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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric methane source and sink sensitivity analysis using Gaussian process emulation" *by* Angharad C. Stell et al.

Anonymous Referee #2

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This manuscript describes a method for testing the sensitivity of various atmospheric CH4 metrics to uncertainties in its budget components. The method generates Gaussian process emulators of hemispheric, monthly mean CH4 mole fraction and δ13C-CH4, using as inputs the same parameters used to initialize a 3-D chemical transport model. Once trained on and validated against 3-D CTM output, emulators are compared against multiple linear regression and a measure of the CTM uncertainty. The emulators are then applied by conducting millions of simulations covering the full parameter space of the 28 inputs. The sensitivities of CH4 and δ13C-CH4 to each input are quantified based on the emulator simulations.

Overall, this manuscript presents a novel, insightful approach to understanding uncertainties in the simulated atmospheric CH4 budget. The text is well organized and clearly written, references to the previous literature thorough, and presentation of figures clear. The topic is pertinent to ACP; after the authors have considered several comments, detailed below, this article should be suitable for publication.

Major Comments

My only major suggestion is that perhaps the observational dataset, currently just used to show that the CTM simulations encompass realistic values, could be incorporated into evaluation of the emulator simulations. For instance, looking at Fig. 4, the CTM appears to underestimate the observed global mean δ13C-CH4 value considerably (panel b). Could the emulator simulations be used to posit the drivers of the CTM underestimate? I understand that it would be unreasonable to meaningfully look at millions of simulations one-by-one, but perhaps the optimal values of the largest drivers of global mean δ13C-CH4 (from Fig. 7b) could be identified? I.e., which combinations of inputs are needed to close in on the observed global mean δ13C-CH4? This could be done for all the observed metrics shown in Fig. 4, if sorting through the emulator simulations to find observation-matching values is feasible.

Minor Comments

L78: While Gaussian process emulation has not been used for study of the methane budget specifically (as far as I am aware), it was recently used to evaluate the CH4 lifetime due to loss by OH. Please see and cite Wild et al., Global sensitivity analysis of chemistry–climate model budgets of tropospheric ozone and OH: exploring model diversity, https://doi.org/10.5194/acp-20-4047-2020

L93: The authors allude here to the Gaussian process inputs being maintained in their original spatial resolution. Does this mean all inputs are 2-D fields at 12x11.25 degrees resolution? Or are some 3-D? An explicit statement of exactly what is being fed into the Gaussian process emulators would be helpful, particularly regarding the inputs'

dimensionality.

Table 2: For the Trend values in the final column, the units are given as "%". Since trends are usually expressed as a rate, I would recommend noting the time period (I believe 2000-2012, based on my interpretation of the text) in the Table header information.

L195: I would hesitate to say that the loss of CH4 by OH is linear; the abundance/loss of CH4 has a feedback on the abundance of OH (see, e.g., Holmes et al., JAMES, https://doi.org/10.1002/2017MS001196). This would likely only influence results regarding large perturbations to CH4, so may not be relevant here, but it should probably be noted.

L360: It would be interesting to assess the role of altered spatial distributions of OH, both in the horizontal (i.e., more NH OH as many global models simulate) and in the vertical (i.e., what if there's more OH in the free troposphere than anticipated by Spivakovsky et al.?). It is understandable if this is beyond the scope of the current study but would make a good future direction.

L380: "are a serious" should be "is a serious"

L388: I would be interested to see a bit more discussion regarding the freshwater source of CH4. Some context regarding what is known about these emissions (that these are distinct from wetlands, what we know about the mechanism (bacteria?), that they are perhaps close in magnitude to wetlands emissions, etc.) would be helpful to the reader without them having to refer back to Saunois et al. This is potentially a very interesting finding, and some context could help raise awareness of this issue in the community.

NB: both "fresh water" and "freshwater" are used in several locations; I suggest maintaining consistency.

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