

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

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We thank the referee for his/her valuable comments, which we address below. Referee comments are in italics followed by our responses. Page and line numbers correspond to the revised manuscript.

*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*

C701

*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.*

It is difficult to show the a priori time series (or any time series for two years) in a figure in the main text. This is because at the size of the figure in the core paper, no features would be able to be discerned, as everything would appear small and crowded. It would not be possible to see how the prior and posterior compare to the observations. For this reason (and addressing comments from another reviewer to make the figures even bigger), these figures are in the Supplement and have been made much larger than would be possible in the core paper.

We have added a set of additional (large) figures to the supplement that show the prior time series against the observations. The plots have also been split by year in order to see even more clearly the comparison between model, prior and observations.

In the text we have written on Page 10 Line 304, ‘In addition, the simulated posterior and prior time series, baselines and comparison with observations are provided in the supplement.’

A discussion of the changes to the boundary conditions has been added to the Results section. The following text has been added on page 10 line 328: ‘Boundary conditions from the WSW, WNW, NNW, NNE and the two upper air directions were the most constrained, as reflected by the significant uncertainty reduction from the prior (over 50%), while air from the other directions were almost never sampled and thus reflected the prior distributions.’

C702

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.

We agree that the boundary conditions are vital for robustly estimating emissions. The majority of 'cross-talk' between emissions and boundary conditions will occur in the outer regions of the domain. Errors in boundary conditions will largely be 'absorbed' into the emissions of those outer regions and these regions are therefore not included in the analysis. The analysis is limited to a small fraction of the inversion domain (the UK and Ireland only).

The boundary conditions are generally the most constrained parameters in an inversion, because many observations are sensitive to these parameters (to the BC corresponding to the direction air had come from at the time). Therefore, the uncertainties are generally the smallest of any other state vector element. However, some directions will be better constrained than others, and those that are not constrained because air had not come from that particular direction very often therefore would not influence the mole fractions at the sites.

See General Comment #1 about the constraint on the boundary conditions.

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

C703

*significance should be included.*

The results for the hyper-parameters (uncertainties derived for each site and the correlation scales) were discussed in detail (now labeled Section 5.2 Covariance hyper-parameters).

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.

The polynomial fit was for the WSW boundary and each other horizontal boundary was found as an offset to WSW. The prior offset for the horizontal boundaries was 0, effectively assuming the Mace Head baseline everywhere.

The following has been added to the text on page 9 line 296, 'A priori, it was assumed that offsets to the horizontal boundary conditions was zero (i.e., the MHD baseline was assumed for all horizontal directions). For upper-air boundary conditions, the mean fraction-weighted (based on sensitivities derived by the NAME model) difference between upper-air influenced observations and baseline was assumed.'

Figure 1 from the Supplement has been moved in to the main text (now Figure 2) and refined for clarity, as described in one of the specific comments below about the inner and outer boundaries.

C704

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

We took the view to use the available measurements that were part of the DECC program, for the reason that the maximum sensitivity of UK and Irish emissions will be at these sites. Regarding the specific validation sites proposed above: The Shetland Islands is located quite far from the UK and would likely only sample baseline air, as it is far north of northern Scotland, where emissions are very low. The second site, Ile Grande, though potentially sensitive to UK emissions, would also be significantly influenced by French and other European emissions, which will not be strongly constrained by the DECC sites. Therefore, this site would largely reflect the prior for those regions and would make the validation more complicated. Internal sites (e.g. new GAUGE sites at Heathfield and Bilsdale) could be used for validating these emissions, as they are perfectly situated south of London and in Yorkshire; however, data from these sites have not yet been published and are not available for use at this time. In future work, we will investigate this validation; however, we very much agree with the reviewer that validation is important.

#### *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.*

'dense monitoring network' has rephrased to 'national-scale monitoring network.'

C705

*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.*

We have added/clarified some text on the significance of our results, when considering the uncertainties:

On page 10 line 309, 'Both UK CH4 and N2O emissions were generally lower than the total and anthropogenic a priori emissions. The difference in CH4 emissions is statistically significant (with the prior outside of the uncertainty of the posterior) but the N2O difference is not significant when accounting for uncertainties. Natural emissions, which are only 5-10% of the prior for both gases, may explain some of the difference, but are not large enough to account for all of it. Emissions from Ireland were consistent with the prior for both gases.'

On page 11 line 333, 'Though the a priori emissions have a small seasonal cycle due to the natural soil and oceanic sources of N2O, the derived amplitude of approximately 0.05 Tg/yr is much larger in the posterior estimates and is statistically significant. We discuss this seasonality further below. A small seasonality was found in Ireland's N2O emissions but this seasonality was not significant relative to the uncertainties.'

Regarding the role of natural emissions (similar to Reviewer 3 comments):

C706

We have changed the prior to provide a more realistic estimate of natural emissions in the UK/Europe. In our original inversion, we used natural emissions compiled from a variety of global inventories. These represent the most up-to-date published inventories, however, there are certain limitations. For example, Saikawa et al., 2013 did not mask out agricultural land in the N<sub>2</sub>O natural soil inventory, therefore, the natural emissions are likely to be overestimated. In the new inversion, we have scaled the natural emissions to be consistent with the percentage of natural land in the UK/Europe. We used published land cover maps, which have apportioned land to agricultural, developed and natural sources. We expect that this scaling will account for the areas that were classed as natural in the inventories but are used for other purposes.

It should be noted that the posterior solution for the majority of the UK and Ireland is largely independent of the prior. As described in detail in General Comment #1, we show through three sensitivity inversions, that the posterior solutions for the UK and Ireland totals have very little sensitivity to the choice of prior.

In our manuscript, we have used the inversion using the scaled-natural prior as our main results. We make all of our comparisons between prior and posterior relative to the scaled-natural prior. Therefore, we have added additional discussion about the role of natural emissions and also included tables in the paper that describe the contribution of the majority of sources in the prior to UK and Ireland emissions.

The following has been added on page 9 line 290, 'Natural emissions were compiled from a variety of sources outlined in Tables 1 and 2. To account for anthropogenic land that was classed as natural in these inventories (for example, the natural soil

C707

N<sub>2</sub>O source did not mask out agricultural land), natural emissions were scaled by the fraction of natural land in each UK and European country based on land cover maps [Morton et al., 2011, EEA 2007]. The contributions of the major source sectors to the UK and Ireland totals are presented in Tables 3 and 4. Anthropogenic sources were approximately 90% of the total for both gases.'

Regarding the comparison of posterior emissions to the prior or anthropogenic inventory:

Throughout the text, we have clarified that comparisons are relative to the prior and/or the anthropogenic inventory.

In abstract, 'We found that N<sub>2</sub>O emissions were consistent with both the prior and anthropogenic inventory but we derived a significant seasonal cycle in emissions.'

On page 10 line 309, 'Both UK CH<sub>4</sub> and N<sub>2</sub>O emissions were generally lower than the total and anthropogenic a priori emissions. The difference in CH<sub>4</sub> emissions is statistically significant (with the prior outside of the uncertainty of the posterior) but the N<sub>2</sub>O difference is not significant when accounting for uncertainties. Natural emissions, which are only 5-10% of the prior for both gases, may explain some of the difference, but are not large enough to account for all of it.'

On page 11 line 361, 'The small natural component, which is less than 10% of the total prior, could also be overestimated, but this would not entirely explain the difference between the prior and the posterior emissions.'

C708

In the conclusions, 'We found that the prior (largely from anthropogenic sources) was higher than our estimates for CH<sub>4</sub> emissions and likely overestimated from the agriculture sector. The small natural sources in the UK are not likely large enough to account for the full discrepancy between the prior and posterior emissions. Average posterior N<sub>2</sub>O emissions were consistent with the prior and the anthropogenic inventory but had an enhanced seasonal cycle, likely from fertilizer application.'

*P858, L25: Do the authors have a suggestion as to why the correlation timescale for N<sub>2</sub>O is more than twice as long as that for CH<sub>4</sub>, considering that the transport is the same?*

A discussion of the correlation scales for CH<sub>4</sub> and N<sub>2</sub>O was included in the Results section (Section 5.2 Covariance Hyper-parameters).

We described the interpretation of the correlation scales: 'The correlation scales are related to a number of factors: errors in the model transport (e.g., a misplaced weather front at one time will likely be misplaced a short time later) as well as unresolved emissions processes (e.g., errors in the assumption of constant emissions). The two sources of correlated errors cannot be disentangled but the time and length scales derived in the inversion are a measure of the scales of the missing or erroneous processes...The correlation timescale is smaller for CH<sub>4</sub> than for N<sub>2</sub>O. Though there are differences in the two networks (i.e., N<sub>2</sub>O is not measured at Angus), a CH<sub>4</sub> inversion in which Angus was excluded was also performed and similar correlation scales were derived (Supplement), suggesting that the network differences are not the source of differences in correlation scales. Furthermore, because the same transport model was used for the two studies, model errors were expected to be similar for the two gases so the differences are likely due to unresolved emissions in the prior. We noted the increased variances at Tacolneston and speculated that this was due

C709

to sporadic emissions from landfills and offshore gas that were not modeled by the constant prior emissions field and not resolved in the inversion. The longer timescale for N<sub>2</sub>O suggests that unresolved emission characteristics from fertilizers acts on a slightly longer timescale (several days).'

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

Long-lived has been inserted.

*P859, L11: Please state that the CO<sub>2</sub>-equivalency is by global warming potential, if that is indeed the case.*

The text has been modified to state that the CO<sub>2</sub> equivalency is by GWP.

*P859, L24-27: What are the proportions of biogenic (natural) emissions of CH<sub>4</sub> and N<sub>2</sub>O 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?*

As mentioned above, Tables 3 and 4 have been added that give the percentage breakdown of the major sources.

*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?*

C710

Measurements were assimilated from all time periods, including nighttime measurements. At all times, measurements from the two heights were averaged. Average differences (the average of daytime versus the average of nighttime measurements) were less than 4 ppb for CH4 and 0.04 ppb for N2O.

The upper height at Taconeston, because of its interaction with the boundary layer, may not always be representative of the well-mixed boundary layer, and therefore we would have to pre-select times where we believed the model simulates the boundary layer well when it is near the upper height. We agree with the reviewer that this is important to do and is the subject of future work. Furthermore, the time series of observations at the upper height is 6 months shorter than for the two lower heights.

The text now states 'Measurements were averaged... both day and night.'

We have also included the following statement on page 4 line 109, 'measurements from 185 m.a.g.l at Taconeston were not used due to the additional complexity of representing this height in the boundary layer'

*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?*

The resolution of the model is 0.352x0.253 degrees (mentioned on Page 5 Line 137), so the 9 grid boxes roughly covers 50 km around the site. The reason this criterion

C711

is used is because when air is relatively stagnant, there will be high sensitivity in the vicinity around the site, and at this time, any local influences (i.e. sub grid scale) will significantly affect the measurement. So the 9 grid boxes are used as a proxy for removing observations from times that could be significantly locally influenced, as the model does not resolve these processes. From MHD, RGL, TAC and TTA, 17, 13, 7 and 4% of CH4 data and 17, 15 and 9% of N2O data were filtered, respectively.

This has been clarified in the text, which now states on Page 4 line 117 that, 'Measurements corresponding to times when there was a high sensitivity of mole fractions to emissions from the nine grid cells surrounding the station were removed from analysis, as they are more likely to be affected by local processes due to the more stagnant air. Approximately 17, 14, 8 and 4% of data was filtered from MHD, RGL, TAC and TTA, respectively.'

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

Text has been changed to, 'Model errors (due to transport errors as well as errors due to unresolved processes) were estimated as part of the inversion framework.'

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

The resolution of the outer domain was 0.563x0.375 degrees and this has been clarified in the text on Page 5 line 142. 'For the purposes of estimating boundary conditions, a second larger domain (9-81N and -100-46E at resolution 0.563x0.375 degrees) was used to identify the origins of air masses that entered the smaller inversion domain'

C712

*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?*

The outer domain was used to track the origin of the air masses that entered the inversion domain. The reason a larger domain was used (instead of the inversion domain) is because the large-scale origin of the air mass is not always evident from the smaller inversion domain. For example, air that is southerly in origin (i.e. with Southern Hemispheric background values) often turns and enters Mace Head from the west. This is only evident when looking at a larger domain. Therefore, the boundary condition that is estimated represents the starting concentration of air entering from that larger domain.

This description has been added on page 7 line 194, 'The boundary conditions represent the concentrations on the boundaries of the outer domain, which is thought to be the direction associated with the 'source' of the air mass (e.g., winds that enter the inner inversion domain from the west sometimes originate from the south). Therefore, the concentrations entering the inner inversion domain are formed by the concentrations on the outer boundaries plus the effect of any emissions in between the two domains. For some directions (in particular the Northeast), there could be significant emissions sources, however, from the predominant directions (Southwest and Northwest), emissions sources are expected to be smaller. These emission sources do not affect the results of the inversion, which require boundary conditions to simulate the net concentrations outside of the inversion domain; however, physical interpretation of the boundary conditions must account for these emissions.'

C713

*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.*

The text has been revised to say, 'The standard deviation was chosen such that the 16th to 84th (cf., 1-sigma of a Gaussian distribution) percentile range was equal to 100% of the median emissions.' Just a note, that there was a typo and it should be 16-84th rather than 5-95th percentiles.

*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?*

Thompson et al., 2014 study found a seasonal cycle for Europe, the timing of which is consistent with what we derive in this study (peak in July). The magnitude of the seasonal cycle largely matches the prior that was used, which included both natural and agricultural soils. However, the amplitude was larger (1 Tg/yr versus 0.5 Tg/yr) than predicted in our study. The seasonal cycle we derived is largely independent of the prior that was used, suggesting that the increased seasonal cycle in Thompson et al., 2014 may be due to the presence of more natural soils in Europe as a whole versus the United Kingdom.

We have added the following text on page 11 line 336, 'Thompson et al., 2014 found a seasonal cycle over Europe with a timing consistent with our findings, however the magnitude of the seasonal cycle was larger and matched closely with the prior that was used. The difference in amplitude is likely to do with the greater prevalence of natural soils in Europe as a whole rather than the UK.'

C714

*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?*

If the values from Figure 1 were averaged into seasonal values (DJF, MAM, JJA, SON), there would be a small seasonal cycle, as reflected in Figure 5 (peak in JJA, min in DJF). However, when the uncertainties are considered on the seasonal emissions, it is not statistically significant.