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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantifying methane and nitrous oxide emissions from the UK using a dense monitoring network" *by* A. L. Ganesan et al.

Anonymous Referee #1

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General comments

In this study, Ganesan et al. use atmospheric measurements of CH4 and N2O in a hierarchical Bayesian inversion framework to optimize fluxes of these two species over the UK. They found emissions of CH4 and N2O that are comparable to other inverse estimates for the UK. The inversion approach is fairly novel and represents an interesting advancement. In general, the methods used are sound and the paper is well written, however, there are some important pieces of information, which appear to be missing in the main text as well as a few points that need clarification. Therefore, I recommend this study for publication after minor changes.

1) The comparison of the modelled and observed concentrations at the 4 sites is an

important aspect of the study, since how well the prior model performs (in terms of transport, boundary conditions etc.) will also determine the accuracy of the optimized fluxes. Therefore, I think that the 2 figures in the supplement showing the concentration time series should be moved to the paper. Furthermore, I think these figures should include both the prior and posterior modelled concentrations as well as the prior and posterior boundary conditions. Also, it is not discussed anywhere how much the boundary condition changes after optimization and whether this is well constrained or not. Such a discussion should be included in the Results section.

2) Also related to the above comment, how well constrained are the boundary condition parameters (17 total)? In other words, how important do the authors consider "cross-talk" between boundary condition parameters and flux parameters to be in the optimization. This is important as only a few ppb for CH4 and a few tenths of a ppb for N2O error in the boundary condition can bias the fluxes significantly. This should be discussed.

3) There is no discussion of the results for the hyper-parameters (which were also optimized in the inversion). A discussion of the changes in these parameters and their significance should be included.

4) It is not stated in the main text how the prior parameters for the boundary condition polynomial were found. The only mention of this is in Table 1 (or 2 for N2O) where the authors state that it was from a fit to the statistically determined Mace Head baseline. Is it the case then that all 8 horizontal boundaries were fitted to Mace Head baseline? I think this should be mentioned in the main text. Also, I think it would be useful to move Fig. 1 from the supplement to the paper.

4) Although it is difficult to independently validate the optimized emissions, it would add confidence to the author's result to show how the optimized fluxes perform when coupled to the transport model and compared to independent measurements. Such independent measurements could be e.g the French site, Ile Grande (LPO), or the

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Shetland Islands site (SIS), both of which should be sensitive to UK emissions.

Specific comments

Title: I think describing a network of 4 sites across the UK as “dense” is perhaps a little misleading, and suggest that this word be removed from the title.

P858, L13-15: The uncertainty ranges given for the posterior UK CH4 and N2O emissions include the NAEI estimates (P859, L19-20). Do the authors consider the difference between NAEI and the inversion estimates to be significant, or rather that the two estimates are in agreement within the uncertainties? Also, Fig. 2 indicates a larger prior estimate than that of NAEI – is this difference due to the contribution of the natural emissions? Please also see comment below about the importance of natural emissions in the UK and how these were accounted for in the comparison with NAEI.

P858, L25: Do the authors have a suggestion as to why the correlation timescale for N2O is more than twice as long as that for CH4, considering that the transport is the same?

P859, L5: Insert “long-lived” before “greenhouse gases” to exclude water vapour.

P859, L11: Please state that the CO2-equivalency is by global warming potential, if that is indeed the case.

P859, L24-27: What are the proportions of biogenic (natural) emissions of CH4 and N2O in the UK? In Tables 1 & 2, prior sources of biomass burning and natural emissions are mentioned, how important are these in the UK and were these accounted for?

P862, L10: Were measurements assimilated from all time periods, i.e. were nighttime measurements included. If so, what were the typical nighttime differences between the measurements at the two heights were averaged? Also, what was the motivation for using the lower two heights rather than the uppermost height, which may be more representative of the well-mixed boundary layer?

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P862, L18: What was the resolution of the transport model, i.e. how large is the area covered by the 9 grid cells? And for what reason was this criterion chosen – please explain how having a high influence from the neighbouring 9 grid cells could lead to artifacts in the inversion. Lastly, how much data were filtered using this criterion?

P862, L25: Please specify which model errors, i.e. transport errors or other?

P863, L16: What is the resolution of the outer domain?

P863, L15-18: It is not clear to me how this outer domain was used, was it used to determine the boundary conditions? In the supplement, the authors state that the footprints along the boundary edges were summed to determine the fractional contribution from each boundary condition (10 in total). What is the connection between this calculation and the outer domain if any?

P869, L5: I am confused by this sentence, perhaps a simple rewording would make it clearer how the SD of the hyper-parameters were calculated.

P871, L18-22: How does the seasonal cycle in N2O found in this study compare to that found by other inversions in Europe, e.g. TransCom study of Thompson et al., ACP, 2014?

Fig. 2. It is interesting that there is no apparent cycle in the Irish N2O emissions in this figure, although from Fig. 5 there does appear to be a seasonal variation. Could the authors please comment on why this is?

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 857, 2015.