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Supplement of

Estimation of trace gas fluxes with objectively determined basis functions using reversible-jump Markov chain Monte Carlo

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S1 Accounting for the background mole fraction

Footprints from the Numerical Atmospheric Modelling dispersion Environment (NAME, Jones et al., 2007; Manning et al., 2011) were used to explain changes in the measured mole fractions due to changes in emissions from within the NAME inversion domain. However, in order to solve for the rest of the methane mole fraction that was not accounted for by the NAME footprints, we simulated the global transport of methane using the Eulerian Model for OZone And Related Tracers (MOZART, Emmons et al., 2010). The model was run for over 10 years prior to 2014, creating a realistic three dimensional distribution of atmospheric methane, using global anthropogenic emissions from the Emissions Database for Global Atmospheric Research (EDGAR, EC-JRC/PBL, 2011), wetland emission sources from Bloom et al. (2012), biomass burning emissions from GFED3 (van der Werf et al., 2010) and other sources and sinks from Fung et al. (1991). Loss due to reaction with OH was based on monthly mean climatologies from Spivakovsky et al. (2000). Combined stratospheric loss due to O^1D and Cl was based on monthly mean climatology output from the Cambridge 2D model (Velders, 1995).

The simulated monthly average mole fractions were used to create "curtains" around the edges of the regional inversion domain, shown by the schematic diagram in Fig. S1. These curtains varied with height and latitude on the east and west boundaries, and height and longitude along the north and south edges. The MOZART mole fractions along the edges were interpolated from their coarser $2.5° \times 1.875°$ output resolution to the native NAME resolution of $0.352° \times 0.234°$, and the curtains contained 20 vertical levels. These curtains were multiplied by the normalised information on what height and latitude (for E and W edges) or longitude (S and N edges) particles left the NAME domain, to create appropriate baseline sensitivity terms. For each baseline sensitivity term there was a baseline parameter variable, to be solved for in the inversion.

Four baseline terms were included in the parameters vector, representing baseline contributions from the North, South, East and West domain edges. In effect this meant scaling the entire mole fraction curtain of each edge up or down. These terms were not included in the transdimensional scheme but they were still solved for in a conventional Metropolis Hastings MCMC sense.

In addition, the large NAME regional domain size made repeated partitioning of the domain into basis functions less computationally efficient than desired. Given that the magnitude of the sensitivity information would be expected to decay as an inverse square law, the data would be unable to provide sufficient information on much of the computational domain. As such, the domain on which the transdimensional inversion was performed was chosen to be a smaller sub-grid within this larger computational domain. Outside of the sub-domain the sensitivity terms were aggregated into six basis functions, which were treated as fixed in space, and therefore not included in the transdimensional scheme.

The six fixed regions contributed on average less than 10% of the total modelled mole fractions explained by the NAME computational domain (not including the baseline mole fractions). Although small, this highlights the importance of including such far-field influences when estimating regional emissions. Similarly the variations in the modelled baseline were far from insignificant, and highlighted that slight differences in the long-range transport of the particles can have a major impact on the portion of the mole fraction not explained by emissions from within the NAME computational domain. The mole fraction time series in Fig. S2, for each of the stations show how, by accounting for

the location of the particles on entering the domain, troughs in the data may be replicated by the model, and the baseline may be considered far from a static mole fraction. Figure S2 shows the data used in Sect. 5 of the main manuscript.
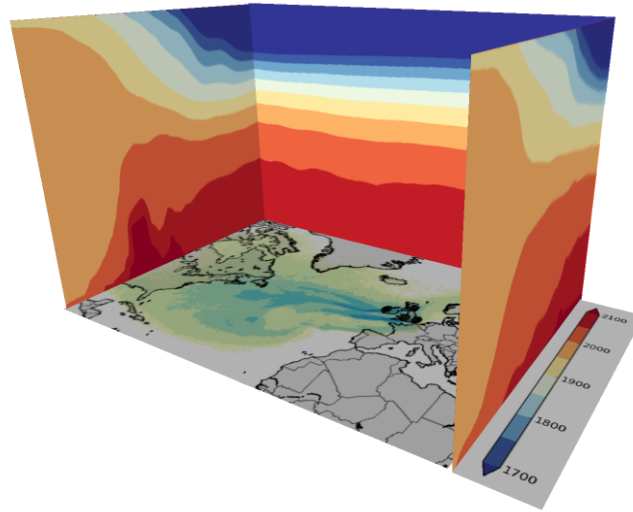

Figure S1: A schematic diagram of the NAME inversion domain, surrounded by MOZART generated mole fraction curtains at the edges of the domain.
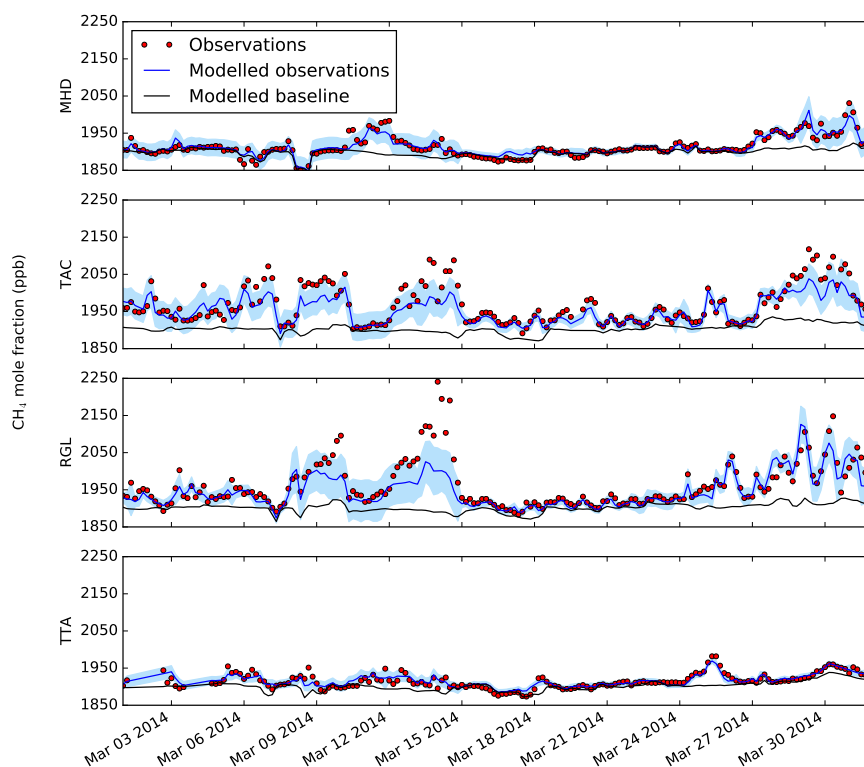
Figure S2: Time series comparison plots of the observed (red dots) and modelled mole fractions (blue line) at each of the measurement sites. From top to bottom: MHD (Mace Head, Irealand), TAC (Tacolneston, England), RGL (Ridgehill, England) and TTA (Angus, Scotland). The blue shading shows the estimated model uncertainty, which varied each 7 day period. The posterior baseline estimation is shown by the black line.

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