

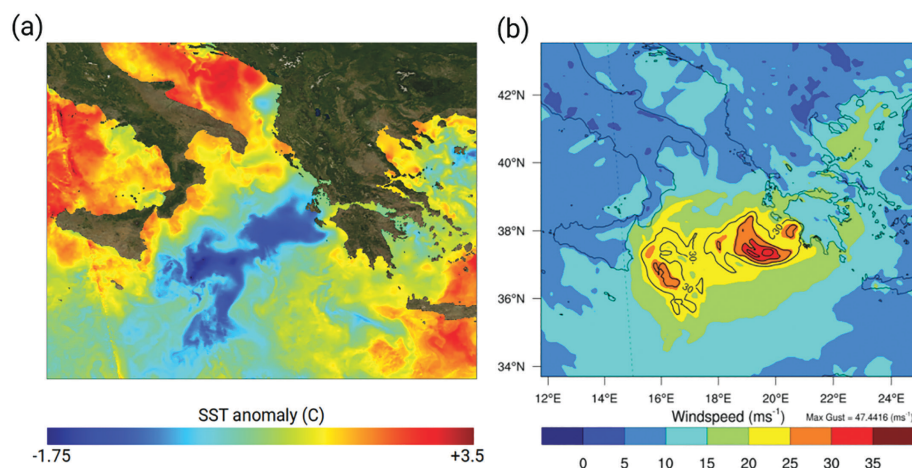


Figure 2. The storm induced sea surface temperature (SST) anomaly compared with modelled storm wind footprint. (a) Satellite observed SST anomaly valid 0000 UTC on 20 September (source: <https://marine.copernicus.eu/about-us/about-producers/sst-tac/>, accessed 21 September 2020). (b) Time-integrated maximum 10-m mean wind speed (shaded) and diagnosed gusts (contours every 5ms^{-1} starting at 30ms^{-1}) obtained from a 72h Weather Research and Forecasting (WRF) simulation initialised with ECMWF ERA5T reanalysis data at 0000 UTC 16 September 2020. (Credits: EU Copernicus Marine Service/ ECMWF. Contains modified Copernicus Climate Change Service Information 2020.)

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Correspondence to: David Smart

d.smart@ucl.ac.uk

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Development of an urban greenhouse gas modelling system to support a London monitoring network

D Hoare¹ , RL Jones², NRP Harris³, V Ferracci³, D Carruthers⁴, A Stidworthy⁴, E Forsyth⁴ and M Rigby¹

¹School of Chemistry, University of Bristol, Bristol, UK

²Department of Chemistry, University of Cambridge, Cambridge, UK

³Centre for Environment and Agricultural Informatics, Cranfield University, Cranfield, UK

⁴Cambridge Environmental Research Consultants, Cambridge, UK

Introduction

Under the United Nations Framework Convention on Climate Change (UNFCCC), developed countries are required to report their national emissions using greenhouse

gas (GHG) inventories, which combine data on GHG-producing activities (e.g. energy production or waste management) with emissions factors for each activity. While it is considered best practice for these 'bottom-up' methods to be evaluated using atmospheric data-based 'top-down' techniques, currently, only the United Kingdom (UK), Switzerland and Australia have included such methods in their National Inventory Reports (Brown *et al.*, 2019).

Cities are becoming a more important modelling scenario as various sub-national groups, including cities, universities and large companies, declare climate emergencies and produce policies designed to reduce GHG emissions in line with a +1.5 degC target (Masson-Delmotte *et al.*, 2018). The mayor of London has developed and begun enacting plans to make London carbon neutral by 2050 (Greater London Authority, 2018). These policies, the size of the city and the presence of a national-scale network (Stanley *et al.*, 2018) make

London a prime case study for the development of top-down, urban inverse modelling techniques.

Inverse models calculate the unknown cause of an observed effect rather than the more straightforward problem of predicting the effect of a known cause. For estimating GHG emissions, this means using observations of atmospheric composition, with atmospheric transport and statistical models, to calculate the causal GHG emissions. Atmospheric transport models can introduce substantial uncertainty into the process. For this reason, the models need to be evaluated in the context of new scenarios such as cities.

The two most abundant long-lived GHGs are carbon dioxide and methane, and both gases have been the subject of top-down investigations in other cities (using different models than those used in this study), such as Boston, USA (McKain *et al.*, 2015; Sargent *et al.*, 2018); Los Angeles, USA (Verhulst *et al.*, 2017); and Paris, France

(Stauffer *et al.*, 2016). Because emissions inventories only quantify anthropogenic sources, any influence from the biosphere must be accounted for. However, it is an ongoing research challenge to isolate anthropogenic carbon dioxide emissions in top-down studies as the biosphere (the photosynthesis and respiration of plants) can dominate the observations (White *et al.*, 2019). For this reason, our study focuses on modelling methane, whose emissions are overwhelmingly anthropogenic in London and the southeast of the UK. Furthermore, methane inventories are thought to be more uncertain than those of carbon dioxide ($\pm 16.7\%$ for methane vs $\pm 2.9\%$ for carbon dioxide in the UK inventory (Brown *et al.*, 2019)) and could benefit more from top-down evaluation.

London's methane emissions are estimated in the 2016 National Atmospheric Emissions Inventory (NAEI) to be 62kT per year, which is 3.1% of the UK total, despite London only occupying 0.64% of the UK's land surface (using Office for National Statistics data). Cities across the UK are similar hotspots of methane emissions due to the density of gas transmission infrastructure and waste management facilities. Verified emissions reductions in London may provide evidence for similar policies to be used in other cities.

The London GHG network 'London GHG' will comprise around 10 high-frequency instruments distributed across the city. To minimise difficulties in modelling the urban roughness layer, instruments are primarily being set up on buildings that lie high above the local urban canopy, such as lone tower blocks and tall church spires. In this paper, we will present and discuss early results from the measurements and modelling of a test site in central London. These results will inform future stages of the London GHG project.

Methods

We have established an initial measurement site at the Thames Barrier in central London (51.497°N, 0.037°E). This site measures carbon dioxide and methane using a Picarro G2401 cavity ringdown spectrometer, which performs a measurement every 5s with a precision of approximately 50 parts per billion (ppb) for carbon dioxide and 1ppb for methane. These measurements are known as mole fractions, which is the atmospheric concentration of the gas measured as the fraction of particles in the air of the gas being observed. This instrument is similar to those installed in the national-scale UK Deriving Emissions linked to Climate Change (UK DECC) network (Stanley *et al.*, 2018). In this article, we will examine the initial period of data collected from 5 May 2018 to 31 July 2018.

We combine two bottom-up inventories to use as our emissions in this work, with NAEI used over the UK and Emissions Database for Global Atmospheric Research (EDGAR) used in surrounding countries. The NAEI is a gridded inventory produced by the UK government and provides a resolution of $1\text{km} \times 1\text{km}$, which can identify emissions within London, while EDGAR is produced by the European Commission Joint Research Centre at $0.1^\circ \times 0.1^\circ$ (approximately $10\text{km} \times 10\text{km}$ in the UK). The latest versions of both inventories available at the time of writing are used, which are 2016 for the NAEI and 2012 for EDGAR. Both inventories provide annual mean estimates but unfortunately do not include any seasonal or diurnal time variations nor spatial uncertainty estimates.

Within London, NAEI methane emissions are predominantly due to waste water treatment and leakages in the domestic gas distribution system. Emissions from the gas network are roughly distributed by population in the inventory, while waste emissions are centred on multiple emission hotspots across the city, as shown in Figure 1. These hotspots may provide a challenge for atmospheric modelling as they are of a size similar to, or smaller than, the model resolution.

Two models are required to infer GHG emissions from atmospheric concentrations: a physical model and a statistical

model. The physical model is usually an atmospheric or chemical transport model that estimates the atmospheric concentration at a given location and time using emission (flux) data and meteorological input. The statistical model compares the modelled and observed concentrations and calculates the emissions field that enables the model to best replicate observations, subject to various constraints (Ganesan *et al.*, 2014). In this work, we focus on analysing and comparing the performance of the two physical models in an urban environment. We use two models that work quite differently in order to identify the best path forward for future modelling in the London GHG project.

The first of the two physical models used in this work is the Met Office Lagrangian particle dispersion model, the Numerical Atmospheric-dispersion Modelling Environment (NAME) (Jones *et al.*, 2007). Atmospheric transport is simulated in NAME as the advection and diffusion of thousands of particles, which are tracked backwards in time from the measurement location, recording where they pass near (within 40m of) the surface – the assumed source of emissions (Manning *et al.*, 2011) (Figure 2). The model provides estimates of observation sensitivities known as 'footprints', which are 2D fields that map how much the different regions in the emissions

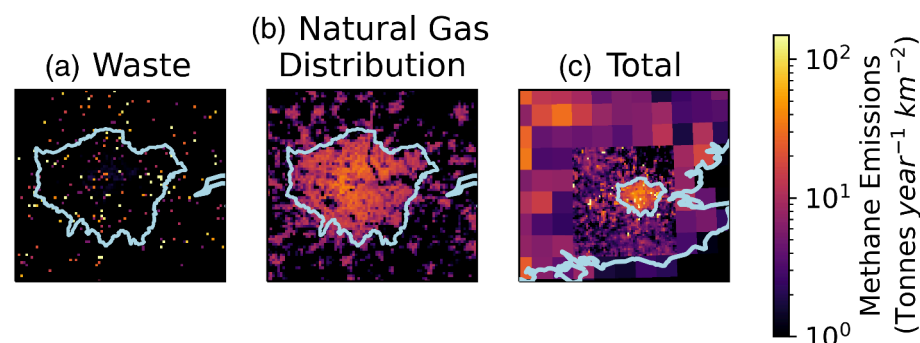


Figure 1. Methane emissions from NAEI 2016 dataset across London for the two largest sectors; (a) waste treatment and disposal, (b) natural gas distribution – labelled 'offshore' in the inventory and (c) the total methane emissions on the model grid. Outlines show UK coast and London boundaries.

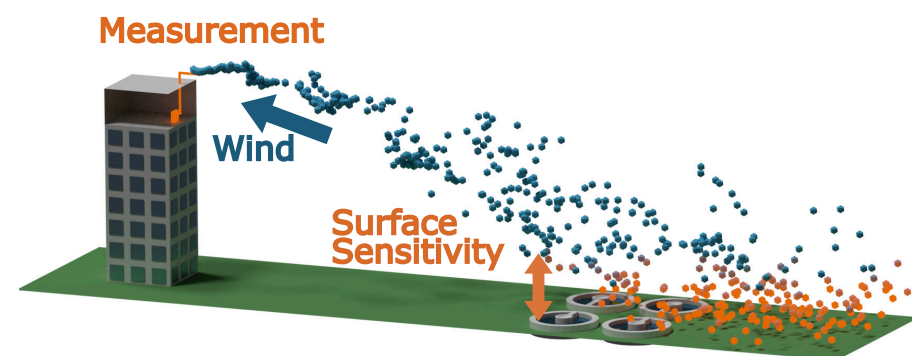


Figure 2. Schematic depiction of a Lagrangian particle dispersion model, such as NAME. Each sphere represents a modelled particle, which is released from the measurement location and transported backwards in time through advection and diffusion, and its passage near the surface is recorded to estimate where the air may have picked up methane emitted from the surface.

field contribute to the observed atmospheric concentration of the gas for each measurement. The model also estimates where and when particles leave the domain so that boundary conditions can be accounted for. Mole fractions at the measurement site can be estimated as the product of each footprint and the emissions field, plus any contribution from the mole fraction at the boundary of the domain. The domain and boundaries used in this work are shown in Figure 3. The boundary conditions are taken from the Copernicus Atmosphere Monitoring Service global methane products, which use satellite measurements and models to produce global four-dimensional methane fields (Inness *et al.*, 2019), adjusted to better match background measurements at Mace Head, Ireland.

The NAME model was run offline using Met Office Unified Model meteorology. We use the high-resolution (1.5km) UKV meteorological data, where available, and the approximately 12km-resolution global dataset elsewhere. While the UKV meteorology has a high enough resolution to resolve urban-scale phenomena such as the urban heat island, NAME itself does not explicitly account for urban turbulent transport. Footprints and emissions are combined in a multiple-resolution grid shown in Figure 1(c), with London and its surroundings in a high-resolution ($0.032^\circ \times 0.021^\circ$, ~2.5km) grid embedded in a low-resolution ($0.352^\circ \times 0.234^\circ$, ~25km) grid used for previous national modelling (Lunt *et al.*, 2016).

The second physical model used is ADMS-URBAN produced by Cambridge Environmental Research Consultants (Stocker *et al.*, 2012; Hood *et al.*, 2018). This model is designed specifically to model urban environments at a very high (street level) resolution, taking account of complex features such as the effect of buildings. ADMS-URBAN differs from NAME in several key ways: ADMS-URBAN can explicitly represent large numbers of individual sources, including point sources (with specified heights) and road sources, but is limited in domain, and the concentration downstream of each source is represented by an analytic distribution that, for point sources, is Gaussian in neutral and stable conditions and skewed Gaussian in unstable conditions but has other more complex forms for road sources. The concentration distribution is stationary in time for each successive hour and may use single-site or gridded meteorology to calculate the footprint. Here, we drive ADMS-URBAN with meteorological measurements from Heathrow Airport. These measurements are internally modified according to the difference in roughness lengths from the urban landscape at Heathrow and the Thames Barrier, resulting in a lower windspeed. This is the same setup that has been successfully used for model-

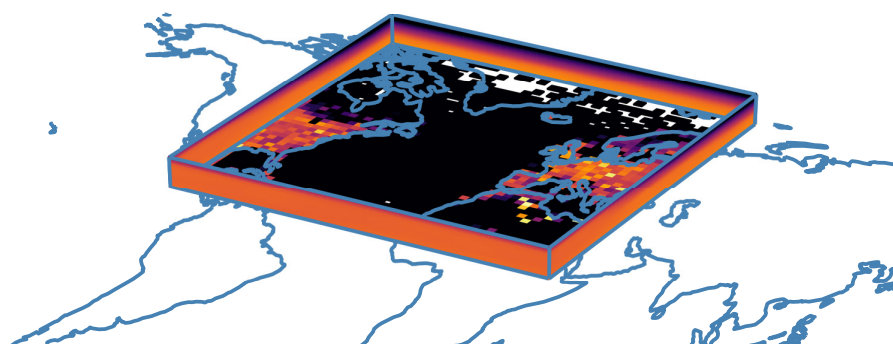


Figure 3. Domain and boundaries for NAME; height not to scale.

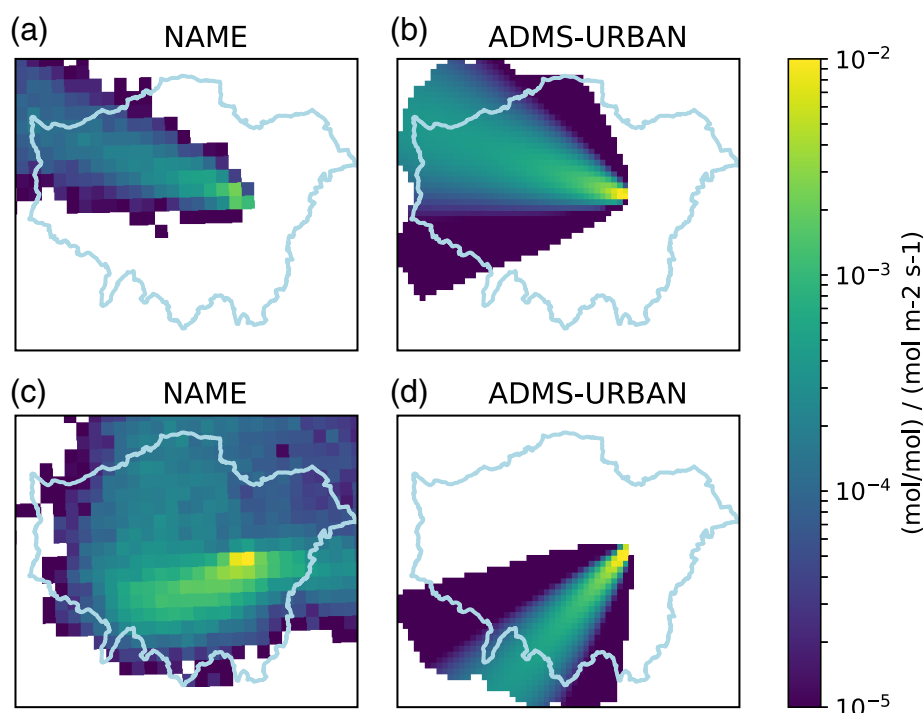


Figure 4. NAME and ADMS-URBAN footprints over London for (a, b) calm weather 1500 UTC, 10 May 2018, and (c, d) a passing front at 1500 UTC, 24 May 2018. Single-site meteorology ADMS-URBAN agrees qualitatively with NAME using gridded meteorological input in most weather conditions over London, but differences are found in the overall magnitude of the footprint and during complex meteorological conditions (e.g. passage of fronts).

ling air quality in London (Hood *et al.*, 2018). The boundary layer height is calculated internally as opposed to NAME, which uses the value diagnosed in the Unified Model. In this study, the domain for ADMS-URBAN is the same as that used by the London Atmospheric Emissions Inventory, which encompasses all London boroughs and everything within the M25. As ADMS-URBAN does not estimate the influence of fluxes outside London or regional boundary conditions, the ADMS-URBAN footprint requires additional information so that the total methane concentration can be simulated. In this study, we embedded ADMS-URBAN footprints within the larger-scale NAME footprints. The ADMS-URBAN footprints are coarsened to match the NAME high-resolution grid (~2.5km) and thus loses some spatial information as the grid carto-

graphic projections are otherwise incompatible. The geographic extent of London used throughout the paper is taken from the OpenStreetMap London administration polygon, rasterised onto the NAME high-resolution grid.

Results and Discussion

Examples of NAME and ADMS-Urban footprints are shown for two different meteorological conditions in Figure 4. The top row shows footprints under steady westerly winds at 1500 UTC, 10 May 2018, whereas the bottom row shows footprints at 1500 UTC, 24 May 2018, under more complex conditions, with fronts passing over London (Figure 5). Under the steady westerly winds, both footprints are qualitatively similar, with observations at the Thames Barrier being influenced

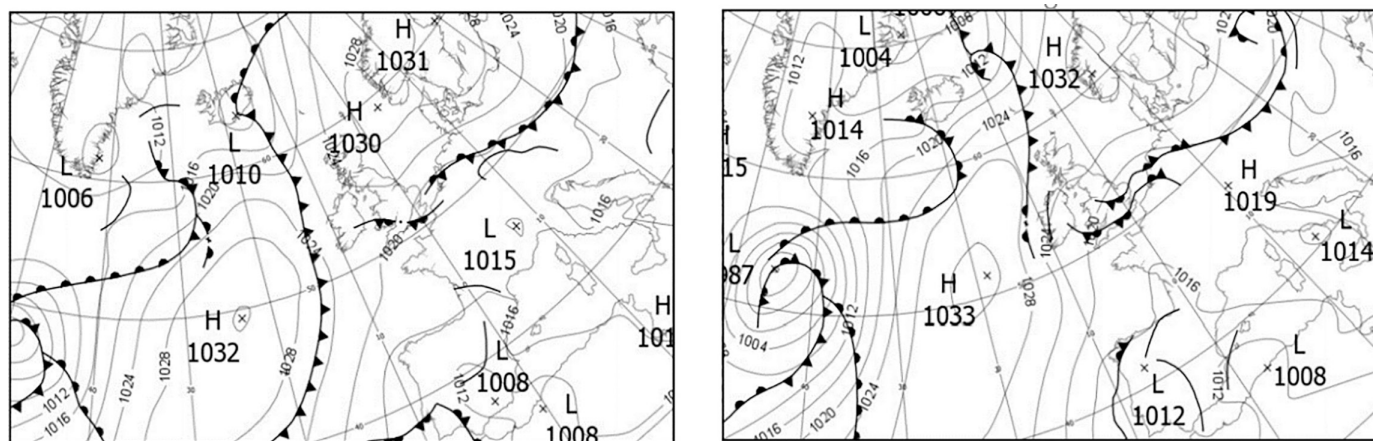


Figure 5. Met Office analysis charts for 1200 UTC 24 May and 0000 UTC 25 May 2018, showing the passage of fronts through London. (Reproduced from the Met Office Daily Weather Summary 2018, © Met Office Crown Copyright.)

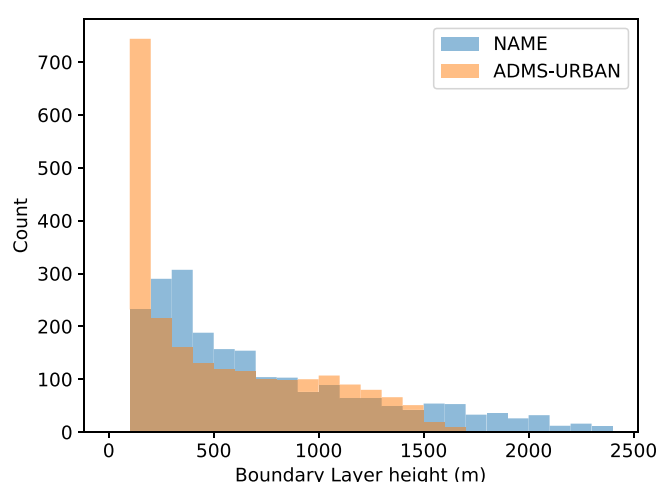


Figure 6. Histogram of boundary layer heights used by NAME and ADMS-URBAN. The peak in ADMS-URBAN data is due to the lower limit of 120m used by the model. Lower boundary layer heights result in higher sensitivity to emissions, all else being equal.

by fluxes from western and central London, although the ADMS-URBAN footprint is four times more sensitive to emissions when both models are integrated over London. Under the more complex meteorological scenario, the NAME footprints indicate sensitivity to a wider area of London with nearly twice the total London sensitivity as ADMS-URBAN, presumably reflecting the range of wind directions experienced by the model particles, whereas ADMS-URBAN shows sensitivity to a narrower region upwind of the measurement site.

On average, ADMS-URBAN is about twice as sensitive to London fluxes as NAME, with a mean (5th–95th percentile) total London sensitivity of 0.97 (0.24 – 2.96) ($\text{mol m}^{-2} \text{s}^{-1}$)⁻¹ compared to 0.43 (0.09 – 1.39) ($\text{mol m}^{-2} \text{s}^{-1}$)⁻¹ for NAME. One possibility for the difference in sensitivity is the internal boundary layer height used by each model. Figure 6 shows a histogram of the boundary layer heights, demonstrating ADMS-URBAN's overall shallower boundary layers. The boundary layer is important in determining surface sensitivity as it limits the vertical mixing of air. In the

models, this increases surface sensitivity, reflecting how low boundary layers trap GHGs and increase their atmospheric concentration near the surface.

Figure 7(a) shows the hourly median and 33rd–66th and 5th–95th percentile ranges of methane observations at the Thames Barrier between 5 May 2018 and 31 July 2018 inclusive. Observed mole fractions are generally higher and more variable at night than during the day, and the lowest values observed are typically observed during the daytime. This difference is thought to be largely due to diurnal changes in atmospheric stability, with stable nocturnal boundary layers trapping locally emitted methane in contrast to strong mixing of nearby sources during the day (Stull, 1988).

Figure 7(b) shows the mean observed mole fractions as a function of wind direction and wind speed (from the Met Office UM analysis meteorology as measurements were not made at the Thames Barrier), which highlights that the highest observed concentrations occur at low windspeeds

and/or from an easterly direction, with a spot of high emissions from the northeast. There are several possibilities why easterly winds are associated with higher methane concentrations. The first reason is that these winds are likely to be carrying emissions from mainland Europe, with the Benelux region being particularly high in emissions according to the EDGAR inventory. In contrast, when winds come from the west, they arrive in the UK or Ireland with mole fractions consistent with the hemispheric background. A contribution from local sources is also possible, with several large methane emission hotspots within several kilometres of the Thames Barrier, according to the NAEI. For example, emissions from the Beckton Sewage Treatment Works approximately 4 km away may be consistent with the maximum rise in the mole fraction at around 50° . Mole fractions associated with this wind direction tend to be highly variable, suggesting a nearby plume impinging on the measurement site, rather than a more well-mixed regional source. Data from the addition sites planned around London could help distinguish between these two cases by providing different viewpoints on local emissions.

By combining the footprints for NAME or ADMS-URBAN (embedded within NAME) with the NAEI and EDGAR emissions fields, we can produce a modelled time series that can be compared to the Thames Barrier data. An example for a typical 2-week period is shown in Figure 8. The modelled mole fractions are attributed to three different factors: fluxes from within London, fluxes outside London and contribution from the boundary conditions at the edge of our NAME domain. The two models only differ in their modelled London contribution as the ADMS-URBAN footprints are embedded into the NAME-derived regional footprints and boundary conditions. The full period mean and 5th–95th percentiles of the mole fraction due to sources within London for

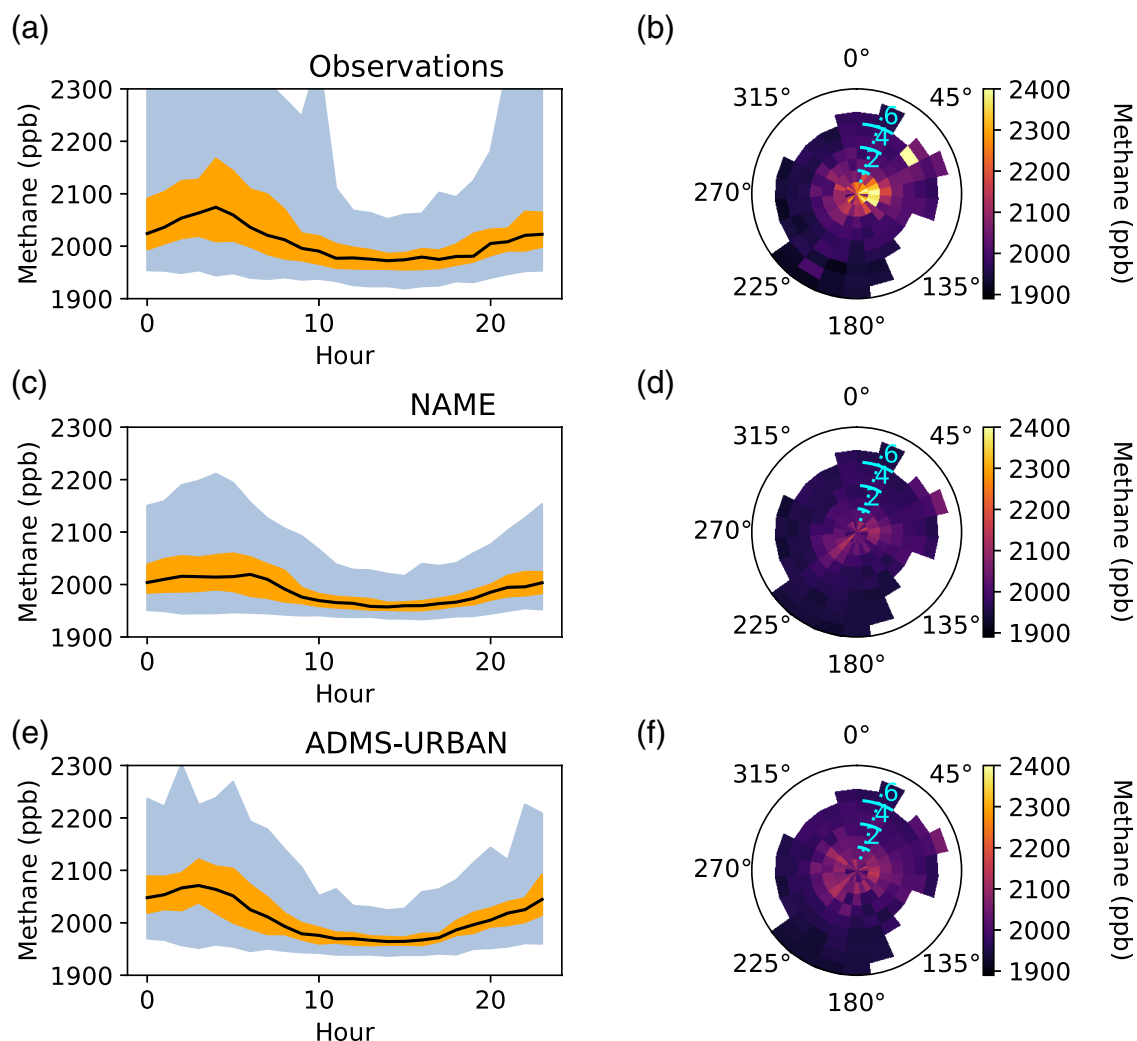


Figure 7. (a, c, e) Methane hourly median (black line) and 33rd to 66th (orange area) and 5th–95th (blue area) percentile range mole fractions and (b, d, f) rose plot (angular: wind direction, radial: wind speed in ms^{-1}) for the Thames Barrier (51.497°N , 0.037°E) for (top) observations, (middle) NAME and (bottom) ADMS-URBAN between 5 May 2018 and 31 July 2018. Extreme values in a are not shown to allow a clearer comparison to model values.

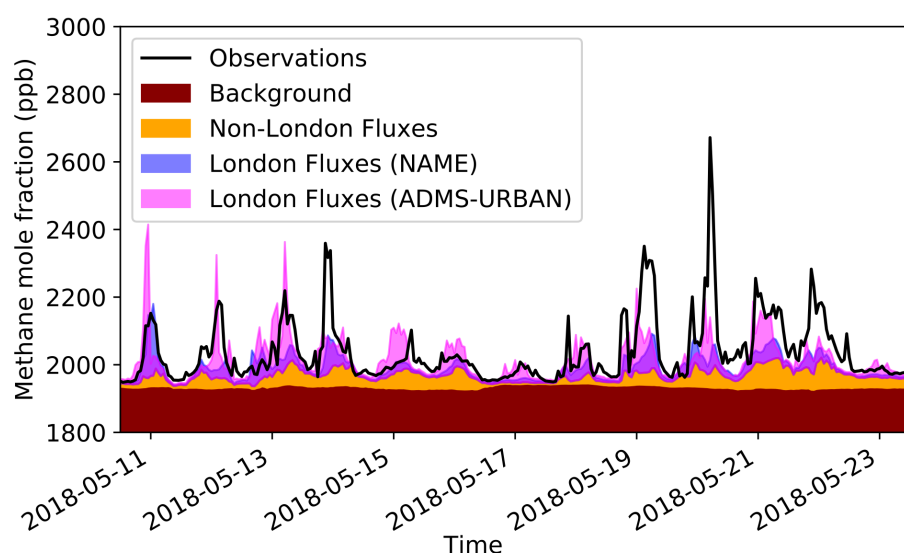


Figure 8. Time series comparison of modelled and observed fluxes for both NAME and ADMS-URBAN embedded in NAME. The different colours represent the contribution from different geographical regions, with the Background and Non-London Fluxes being the same between the two models. The purple segments show where the two London contributions overlap.

NAME and ADMS are 34.2 (4.37–121) ppb and 55.9 (9.30–173) ppb, respectively, compared to 45.2 (9.67–113) ppb from regional

sources and 1921 (1910–1937) ppb from the boundary conditions. The modelled concentrations generally capture the observed

diurnal cycle, although the magnitude of the night-time peaks can differ from the observed data by around a factor of two or more. NAME mostly underpredicts methane concentration, while the ADMS-URBAN model underpredicts on some nights and overpredicts on others.

Figure 7(c–f) shows the hourly medians and wind dependence for the observed, NAME and ADMS-URBAN modelled mole fractions. From the hourly medians, the night-time underestimation seen in Figure 8 is more evident. Both models show an increase in mean mole fractions at low wind speeds, but at a much lower magnitude than in the observations. This finding could be because nearby sources (within a few km) are larger than estimated in the inventory, or it could show that the models tend to overestimate mixing during low-wind conditions, with both possibilities suggesting the high observations are not primarily due to the Benelux region. The hotspot to the northeast is also not captured in the models, which may indicate that a source in this direction is not present or underestimated in the inventory, or it could show that

model transport is generally too dispersive for this wind sector.

Figure 9 shows the modelled mole fractions plotted against the observations for the two dispersion models, for total concentrations and the London contribution only. For this analysis, the data were filtered to retain only points where the observational variability within each hour period was less than one half of the modelled London contribution. This removes points heavily influenced by local emissions that the models are not expected to capture accurately. Summary statistics are shown in Table 1. Overall, the models show broadly similar correlations with the data, despite their very different architectures. The NAME model has a slope of regression greater than 1, suggesting that the emissions or modelled sensitivities are underestimated. The opposite is true for the ADMS model, although the line of regression is skewed by a small number of points where the

model greatly overpredicts methane concentrations. For both models, the R^2 value decreases when looking at just the London contribution, perhaps because they struggle to accurately represent complex urban meteorology or because of errors in the distribution of nearby emissions sources in the NAEI. During the most well-mixed conditions (between 1100 and 1700, when hourly observation variability is below 5 ppb), the models are in closer agreement but show lower sensitivity to London emissions than at other times. Overall, model output from NAME correlates more strongly with the observations than ADMS-URBAN, perhaps due to the use of three-dimensional meteorology compared to single-site meteorology. However, ADMS-URBAN better captures the diurnal cycle present in the observations, possibly due to the different boundary layer height calculations used, although there could be many factors that contribute to both differences between the models.

These simulations show that NAME and ADMS combined with the NAEI can capture some of the major features in a methane mole fraction time series at an urban site. The two models show similar features in their simulated mole fractions, despite a different modelling approach and driving meteorology, which suggests that a substantial portion of the model–measurement mismatch is due to the differences between the truth and inventory emissions magnitude, distribution and/or temporal variability. The next step in the development of a modelling system to support the London GHG network is to develop a new statistical model, an inverse modelling system that can determine whether changes in emissions and their distribution can improve the fit between the model and the data (Lunt *et al.*, 2016). The differences between the models will lead to differences in inferred emissions from an inverse modelling system. These differences will capture some of the sensitivity of the inverse models to atmospheric transport error and can help better inform interpretation of inferred emissions as a result.

Conclusion

As the first step in the development of a network for monitoring of London's carbon dioxide and methane emissions, we have established a continuous measurement site on the Thames Barrier. We analysed methane data from this site during the summer of 2018 and compared the observations to two distinct atmospheric transport models, NAME and ADMS-URBAN. Results showed that, over a 3-month period, the models could capture some of the broader features in the data, such as the diurnal cycle and wind direction dependence. The consistency of the difference between the model prediction of some of these features and the data suggests that a substantial proportion of the model–observation discrepancy is due to errors in the emission inventories.

We will use both models in a future emissions estimation framework to provide some estimate of the sensitivity of the derived emissions to atmospheric transport model errors. Further work towards a London GHG

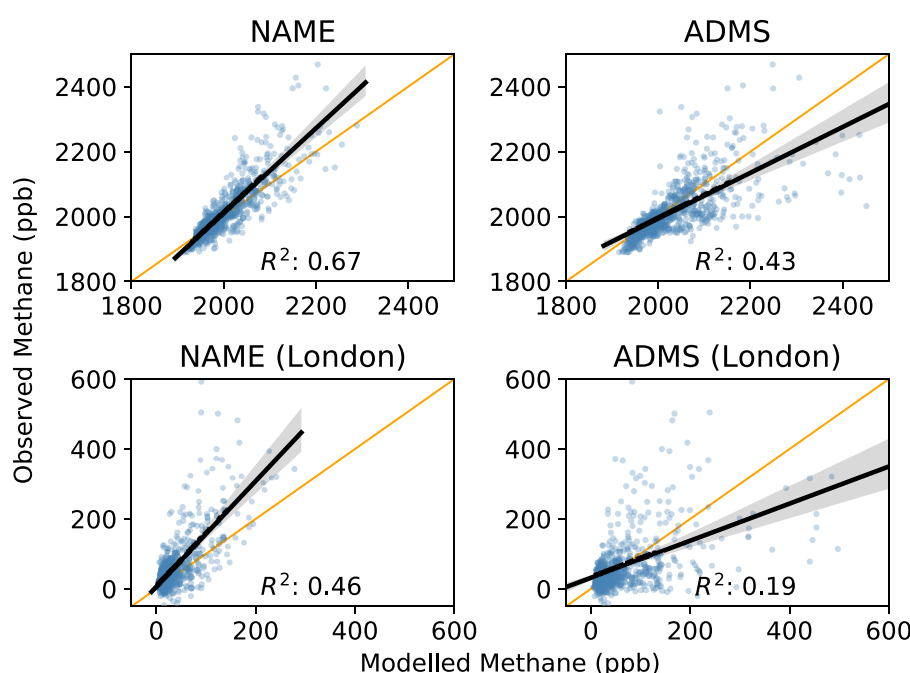


Figure 9. The top row shows the hourly mean mole fractions (contributions from London, regional sources and boundary conditions) for NAME (left) and ADMS (right). The bottom row shows the London contribution only, by subtracting the modelled non-London component from both the data and the model.

Table 1.

Linear fit statistics for model–observation comparison. The slope is derived with the model-predicted mole fraction as the independent variable (as in Figure 9), and the bias is the mean of the data minus the model (i.e. a positive bias, or a slope greater than one shows a model underestimate and vice versa).

Model configuration	Slope	R^2	Bias (ppb)	Standard deviation (ppb)
NAME	1.3	0.67	+12	59
ADMS embedded in NAME	0.71	0.43	–17	79
NAME (London)	1.5	0.46	+28	72
ADMS embedded in NAME (London)	0.53	0.19	–2.9	92
NAME (well mixed)	1.2	0.77	+1.8	16
ADMS embedded in NAME (well mixed)	0.95	0.63	–6.6	20

monitoring network will involve the set-up of additional measurement sites across the city and the development of an urban-scale inverse modelling system that will use the transport models from this work to obtain top-down emissions estimates for London.

Provided that the network can be supported over the coming years, the results from these estimates will be supplied to policymakers to help determine whether London's emissions reduction targets have been successful. The London GHG system also has the potential to identify missing sources or spatial discrepancies in the NAEI and may be able to give some insight into the temporal variability in emissions not accounted for in the bottom-up inventories.

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Correspondence to: D. Hoare

daniel.hoare@bristol.ac.uk

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Weather in my life – Kathryn Chalk

Flood Forecasting Centre, Met Office



When did you first become interested in the weather?

Two notable people sparked my interest in the weather, the first of which is my Nan. For as long as I can remember, we have not had a conversation where one of us has not

mentioned the weather, and who can blame her; us Brits love nothing better than talking about it! My second influence is a slightly unique one. When I was 8 years old, I got the opportunity to sing on a well-known morning TV show with my school choir, and just

before we aired, I watched the production of a live weather forecast broadcast. From then on, I have been following my goal to work in meteorology, and ultimately, I have landed a job communicating the forecast through the media.



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