

Referee # 1:

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 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.”

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 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 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.

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."

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):

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.

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.”

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 N₂O 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 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.”

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. ”

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?

A discussion of the correlation scales for CH₄ and N₂O 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 CH₄ than for N₂O. Though there are differences in the two networks (i.e., N₂O is not measured at Angus), a CH₄ 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 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 N₂O 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 CO₂-equivalency is by global warming potential, if that is indeed the case.

The text has been modified to state that the CO₂ equivalency is by GWP.

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?

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?

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 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. 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 Tacolneston 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 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”

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.”

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.”

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.

Referee # 2

We appreciate the helpful comments and feedback from the reviewer and address them below. Reviewer comments are italicized followed by our responses. Page and line numbers correspond to the revised manuscript.

Comments

• *Abstract, P858 L1-8: These three sentences do not really belong in an abstract, but rather in the introduction.*

The abstract has been changed to: The UK is one of several countries around the world that has enacted legislation to reduce its greenhouse gas emissions. In this study, we present top-down emissions of methane (CH₄) and nitrous oxide (N₂O) for the UK and Ireland over the period August 2012 to August 2014. These emissions were inferred using measurements from a network of four sites around the two countries.

• *P858 L21: It would improve the readability to start a new paragraph here, between the discussion of emissions and the discussion of uncertainty.*

Thanks for the suggestion but we prefer the format of one paragraph for the abstract.

• *P859 L21: I find the wording of this sentence odd; perhaps 'N₂O has the highest emission uncertainty of all the gases in the inventory' or similar.*

The sentence has been reworded and now states “Of all the gases in the inventory, N₂O has the highest emission uncertainty.”

• *P859 L24: Table 1 does show where the emission estimates for CH₄ come from in terms of citations but it does not show that anthropogenic sources dominate, as indicated here in the text. It would be useful to add to Tables 1 and 2, or as a new table, the total emission estimates for all these categories (natural and anthropogenic) for CH₄ and N₂O.*

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.”

• *P861 L21-P862 L15: The information here is quite hard to follow and would be easier to comprehend as a table, with columns (for example): CH₄ instrument, CH₄ measurement period, N₂O instrument and measurement period, sampling heights available and used, and altitude of each site.*

Thank you for this comment. We have added Table 1 to make this information more clear and concise.

• *P862 L16-26: Why was two-hour averaging chosen? Did data analysis or a previous publication suggest no significant changes within this time, or is it rather a compromise for the amount of data that can feasibly be handled?*

The two-hour period was chosen for two reasons: (1) To minimize not only data volume but the amount of model footprints that would have to be generated and used in the inversion; (2) this is consistent with the measurement period of other measurements made on site (Medusa –GCMS system for halocarbons);

In the text on page 4 line 115, we state, “This period was chosen to minimize data volume and to be consistent with the sampling period of the halocarbon measurement system in the network.”

• *P862 L24: The abbreviation SD is used several times in the paper and not defined. Although it is relatively common it should still be defined here at the first instance.*

We have defined SD on Page 4 Line 124. This notation was implemented by the journal during the typesetting process.

• *P863 L5: Why were particles tracked for 30 days? Surely most particles would exit the UK domain and even the extended Europe domain long before 30 days.*

Due to the size of the extended domain, which will now be clear in Figure 2 (Figure 1 from original supplement), 30 days was used to ensure that particles would have left the domain. We have done tests to find out what percentage error would occur by particles remaining in the domain at the end of the simulation and found 30 days to be adequate in this domain.

• *P869 Section 5: Should be titled ‘Results and Discussion’ as no separate discussion section is included. In addition it would improve readability if Section 5 were broken up into a few subsections, according to the different topics ie. total emissions, sectoral comparison, uncertainty...*

Section 5 has been retitled Results and Discussion and subsection titles have been inserted.

• *P869 L24: How much larger were the uncertainties? It would be interesting to know quantitatively how much difference the extra site makes to the total emission estimates.*

Uncertainties were on average approximately 36% larger on UK CH₄ emissions during January-May 2013 and 50% larger on UK N₂O emissions during December 2013-January 2014, than the average of months sampled by the full network.

• *P871 L4-22: The discussion here is a little hypothetical and somewhat meaning- less. It is clear that the prior disagrees regarding seasonality, as it is annually resolved, so no information can be found from the seasonal differences to the prior. It is also well-known from countless studies that fertilisation leads to N₂O emissions, and that factors such as*

fertiliser and climate affect agricultural N₂O emissions. It would be interesting to know if fertiliser is in fact applied earlier (ie. Spring) in eastern England than in central England (summer) in agreement with the posterior modelled seasonality in the emission distribution; or whether climate such as rainfall patterns may be able to explain the different seasonality between these regions in the posterior. This may provide new information on whether the seasonality seems dominated by fertiliser or climate for the modelled years in the UK.

We agree with the reviewer that it is well known that fertilizers are linked to N₂O emissions. However, the seasonal patterns of fertilizer application in the UK are not necessarily well known, and unfortunately, fertilizer application information (type and rate used) in the UK is proprietary information that was not available to be used as part of this study. There is limited information released on ‘recommended’ fertilizer application – type and rates based on crop type, however, extrapolating this to real concentration data is challenging – the meteorological conditions of the two years of this study have been quite different, with 2014 being one of the wettest on record with significant portions of the UK being flooded and this would likely impact when and how fertilizers were applied, that would deviate from the ‘recommendation’. We very much agree with the reviewer that disentangling the sources of the variations in the UK, spatially and temporally, through the use of process-models and/or regression analyses is very interesting and very important, and this is the subject of future work.

We have added on page 12 line 378, “Further elucidating the drivers of this seasonality requires process or empirical models of N₂O production.”

• *P871 L24: It is difficult to see if this is true from the referenced figures, as the uncertainties in the figures are relative to the median emissions. In fact it looks like emission uncertainty is lowest across Ireland and south-east England, and quite high around Mace Head and Tacolneston, but perhaps this is due to the magnitude of emissions as well. It may be useful to include a fourth panel to each figure showing the absolute uncertainty in the posterior emissions, or the uncertainty reduction relative to the prior uncertainty.*

A panel (d) has been added to Figures 4 and 5 that shows uncertainty reduction from the prior. This panel will further indicate the regions that the observations are most sensitive toward.

• *P872 L24: Insert a line break and start a new paragraph before switching the discussion from CH₄ to N₂O.*

We feel that this paragraph is about the uncertainties derived for both gases, as we compare the two together to gain insight into the sources of discrepancy. For this reason, we feel it is best to remain in the same paragraph.

• *P872 L24-27: The ratio between uncertainty at RGL and at TAC is ~0.78 for CH₄, while for N₂O it is ~0.80 (as far as I can tell from the figure). It is therefore not really true that the uncertainties are similar for N₂O and different for CH₄ at the two sites. I would say the uncertainty is higher at TAC for both gases. This may even suggest that it*

is model error rather than unresolved emission processes - opposite to what the authors propose at L26-27.

The ratio of RGL to TAC is 0.78 for CH₄ and 0.95 for N₂O, so there might have been an error interpreting the numbers from the figure.

• P874 L16: The inclusion of isotope measurements was not discussed in this paper at all. It would of course be interesting to have a discussion of how much isotope measurements may improve results in the current set-up, although it may be beyond the scope of this paper. Otherwise, a paper showing that isotope measurements can improve modelling results should be cited (eg. Rigby et al. 2014?).

The analysis of the additional constraint provided by isotope measurements is beyond the scope of the paper because these measurements do not currently exist. However, we have included the citation for the Rigby et al., 2012 publication showing the value of isotopologue measurements for source partitioning.

• Conclusions: Other changes mentioned throughout the results should be reflected in updated conclusions.

- Ireland emissions are included in conclusions.
- Regarding natural sources: “The small natural sources in the UK are not likely large enough to account for the full discrepancy between the prior and posterior emissions”
- NAEI has been changed to anthropogenic inventories to include Ireland in the conclusions

• Figure 1: I would find it useful to see the total emission distributions for CH₄ and N₂O, as well as the major sectoral emissions.

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 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.

Referee # 3:

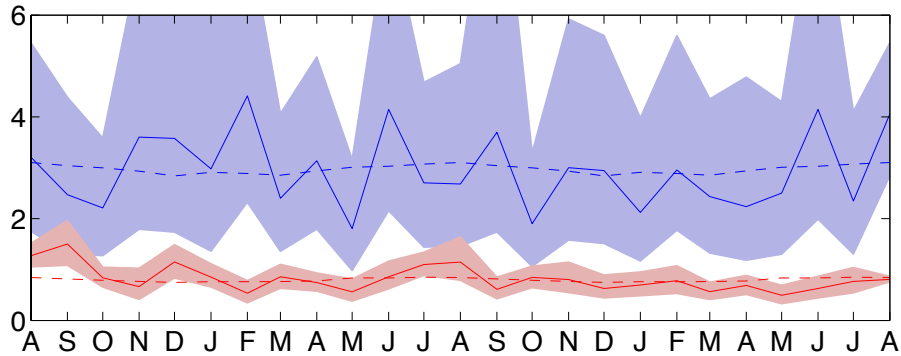
We thank the reviewer for the thorough comments, which we address below. Reviewer comments are italicized followed by our responses. Page and line numbers correspond to the revised manuscript.

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 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.



CH₄ emissions (Tg/yr) using Mace Head observations only. Posterior (solid), prior (dashed), UK (blue), Ireland (red). Uncertainties indicated by shading.

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⁻¹ prior total emissions for the UK, while reported anthropogenic emissions are 2.4 Tg CH₄ yr⁻¹. I assume that the difference of ~0.7 Tg CH₄ yr⁻¹ 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. 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 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.*

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:

- Title now includes Ireland
- Table 4: Percent contribution of a priori sources to Ireland emissions
- Results include Ireland emissions
- 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’ -> ‘total emissions’

‘emissions’ has been changed to ‘total emissions’

Introduction, page 859, line 5-6: ‘global warming potentials over a 100 year time horizon 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⁻¹ of CH₄ with an uncertainty of 20% and 0.116 Tg yr⁻¹ 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.

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⁻¹ (Prather et al., 2012)': The unit here is Tg-N yr⁻¹ for N₂O (and not Tg N₂O yr⁻¹ 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⁻¹ 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⁻¹ CH₄ and 0.070 (0.055–0.090) Tg yr⁻¹': 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⁻¹) 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 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⁻¹ for CH₄ and 0.07–0.17 Tg yr⁻¹ 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⁻¹ for CH₄ and 0.07–0.17 Tg yr⁻¹ for N₂O, 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.”

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 185m.a.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 ?

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 0.3ppb N₂O.” We changed the word 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 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, 'Matérn covariance function': would be useful to give a reference for the Matérn 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 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 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⁻¹ CH₄ prior emissions for the UK, while the authors seem to use the reported emissions of 2.4 Tg yr⁻¹ CH₄ as prior for the anthropogenic emissions. Therefore, I assume that the prior natural CH₄ emissions are about 0.7 Tg CH₄ yr⁻¹ ? Likewise, also the N₂O emissions shown in Fig. 2 (~0.14 Tg N₂O yr⁻¹) seem to include a significant contribution of natural sources (as reported N₂O emissions are 0.116 Tg N₂O yr⁻¹).

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

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 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.

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 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.