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Anonymous Referee #1

Received and published: 9 January 2019

1 Overview:

Review of "Estimates of sub-national methane emissions from inversion modelling" by Connors et al.

Connors et al. present an analysis from a year of methane measurements at 4 sites in East Anglia. They describe the 4 new instruments that are mounted on churches or other tall towers. These instruments are then used to constrain methane emissions in between June 2013 and May 2014. The description of the network is generally good and the figures are all reasonably well made. However the inversion portion of the manuscript needs quite a bit of work. I have some major concerns with the seemingly unsubstantiated choices, poor description, and potential overfitting. I think there is interesting data here that should eventually be published, but there are some major issues that need to be dealt with first.

2 Major Comments

The section on inversion modelling (Section 2.2) could use a major re-write to describe what was actually done and justify choices made.

2.1 Over-fitting?

A **major** drawback with a least squares cost function is over-fitting. This is precisely why most inversions use a regularization or a prior. I don't see any discussion of overfitting. How do the authors combat over-fitting? I strongly suspect this is why they find a 'dipole effect' (see Minor Comment #1).

Response:

Thank you, we acknowledge the limitation of the least squares cost function. However there are also limitations with using a prior with high levels of unknown uncertainty, particularly at such fine spatial resolution on the sub-national scale. Therefore, in this study we are assuming that the prior uncertainty is so high as not to add information to the inversion system. It is also important to recognize that if one uses prior information then the result is not independent of the prior and therefore cannot be used to verify the prior emissions, which is what we wanted to do in this study. Overfitting is definitely a potential hazard that is why the inversion was repeated with many different settings to explore the uncertainties in the system: these are discussed at length in Connors, PhD Thesis, 2015.

Please see Minor comment #1 response for discussion on the 'dipole effect'.

2.2 Simulated Annealing

2.2.1 Use of Least-Squares and Simulated Annealing?

A least squares cost function (also known as a maximum likelihood estimate), as the authors use, has a closed form solution for the optimal solution. Using the author's notation, the optimal solution ($\hat{x}$) would be:

$$\hat{x} = \square K_T \_^{-1} \_ K \_^{-1} K_T y_0. \quad (1)$$

where K is the dilution matrix, __ is the error covariance matrix, y_0 is the vector of observations after removing the background concentrations: $y_0 := y - b$.

There are four cases I could envision using something like Simulated Annealing:

1. if the system is non-linear (e.g., if K is a function of x)
2. to regularize the solution (e.g., by coarsening K as part of the inversion)
3. there are additional constraints being applied (e.g., non-negativity)
4. for computational expediency

In the case of additional constraints (such as a non-negativity constraint), it seems that bounded optimization would be preferable to a stochastic method such as simulated annealing. Gradient-based methods like L-BFGS-B ("https://en.wikipedia.org/wiki/Limited-memory_BFGS#L-BFGS-B") tend to be much faster for convex optimization problems such as this one. A stochastic method like Simulated Annealing would probably be better for a non-convex optimization problem.

Response:

Thank you for this comment, we acknowledge that the simulated annealing method is not the fastest for convex optimization and we certainly do not want to claim that this is the most efficient solution method. At no point in the manuscript have we stated this. Simply, this is one method that can be used whilst imposing the constraint of a non-negative solution.

2.2.2 Error statistics from simulated annealing

Regarding the use of Simulated Annealing, it's unclear to me why the authors chose to use a technique like simulated annealing here. Simulated annealing is an optimization technique that is quite efficient, so it works rather well in high-dimensional

problems. However, the samples obtained from Simulated Annealing are inconsistent with the true posterior statistics (the uncertainties will be smaller that the true uncertainties).
So reporting error statistics from Simulated Annealing strikes me as dubious at best. Using something like an adaptive MCMC or a reversible jump MCMC (rjMCMC) as some of the co-authors here have previously done seems far superior to simulated annealing.

Response:

This comment questions the choice of the solution method commenting on its inefficiency (see previous response) and ability to resolve all uncertainties. We agree with the reviewer that the error statistics are a weakness in the study and we have stated this in the discussion. However, even MCMC methods cannot claim to capture all uncertainties and they are still limited by the posing of the uncertainties attached to the observations, transport modelling and prior, and have equal problems dealing with biases.

Related to this, it's entirely unclear why Manning et al. (2003) would be the citation for simulated annealing on Page 8, Line 26. This is not a technique or method that Manning developed. For example, the Machine Learning textbook from Christopher Bishop (Bishop, 2007) would be a much more appropriate citation for someone interested in how Simulated Annealing works.

Response:

Thank you for the suggestion but our method originates from the 'Numerical Methods' discussed in Manning 2003, so we have not changed the reference.

2.3 Section 2.2.3: A priori emission estimates

It's not clear why this section is necessary at all. As mentioned above, if the system is linear (the dilution matrix is predetermined and does not change) and the authors choose a least-squares cost function then the solution can be directly computed. There would be no need for a "random, non-negative emission field". How is this being used? Is this the initial starting point for the simulated annealing? If so, then it should be referred to as such and probably not be discussed in a 2-sentence section on prior emissions.

Response:

Thank you for this point and suggestion. Yes, the subsection describes the initial starting point of the simulated annealing. To aid clarity and minimize any confusions, the subsection has therefore been removed and the following sentence has been added to Section 2.2.2 InTEM$_{2014}$ inversion model:

"The initial starting point for the simulating annealing process is a random, non-negative emission field, which assumes no a priori knowledge of the location or magnitude of emissions."

2.4 Regarding the resolution of the solution grid

There has been an abundance of work looking at how to define a multi-scale state vector. The textbook from Rodgers (2000) talks about this. Bocquet et al. (2011; QJRMS) is entirely devoted to this topic. Other work like Turner & Jacob, (2015; ACP), Lunt et al., (2016; GMD), Henne et al. (2016; ACP), and Bousserez & Henze, (2017; QJRMS) also talk at length about how to construct this multiscale formalism. Briefly, by coarsening (or restricting) the grid you are applying a hard constraint on the inversion. Basically, sub-elements are no longer allowed to vary independently. This is a form of regularization. In the most extreme case you could coarsen to a single state vector element. The degree of coarsening can change the problem from an under-determined problem to an over-determined problem. This ties back to an earlier question, how do you deal with potential over-fitting? Was there any cross-validation done? At the bare minimum, Bocquet et al. (2011) should be cited.

Response:

Thank you for the suggested citations and the introductory text on calculating this multi-scale vector / solution grid. Bousquet et al., (2011 ; QJRMS), Lunt et al., (2016; GMD) and Henne et al. (2016; ACP) have been added to the text. Although not shown in this manuscript multiple sensitivity runs were conducted varying the solution grid resolution. From a coarse solution grid with only 15 boxes (based on Figure 4a) to a more spatially fine resolution containing ~250 boxes. A short description of this analysis has been added to the text. This is also discussed in Connors, PhD Thesis, 2015 (add to the text).

2.5 Computing the Baseline

This section needs more work. Computing the baseline is a non-trivial task for any regional inversion and many studies are devoted entirely to estimating the baseline. At the bare minimum, the authors should provide some justification for their choice of the 18th percentile. Other studies such as Henne et al. (2016; ACP) provided an extensive analysis on the choice of background including precomputing it (as Connors et al. have done) vs jointly estimating it as part of the inversion.

Response:

We agree with the reviewer that this section should be developed and we apologise for this not being present in the submitted manuscript. The section has been edited and expanded. We hope this is now to the satisfaction of the reviewer. Please also see that the discussion has been expanded to discuss the limitations of the baseline more fully.

2.6 Evaluation of meteorology?

I didn't see any mention of evaluating the meteorology. This is a crucial step in atmospheric inversions using real measurements that seems to be missing.

Response:

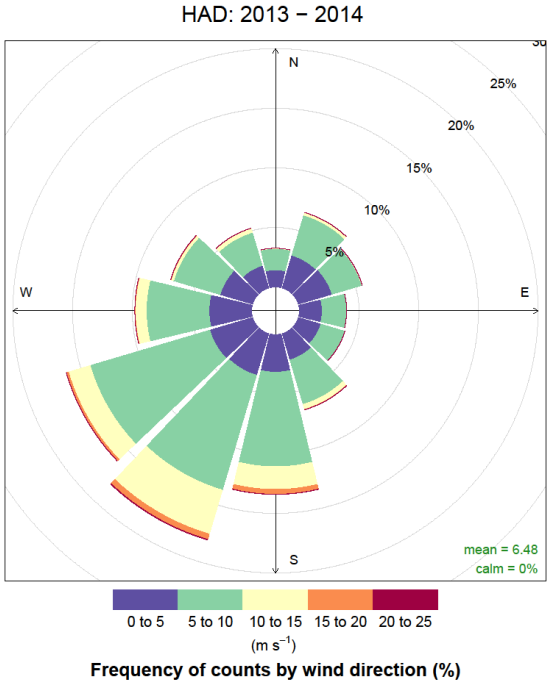
We have added the following sentence to the manuscript: To explore the sensitivity of the inversion results to the uncertainties in the 3-D time-varying modelled meteorology, inversion analysis can be conducted using different numerical weather prediction model meteorologies. This sensitivity analysis was beyond the scope of the work presented here.
To our knowledge, there are no other UK NWP meteorology data available at 1.5km spatial resolution, so a like-for-like comparison would not be possible. A comparison between 1.5km and 25km resolution in the NAME model was undertaken in Connors, PhD Thesis, 2015, but this sensitivity was not conducted with InTEM. One of the reasons East Anglia was purposefully chosen for this study was its flat orography, where the meteorology is easier to model, and so associated meteorological uncertainties were minimised. We have added a reference on the 1.5km spatial resolution to the manuscript (Tang et., al., 2013) in Section 2.2.1 on page 7 and Figure 3 caption on page 8 of the track changed manuscript.

2.7 Figure 3 seems odd

The dilution matrix as Connors et al. refer to it (also commonly known as the footprint matrix, transport operator, Jacobian, etc.) looks pretty Gaussian. Is there no dominant wind pattern at Haddenham? I would usually expect some dominant wind pattern (i.e., more sensitivity to the upwind region). A wind rose showing that, indeed, the winds are roughly uniformly represented from all sectors here would be useful. Otherwise, turning this into a 3 panel figure (current figure as a large column on the left and two subpanels on the right column) with two illustrative examples of dilution matrices from two days. I would expect the illustrative days to show strong sensitivity upwind of the site for that day. Basically, I'm curious if this was computed correctly.

Response:

Thank you, we have checked this and the figure calculation is correct. There is a prevailing wind direction as can be seen in the SW bias in the footprint. (The log-scale in the plot does obscure this somewhat but is needed to show the full range of particle densities.) To illustrate this, we show a windrose of the met data used in Figure 3 to show the prevailing south-westerly wind direction. We do not feel that the addition of this figure substantially increases the value of the paper, the subsection it appears in focuses on the footprint matrix description for its use in the inversion model and is not a discussion of the meteorology experienced at the site. To aid clarity, the following sentence has been added to *the figures caption, which now more clearly states that the figure shows an annual average, whereas in the inversion timestamps of 1 hour are used (also in response to reviewer 2's comment on this): "Figure shows the averaged footprint map over one year whereas timestamps of 1 hour are used as input data within InTEM2014."*



2.8 Agreement with NAEI:

The authors mention good agreement with NAEI (to within _5%). However, Figure 6 looks strikingly different. Is this because they've further coarsened the emissions before this comparison? I don't see how they are getting a 5.

Response:

The 5% refers to the comparison between the county level estimates of Norfolk (region 1) and Suffolk (region 2). When aggregated, these areas are within 5% of each other. This is not the case for the county of Cambridgeshire (Region 3), where the text states there is over 20 % difference in the estimates. Please note, the scales in Figures 6 and 7 are logarithmic. We have added an extra reference in the text to Table 2 where the 5% value is given.

3 Minor Comments

3.1 Dipoles and overfitting in the solution

The authors discuss a 'dipole effect' (e.g., Page 11, Line 21) in the inversion results. These are common in solutions with overfitting. The inversion is attempting to fit a high value at a measurement site, so it inflates the emissions to a very large value at that one location and then compensates by reducing the emissions in a neighboring grid cell where the observations have weak constraints. Basically, this is what happens:

• Location A: concentration too low -> increase emissions in just this location

• Location B: no constraint on concentration, domain wide emissions too high because of Location A -> reduce emissions. This is combatted in most inversion systems by having a prior or regularization that includes some off-diagonal relationships (e.g., emissions from Location A and Location B should be weakly correlated).

Response:

Thank you for this comment. Neither $InTEM_{2014}$ nor $InTEM_{2018}$ have non-negative off-diagonal elements in the prior uncertainty matrix due to the large variability in emissions from grid box to grid box in the NAEI. At 1 km resolution, the NAEI can have occurrences of very large emissions next to areas of very low (for example, landfills in the waste sector part of the inventory) because of the different sectors it is trying to represent. Therefore, strong variability of emissions from grid box to grid box is possible. Investigating this was one of the aims of the study. By making the assumption that adjacent grids have emissions that are correlated you are adding information to the system, but where does this knowledge come from? Priors such as the NAEI can be averaged out to some degree, for example as the location of agricultural emissions can be imprecisely known. But, ideally, this should be dealt with by testing the impact of merging the dipole grids, i.e., is this dipole true or is it an artifact? Inversions using different grids were undertaken in Connors, PhD Thesis, 2015. *Further analysis could help investigate this, but we consider this beyond the scope of this manuscript.*

3.2 Suggestion for title

It would be nice if the authors specified where the work was done in the title. I'd suggest adding "in East Anglia" or "in the United Kingdom" (or something to that effect). Maybe something like: "Estimates of sub-national methane emissions from inversion modelling in East Anglia"

Response:

Thank you, we agree and have edited the title to now read 'Estimates of sub-national methane emissions in the United Kingdom using inversion modelling'.

3.3 Proof of concept

On Page 3, Line 1 the authors motivate the work as a "Proof of Concept" that inversion schemes can work at sub-national scales. Although this has been shown numerous times in the past. Examples include Scot Miller's 2013 PNAS paper for methane in the US, Stephan Henne's 2016 ACP paper for methane in Switzerland, work from Kathryn McKain on methane emissions in Boston, work from Ken Davis' group on urban inversion modelling in Indianapolis for CO_2, and work from Thomas Lauvaux on CO_2 at urban scales. So I don't find a "Proof of Concept" to be a particularly compelling motivation. The work is definitely interesting, but I don't think this should be a major motivation for the reader.

Response:

Thank you we have taken this into account, the 'proof of concept' sentence has been removed from the paragraph. The two studies that focus on methane suggested by the reviewer have been incorporated into the introduction when citing studies which estimate methane using inversions across different spatial scales.

3.4 Table 1:

The instrument acronyms are rather confusing in Table 1. I would remove "UCAM" and "UEA" from the table and instead add a different column that lists the sampling rate. Alternatively, the authors could have footnotes under that table that explain the acronyms. As it stands, the reader needs to scan the text to try and figure out what the acronyms mean.

Response:

This has been taken into account. The following footnotes have been added:

[1]UCAM refers to the GC-FID instrument installed by University of Cambridge, with a sampling rate of 1-2 minutes.

[2]UEA refers to the GC-FID instrument installed by University of East Anglia, with a sampling rate of ~20 minutes.

3.5 "Pseudo-observations":

I would avoid using the term "pseudo-observations" because it sounds like the authors are doing a synthetic-data study (i.e., an OSSE). "Simulated", "modelled", or "predicted" concentrations would be preferable.

Response:

We agree, occurrences of pseudo have been replaced with simulated.

3.6 Equation #1:

Do not present an equation in this form. Use this equation to introduce your nomenclature for later.

Something like this would be preferable:

$y = Kx + b$ (2)

where y is an n × 1 vector of concentrations (units: g m^{-3}), K is an n × m dilution matrix (units: s m^{-1}), x is an m×1 vector of gridded emissions (units: g s^{-1} m^{-2}), and b is an n × 1 vector of concentrations upwind of the modelling domain (units: g m^{-3}).

Response:

We agree, thank you, the equation has been changed.

3.7 Table 3:

"Simulated annealing" is not a cost function type. The cost function is a least-squares or maximum likelihood estimate.

Response:

We agree, thank you, the typo has been changed.

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.

Thank you for this very valid question. We acknowledge that including unfiltered observations is not a common approach in inversion modelling but we have several reasons why we did this. Firstly, all observations contain information and thus have the potential to inform the inversion. High measurements recorded at night when boundary layers are low can be from nearby sources. As our study was an attempt to run inversion modelling at high spatial resolution, using 1.5km meteorology, we wanted to include this information in our runs. However, as the reviewer rightly points out larger meteorological uncertainties exist at low boundary layer, nighttime periods. For this reason, we applied a de-weighting uncertainty estimate on observations that varied with time and at each individual site. The standard deviation of the hourly measurement was included as an uncertainty estimate (stated in section 2.2.2 on page 9 of the revised, tracked-changed manuscript.) During the night, when measurement values would vary more widely, these measurements would be assigned higher uncertainties. Additionally, we purposefully chose East Anglia, an area of flat orography, to reduce errors in the meteorological data and we used data at a much finer spatial resolution (1.5km compared to the global 25km UM data). Finally, we attempted to locate the measurement sites not immediately close to large sources of methane (i.e. in villages off the main gas grid, away from livestock farms and at least 5km away from landfills). We acknowledge that installing sites at higher altitudes would reduce any remaining local sources and minimize resulting issues with meteorological uncertainty. Measurements were taken from existing infrastructure and we installed out sites at the highest available locations in the chosen villages (church towers) to minimize this issue as far as practically possible. We hope this explanation is sufficient for the reviewer.

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?

Response:

Thank you, we acknowledge the limitation of the least squares cost function however there are limitations with using a prior with high levels of unknown uncertainty, particularly at such fine spatial resolution on the sub-national scale. Here we are assuming that the prior uncertainty is so high as not to add information to the system. It is also important to recognize that if one uses prior information then the result is not independent of the prior and therefore cannot be used to verify the prior emissions, which is what we wanted to do in this study. It should also be noted that InTEM2018 was not available at the time of this work and that it is still not set up for such fine spatial scales. Thus, InTEM2014 is the more suitable for this study.

Minor comments

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

Response:

Thank you for pointing out the vagueness of this statement. The sentence has now been re-written to "Resulting InTEM$_{2014}$ methane estimates for the East Anglia region overlap with the NAEI when uncertainties are accounted." We hope this is more informative for the reader.

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.

Response:

Agree, the citation has been added.

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.

Response:

We prefer not to make a comparative statement on whether it is easier or not to achieve than reducing CO2 emissions, as there are substantial barriers to achieving deep reductions in methane emissions which we have not assessed. We have added a sentence on the potential for mitigation given its short lifetime. We hope this is acceptable to the reviewer. The paragraph now reads:

"Anthropogenic emissions (principally fossil fuels, agriculture and waste, and biomass burning) constitute approximately 60% of the current emissions (Saunois et al., 2016) and so there is a large mitigation potential from reducing methane emissions. While the relatively short lifetime of methane in the atmosphere compared to that of carbon dioxide gives argument for the potential to further mitigate warming in the near term, there are still sources of methane that have barriers to mitigation, for example, in the agriculture sector (Rogelj et. Al., 2018).

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.

Response:

Thank you for the suggestion. The text has been modified to "Knowing what causes and where these emissions occur allows the design of efficient reduction strategies. Quantifying emissions using atmospheric observations offers independent validation to assessing if reduction measures are successfully being met."

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

Response:

Unless we are mistaken, this is not stated in the NAEI, only that this is a UK total estimate for that year. This is an uncertainty calculated from the known uncertainties, any missing uncertainties are not included in the calculation. Uncertainties at the sub-national level will be substantially higher, as stated in the text.

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

Response:

Thank you. Two citations that focus on the subnational scale have been added: Henne, 2016 in ACP and Miller, 2013 in PNAS.

p3,l3: Why are only preliminary findings presented? Only final results should be pub-lished in a peer-reviewed manuscript! I suggest rewording.

Response:

We agree, the text has been rephrased to say 'This paper presents the results of the work and...'.

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.

Response:

Thank you, we agree with your comment and so have revised the second bullet point to "The presence of different dominating emission sources within the inversion domain. This allows the inversion system to highlight potential issues in the NAEI, for example, biased emission factors for different processes."

p3,l32: What about other local sources typical for rural environments? Any livestock in these towns? Waste water treatments?
Response:
Several sources of methane exist in East Anglia, we have added a short description of the main sources found near to the sites but a more in depth discussion on the influence of nearby sources can be found in Section 3.2.1. Our sentence in Page 3 Line 33 is to rationale why the measurement sites were not installed at ground level.
p3,l33: Any coastal wetlands or marshes nearby?
Response:
Several sources of methane exist in East Anglia, we have added a short description of the main sources found near to the sites but a more in depth discussion on the influence of nearby sources can be found in Section 3.2.1.

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?
Response:
Calibrations were done at half hourly intervals. All sites were one point calibration with the exception of the instruments at Weybourne (Site 3), which was a multi-point calibration. This information has been added to the text, however there was not an additional independent target gas used to estimate the performance of the calibration, only the cross inter-calibration experiments done between the NPL and NOAA calibration gases, which is already stated in the text.

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.
Response:
Thank you for this very valid comment. To minimize this acknowledged issue we did two things: the first was to use an average of the two measurement altitudes at Tacolneston (54m and 100m, as stated on page 4 in section 2.1.1). And secondly, by assigning uncertainty based on the variability across the two heights to the hourly averaged observations (see response to major comment) we de-weight any measurements that showed large differences in readings at the two altitudes. So, during stable boundary conditions, the two altitude measurements would likely differ more, thus the observational uncertainty would be higher and the hourly measurement de-weighted. All measurements were averaged on an hourly timescale (with roughly 30 measurements taken within an hour). Instances where the boundary layer was well mixed are likely periods when the observations within the hour show little variation and thus have low uncertainty. We recognize that this approach could have been further improved but unfortunately it is not possible to run further sensitivity analysis on this matter. We hope that our attempt to de-weight potential instances of this goes some way to mitigate the point raised by the reviewer, although it is a valid one.

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.
Response:
We agree, instances where the text refers to 'dilution' have been modified to 'footprint'. A sentence in the figure caption has been added to clearly state the figure presents average conditions whereas hourly are used in 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!
Response:
Thank you, the equation and text have been modified to:
$$y = Kx + b \quad (1)$$
where y is an n × 1 vector of concentrations (units: g m^{-3}), K is an n × m dilution matrix (units: s m^{-1}), x is an m×1 vector of gridded emissions (units: g s^{-1} m^{-2}), and b is an n × 1 vector of concentrations upwind of the modelling domain (units: g m^{-3}).

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!
Response:
Thank you for this comment. We agree with that the simulated annealing method is not the most efficient of solvers but that does not mean it is wrong. And although more efficient alternatives exist, we see no fundamental error in using this technique.

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.

Response:

Uncertainty sigma is calculated as the sum of the squares - the denominator in Equation 2 contained a typo – we apologise for this. The i in the denominator is now within the bracket. The sigma is different for each of the four sites, which will result in different cost scores for each site. The sites, Weybourne and Tacolneston had lower assigned uncertainties compared to the other two sites. A large sigma for an individual station will de-weight that particular station.

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

Response:

Thank you for pointing out the lack of clarity here. We hope the following edit is clearer and more useful to the reader: "The 18th percentile produces results with the lowest standard deviation of a posteriori emissions of all baselines tested and with the lowest cost score of all the baselines tested."

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

Response:

No, the baseline is not part of the state vector. It is pre-calculated from the observational timeseries and dilution matrix. The baseline cost score refers to resulting cost score from the InTEM$_{2014}$. This section has been edited to make this clearer in the text.

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

Response:

Yes, this is correct. This has now been explicitly the first time the term cost score is used (Section 2.2.2, page 8).

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?

Response:

The NEAI did not supply a sub-national uncertainty estimate with its dataset. It would be higher than 40% but that number is unknown. We have added a footnote (shown below) that expressed the NAEI uncertainty as 40% but also states that this would be higher but the specific value is unknown.

"Uncertainty calculated using 40% estimate provided with NAEI for the whole of the UK. Sub-national uncertainty estimates were not provided."

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.

Response:

Thank you, we agree. The sentence has been changed to now read "…but local studies using additional measurements, Gaussian plume, and local scale Lagrangian particle dispersion modelling do show that…".

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)?

Response: We do not think that this emission would be from outside the UK. Firstly any concentrations that were not removed by the baseline can be allocated into the border regions shown in Figure 4a (also please see the revised section 2.2.4 on the Baseline). Secondly, there is no land-sea mask applied in InTEM2014 (or 2018) and so the inversion is free to allocate emissions further afield if this will reduce the cost score. As the results have not done this we have no reason to believe or evidence to suggest this is wrongly allocated. Closer potential sources of methane in that area include Bacton (the port which imports natural gas from the North Sea) but this is still 40km north-east from the methane emissions shown in InTEM or Norwich, which is roughly 10-15 km north of the area. As an aside, and referring back to the earlier discussion on unfiltered data, we think that the 'surprisingly well resolved' high emissions near Haddenham may not have been seen so clearly without using unfiltered data.

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.

Response:

Thank you, we apologise for not including the units before. These (kt yr^{-1}) have been added into the text and into the headers of Table 2. Regions where methane estimates differ more are also regions where there is larger standard deviation ranges from the InTEM$_{2014}$ results, therefore making it harder to compare. InTEM$_{2014}$ resolves the Cambridgeshire region (Region 3) more stably but has a larger difference with the NAEI than compared with Norfolk and Suffolk (Regions 1 & 2).

p11,l14f: Unclear why a 'footprint radius' of 50 km would be implied.
Response:
The quantity stated in the text is a visual estimate, after the results show more consistent stable emissions in areas that have observation sites (than compared to areas further away). Inversion runs using only 1 observation site of data would also show emissions estimates from an area of roughly this magnitude, and have low emission estimates further away - although this is not discussed in this paper. As the quantity stated is arbitrary, we have changed the text to state 'local'.

Figure 6: It would also be interesting to see a difference map between InTEM results and NAEI inventory.
Response:
Thank you for the suggestion. We have added the difference plot to Figure 6. Please not the map overlap was not possible to add due to a change in licensing of the software being used.

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?
Response:
The reviewer makes a good point to question the comparability between landfill emissions in Taiwan and the UK. A further review of the literature shows this is not a highly studied area but two citations have been added with similar examples based in Sweden (Börjesson et al, 2001) and the US (Kelly et al, 2006).

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.
Response: Thank you for this comment but we would like to point out that the NAEI has at least 40% uncertainty at the national level and much higher uncertainties at the finer spatial resolution (as stated in the manuscript). Therefore assuming the NAEI is correct could be unjustified. We do not have the calculated value the reviewer requests for this area of the map but we do have the area total for Cambridgeshire (that includes the requested area from the reviewer). The emissions for the NAEI and InTEM2014 are 26.5 kt yr^{-1} and 20.5 ±2.1kt yr^{-1}, respectively, so Cambridgshire estimates are lower in InTEM than the NAEI.

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.
Response:
We thank the reviewer for the interest in this section unfortunately we are unable to answer these specific questions as we have not visited the area discussed and so we have limited information about this area of land. We have edited the text to said 'irrigation or drainage' channels to reflect this. The suggestion of an inversion grid that would reflect different land cover types has been added to the discussion section around future work.

p13,l19: This does not fit to the area labels in Figure 4! Why were only these areas considered for the sensitivity analysis?
Response:
The outer border regions shown in figure 4 are not shown in figures 6 due to uncertainty in their emissions from the baseline setup. In InTEM2014, any below-baseline concentrations are set to a value of zero, as a way of avoiding negative emissions. If a baseline is set too high it results in reducing peak sizes in the observed data and thus an under-estimation of a posteriori emissions. Conversely, a too low baseline will result in higher a posteriori emissions, particularly around the edges of the inversion domain because any signal originating from outside the inversion domain, that has not been correctly assigned to the baseline, are placed as emissions in the boxes furthest away from the observation sites. For this reason, a posterior emissions from the border regions shown in Figure 4 are always discounted in the $InTEM_{2014}$ emissions analysis (Section 3).
We apologise for this not being described in the submitted manuscript version – the corresponding section (Section 2.2.4) has been updated. The following has been added to the caption of Figure 6: The outer border regions as shown in Figure 4 are not displayed here due to baseline uncertainty as discussed in Section 2.2.4.

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.
Response: Thank you for this comment. We would like to refer in part to our response to the major comment above. We repeat that all observations contain information and as our study was an attempt to run inversion modelling at high spatial resolution, using 1.5km meteorology, we wanted to include nighttime information in our runs, but using a de-weighting uncertainty estimate to help reduce the errors that can occur from nighttime meteorology modelling. During the night, when measurement values would vary more widely, the measurements would be assigned higher uncertainties. Some basic sensitivity analysis

comparing daytime and nighttime observations within InTEM was conducted in Connors, PhD Thesis, 2015 – which showed local information being resolved using nighttime observations and more regional information from the day time observations. We used all observations within our InTEM runs for this study. As our aim for this research was to investigate spatial resolution, as well as the differences in emission estimates when altering the number of observation sites, we have not included the day vs night plots as we consider it beyond the scope of the study.

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?
Response:
Thank you for noting this. The colour scale has been modified so they are now comparable.

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.
Response:
Thank you, Section 2.2.4 on the baseline calculation has now been expanded to cover this in more detail, and in addition the discussion has been expanded to discuss limitations with the baseline more fully. The discussion text now reads: "Despite using a measurement-based approach to define the baseline, the level of knowledge of the methane concentration in the air entering East Anglia is a major cause of uncertainty in our analysis. The static value of 5 ppb to account for errors in the defined baseline is rudimentary and should vary with respect to time to reflect associated uncertainties. Approaches in which East Anglia is nested within a larger scale inversion, and thereby moving the boundary conditions further away, would be preferable (as was done in Section 2.2.5 and in (Manning et al., 2011, for example). Additionally, further work to improve the baseline calculation method could include solving for the baseline within the inversion (Lunt et. Al., 2016)."

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?
Response:
Analysis using $InTEM_{2018}$ solves on a much low resolution spatial grid compared to $InTEM_{2014}$ (25 km vs 4 km). Analysis assessing sensitivity when varying the number of observations sites was also not conducted in $InTEM_{2018}$, nor are there any plans to do so.

p16,l32: 'finer resolution' This is not visible in Fig 9, which seems to show the same spatial resolution for both InTEM2018 runs, with or without EA sites.
Response:
Thank you for pointing this out, this is an error left form a previous draft of the manuscript. The text has been removed.

Technical comments

Figure 1: Improve resolution of googleMaps image. Latitude and Longitude axis labels should also contain units.
Response:
Units have been added to the axis. A vector file will be sent with final production of the paper to improve resolution issues.

Table 1: Units for longitude and latitude.
Response:
The units have been added.

p5,l2: 'psig' not SI units.
Response:
Pounds per square inch gauge has been converted to 234 kPa gauge.

Figure 6 and 7: The colours on the map are not very clear (line patterns). Looks like something went wrong during conversion to pdf.
Response:
The lines come from how the plot is created, essentially it shows the gridded spatial resolution (from the NAME output grid) as each grid is plotted individually (with a thin boarder). As these lines are informative we see value in keeping them in the figures. For the final version of the paper we can supply the vector form of these figures (so any of the artificial irregular appearances of the lines will be removed – this is visible in Figures 6 only). A sentence explaining the lines has been added to the captions of Figures 6 & 7.

Estimates of sub-national methane emissions in the United Kingdom ~~from~~ using inversion modelling

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Abstract. Methane is a strong contributor to global climate change, yet our current understanding and quantification of its sources and their variability is incomplete. There is a growing need for comparisons between emission estimates produced using 'bottom-up' inventory approaches and 'top-down' inversion techniques based on atmospheric measurements, especially at higher spatial resolutions. To meet this need, this study presents using an inversion approach based on the Inversion Technique for Emissions Modelling (InTEM) framework and measurements from four sites in East Anglia, United Kingdom. Atmospheric methane concentrations were recorded at 1-2 minute time-steps at each location within the region of interest. These observations, coupled with the UK Met Office's Lagrangian particle dispersion model, NAME (Numerical Atmospheric dispersion Modelling Environment), were used within $InTEM_{2014}$ to produce methane emission estimates for a 1-year period (June 2013 - May 2014) in this eastern region of the UK (~100 x 150 km) at high spatial resolution (up to 4 x 4 km). $InTEM_{2014}$ was able to produce realistic emissions estimates for East Anglia, and highlighted potential areas of difference from the UK National Atmospheric Emissions Inventory (NAEI). As this study was part of the UK Greenhouse gAs Uk and Global Emissions (GAUGE) project, observations were included within a national inversion using all eleven measurement sites across the UK to directly compare emission estimates for the East Anglia Region. Resulting~~s show similar~~ $InTEM_{2014}$ methane estimates for the East Anglia region overlap with the NAEI when uncertainties are accounted. Methane emissions from Norfolk and Suffolk show good agreement with the estimates in NAEI, with differences of ~5%. Larger differences are found for Cambridgeshire where our estimate is 22.5% lower than that of NAEI. The addition of the EA sites within the national inversion system enabled finer spatial resolution and a decrease in the associated uncertainty for that area. Further development of our approach to include a more robust analysis of the methane concentration in the air entering this region and the uncertainty associated with the resulting emissions would strengthen this inverse method. Nonetheless, our results show there is value in high spatial resolution measurement networks and the resulting inversion emission estimates.

1. Introduction

Methane is a potent greenhouse gas (GHG) whose atmospheric concentration has quadrupled over the past 20,000 years and now lies well outside the variability observed over the past 800,000 years in the ice core record (Brook and Buizert, 2018). This rise became appreciable around the time of the Industrial Revolution and continues up to the present day. There is considerable dispute about what is driving the recent rise with possible causes including tropical wetland expansion, increased fossil fuel emissions, and a decrease in the atmospheric removal rate (e.g., ~~(~~Nisbet et al., 2016; Rice et al., 2016; Rigby et al., 2017; Thompson et al., 2018; Turner et al., 2017). Anthropogenic emissions (principally fossil fuels, agriculture and waste, and biomass burning) constitute approximately 60% of the current emissions (Saunois et al., 2016) and so there is a large mitigation potential from reducing~~reductions~~ in methane emissions ~~are feasible~~.

Methane has a global warming potential of 28 over a 100 year timescale (Harris et al., 2014) and will play a vital role in any attempt to limit global temperature increase to 1.5°C or even 2.0°C (e.g., Comyn-Platt et al. 2018). While the relatively short lifetime of methane in the atmosphere compared to that of carbon dioxide gives argument for the potential to mitigate warming more in the near term, there are still sources of methane that have barriers to mitigation, for example, in the agriculture sector (Rogelj et al., 2018).

~~There is thus great interest in reducing its emissions and atmospheric concentrations in the near future. An essential part of this is, first, knowing what and where the emissions are (source, location, and magnitude) and, second, knowing that these emissions are reducing.~~ Knowing what causes and where these emissions occur allows the design of efficient reduction strategies. Quantifying emissions using atmospheric observations offers independent validation to assessing if reduction measures are successfully being met. ~~Atmospheric observations are an ideal way of providing evidence for both as, through their addition into inversion methods, they can identify and quantify emission sources, which then can be monitored over time.~~

National emission inventories are produced as part of the UNFCCC process, which require nations to submit annual estimates of their GHG emissions. These contain detailed information, often available at sub-national scales, and is produced by so-called 'bottom-up' methods, which provide national inventories for multiple emission source sectors. The calculations involve using defined emission factors based on recommended values or field measurements together with activity data. The UK's National Atmospheric Emissions Inventory (NAEI - Brown et al. 2018) for methane contains annually averaged estimates on a 1x1 km or 5x5 km grid resolution. Emissions are categorised into different SNAP (Selected Nomenclature for sources of Air Pollution) sectors which include 'agric' (SNAP 10 - agriculture, forestry and land-use change), 'waste' (SNAP 09 - waste treatment and disposal), and 'offshore' (SNAP 05 - extraction and distribution of fossil fuels). While the total uncertainty for UK methane emissions in the NAEI is estimated at 40%., the sub-national scale the uncertainty is much larger. The NAEI also does not include seasonal variations or natural emissions. Inversion, or 'top-down', techniques provide an alternative way of estimating GHG emissions. Emission fluxes are estimated using atmospheric measurements and a meteorological dispersion model that can simulate source to receptor dispersion. Methane emissions have been estimated using many inversion methods at global (e.g., (Bousquet et al., 2011; Houweling et al., 2014), European (e.g., Bergamaschi et al., 2005, 2018), ~~and~~ national (e.g., Ganesan et al., 2014; Rigby et al., 2011) and sub-national (e.g., ~~(~~Henne et al., 2016; Miller et al., 2013)~~ ~~scales. These approaches provide an independent way of checking the national inventory totals and can assess emission changes over varying timescales (Brown et al., 2018).

This study, performed as part of the Natural Environment Research Council's Greenhouse gAs UK and Global Emissions project (NERC GAUGE) project (Palmer et al., 2018), explores the possibility of producing top-down methane emission estimates on the sub-national scale to be directly compared with the 2012 NAEI. The approach taken is to make observations at four sites in Eastern England which are tens of km apart. To achieve this, three additional sites measuring

atmospheric methane were installed around an existing site, Tacolneston, which is part of the UK DECC (Deriving Emissions linked to Climate Change) network. Emission estimates were synthesised using a top-down sub-national inversion method developed by the UK Met Office (Manning et al., 2003, 2011). This method is a previous version of the approach known as InTEM (Inversion Technique for Emissions Modelling) as used in Arnold et al. (2018) and will henceforth be referred to as $InTEM_{2014}$. $InTEM_{2014}$ was chosen for two reasons. First, it has been used to produce annual national methane emissions estimates dating back to 1990 (Manning et al., 2011) and so it provides some traceability to the national estimates. Second, we had experience in adapting it in the development of a novel method to estimate $CHBr_3$ emissions around Malaysia (Ashfold et al., 2014). ~~The initial aim of this project was to establish a 'proof of concept' that $InTEM_{2014}$ (and by implication other inversion schemes) could be used at the sub-national scale.~~

This paper presents the ~~preliminary~~ results~~findings~~ of the work and discusses ways to improve the current setup. Section 2 describes the methodology underlying the measurements and the inverse modelling used. The results are presented and discussed in Section 3, with particular emphasis on the causes of uncertainty in the adopted approach. In addition, the results of a model calculation performed as part of an inversion incorporating all of the UK measurements collected within GAUGE and DECC, i.e., with the East Anglian measurements nested within the DECC/GAUGE tall tower network, are presented as a possible way forward.

2. Methodology

Our approach requires two main elements: (i) calibrated measurements from the four sites; and (ii) an inversion model to provide estimates of the emissions and their uncertainties.

2.1 Measurements

2.1.1 Sites

The measurement sites for this trial project were located in East Anglia, United Kingdom. This region was chosen for three reasons:

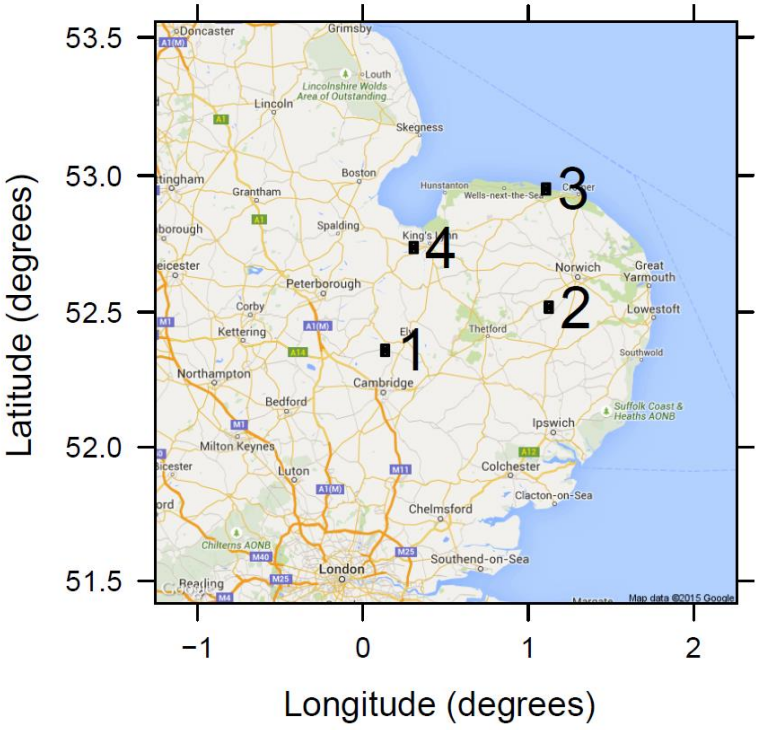
a) The relatively flat topography. Turbulence in the boundary layer and low troposphere is hard to model at the high resolution required for this study. East Anglia is flat and low-lying, with a highest elevation of 146 m. Uncertainties in the small scale meteorological turbulence that is parameterised, not explicitly modelled, in the dispersion model (see Section 2.2.1) are reduced in this simpler topography compared to more heterogeneous terrains. The calculated trajectories are thus in principle more accurate than those calculated for areas of the UK with more complex topography.

b) ~~The existence of gradients in the NAEI emissions fields across East Anglia, provides a better test of the inversion system than would a region with homogenous emissions.~~ The presence of different dominating emission sources within the inversion domain. This allows the inversion system to highlight potential issues in the NAEI, for example, biased emission factors for different processes.

c) Its close proximity to Cambridge, and thus has ease of access to the measurement sites for logistical maintenance and calibration.

The sites' locations were nested within the pre-existing UK DECC tall tower network, which has since been expanded to include two new tall tower sites established under GAUGE (Stavert et al., 2018). The aim was to develop a stand-alone inversion scheme in the first instance and then to integrate these East Anglian measurements into a UK-wide inversion analysis, so the ability to link the calibration of the East Anglian and national networks was vitally important.

The four measurement locations are shown in Figure 1 and some characteristics of the sites are given in Table 1. Sites 1 (Haddenham) and 4 (Tilney) are churches in villages away from the national gas grid and have inlets ~ 25 m above the ground to reduce the influences of local methane sources. The Weybourne Atmospheric Observatory (Site 3, hereafter Weybourne) is coastal, to the North of East Anglia, and has a 10 m mast for its inlet. Two instruments were run in tandem at the Weybourne site and the data combined to ensure data collection in case of instrument failure (Section 2.1.2). Finally, Site 2 is the tall tower measurement site at Tacolneston which has inlets at three heights, 54 m, 100 m and 185 m above the ground. This study uses an average of the 54 m and 100 m observations as a method to reduce local source influences. Differences in inlet altitude amongst the observation sites were represented in the atmospheric dispersion model (Section 2.2.1). East Anglia has multiple sources of methane, which are dominated by emissions from the waste sector (for example, landfills are irregularly found throughout the area) and the agricultural sector (farmland mainly located in the centre and eastern locations of the four sites). Fenlands can be found in the northern and western areas around the four sites (Brown et al., 2018).



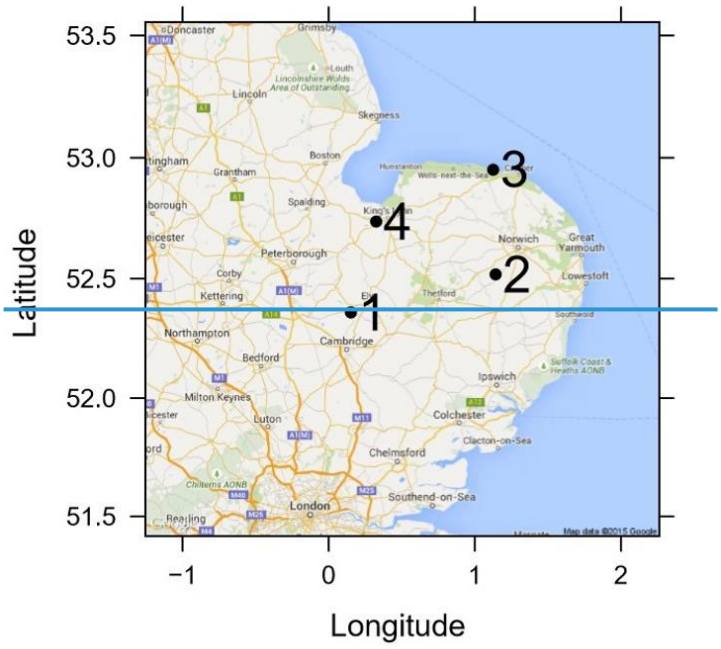


Figure 1: Map of the East of England showing site locations. 1=Haddenham, 2=Tacolneston, 3=Weybourne, 4= Tilney. (Google Maps, 2015). See also Figure 1 in Palmer et al. (2018) for location of all UK monitoring sites, which are used in Section 3.5 in this paper.

Table 1: Overview table of the East Anglian measurement site information.

Site	*Site name*	*Latitude, Longitude (degrees)*	*Inlet height (m agl)*	*Instrument*	*Running dates*
1	Holy Trinity Church, Haddenham	52.359, 0.149	25	GC-FID	06/2012-Present
2	Tacolneston tall tower	52.518, 1.139	54, 100	Picarro CRDS	07/2012-Present
3	Weybourne	52.950, 1.122	10	GC-FID UCAM[1]	02/2013-05/2014
				GC-FID UEA[2]	03/2013-05/2018
4	All Saints Church, Tilney	52.737, 0.321	25	GC-FID	06/2013-Present

2.1.2 Instrumentation

The Tacolneston measurements were made using G2301 (Picarro Inc., USA) Cavity Ring-Down Spectrometer (Crosson, 2008) in the set-up described in Stanley et al. (2018). All other locations used gas chromatography coupled with flame ionisation detectors (GC-FIDs). At these locations, a stainless steel mesh (2 μm) was fitted to the inlet tube to filter any larger impurities from damaging the GC and reducing the air flow. The Weybourne site hosted two GC-FID instruments that shared the same inlet tube. The setup below describes the GC-FID installed by University of Cambridge (GC-FID UCAM). The setup of the second GC-FID, maintained by the University of East Anglia (GC-FID UEA), can be found in Forster (2013). The two data sets are combined in this project, noting differences in sampling intervals (1-2 minutes UCAM, ~20 minutes UEA).

A schematic of the GC-FID instrumental setup used at Sites 1, 3 (UCAM only) and 4 is shown in Figure 2. Nitrogen was used as a carrier gas. Two other gases, pressurised air and hydrogen, were used to fuel the flame within the FID. All three gases first passed through a molecular sieve to filter out water and hydrocarbons (labelled W and HC). The nitrogen

[1] UCAM refers to the GC-FID instrument installed by University of Cambridge, with a sampling rate of 1-2 minutes.
[2] UEA refers to the GC-FID instrument installed by University of East Anglia, with a sampling rate of ~20 minutes.

carrier gas was additionally scrubbed for oxygen to protect the column from oxidation (labelled O). Inlet and calibration tubes were filtered using a desiccant-based Nafion dryer (labelled ND). The GC was run at an internal temperature of 100°C and a column gauge pressure of ~~34 psig~~234 kPa. This setup allowed for a fast methane elution time (< 1 minute). Samples were taken every 1-2 minutes at all GC-FID sites. The raw data were analysed using the commercially available software 'Igor Pro' (WaveMetrics, 2012), which automatically detects and measures the desired peak height. Relative standard deviation (precision) values were defined half hourly. Average values showed precision to be below 0.3% (of the relative standard deviations).

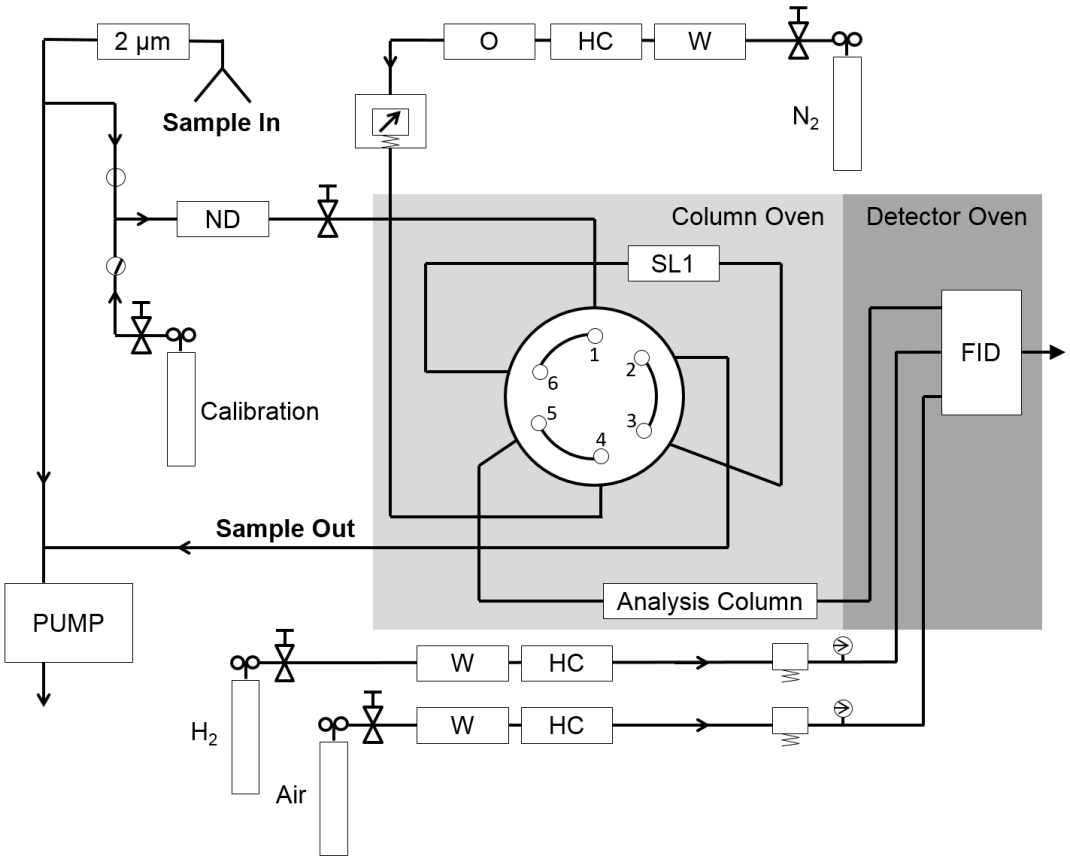


Figure 2: Ellutia GC-FID 200 Series system flow diagram. Nitrogen carrier gas is filtered for water (W), hydrocarbons (HC), and oxygen (O) before entering the column. Hydrogen and compressed air are used to fuel the flame ionisation detector (FID). Both are filtered for W and HC. A funnel filled with stainless steel-mesh (2 µm) is attached to the inlet tube, which is faced down to protect from rain and large particulates. Solenoid valves allow the GC to sample either the inlet air or the calibration gas. A pump is attached to draw in the inlet air and draw out the sample from the GC-FID. NB: SL1 = sample loop 1. ND = Nafion dryer.

2.1.3 Calibration

All GC-FID sites were calibrated using an NPL calibration gas (0.28% precision) at half hourly intervals. All sites were one point calibration with the exception of the instruments at Weybourne (Site 3), which was a multi-point calibration Inter-calibration experiments between our NPL-calibrated instruments (Site 1, GC-FID UCAM at Site 3, Site 4) and the NOAA-calibrated instruments (Site 2, GC-FID UEA at Site 3) showed an offset of -4.9 ppb (average of three calibration experiments). Although both the stated and derived calibration concentrations for the NPL standard were within the ranges of the calibration gas uncertainties plus GC-FID precision, all NPL measurements were converted to the NOAA scale, consistent with the DECC network (Stanley et al. 2018).

2.2 Inversion Modelling

2.2.1 NAME trajectory calculation

The UK Met Office's Numerical Atmospheric dispersion Modelling Environment (NAME) model (Jones et al., 2007) is used to estimate air flow from potential methane sources to the measurement sites. NAME was originally developed by the UK Met Office for modelling the long-range dispersion of radioactive material from nuclear power stations (Maryon et al., 1991). It is a Lagrangian model which uses the 3-D meteorological fields produced by the UK Met Office's numerical weather prediction model, the Unified Model (UM; Cullen 1993). When run backwards in time, NAME dispersion trajectories are used in the inverse modelling of atmospheric emissions (Ashfold et al., 2014; Manning et al., 2003). This project used two resolutions of UM meteorological fields: global (3 hourly, ~25 km horizontal, 8 levels in the lowest 500 m vertical[3]) and UK (hourly, ~1.5 km horizontal, 12 levels in the lowest 500m vertical, ~~(~~Tang et al., 2013) ~~)~~, the 1.5 km UK fields were nested within the global data when running NAME.

The model setup at each site is identical except for the particle-source release location (latitude, longitude) and height (m above ground level). The three sites with inlets between 15-27 m (1-Haddenham, 3-Weybourne and 4-Tilney) have a modelled release altitude of 25 m (±25 m) above sea level. Tacolneston (Site 2), with inlets at 54 m and 100 m is assigned a release altitude of 75 m (±25 m).

NAME produces a modelled representation of the contributing 'surface influence' (defined as the lowest 100 m above ground level in NAME) at a particular location (one of the measurement sites) by releasing chemically inert particles (10,000 h^{-1}) from the *x*, *y*, *z* coordinate of that measurement site. NAME computes the movements and geolocation of each particle every minute for 5 days backwards in time. Each location releases mass at a rate of 1 g s^{-1} equally distributed across the particles. A time integrated particle density map (units g s m^{-3}; resolution 1.5 × 1.5 km) is produced for each measurement location that shows, on a gridded output, the relative contribution that each grid square has made over the preceding 5-day period (Manning et al., 2011). After conversion, the resulting metric (units of s m^{-1}) can be described as the mean time that particles reside in each grid cell for a 1 hour particle release period. This metric corresponds to the multiplying factor by which emissions are diluted from their initial source to being monitored at the measurement location. This relationship is given in Equation 1 and a ~~dilution~~footprint map for the measurement site Haddenham is shown in Figure 3. ~~Dilution~~Footprint maps are calculated hourly at each measurement location over the monitoring period (of two years). Results are compiled into a '~~dilution~~footprint matrix' (also referred to as the Jacobian matrix), which shows how the ~~dilution~~footprint values changed over time, and is an input into the inversion system.

The model domain limits, shown in Figure 3, are centred on East Anglia but span most of the south east of England. With the model particle lifetime set at five days, this is long enough for the vast majority of particles to leave the domain of interest, and thus capture all surface influence within the geographical domain.

[3] UM vertical resolution levels decrease as the altitude increases.

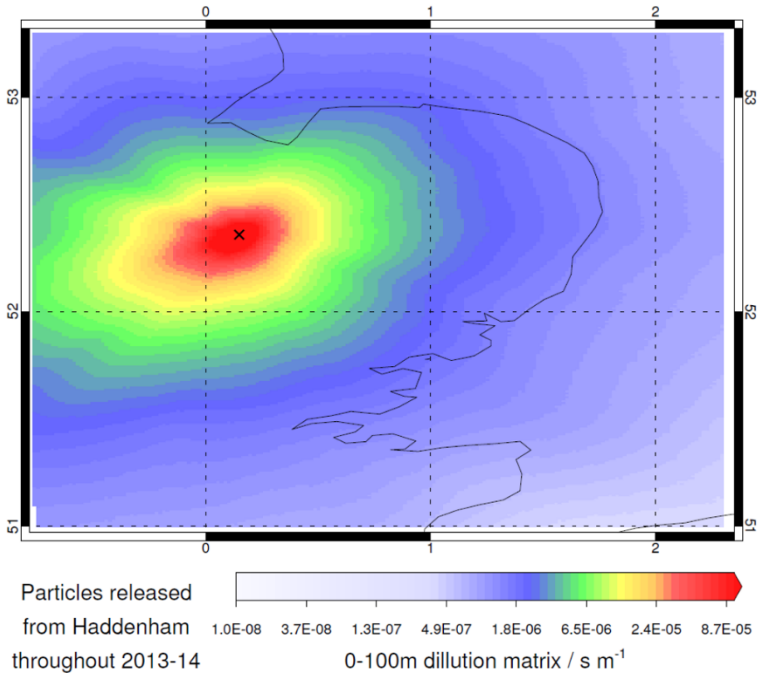


Figure 3: ~~Mean dilution~~Footprint matrix for the Haddenham site (Site 1, location marked with an X) for 2013 and 2014, as calculated by the NAME particle dispersion model. 3-D meteorological fields produced by the UK Met Office's numerical weather prediction model, at 1.5 km regional resolution nested within 25 km global resolution are used when running NAME. Sources are released for a one hour duration period, every hour from 01 January 2013 to 31 December 2014. Particles' geolocation is calculated every minute for 5 days backwards in time. Each location releases particles at a rate of 1 g s^{-1}, resulting in a time integrated particle density map (units g s m^{-3}; resolution 1.5 × 1.5 km, (Tang et al., 2013)) that shows the relative contribution of each grid square over the preceding 5-day period (Manning et al., 2011). Figure shows the averaged footprint map over one year whereas timestamps of 1 hour are used as input data within $InTEM_{2014}$. A conversion to the '~~dilution~~footprint matrix' (units of s m^{-1}) can be described as the mean time that particles reside in each grid cell given a 1 hour particle release period.

2.2.2 $InTEM_{2014}$ inversion model

$InTEM_{2014}$ uses methane measurements and the corresponding ~~dilution~~footprint maps to estimate emissions within a given domain according to the relationship expressed in Equation 1, where *y* is an n × 1 vector of concentrations (units: g m−3), *K* is an n × m footprint matrix (units: s m^{-1}), *x* is an m×1 vector of gridded emissions (units: g s^{-1} m^{-2}), and *b* is an n × 1 vector of concentrations upwind of the modelling domain (units: g m^{-3}).

$$y = Kx + b \quad (1)$$

~~emission (g s^{-1} m^{-2}) x dilution (s m^{-1}) = concentration (g m^{-3})~~

Through an iterative process known as simulated annealing (Manning et al., 2003), ~~pseudo~~modelled-observations calculated from simulated emissions fields for specific times and locations are quantitatively compared with the measured observations using cost functional analysis. The resulting $InTEM_{2014}$ emissions estimate will be the emissions field with the lowest cost score.

~~Pseudo~~These simulated-observations are calculated by multiplying the emission estimates (from the 2012 NAEI) for each grid cell by their corresponding ~~dilution~~footprint value (taken from NAME) at each timestep. A least-squares cost function used (Equation 2) to quantitatively compare the two observational time series, similar to cost functions used in work such as Manning et al. (2003), Ashfold et al. (2014), and (Fang et al., 2016). Bayesian approaches have become more regularly used in top-down emission estimates, which incorporate pre-defined uncertainty estimates that result in calculated

uncertainties for the final emissions (e.g., Arnold et al., 2018; Bousquet et al., 2011; Feng et al., 2018). Hierarchical Bayesian cost functions have since been developed where both the uncertainty values associated with the prior estimates, and the model uncertainty estimates can be derived within the inversion itself (Ganesan et al., 2014; Lunt et al., 2016). Estimating realistic and rigorously derived emission uncertainties remains a major challenge in inversion studies.

The cost function used here incorporates defined uncertainties associated with the observations and the model but does not include the use of a prior (see Section 2.2.3).

$$r_i = \sum_{i=1}^{n} \left(\frac{(y_i - (Kx_i)^2)}{\left((\sigma_\varepsilon)_i\right)^2 (\sigma_{\varepsilon})_i^2} \right) \quad (2)$$

K_i is the forward model and x_i is the measured concentration at a particular timestep (i). At all timesteps the difference between the ~~pseudo~~simulated-observation (Kx_i) and the measured observation (y_i) is squared and then divided by the uncertainty variance $(\sigma_\varepsilon{}^2)_i$. This uncertainty is the sum of all assumed errors in observations, modelling and baselines for each hourly timestep (i). Observational uncertainty is defined as the sum of the hourly instrument precision, the calibration gas uncertainty, and the standard deviations of the hourly concentrations plus 5 ppb. The value of 5 ppb is an estimate of the uncertainty in the baseline value in any given hour (Section 2.2.4). Dividing by the total uncertainty (i.e., variance) de-weights uncertain observations. The lower the resulting cost score (r in Equation 2) the smaller the difference between the ~~pseudo~~simulated and measured observations, implying a more accurate emissions estimate than one with a higher cost score.

The simulated annealing method in InTEM$_{2014}$ iteratively converges on the best solution and the final result is limited by the available computer resource. Therefore, the derived final output is close to but may not be the best possible solution, i.e., with the lowest possible cost score. For this reason, and due to the stochastic nature of the convergence within the simulated annealing process, the InTEM$_{2014}$ runs were repeated multiple times and the resulting emission results averaged. Sensitivity analyses showed that 25 repeats were sufficient to produce consistent methane emission estimates, standard deviations and cost scores. The initial starting point for the simulating annealing process is a random, non-negative emission field, which assumes no a priori knowledge of the location or magnitude of emissions.

~~**2.2.3 A priori emission estimates**~~

~~Unlike Bayesian inversions, InTEM$_{2014}$ does not use a prescribed *a priori* emission estimate, which includes boundaries to the emission magnitudes based on uncertainty assumptions. In this study, we used a random, non-negative emission field, which assumes no a priori knowledge at the location of emissions.~~

2.2.43 Solution grid

There has been deep discussion around defining the multi-scale state vector used in inversion modelling (Bousquet et al., 2011; Henne et al., 2016; Lunt et al., 2016). For this study, ~~The~~ methane emission estimates are resolved on a more spatially coarse grid than the NAME model output to reduce the computational cost and decrease modelling uncertainties (Manning et al., 2003). This so-called 'solution grid', of which the a posteriori estimate is resolved, is irregular and is constructed using the ~~dilution~~footprint matrix (Section 2.2.1) and the 2012 NAEI for methane, but the NAME output grid can first be divided into broader regions to calculate emission totals. These regions are based on the East Anglia county

boundaries, providing rough county-wide estimates of methane emitted over the given period of time. The solution grid resolution is a sub-division of these county areas, which has a spatial resolution that is between the county-based starting regions and the NAME grid. The solution grid resolution is determined through two factors: the ~~dilution~~footprint matrix and the NAEI methane emissions values.

For the ~~dilution~~footprint grid matrix, areas where trajectories spent relatively long periods of time will have a finer spatial resolution, as more data are available and thus there is greater sensitivity to resolve the emissions from that area. A pre-defined ~~dilution~~footprint threshold subdivides regions into finer grids based on the ~~dilution~~footprint matrix (Manning et al., 2011). Generally, areas nearer to the measurement sites are more finely resolved than more distant areas. Complementing this, the NAEI methane emissions are also incorporated to define the solution grid resolution. The NAEI emission magnitudes are used as a linear scaling factor to define the grid resolution (as in other inversion techniques such as Rigby et al. (2011). High methane emission sources in the NAEI (e.g., landfills) are more finely resolved than low methane emission areas. The resulting solution grid resolution can be seen in Figure 4.

Sensitivity analysis conducted by varying the spatial resolution of this solutions grid. Solving on a more finely spatially resolved solution grid helps to minimise aggregation error, but increasing resolution comes at a computational cost Furthermore, aggregating emission estimates can help account for trajectory uncertainties from meteorological data errors. $InTEM_{2014}$ was repeatedly ran using solution grids consisting of between 15 grid boxes (Figure 4a) and 250 grid boxes. More spatially fine results showed indistinguishable differences in emission estimates for each sub-regional areas (Figure 4a), as all estimates with within 1 standard deviation. The final solution grid resolution (Figure 4b) was fine enough to resolve nearby point sources without being prohibitively computationally intensive.

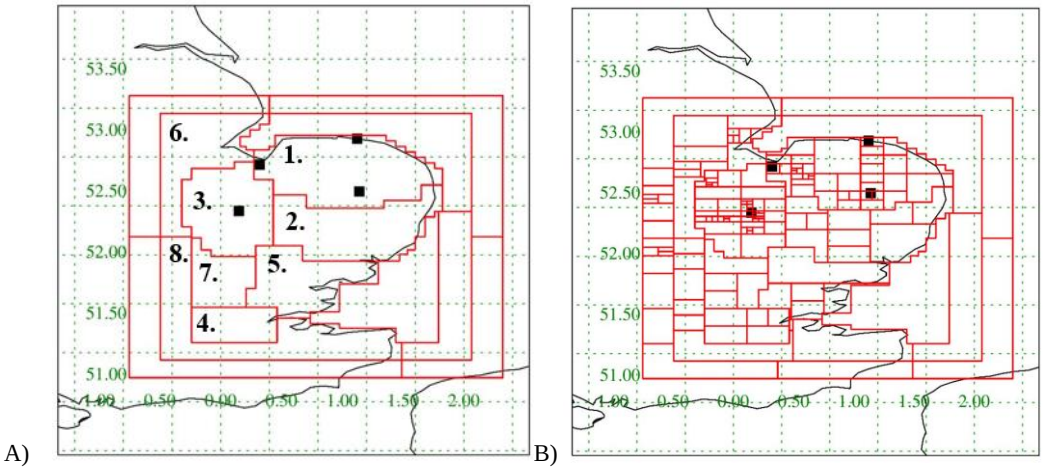


Figure 4: A) Map showing the starting resolution of the inversion. These regions are loosely based on the county regions in East Anglia. As an output, InTEM provides statistical information on the emissions for each of these regions. B) Map showing the solution grid resolution. This grid resolution is computed based on information from the ~~dilution~~footprint matrix (Figure 3) and the 2012 NAEI for methane (Brown et al. 2018). Numbering refers to counties or other more arbitrary areas: 1= Norfolk; 2= Suffolk; 3= Cambridgeshire; 4 = London; 5 = Essex; 6 = Lincolnshire; 7 = Buckinghamshire; 8 = South west area.

2.2.~~5~~ 4 Baseline

Within inversion modelling a baseline, representing incoming atmospheric concentrations at the edge of the inversion domain must be defined. Multiple methods for calculating baselines have been reported in the literature. Predominantly, baselines are either pre-computed (e.g., ~~(~~Henne et al., 2016) or resolved as part of the inversion (e.g., ~~(~~Ganesan et al., 2014).

~~A baseline that represents the atmospheric methane concentration arriving at the edge of the inversion domain (Figure 4) must be defined within InTEM$_{2014}$.~~ For this study, a pre-computed, statistical baseline was calculated from the measured observations that also incorporated the particle trajectory / footprint matrix analysis from NAME, to account for varying baseline methane concentrations entering the inversion domain from different wind directions. Individual baselines for each site were created based upon their air-mass history contributions of the sites for each hourly time stamp. Methane concentrations from the four measurement sites were divided into time series depending on whether their trajectory origin was dominated by a certain direction (e.g., from the NNE, ENE, ESE, NNW etc.). These eight individual time series, representing concentrations from the eight different compass directions, were used to estimate eight statistically derived baselines, each calculated by passing a rolling 18th percentile, spanning one week, through each dataset. Sensitivity analysis, which assessed the timeframe and the percentile of the statistical filtering was conducted. The 18th percentile is chosen from a sensitivity analysis, in which the rolling percentile was varied from the 5th to the 45th percentile. The 18th percentile produces consistent ~~emission~~ results with the lowest standard deviation of ~~consistently stable~~the a posteriori emissions from all baselines tested, and with the lowest cost score of all the baselines tested[4]. Baselines for the four measurement sites were then created using the NAME trajectory analysis to weight a combination of the eight direction dependent baselines.

In InTEM, any above-baseline concentrations are set to have a value of zero, as a way of avoiding negative emissions. If a baseline is set too high it results in reducing peak sizes in the observed data and thus an under-estimation of a posteriori emissions. Conversely, a too low baseline will result in higher a posteriori emissions, particularly in around the edges of the inversion domain, as any measured methane has actually originated from outside the inversion domain (and has not been correctly filtered out in the baseline) are placed in the boxes furthest away from the observation sites. For this reason, a posterior emissions from the border regions shown in Figure 4 are always discounted in the InTEM$_{2014}$ emissions analysis (Section 3).

3. Results

This section covers results from the InTEM$_{2014}$ inversion analysis. For analysis of the concentrations from the measurement sites, including their daily, weekly and intra-annual variability, please refer to Figure 3 in Palmer et al. (2018).

3.2 InTEM$_{2014}$ emission estimate results

The following results were derived using a one year dataset from all four sites covering the period from June 2013 to May 2014 (inclusive) with hourly observations. This period was chosen as it marks the first full year where all four sites were operational. A subset of the resulting measured and ~~pseudo~~simulated-observations for 01-30 July 2013 is shown in Figure 5. The equivalent 2012 NAEI ~~pseudo~~simulated-observations are included for comparison. From the measured observations, it is clear that the Haddenham and Tilney sites observe short periods of elevated methane, usually during nocturnal hours when the boundary layer height is low, which suggests the presence of local methane sources. Several landfill sites can be found close to Haddenham and Tilney (<10 km) which can be large point sources of methane (NAEI, Brown et al., 2018). Isotopic analysis by Riddick et al. (2017), confirmed a methane signal from the Waterbeach Waste Management Park being present at the Haddenham site at times of elevated methane.

Figure 5 shows a large fraction of the *a posteriori* estimates lie outside the uncertainty range of the measured observations, although without a more thorough description of a priori errors this is difficult to fully diagnose. As it stands, this implies

[4] Cost score refers to the resulting cost score from InTEM$_{2014}$ when run using each tested baseline as an input.

that either the prescribed InTEM$_{2014}$ uncertainties are too small or the resulting emission field needs be more resolved (in time and space) in order to better represent the concentrations being measured (currently, the spatial resolution of the resulting emission grid could be too coarse to fully capture the peaks and troughs of the measured time series). It should be noted that the ~~pseudo~~simulated-observations calculated using the NAEI are substantially outside the observation uncertainty ranges, and that neither are able to replicate the high concentrations measured at the Haddenham and Tilney sites. A scatterplot of a posteriori enhancements vs. observed enhancements as calculated by InTEM$_{2014}$ at the Haddenham site can be found in Riddick et al 2017 (Figure SM2.2). The InTEM$_{2014}$ resulting emissions field has a lower (i.e., better) cost score than the emissions grid from the NAEI (12.5 compared to 14.9). For comparison, the 2009 NAEI emissions grid yielded a higher cost score of 15.8, showing that the methane emissions distribution produced by InTEM$_{2014}$ fits the measured observations better than the 2009 or the 2012 NAEI.

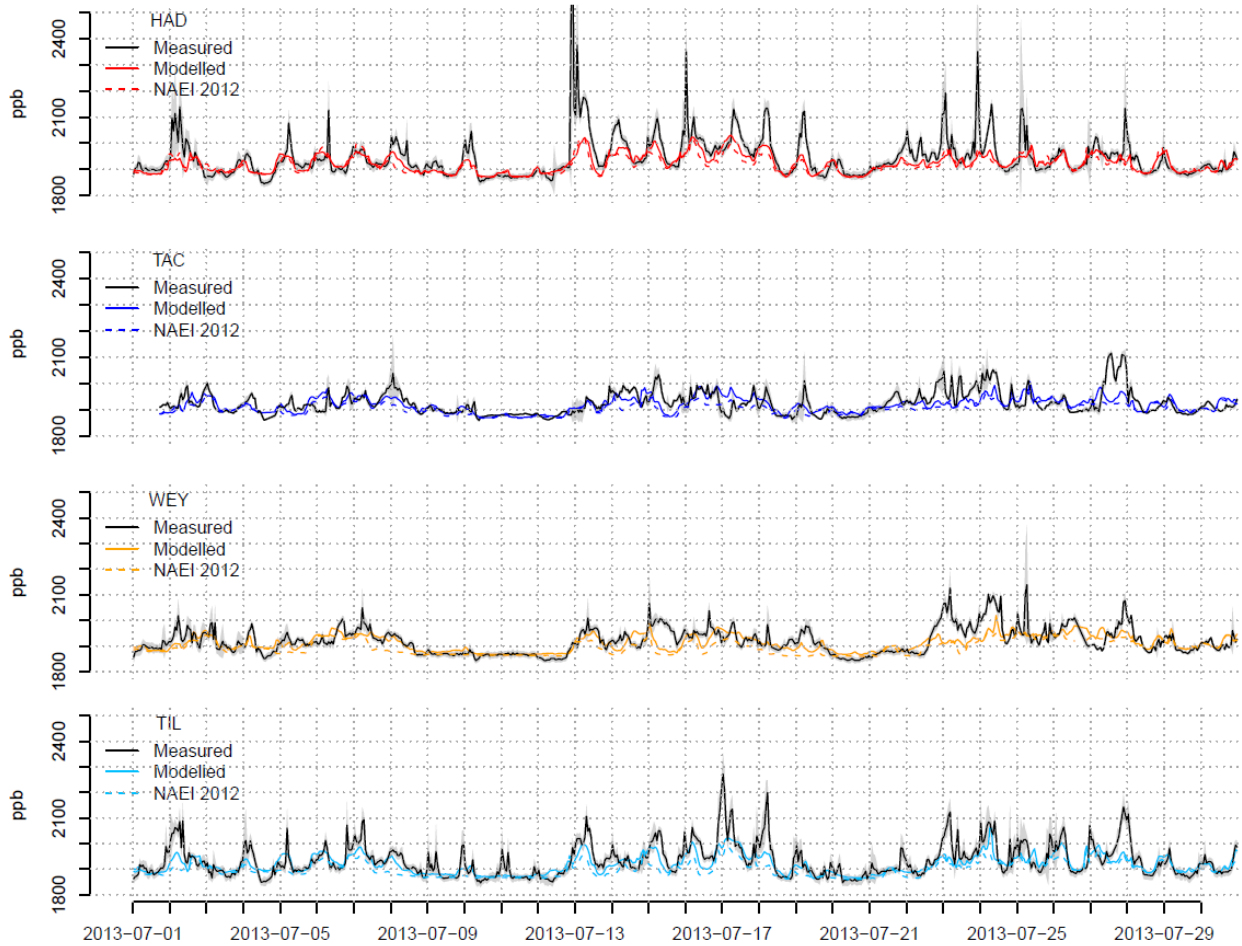


Figure 5: Time series of measured and *a posteriori* modelled methane (using inversion estimated emission distribution) mole fractions for all 4 observations sites for 01-30 July 2013. The equivalent ~~pseudo~~simulated-observations calculated using the 2012 NAEI emissions inventory (Brown et al. 2018) have been added for comparison.

3.2.1 Regionality

Figure 6 shows the inversion emission map and the 2012 NAEI methane emission map, both plotted to the same spatial resolution (see Section 2.2.4: Solution grid). The inversion produces comparable emissions estimates for East Anglia with total estimates being within 15% of the NAEI (NAEI estimates 280 ±112[5] kt yr^{-1}, InTEM$_{2014}$ estimates 310 ±63.0 kt yr^{-1}, rounded to 2 s.f.). Similarities between the spatiality of emission are visible, with both maps showing large emissions in

[5] Uncertainty calculated using 40% estimate provided with NAEI for the whole of the UK. Sub-national uncertainty estimates were not provided.

the London area, point sources around Haddenham, and lower emissions along the southern East Anglian coast. Discrepancies appear between some of the magnitudes in the finely resolved emissions maps, but local studies using additional measurements, Gaussian plume, and local scale Lagrangian particle dispersion~~WindTrax~~ modelling do show that the high point source emissions near Haddenham are real (Riddick et al., 2017).

Table 2 shows the inversion area emission totals (labelled in Figure 4). The East Anglian areas are loosely based on the UK counties (Suffolk - 4, Norfolk - 10 and Cambridgeshire - 15). A positive relationship between area standard deviations and the distance from the measurement sites can be seen in Table 2. For example, the areas close to London and in the south west of the regions have standard deviations of 18.0 kt yr^{-1} and 59.2 kt yr^{-1}, respectively, but areas representing the east Anglian counties are all below a standard deviation of 2.5 kt yr^{-1}. This implies that $InTEM_{2014}$ is able to more robustly resolve emission totals for areas close to measurement sites, although individual site biases apply (see Section 3.4). This analysis implies that the ~15-25 m a.g.l. EA measurement sites have an effective local footprint~~s of roughly a 50 km radius~~. Our estimates for methane emissions from Norfolk and Suffolk show good agreement with the estimates in NAEI, with differences of ~5%. Larger differences are found for Cambridgeshire where our estimate is 22.5% lower than that of NAEI. Percentage differences for regions that are further away from the measurement sites range from 10.8% (region 11, London area) to 66.1% (region 1, south west area). All land area estimates are within a factor of two of the NAEI.

Compared to the NAEI, $InTEM_{2014}$ emissions have a 'dipole effect' in some areas. For example, a large methane source is shown to the south west of Tacolneston but low emissions are estimated in the surrounding area. The NAEI also shows an increased emission level south west of Tacolneston, but the overall emission ranges are less extreme. It is unclear if these dipoles are 'true' signals, or a product of $InTEM_{2014}$'s inability to fully resolve emissions on this spatial and temporal scale. Differentiating between false dipoles and real point sources is not straightforward in this analysis. Intermittent source emissions, as well as uncertainty in the meteorological analyses used to run NAME could account for $InTEM_{2014}$ being unable to pinpoint some emission sources. In principle, this could be overcome by introducing point sources into the priori, as used in some Bayesian approaches (Rigby et al., 2017).

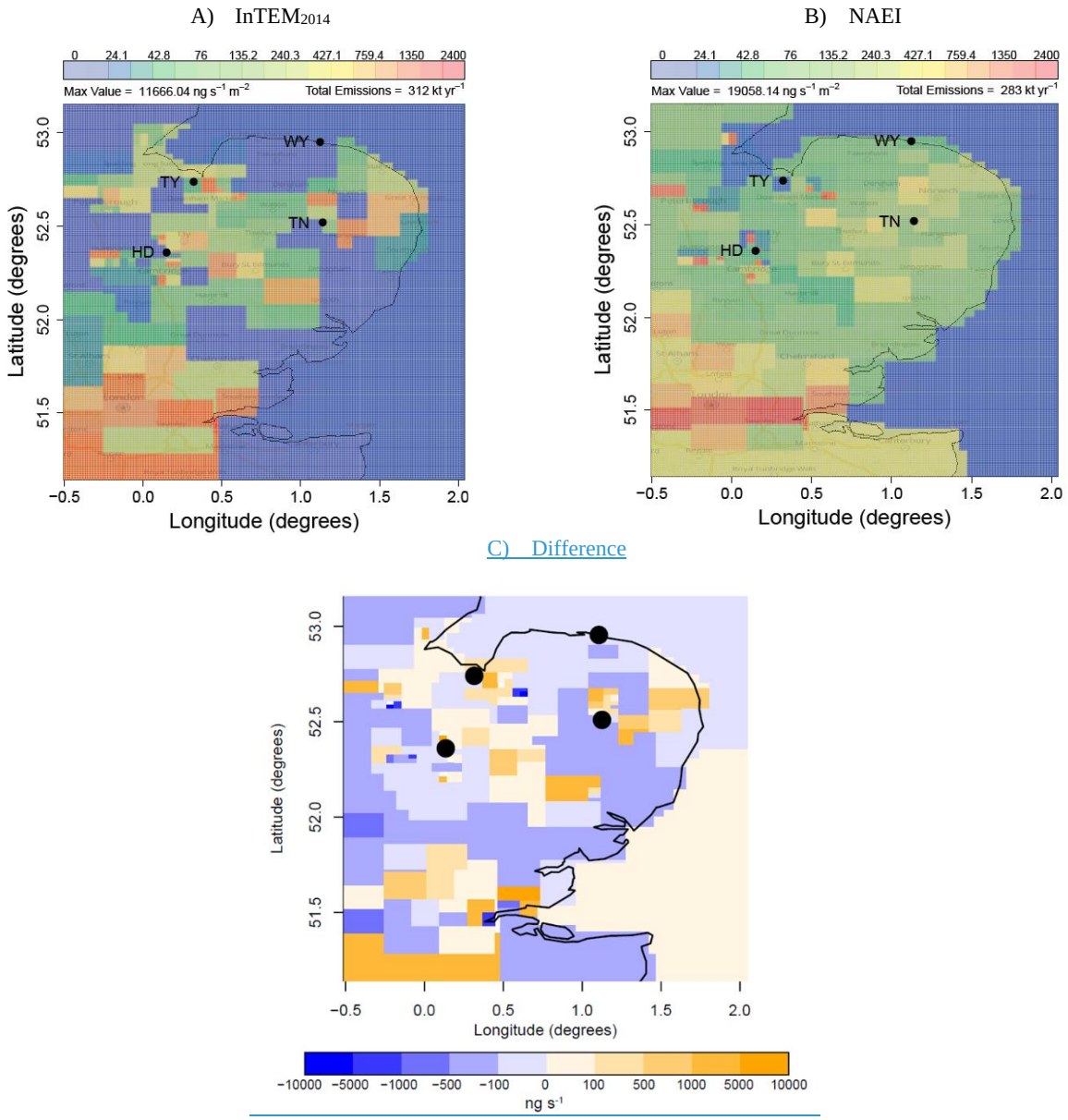


Figure 6: A) Methane emission map, from June 2013 to May 2014, produced by an InTEM inversion run using all 4 sites' observational data. B) The 2012 NAEI (Brown et al. 2018) re-gridded to the inversion grid resolution (see Figure 5). C) Difference plot of the *a posteriori* InTEM$_{2014}$ estimates minus the NAEI. [CS2]**Sites are labelled for reference: HD = Haddenham, TN = Tacolneston, WY = Weybourne, TY = Tilney. NB: Logarithmic colour scale. Difference between orange / red is roughly a factor of 100 larger than the difference between blue / green. Gridlines in 6.A and 6.B represent the spatial resolution of the NAME model output. The outer border regions as shown in Figure 4 are not displayed here due to baseline reasons as discussed in Section 2.2.4.**

Table 2: Emission totals (kt yr^{-1}) resulting from InTEM inversion using all 4 observational site data for the period June 2013 to May 2014. Emission totals are for 'regions' shown in Figure 5. Equivalent totals of the 2012 NAEI (Brown et al. 2018) per region and their differences as percentages are shown for comparison. One standard deviation (1.s.d.) is shown below regional estimates. Sea regions from Figure 5 not shown here but emissions totals were 0.4 kt yr^{-1} (0.64 1.s.d.) compared to 0.7 kt yr^{-1} from the NAEI (57.1% difference).

#		*NAEI* (kt yr^{-1})	*InTEM ± 1 standard deviation* (kt yr^{-1})	*% difference* (kt yr^{-1})
1	*Norfolk*	38.9	37.1 ±1.7	4.7
2	*Suffolk*	24.1	22.8 ±1.9	5.6
3	*Cambridgeshire*	26.5	20.5 ±2.1	22.5
4	*London*	51.2	45.7 ±18.0	10.8
5	*Essex*	24.5	19.6 ±8.1	19.9
6	*Lincolnshire*	17.6	9.1 ±4.1	48.3
7	*Buckinghamshire*	20.5	30.6 ±7.3	-49.1
8	*South west area*	75.0	124.5 ±59.2	-66.1
	TOTAL	278.3 ±112.3[6]	310.5 ±63.0	-11.4

3.2.2 Methane emission estimates surrounding Haddenham

Due to how the spatial resolution of the emission grid is calculated (Section 2.3.3), finer spatial resolution is available in areas around the measurement sites. Figure 7 shows a magnified section of the emission maps in Figure 6 centred around the Haddenham site (Site 1) from (a) the inversion study and (b) the NAEI. Although, the uncertainty associated with point sources is high (inversion standard deviations can be ~100% or larger for individual point sources, (Riddick et al., 2017), interesting aspects are discernible from the existence and locations of some of these emissions. Firstly, all point sources in the NAEI (Figure 7b) correspond to landfill sites, with the exception of the most southerly point source, which is the city of Cambridge. The inversion resolved these emissions, although all emissions west of Haddenham are lower than the NAEI. Furthermore, our analysis finds fewer emissions in the area labelled "1" compared to the NAEI. This area corresponds to 'historic' landfills that are no longer in use (decommissioned in the late 1980s / early 1990s, Environment Agency, (UKEA, 2015) yet are still estimated to be emitting methane in the NAEI. Some studies have recorded methane emissions from landfills peaking and substantially reducing after a few years after burial (Börjesson et al., 2001; Hegde et al., 2003; Kelly et al., 2006). For example, Hegde et al. (2003) investigated methane emissions from a landfill in Taiwan and observed that buried waste had a peak emission between two and three years after burial and that emissions after five years were 0.63% of the maximum values measured. Additionally, (Börjesson et al., (2001) measured methane oxidation between 60 and 94% from soils in an 11 year old landfill compared to be between 41 and 50% from a new landfill, implying lower methane emissions from older sites. This analysis suggests emissions to be lower than calculated in the NAEI, although model uncertainties are significant. The area labelled "2" in Figure 7 shows another discernible difference between the inversion and NAEI emissions, with the inversion results showing larger methane emissions. This area corresponds to managed and unmanaged fenland with multiple irrigation or drainage channels structured throughout. Areas of near-stagnant water can potentially be large methane emitters (Minkkinen and Laine, 2006) but few methane sources are estimated in the NAEI in this region (Brown et al., 2018). With this InTEM$_{2014}$ setup, our results suggest a potentially missing, or underestimated methane source in the NAEI for this area, although a more quantified uncertainty

[6] Uncertainty calculated using 40% estimate provided with NAEI for the whole of the UK. Sub-national uncertainty estimates were not provided.

analysis would be needed as part of further work to resolve these emissions more fully. For example, using an inversion grid structure based on the dominating land cover types could help to distinguish different source processes.

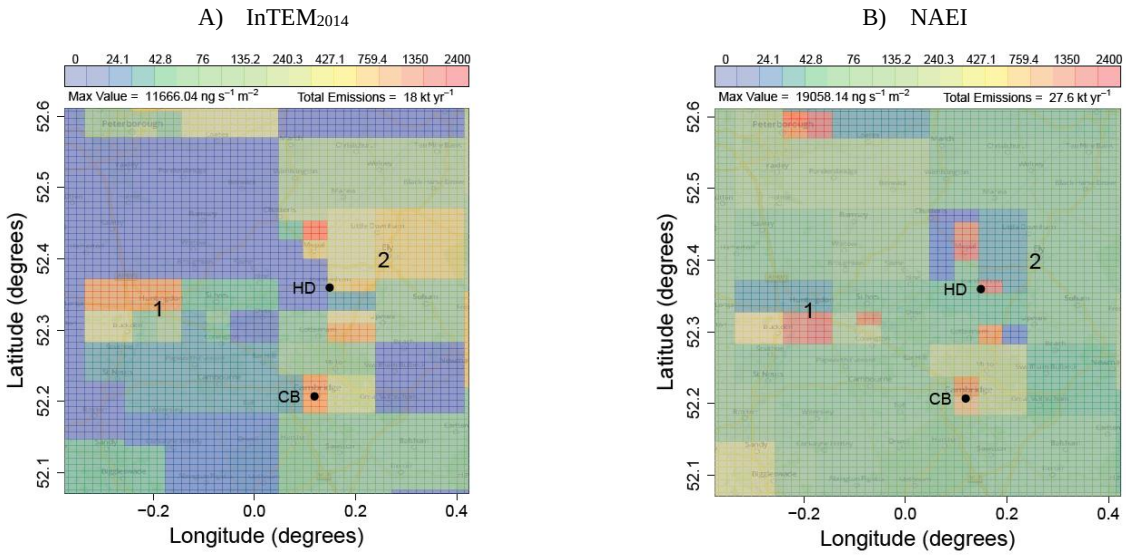


Figure 7: A) Methane emission map, from June 2013 to May 2014, produced by the inversion using all 4 sites' observational data, zoomed to the Cambridgeshire area. B) The 2012 NAEI methane emissions (Brown et al. 2018) re-gridded to the inversion grid. Cambridge (CB) and Haddenham (HD) are labelled for reference. Label 1 refers to an area of active landfills. Label 2 refers to an area of manufactured irrigation channels, where stagnant water can accumulate. Gridlines represent the spatial resolution of the NAME model output.

3.4 InTEM$_{2014}$ sensitivity to the number of observation sites

The final part of our analysis investigates InTEM$_{2014}$'s sensitivity to the number of measurement sites used within the inversion. For this analysis InTEM$_{2014}$ was run as described in Section 2.2 (one year period, June 2013 - May 2014) but the inversion was repeated using observation data from a subset of 1-3 measurement sites (all combinations were assessed). The InTEM$_{2014}$ emission estimates for the Norfolk, Suffolk and Cambridgeshire areas (areas 4, 10, 15) are plotted in Figure 8 (referred to as NSC). This figure shows the range of NSC emissions totals is reduced as more measurement sites are incorporated in the inversion run. Furthermore, we can see that the inversion method is influenced by the specific sites' measurement data. For example, the sites which experience the lowest range of methane concentrations (Weybourne, Tacolneston) produce lower emission estimates for the NSC region (see Figure 5, and Figure 3 in Palmer et al. 2018). Similarly, sites with more local point sources (Haddenham, Tilney) produce higher regional emissions maps.

NSC total estimates using a single measurement site in the inversion are further away from the NAEI but closer when all four measurement sites are used. However, in each inversion result using only one measurement site, the county estimates for the county that the single site resides compares more closely to the NAEI than other county estimates.

Poorly resolved local influences are diminished with the incorporation of other sites' data but not removed entirely. For example, inversions using Haddenham data always produces the higher NSC total emission estimates. These results strengthen the argument for incorporating multiple sites within inversion analysis but the number of required sites is dependent on the size and resolution, both spatial and temporal, of the desired region for analysis.

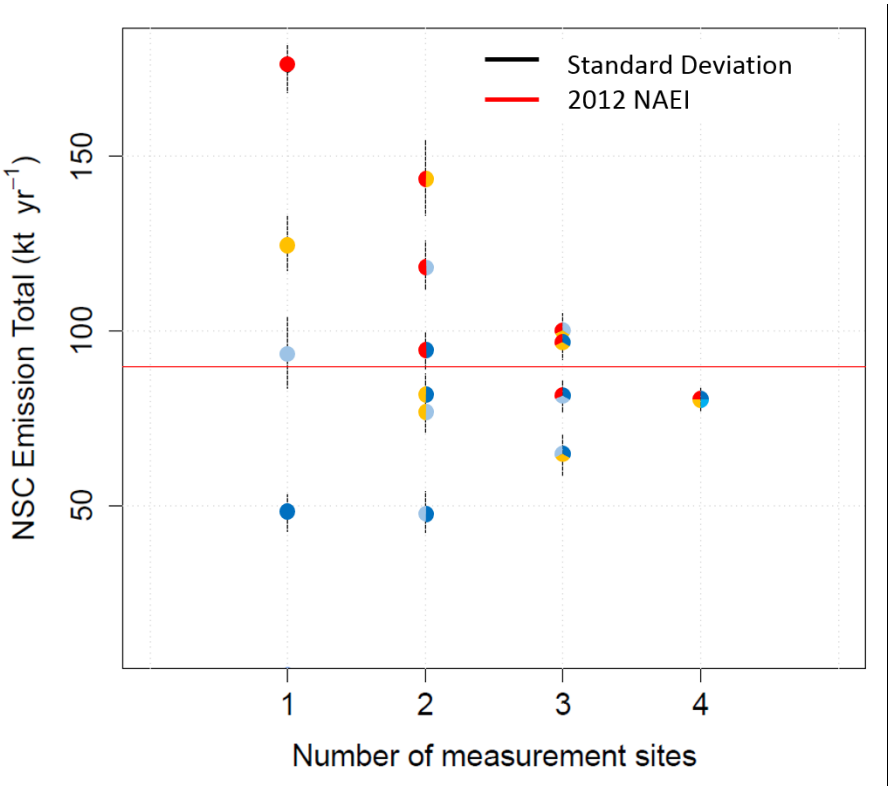


Figure 8: Emission totals (kt yr^{-1}) for the three areas approximately corresponding to Norfolk, Suffolk, and Cambridgeshire (NSC), as shown in Figure 4. Emission result from InTEM inversions being run with 1-4 observational site(s) data. *x*-axis shows number of observation sites used in each InTEM inversion. Vertical black lines represent one standard deviation. Horizontal red line shows NAEI emissions total for the NSC area. Colours shown on each dot correspond as Haddenham (red), Tacolneston (dark blue), Weybourne (light blue) and Tilney (orange). Colours also correspond to those used in Figure 5).

3.5 Effect of including East Anglian sites in a national inversion

To investigate the influence of the East Anglia (EA) measurement network, InTEM$_{2018}$, as described in Arnold et al. (2018), was run both with and without the inclusion of the sites within the National UK network. Arnold et al., (2018) uses an updated version of InTEM for estimating national emissions and the main differences with InTEM$_{2014}$ are summarised in Table 3.

Figure 9 shows the resulting methane emissions map centred over the east of England for A) the UK network including the EA measurement sites and B) without the EA network. Please note that Tacolneston is included in both inversions as it is a tall tower measurement site, however the measurement inlet heights vary for the different inversions (Table 3). Table 4 show the InTEM$_{2018}$ estimated emission totals for an area closely corresponding to the counties Norfolk, Suffolk and Cambridgeshire, the three closest counties to the EA sites, calculated by InTEM$_{2014}$, both with and without the inclusion of the EA measurement sites. From this table, it is clear that the additional inclusion of the EA sites into InTEM$_{2018}$ does not greatly alter the emission estimates for the area but the uncertainty has been reduced (84.9 kT yr^{-1}, with 1.s.d. of 12.8, including the EA sites compared with 82.3 kT yr^{-1} with 1.s.d. of 23.6 without). This is reassuring as it implies robustness of the inversion results to additional data. Both InTEM$_{2018}$ estimates show a 45% increase compared to the 2015 NAEI. Additionally, the inclusion of the EA sites seems to resolve differences in emissions around the measurement locations,~~allows for finer spatial resolution to be resolved in the national inversion, and thus~~ perhaps providinges ~~further~~ new information. For example, Figure 9a shows a latitudinal band of larger methane emissions just north of Tacolneston (Site 2), a feature also visible in the InTEM sub-national inversions (Figure 6a).

In Table 4, the geographical boundaries between the InTEM$_{2014}$ and InTEM$_{2018}$ emission totals for the EA area vary slightly due to differences in the spatial resolution of the emissions grid, which result in the areas not being directly

comparable. Nevertheless, a rough comparison shows similar totals, again demonstrating the stability of the inversion results, and both estimate higher emissions compared to the 2015 NAEI and lower compared to the 2012 NAEI.

Table 3: differences in InTEM$_{2014}$ (Connors et al 2018) and InTEM$_{2018}$ (Arnold et al 2018). NB: a.g.l = above ground level.

	InTEM$_{2014}$	***InTEM$_{2018}$***
Observations:		
Tacolneston measurement height	Average of 54 and 100m	Average of 54, 100, and 185 m
NAME dispersion model:		
Definition of 'surface influence'	lowest 100 m a.g.l	lowest 40 m a.g.l
# of inert particles released	10 000 hr^{-1}	20 000 hr^{-1}
Particle tracking timestamps	5 days, every 1 minute	30 days, every 1- 6 minutes
Grid resolution	~1.5 km x 1.5 km	~ 25 km x 25 km
Inversion framework:		
Prior	none	2015 NAEI (40% UK uncertainty)
Cost function type	Least Squares~~Simulated annealing~~	Bayesian
Solution grid resolution	≥4 km x 4 km	≥25 km x 25 km

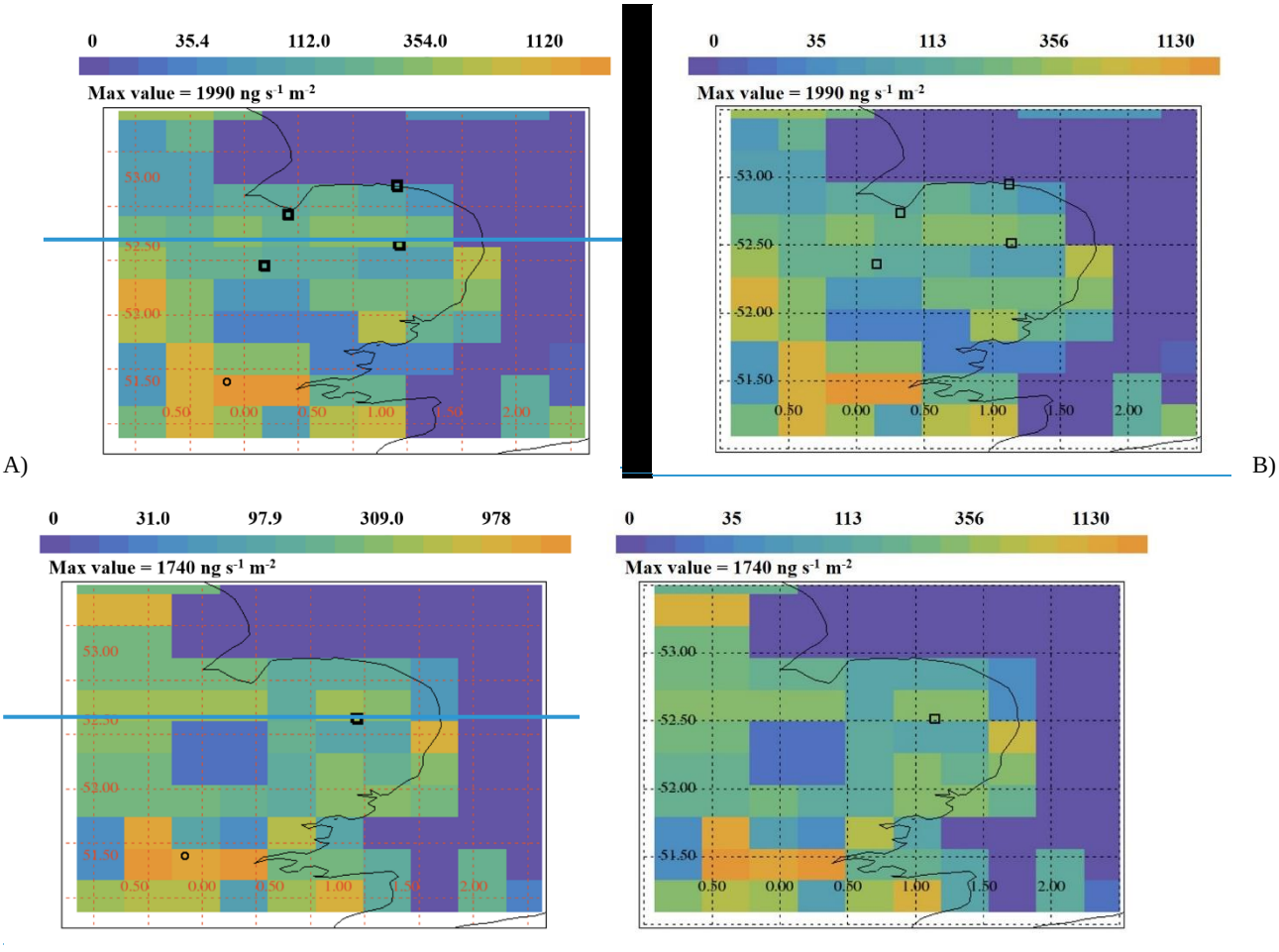


Figure 9: InTEM$_{2018}$ emission results from the national inversion A) using Mace Head, tall towers, and East Anglian station data, and B) using just Mace Head and tall tower station data. NAEI 2015 emission estimates on a 25 km grid (UK assumed 40% uncertain) are used as prior for inversion.

Table 4: Comparison of methane emission totals for an area roughly corresponding to Norfolk, Suffolk and Cambridgeshire. NB: Due to differing spatial resolution of the emissions grid, areas are not equal and thus are not directly comparable. Additionally, different cost functions were used in the two inversion methods and the standard deviation from InTEM$_{2014}$ (marked with a *) is not directly comparable with InTEM$_{2018}$.

	InTEM$_{2014}$	***InTEM$_{2018}$ without EA sites***	***InTEM$_{2018}$ with EA sites***
NSC area (kT yr^{-1})	80.4	82.3	84.9
1 standard deviation	3.3*	23.6	12.8

4. Summary and Discussion

We have employed a network of observations and an inversion system to estimate methane emissions over three counties in eastern England. This approach is conceptually similar to the ones used to estimate N_2O emissions in the United States mid-west (Nevison et al., 2018) and in California (Jeong et al., 2018) though ours is run on a smaller geographic scale and is trying to produce emissions at finer spatial resolution. To achieve this, measurements of methane from 4 sites in East Anglia were operated from 2012. The impact of local sources on the measurements was minimised by locating the inlets high in church towers in villages that are not part of the national gas distribution network.

These measurements were interpreted using a regional inversion approach based on the NAME inversion methodology and high resolution Met Office 3-D meteorological analyses at 1.5 km x 1.5 km horizontal resolution nested within coarser analyses at 25 km x 25 km horizontal resolution. Baseline values were calculated using measurements on the upwind side of the area being studied. This approach produces emission estimates with fine spatial resolution (up to 4 km x 4 km). The resulting total emission estimates are in good overall agreement with the UK NAEI bottom-up estimates with several notable differences in the distribution of emissions. One difference is in the Fens region of East Anglia where we find higher emissions. This could be due to emissions from managed wetlands currently being underestimated in the NAEI.

The NAEI contains a number of point sources (such as landfills) whose presence can be clearly seen in the inversion analysis, even though using no emissions prior is used within the inversion. This is borne out in a case study examining methane emissions from the Waterbeach landfill site (Riddick et al., 2017). It implies that there is real spatial information in the inversion results, and that a more refined uncertainty analysis would allow emission estimates from point sources to be derived from larger-scale analyses. Despite using a measurement-based approach to define the baseline, the level of knowledge of the methane concentration in the air entering East Anglia is a major cause of uncertainty in our analysis. The static value of 5 ppb to account for errors in the defined baseline is rudimentary and should vary with respect to time to reflect associated uncertainties. Approaches in which East Anglia is nested within a larger scale inversion, and thereby moving the boundary conditions further away, would be preferable (as was done in Section 2.2.5 and in ~~Approaches in which East Anglia is nested within a larger scale inversion would be preferable (~~Manning et al., 2011). Additionally, further work to improve the baseline calculation method could include solving for the baseline within the inversion (Lunt et al., 2016).

We have also investigated the impact of including the additional measurement sites in EA on calculated methane emission estimates in East Anglia using the national inversion approach InTEM$_{2018}$. Results from the inversion, which included the national GHG network stations (UK DECC network, Stanley et al., 2018; GAUGE tall towers, Stavert et al., 2018) and the EA network, show consistent results to those just using the EA network, demonstrating a stability in the inversion 'top-down' estimates. Benefits of the addition of the EA sites within InTEM$_{2018}$ were the ability to provide finer spatial resolution and to decrease the associated uncertainty for that area.

Author contributions

S. Connors, A. J. Manning, and N. R. P. Harris designed the experiments and S. Connors, A. J. Manning, and A. D. Robinson, carried them out. S. Connors and A. J. Manning developed the model code and performed the simulations. A. D. Robinson, S. N. Riddick and R. L. Skelton sources, installed and ran the measurement instrument in Sites 1, 3 & 4 (Haddenham, Weybourne – UCAM instrument, and Tilney) and provided data for the analysis. G. L. Forster, D. E. Oram, and S. Humphrey ran the measurement instrument in Site 3 (Weybourne – UEA instrument) and provided data for the analysis. Anita Ganesan, Aoife Grant, Kieran Stanley, and Ann Stavert, ran the measurement instrument in Site 2 (Tacolneston) as well as other data for the GAUGE network and provided data for the analysis. N. R. P. Harris and P. I. Palmer were project leads and gave scientific oversight and guidance throughout the planning, implementation, collection, and analysis of the data. S. Connors, A. J. Manning, and N. R. P. Harris prepared the manuscript with contributions from all co-authors.

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