Atmos. Chem. Phys. Discuss., 15, C274–C281, 2015
www.atmos-chem-phys-discuss.net/15/C274/2015/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #3

Received and published: 18 February 2015

The discussion paper of Ganesan et al. presents an inverse modelling study, estimating CH_4 and N_2O emissions from the UK and Ireland for a 2 years period (August 2012 - August 2014). The study builds on previous work reported by Manning et al. [2011] (and earlier work) based on the NAME model, but uses an improved methodology (hierarchical Bayesian inverse framework), which includes the optimization of prior uncertainties and correlations, aiming at an optimal statistical consistency of the inversion. A futher novel element of the study is the use of observations from 3 new monitoring stations in the UK (Ridge Hill, Tacolneston, Angus).

Overall the paper is well presented and clearly written. However, I have several comments, which should be addressed before publication in ACP:

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.

(2) The paper should discuss more clearly the impact of natural emissions. The authors state that their results yield CH_4 and N_2O 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 $\sim$3.1 Tg CH_4 yr^{-1} prior total emissions for the UK, while reported anthropogenic emissions are 2.4 Tg CH_4 yr^{-1} . I assume that the difference of $\sim$0.7 Tg CH_4 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.

(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 CH_4 and N_2O not used in the inversion, or specific transport tracers, such as ^{222}Rn. I realize

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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.

(4) Regional scope of the paper: The paper includes a discussion of CH_4 and N_2O 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).

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

abstract, page 858, line 12: 'emissions' -> '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 (CH_4) and 265 (N_2O)).

Introduction, page 859, line 9-11: 'legally binding target to reduce the country's CO_2 equivalent emissions to 80% of 1990 levels by 2050.' The target is to reduce CO_2 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.'

Introduction, page 859, line 19-20: 'In 2012, the UK reported 2.42 Tg yr^{-1} of CH_4 with an uncertainty of 20% and 0.116 Tg yr^{-1} of N_2O with an uncertainty of 69%': would

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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).

Introduction, page 859, line 24-27: 'The principal sources of CH_4 in the UK...' I would suggest to modify to: 'The principal anthropogenic sources of CH_4 in the UK...'

Introduction, page 860, line 20: '554$\pm$56 and 15.8$\pm$1.0 Tg yr^{-1} (Prather et al., 2012)': The unit here is Tg-N yr-1 for N_2O (and not Tg N_2O yr^{-1} which is otherwise used in this paper'. Please check the number: In table 1 of (Prather et al., 2012) a value of 15.7$\pm$1.1 Tg-N yr^{-1} is given.

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} CH_4 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 CH_4 (1.72-2.47 Tg CH_4 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)).

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 CH_4 and 0.07–0.17 Tg yr^{-1} for N_2O, 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 CH_4 and N_2O emission estimates than the other 3 models used in this comparison.

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)).

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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.

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

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 representatitve (less effected by local emissions and better represented by atmospheric models).

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

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.

Measurements, page 862, line 24-25: 'Typical instrumental uncertainties were 10 ppb CH_4': 10 ppb as instrumental uncertainy of a Picarro instrument seems very high - should be typically in the order of 1 ppb or better.

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 boudary conditions, but also on the emissions derived for continental Europe. This requires some discussion in the paper.

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 ?

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

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

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.

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.

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

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

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)).

Results, page 869, line 19-20, 'Both UK CH_4 and N_2O 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 $\sim$3.1 Tg yr^{-1} CH_4 prior emissions for the UK, while the authors seem to use the reported emissions of 2.4 Tg yr^{-1} CH_4 as prior for the anthropogenic emissions. Therefore, I assume that the prior natural CH_4 emissions are about 0.7 Tg CH_4 yr^{-1} ? Likewise, also the N_2O emissions shown in Fig. 2 ($\sim$0.14 Tg N_2O yr^{-1}) seem to include a significant contribution of natural sources (as reported N_2O emissions are 0.116 Tg N_2O yr^{-1}.

Results, page 870, line 6, 'oceanic sources of N_2O': Explain, how oceanic / offshore emissions are attributed to the UK (or other countries) ?

Results, page 870, line 9ff, 'fractional uncertainties' / Figures 3+4: would be useful to

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include also uncertainties of the prior.

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 CH_4 emissions? It would be useful to include also a map with the assumed prior natural emissions (e.g. in Figure 1).

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).

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 CH_4 and N_2O not used in the inversion, or specific transport tracers, such as ^{222}Rn (see general comment (3)). This should be included in the discussion.

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).

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

Table 1/2: - give numbers for annual (a priori) CH_4 and N_2O emissions of UK for dif-

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ferent categories. - In addition, it would be useful to list here also the emissions of Ireland.

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)

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)

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