



Supplement of

Quantifying methane and nitrous oxide emissions from the UK and Ireland using a national-scale 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. 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.

2 Simulated mole fractions and observations

Figures 1 through 16 show the simulated posterior and prior mole fractions by year in comparison with observations at each site, with the net effect of the baseline shown alongside. The net baseline was computed by summing the fractional portion coming from each boundary at each particular time.

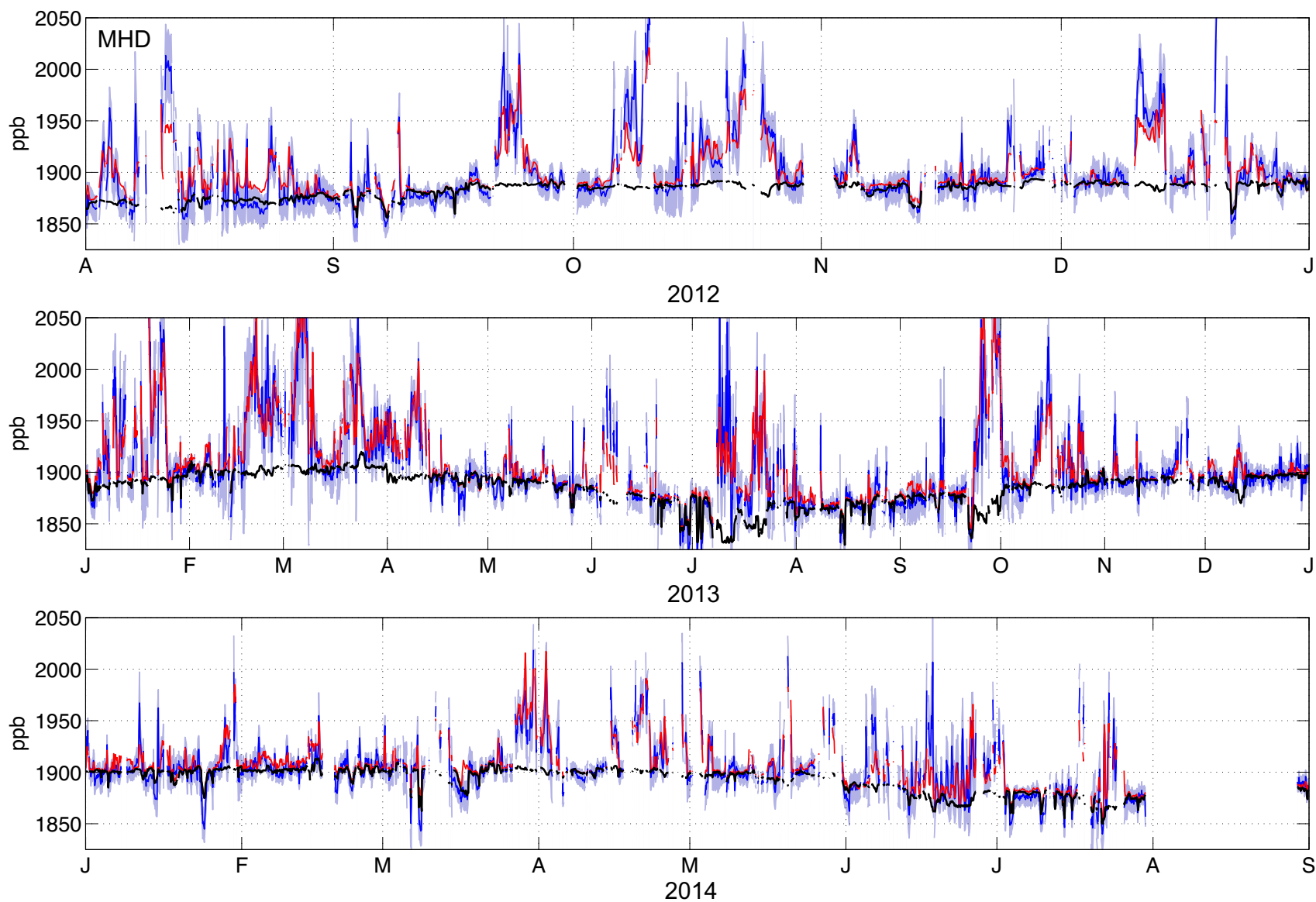


Figure 1: Simulated posterior CH_4 mole fractions from MHD by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

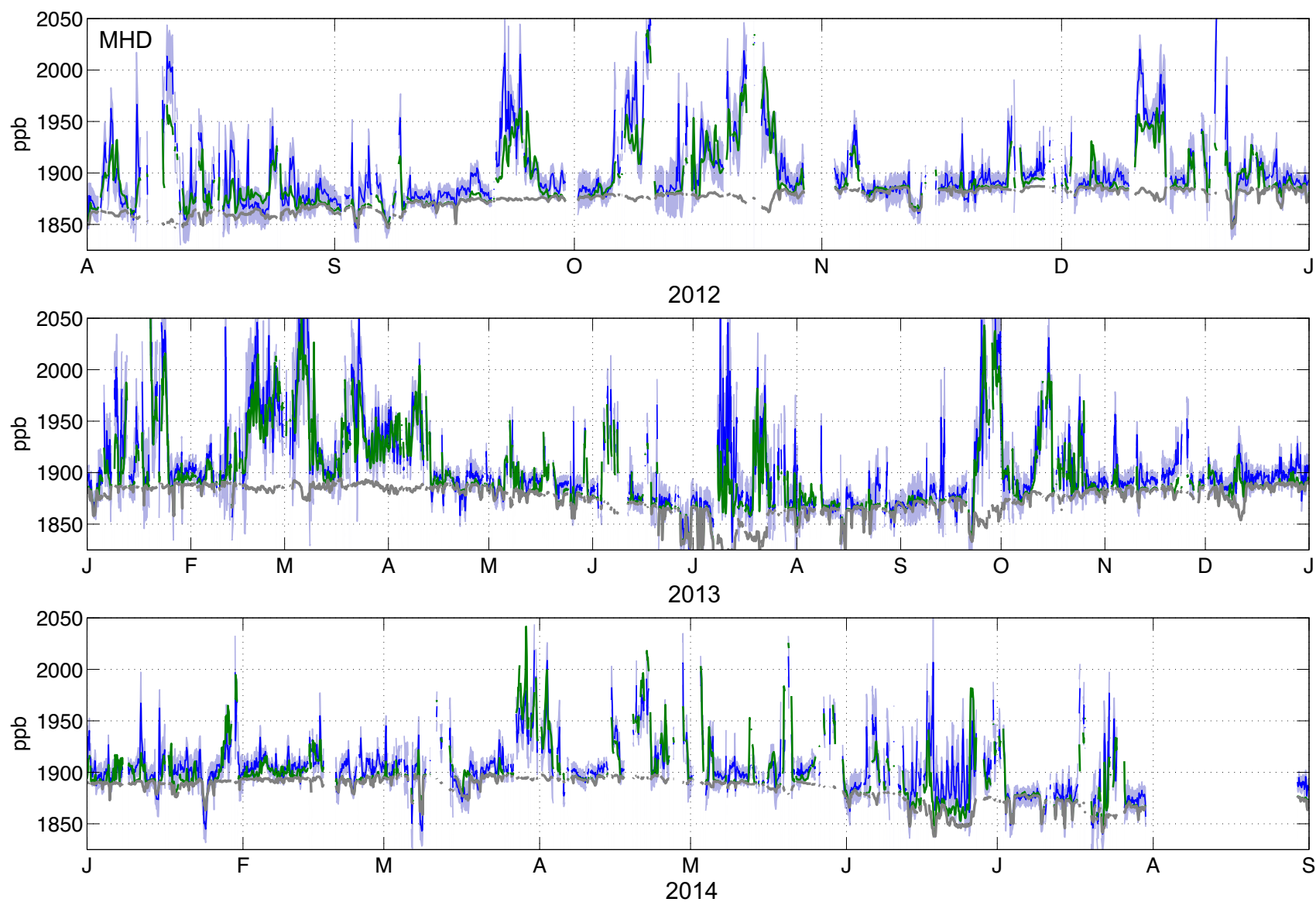


Figure 2: Simulated prior CH₄ mole fractions from MHD by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

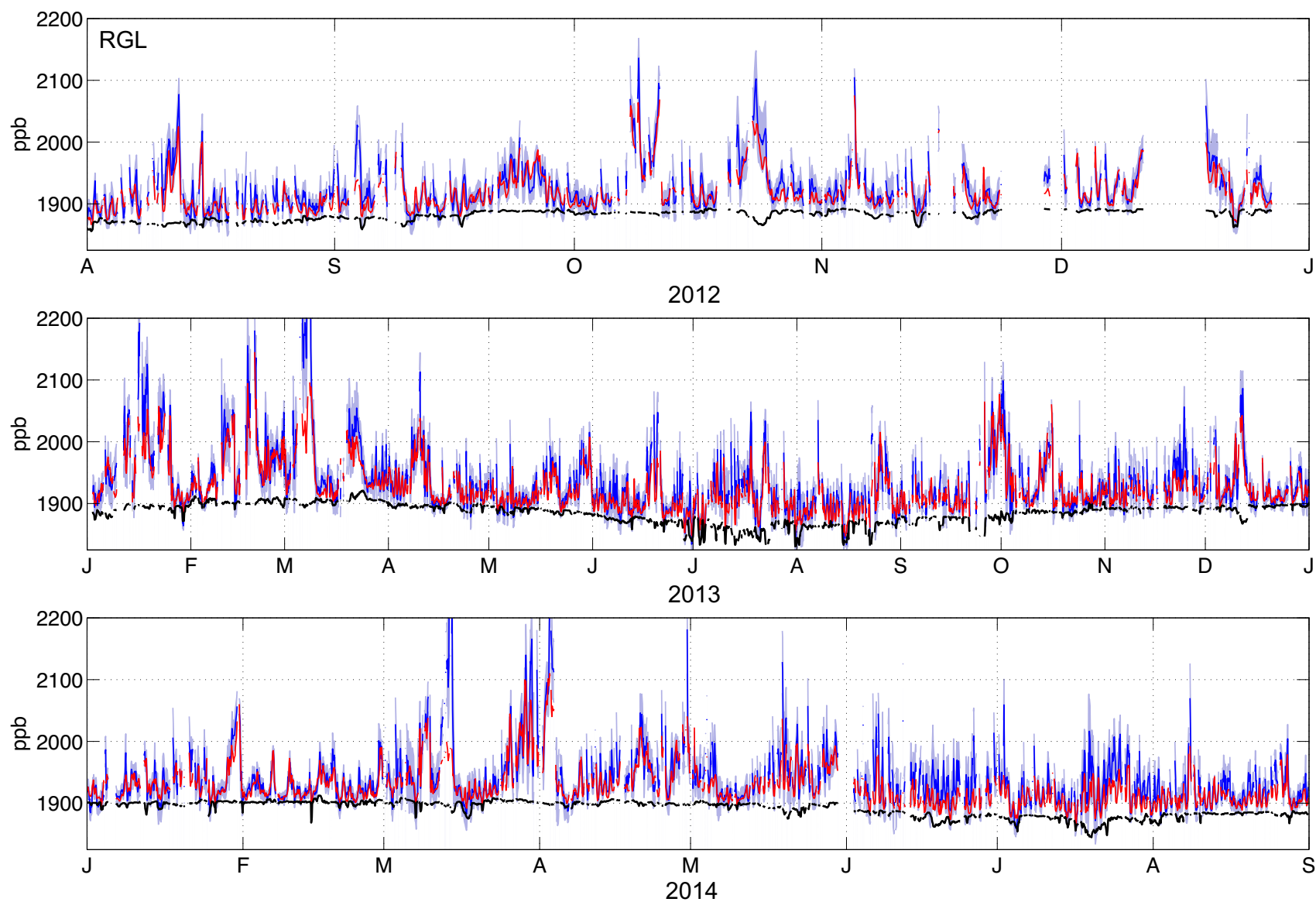


Figure 3: Simulated posterior CH_4 mole fractions from RGL by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

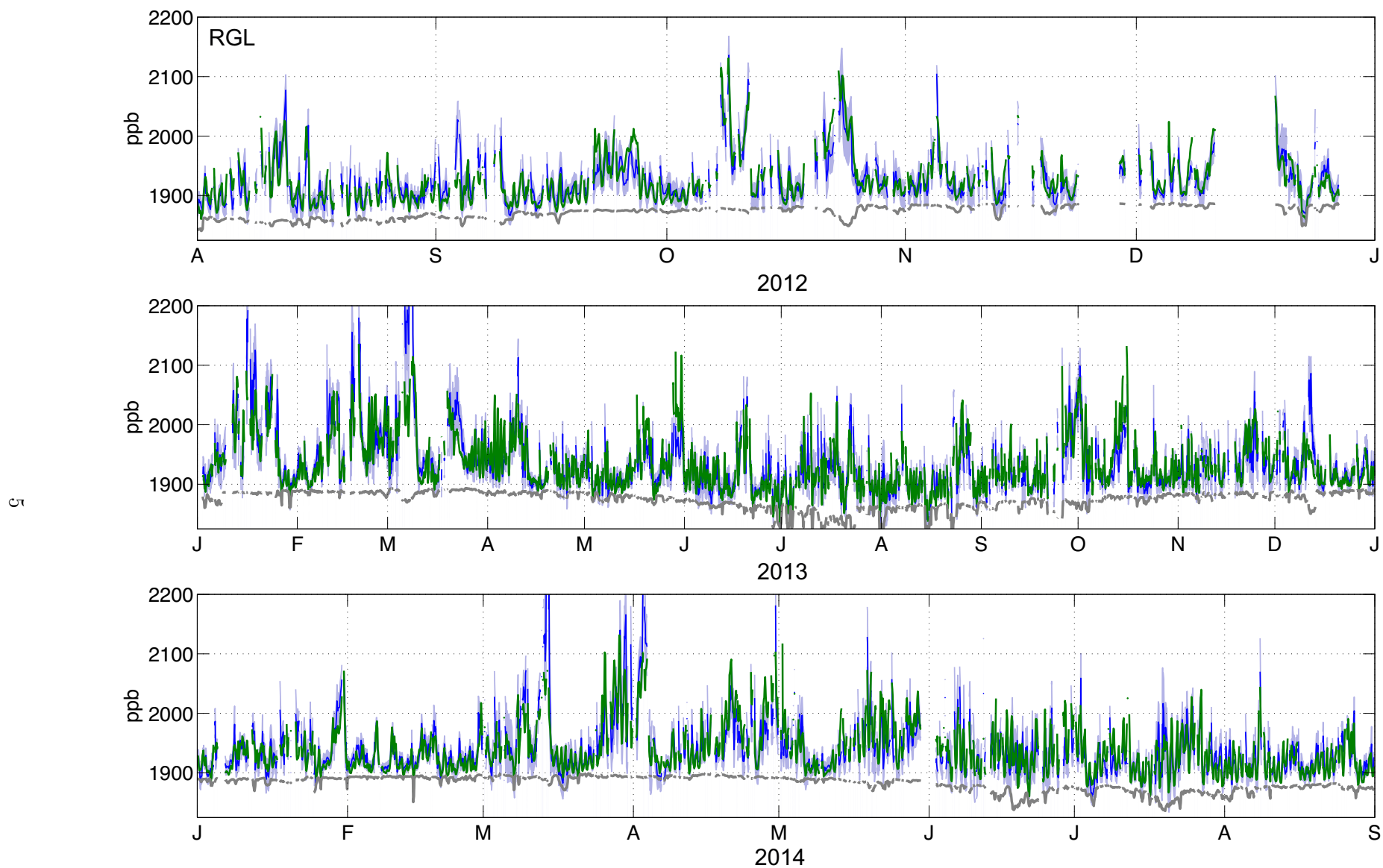


Figure 4: Simulated prior CH₄ mole fractions from RGL by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

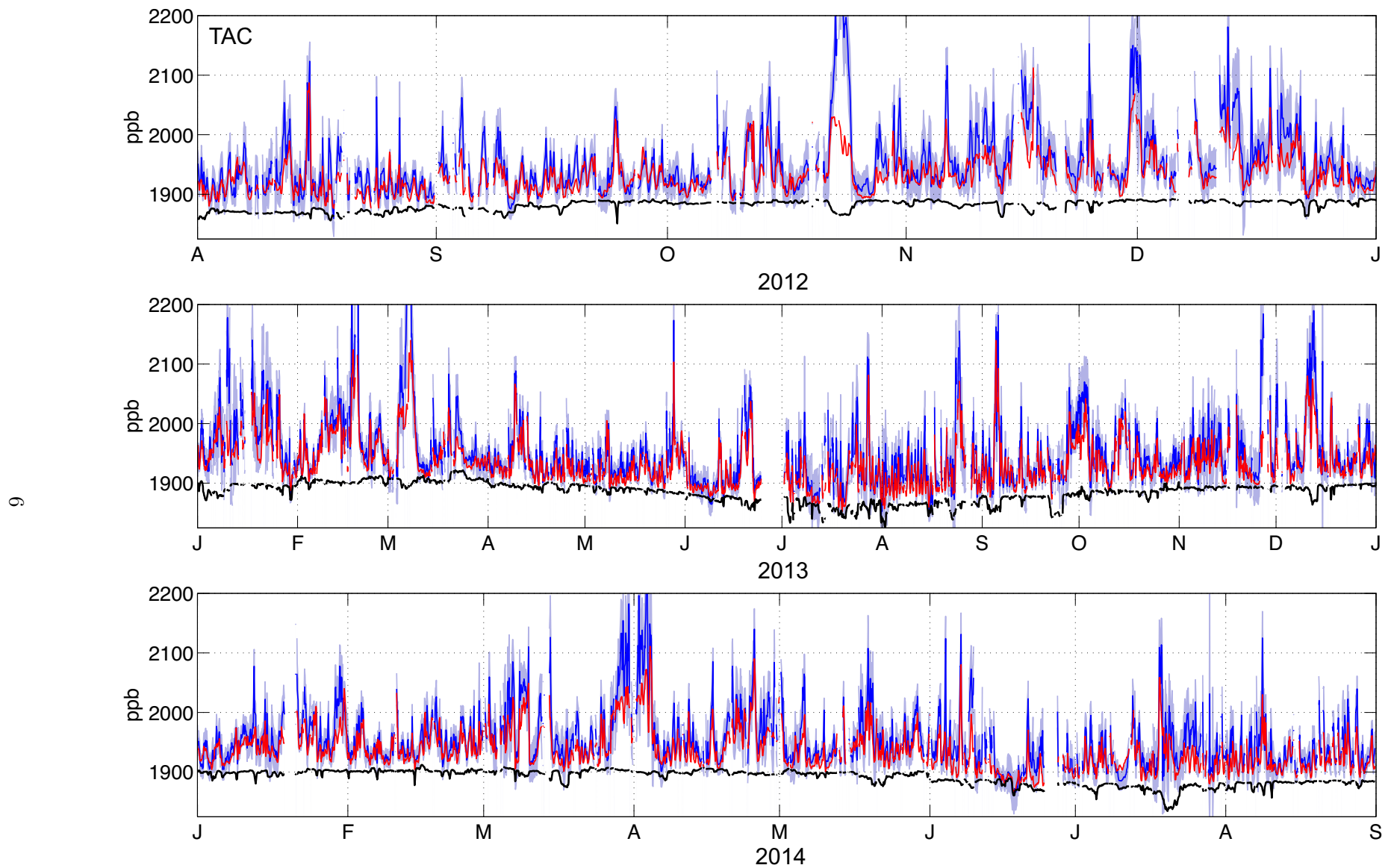


Figure 5: Simulated posterior CH_4 mole fractions from TAC by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

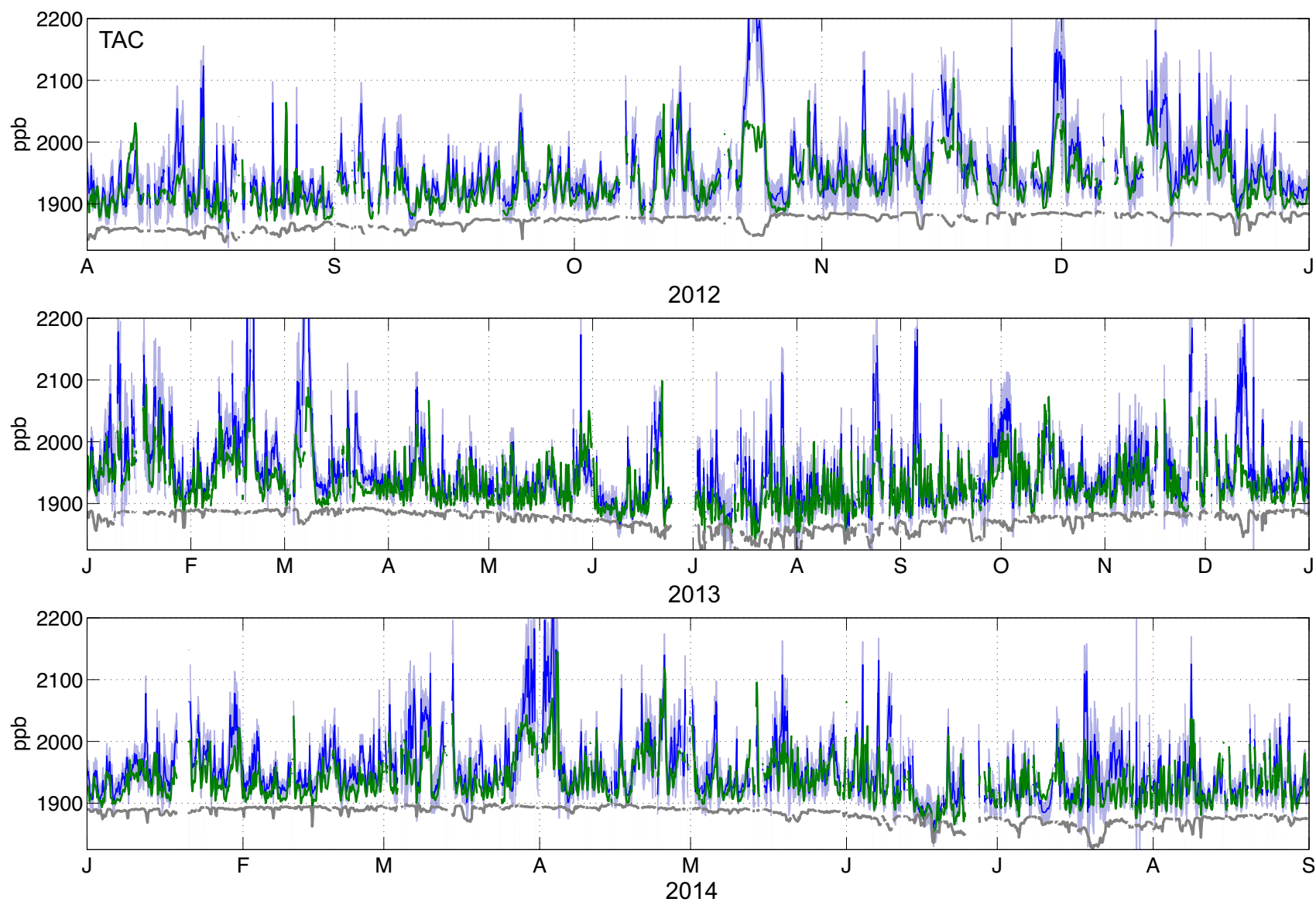


Figure 6: Simulated prior CH₄ mole fractions from TAC by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

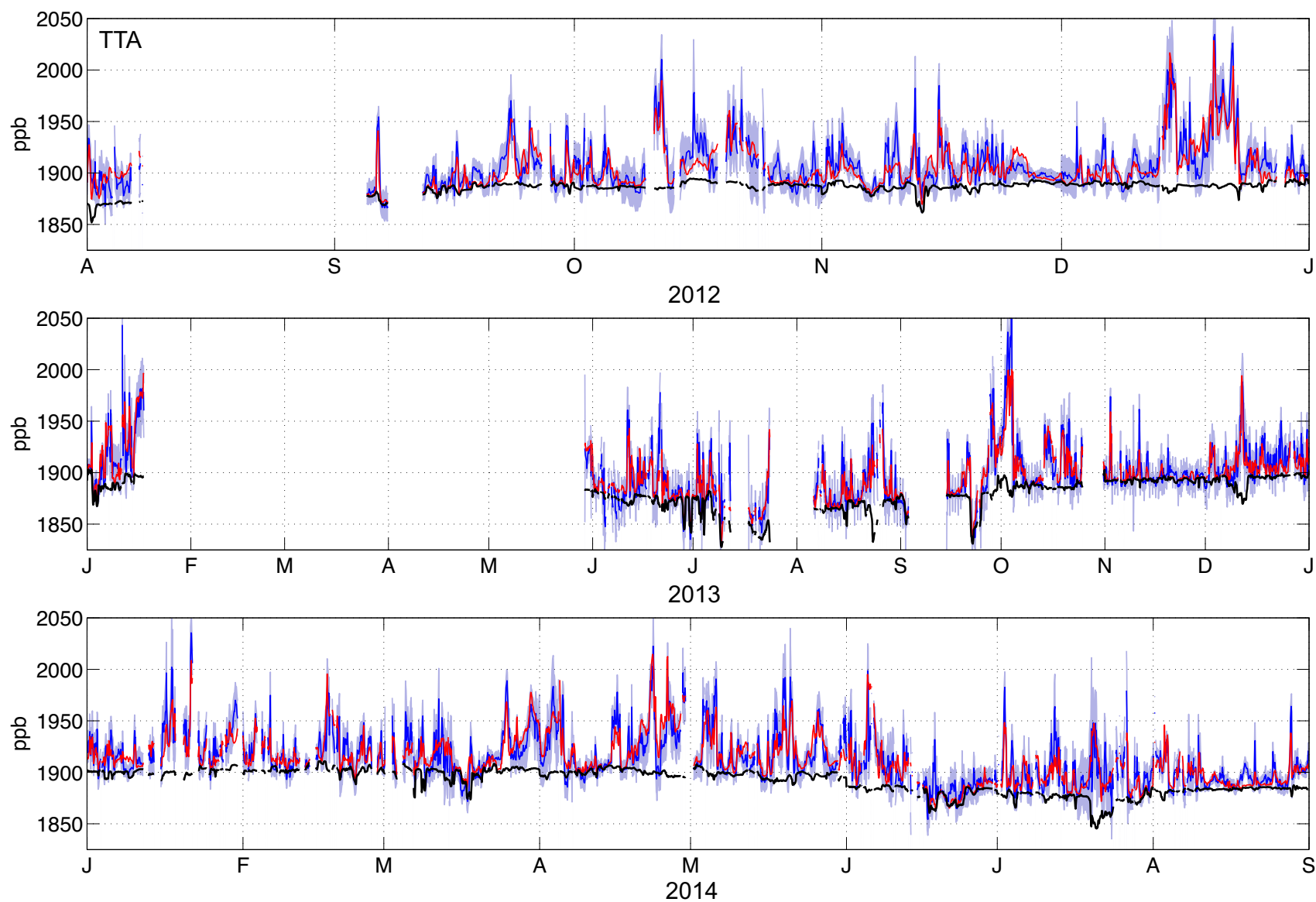


Figure 7: Simulated posterior CH_4 mole fractions from TTA by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

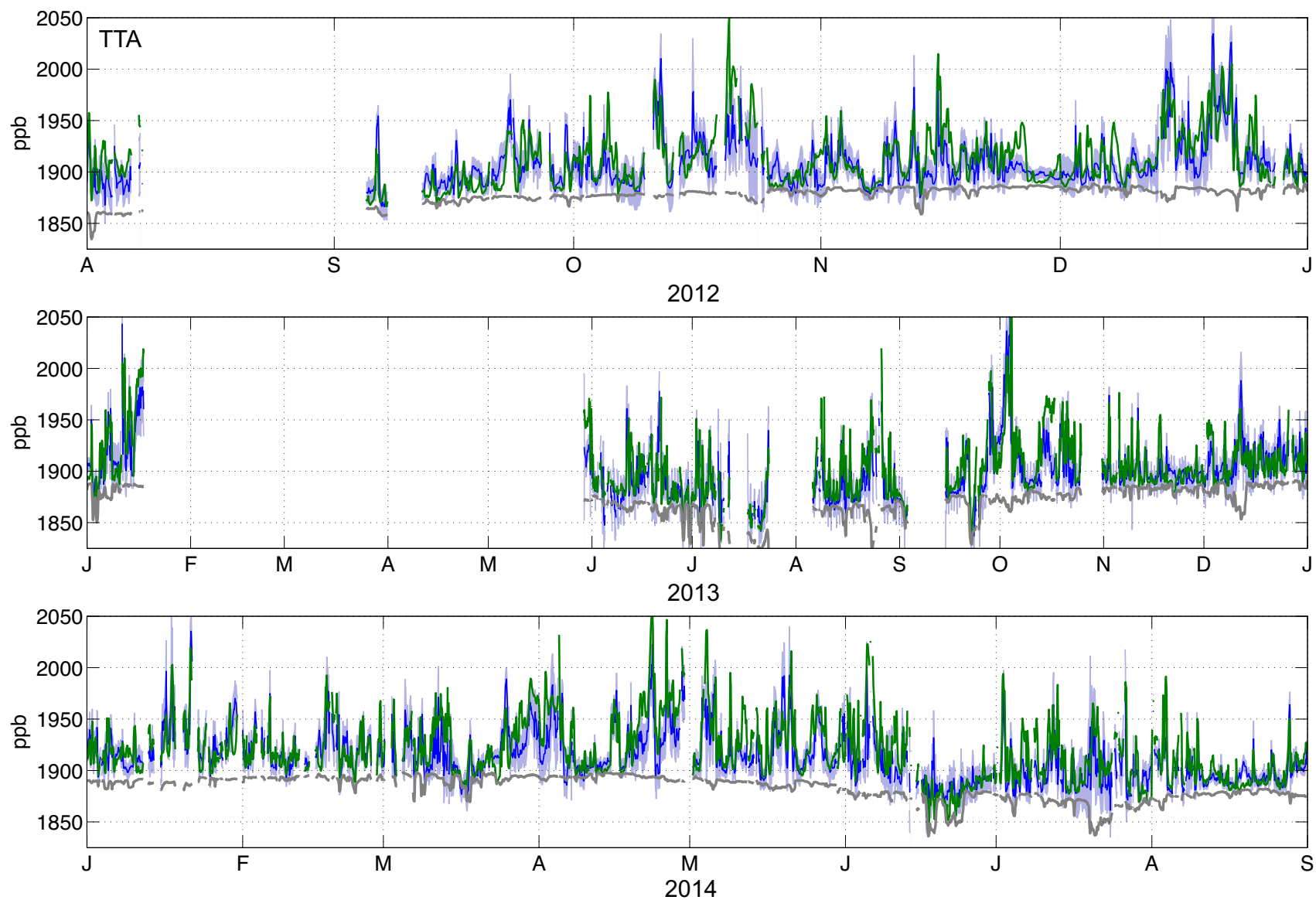


Figure 8: Simulated prior CH₄ mole fractions from TTA by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

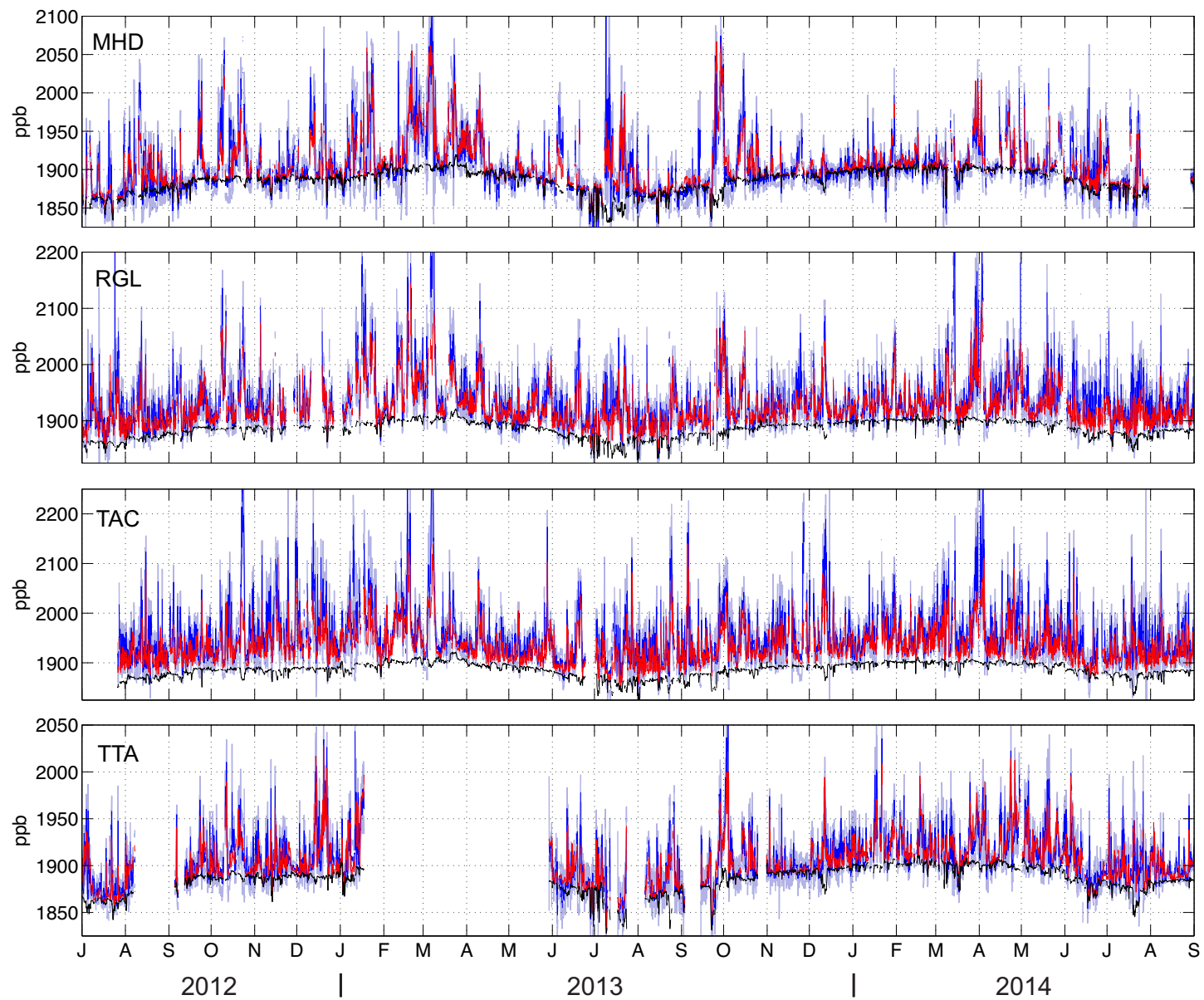


Figure 9: Simulated posterior CH₄ mole fractions for all sites and all years (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

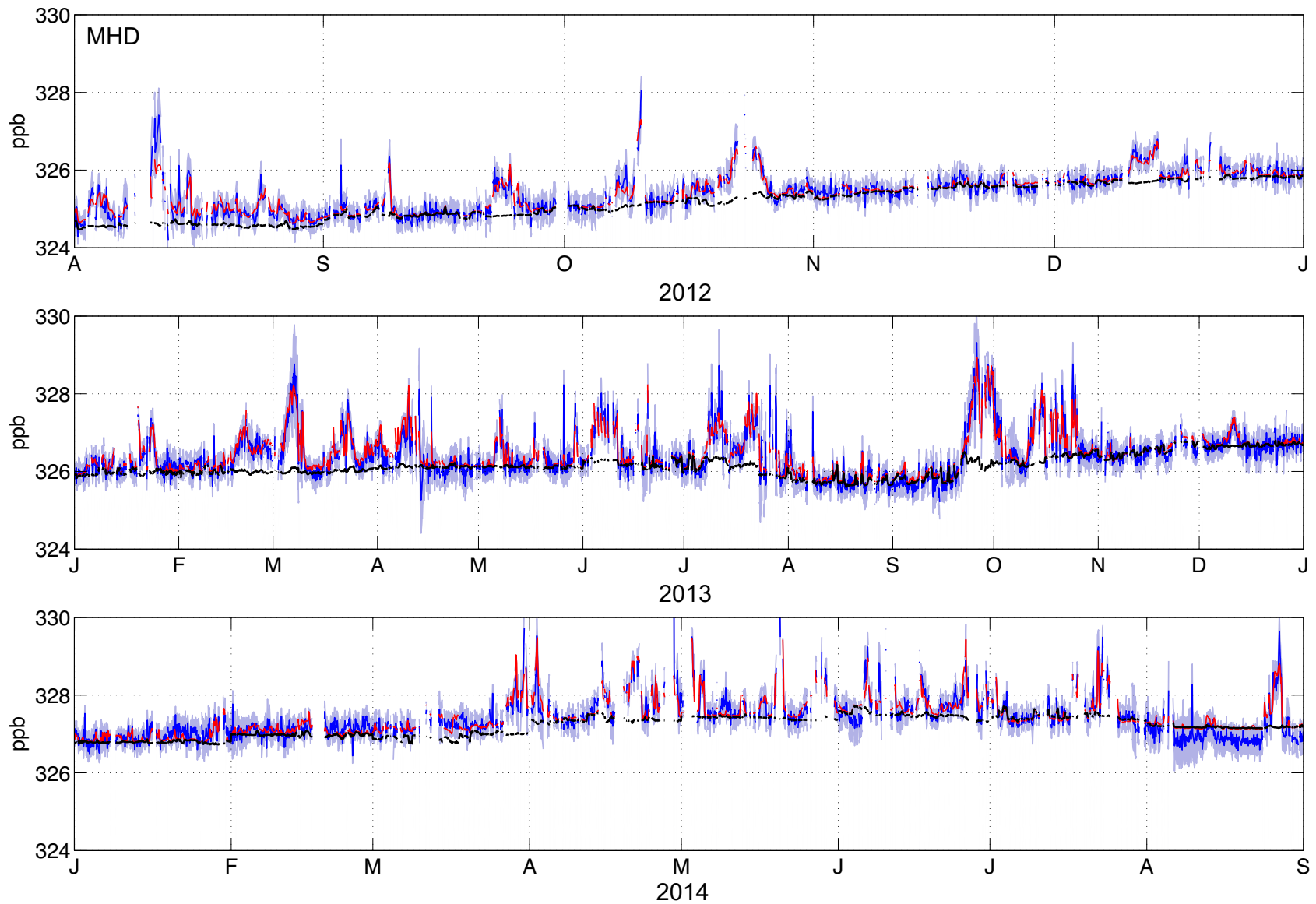


Figure 10: Simulated posterior N₂O mole fractions from MHD by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

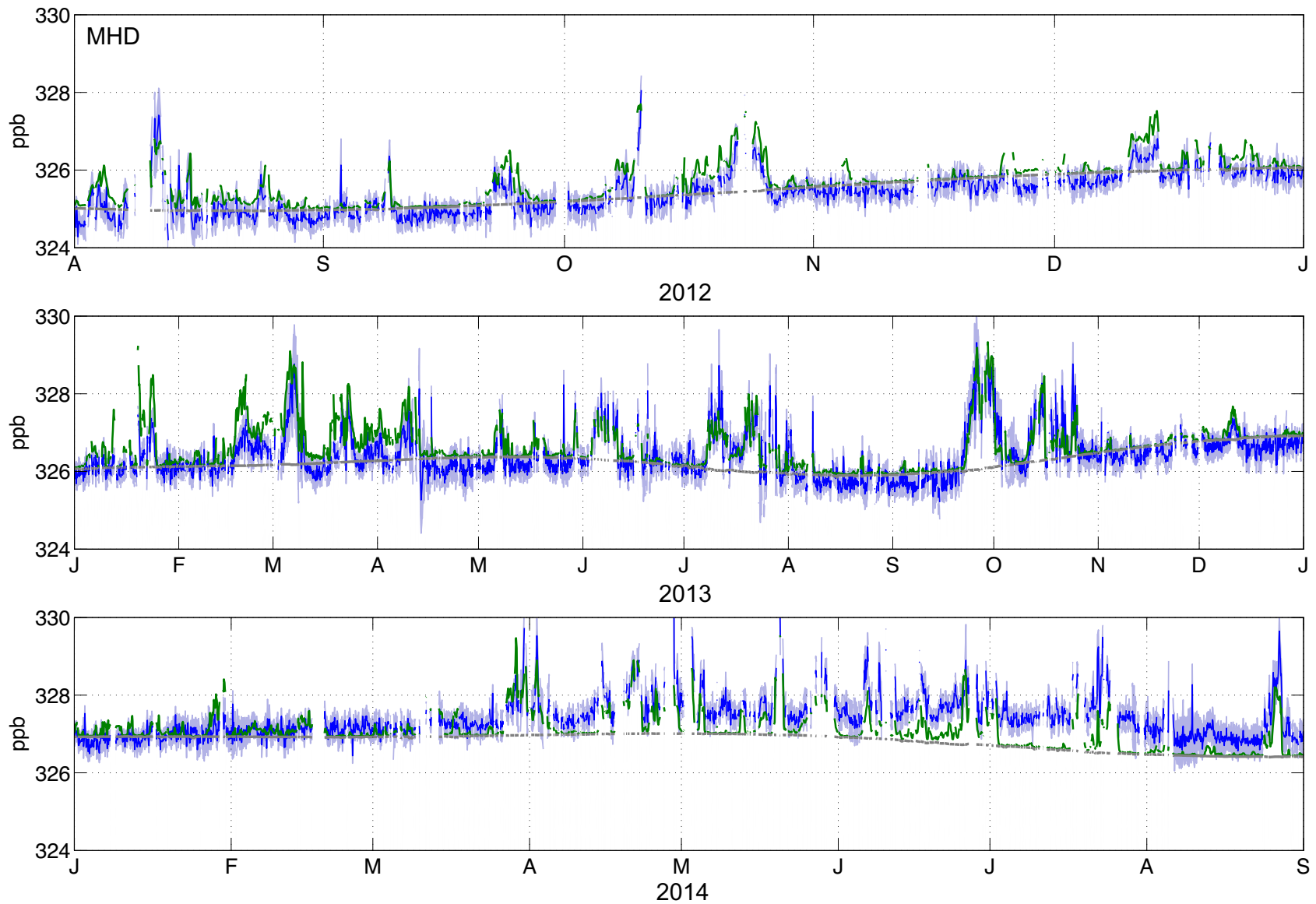


Figure 11: Simulated prior N₂O mole fractions from MHD by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

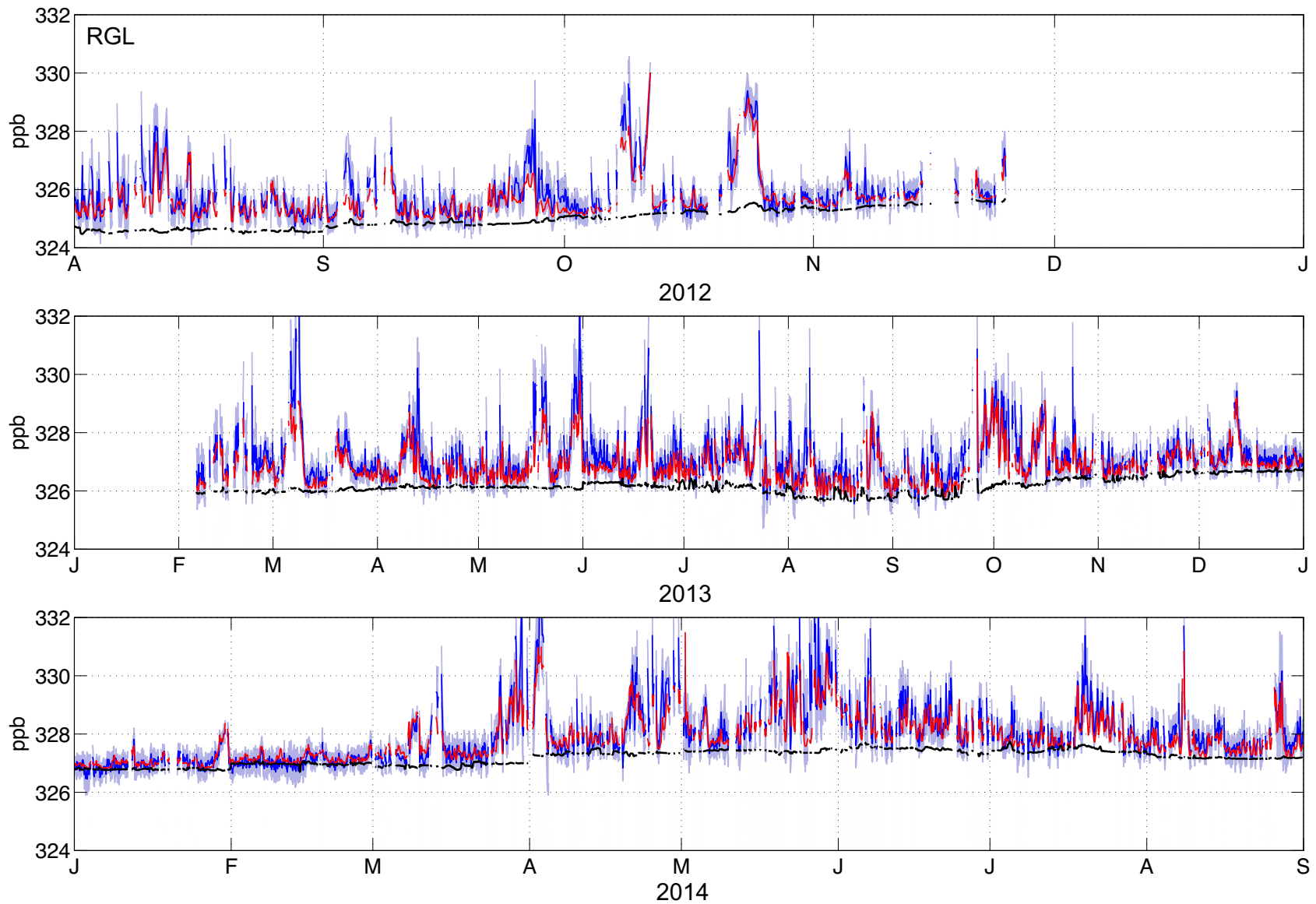


Figure 12: Simulated posterior N₂O mole fractions from RGL by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

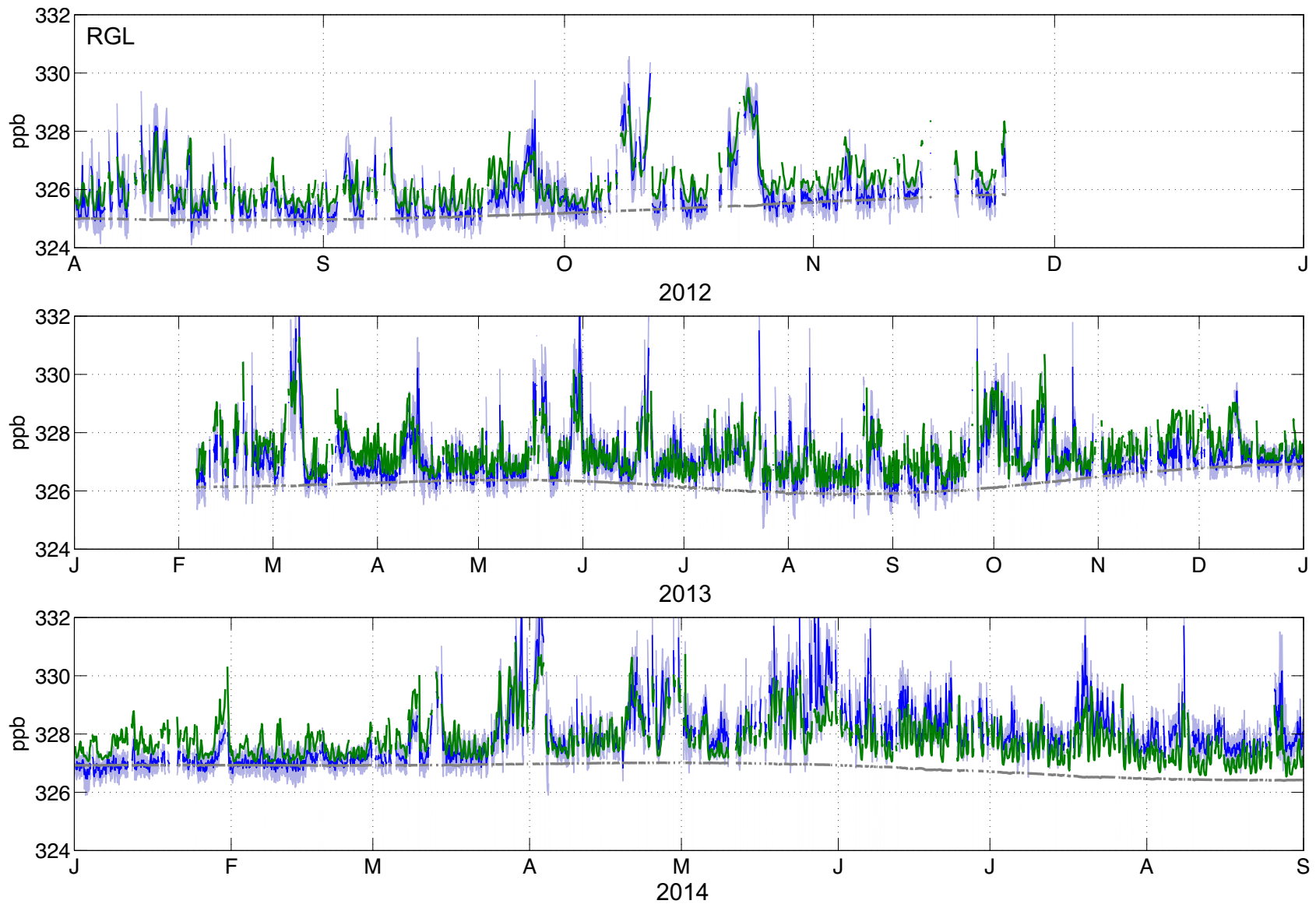


Figure 13: Simulated prior N₂O mole fractions from RGL by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

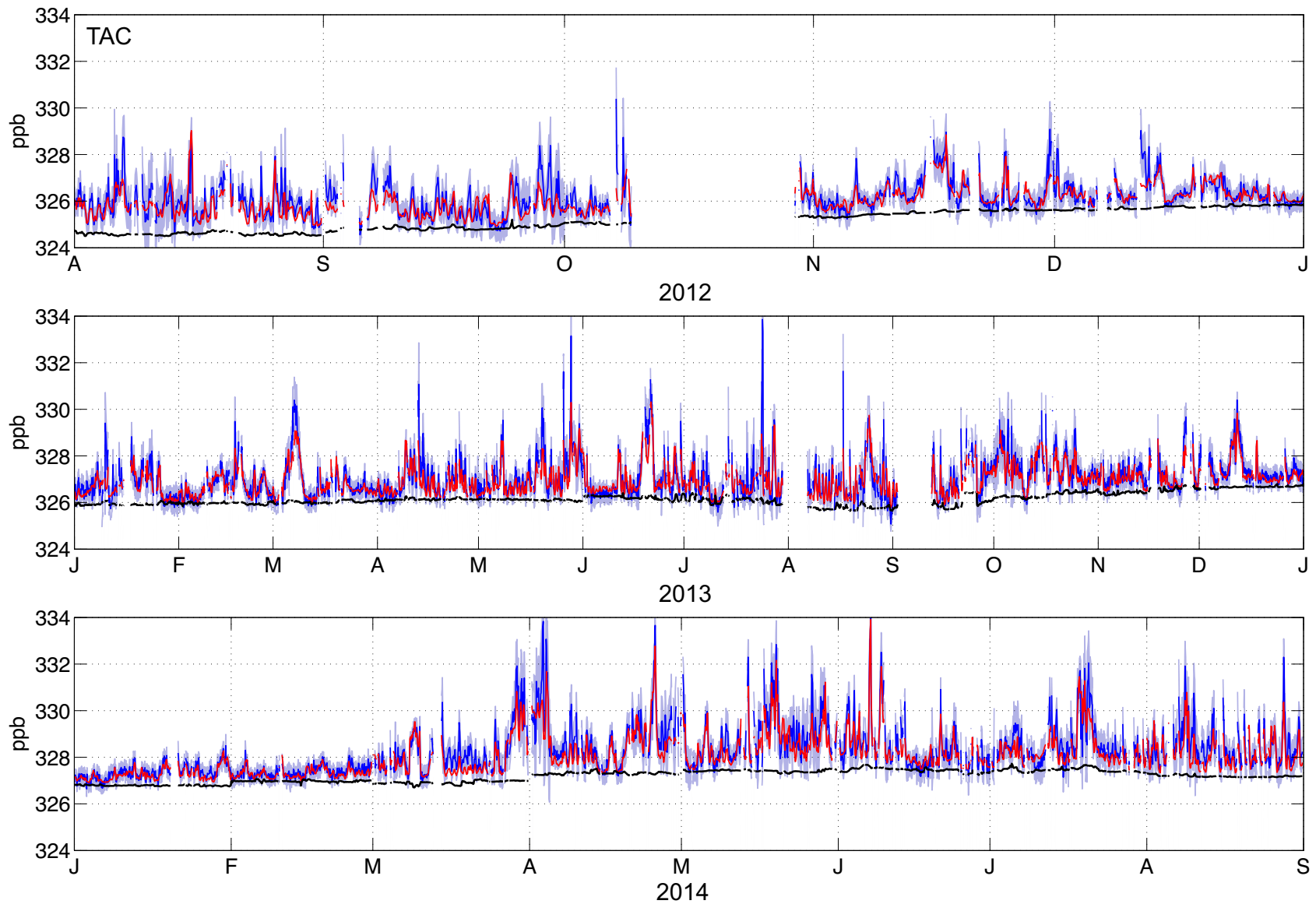


Figure 14: Simulated posterior N₂O mole fractions from TAC by year (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

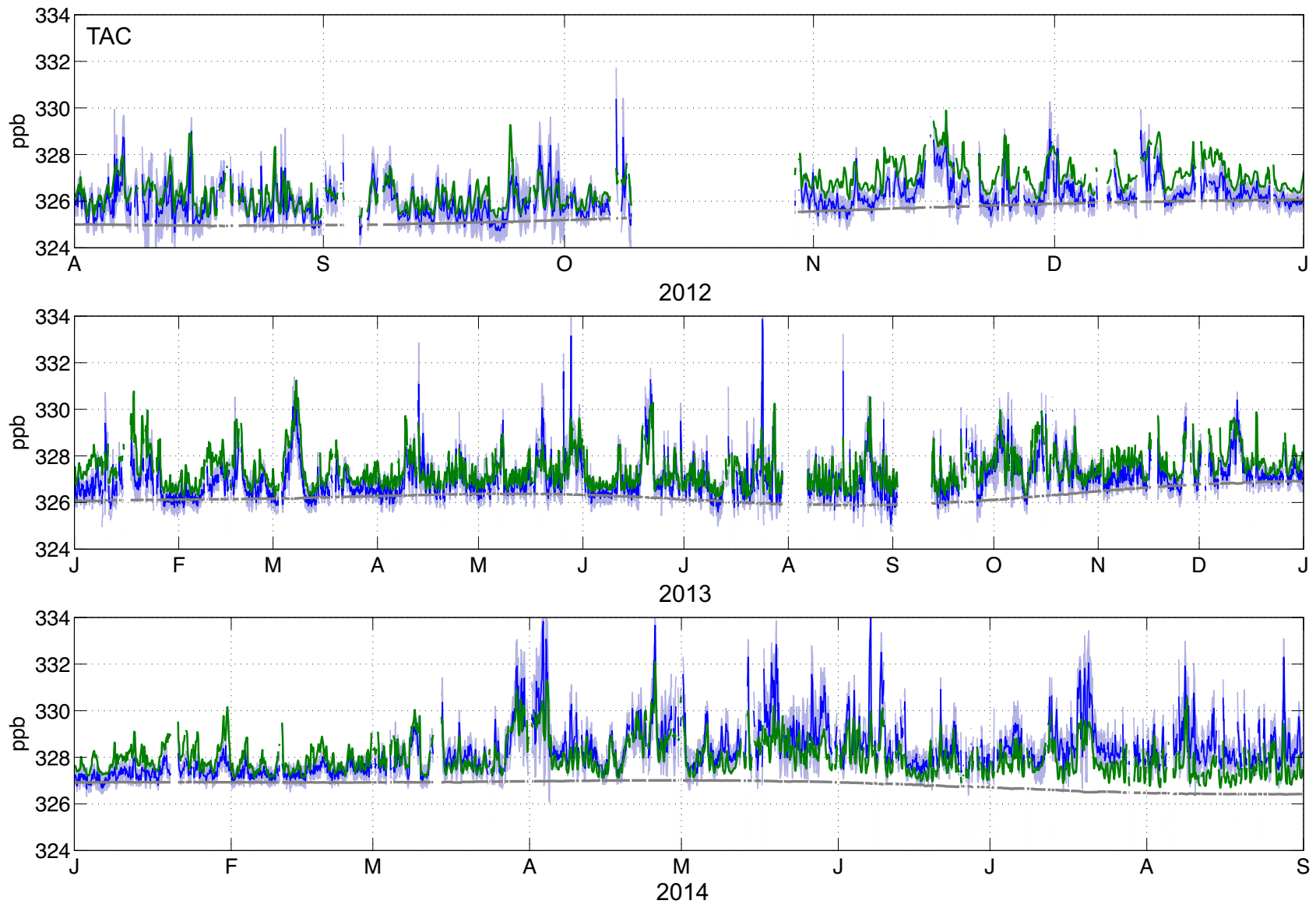


Figure 15: Simulated prior N₂O mole fractions from TAC by year (green), observations (blue) and derived baseline (grey). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

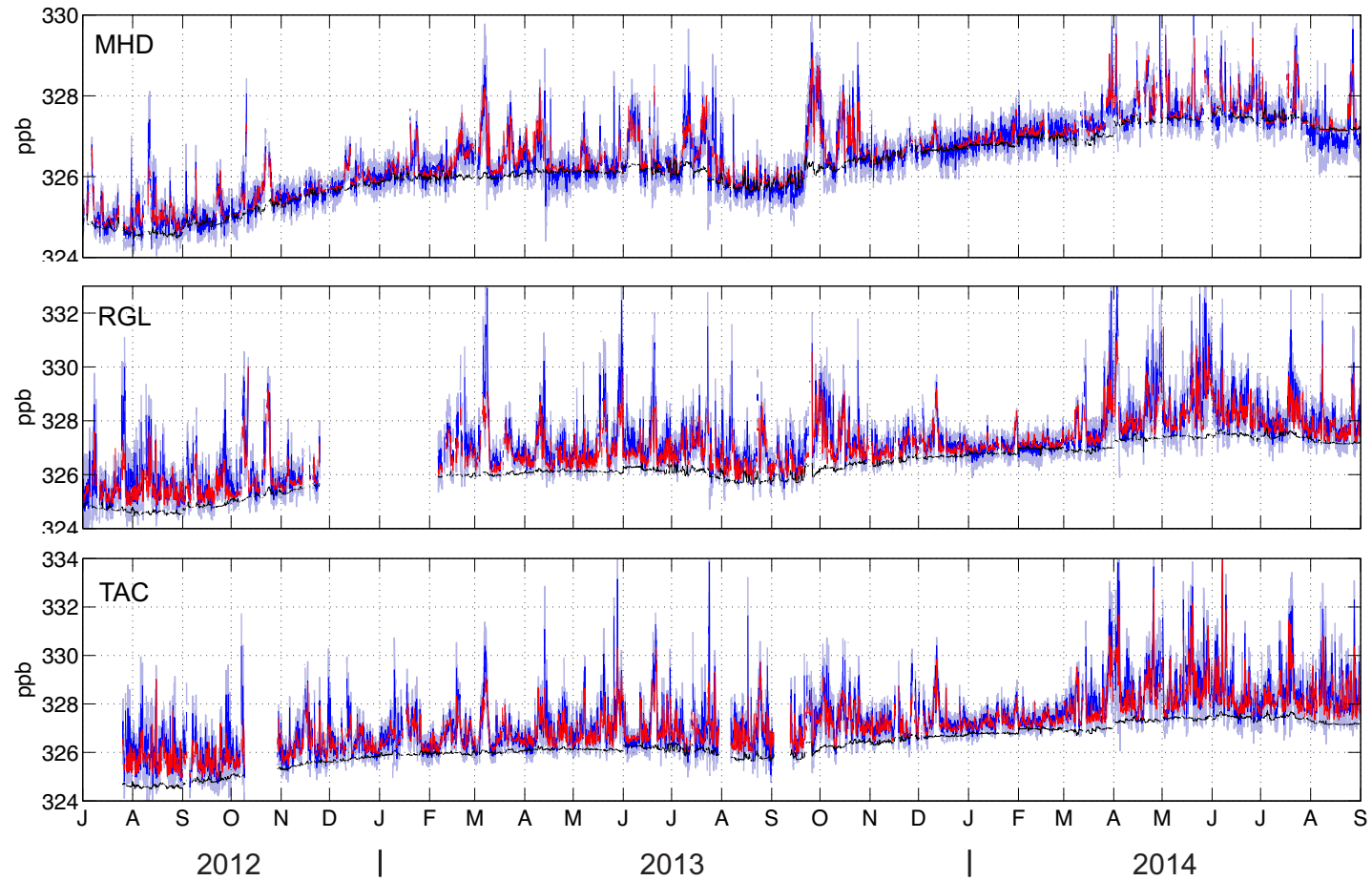


Figure 16: Simulated posterior N₂O mole fractions for all sites and all years (red), observations (blue) and derived baseline (black). Shading indicates the posterior 5th to 95th percentile model errors plus measurement uncertainties.

3 CH₄ sensitivity study

The CH₄ study was also performed excluding measurements from Angus in order to provide a direct comparison with the N₂O study. Figure 17 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 CH₄ and N₂O are not simply due to differences in the network.

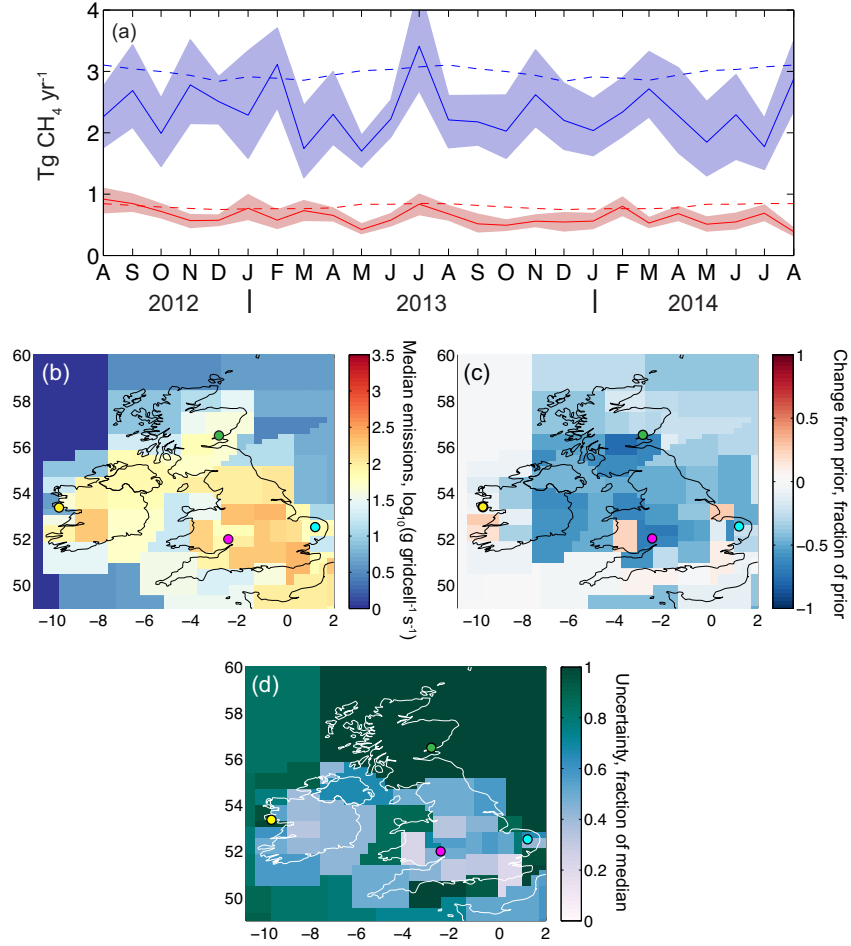


Figure 17: Results derived for the CH₄ sensitivity study in which Angus measurements were excluded. (a) Total national CH₄ emissions for the UK (blue, solid) and Ireland (red, solid) in Tg yr⁻¹. Prior emissions for each country are shown in the dashed lines. Shading corresponds to the 5th to 95th percentile range. (b) Median posterior CH₄ 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).

4 Sensitivity to prior emissions

Two sensitivity studies were performed to assess the effect of the prior emissions on the posterior solution. In the first, it was assumed that there were no natural sources in the prior, therefore, only anthropogenic sources were included. In the second inversion, we assume that the prior emissions consisted of anthropogenic emissions and natural emissions, but the natural emissions were not scaled based on land cover in each country. In the original inversion (including natural sources), we inferred average UK total emissions of 2.09 (1.65–2.67) Tg yr⁻¹ CH₄ and 0.101 (0.068–0.150) Tg yr⁻¹ N₂O and Ireland emissions of 0.62 (0.50–0.74) Tg yr⁻¹ CH₄ and 0.025 (0.019–0.033) Tg yr⁻¹ N₂O.

In the sensitivity study with anthropogenic prior only, we derived 2.03 (1.60–2.62) Tg yr⁻¹ CH₄ and 0.100 (0.066–0.145) Tg yr⁻¹ N₂O and Ireland emissions of 0.59 (0.48–0.72) Tg yr⁻¹ CH₄ and 0.024 (0.018–0.032) Tg yr⁻¹ N₂O. In the sensitivity study with natural emissions that were not scaled, we derived 2.08 (1.57–2.72) Tg yr⁻¹ CH₄ and 0.107 (0.071–0.157) Tg yr⁻¹ N₂O and Ireland emissions of 0.62 (0.48–0.78) Tg yr⁻¹ CH₄ and 0.027 (0.02–0.034) Tg yr⁻¹ N₂O.

We show through the three inversions that the four station network has enough data density to constrain the UK and Ireland totals. While Northern Scotland is not very sensitive in the network, by design this is an area with low emissions and therefore does not significantly impact the UK total.

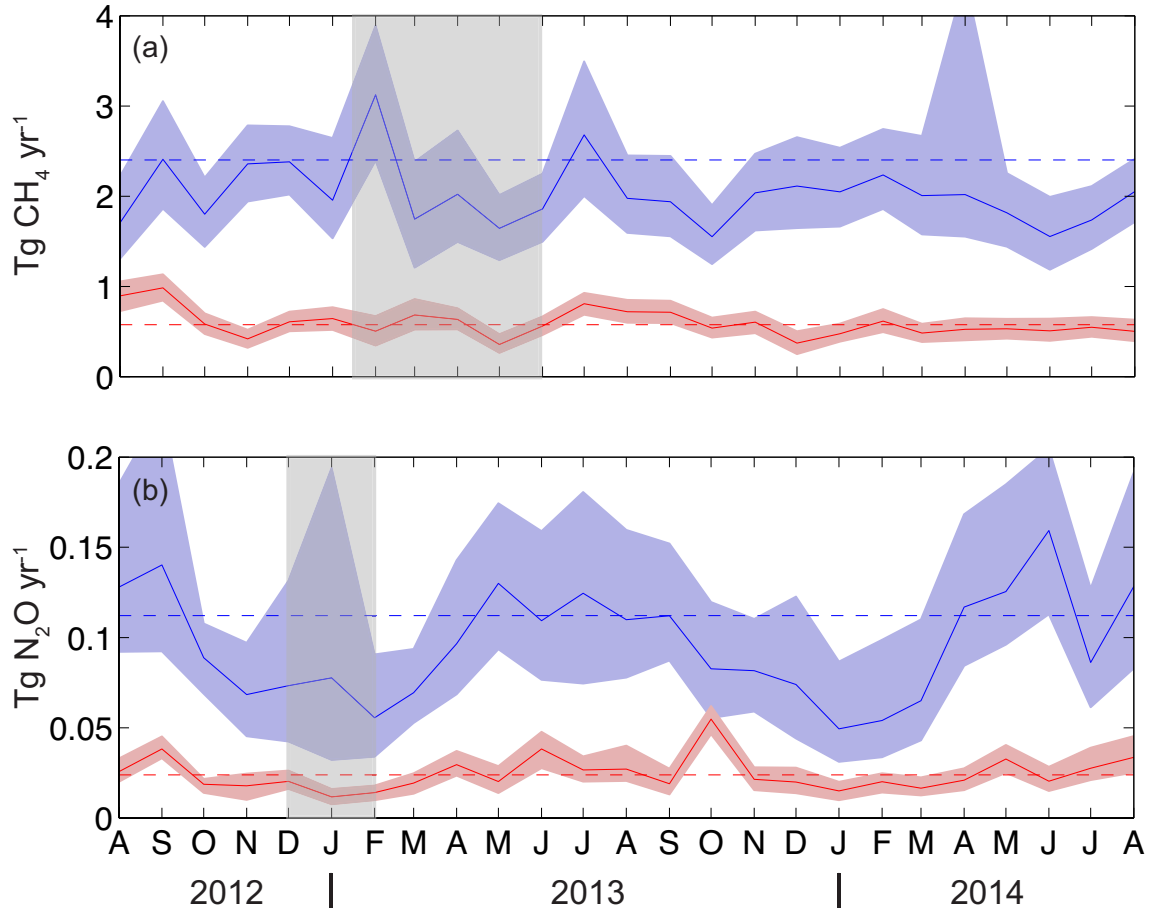


Figure 18: (a) Total national CH₄ and N₂O emissions for the UK (blue, solid) and Ireland (red, solid) in Tg yr⁻¹. Prior (anthropogenic only) emissions for each country are shown in the dashed lines. Shading corresponds to the 5th to 95th percentile range.

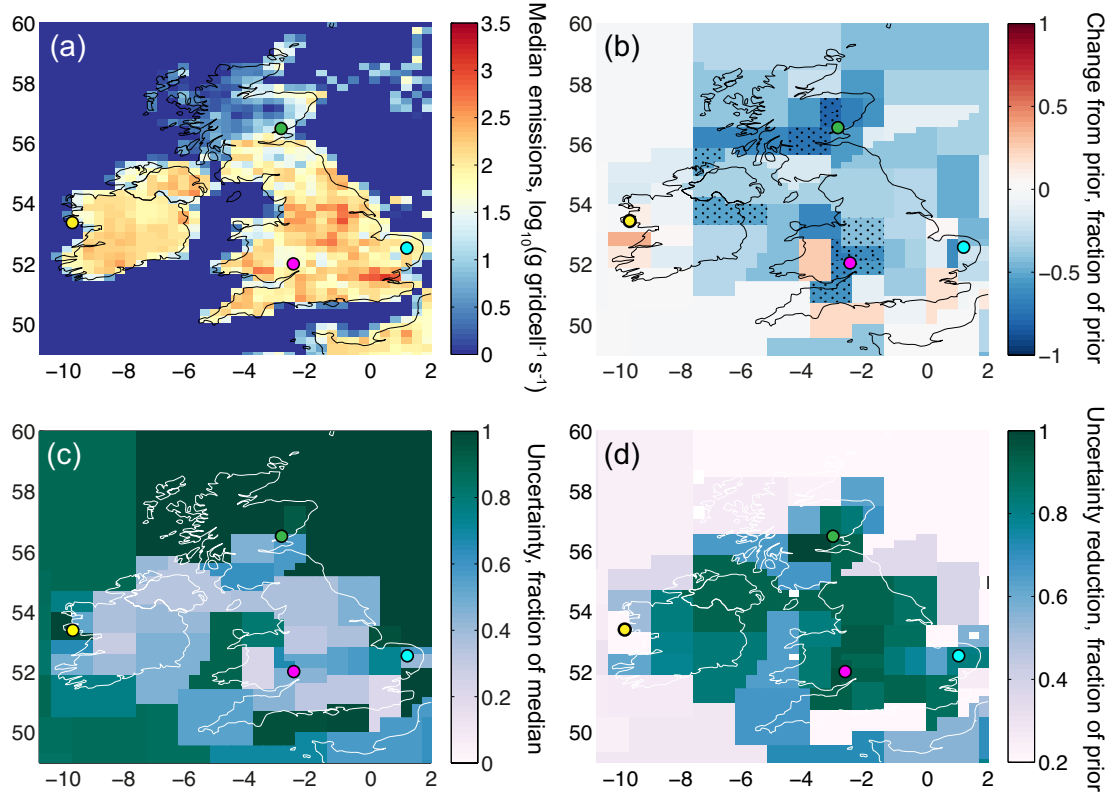


Figure 19: Results derived for the CH₄ sensitivity study in which the prior assumed anthropogenic emissions only. (a) Median posterior CH₄ emissions shown on a logarithmic scale. (b) Fractional difference of the median posterior emissions from the prior. (c) Fractional posterior emissions uncertainty, which corresponds to the difference between the 5th and 95th percentiles, relative to the median. Colored circles show the measurement stations (d) Uncertainty reduction from the prior, relative to the prior (MHD, yellow; RGL, magenta; TAC, cyan; TTA, green).

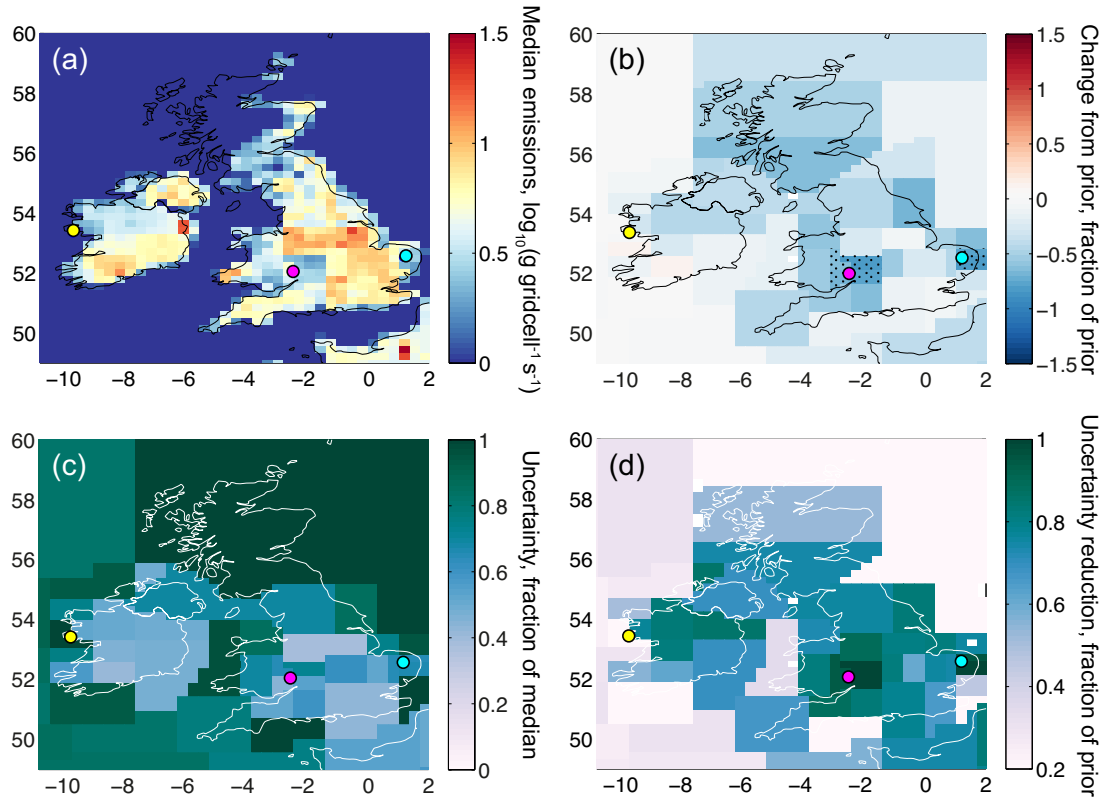


Figure 20: Results derived for the N_2O sensitivity study in which the prior assumed anthropogenic emissions only. (a) Median posterior N_2O emissions shown on a logarithmic scale. (b) Fractional difference of the median posterior emissions from the prior. (c) Fractional posterior emissions uncertainty, which corresponds to the difference between the 5th and 95th percentiles, relative to the median. Colored circles show the measurement stations (d) Uncertainty reduction from the prior, relative to the prior (MHD, yellow; RGL, magenta; TAC, cyan).

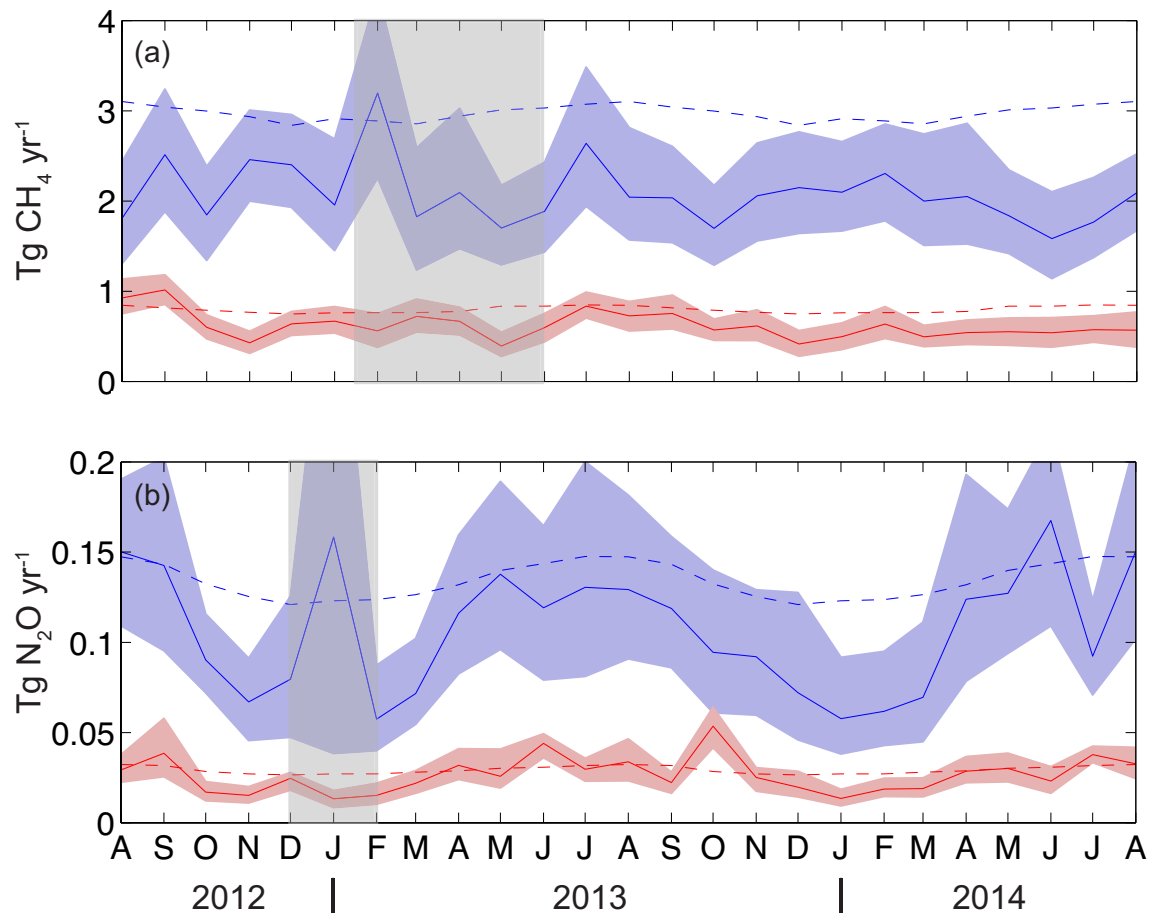


Figure 21: (a) Total national CH_4 and N_2O emissions for the UK (blue, solid) and Ireland (red, solid) in Tg yr^{-1} . Prior (natural emissions not scaled) emissions for each country are shown in the dashed lines. Shading corresponds to the 5th to 95th percentile range.

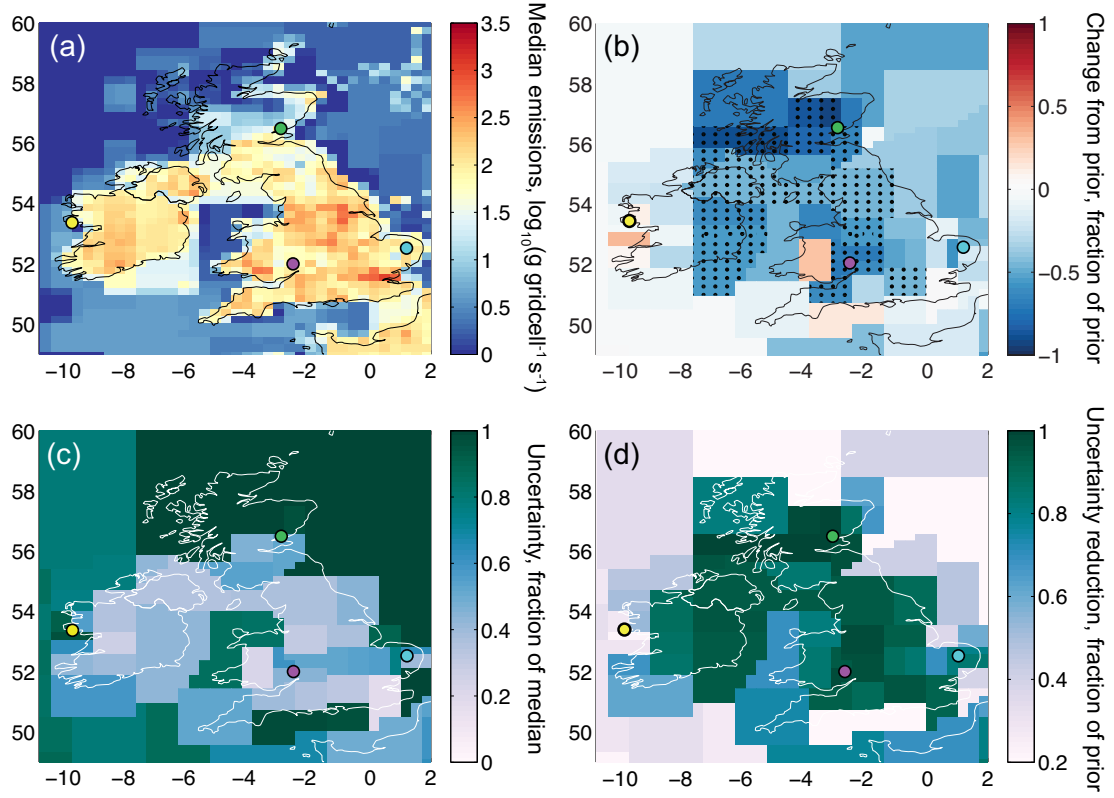


Figure 22: Results derived for the CH₄ sensitivity study in which the prior assumed anthropogenic and natural emissions (not scaled). (a) Median posterior CH₄ emissions shown on a logarithmic scale. (b) Fractional difference of the median posterior emissions from the prior. (c) Fractional posterior emissions uncertainty, which corresponds to the difference between the 5th and 95th percentiles, relative to the median. Colored circles show the measurement stations (d) Uncertainty reduction from the prior, relative to the prior (MHD, yellow; RGL, magenta; TAC, cyan; TTA, green).

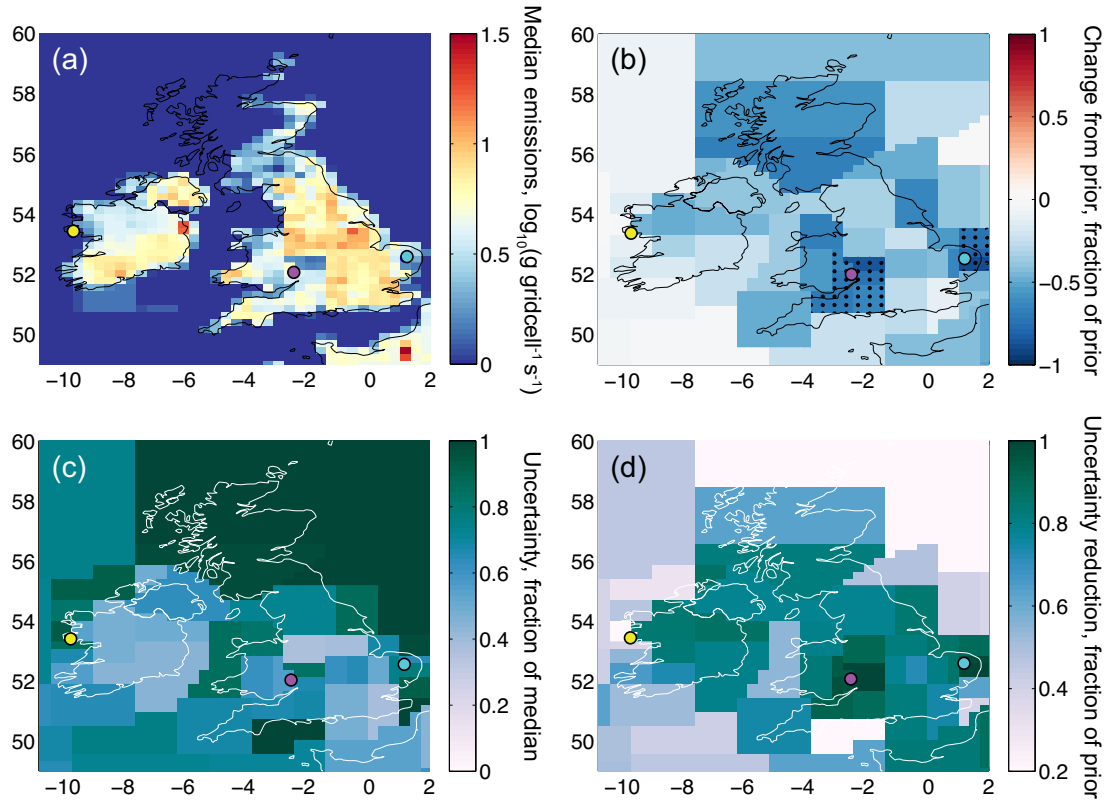


Figure 23: Results derived for the N_2O sensitivity study in which the prior assumed anthropogenic and natural emissions (not scaled). (a) Median posterior N_2O emissions shown on a logarithmic scale. (b) Fractional difference of the median posterior emissions from the prior. (c) Fractional posterior emissions uncertainty, which corresponds to the difference between the 5th and 95th percentiles, relative to the median. Colored circles show the measurement stations (d) Uncertainty reduction from the prior, relative to the prior (MHD, yellow; RGL, magenta; TAC, cyan).