

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 reviewer for the thorough comments, which we address below. Reviewer comments are italicized followed by our responses. Page and line numbers correspond to the revised manuscript.

General comments:

(1) It would be useful to compare in more detail with the previous studies of Manning et al. [2011] based on the NAME model, which used only a single station (Mace Head) to infer emissions from the UK and Ireland. In particular, it would be interesting to

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analyze separately the impact of the new methodology (hierarchical Bayesian inverse framework and further updates such as the treatment of boundary conditions) vs. the impact of the additional 3 monitoring stations. This could be easily done by performing an additional inversion with the new methodology but using only the observations from Mace Head. The proposed sensitivity experiment would allow a more systematic analysis of the differences compared to the previously reported inversions based on the NAME model.

We have done an inversion using our methodology with Mace Head data alone. However, there are issues with trying to directly compare Manning et al., 2011 results with this case study. Firstly, the results generated using the hierarchical Bayesian method are at monthly resolution, using a prior. In a given month only a fraction of Mace Head observations are sensitive to UK/Ireland emissions (i.e., Mace Head often sees baseline) and the result is that the posterior solution generally tracks the prior (see figure below). Manning et al., 2011 does not use a prior, but to get around the issue of having an under-determined system with only one station, the inversion is conditioned using 3-year moving averages (i.e., 3 years of observations that surround each month are used to define that month and then the month is stepped forward). The effect of this moving window is that many more observations are used and the emissions are smoothed over 3 years. A more minor consideration is that Manning et al., 2011 results only go to the year 2007 and the results of this study are for 2012–2014, making the comparison more indirect.

The only meaningful comparison between the two methodologies would be between the results of this study and the methodology of Manning et al., 2011 (applied without using a 3 year smoother) applied to 2012–2014 using all four stations. Unfortunately, these results have not been published and are not available for inclusion in this study. While we agree that a comparison between the two methods would be very useful and

important to isolate the effects of methodology versus additional stations, we do not have the appropriate comparison, as it would mainly show the differences between Manning et al., 2011 and the prior used in this study.

We note that because our system does monthly resolution inversions, it does depend on having high enough data density. We show through our three inversions (the first, which uses a natural + anthropogenic prior, the second which uses a scaled natural + prior and the third which uses an anthropogenic prior only), that the posterior solution is largely independent of the prior for the majority of the UK and Ireland.

We have added the following text to the main Results section on page 12 line 393, 'Two sensitivity studies are provided in the Supplement to show the effect of the prior on the posterior solution. Two additional inversions were performed: the first in which the prior assumed anthropogenic emissions only and the second which assumed that the natural emissions were not scaled by land use statistics. We found that the majority of the UK and Ireland were largely insensitive to the choice of prior and that the four station network has enough data density to constrain the UK and Ireland totals. While Northern Scotland is not very sensitive in the network, by design this is an area with low emissions and therefore does not significantly impact the UK total.'

(2) The paper should discuss more clearly the impact of natural emissions. The authors state that their results yield CH₄ and N₂O emissions 'generally lower than the inventory'. However, for comparison of the emissions derived in the inversions with the UK National Atmospheric Emissions Inventory reported to UNFCCC, the impact of natural emissions needs to be discussed in more detail. The authors use various scientific inventories for the natural sources (listed in Tables 1 and 2), but without giving numbers of the natural emissions for the UK / Ireland. Figure 2 shows 3.1 Tg CH₄/yr prior total emissions for the UK, while reported anthropogenic emissions are 2.4 Tg

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CH₄ /yr . I assume that the difference of 0.7 Tg CH₄ /yr is due to the applied prior natural emissions - but this needs to be discussed better (and more quantitatively) in the paper. Furthermore, the comparison between inventories and inverse modelling estimates should take into account the estimated uncertainties, i.e. it should be stated if the differences are considered statistically significant.

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 N₂O 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.

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

We have also 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:

In abstract, 'We found that N₂O emissions were consistent with both the prior and anthropogenic inventory but we derived a significant seasonal cycle in emissions.'

On page 9 line 290, 'Natural emissions were compiled from a variety of sources

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outlined in Tables 1 and 2. To account for anthropogenic land that was classed as natural in these inventories (for example, the natural soil N₂O 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.'

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.'

In the conclusions, 'We found that the prior (largely from anthropogenic sources) was higher than our estimates for CH₄ 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 N₂O emissions were consistent with the prior and the anthropogenic inventory but had an enhanced seasonal cycle, likely from fertilizer application. '

(3) Unfortunately the paper provides only very limited information about the validation of the NAME model. A more detailed validation of model transport is essential to evaluate the performance of the inversion, e.g. using independent observations of CH₄ and N₂O not used in the inversion, or specific transport tracers, such as 222Rn. I realize that such validation is limited by the availability of independent observations during the target period of this study. In any case, however, I would recommend to emphasize in the paper the need of more detailed validation in the future, especially regarding vertical mixing.

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We largely agree with the reviewer that a detailed validation study is important and would require independent observations (where the sources and sinks are well known) as well as methods to identify key components of the model that need to be improved. The latter is a subject of our future work. 222Rn measurements are not available at these sites, however. We have currently written, 'NAME has previously been validated against tracer release experiments, surface and balloon measurements but parametric and structural uncertainties are not well known (Morrison and Webster, 2005; Ryall and Maryon, 1998). While the results of this study cannot discern specific sources of error in the model, this is a subject of great interest and future work'.

We have added more emphasis on the fact that these validation exercises were not done during the period of our study and have modified this sentence to, 'NAME has previously been validated against tracer release experiments, surface and balloon measurements but parametric and structural uncertainties are not well known (Morrison and Webster, 2005; Ryall and Maryon, 1998). Further, validation exercises have not been conducted during the period of this study. While the results of this study cannot discern specific sources of error in the model, this is a subject of great interest and future work.'

(4) Regional scope of the paper: The paper includes a discussion of CH₄ and N₂O emissions from Ireland. However, in some parts of the paper the discussion is limited to the UK, and also the title suggests that the focus of the paper is only the emissions from the UK. I would suggest to include Ireland in the discussion throughout the paper (and extend the title of the paper accordingly).

The main extensions we have made in the discussion for Ireland:

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Title now includes Ireland

Table 4: Percent contribution of a priori sources to Ireland emissions

Ireland emissions are presented in the Results

On page 11 line 339, 'A small seasonality was found in Ireland's N₂O emissions but this seasonality was not significant relative to the uncertainties.'

Conclusions reflect Ireland totals

Further specific comments

title: 'dense' monitoring network: certainly the availability of 4 stations is a significant improvement compared to previous studies (relying largely on Mace Head for the UK / Ireland), but I would not really consider the 4 stations as 'dense' network

Title has been changed to, 'Quantifying methane and nitrous oxide emissions from the UK and Ireland using a national-scale monitoring network.'

abstract, page 858, line 12: 'emissions' to 'total emissions'

'emissions' has been changed to 'total emissions'

Introduction, page 859, line 5-6: 'global warming potentials over a 100 year time hori-

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zon of 34 and 298'. The authors refer here to the AR5 GWP including climate-carbon feedback - this should be explicitly mentioned (the corresponding numbers without inclusion of climate-carbon feedback are 28 (CH₄) and 265 (N₂O)).

It has been clarified that these numbers include climate-carbon feedback.

Introduction, page 859, line 9-11: 'legally binding target to reduce the country's CO₂ equivalent emissions to 80% of 1990 levels by 2050.' The target is to reduce CO₂ equivalent emissions by 80%, i.e. to 20% of 1990 emissions see: <http://www.legislation.gov.uk/ukpga/2008/27/section/1> : '(1)It is the duty of the Secretary of State to ensure that the net UK carbon account for the year 2050 is at least 80% lower than the 1990 baseline.'

Thank you for catching this (important) typo! The 80% has been changed to 20%.

Introduction, page 859, line 19-20: 'In 2012, the UK reported 2.42 Tg /yr of CH₄ with an uncertainty of 20% and 0.116 Tg /yr of N₂O with an uncertainty of 69%': would be useful to mention here also the numbers for Ireland. Please add reference for the given uncertainty estimates (as they are not reported in the the common UNFCCC CRF tables).

The uncertainties of 20% and 69% are located in Annex 7 of the National Inventory Report for the UK. Uncertainties for Irish emissions are found in the Ireland National Inventory Report as well. Calculating the net uncertainties results in an approximately 20% uncertainty on CH₄ emissions and approximately 88% uncertainty on N₂O emissions.

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We have added this information to the text on page 2 line 38, 'In 2012, the UK reported 2.42 Tg/yr CH₄ with an uncertainty of 20% and 0.116 Tg/yr N₂O with an uncertainty of 69% in the UNFCCC 2014 UK National Inventory Report...In the same year, Ireland reported 0.575 Tg/yr of CH₄ with an uncertainty of 20% and 0.024 Tg/yr of N₂O with an uncertainty of 88% in the Ireland National Inventory Report.'

Introduction, page 859, line 24-27: 'The principal sources of CH₄ in the UK...' I would suggest to modify to: 'The principal anthropogenic sources of CH₄ in the UK...'

'Sources' has been changed to 'anthropogenic sources.'

Introduction, page 860, line 20: '554 ± 56 and 15.8±1.0 Tg /yr (Prather et al., 2012)': The unit here is Tg-N /yr for N₂O (and not Tg N₂O /yr which is otherwise used in this paper'. Please check the number: In table 1 of (Prather et al., 2012) a value of 15.7±1.1 Tg-N /yr is given.

I have double-checked Prather et al., 2011 and it is indeed 15.7±1.1, however, it is presented as 15.8±1.0 in chapter 6 of the IPCC Fifth Assessment Report. We have revised our manuscript to reflect the correct value and units and thank the reviewer for spotting this error.

Introduction, page 860, line 21ff: 'Manning et al.(2011)...emissions for the UK...in 2007 to be 1.88 (0.8-3.3) Tg /yr CH₄ and 0.070 (0.055-0.090) Tg /yr': It should be discussed later in the paper (in 'results' section) in more detail, to which extent the lower uncertainty range found in this study for CH₄ (1.72-2.47 Tg CH₄ /yr) is due to the use of 4 monitoring stations compared to a single station only (Mace Head) by Manning et al. (2011) and to which extent to the new methodology (see general

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comment (1)).

As we discuss above, it is not possible to make a meaningful comparison between the Manning et al., 2011 results and the results of this study. Please see above discussion (general comment #1).

We have added the following text on page 10 line 316, 'The CH₄ emissions derived in this study are statistically consistent with the 2007 emissions estimated by Manning et al., 2011 while the N₂O emissions are slightly higher. The uncertainties derived in this study are smaller for CH₄ but larger for N₂O and the differences in uncertainties for the two studies is likely due to the different methodologies used as well as the additional measurement stations in this study. The hierarchical method provides a framework for more completely and rigorously characterizing random uncertainties in the system, but does not account for systematic uncertainties. The emissions and uncertainties derived here lie in the lower range of results obtained by Bergamaschi et al., 2014 continuing to point to large systematic differences between models.'

Introduction, page 860, line 24ff, 'Bergamaschi et al. (2014), using a variety of global and regional approaches, derived 2006-2007 emissions for the UK and Ireland that ranged between 2.5-5 Tg /yr for CH₄ and 0.07-0.17 Tg /yr for N₂O, depending on the inversion method and chemical transport model': It should be mentioned that also the NAME model participated in this model comparison, yielding typically lower CH₄ and N₂O emission estimates than the other 3 models used in this comparison.

We have added the following text on page 3 line 70, "Bergamaschi et al. (2014), using a variety of global and regional approaches, derived 2006-2007 emissions for the UK and Ireland that ranged between 2.5-5 Tg /yr for CH₄ and 0.07-0.17 Tg /yr for N₂O,

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depending on the inversion method and chemical transport model (with NAME derived emissions generally being lower than those from the other studies).'

Introduction, page 861, line 1-2: 'highlights the need for robust uncertainty quantification and investigation into systematic model errors.' It should be discussed later in the paper (in 'results' section) in more detail, how the error estimates have improved and how the new estimates (with their uncertainties) compare with the previous estimates of other, independent inverse models (e.g. model comparison of Bergamaschi et al. (2015)).

We have added the following text on page 10 line 316, 'The CH₄ emissions derived in this study are statistically consistent with the 2007 emissions estimated by Manning et al., 2011 while the N₂O emissions are slightly higher. The uncertainties derived in this study are smaller for CH₄ but larger for N₂O and the differences in uncertainties for the two studies is likely due to the different methodologies used as well as the additional measurement stations in this study. The hierarchical method provides a framework for more completely and rigorously characterizing random uncertainties in the system, but does not account for systematic uncertainties. The emissions and uncertainties derived here lie in the lower range of results obtained by Bergamaschi et al., 2014 continuing to point to large systematic differences between models.'

Measurements, page 862, line 2-3: 'No sample drying was employed at this site.': correction for water vapour (measured by Picarro instrument) should be mentioned.

The text now states on page 4 line 102, 'A water vapor correction (as measured by the instrument) was used at all sites and all measurements were calibrated using dry standards filled in aluminum cylinders.'

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Measurements, page 862, line 6-7: 'calibration factor of 1.0003.': give reference

The reference for Dlugokencky et al., 2005 has been included.

Measurements, page 862, line 9-10: 'In this study, an average measurement of the two lowest heights was used (measurements from 185ma.g.l. at Tacolneston were not used)': Explain the motivation to use the average of the two lowest heights. In general, higher measurements are considered more representative (less effected by local emissions and better represented by atmospheric models).

The upper height at Tacolneston, 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. A small error could either place this height inside or outside of the boundary layer. 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.

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

Measurements, page 862, line 16: 'Measurements were averaged over each two hour period...': Were both day and nighttime data used ?

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Measurements were assimilated from all time periods, including at night. At all times, measurements from the two heights were averaged. The text now states on 'Measurements were averaged. . . both day and night.'

Measurements, page 862, line 17ff: 'Measurements corresponding to times when there was a high sensitivity...were removed': would be useful to mention here the chosen threshold value.

Rather than mention the threshold value (as it is in units that are not physically intuitive), we specify the number of observations that were removed from analysis, for readers to get a sense of the threshold. We have added the following 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.'

Measurements, page 862, line 24-25: 'Typical instrumental uncertainties were 10 ppb CH4': 10 ppb as instrumental uncertainty of a Picarro instrument seems very high - should be typically in the order of 1 ppb or better.

We have written in the text, 'For CH₄ observations, measurement uncertainty described the variability of one-minute data in the two-hour averaging period. For N₂O observations, this uncertainty was the quadratic sum of the instrument precision (calculated as the standard deviation, SD, of the approximately hourly measurements of the standard each day) and the variability in the averaging period. Typical measurement uncertainties were 10 ppb CH₄ and 0.3 ppb N₂O.' We changed the word

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instrumental to measurement to avoid confusion.'

Atmospheric transport model, page 863, line 13-14, 'The inversion domain extended from approximately 36 to 67N and -14 to 31E': The chosen domain includes large parts of Europe. Hence, the derived emissions for the UK (and Ireland) depend not only on the boundary conditions, but also on the emissions derived for continental Europe. This requires some discussion in the paper.

We have added the following text 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.'

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.'

Inversion framework, page 864, line 9, 'individual grid cells' - what is the size of the individual grid cells - is this at the 0.352 x 0.234 resolution mentioned above for the

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meteorology or is a different resolution chosen for the emissions ?

This is at the 0.352x0.234 degree resolution. We have written on page 4 line 117, 'Data was filtered for local influence using a transport model. Measurements corresponding to times when there was a high sensitivity of mole fractions to emissions from the nine grid cells (at 25 km resolution) surrounding the station were removed from analysis...'

Inversion framework, page 864, line 20, 'Matern covariance function': would be useful to give a reference for the Matern covariance function.

A citation for the Matern covariance function (Stein, 1999) has been included on page 6 line 168.

Inversion framework, page 866, equation (4): I assume that stochastic error term ϵ accounts only for model errors ?

The stochastic error term accounts for all random errors in the model-measurement mismatch.

Inversion framework, page 866, line 25, 'if measurements were made daily...': how many measurements per day are used ? In section 2 it had been stated that measurements were averaged over 2 hours.

This example was used to illustrate the point, but we have changed the numbers so that it directly applies to this problem as well. The text now states on page 8 line 230 that 'For example, if measurements were made every two hours over a year at four

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sites, there would be 4380 (365 x 12) possible measurements at four locations and T would be of size 4380 and S of size four.'

Inversion framework, page 867, line 12-13, 'Through MCMC, these PDFs are sampled from and used to form the posterior PDF': would be useful to explain better, how the prior PDFs are optimized.

Optimized translates to choosing a statistic of the posterior distribution to represent that distribution. We have chosen to use the median and 5th and 95th percentiles to describe the posterior PDF. On page 10 line 302, we have written, 'Results are presented as the median of the posterior PDFs and uncertainties for all parameters correspond to the 5th to 95th percentile range.'

Inversion framework, page 867, line 15, 'A uniform distribution (U)': not clear to me, what exactly is meant by this uniform distribution.

The discrete Uniform distribution is a commonly used distribution in statistics (with a probability of 1 between a certain range, and 0 outside the range). Because it is quite often used (as often as Gaussian and lognormal distributions are used) we have left it to the reader to find this information.

Inversion framework, page 868: The description of the 4 covariance matrix properties could be moved to the supplement.

We feel that these properties are important for the reader to understand why we made the assumption of separability in the covariance matrix. These are important

C740

properties that make the scope of the problem computationally feasible, which is important for MCMC.

Inversion framework, page 869: line 6ff, 'Gridded anthropogenic emissions for the UK were from the NAEI for 2012...': The natural emission inventories should also be mentioned here - since they seem to contribute significantly (see general comment (2)).

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 N₂O 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.'

Results, page 869, line 19-20, 'Both UK CH₄ and N₂O emissions were almost continuously lower than the prior.': The difference seems to be also due to relatively high natural emissions. E.g., Figure 2 shows 3.1 Tg /yr CH₄ prior emissions for the UK, while the authors seem to use the reported emissions of 2.4 Tg /yr CH₄ as prior for the anthropogenic emissions. Therefore, I assume that the prior natural CH₄ emissions are about 0.7 Tg CH₄ /yr ? Likewise, also the N₂O emissions shown in Fig. 2 (0.14 Tg N₂O /yr) seem to include a significant contribution of natural sources (as reported N₂O emissions are 0.116 Tg N₂O /yr.

Please see general comment # 2 about the role of natural emissions.

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Results, page 870, line 6, 'oceanic sources of N₂O': Explain, how oceanic / offshore emissions are attributed to the UK (or other countries) ?

The grid cells comprising the UK were identified such that almost all of emissions reported in the NAEI occurred within what was defined as the 'UK'. Grid cells identified as UK through this method but which, geographically, largely fall within another country (principally Ireland) were re-classified as non-UK. The NAEI includes parts of the Atlantic Ocean, North Sea, Irish Sea and English Channel, where offshore sources are reported to exist. Other European countries were defined based purely on geographical boundaries over land, over the seas/oceans they were extended by 2 or 3 grid cells to catch potential offshore emissions.

Results, page 870, line 9ff, 'fractional uncertainties' / Figures 3+4: would be useful to include also uncertainties of the prior.

An extra panel (d) has been added to Figures (now) 4 and 5 that shows the uncertainty reduction from the prior, and further indicates which areas have the most constraint in the network. This new panel also provides an indication of uncertainty, relative to the prior which complements panel c, showing the uncertainty relative to the posterior.

Results, page 870, line 26ff, 'the largest difference, as a percentage of the prior, occurs throughout Scotland, western England and eastern Ireland': Maybe this is partly also due to the assumed natural CH₄ emissions? It would be useful to include also a map with the assumed prior natural emissions (e.g. in Figure 1).

We have provided maps of the dominant anthropogenic sources along with a map of the total annual average prior. We have also included Tables 3 and 4, with the

C742

percentage breakdown of each source to the UK and Ireland totals (natural sources are 5-12%). Together, the reader will understand the contribution of natural emissions in the UK and Ireland.

Results, page 871, line 23ff, 'Analysis of the uncertainties derived in the inversion (panel c of Figs. 3 and 4) shows the greatest observational constraint in the 100 km around the stations': To me the described patterns is not very clearly visible in the Figures. As mentioned above, it would be useful to show also the uncertainties of the prior which should demonstrate the uncertainty reduction more clearly (or alternatively show maps with the uncertainty reduction).

As mentioned above, an extra panel (d) has been added to Figures (now) 4 and 5 that shows the uncertainty reduction from the prior. This panel shows where the largest constraint is in the network, and directly corresponds to the dotted regions (statistically significant change from the prior) in panel b.

Results, page 872, discussion of model errors: I assume that the method used mainly estimates the model representation errors and only those components of the transport errors which are reflected in a mismatch between observations and model. However, there might be further systematic model transport error components, such as potential model biases in vertical transport, which might not be diagnosed by the applied methodology - since such transport errors can be (at least partly) compensated in the inversion by erroneous emission increments. Therefore, further independent validation is required, e.g. using independent observations of CH₄ and N₂O not used in the inversion, or specific transport tracers, such as ²²²Rn (see general comment (3)). This should be included in the discussion.

C743

We agree that there could be systematic errors in the model, which is the subject of great interest and future work. At the moment, the methodology in this paper only accounts for random uncertainties but we are working on methods to incorporate systematic errors into this framework. We very much also agree that additional validation methods are needed (see response to General comment # 3).

Results, page 874, line 3, 'As measurement networks around the world dramatically grow...': I would not consider the current increase of monitoring stations 'dramatic', but still rather slow (and many regions, even in Europe, not well covered).

The word 'dramatic' has been removed.

Conclusions, page 874, line 25, 'considerations that need to made': check wording

The typo has been corrected to 'considerations that need to be made'.

Table 1/2: - give numbers for annual (a priori) CH₄ and N₂O emissions of UK for different categories. - In addition, it would be useful to list here also the emissions of Ireland.

Tables 3 and 4 provide the breakdown of the major a priori sources.

Figure 3/4: Maybe it would be more useful to show the difference between posterior and prior (Figure (b)) in absolute units rather than as fractional differences (as in areas of low emissions a larger fractional difference has only a limited meaning)

We have chosen to plot the difference as a fraction so that the regions where there
C744

are significant changes from the prior would be highlighted. In absolute units, regions with larger emissions would appear to have larger changes, even if there was only a small influence from the observations. Similarly, even large changes in areas of low emission would not appear large in absolute units, again making it difficult to see the effect of the inversion. Therefore, it only has a limited meaning to plot absolute changes. We have also added an additional panel showing uncertainty reduction from the prior, relative to the prior, so that it is even clearer where the largest constraints are in the domain.

Supplement: Figures 2 and 3: would be useful to show the time series at higher temporal resolution in order to better visualize, how well the model represents the measurements (including e.g. diurnal cycles). Furthermore, it would be useful to give an overview about the overall model performance (e.g. mean bias and RMS for each station)

We have reformatted the supplement to show the prior and posterior time series at higher resolution (yearly plots for each site) so that it is more clear how the model/prior/posterior perform at each site.