Supplementary Material

Table S1 lists the aircraft instrumentation on the Y12 aircraft during ARIAs. For synchronization of aircraft measurements, we first synchronized Picarro measurements of CO_2, CO and CH_4, which have a 6-second lag time that takes for a plume to transport from the aircraft sample inlet to the Picarro cavity to be detected by the analyzer. The lag time was determined by introducing a pulse of CO_2/CO/CH_4 calibration standard into the sample inlet and then measuring the time it takes for the Picarro to detect the pulsed signals. All other measurements were then synchronized based on concurrent peak appearance of these measurements and the Picarro measurements.

Table S1. UMD Aircraft Instrumentation

Variable	**Method**	**Sample Frequency**	**Precision/Accuracy***
Position	GPS	1 s	Horizontal: ~1 / ±2.5 m Vertical: ~1 / ±3.75 m
Meteorology (T, RH, P, 2-D Wind)	Cloud water inertial probe (CWIP): Hotwire, advanced heading reference system, 5-hole gust probe	1 s	T: 0.2 / ±0.5 °C P: 2.6 hPa / ±0.25% of FS RH: 1 / ±2% WS: 0.5 / ±1.0 m/s WD: 5 / ±10 °
Greenhouse gas CO_2/CH_4/CO/H_2O	Cavity Ring Down Spectroscopy Picarro Model G2401-m	2 s	CO_2: 0.02 / ±0.1 ppm CH_4: 0.2 / ±1 ppb CO: 4.2 / ±10 ppb
Ozone (O_3)	UV Absorption	10 s	1 ppb / ±1%
Sulfur Dioxide (SO_2)	Pulsed Fluorescence	10 s	0.1 ppb / ±3%
Nitrogen Dioxide (NO_2)	Cavity enhanced absorption spectroscopy, Los Gatos	1 s	0.05 ppb / ±5%
Reactive Nitrogen (NO-NOy)	Chemiluminescence	10 s	0.05 ppb / ±3%
Aerosol Scattering, b_{scat} (450, 550, 700 nm)	Nephelometer	1 s	±5x10^{-7} m^{-1} / ±5%
Aerosol Absorption, b_{abs} (565 nm)	Particle Soot Absorption Photometer (PSAP)	1 min	±5x10^{-7} m^{-1} / ±5%
Black Carbon (370, 470, 520, 590, 660, 880, 950 nm)	Aethalometer	2 min	0.05 μg/m^3 / ±5%
VOCs	Grab Canisters/GC-FID	5-6 / flight	Species dependent
Formaldehyde	Wet chemistry and fluorescence detection	90 seconds	0.1 ppb / ±5%

*The precisions are from the instrument specifications provided by the manufacturers while the accuracies are estimated from the uncertainties in calibration standards and mass flow controllers that are used to control flow rates of zero air and calibration gas.

Figure S1. ARIAs flights over the NCP. Eleven Research Flights were conducted in May to Mid-June 2016.

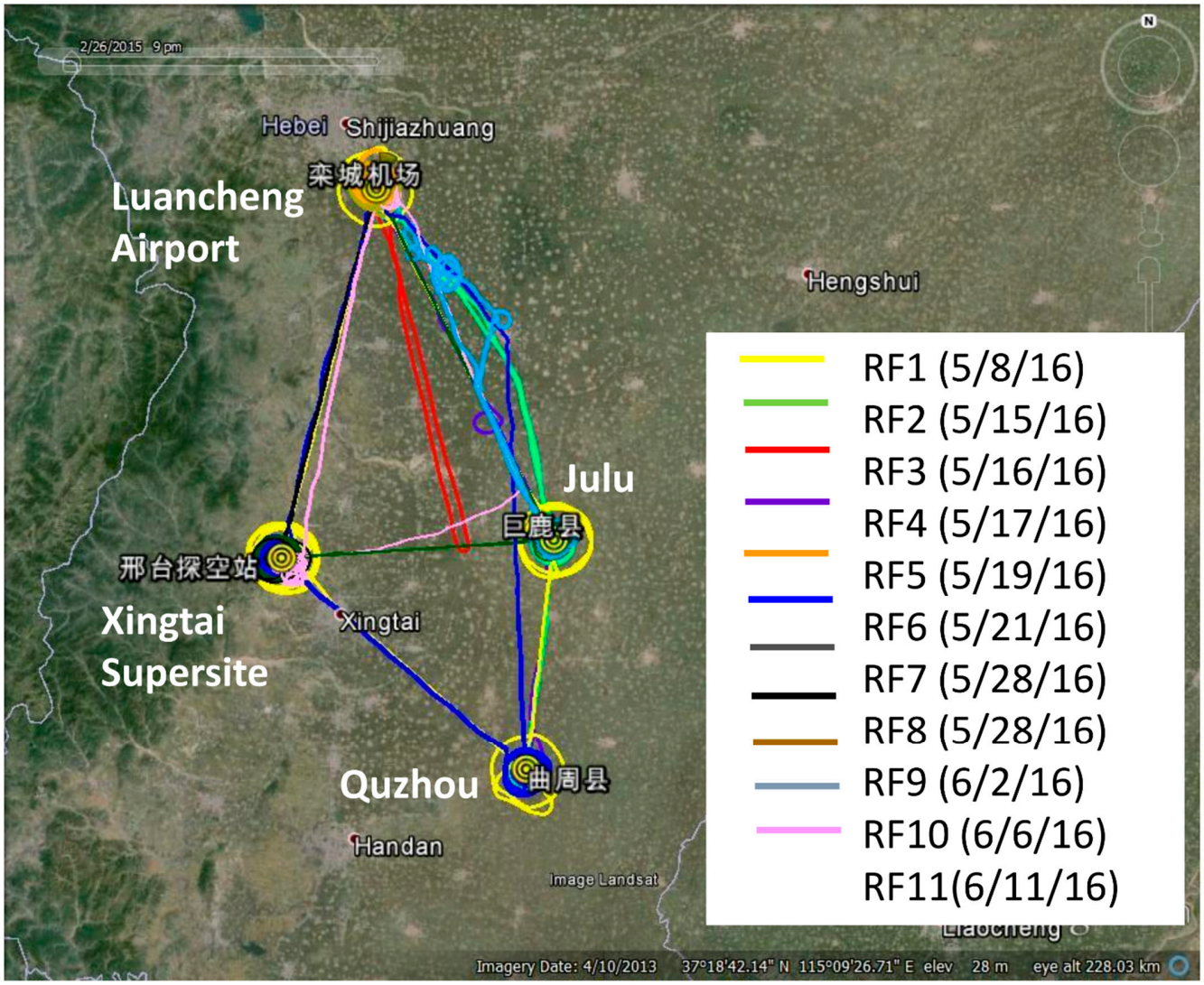


Figure S2. CMEE Air quality monitoring sites in 2016

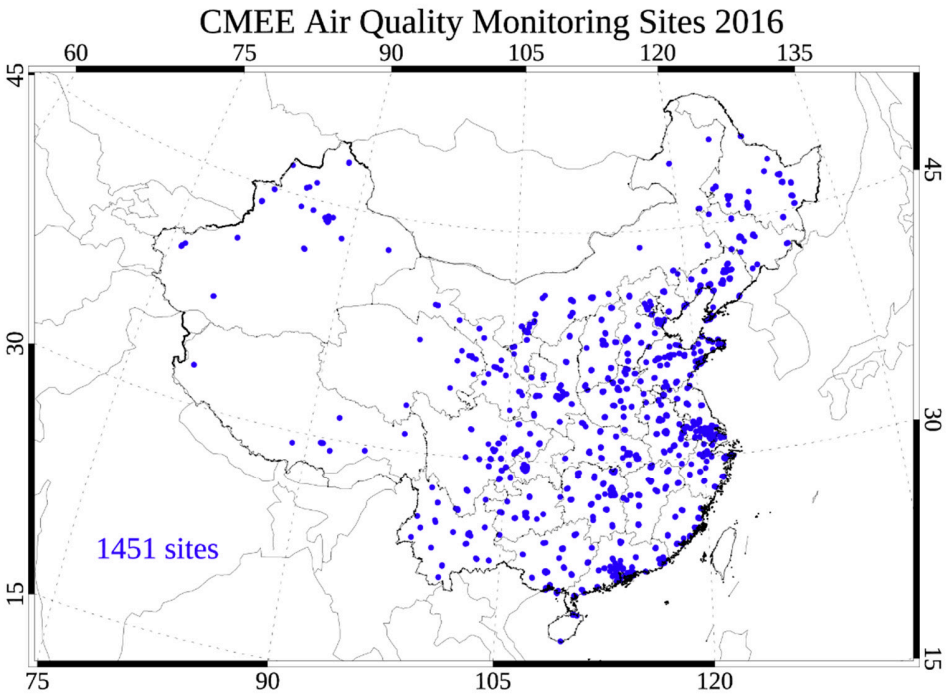


Figure S3. A plume observed over Xingtai during the flight on June 11, 2016. The height of mixing layer is around 1500 m AGL. Secondary peaks NO_2 at 500 m AGL and CO at 1500 m AGL were observed.

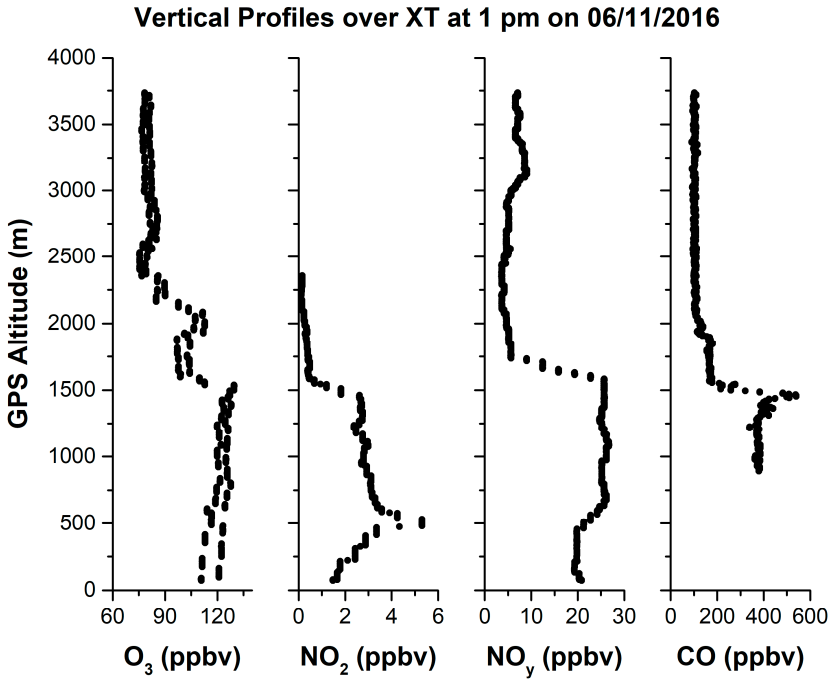
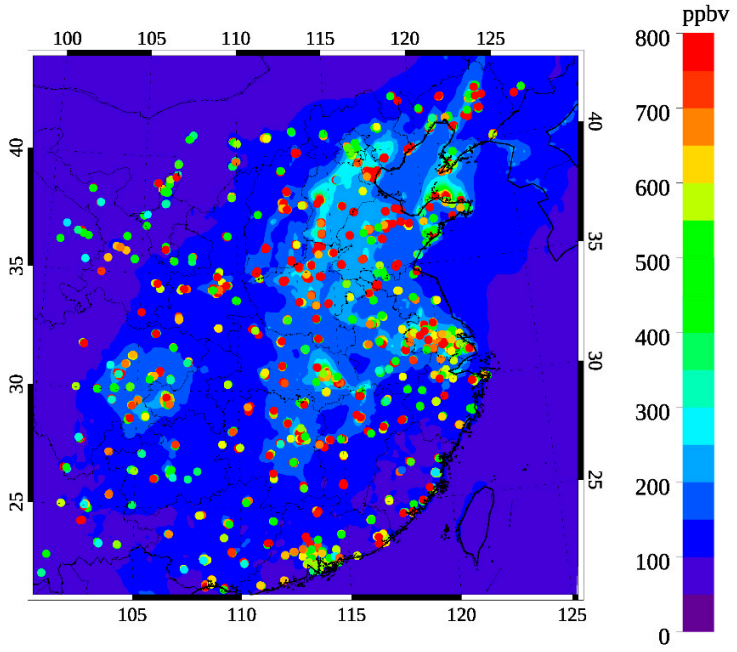
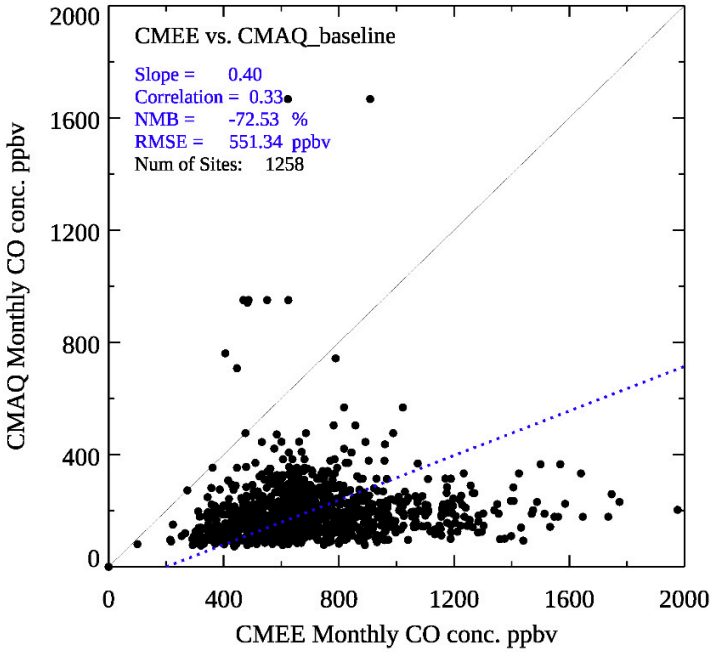


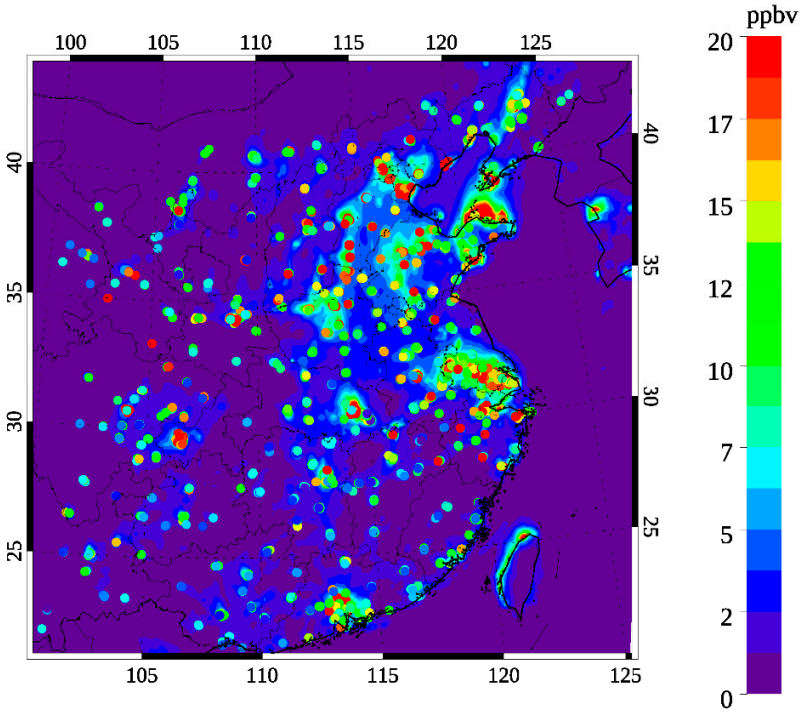
Figure S4. Comparison of mean CO and NO_2 concentrations in May and June 2016 over eastern China. Left: Contour plot, the background stands for mean concentrations from the CMAQ_baseline run; the dots represent observed values from the CMEE network. Right: Scatter plot, blue line is the linear regression fitting. a) CO, b) NO_2.

a)





b)


ppbv
20
17
15
12
10
7
5
2
0
100
105
110
115
120
125
40
35
30
25

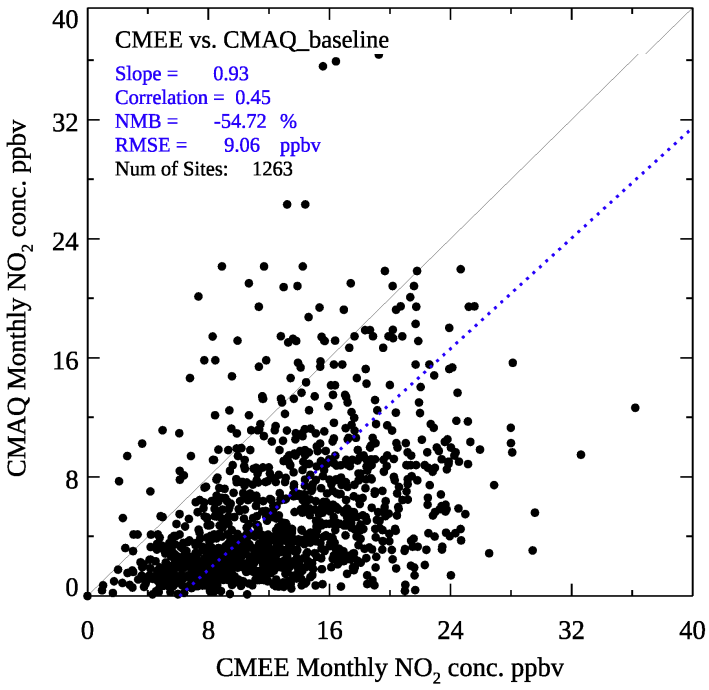
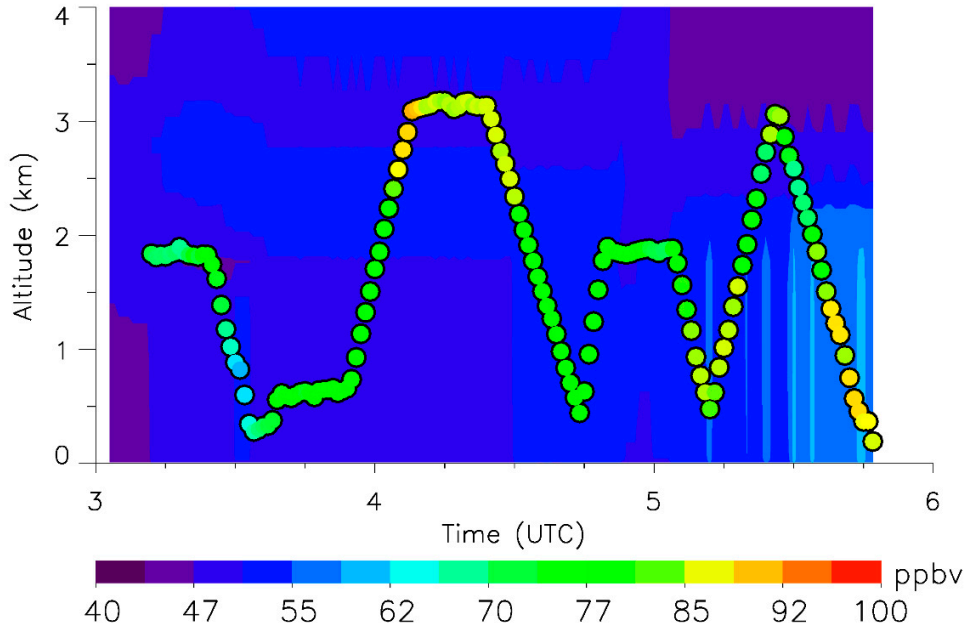
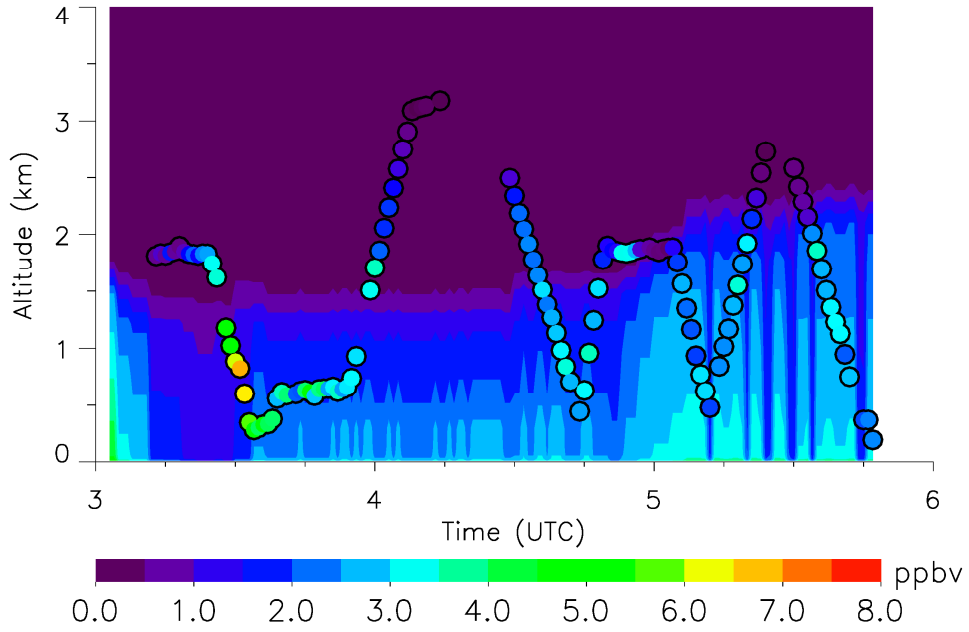

CMEE vs. CMAQ_baseline
Slope = 0.93
Correlation = 0.45
NMB = -54.72 %
RMSE = 9.06 ppbv
Num of Sites: 1263
CMAQ Monthly NO2 conc. ppbv
CMEE Monthly NO2 conc. ppbv
0
8
16
24
32
40

Figure S5. A case study comparing aircraft observations and the CMAQ_baseline run results on June 11, 2016. Background: CMAQ simulations. Overlay: 1-min Y12 measurements. a) O_3, b) NO_2, c) NO, d) NO_y, e) CO.

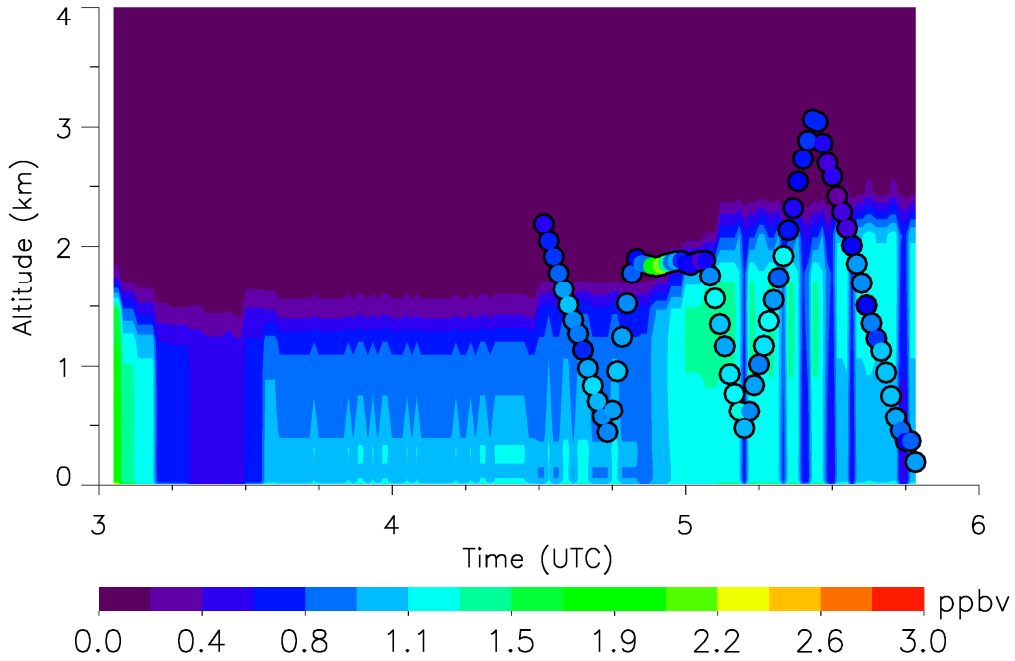
a)



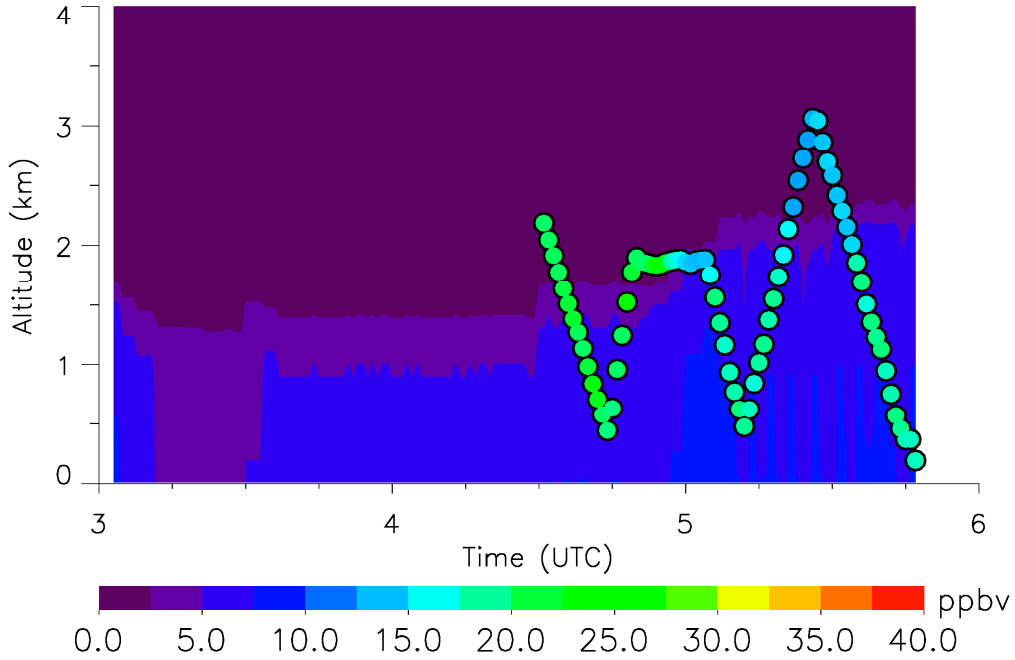
b)



c)


Altitude (km)
0
1
2
3
4
3
4
5
6
Time (UTC)
ppbv
0.0
0.4
0.8
1.1
1.5
1.9
2.2
2.6
3.0

d)


Altitude (km)
0
1
2
3
4
3
4
5
6
Time (UTC)
ppbv
0.0
5.0
10.0
15.0
20.0
25.0
30.0
35.0
40.0

e)

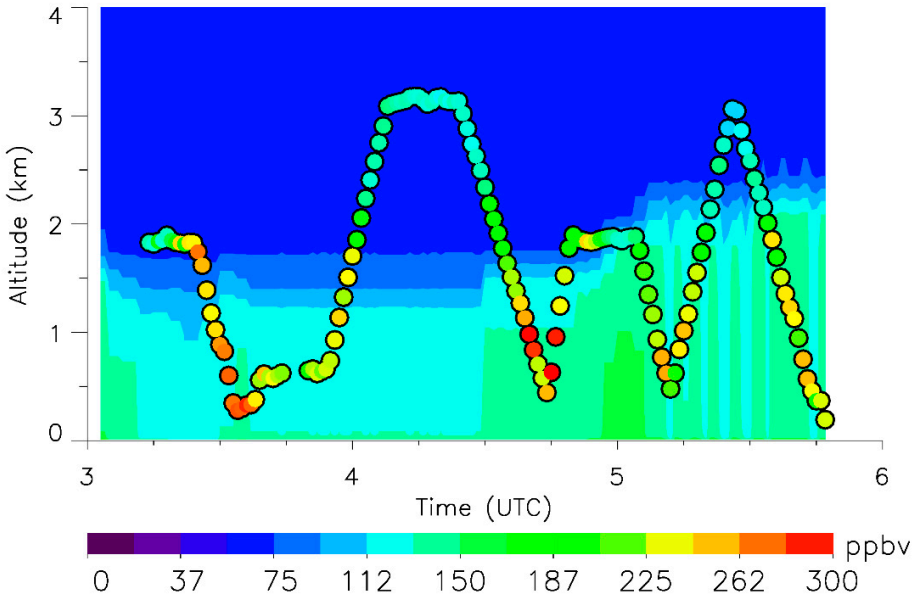
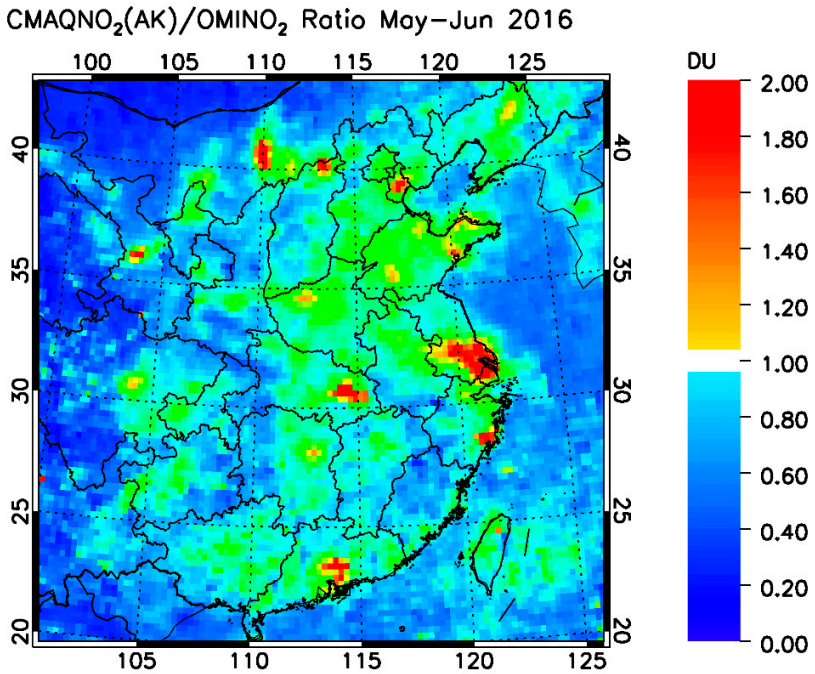
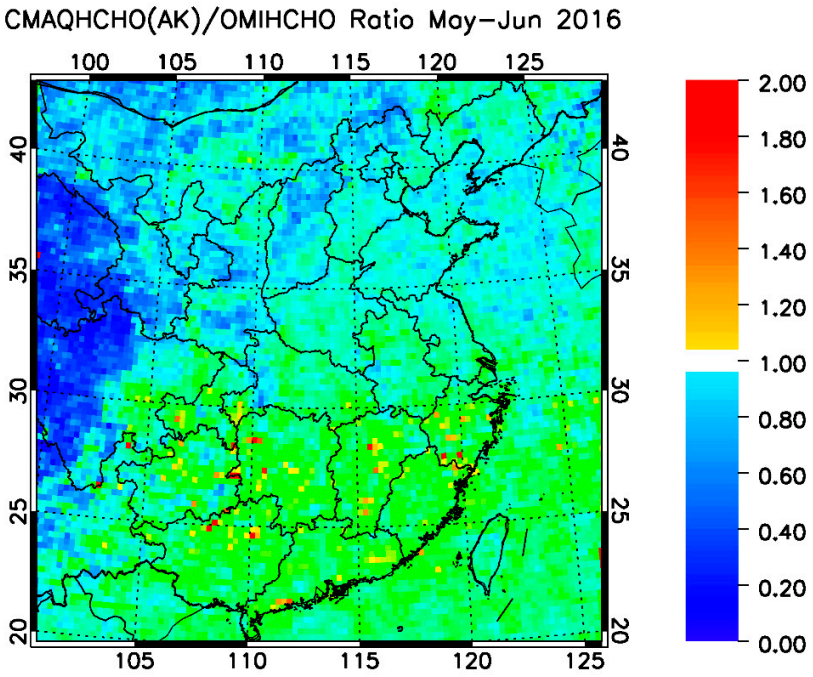


Figure S6. Ratios of column contents of the CMAQ_baseline simulations and satellite observations. a) CMAQ/OMI NO_2; b) CMAQ/OMI HCHO; c) CMAQ/MOPITT CO.

a)



b)


CMAQHCHO(AK)/OMIHCHO Ratio May–Jun 2016
100
105
110
115
120
125
40
35
30
25
20
2.00
1.80
1.60
1.40
1.20
1.00
0.80
0.60
0.40
0.20
0.00

c)

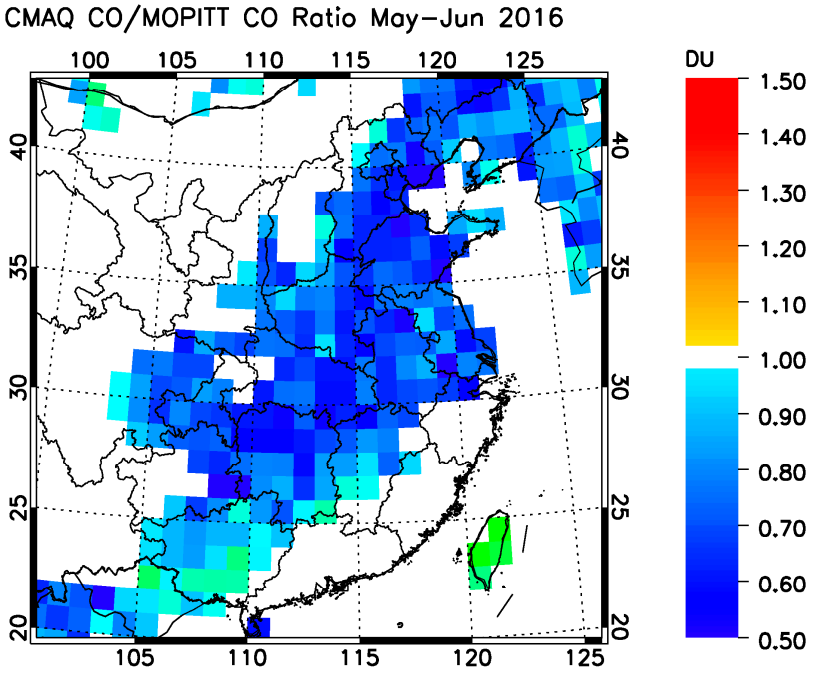
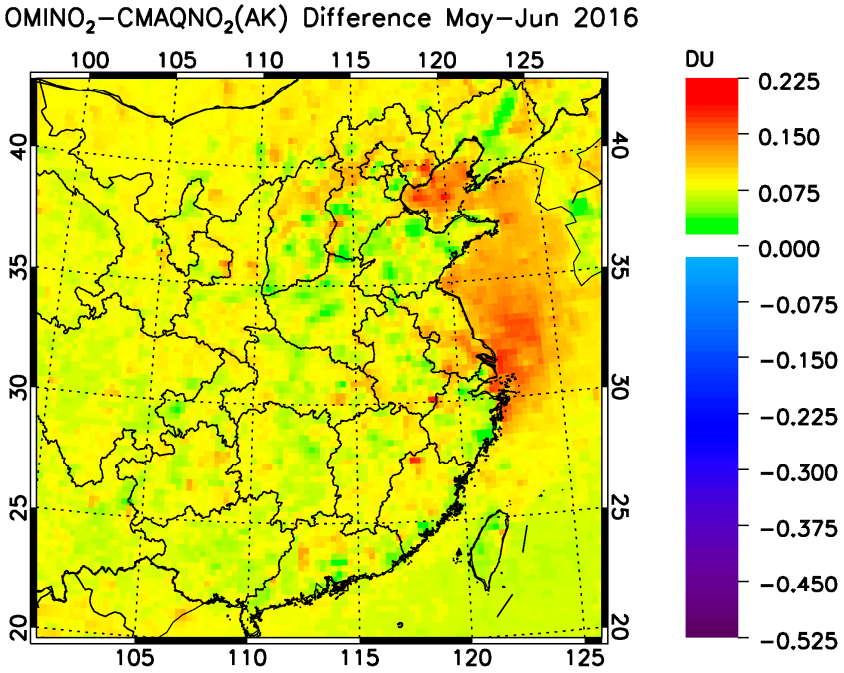
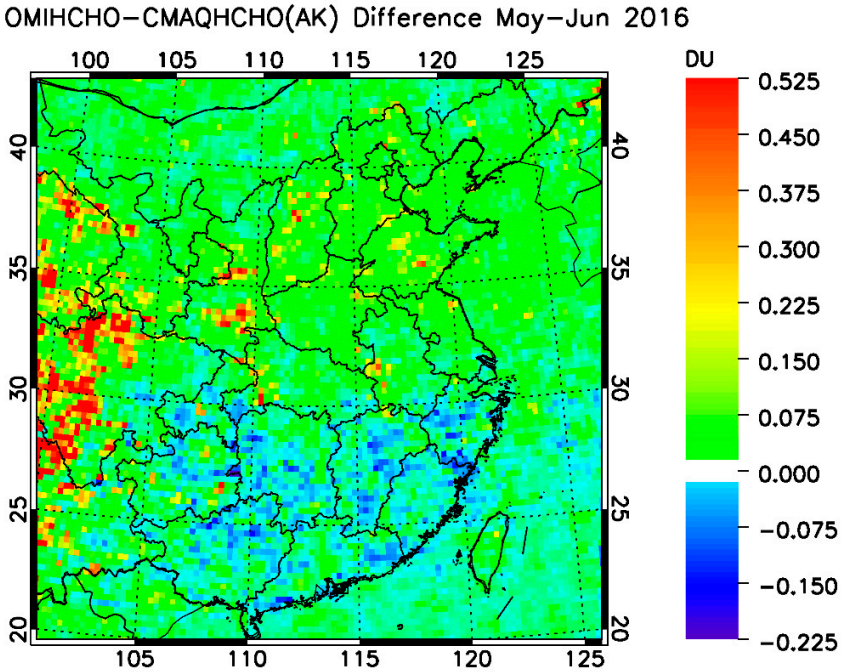

CMAQ CO/MOPITT CO Ratio May–Jun 2016
DU
100
105
110
115
120
125
40
35
30
25
20
1.50
1.40
1.30
1.20
1.10
1.00
0.90
0.80
0.70
0.60
0.50

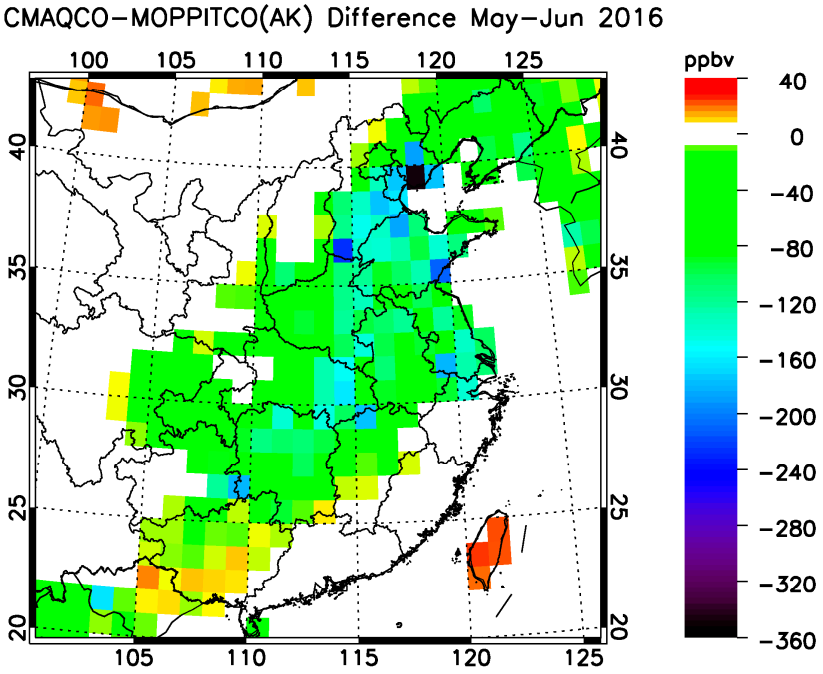
Figure S7. Similar as Figure 8, but shows differences between satellite observations and CMAQ_all simulations in May and June 2016. a) difference of NO_2 column; b) difference of HCHO column; c) difference of near surface CO.

a)



b)

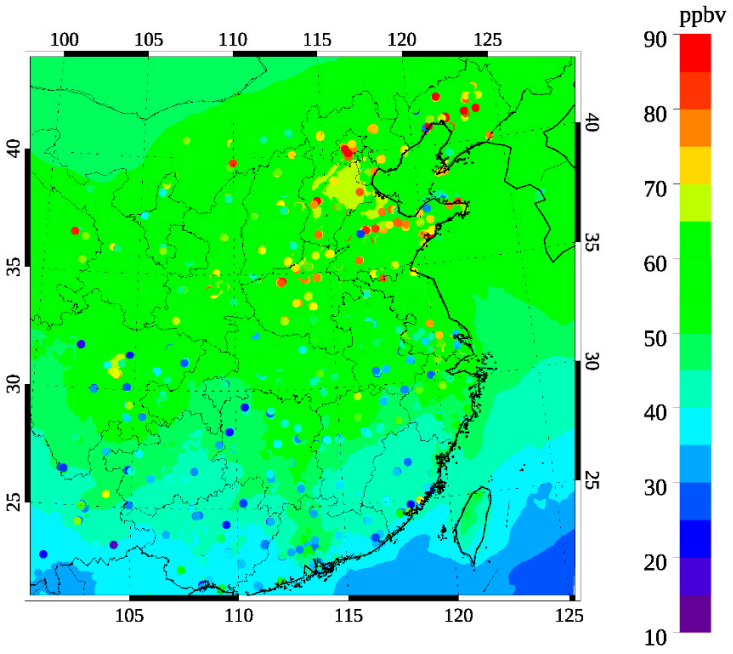


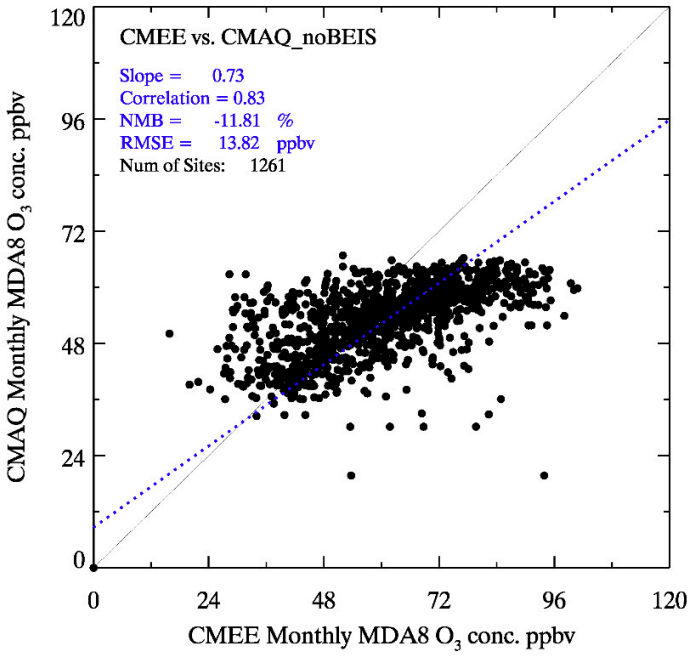


c)

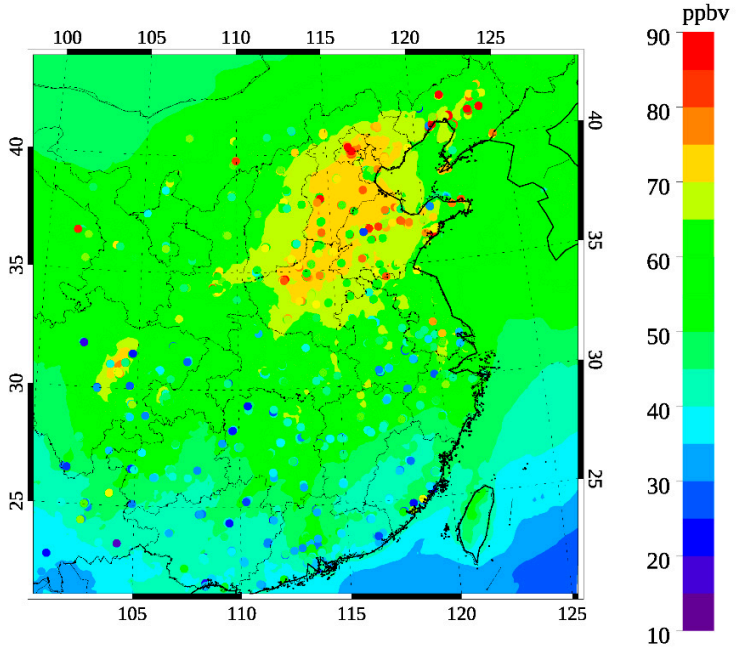
Figure S8. Similar as Figure 11, but compares CMEE observations and other CMAQ runs. a) CMAQ_noBEIS, b) CMAQ_CO, c) CMAQ_NOx, and d) CMAQ_VOC.

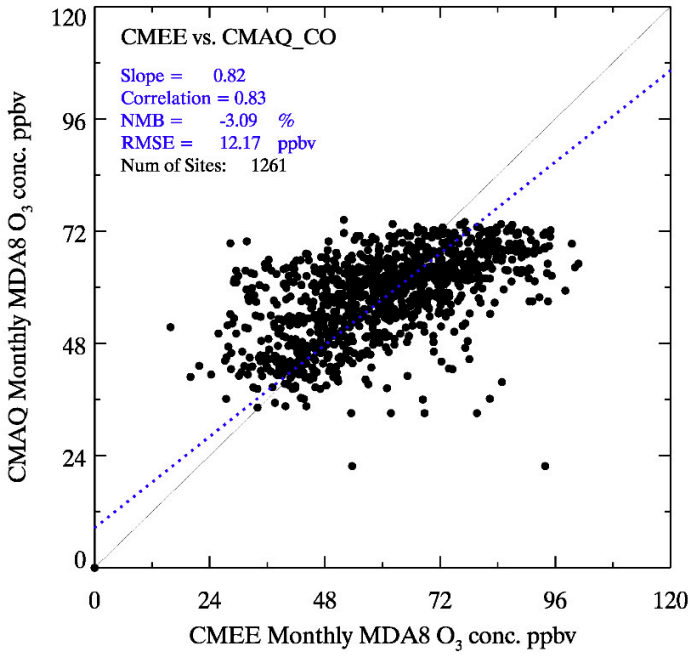
a)



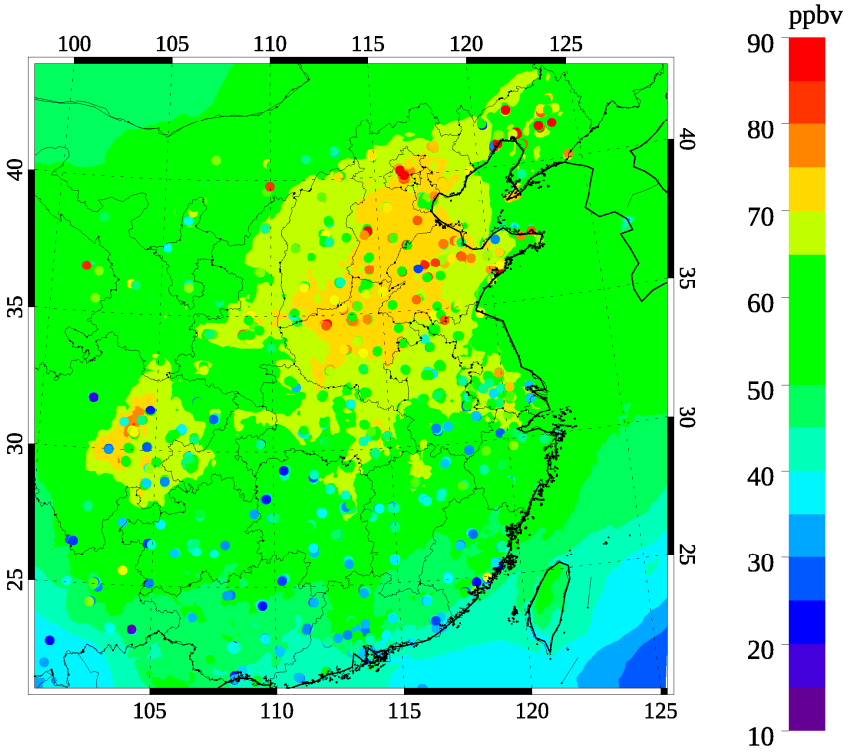

CMEE vs. CMAQ_noBEIS
Slope = 0.73
Correlation = 0.83
NMB = -11.81 %
RMSE = 13.82 ppbv
Num of Sites: 1261
CMAQ Monthly MDA8 O3 conc. ppbv
CMEE Monthly MDA8 O3 conc. ppbv
0
24
48
72
96
120

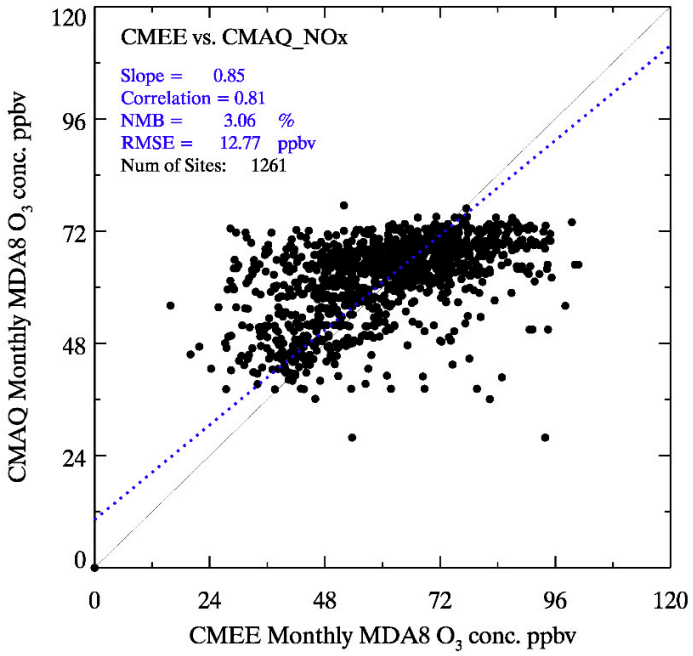
b)


ppbv
90
80
70
60
50
40
30
20
10
100
105
110
115
120
125
40
35
30
25

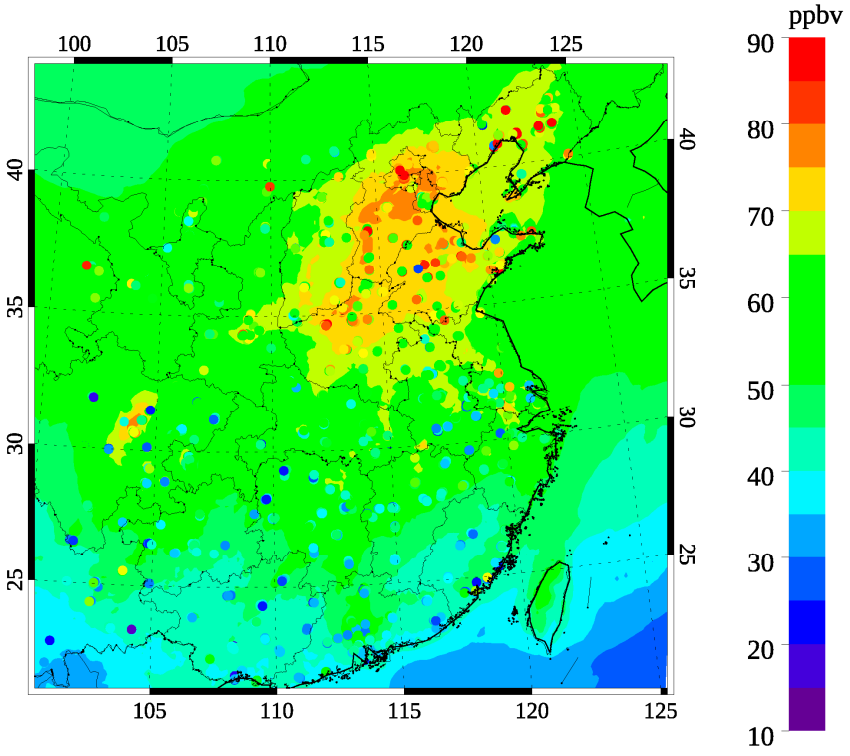

CMEE vs. CMAQ_CO
Slope = 0.82
Correlation = 0.83
NMB = -3.09 %
RMSE = 12.17 ppbv
Num of Sites: 1261
CMAQ Monthly MDA8 O3 conc. ppbv
CMEE Monthly MDA8 O3 conc. ppbv
0
24
48
72
96
120

c)


ppbv
90
80
70
60
50
40
30
20
10
100
105
110
115
120
125
40
35
30
25


CMEE vs. CMAQ_NOx
Slope = 0.85
Correlation = 0.81
NMB = 3.06 %
RMSE = 12.77 ppbv
Num of Sites: 1261
CMAQ Monthly MDA8 O3 conc. ppbv
CMEE Monthly MDA8 O3 conc. ppbv
0
24
48
72
96
120

d)


ppbv
90
80
70
60
50
40
30
20
10
100
105
110
115
120
125
40
35
30
25

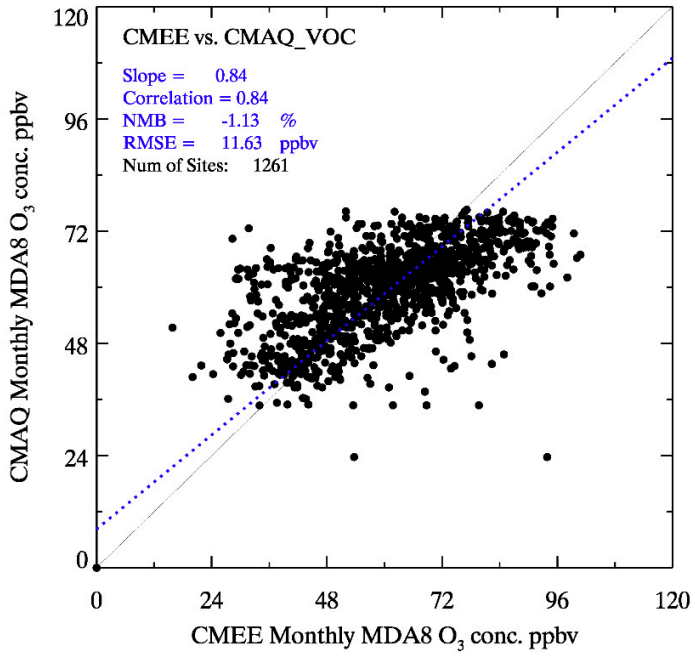
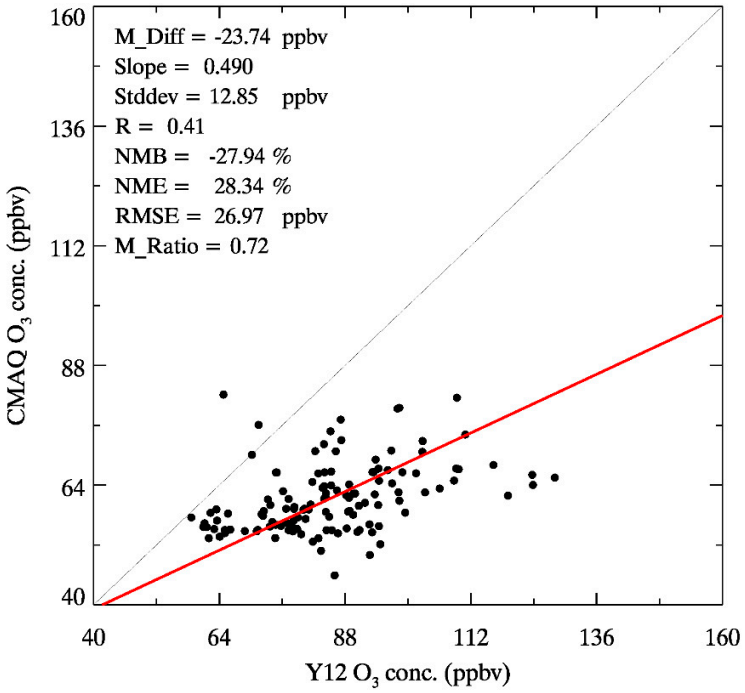
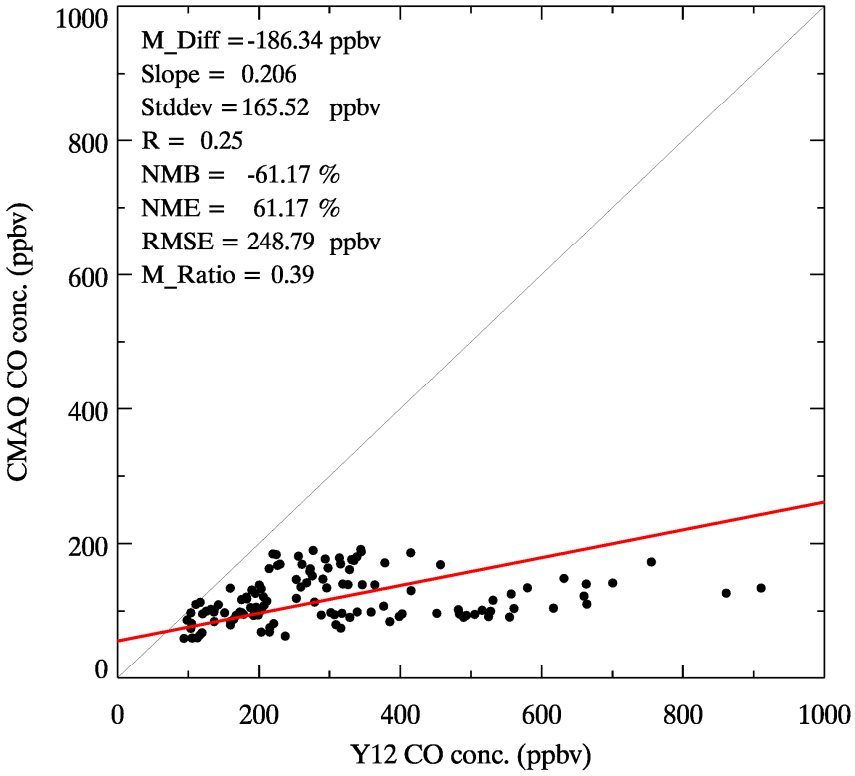
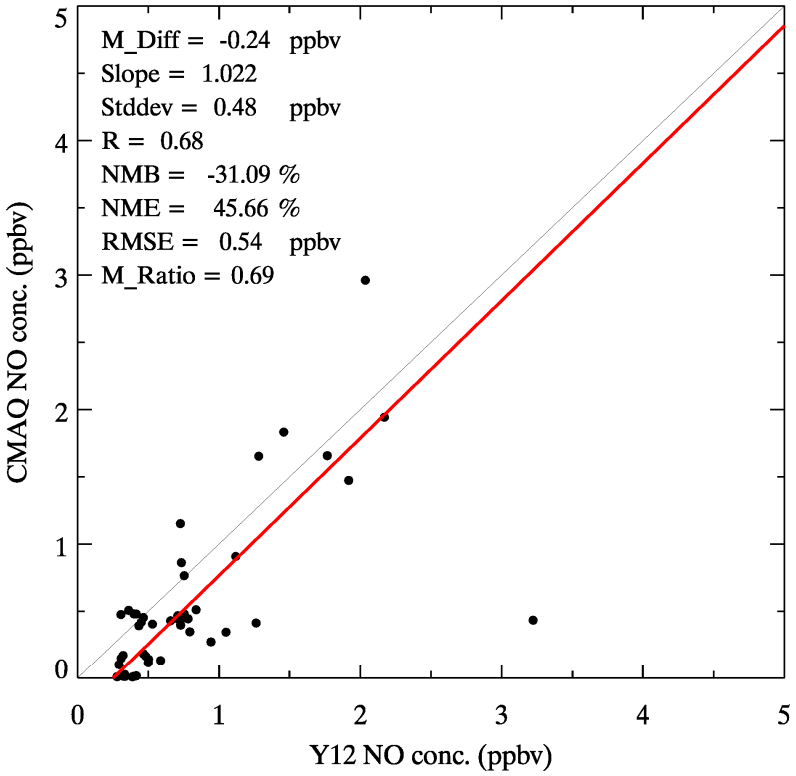


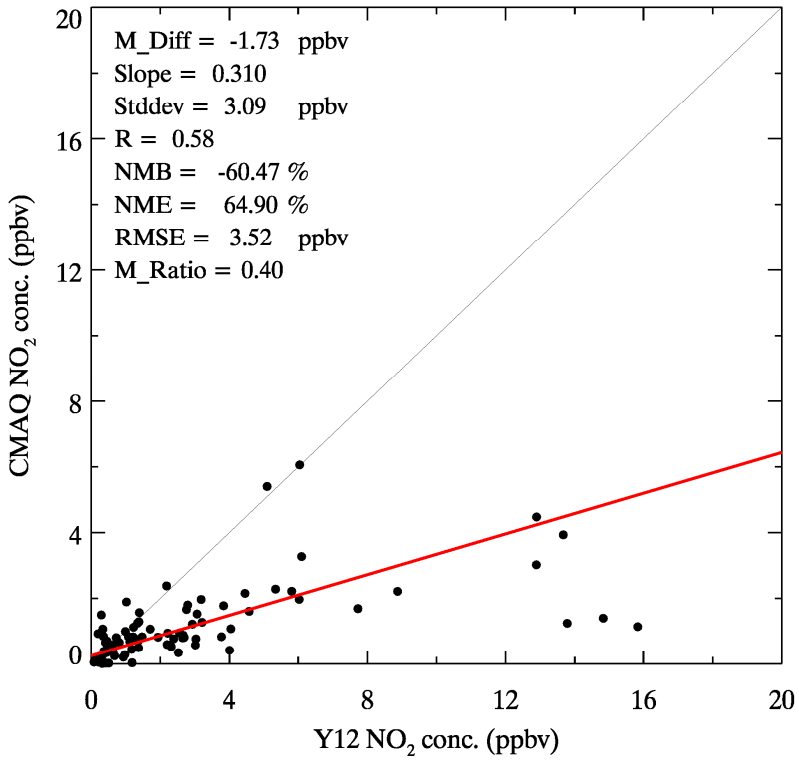
Figure S9. Similar as Figure 5, but shows results from other CMAQ sensitivity experiments in the campaign region.

a) CMAQ_noBEIS

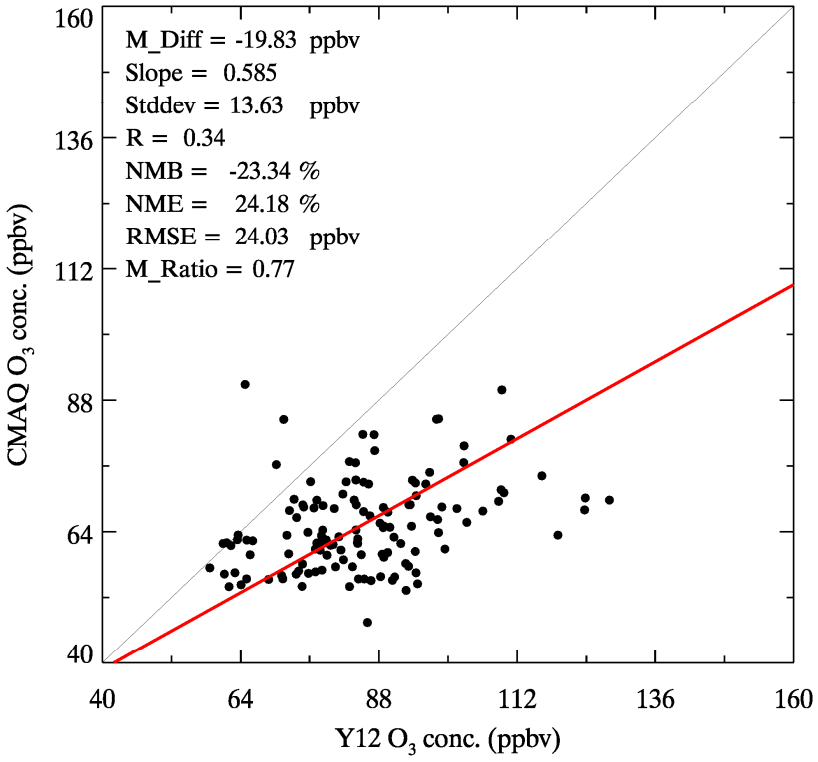


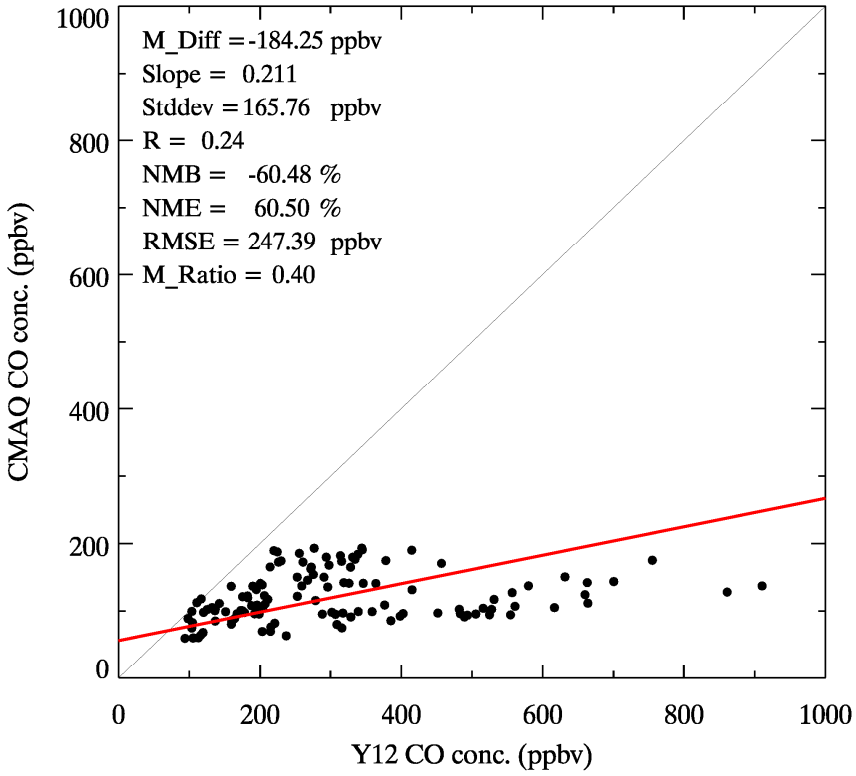

M_Diff = -186.34 ppbv
Slope = 0.206
Stddev = 165.52 ppbv
R = 0.25
NMB = -61.17 %
NME = 61.17 %
RMSE = 248.79 ppbv
M_Ratio = 0.39
CMAQ CO conc. (ppbv)
Y12 CO conc. (ppbv)
0
200
400
600
800
1000

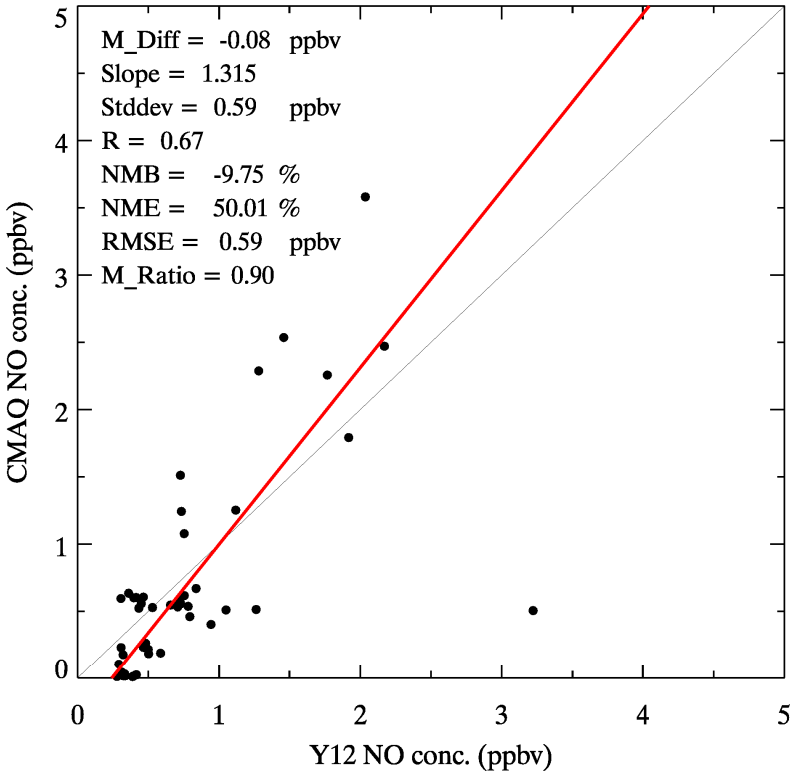

M_Diff = -0.24 ppbv
Slope = 1.022
Stddev = 0.48 ppbv
R = 0.68
NMB = -31.09 %
NME = 45.66 %
RMSE = 0.54 ppbv
M_Ratio = 0.69
CMAQ NO conc. (ppbv)
Y12 NO conc. (ppbv)
0
1
2
3
4
5

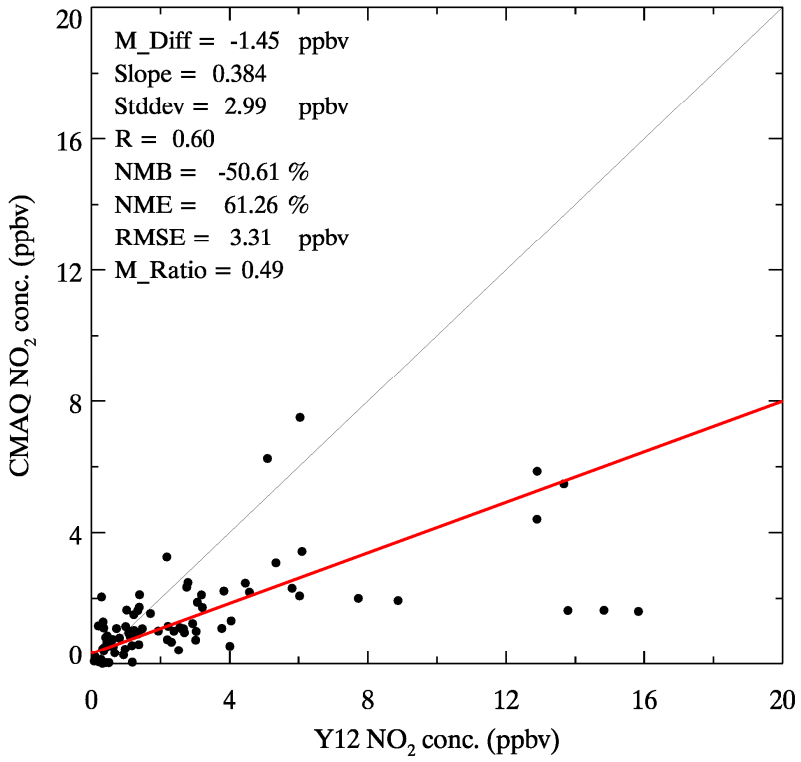

M_Diff = -1.73 ppbv
Slope = 0.310
Stddev = 3.09 ppbv
R = 0.58
NMB = -60.47 %
NME = 64.90 %
RMSE = 3.52 ppbv
M_Ratio = 0.40
CMAQ NO2 conc. (ppbv)
Y12 NO2 conc. (ppbv)
0
4
8
12
16
20

b) CMAQ_NOx

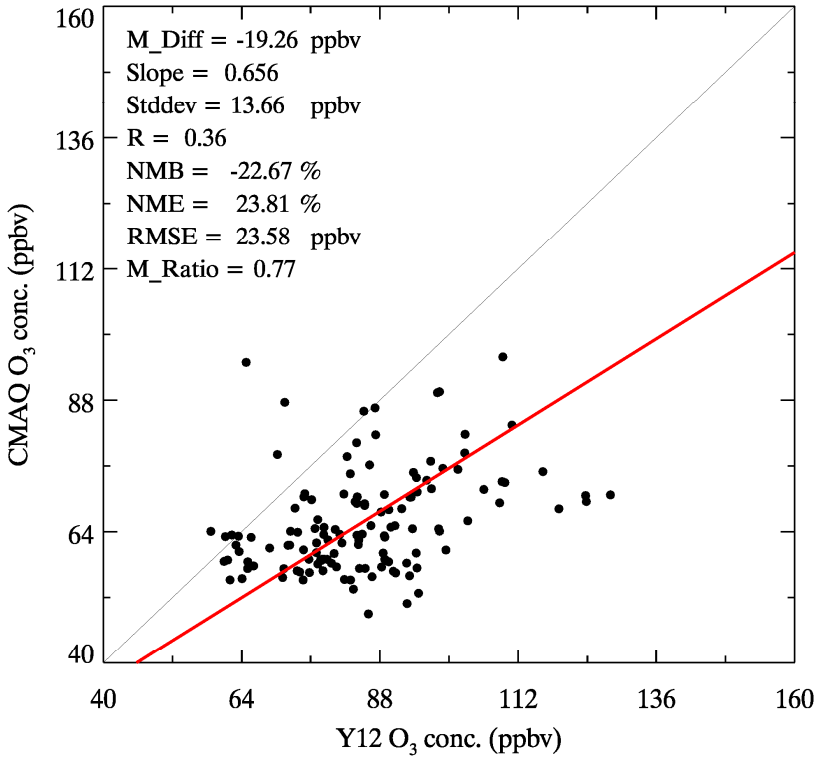

M_Diff = -19.83 ppbv
Slope = 0.585
Stddev = 13.63 ppbv
R = 0.34
NMB = -23.34 %
NME = 24.18 %
RMSE = 24.03 ppbv
M_Ratio = 0.77
CMAQ O3 conc. (ppbv)
Y12 O3 conc. (ppbv)
40
64
88
112
136
160

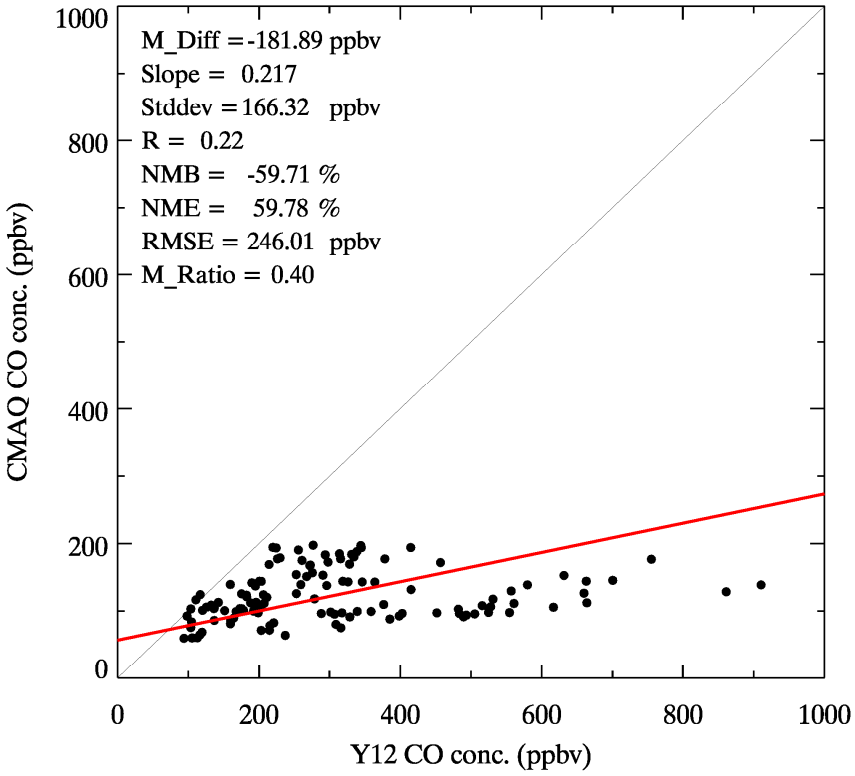

M_Diff = -184.25 ppbv
Slope = 0.211
Stddev = 165.76 ppbv
R = 0.24
NMB = -60.48 %
NME = 60.50 %
RMSE = 247.39 ppbv
M_Ratio = 0.40
CMAQ CO conc. (ppbv)
Y12 CO conc. (ppbv)
0
200
400
600
800
1000

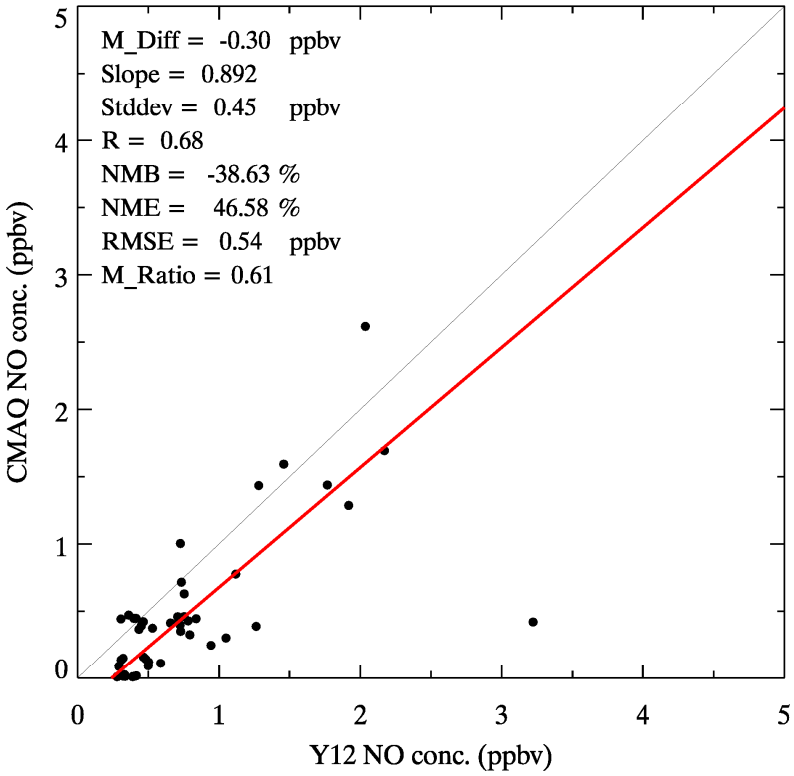

M_Diff = -0.08 ppbv
Slope = 1.315
Stddev = 0.59 ppbv
R = 0.67
NMB = -9.75 %
NME = 50.01 %
RMSE = 0.59 ppbv
M_Ratio = 0.90
CMAQ NO conc. (ppbv)
Y12 NO conc. (ppbv)
0
1
2
3
4
5

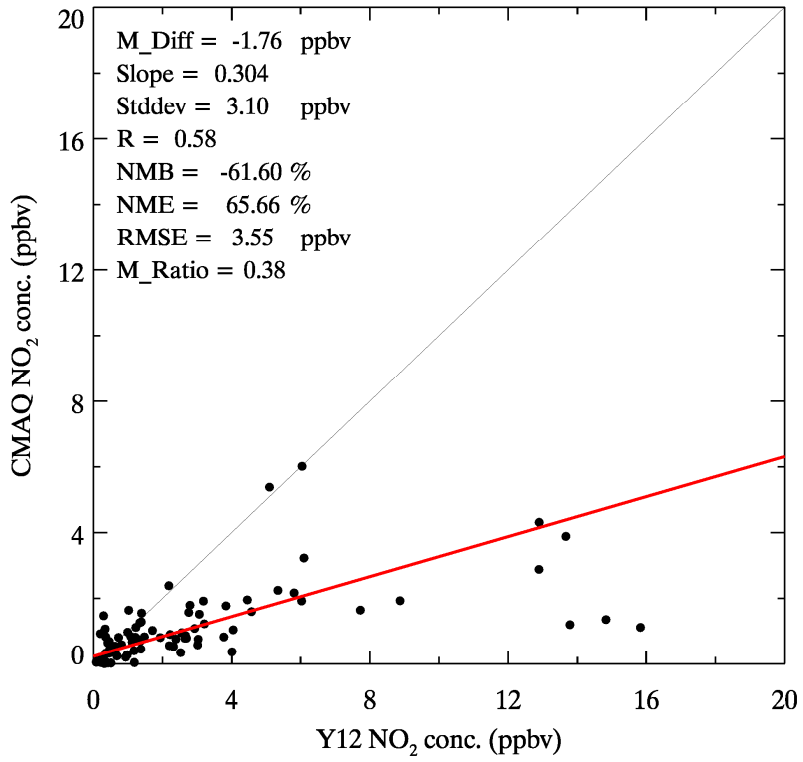

M_Diff = -1.45 ppbv
Slope = 0.384
Stddev = 2.99 ppbv
R = 0.60
NMB = -50.61 %
NME = 61.26 %
RMSE = 3.31 ppbv
M_Ratio = 0.49
CMAQ NO2 conc. (ppbv)
Y12 NO2 conc. (ppbv)
0
4
8
12
16
20

c) CMAQ_VOCs

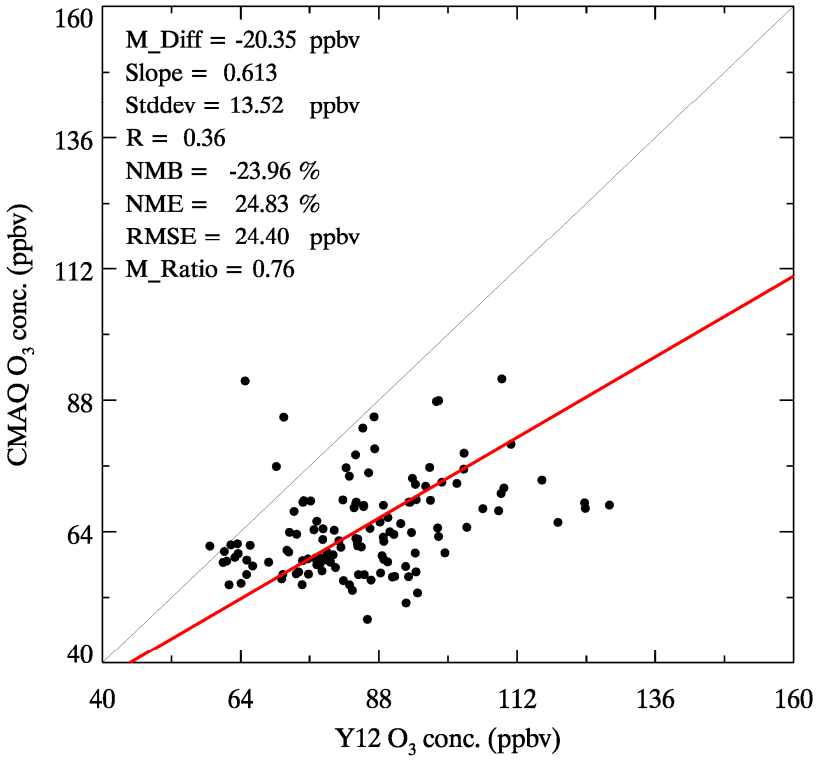

M_Diff = -19.26 ppbv
Slope = 0.656
Stddev = 13.66 ppbv
R = 0.36
NMB = -22.67 %
NME = 23.81 %
RMSE = 23.58 ppbv
M_Ratio = 0.77
CMAQ O3 conc. (ppbv)
Y12 O3 conc. (ppbv)
40
64
88
112
136
160

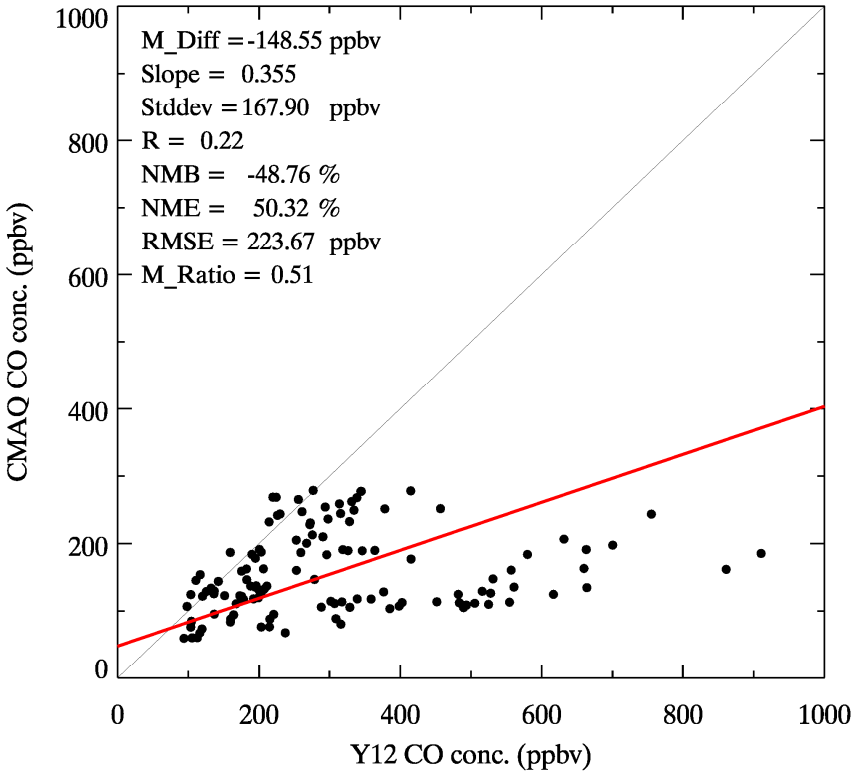

M_Diff = -181.89 ppbv
Slope = 0.217
Stddev = 166.32 ppbv
R = 0.22
NMB = -59.71 %
NME = 59.78 %
RMSE = 246.01 ppbv
M_Ratio = 0.40
CMAQ CO conc. (ppbv)
Y12 CO conc. (ppbv)
0
200
400
600
800
1000

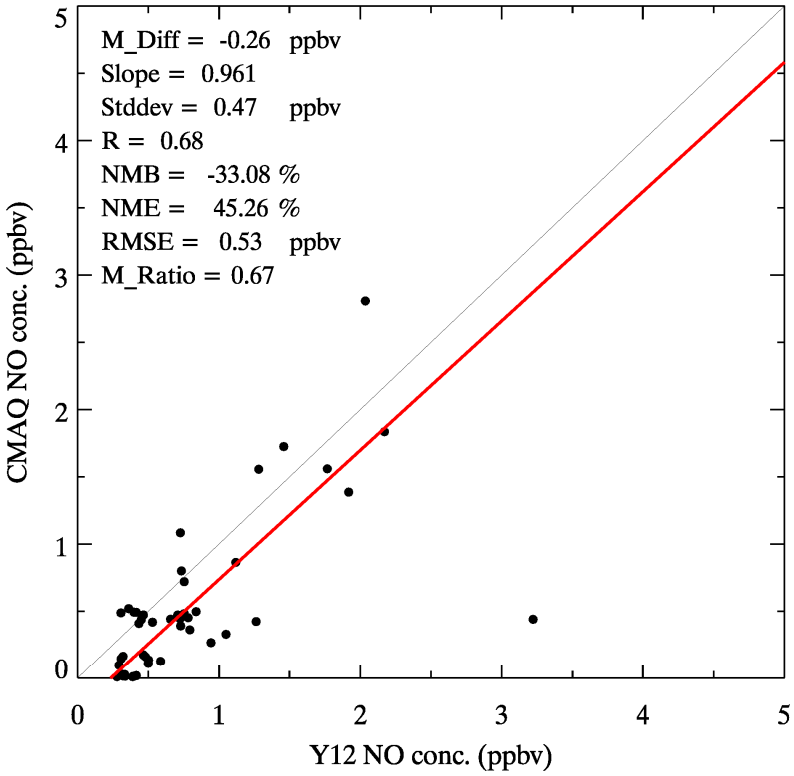

M_Diff = -0.30 ppbv
Slope = 0.892
Stddev = 0.45 ppbv
R = 0.68
NMB = -38.63 %
NME = 46.58 %
RMSE = 0.54 ppbv
M_Ratio = 0.61
CMAQ NO conc. (ppbv)
Y12 NO conc. (ppbv)
0
1
2
3
4
5

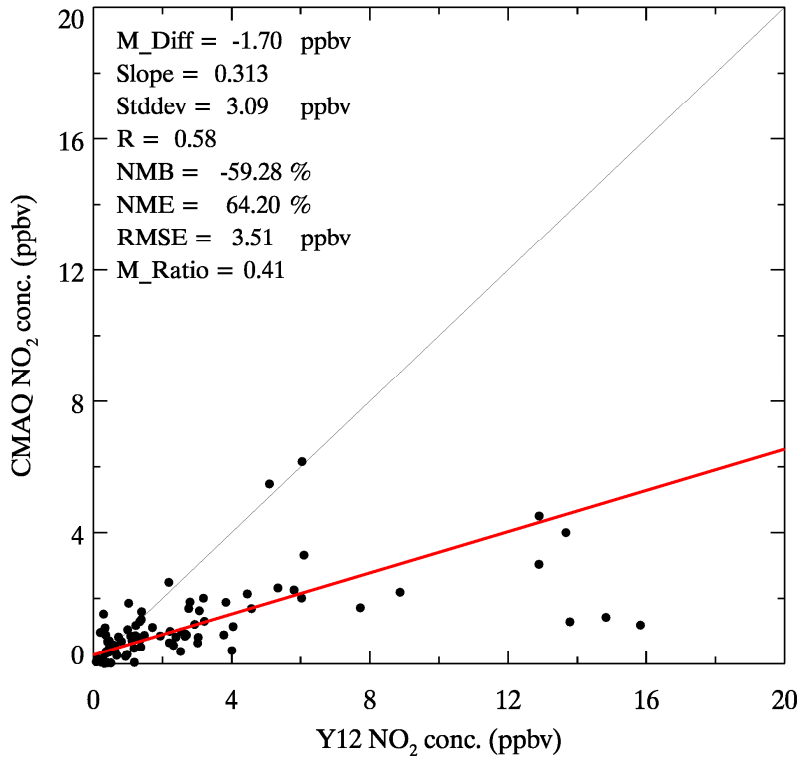

M_Diff = -1.76 ppbv
Slope = 0.304
Stddev = 3.10 ppbv
R = 0.58
NMB = -61.60 %
NME = 65.66 %
RMSE = 3.55 ppbv
M_Ratio = 0.38
CMAQ NO2 conc. (ppbv)
Y12 NO2 conc. (ppbv)
0
4
8
12
16
20

d) CMAQ_CO

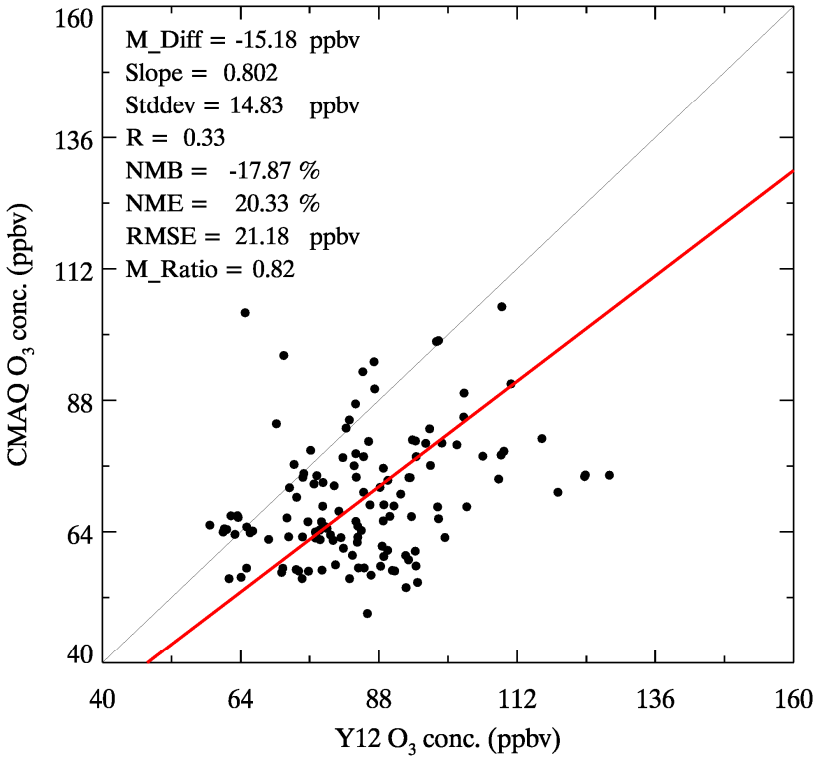

M_Diff = -20.35 ppbv
Slope = 0.613
Stddev = 13.52 ppbv
R = 0.36
NMB = -23.96 %
NME = 24.83 %
RMSE = 24.40 ppbv
M_Ratio = 0.76
CMAQ O3 conc. (ppbv)
Y12 O3 conc. (ppbv)
40
64
88
112
136
160

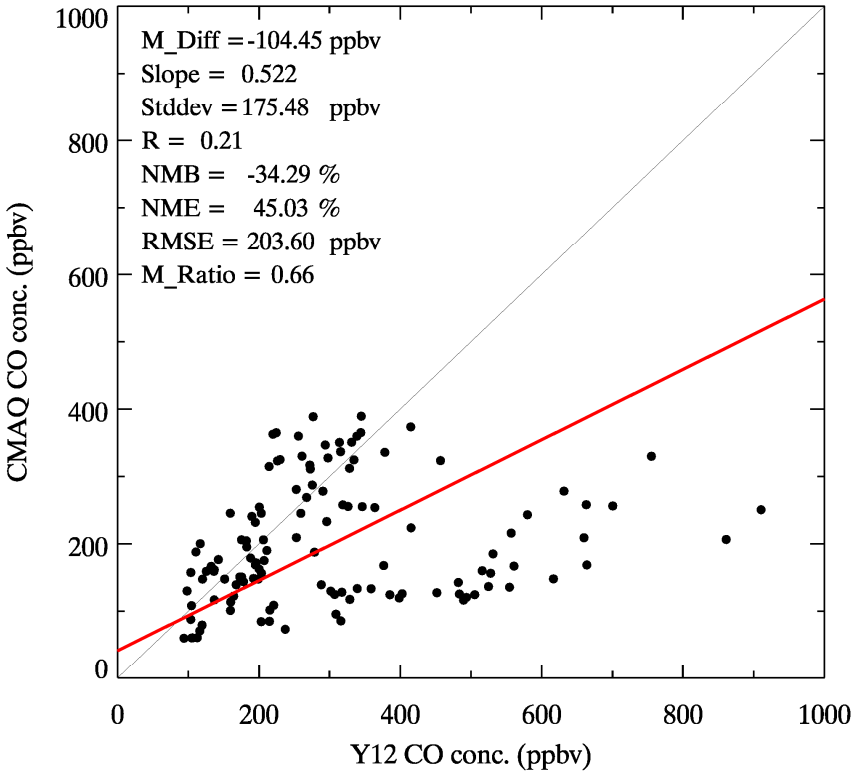

M_Diff =-148.55 ppbv
Slope = 0.355
Stddev =167.90 ppbv
R = 0.22
NMB = -48.76 %
NME = 50.32 %
RMSE = 223.67 ppbv
M_Ratio = 0.51
CMAQ CO conc. (ppbv)
Y12 CO conc. (ppbv)
0
200
400
600
800
1000

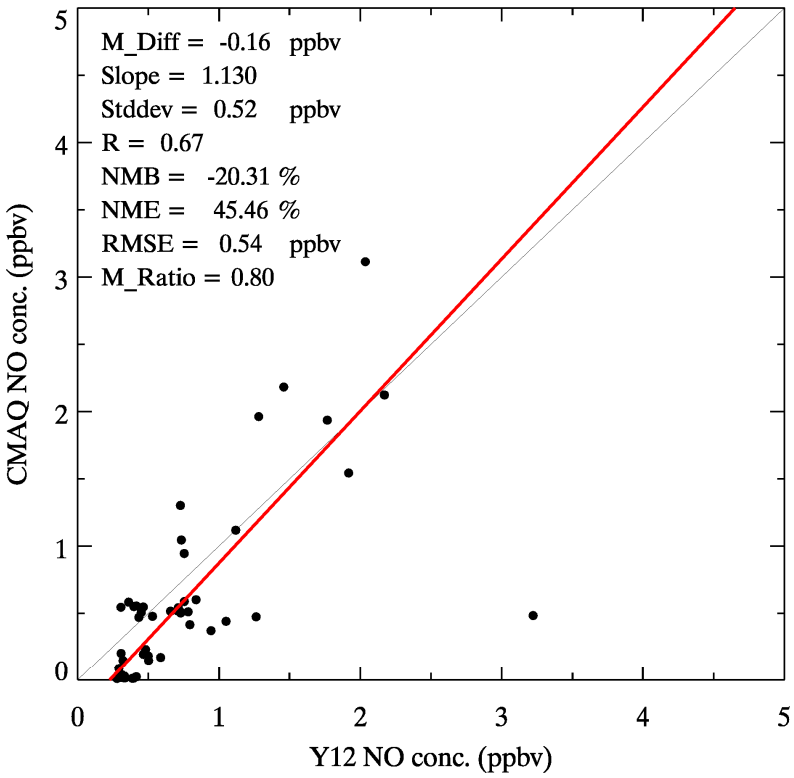

M_Diff = -0.26 ppbv
Slope = 0.961
Stddev = 0.47 ppbv
R = 0.68
NMB = -33.08 %
NME = 45.26 %
RMSE = 0.53 ppbv
M_Ratio = 0.67
CMAQ NO conc. (ppbv)
Y12 NO conc. (ppbv)
0
1
2
3
4
5

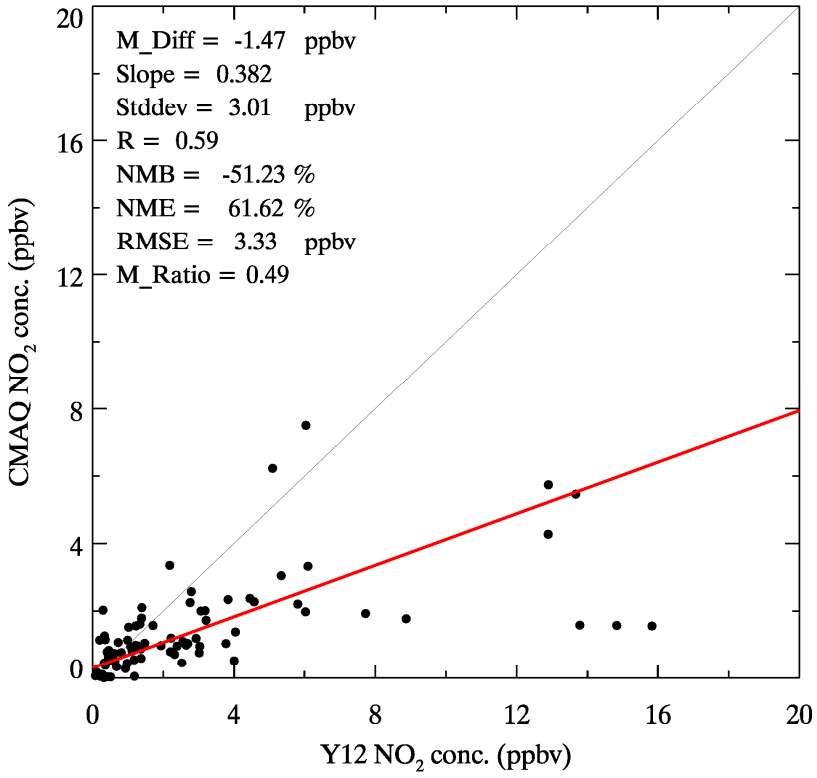

M_Diff = -1.70 ppbv
Slope = 0.313
Stddev = 3.09 ppbv
R = 0.58
NMB = -59.28 %
NME = 64.20 %
RMSE = 3.51 ppbv
M_Ratio = 0.41
CMAQ NO2 conc. (ppbv)
Y12 NO2 conc. (ppbv)
0
4
8
12
16
20

e) CMAQ_all


M_Diff = -15.18 ppbv
Slope = 0.802
Stddev = 14.83 ppbv
R = 0.33
NMB = -17.87 %
NME = 20.33 %
RMSE = 21.18 ppbv
M_Ratio = 0.82
CMAQ O3 conc. (ppbv)
Y12 O3 conc. (ppbv)
40
64
88
112
136
160


M_Diff =-104.45 ppbv
Slope = 0.522
Stddev =175.48 ppbv
R = 0.21
NMB = -34.29 %
NME = 45.03 %
RMSE = 203.60 ppbv
M_Ratio = 0.66
CMAQ CO conc. (ppbv)
Y12 CO conc. (ppbv)
0
200
400
600
800
1000


M_Diff = -0.16 ppbv
Slope = 1.130
Stddev = 0.52 ppbv
R = 0.67
NMB = -20.31 %
NME = 45.46 %
RMSE = 0.54 ppbv
M_Ratio = 0.80
CMAQ NO conc. (ppbv)
Y12 NO conc. (ppbv)
0
1
2
3
4
5


M_Diff = -1.47 ppbv
Slope = 0.382
Stddev = 3.01 ppbv
R = 0.59
NMB = -51.23 %
NME = 61.62 %
RMSE = 3.33 ppbv
M_Ratio = 0.49
CMAQ NO2 conc. (ppbv)
Y12 NO2 conc. (ppbv)
0
4
8
12
16
20