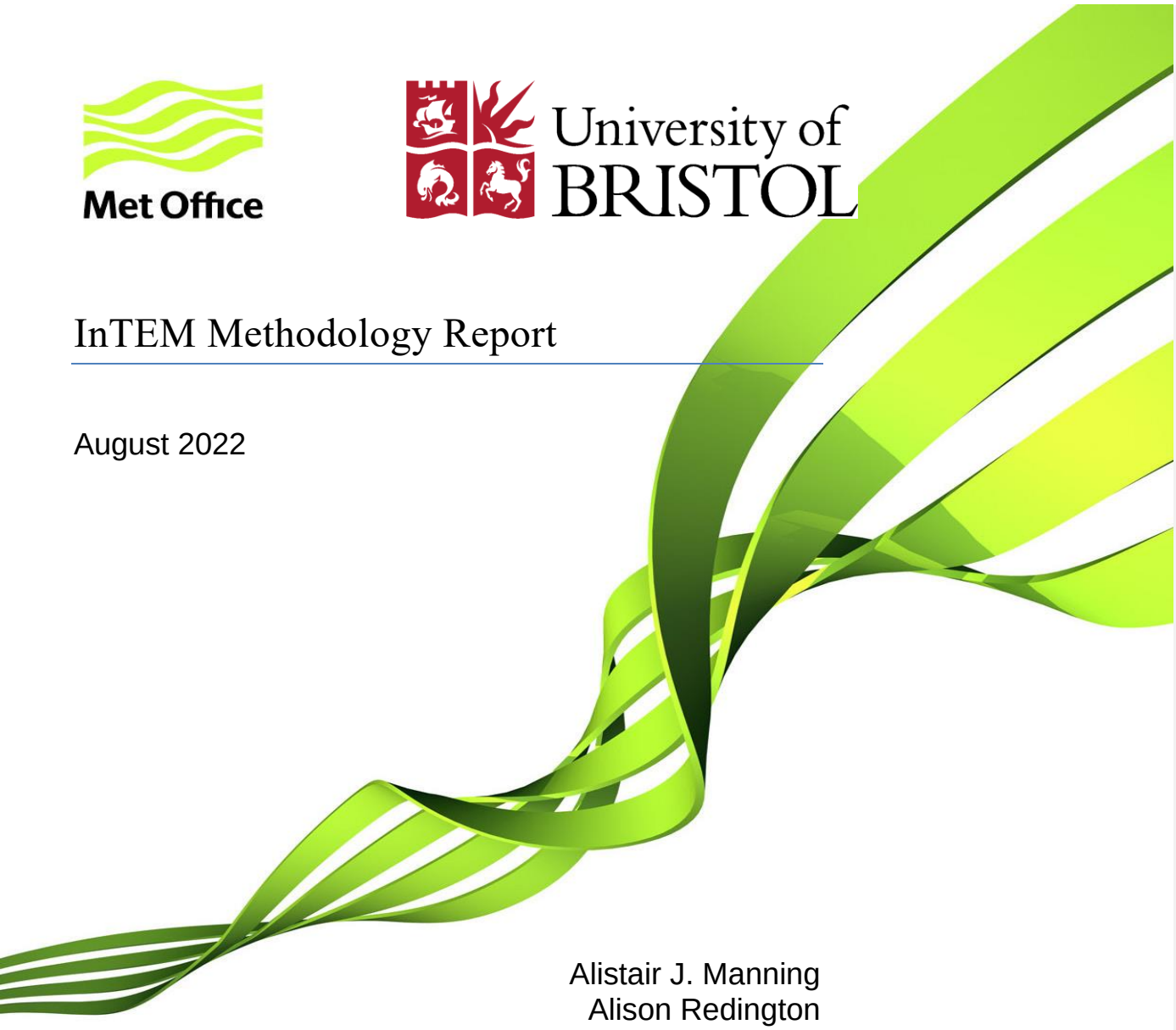


InTEM Methodology Report

August 2022

Alistair J. Manning
Alison Redington

Version 2022.1

Contents

1. Atmospheric Modelling

This section describes how we understand and estimate emissions (model) through investigation of where the air has passed over as it travels to the atmospheric observation station. We use the same three-dimensional time-varying description of meteorology that is used by the UK Met Office to understand our current weather and from which weather forecasts are produced. Wind speed and wind direction that vary in time, location and height, along with other important weather variables such as 'boundary-layer height', surface heating, atmospheric temperature and pressure, are extracted from this model and are used as input to the Met Office's Numerical Atmospheric dispersion Modelling Environment (NAME). This model describes how pollution moves and dilutes in the atmosphere.

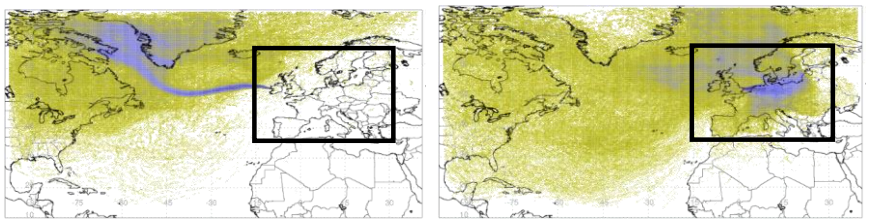
1.4 Meteorological Model

The time-varying three-dimensional weather data comes from the Met Office's operational weather forecast model called the Unified Model (UM). The UM is run every six hours and it predicts the weather globally. It is a grid-based model and currently it has a global horizontal resolution of 12 km and a UK horizontal resolution of 1.5 km. It has a vertical resolution of tens of metres near to the ground and then increases with height. It is under constant review and improvement and is one of the world's leading weather forecast models.

1.5 Atmospheric Transport Model

NAME is the UK Met Office's particle atmospheric transport model [*Jones et al.,* 2007]. It is a world-leading model that is operationally employed by the UK government to respond to a wide range of atmospheric dispersion applications including volcanic ash, dust, fire plumes, nuclear accidents and biological diseases such as foot and mouth virus [*Gloster et al.*, 2007; *Leadbetter et al.*, 2011, *Ryall et al.*, 1998, *Witham and Manning*, 2007]. It is actively developed and improved and is widely used across the UK research community. It principally uses the Met Office's UM time-varying, three-dimensional meteorology, but it can also use three-dimensional weather information from other meteorological centres such as the ECMWF (European Centre for Medium Range Weather Forecasting). NAME follows theoretical particles in the modelled three-dimensional atmosphere defined by the UM meteorology. By following many thousands of such particles, you gain an understanding of the likely spread and dilution of pollution in the atmosphere from different emission sources.

NAME is run backwards in time to estimate the previous 30-day history of the air before it arrives at *each measurement height* at *each observation station* in the UK DECC network (currently; Mace Head [MHD], Tacolneston [TAC], Ridge Hill [RGL], Bilsdale [BSD], Heathfield [HFD]) for each hour from 1989 to the current day (see Figure 1 for two examples). UK Met Office UM 3-hourly global data are used from August 2002 – current day. Its global horizontal resolution has improved from 40 km in 2002 to 12 km in 2017 and the vertical resolution has also increased from 31 levels in 2002 to 59 levels in 2015. From 1989 to Aug 2002 ECMWF (European Centre for Medium-range Weather Forecasting) ERA-Interim (Re-Analysis) data, with a ~80 km horizontal resolution and 37 vertical levels, are used. The ERA-Interim data are preferred over older UK Met Office UM data (pre-August 2002) as it uses more up-to-date physics, is run at a higher resolution and, as it is a re-analysis, its physics is constant over time. Over the UK, from 2012, we make use of high resolution (1.5 km, hourly) UM data nested inside the global data. The horizontal resolution of the NAME output is set at 25 km throughout (the Lagrangian nature of NAME means that this is independent of the resolution of the meteorology, higher resolution meteorology simply improves the description of the flow) and estimates the surface (0-40 m) impact of a large regional domain stretching from North America to Russia (-98º to 40º longitude) and North Africa to the Arctic circle (10º to 79º latitude) and extends from the ground to more than 19 km vertically. The three-dimensional locations and times when the model particles leave this regional domain are also recorded and used in the emission estimation process to improve the background (baseline) estimate.



(a) (b)

Figure 1: Examples of 2-hour air history maps derived from NAME (a) for MHD, baseline period (b) for TAC, regionally polluted period. The air-history maps describe which surface areas (0-40m) in the previous 30-days impact the observation point within a particular 2-hour period. The black box indicates the geographical domain used for the detailed emission estimates.

The NAME model is three-dimensional, therefore it is not just surface-to-surface transport that is modelled. The NAME particles are released in a 20 m vertical line centred on the height of the observation. An air particle can travel from the surface to a high altitude and then back to the surface. However only those times when an air particle

is within the lowest 40 m above the ground will it be recorded in the 'air-history' maps. Particles leaving the regional domain (shown above) at all elevations are recorded to improve the background (baseline) estimate at each station. For each hour, 20,000 inert model particles are used to describe the atmospheric dispersion. Running NAME backwards is very computationally efficient as every modelled particle has a direct impact on the air history maps produced. The time-integrated maps for each individual hour are also recorded and can be used to impose a short atmospheric lifetime on a gas, e.g. HFOs, or describe hourly varying fluxes, e.g. of CO_2 (future work).

1.6 Estimating Greenhouse Gas Emissions using Observations

The UK and European emission estimates are calculated using a sophisticated inversion methodology referred to as InTEM (Inversion Technique for Emission Modelling) and described in detail in *Manning et al.*, [2021], *Arnold et al.*, [2018], *Manning et al.*, [2011], and previously in *Manning et al.* [2003]. InTEM looks for the geographical emission map that, when diluted through atmospheric mixing produces modelled time-series' at each of the observation stations that best matches the actual observations recorded. The uncertainties in the observations and the modelling are taken into consideration when matching the modelled and observed time-series. Times when the model or the observations are more uncertain are made less important, through being de-weighted in the inversion, compared to those times when both the model and the observations are considered to be more certain.

Commented [MA1]: New reference

The InTEM methodology is flexible in that where observations (and uncertainties) are available from other additional monitoring stations they can be readily incorporated into the inversion system to better quantify, spatially and temporally, the estimated emissions. This flexibility has been readily demonstrated in our work on the EU project InGOS (Integrated non-CO_2 Greenhouse gas Observing System), using data from more than twenty stations across Europe, in the NERC (UK National Environmental Research Council) GAUGE (Greenhouse gAs UK and Global Emissions) project and the current NERC DARE-UK (Detecting and Attributing Regional Emissions in the UK) project. The Met Office was one of five inverse modelling groups involved in InGOS. The work in all of the projects clearly demonstrates the need for measurements that are robust and inter-comparable. Any biases in any of the measurement systems lead to errors in the emission estimation results. One of the key points is that all observations from across the UK DECC network are regularly and systematically inter-compared to ensure an un-biased, and robust, data set of observations.

The observations from each station are averaged (where the frequency of observation is greater than four hours) over each four-hour period and the standard deviation of the observations (provided there are at least three) over the three four-hour periods centred on the current period is used as part of the observational uncertainty. At frequent intervals, the observation systems measure specific tanks of air ('standards') to assess how the raw instrument signal is changing over time. We also use these multiple measurements of the standard to calculate individual measurement precision, which can then be applied to the air measurements (and makes up the second part of the observational uncertainty).

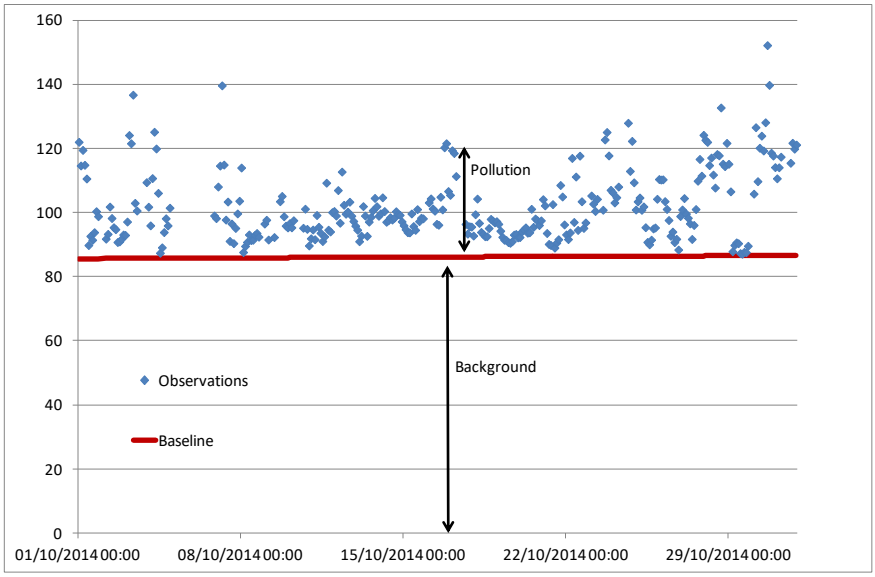


Figure 2: Time-series of observations of HFC-134a (greenhouse gas used in car air-conditioning) at a measurement site in the UK. The blue dots are the actual observations (in ppt). The red line is the estimated Northern Hemisphere background concentration (baseline). The difference between the measured concentration and the background concentration reflects the amount of regional pollution at this point in time.

The observed concentrations comprise two distinct components; (a) the mid-latitude Northern Hemisphere background concentration, referred to as the background or baseline, that changes only slowly over time, and (b) rapidly varying perturbations above the baseline (see Figure 2). These observed deviations above background are assumed to be caused by emissions on regional (European) scales that have yet to be fully mixed on the hemispheric scale. The magnitude of these deviations from baseline and, crucially, how they change as the air arriving at the stations travel over different areas, is the key to understanding where the emissions have occurred. The inversion system

considers all of these changes in the magnitude of the deviations from baseline as it searches for the best match between the observations and the modelled time-series. The best match should occur when the actual emissions most closely match the estimated modelled emissions.

The observation time-series, together with the NAME model output predicting the recent history and dilution of the air, are used within InTEM to estimate the geographic map of emissions. The minimisation technique, Non-Negative Least Squares Fit [*Lawson and Hanson*, 1974], is used within InTEM to derive these regional emission estimates based on a statistical skill score (cost function) comparing (best-fitting) the observed and modelled time-series at each observation site. The cost function is 'Bayesian' (i.e. improving a prior emission distribution through additional knowledge provided by the observations) and all of the uncertainties are defined as Gaussian (the statistical distributions are symmetric about the mean value, with an equal likelihood of being above or below the mean (truncated at zero)). The mathematical equation is given below.

$$C = (M'e' - y)^T R^{-1} (M'e' - y) + \left(e' - e'_p\right)^T B^{-1} \left(e' - e'_p\right)$$

Equation 1: Bayesian cost function used to minimise the mismatch between model and observations

$$C = \begin{pmatrix} Mismatch\ between \\ Modelled\ timeseries\ and \\ Observations \end{pmatrix} + \begin{pmatrix} Mismatch\ between \\ Emission\ solution\ and \\ Prior\ estimate \end{pmatrix}$$

Where,

C = Cost function score (the aim is to minimise this score)

M' = Dilution matrix from NAME

e' = Estimated emission map and baseline adjustment parameters

y = Measurements

R = Observation – Model uncertainty matrix

e_p' = Prior estimate of the map of emissions

B = Uncertainty of the prior estimate.

This equation is in two parts. The left-hand part describes the mismatch (fit) between the modelled time-series and the observed time-series at each observation station. The right-hand part describes the mismatch (fit) between the estimated emissions and any prior estimated emissions that might be available. In this work, because we want to

estimate emissions that are independent of the inventory process, we routinely remove this second part of the equation (by making the uncertainty of the prior, *B*, extremely large). In theory an independent prior could be used, however in practice all the inventories available use similar statistics, for example, population, and so they are not totally independent of the UNFCCC estimates. In specific circumstances we can include the right-hand part of the equation to investigate the impact of using all the available knowledge. The uncertainty matrix, *R*, is a critical part of the equation. It describes, for each four-hour time period, the uncertainty of the model and the observation. The different components of the uncertainty matrix are described in a later section.

The aim of the inversion method is to estimate the spatial distribution of emissions across a defined geographical area. The emissions are assumed to be constant in time over the inversion time period. The inversion time period depends on the number of measurements that are available. If there are only measurements from the MHD station, then the inversion time window is set to be two or three years. If there are measurements from two stations, then the inversion window is set to two years. For InTEM to estimate robust emissions of the UK it needs to be informed by sufficient information from the UK. Since MHD, on the west coast of Ireland, only sees the UK 20-30% of the time, it takes several years worth of data to build up sufficient information, hence the two- or three-year inversion window. More stations, especially those based in the UK, add significant quantities of information about UK emissions to enable shorter time periods to be used. For the special cases of methane (CH_4), nitrous oxide (N_2O) and sulphur hexafluoride (SF_6), where there are measurements available from four or more UK or Irish stations (2012 onwards), the inversion time period can be set to three-months or smaller. However, assuming the emissions are invariant over long periods of time is a simplification, but is necessary given the scarcity of the observations available. To compare the measurements and the NAME model time-series, the latter are output as a mixing ratio (e.g. parts per million [ppm]) using the modelled temperature and pressure.

The inversion domain is chosen to be a smaller subset of the full domain used for the air history maps. It covers 14º W – 31º E longitude and 36º N – 79º N latitude and is shown as the black box in Figure 1. The smaller inversion domain covers all of Europe and extends a reasonable distance into the Atlantic. It is good for the inversion domain to be smaller than the full domain to ensure re-circulating air masses (air that leaves the inversion domain but then re-enters at a later time) are properly represented and also it is computationally efficient to do so. The inversion domain is however large enough so

that emission sources outside, and very distant from the UK, have little discernible impact on the concentrations measured at the UK DECC network stations, i.e. the observed signal from sources outside the inversion domain are weak and rarely seen. The emissions in the six outer regions between the inversion domain boundary and the full domain boundary are also solved for in InTEM as 6 outer regions as shown in Figure 3.

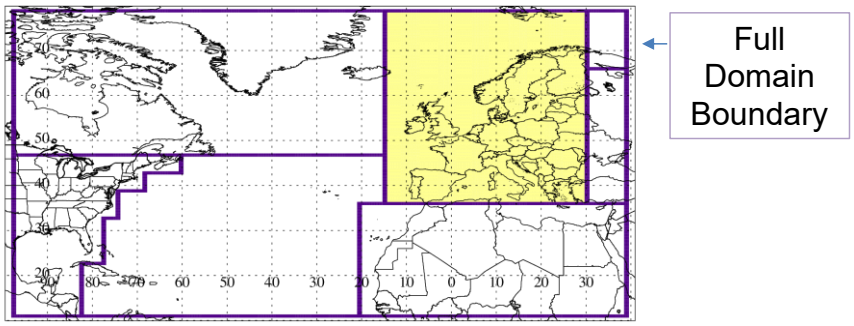


Figure 3: Map showing the 6 outer regions solved for in the inversion surrounding the main inversion domain, shown in yellow.

Monthly and annual estimates of emissions per gas per geographical region (UK, North-West Europe [NWEU]) are reported by calculating the mean of all the solutions that contain that month or year within the solved-for time period (e.g. any three-year inversion period that encompasses the year in question are included in the statistics for that year). An uncertainty standard deviation is estimated for each month or year for each gas and geographical region by calculating the average (assuming correlated uncertainties within the same time period) of the corresponding uncertainties. The inversion results will be compared to available inventories. Figure 4 shows an example of the time-series of emission output for the UK that is generated for methane using a variety of different inversion time-windows and station combinations.

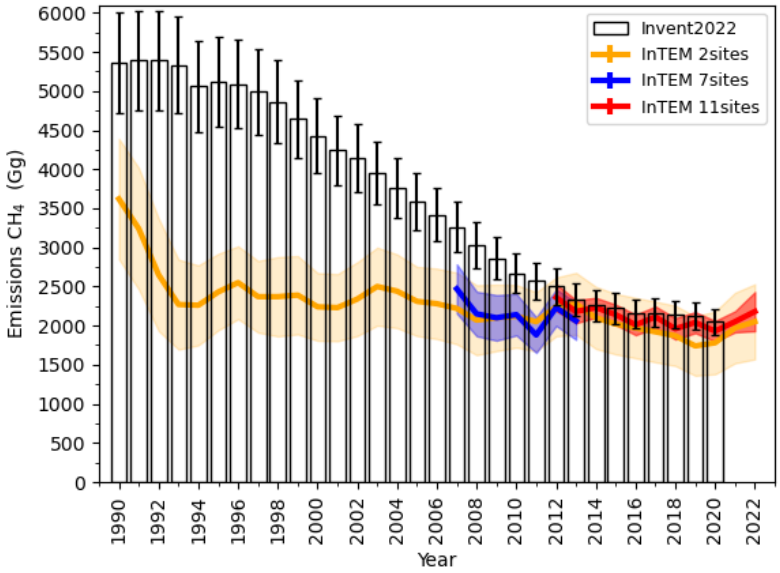


Figure 4: CH_4 emission estimates for UK (Gigagrams per year). Black columns are the UK UNFCCC submissions (April 2022) with uncertainty bars. The orange line is the inversion mean value for MHD and Cabauw (CBW) 2-year inversions and associated uncertainty. The blue line are annualised values for 2-month inversions with 7-sites (MHD, CBW, TTA, LUT, TRN, JFJ, CMN) and the red line are annualised values for 1-month inversions using the full UK DECC network and 3 EU AGAGE sites (11 stations), both with associated uncertainty.

3.3.1. Atmospheric 'Baseline' Northern Hemisphere Concentration

Baseline concentrations are defined as those that have not been influenced by significant emissions within the regional (inversion) domain (Figure 1a) as the air travels to MHD on the west coast of Ireland. These times are when the atmospheric concentrations are well mixed and therefore are representative of the mid-latitude Northern Hemisphere background concentrations. The analysis considers the long-term trend of the monthly and annual baseline mole fractions, their rate of growth and their seasonal cycle.

A two-hour period will be classed as 'baseline' if it meets certain criteria assessed using the NAME air history output:

- Local emissions particularly over land do not significantly contribute.
- Populated regions do not significantly contribute.
- The US, African and Eastern European 'Outer Regions' (see Figure 3) do not significantly contribute.
- The air mass has a dominant contribution from the north-west sector (chosen because southerly and south-westerly trajectories (potentially from the tropics)

can be depleted in trace gas concentrations, or influenced by the east coast of the USA, and easterly trajectories are influenced by Europe).

As an example, Figure 5 shows a three-month extract of the CH_4 observations measured at MHD. The observations have been colour-coded to indicate whether, using the above classification, the air mass they were sampled from was considered baseline. For the baseline analysis all non-baseline observations are removed.

The points defined as baseline using the above methodology still have a certain level of noise e.g. from unexpected emissions (forest fires in Canada or shipping); or incorrectly modelled meteorology or transport. To capture such events the daily mean of the observations when the baseline criteria are satisfied are used to estimate a daily baseline value. The daily values (when they exist) are fitted to a 4th order polynomial within a 180-day moving window centred on each day in turn. If there are insufficient data points in the 180-day window the fit is made either quadratic or linear. The uncertainty of the baseline is the root mean square error (rmse) of the fit. Each day therefore has 180 estimated baseline values. The daily baselines estimated when the current day is within 15 days of the centre of the fitted time window are averaged to estimate the baseline for the current day. The baseline uncertainty (σ_b) for the current day is the maximum of the rmse values within these 31 estimates. Days with fewer than 10 estimated daily values are defined to not have a baseline. The resulting daily time-series is then averaged up to monthly and annual values and presented to BEIS (e.g. Figure 6a).

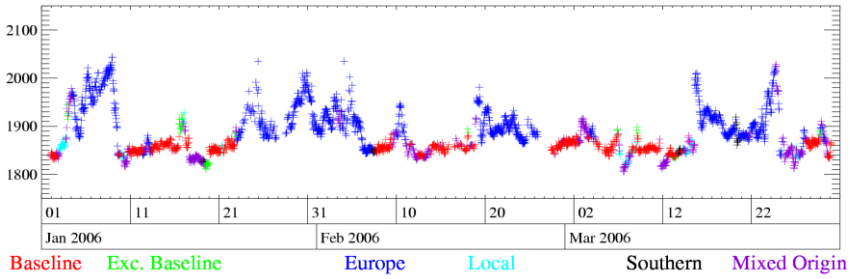


Figure 5: Example three-month time-series of Mace Head CH_4 observations (ppb) showing the impact of the baseline and non-baseline classification. The baseline observations are shown in red; Green = Excluded from baseline; Navy Blue = Polluted; Cyan = Locally influenced; Black = Southernly influenced; Purple = Non-baseline, mixed origin.

Monthly growth rates are estimated for each gas so that changes in the underlying trends can be identified and investigated. The daily baseline data are de-seasonalised

(three methods) before a local annual growth rate is defined for each day. These daily growth rate values are then averaged per month (and per year) to estimate the annual growth rate per month (and per year) for each gas and reported to BEIS (e.g. Figure 6b).

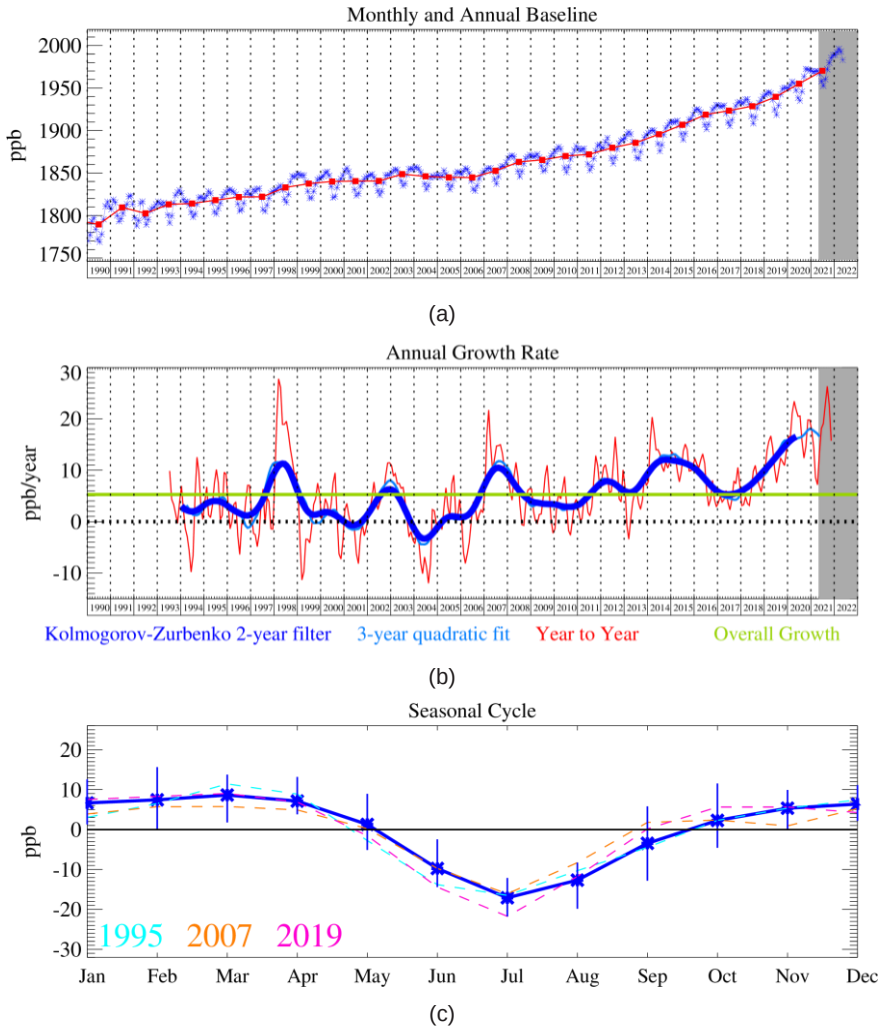
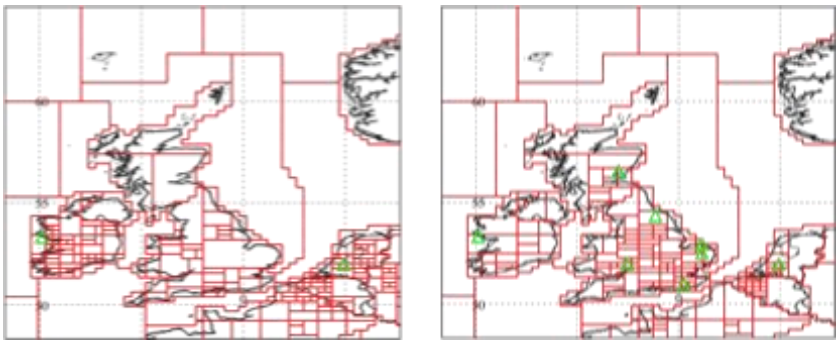


Figure 6: Example baseline analysis plot for methane (CH_4). (a) monthly (blue) and annual (red) mid-latitude Northern Hemisphere baseline mole fractions. (b) Monthly (seasonal cycle removed) baseline growth rate (3 methods) and overall growth rate (green). (c) Average seasonal cycle with year-to-year variability (uncertainty bars), the first (light blue), middle (orange) and last (pink) year of complete data are also shown. Grey shaded area indicates currently un-ratified data.

The seasonal cycle is estimated by subtracting the baseline concentration from the underlying trend value (as discussed in the previous paragraph). Estimates of the monthly seasonal cycles of each gas are also reported to BEIS (e.g. Figure 6c).

3.3.2. Estimating Emissions on a Gridded Domain

To obtain robust emission estimates for every area, an 'area' being defined as a collection of the native 25 km grid-boxes, within the inversion domain, each area needs to significantly contribute to the concentrations recorded by the observation network on a reasonable number of times. The model signal is calculated using a combination of the NAME model footprint and the current prior emission map. If the signal from an area is only rarely or poorly seen in the observation time-series then its impact on the cost function is minimal and the inversion method has little skill at determining emissions from that area. The contribution that different areas make to the observed concentration varies from area to area. Atmospheric dispersion makes areas that are distant from the observation sites contribute relatively little to the observations, whereas those that are in close proximity can have a large impact. To more equally balance the contribution from different areas, those areas that make a large contribution to the signal seen at the measurement station are split (starting from the country scale) into progressively smaller areas (the smallest grid used in this work is defined as 25 km). The starting areas are defined by country boundaries or collections of countries (e.g. Belgium and Luxembourg). Figure 7a shows an example of the grid that results when only MHD and CBW data at two-year resolution are used. Figure 7b shows an example grid with improved geographical resolution, particularly over the UK, when data from the full DECC network plus 3 European AGAGE stations are used at one-month resolution. The splitting varies for each inversion time period considered and between the different gases due to varying meteorology, the impact of missing observations and different priors.



(a) (b)

Figure 7: Example of the distribution of the different sized regions used by the inversion method to estimate regional emissions (smallest grid ~25 km): (a) MHD+CBW 2-year inversion period (b) Example 11-site UK DECC network + 3 AGAGE stations 1-month inversion period.

Integral to InTEM, as part of the setup process, the grid used to estimate the emission field is refined. A prior, with the NAME footprints, is used to calculate the starting grid. InTEM is then solved using the Bayesian function given above to estimate an emission distribution that best fits the observations given this grid. This InTEM solution is then used to calculate an improved grid. InTEM is then solved again with this new grid (note the original prior is still retained for this calculation). This is repeated multiple times until the grid resolution does not change. This iterative re-gridding allows significant point emission sources to be better resolved and estimated by InTEM.

3.3.3. Direction Specific Baseline

With the introduction of a network of stations it is necessary to define a baseline for each station across the network. A baseline for each station cannot be estimated in the same way as for MHD because the other stations within the network do not receive air that is unaffected by UK (and regional) emissions. The MHD baseline cannot be used directly for each observation station as the different stations do not receive air from the same direction and height as MHD at the same time. For example, TAC in East Anglia may be observing air from the north at the same time as MHD is observing air from the south-west. The impact of air from the upper troposphere is also important and is variable across the network at different times. Gas concentrations usually have a vertical and latitudinal gradient due to heterogeneous global emissions, i.e. more emissions may occur in the Northern Hemisphere where the majority of the land mass (and people) is located. It is therefore important to reflect these differing baselines within the inversion system. InTEM has a method that directly solves for adjustments to the MHD baseline

depending on the direction and height the air entered the modelled domain, so as these contributions differ across the network each station has a unique baseline time-series.

The direction and height the air enters the modelled domain is recorded for each observation time-step (e.g. 4-hours) for each observation station within the network. This information is interrogated and the percentage contribution from eleven different directions and heights are determined for each observation time-step for each station. The eleven directions are: WSW, WNW, NNW, NNE, ENE, ESE, SSE, SSW (all below 3 km); From the south 3-8 km; From the North 3-8 km; Above 8 km. Figure 8 shows a schematic of the different directions used in InTEM. These extra eleven variables are solved for in the cost function (in vector *e'* in Equation 1) which allows InTEM to separately describe the time-series of baseline influence at each observation station. In addition, the six outer regions shown in Figure 3 are also solved for in the inversion. In addition, for each station an additional two variables are solved for to account for any systematic negative or positive bias. Such a bias may arise through the observations or the transport model.

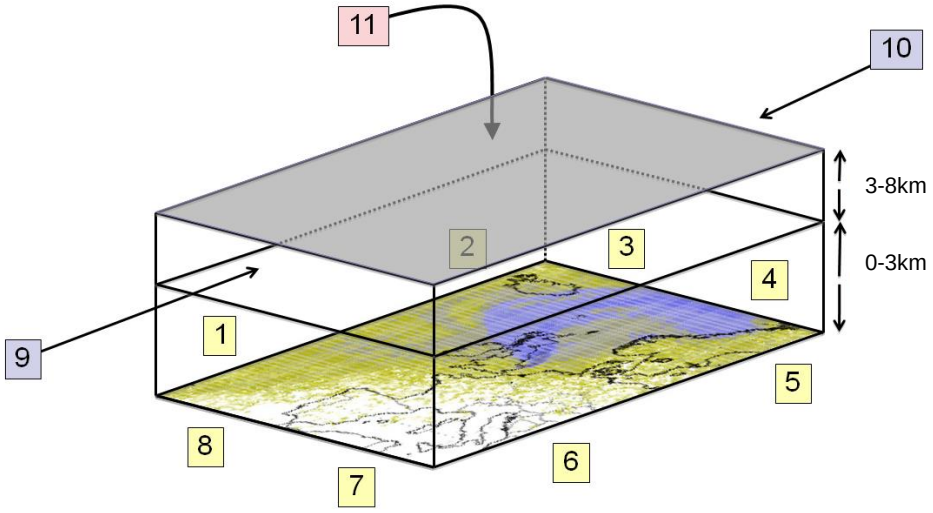


Figure 8: Schematic of the 11 different directions used within InTEM to describe the baseline.

1.7 Uncertainty

A Bayesian framework is used to assess the uncertainties in the inversion system. This framework has a rigorous mathematical method for estimating the uncertainty reduction due to increased knowledge, in this case more observations. The errors are assumed Gaussian (i.e. there is an equal and symmetric likelihood of the error being positive or

negative), this is an assumption that is widely used. For most of the gases investigated (methane being a possible exception) the emissions are always positive, i.e. the gas is emitted rather than being absorbed by the surface, therefore we truncate the errors so that the final emission cannot be negative (using a non-negative solver). A key point to note is that the uncertainties in the observations, in the model transport and also in the prior emissions are assumed well characterised and known. Robustly quantifying these three areas of uncertainty is extremely challenging and is an area of on-going research. The overall model-observation uncertainties are combined together to define the uncertainty matrix *R* (Equation 1). It is assumed that each four-hour period is correlated with those around it. This simulates the fact that any error in a four-hour period, say from the meteorological model, is likely to be felt in the preceding and following four-hour periods. This correlation is assumed to exponentially decay with a time correlation of twelve hours. Likewise, observation stations close to each other are potentially affected by similar errors and do not provide entirely independent information. Therefore, an exponentially decaying, geographical distance correlation of 200 km is also applied.

It is usually the case that the model uncertainty is much greater than observation uncertainty. The values in *R* vary from gas-to-gas and from one model-observation time-step (four-hours) to the next depending on different time-varying factors (described below), so there is no single number per gas. As a result, the uncertainty per annual UK estimate from InTEM varies from gas-to-gas and from year-to-year, but usually falls within 30-100% assuming very uncertain prior uncertainty estimates.

3.4.1. Uncertainty in the Observations

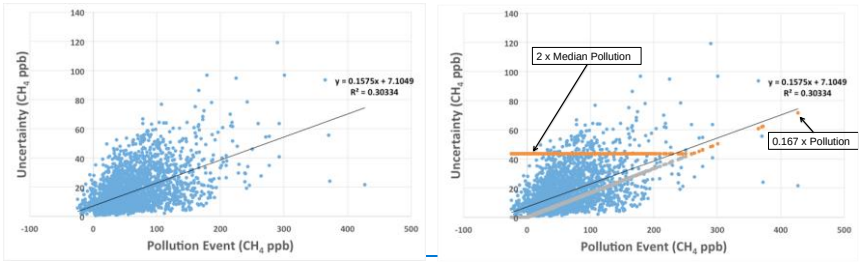
The observation uncertainty is derived from a combination of repeatability uncertainty and averaging uncertainty. The former is from the instrument and is the variability (expressed as a standard deviation (σ_r) observed when the same tank of air (standard) is repeatedly measured on the same day. The latter is from the variability (expressed as the standard deviation (σ_v) in the observations when they are averaged to the model time window (four-hours). The CH_4 observations are measured at ~0.3 Hz at the tower stations (although there are regular data gaps as different heights are sampled) and 40-minute intervals at MHD. For gases where there are fewer than three observations in a four-hour window, for example, for the gases measured on the Medusa, which takes one sample approximately every two hours, the variability across three contiguous four-hour time periods (the preceding, current and next time periods) is used to estimate this averaging uncertainty.

3.4.2. Selecting well-modelled observations

The model transport uncertainties are very difficult to quantify. They are assumed to be related to the height of the atmospheric boundary layer (ABL), the vertical profile of temperature, the wind speed, and the influence of the area local to the measurement point. When the air is well-mixed it is assumed the model will perform well. The ABL is the volume of air directly in contact with, and affected by, the ground surface, which varies from day to night and depends on the strength of the sun on the ground and the strength of the wind. If it is a hot day in summer a typical ABL in the UK would reach more than 2 km, whereas on a cold still night the ABL can fall below 50 m. The atmospheric turbulence (mixing) in the ABL ensures that ground surface emissions are readily mixed throughout the ABL. The mixing from the ABL to the free-troposphere, the volume of air directly above the ABL extending up to the tropopause, 10-12 km above the ground, can be much slower. Therefore, modelling the ABL correctly is vitally important to be able to understand the ability of the atmosphere to mix and dilute the surface emissions. If the ABL is high (1-2 km), an error in the modelled ABL of 50 m would not have a strong impact on the ability of the model to accurately model the atmospheric dispersion. However, if the ABL is low (50 m) an error of 10-50 m could be extremely significant. Also, times when the ABL is low are associated with times when the wind is weak, during such times local atmospheric flows, for example drainage flows in valleys and land-sea breezes, can be significant and these potentially sub-grid scale flows are not modelled well, if at all in the UM. It is also important to know the height of the ABL with respect to the height of the measurement inlet. If the ABL is close to the inlet height, small errors in the ABL can have a significant impact on the quality of the modelled concentration. The other key factor is the 'localness' factor, this is the sum of the nine grid cells in the NAME air history map centred on the observation location. When this is high, it means that the air is more stagnant around the station and therefore the flow will be more complex to model and more prone to uncertainty. It is akin to a low ABL condition. The mismatch between the modelled and actual topography at the station also complicates the modelling and adds to the uncertainty. At each observation station, four-hour periods when the modelled ABL is > 200m, when the wind speed does not vary much, localness is low (arbitrary 20 ng/m^3), the temperature profile relates to a near-neutral or unstable atmosphere (temperature gradient < 1.5 K/m) and the observations within the same four-hour period are similar at different heights (where available) are considered well-modelled and are used in InTEM.

3.4.3. Model uncertainty

Through analysis of the CH_4 measurements at observation sites in the UK DECC network with two or more inlet heights, it is clear that no one meteorological variable describes the model uncertainty well. It is assumed that the level of atmospheric uncertainty is directly linked to the difference in the observations measured at different heights within each inversion time step (four-hours). When the difference is small it implies the atmosphere is well mixed and can be well modelled, conversely when the difference is large, it implies the atmosphere has strong vertical gradients that are unlikely to be well modelled. The best descriptor of the difference between the two/three heights was found to be the size of the pollution event at any given time ~~(9a)~~. The average linear gradient across all years at all stations with multiple inlet heights was found to be 0.1 of the size of the pollution event. The minimum uncertainty was defined as the median pollution event over the inversion period (or year). This is used to define the model uncertainty (σ_a) that is applied to each accepted four-hour period.



~~**Figure 9: (a) Example of the uncertainty (variability of CH_4 between two observation heights) at TAC compared to the size of the pollution event above baseline. (b) Model uncertainty imposed for CH_4 at each time is shown as the orange points.**~~

3.4.4. Overall Model-Observation Uncertainty

The total model uncertainty is given by:

$$\sigma = \sqrt{(\sigma_r)^2 + (\sigma_v)^2 + (\sigma_a)^2 + (\sigma_b)^2}$$

Where,

σ_b = Baseline uncertainty (mole fraction)

σ_a = Model uncertainty (mole fraction)

σ_r = Observation repeatability uncertainty (mole fraction)

σ_v = Observation variability uncertainty (mole fraction)

The variance (σ^2) is calculated for each time-period (2-hours) and used to populate the main diagonal in the *R* matrix. The off-diagonals of *R* are populated with the cross-correlation terms:

$$\sigma_i \, \sigma_j \, exp\left(\frac{-\Delta t}{12hours}\right) \, exp\left(\frac{-\Delta x}{200 \, km}\right)$$

Where, *i* and *j* are the row and column respectively relating to two different observations, *Δt* is the time difference between the observations and *Δx* is the horizontal distance between the observation stations (if the observations come from different stations).

2. Bibliography

American Laboratory (2012), *Environmental and Atmospheric Monitoring Using Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS)* [Online]. Available: http://www.americanlaboratory.com/913-Technical-Articles/125787-Environmental-and-Atmospheric-Monitoring-Using-Off-Axis-Integrated-Cavity-Output-Spectroscopy-OA-ICOS/

Arnold, T., A. J. Manning, J. Kim, S. Li, H. Webster, D. Thompson, J. Muhle, R. Weiss, S. Park, and S. O'Doherty (2018), Inverse modelling of CF4 and NF3 emissions in East Asia; *Atmos. Chem. Phys.*, 18, 13305-13320, https://doi.org/10.5194/acp-18-13305-2018

Gloster J., P. S. Mellor, A. J. Manning, H. N. Webster, and M. C. Hort (2007), Assessing the risk of windborne spread of bluetongue in the 2006 outbreak of disease in northern Europe, *Veterinary Record*, *160*, 54-56

Jones, A., D. Thomson, M. Hort, and B. Devenish (2007), The U.K. Met Office's next generation atmospheric dispersion model, NAME III, in Air Pollution Modeling and its Applications XVII: Proceedings of the 27th NATO/CCMS International Technical Meeting on Air Pollution Modeling and its Applications, edited by Borrego, C. and A.-L. Norman, pp. 580-589, Springer, New York.

Lawson, C. L., and R. J. Hanson (1974), *Solving least squares problems*. Vol. 161. Englewood Cliffs, NJ: Prentice-hall

Leadbetter S. and M. Hort (2011), Volcanic ash hazard climatology for an eruption of Hekla Volcano, Iceland, *J. of Volcanology and Geothermal Research* , *199*, 230-241

Manning, A. J., D. B. Ryall, R. G.Derwent, P. G.Simmonds, S. O'Doherty, Estimating European emissions of ozone-depleting and greenhouse gases using observations and a modelling back-attribution technique (2003), *Journal of Geophysical Research*, 108, 4405

Manning, A. J., S. O'Doherty, A. R. Jones, P. G. Simmonds, and R. G. Derwent (2011), Estimating UK methane and nitrous oxide emissions from 1990 to 2007 using an inversion modeling approach, *J. Geophys. Res.*, 116, D02305, doi:10.1029/2010JD014763

Met Office (2012), *GC-ECD* [Online]. Met Office. Available: http://www.metoffice.gov.uk/atmospheric-trends/instrumentation/gc-ecd

Picarro Inc. (2015), *Cavity Ring-down Spectroscopy (CRDS)* [Online]. Available: http://www.picarro.com/technology/cavity_ring_down_spectroscopy

Ryall, D. B., R. G. Derwent, A. J. Manning, P.G. Simmonds, S. O'Doherty (2001), Estimating source regions of European emissions of trace gases from observations at Mace Head, *Atmospheric Environment*, Vol. 35, No. 14, 2507-2523

Witham, C., and A.J. Manning (2007), Impacts of Russian biomass burning on UK air quality, *Atmospheric Environment*, Vol. 41, pp. 8075-8090

World Meteorological Organisation (2012), 16th WMO/IAEA Meeting on Carbon Dioxide, Other Greenhouse Gases, and Related Measurement Techniques.

3. Glossary of Acronyms

ABL: Atmospheric Boundary Layer
AGAGE: Advanced Global Atmospheric Gases Experiment
ALE: Atmospheric Lifetime Experiment
BSD: Bilsdale observation station
CFC: ChloroFluoroCarbon
CH_4: Methane
CO: Carbon monoxide
CO_2: Carbon dioxide
CRDS: Cavity Ring-Down Spectrometer
CSIRO: Commonwealth Scientific and Industrial Research Organisation
DECC: Department of Energy and Climate Change
ECD: Electron Capture Detector
Empa: Swiss Federal Laboratories for Materials Science and Technology
ERA-Interim: ECMWF Re-Analysis meteorology – Interim product
EU: European Union
FID: Flame Ionisation Detector
GAGE: Global Atmospheric Gases Experiment
GAUGE: Greenhouse gAs Uk and Global Emissions
GAW: Global Atmospheric Watch
GC: Gas chromatograph
GHG: GreenHouse Gas
H_2: Hydrogen
H_2O: Water
HCFC: HydroChloroFluoroCarbon
HFC: HydroFluoroCarbon
Hg: Mercury
HgO: Mercuric oxide
ICOS: Integrated Carbon Observation System
InGOS: Integrated non-CO_2 Greenhouse gas Observing System
InTEM: Inversion Technique for Emission Modelling
LGR: Los Gatos Research
LSCE: Laboratoire des Sciences du Climat et de l'Environnement
MD: Multi-Detector
MHD: Mace Head observation station
MPI: Max Planck Institute
MS: Mass spectrometry
N_2O: Nitrous oxide
NAME: Numerical Atmospheric dispersion Modelling Environment
NERC: Natural Environment Research Council
NF_3: Nitrogen trifluoride
NOAA: National Oceanic and Atmospheric Administration
NWEU: North West Europe
OA-ICOS: Off Axis-Integrated Cavity Output Spectroscopy
ODS: Ozone Depleting Substance
PFC: PerFluoroCarbon
RGA: Reduction Gas Analyser
RGL: Ridge Hill observation station
SIO: Scripps Institution of Oceanography
SF_6: Sulphur hexafluoride
TAC: Tacolneston observation station
TTA: Tall Tower Angus observation station
UK DECC Network: United Kingdom Deriving Emissions linked to Climate Change Network
UM: UK Met Office Unified Model
UoB: University of Bristol
UNFCCC: United Nations Framework Convention on Climate Change
WMO: World Meteorological Organization