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Atmospheric Chemistry and Physics Discussions

Supplement of

Quantifying methane and nitrous oxide emissions from the UK using a dense monitoring network

A. L. Ganesan et al.

Correspondence to: A. L. Ganesan (anita.ganesan@bristol.ac.uk)

Supplemental material to 'Quantifying the UK's methane and nitrous oxide emissions using a dense monitoring network'

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1 Boundary condition estimation

The total value of the baseline level for each station was calculated as the sum of the fractional portions coming from each boundary (over the 30 day air history) for each two-hour simulation of the study. Figure 1 is a schematic of the ten boundaries that were simulated. Over the horizontal inversion domain, boundary conditions to two vertical boxes were estimated. The first box extended from 0 to 3 km altitude. In this box, eight boundary conditions to each side of the four quadrants of the horizontal domain (Northwest, Northeast, Southeast and Southwest) were estimated, corresponding to air entering the domain from different directions in the lower troposphere. The second box was for air entering from any direction between 3-9 km, which corresponds to the middle and upper troposphere, as well as air from above. Lastly, the boundary condition on the 9 km boundary was estimated, corresponding to upper tropospheric/stratospheric air descending into the domain. The fraction of particles entering the domain through each boundary was tracked using the air histories. For each of the eight horizontal boundaries, the total footprint in the edge boxes along the boundary was summed. This method approximated the number of particles exiting the boundary as it was assumed that once particles reached the edge box, they immediately left the domain (at which point they were removed from the system). This 'edge box' method was similarly used for the horizontal boundaries of the 3-9 km box, but instead of estimating eight independent mid-troposphere boundaries, these fractions were summed into one value for the entire box. This approximation was made for simplicity and reduced computational expense. For the top boundary at 9 km, the total footprint was summed between 9 and 19 km over the entire domain. Because particles would only reach this altitude late in the simulation, this was used as an approximation to the number of particles that traveled above 9 km.

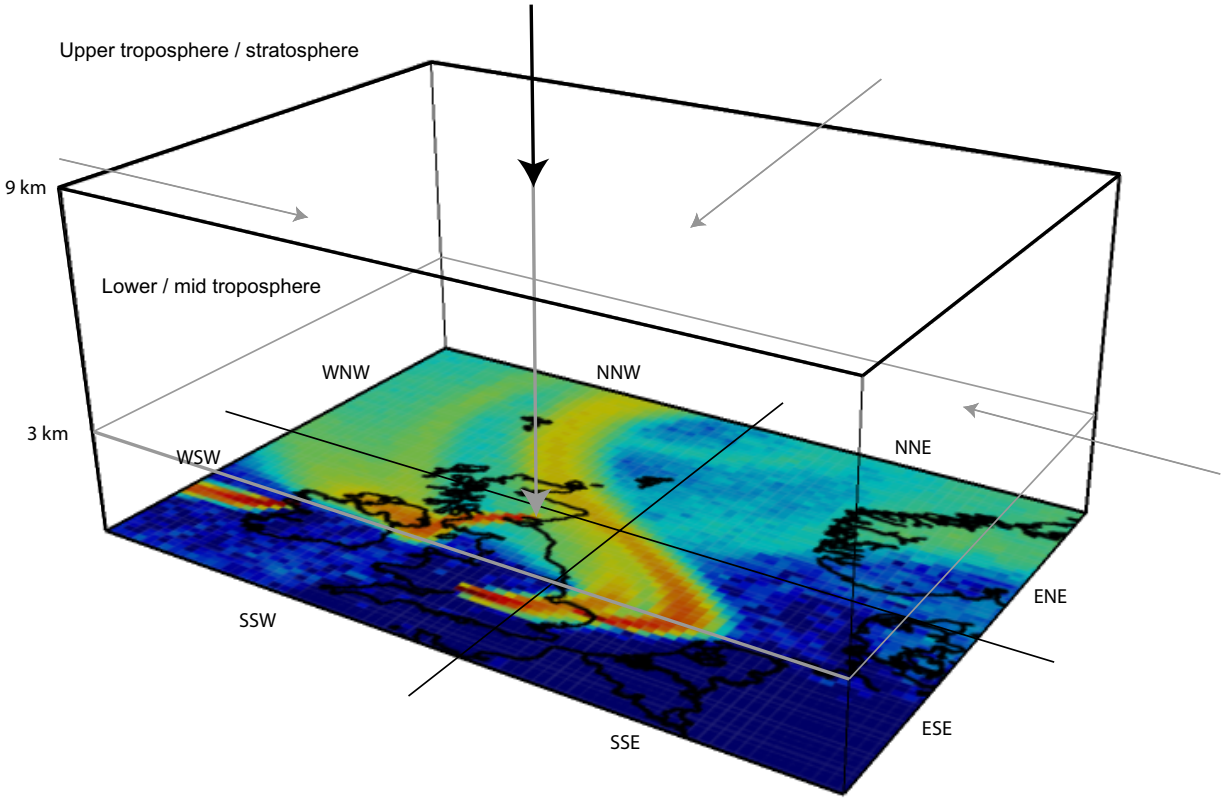

Figure 1: Schematic of boundary conditions to the inversion domain. PDF parameters to seventeen BCs were estimated in total; eight defining the polynomial that governs the WSW boundary and offsets for seven other horizontal boundaries and two upper atmosphere boundaries (arrows in grey correspond to the BC governing air entering the 3-9 km box and the arrow in black corresponds to the BC governing air from above 9 km). The map shows combined air histories for all four sites for a given 2-hour period and illustrates that the stations can sample different 'baselines' at the same time due to differences in their meteorology.

2 Simulated mole fractions and observations

Figures 2 and 3 show the simulated mole fractions in comparison with observations at each site, with the net effect of the derived baseline shown alongside. The net baseline was computed by summing the fractional portion coming from each boundary at each particular time.

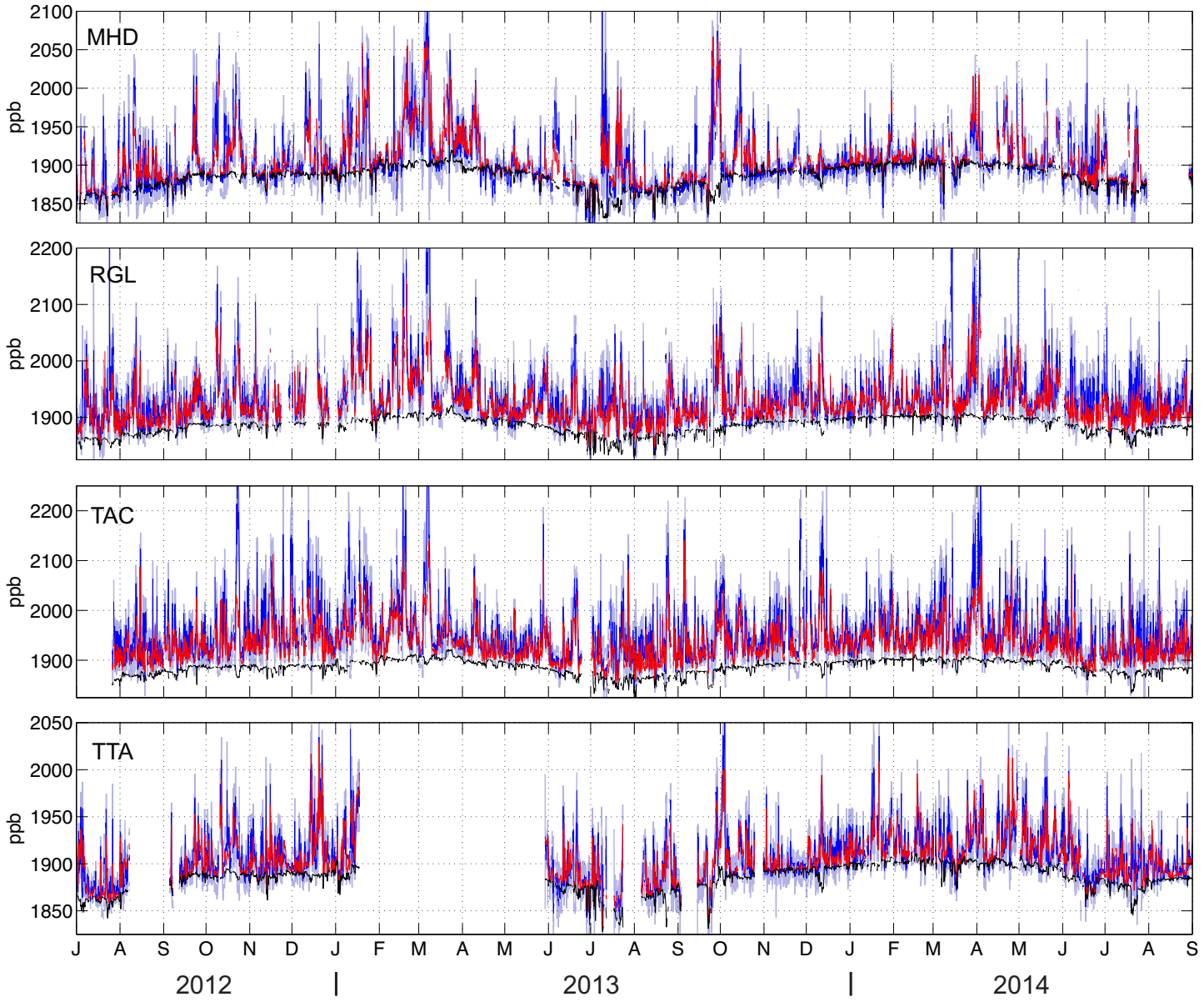

Figure 2: Simulated CH_4 mole fractions (red), observations (blue) and derived baseline (black) for each site. Shading indicates the posterior 5th to 95th percentile model errors as well as instrumental uncertainties.

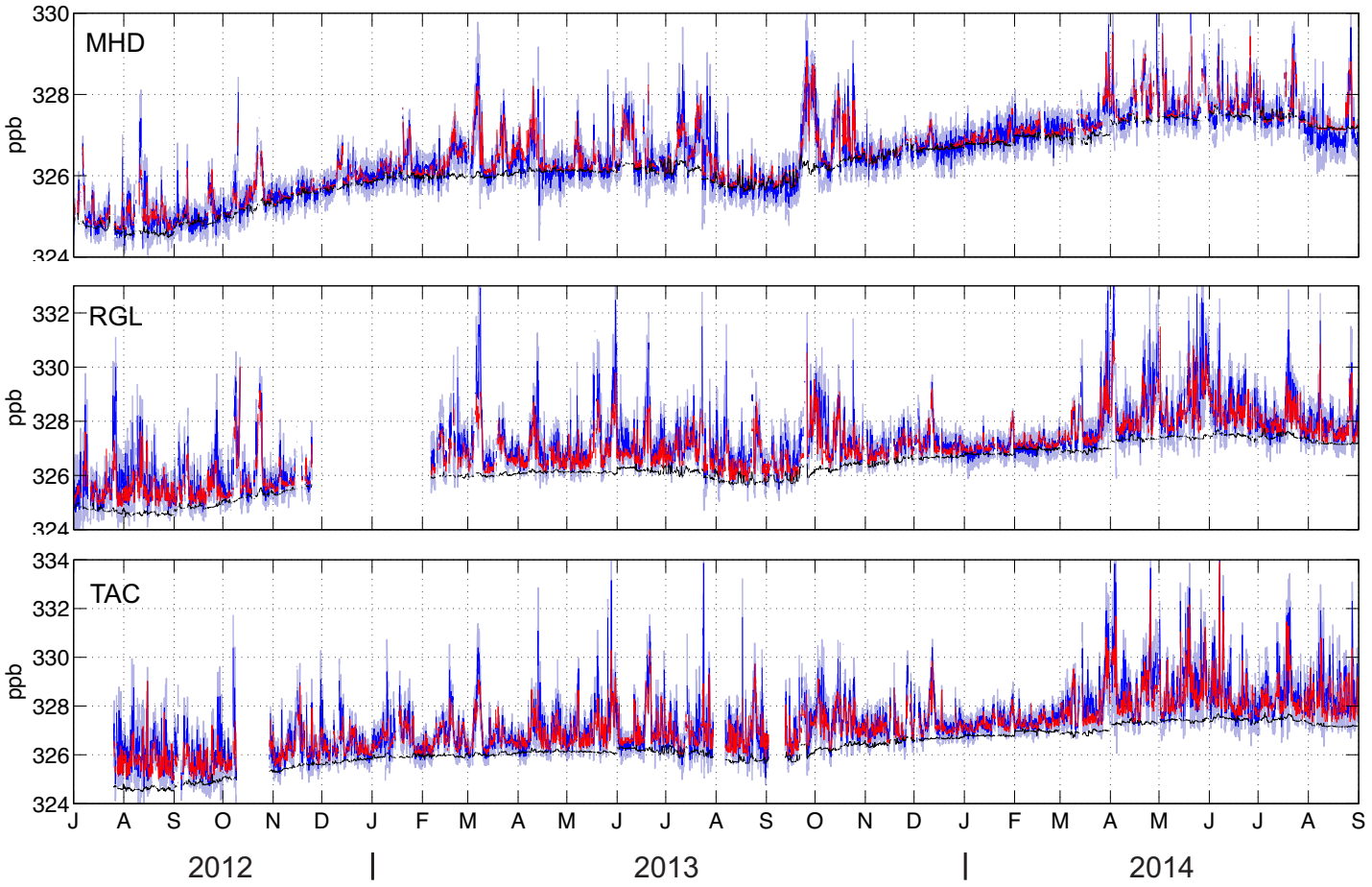

Figure 3: Simulated N_2O mole fractions (red), observations (blue) and derived baseline (black) for each site. Shading indicates the posterior 5th to 95th percentile model errors as well as instrumental uncertainties.

3 CH_4 sensitivity study (excluding Angus)

The CH_4 study was also performed excluding measurements from Angus in order to provide a direct comparison with the N_2O study. Figure 4 shows derived emissions and spatial distributions for this case study. The majority of difference between this sensitivity study and the study including Angus is that there is less adjustment made to Scotland's prior emissions. As a result, total UK emissions are higher (closer to the prior) and uncertainties in Scotland are significantly higher as well. UK total emissions uncertainties are thus larger, highlighting the benefits of including this extra station. The correlation scales derived in this study were 0.98 (0.68-1.49) days and 113 (14-352) km, which are very similar to the correlation scales derived in the main study. These results highlight that Angus measurements serve to primarily constrain Scotland's emissions. The similar correlation scales between the two studies suggest that Angus, being over 600 km away from the other three sites, does not play a significant role in constraining the spatial correlation scale and that the differences in scales derived for CH_4 and N_2O are not simply due to differences in the network.

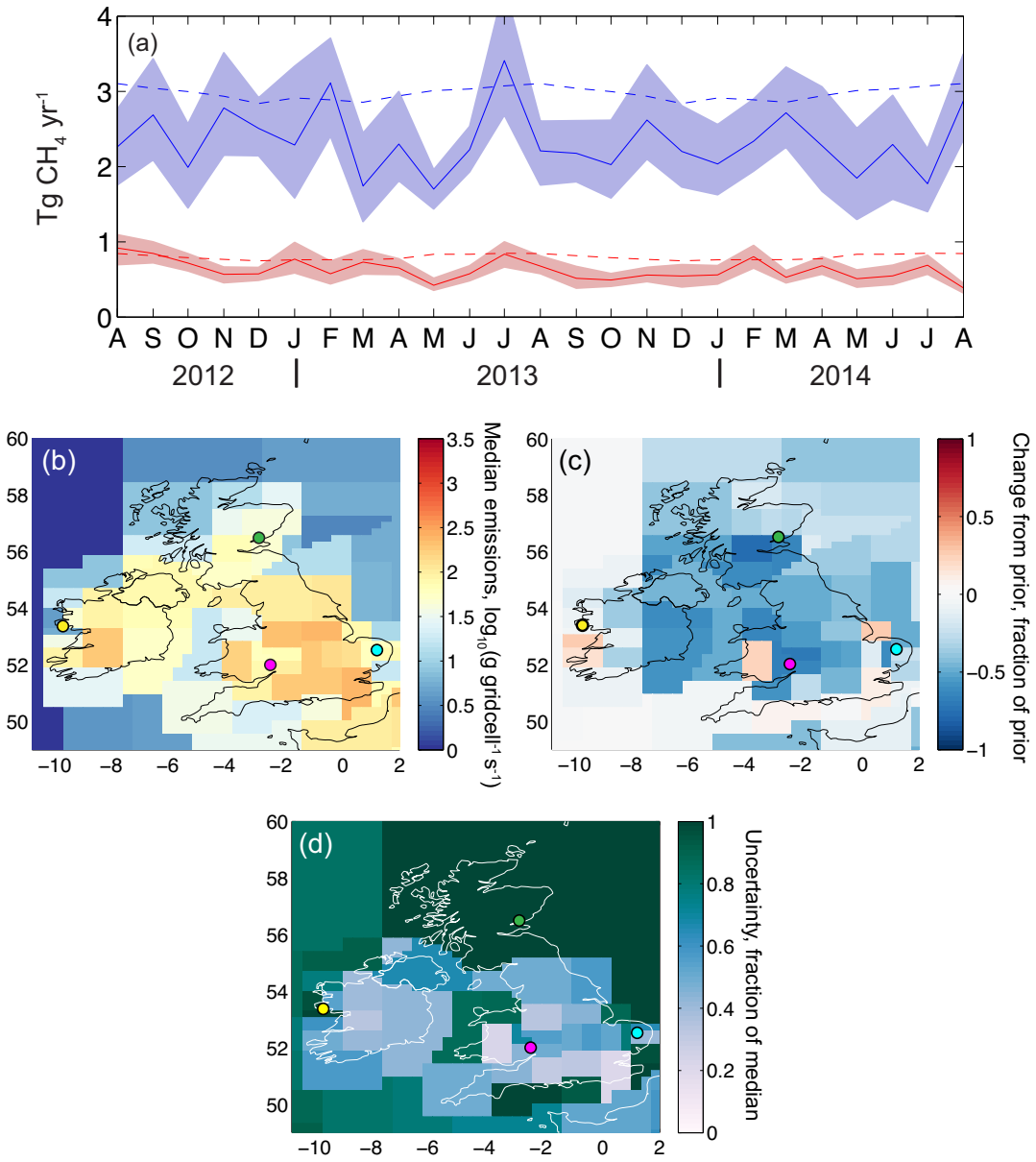

Figure 4: Results derived for the CH_4 sensitivity study in which Angus measurements were excluded. (a) Total national CH_4 emissions for the UK (blue, solid) and Ireland (red, solid) in Tg yr^{-1}. Prior emissions for each country are shown in the dashed lines. Shading corresponds to the 5th to 95th percentile range. (b) Median posterior CH_4 emissions shown on a logarithmic scale. (c) Fractional difference of the median posterior emissions from the prior. (d) Fractional posterior emissions uncertainty, which corresponds to the difference between the 5th and 95th percentiles, relative to the median. Colored circles show the measurement stations (MHD, yellow; RGL, magenta; TAC, cyan; TTA, green, now excluded).