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Supplement of

Inverse modelling of CF_4 and NF_3 emissions in East Asia

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Supporting Text

This describes the method for calculating the well mixed Northern Hemisphere baseline mixing ratios using Mace Head measurements as summarised in Section 2.6 of the main text.

A 2-hour period at Mace Head is classed as 'baseline' if it meets the following criteria:

- The total air concentration from the nine grid boxes centred on and surrounding Mace Head is less than a low (arbitrary) limit. The limit is set so that it is clear that local emissions do not significantly contribute.
- The total contribution from populated areas is less than a low (arbitrary) limit. The limit is set so that it is clear that populated regions have not significantly contributed. The chosen limit is arbitrary but the impact of doubling it is small.
- The percentage of air entering from the north (directions 3 and 4 in Figure 2 in the main text) and west (directions 1 and 2 in Figure 2 in the main text) edges dominates (>90%).
- Less than 4% of the air entering the domain has come from higher than 9km, i.e. from the upper troposphere.

The limits chosen attempt to define a threshold below which any emission sources would generate a concentration at Mace Head that would not be discernible above the baseline noise. The same limit value is used for all of the gases analysed. The points defined as baseline using the above methodology still have a certain level of noise. The principle reasons for this are: unexpected short-lived emissions, local effects that are not identified, incorrectly modelled meteorology or transport, i.e. European or southerly or upper troposphere air defined as baseline by error. Irrespective of the methodology used to identify these events some will inevitably be classed as baseline when it is inappropriate to do so. To capture such events the baseline data are statistically filtered to isolate and remove these non-baseline observations. For each baseline point in turn, the baseline points in a 40-day window surrounding this central value are considered and, provided that there are sufficient points (>11 with at least 4 in each third of the time window or more than 18 in two thirds of the time window), a quadratic is fitted to these values. The standard deviation of the actual points and the fitted curve is calculated (std) and if the current baseline value is more than x std away from the fitted value it is marked for exclusion from the baseline observations. After all baseline points have been considered, those to be excluded are removed. The process is repeated nine times, each time the value for x is gradually reduced from 6 to 2, thus ensuring that those points statistically far from the fitted baseline do not unduly affect the points to be excluded by skewing the fitted curve. If there are insufficient baseline points in a 40-day window the values are only included if the spread in the points is small and there are at least 5 data points

For each hour in the time-series the baseline points in a running 40-day window are fitted using a quadratic function and the value extracted for the current hour in question. The process is then advanced by an hour and repeated. If there are insufficient baseline points well-spaced within the window (at least 3 in each quarter) it is gradually extended up to 150 days.

For each hour within the observation time record a smoothed baseline concentration is estimated by taking the median of all fitted baseline values within a 20-day time window. If there are fewer than 72 baseline values in the time window then the window is steadily increased up to a maximum of 40 days. The noise or potential error in the smoothed baseline concentration ($\sigma_{baseline}$ in Section 2.5 in the main text) is estimated to be the standard deviation of the difference between the observations classed as baseline and the smoothed baseline concentrations at the corresponding times.

Supporting Figures

Figure S1
2010 prior emission map (g m^{-2} yr^{-1}) of CF_4 used in the inversion (top): Emissions are spread evenly within 14 discrete regions over the entire domain. These discrete regions included South Korea, North Korea, East China, West China (China split via a central north-south dividing line), Taiwan and Japan. Emissions were calculated by averaging the Edgar v4.2 emissions database estimates over each of the 14 regions (bottom; Edgar's 0.1°×0.1° displayed on our inversion grid resolution 0.352°×0.234°)

Figure S2
Annual time series (2008-2011) of HFC-23 observations (black), posterior baseline (red) and measurement-model error applied to each observation (grey). Statistics for each comparison year of measurements to model output also shown.

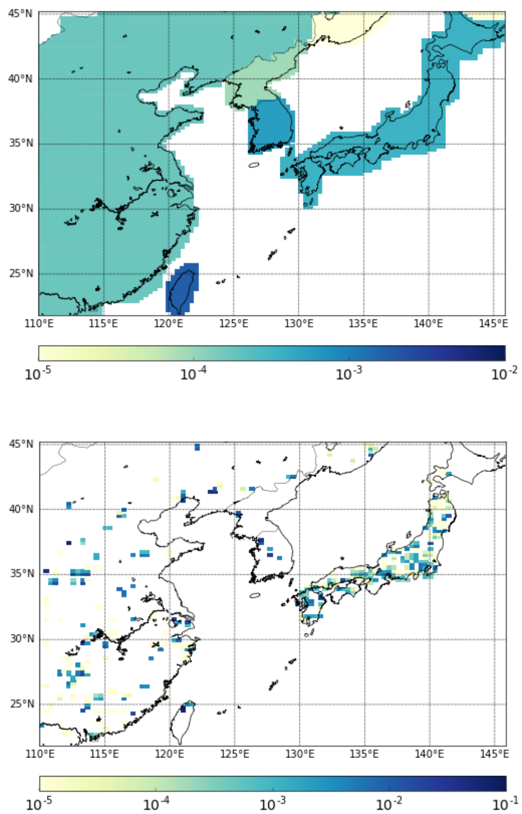
Figure S3
Same as S2 for 2012

Figure S4
Same as S2 for CF_4

Figure S5
Same as S4 for 2012-2015

Figure S6
Same as S4 for NF_3 (no measurements in 2012, and only measurements in the later part of 2013 were made and are not shown here)

Figure S7
Posterior emissions minus uncertainty for all years for CF_4 corresponding to emissions magnitudes shown in Figure 6 in the main text. Results are from inversions with initial uncertainty on the prior emissions field is set to 100 times emissions at each fine grid square. Units in g m^{-2} yr^{-1}.



45°N
40°N
35°N
30°N
25°N
110°E
115°E
120°E
125°E
130°E
135°E
140°E
145°E
10^{-5}
10^{-4}
10^{-3}
10^{-2}
45°N
40°N
35°N
30°N
25°N
110°E
115°E
120°E
125°E
130°E
135°E
140°E
145°E
10^{-5}
10^{-4}
10^{-3}
10^{-2}
10^{-1}

Figure S1

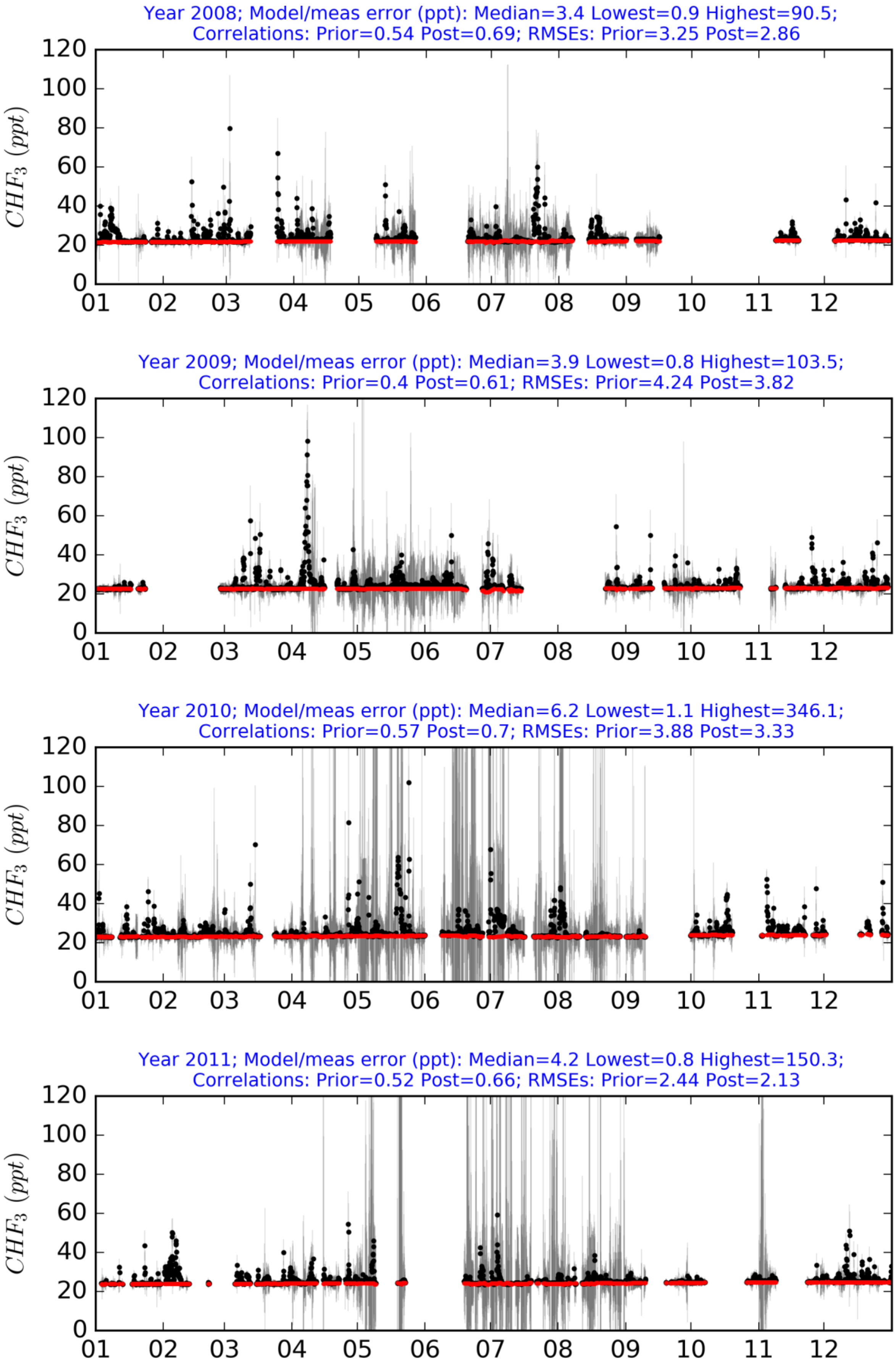
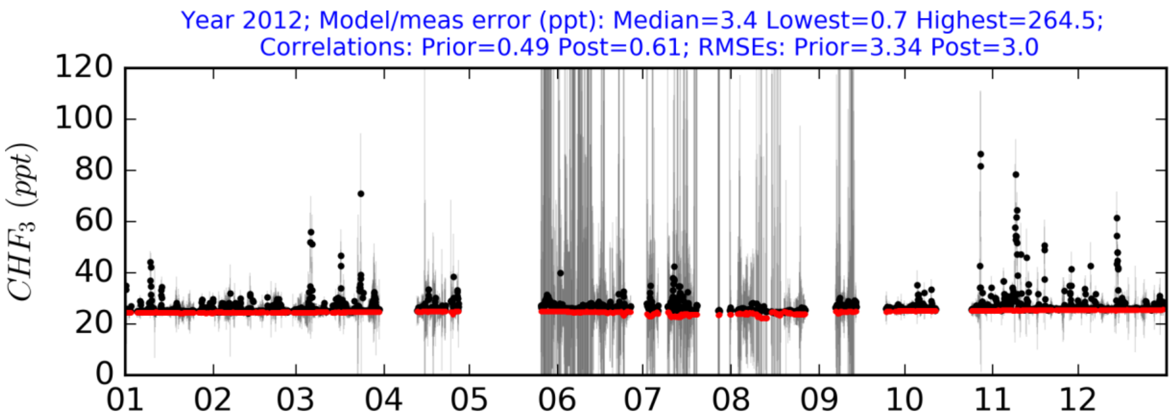

Year 2008; Model/meas error (ppt): Median=3.4 Lowest=0.9 Highest=90.5;
Correlations: Prior=0.54 Post=0.69; RMSEs: Prior=3.25 Post=2.86
$CHF_3\ (ppt)$
Year 2009; Model/meas error (ppt): Median=3.9 Lowest=0.8 Highest=103.5;
Correlations: Prior=0.4 Post=0.61; RMSEs: Prior=4.24 Post=3.82
$CHF_3\ (ppt)$
Year 2010; Model/meas error (ppt): Median=6.2 Lowest=1.1 Highest=346.1;
Correlations: Prior=0.57 Post=0.7; RMSEs: Prior=3.88 Post=3.33
$CHF_3\ (ppt)$
Year 2011; Model/meas error (ppt): Median=4.2 Lowest=0.8 Highest=150.3;
Correlations: Prior=0.52 Post=0.66; RMSEs: Prior=2.44 Post=2.13
$CHF_3\ (ppt)$
0
20
40
60
80
100
120
01
02
03
04
05
06
07
08
09
10
11
12

Figure S2


Year 2012; Model/meas error (ppt): Median=3.4 Lowest=0.7 Highest=264.5;
Correlations: Prior=0.49 Post=0.61; RMSEs: Prior=3.34 Post=3.0
CHF_3 (ppt)
120
100
80
60
40
20
0
01
02
03
04
05
06
07
08
09
10
11
12
Figure S3

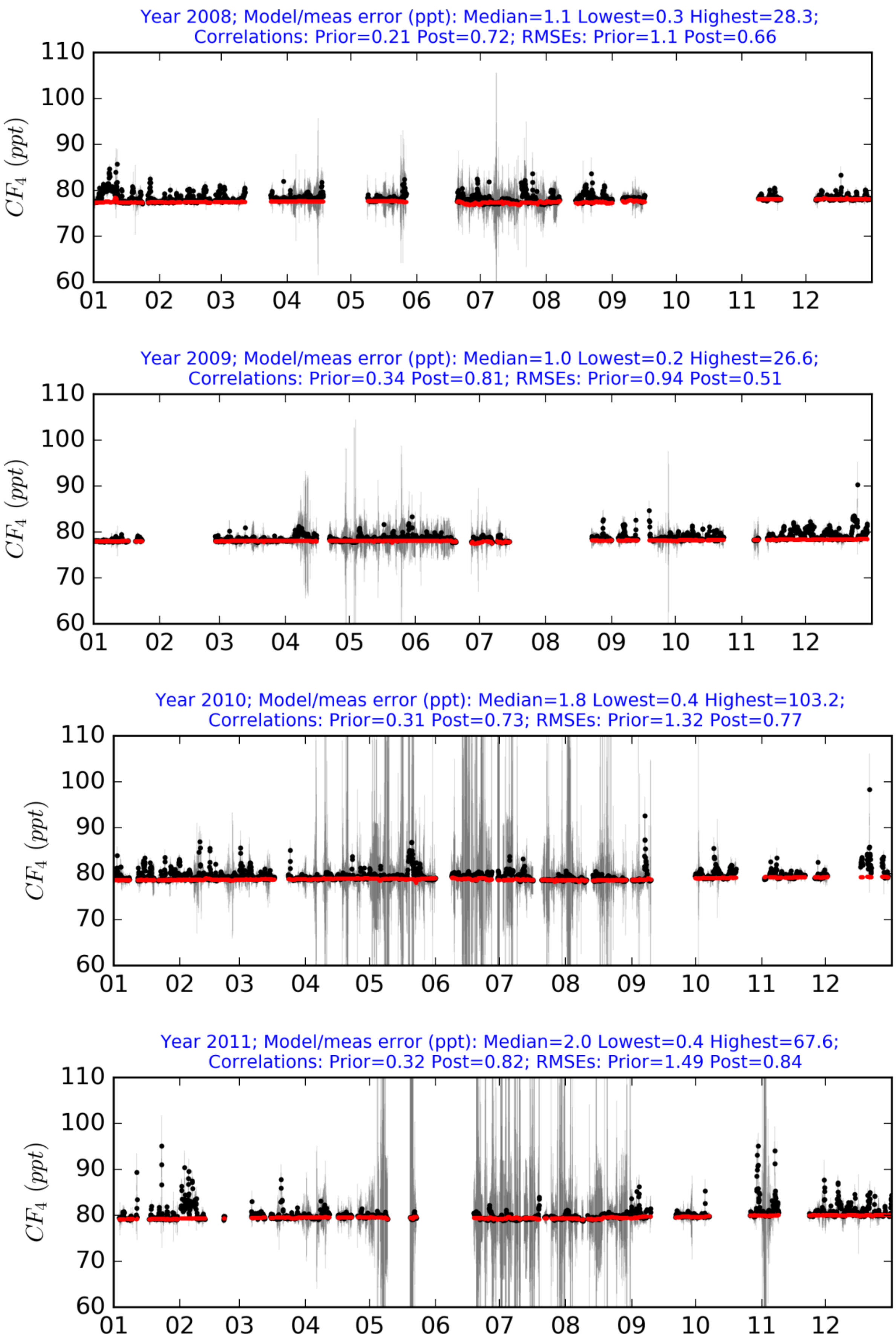
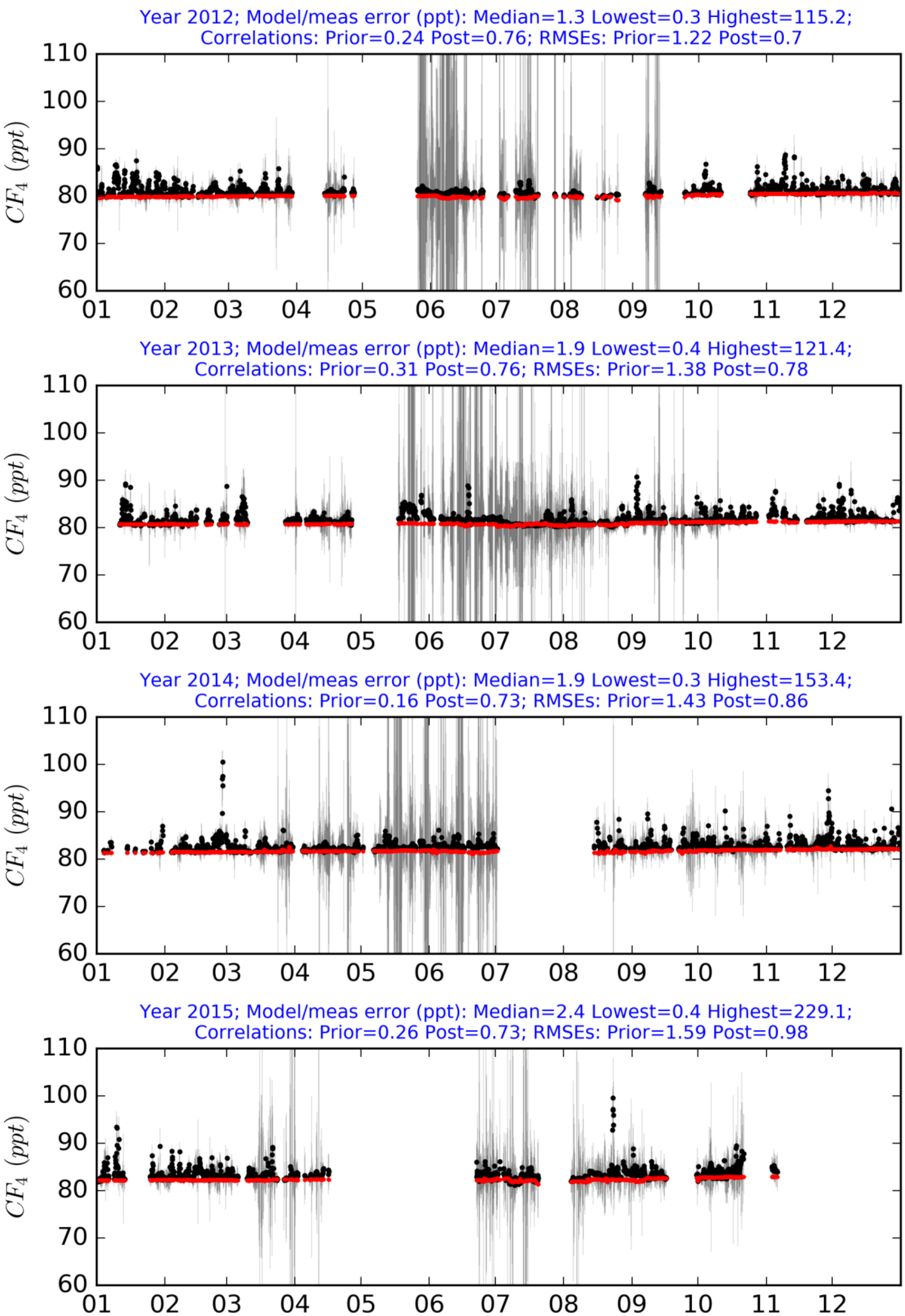

Year 2008; Model/meas error (ppt): Median=1.1 Lowest=0.3 Highest=28.3;
Correlations: Prior=0.21 Post=0.72; RMSEs: Prior=1.1 Post=0.66
Year 2009; Model/meas error (ppt): Median=1.0 Lowest=0.2 Highest=26.6;
Correlations: Prior=0.34 Post=0.81; RMSEs: Prior=0.94 Post=0.51
Year 2010; Model/meas error (ppt): Median=1.8 Lowest=0.4 Highest=103.2;
Correlations: Prior=0.31 Post=0.73; RMSEs: Prior=1.32 Post=0.77
Year 2011; Model/meas error (ppt): Median=2.0 Lowest=0.4 Highest=67.6;
Correlations: Prior=0.32 Post=0.82; RMSEs: Prior=1.49 Post=0.84
CF_4 (ppt)
110
100
90
80
70
60
01 02 03 04 05 06 07 08 09 10 11 12

Figure S4



Year 2012; Model/meas error (ppt): Median=1.3 Lowest=0.3 Highest=115.2;
Correlations: Prior=0.24 Post=0.76; RMSEs: Prior=1.22 Post=0.7
Year 2013; Model/meas error (ppt): Median=1.9 Lowest=0.4 Highest=121.4;
Correlations: Prior=0.31 Post=0.76; RMSEs: Prior=1.38 Post=0.78
Year 2014; Model/meas error (ppt): Median=1.9 Lowest=0.3 Highest=153.4;
Correlations: Prior=0.16 Post=0.73; RMSEs: Prior=1.43 Post=0.86
Year 2015; Model/meas error (ppt): Median=2.4 Lowest=0.4 Highest=229.1;
Correlations: Prior=0.26 Post=0.73; RMSEs: Prior=1.59 Post=0.98
CF_4 (ppt)
60
70
80
90
100
110
01
02
03
04
05
06
07
08
09
10
11
12

Figure S5

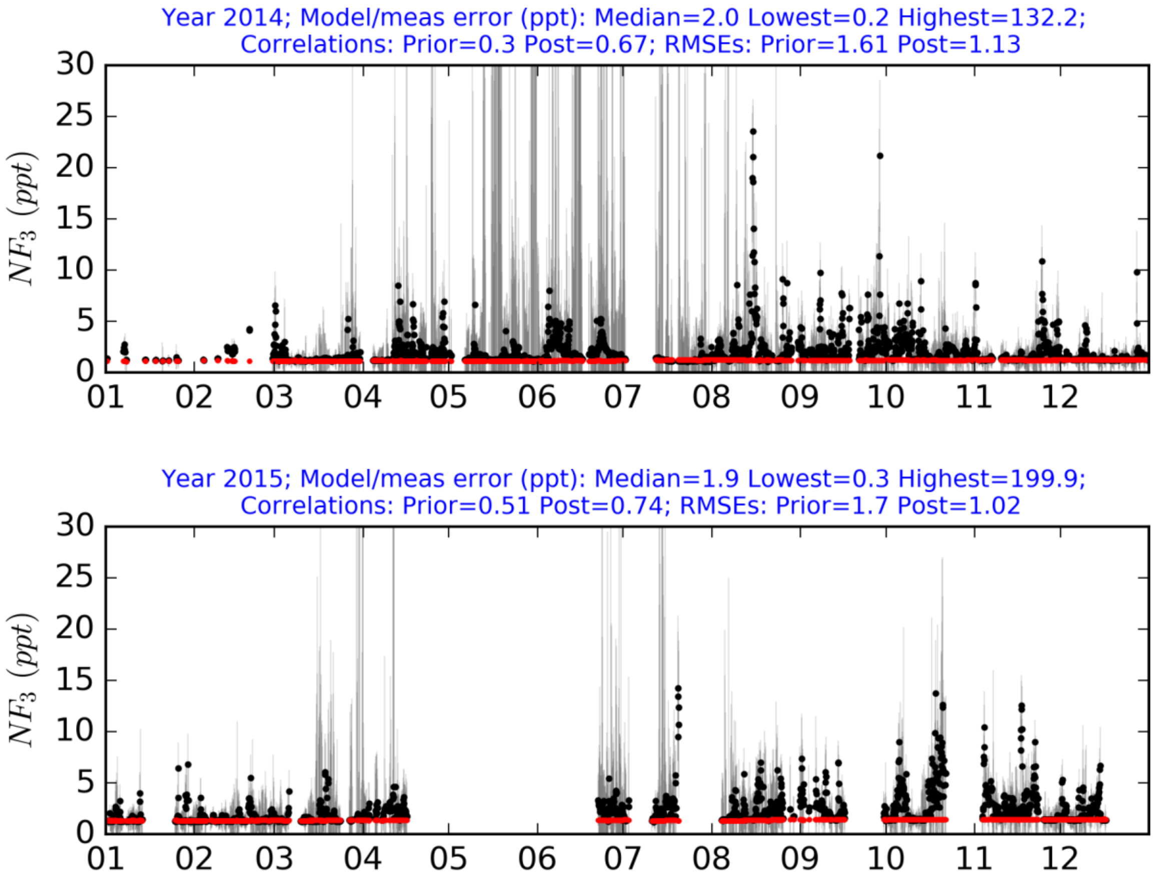

Year 2014; Model/meas error (ppt): Median=2.0 Lowest=0.2 Highest=132.2;
Correlations: Prior=0.3 Post=0.67; RMSEs: Prior=1.61 Post=1.13
NF_3 (ppt)
0 5 10 15 20 25 30
01 02 03 04 05 06 07 08 09 10 11 12
Year 2015; Model/meas error (ppt): Median=1.9 Lowest=0.3 Highest=199.9;
Correlations: Prior=0.51 Post=0.74; RMSEs: Prior=1.7 Post=1.02
NF_3 (ppt)
0 5 10 15 20 25 30
01 02 03 04 05 06 07 08 09 10 11 12

Figure S6

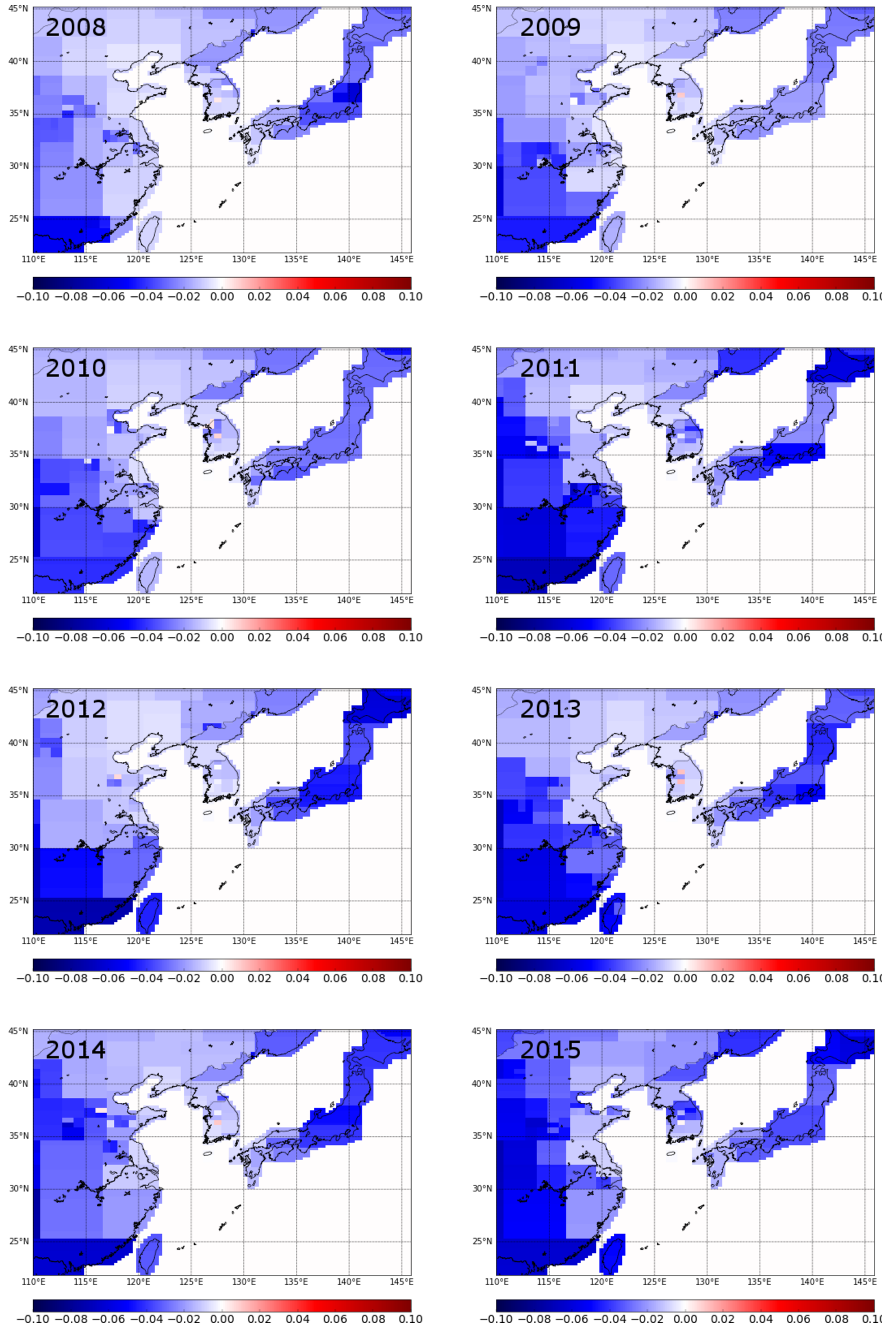

2008
2009
2010
2011
2012
2013
2014
2015
45°N
40°N
35°N
30°N
25°N
110°E
115°E
120°E
125°E
130°E
135°E
140°E
145°E
−0.10 −0.08 −0.06 −0.04 −0.02 0.00 0.02 0.04 0.06 0.08 0.10

Figure S7