Overall comment
Peter Rayner (Referee)

Referee comment on "Variational inverse modelling within the Community Inversion Framework to assimilate δ^{13}C(CH_4) and CH_4: a case study with model LMDz-SACS" by Joël Thanwerdas et al., Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmd-2021-106-RC2, 2022

This paper describes a system for estimating surface sources of methane (CH$_4$) using atmospheric observations and chemical transport models. While this classical inverse approach is well known and well tested, the paper extends it by including isotopic ratios of atmospheric samples and extracting sectoral information on emissions. In particular, they include the isotopic signatures of source categories in their target variables, relaxing the usual assumption of perfect knowledge. They demonstrate the importance of estimating isotopic signatures for the accuracy of estimated fluxes while also pointing out the cost in increased uncertainty.

The paper is a clear exposition of the inverse system. As the authors point out, it is not an analysis of recent methane fluxes, that awaits another paper. The current paper is well written and relatively complete. Since this is a contribution to a discussion I will restrict myself to large-scale suggestions and questions except for one minor point of language.

1. non-Negative Constraints} Is there a non-negative constraint on either emissions or isotopic signatures? I doubt this since it is (or was) not easy to do in the M1QN3 algorithm used here. It is, though possible by routines in the scipy minimisation suite that still offer the same limited memory capability. the advantages can be large since a non-negative constraint removes the risk of large positive-negative flux dipoles which can inflate the posterior uncertainty.

Using Smoothed Observations} I note the comment on Page 10. The observed high-frequency temporal variability
cannot be adequately reproduced by the LMDz-SACS model. Therefore, instead of assimilating the real observations, we used a smooth curve fitting the real observations." This is both striking and concerning. We noted from the earliest days of using high-frequency observations in formal inversions \citep{law02,law03,peylin05} that much of the power of high-frequency measurements came from the interplay between variations in meteorology and concentration. Abandoning this deserves more comment. What evidence do you have of the failure of LMDZ-SACS to simulate such observations? If you are using smoothed concentrations do you smooth the meteorology or the simulated concentration and (potentially) sensitivity the same way?

\paragraph{Spin-up and Spin-down} You noted on Page 18 ` `However, flux and source signature estimations of the 2012-2013 and 2016-2017 periods are not interpreted as the system appears to require a 2-year spin-up (2012-2013) and a 2-year spin-down (2016-2017), over which the inversion problem is not sufficiently constrained and isotopic signatures vary widely over time.". This is intriguing. It occurs, if I understand correctly, despite a long spin-up with 2012 fluxes to roughly equilibrate isotopic ratios at the start of the inversion period. Do you do this for every iteration as the control vector is updated? (I doubt this, it would be \emph{very} expensive.) I am particularly surprised by the spin-down problem. We are used to the idea that CO$_2$ fluxes, at least, are only really constrained by observations a few weeks into the future. After that atmospheric mixing homogenises the Jacobian too much. Hence fluxes too close to the end of a run might lack constraint. There might be a reason why isotopic ratios would have much longer-lasting sensitivities but this isn't obvious to me and deserves some explanation.

\paragraph{Computational Cost} The authors dwell on this a good deal. It seems almost a metric of a given set-up is its convergence rate. I suggest de-emphasising this. While I sure calculation time was frustrating it is mainly caused by the parallellisation limits on LMDZ-SACS. If these restrictions were reduced, as they already are in some other models, this would be a less important point. It is also certain to reduce in importance as models improve.

\paragraph{Minor Grammatical Point} ` `sensibility" should be ` `sensitivity" throughout.

\bibitem [{Law et~al.(2002)Law, Rayner, Steele, and Enting}]{law02}
Law, R.~M., Rayner, P.~J., Steele, L.~P., and Enting, I.~G.: Using high

\bibitem{law03}

\bibitem{peylin05}

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