Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-1187-RC2, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimates of sub-national methane emissions from inversion modelling" *by* Sarah Connors et al.

Anonymous Referee #2

Received and published: 26 February 2019

The manuscript "Estimates of sub-national methane emissions from inversion modelling" by Connors and co-workers presents atmospheric inverse modelling results from a very dense methane surface observation network in the eastern UK, quantifying local to regional sources and investigating the impact of network density. In terms of spatial measurement density and targeted local point sources the study offers some new insights into the feasibility of inverse modelling at this scale. My main concern (details below)with the present study is the use of unfiltered data for the inverse modelling and the use of a less well controllable inversion system. Hence, I suggest some major revisions to the manuscript before it can be published in ACP. Otherwise the manuscript is well written and structured, but a number of figures need to be improved before publication as well (details below).

Major comments

Use of unfiltered observation data in the inverse modelling

The tree additional CH4 observing sites setup for this study have sampling heights between 10 and 25 m above ground. Although the authors indicate that local sources should be small (a notion that is not well documented) one can see considerable concentration increases at all 3 sites during nighttime stable boundary layer conditions (Fig 5). It is well known that atmospheric models face serious challenges in stable boundary layers. Hence, it is not surprising that there exists a large discrepancy between observations and simulations (both using NAEI emissions and optimised a posteriori emissions) for nighttime observations. The comparison during daytime is generally much better. The problem is less evident for the higher sampling height of Tacolneston. Other inverse modelling studies have often used filtered observational data, excluding nighttime stable conditions, to rule out that biases in the transport model impact the emission estimates. Why was this not done or at least explored in the current study? I strongly encourage to explore the impact of observation filtering on the emission results, especially also in the light of the large variability seen in the results when only sub-sets of sites were used.

Inversion scheme

It seems a bit of a shame that the authors chose to use the InTEM2014 inversion system for their main analysis, since the InTEM2018 Bayesian inversion system would have allowed for a more stringent analysis of uncertainties in the inversion. Since InTEM2014 does not prescribe a priori emissions and their uncertainties, it remains unclear how the final uncertainties in InTEM2014 were derived and how representative they are of the real uncertainties. In addition, a fully Bayesian approach could have dealt with what is described by the authors as 'dipole effect', by introducing spatial correlation in the a priori covariance matrix. Another problem of the annealing approach and the used cost function seems to be over-fitting of the data. Without a consistent

uncertainty budget the method seems to be vulnerable to over-fitting and hence produces dipoles but possibly additionally noisy emission fields. Although the authors explain that InTEM2014 was ready to use in their group, it seem one could have learned a bit more with InTEM2018. Furthermore, some InTEM2018 results are presented in section 3.5 anyway. So why not use it for the main analysis as well?

Minor comments

p1,l33: What does 'similar methane estimates' mean in terms of percentage differences? Next sentence mentions 'good agreement' as about 5 %.

p2,l5: The sentence should mention when this rise started and that there was a decade of stabilised CH4 concentrations before. There is another recent publication that should be included in the list: Thompson, R. L., Nisbet, E. G., Pisso, I., Stohl, A., Blake, D., Dlugokencky, E. J., Helmig, D., and White, J. W. C.: Variability in Atmospheric Methane From Fossil Fuel and Microbial Sources Over the Last Three Decades, Geophys. Res. Lett., 45, 11,499-411,508, doi: 10.1029/2018GL078127, 2018.

p2,l9: Maybe one can say that emission reductions are feasible. However, more interesting would be a statement why such reductions are probably more feasible or easier to achieve than reducing CO2 emissions of a similar magnitude.

p2,l13: I would reformulate this sentence towards something that states that knowing what causes and where emissions occur allows to design efficient reduction strategies. Quantifying emissions with atmospheric observation offers independent validation/support tool to assess if reduction measures were successfully met.

p2,l24: Is the 40 % uncertainty of the NAEI on the 1-sigma or 2-sigma confidence level?

p2,l30: There are several other CH4 inverse modelling studies on the national or subnational scale. Not just the ones for the UK.

p3,l3: Why are only preliminary findings presented? Only final results should be pub-

lished in a peer-reviewed manuscript! I suggest rewording.

p3,l22f: Why would that be the case? Also in an area with homogeneous emissions 4 sites at different locations and different meteorology would experience different concentrations and the challenge for the transport and inverse modelling system would largely be the same. What is more interesting from my perspective is the presence of different dominating emission sources in the domain. This allows the inversion system to pick up problems of the inventory in terms of biased emission factors for different processes. Something which is also discussed later by the authors.

p3,l32: What about other local sources typical for rural environments? Any livestock in these towns? Waste water treatments?

p3,l33: Any coastal wetlands or marshes nearby?

p4,l5: Can this number also be given as a mole fraction? 0.3 % of 1900 ppb? So in the order of 5 - 6 ppb?

section 2.1.3: How often was a calibration gas applied? Was this a one point calibration or multi-point calibration? Was an independent target gas used to estimate the performance of the calibration?

p6,l16f: How can the 100 m sampling height be justified at this scale? With a release at 100 m (Tacolneston) and a sampling height of 100 m one will get source sensitivity directly at the receptor, whereas in reality and especially during stable conditions I would expect a plume to take some time and distance before actually touching the ground. Usually this distance should be short compared to the grid scales of the atmospheric inversions, but here this grid scale is in the order of a few km only.

p6,l25 and elsewhere: Usually the term 'footprint' or 'sensitivity map' is used in this context. 'Dilution map' seems to suggest something else. It should also be mentioned that the figure presents average conditions whereas hourly 'footprints' are the ones that are used for the inversion.

Equation 1: Bit of a poor layout for an equation. Please use a more mathematical notation and explain units in text. There is also a sum over the domain required to yield the concentration at the receptor!

Equation 2: It remains unclear why one would need a simulated annealing method to solve for the minimum of equation 2. There should be a very straightforward analytical solution for this! Why does r have an index i if the sum runs over i? Does i run over all observations from all sites or just over time? Why does x have an index i? x is the state vector that does not change with time. K should have an index i or maybe one could write $(Kx)_i$. Also the text says that 'xi' is the measured concentration. That is wrong!

p8,l9f: Are individual uncertainty terms added directly or is a sum of squares used (which would be more appropriate)? What is the final average uncertainty? How does it differ for the different sites? This is important to understand if a given site has more influence on the results than others.

p9,l18: What does stable emissions mean in this context? A posteriori emissions did not change with the choice of percentile threshold?

p9,l18: How is the 'cost score of the baseline' derived? Is the baseline part of the state vector? Or does it remain unchanged?

p10,l15: What is the cost score? r in equation 2?

p11,l4: Give uncertainty estimate for NAEI value here as well. Somewhere it said +/- 40 %. So 112 kt/yr? Or at least since the 40 % was given for the national total. Same question again: What is the confidence level of the uncertainties?

p11,l8: What is WindTrax modelling? Not clear if one does not want to read the reference. It would call it a local scale Lagrangian particle dispersion model.

p11,l8: "high point source emissions near Haddenham". Actually the large point sources around HD seem to be surprisingly well resolved by InTEM and NAEI. I am more concerned about the large emissions in InTEM east of TN. Could this be wrong

attributions from sources outside the UK (for example Benelux region)?

p11,l13: What are the units of the given standard deviations? Looks like these are values from Table2. So mass emissions? Giving a relative uncertainty would make more sense when comparing regions with strongly different total emissions.

p11,l14f: Unclear why a 'footprint radius' of 50 km would be implied.

Figure 6: It would also be interesting to see a difference map between InTEM results and NAEI inventory.

p12,l1: How comparable is a study of landfills in Taiwan with conditions in the UK? Environmental factors will play a large role in the decay processes in a landfill. These factors appear to be quite different between UK (temperate climate) and Taiwan (tropical). Also the question of how much and which kind of organic matter was initially deposited in the land fill, will play a role. Is there no similar study from a European site?

p12,l4f: This looks more like the emissions are less well allocated in InTEM compared to NAEI. The point sources east and north of HD are less intense in InTEM and may be wrongly allocated to the larger grid cells labeled 2. What are the total emissions for a region around the cells labeled 2 but including the point sources north and east of HD? I would expect that the total may be much more similar.

p13,l5: Are these irrigation or drainage channels? How are the managed fenlands used? Rangeland, crop agriculture? If the latter dominates it is likely that these lands are usually well drained and no large emitters of CH4. How large are the unmanaged fenlands in comparison? An inversion grid structure that would reflect different dominating land cover types could have helped to distinguish different source processes.

p13,l19: This does not fit to the area labels in Figure 4! Why were only these areas considered for the sensitivity analysis?

Section 3.4: The large impact/bias introduced by the Haddenham observations could

be a result of poor representation of the nighttime observations (see Fig. 5). I wonder if these results would be more robust if only daytime observations would be used. Also see major comment above.

Figure 9: Please use the same value range for both sub-panels in order to make them comparable. Alternatively, a difference plot would also emphasise the important details. Could the NSC area, as used for table 4, be outlined in the figures?

p16,l26: This (uncertainty of baseline) has not been discussed anywhere above. How do we know that this a major source of uncertainty? How is it quantified? Only an assumption is made for the baseline uncertainty (5 ppb) but there seems to be no justification of this value.

p16,l27: But that is exactly what was done in section 3.5. So why not use this as the main analysis using InTEM2018 instead of the older version?

p16,l32: 'finer resolution' This is not visible in Fig 9. which seems to show the same spatial resolution for both InTEM2018 runs, with our without EA sites.

Technical comments

Figure 1: Improve resolution of googleMaps image. Latitude and Longitude axis labels should also contain units.

Table 1: Units for longitude and latitude.

p5,l2: 'psig' not SI units.

Figure 6 and 7: The colours on the map are not very clear (line patterns). Looks like something went wrong during conversion to pdf.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1187, 2018.