cycling of radiomethane and the “fossil fraction” of the methane source. ACP, 7, 2141-2149) have provided independent evidence that the fraction was about 30%. For this to now also be seen in a careful inverse analysis of the CH_4 concentration and $\delta^{13}C$ data should lead to a significant upward revision of the CH_4 fossil fuel fraction in future IPCC reports.

Suggested changes.

The relationship between this paper and Lan et al, 2021, is important and as they are in different journals it can help to repeat a bit more of what is in Lan et al here. E.g. the range of scenario options that had been considered in Lan et al before this paper adopts C_WL+ as the base for inversion analysis.

In section 3, I see no mention of how stratospheric removal is treated in the TM5 model. As this leads to return of air with some isotopically enriched CH_4 back into the troposphere, and that enrichment varying with latitude, it can effect the $\delta^{13}C$ analysis.

Section 4.1 “Comparison with Thanwerdas et al 2021” seems to have been added after the rest of the paper had been written. I think it would read better if this section was merged into section 3.6 “Comparison to the GCP budget”.

The rest of section 4 starts with a summary of what has been covered in the paper and then moves on to consider options for future development. After comparing this with some other papers in ACP, I would suggest that it would be better to have section 4 just focused on how the results can be developed with future work, and to move the initial summary, currently at the beginning of section 4, to a section 5 on “Concluding remarks” that summarised some of the key points made in the paper.

Technical Details.

The Courtier et al 1998 reference given here would be better as the peer reviewed version published in Quarterly Journal of the Royal Meteorological Society, 124:1783-1807, 1998. https://rmets.onlinelibrary.wiley.com/doi/abs/10.1002/qj.49712455002