

# Supporting information

## 1 Regional division

The analysis in this study was conducted for seven administrative regions: North China (i.e., Beijing, Tianjin, Hebei, Shanxi, and Inner Mongolia), North-east China (i.e., Liaoning, Jilin, and Heilongjiang), East China (i.e., Shanghai, Jiangsu, Zhejiang, Anhui, Fujian, Jiangxi, and Shandong), Central China (i.e., Henan, Hubei, and Hunan), South China (i.e., Guangdong, Guangxi, and Hainan), South-west China (i.e., Chongqing, Sichuan, Guizhou, Yunnan, and Xizang), and North-west China (i.e., Shaanxi, Gansu, Qinghai, Ningxia, and Xinjiang).

## 2 Validation of meteorological simulation

Meteorological parameters simulated by WRFv3.8 were compared with observation data downloaded from the National Climatic Data Center (NCDC). The variables included the temperature at 2 m, wind speed at 10 m, and humidity at 2 m. The statistical indices used include the mean observation (Mean OBS), mean simulation (Mean SIM), mean bias (MB), gross error (GE), root mean square error (RMSE), and index of agreement (IOA). The model shows good agreement with observations and captures the trend of meteorological parameters between the two years. The average temperature of observations in North, North-east, East, South, North-west, and South-west China showed an increase by 1.24°C, 0.81°C, 0.41°C, 0.65°C, 0.26°C, and 0.04°C, while the temperature in Central China showed a decrease by 0.29°C. The simulation showed temperature increases by 0.98°C, 0.90°C, 0.32°C, and 0.44°C in North, North-east, East, and South China, and temperature decreases by 0.24°C, 0.13°C, and 0.09°C in Central, North-west, and South-west China. The increments were slightly underestimated; the inconsistent results for North-west and South-west China were small and within the systematic bias.

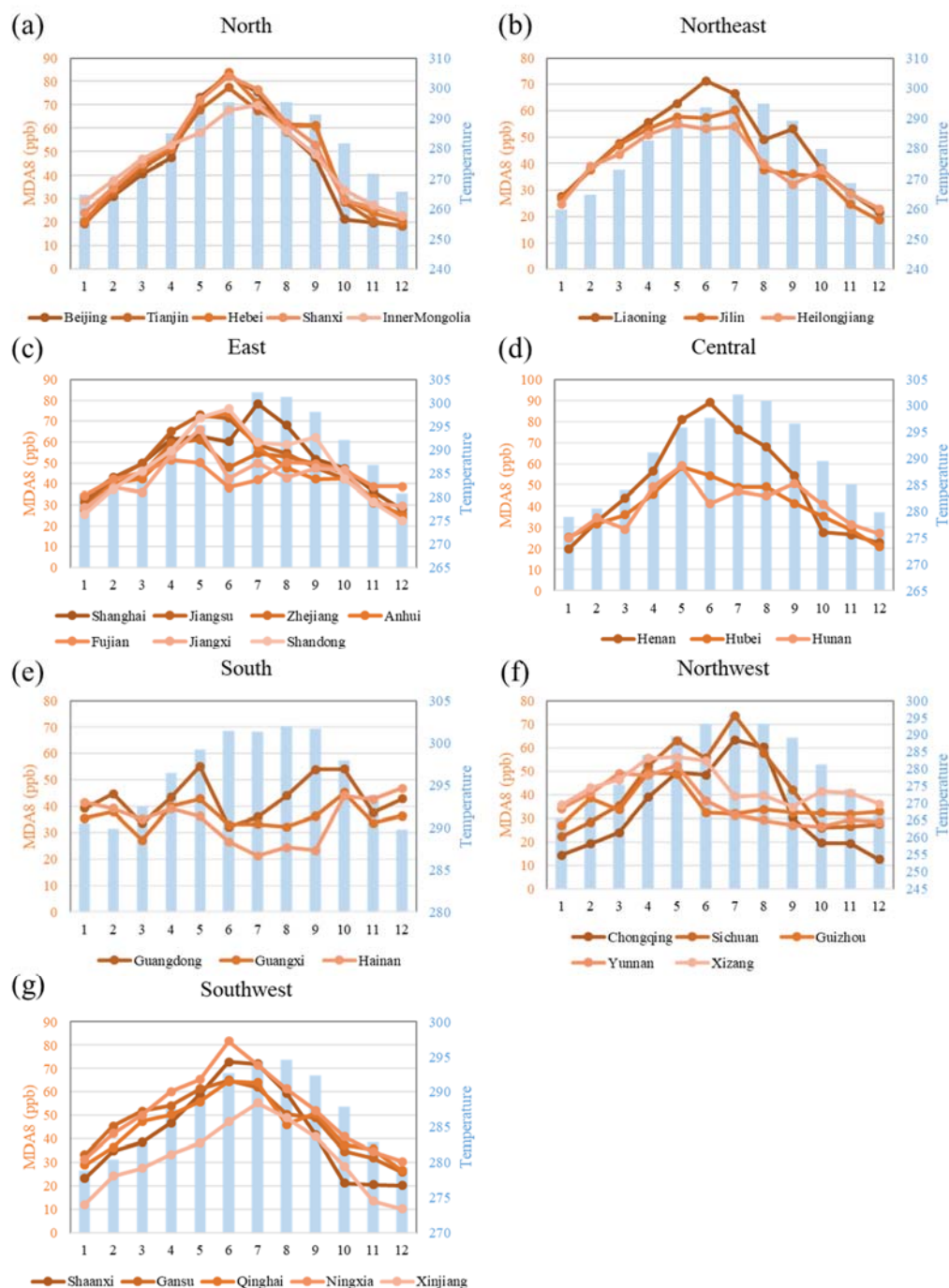


Fig. S1 Monthly variation of observed AvgMDA8 O<sub>3</sub> (ppb) and temperature (K) of each region in 2017: (a) North China; (b) Northeast China; (c) East China; (d) Central China; (e) South China; (f) Northwest China; (g) Southwest China

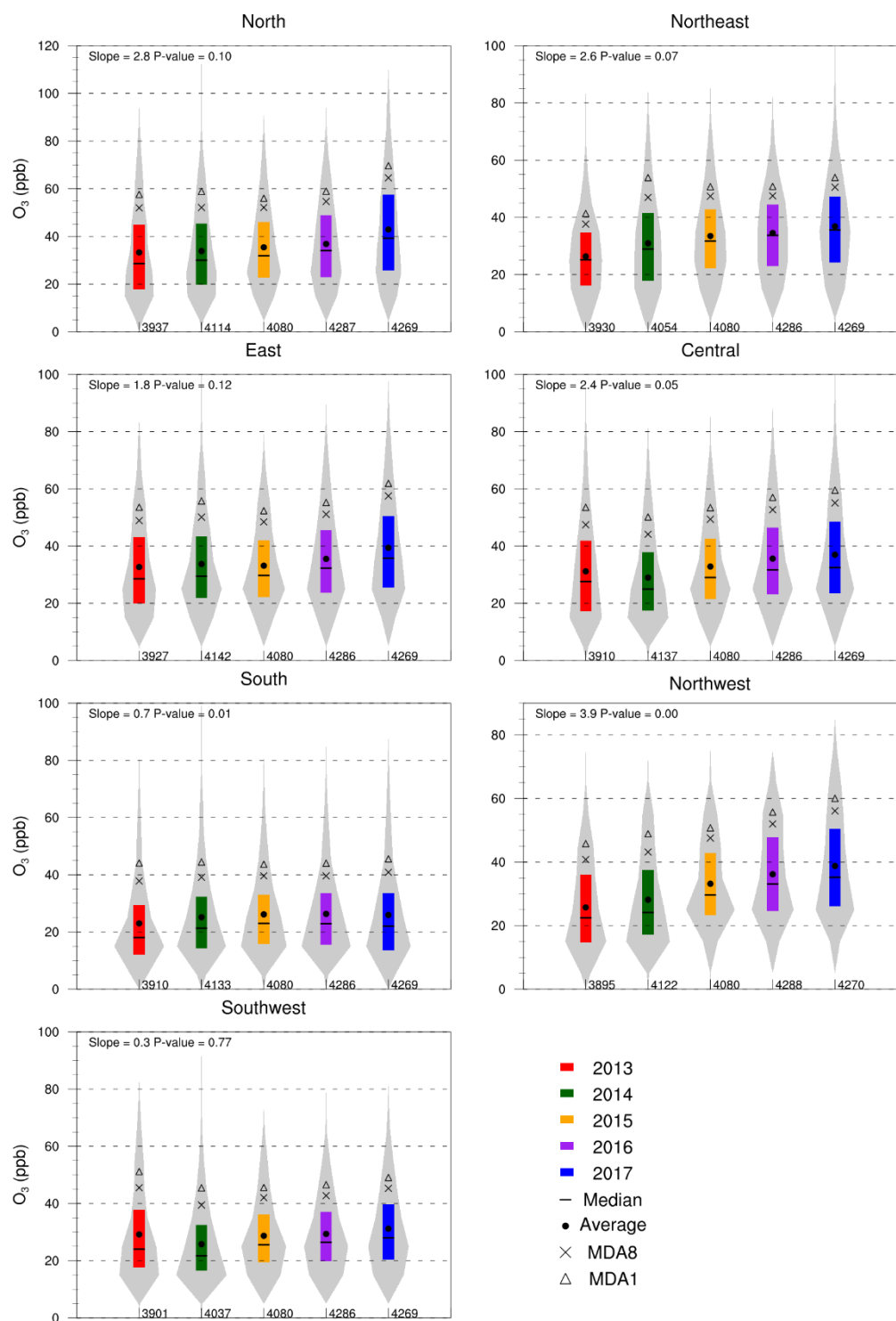


Fig. S2 Inter-annual changes of O<sub>3</sub> concentration in the warm season during 2013–2017

(Grey shading represents the concentration distribution of O<sub>3</sub> observation over all valid hourly data, values on the x-axis represent the numbers of valid hourly data, color boxes show the range of the 25th and 75th percentile values, the solid lines show the median value, the filled dots show the average value, the cross symbols show the AvgMDA8 O<sub>3</sub>, and the hollow triangles show the AvgMDA1 O<sub>3</sub>)

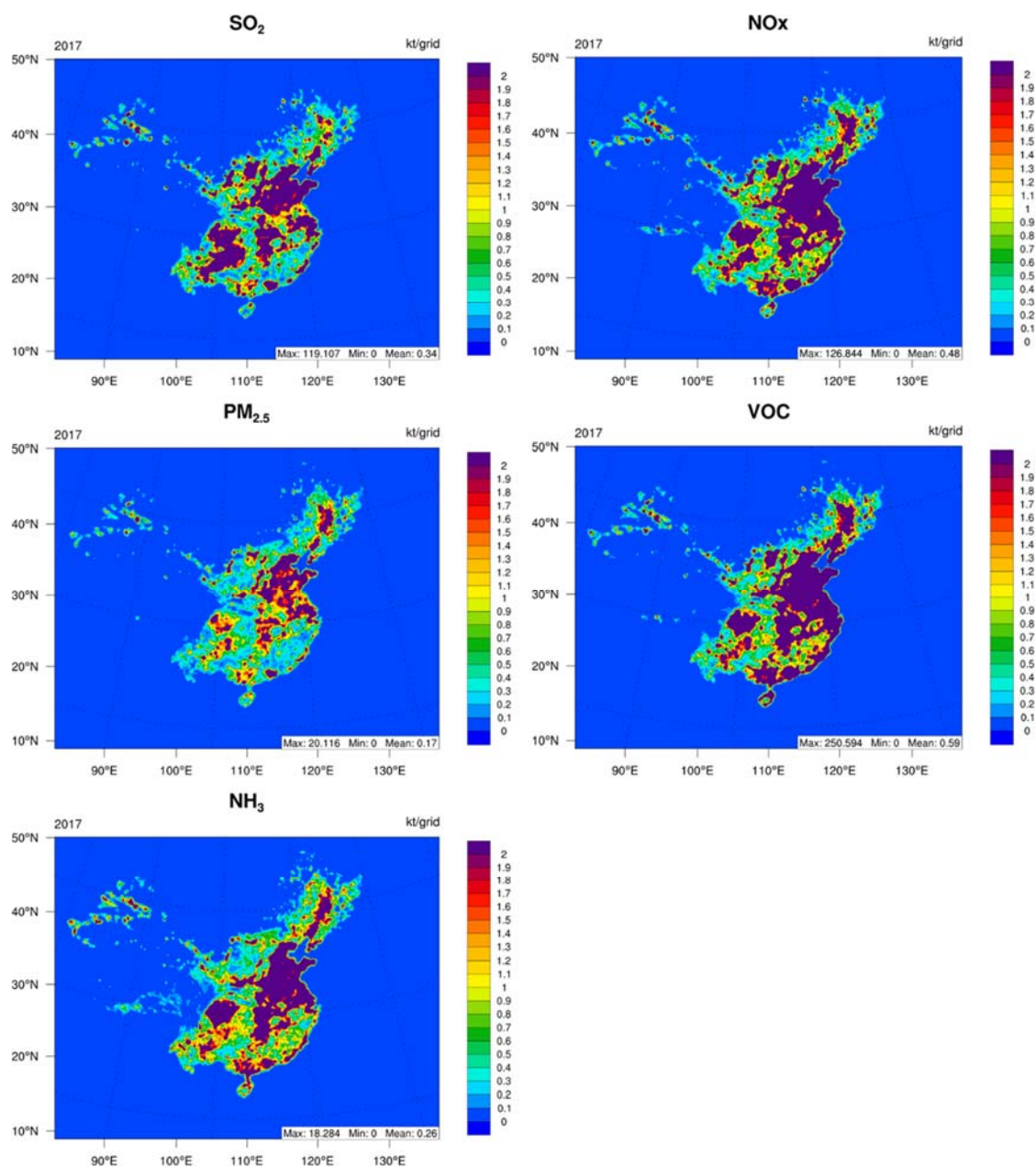


Fig. S3 Spatial distribution of the annual anthropogenic emissions in 2017

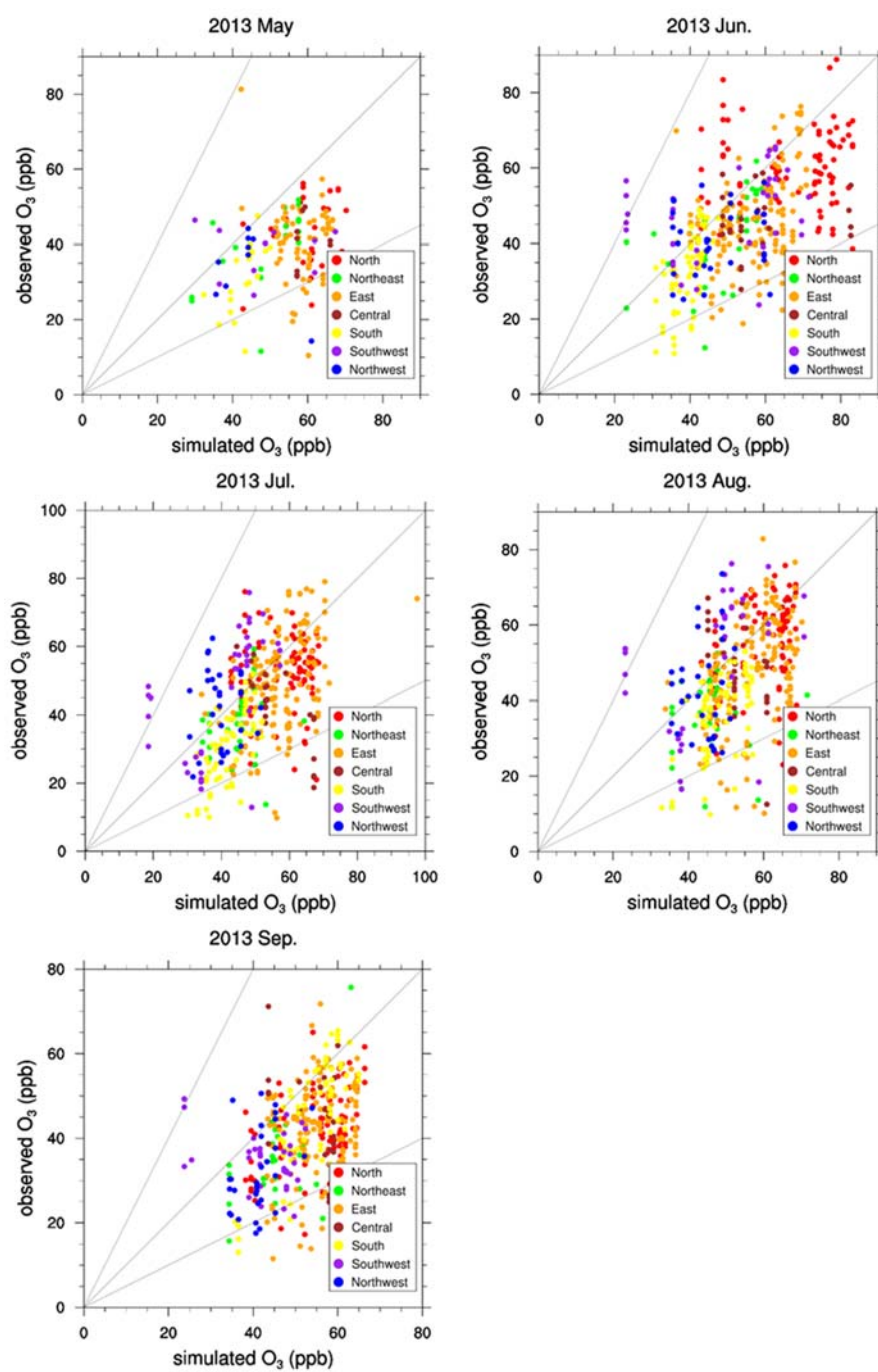


Fig. S4 The CMAQ performance in simulating AvgMDA8  $O_3$  in 2013. The 1:1, 2:1, and 1:2 lines are shown for reference

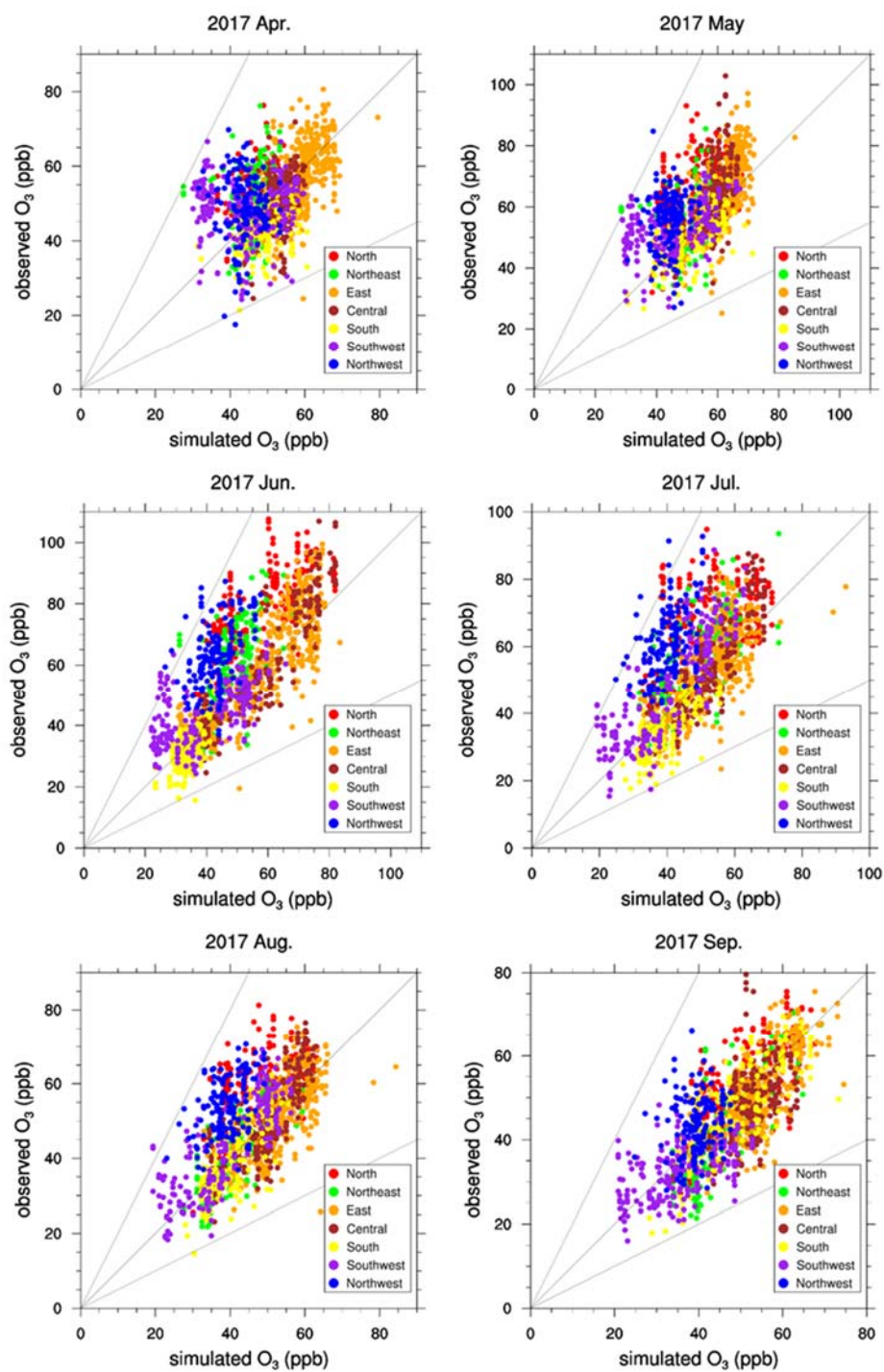


Fig. S5 The CMAQ performance in simulating AvgMDA8 O<sub>3</sub> in 2017. The 1:1, 2:1, and 1:2 lines are shown for reference



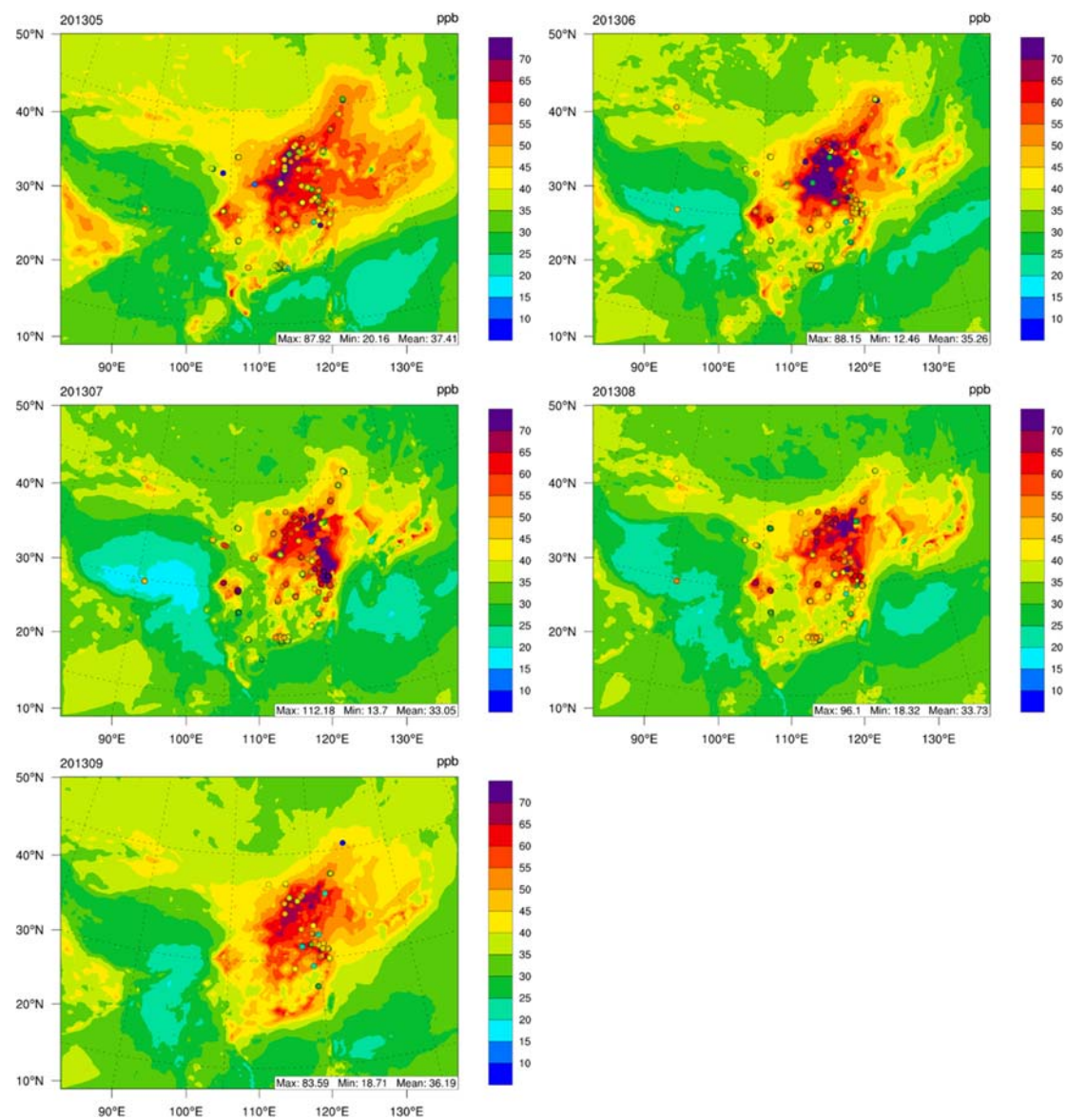


Fig. S6 Spatial distribution of the simulated AvgMDA8 O<sub>3</sub> in 2013 (the background colors represent the simulation and the filled dots represent the observations)

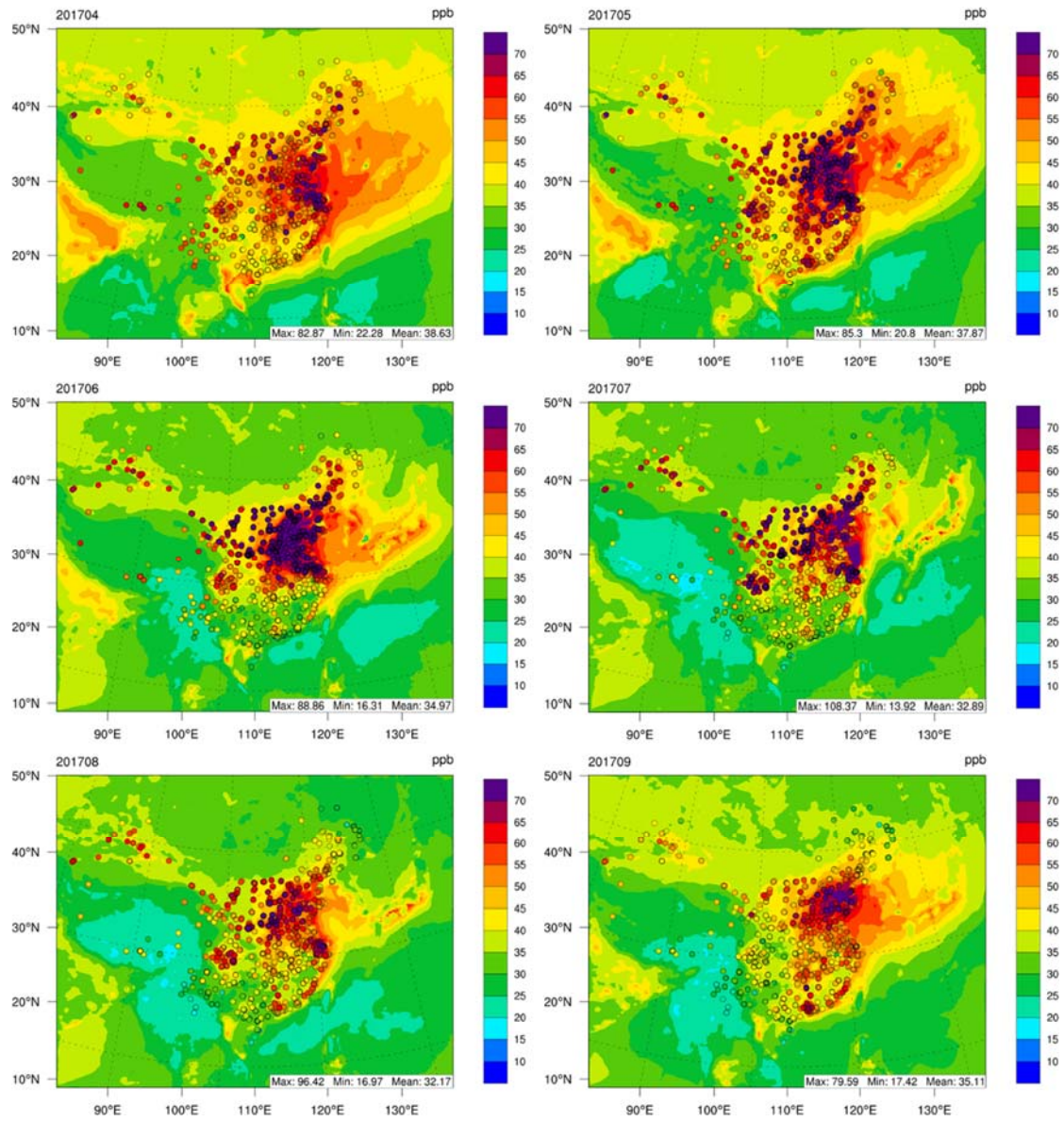


Fig. S7 Spatial distribution of the simulated AvgMDA8 O<sub>3</sub> in 2017 (the background colors represent the simulation and filled dots represent the observations)



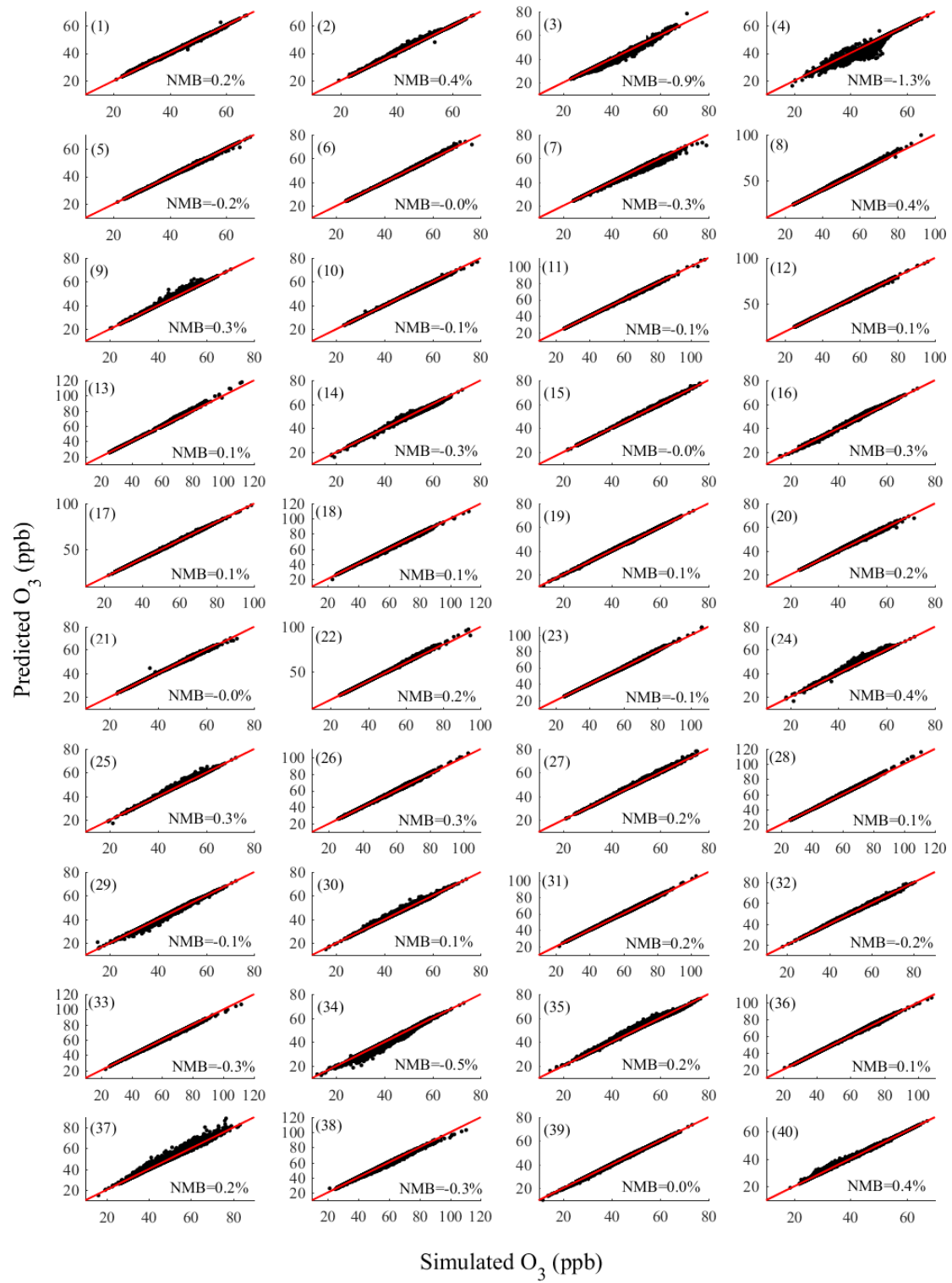


Fig. S8 Leave-one-out comparison of  $O_3$  between pf-RSM prediction and CMAQ simulations in April 2017 (The red line is the one-to-one line that would indicate perfect agreement)

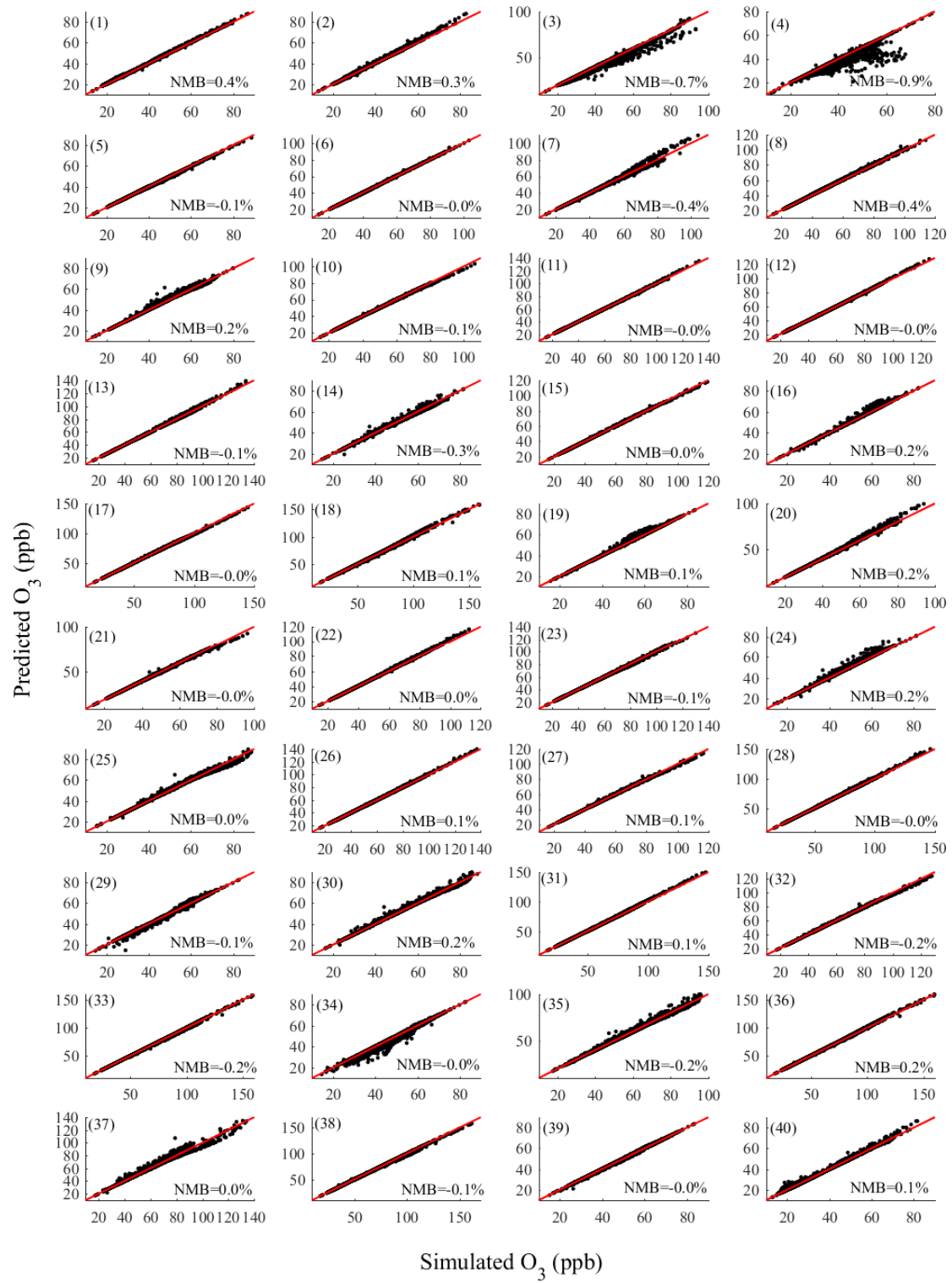


Fig. S9 Leave-one-out comparison of  $O_3$  between pf-RSM prediction and CMAQ simulations in July 2017 (The red line is the one-to-one line that would indicate perfect agreement)

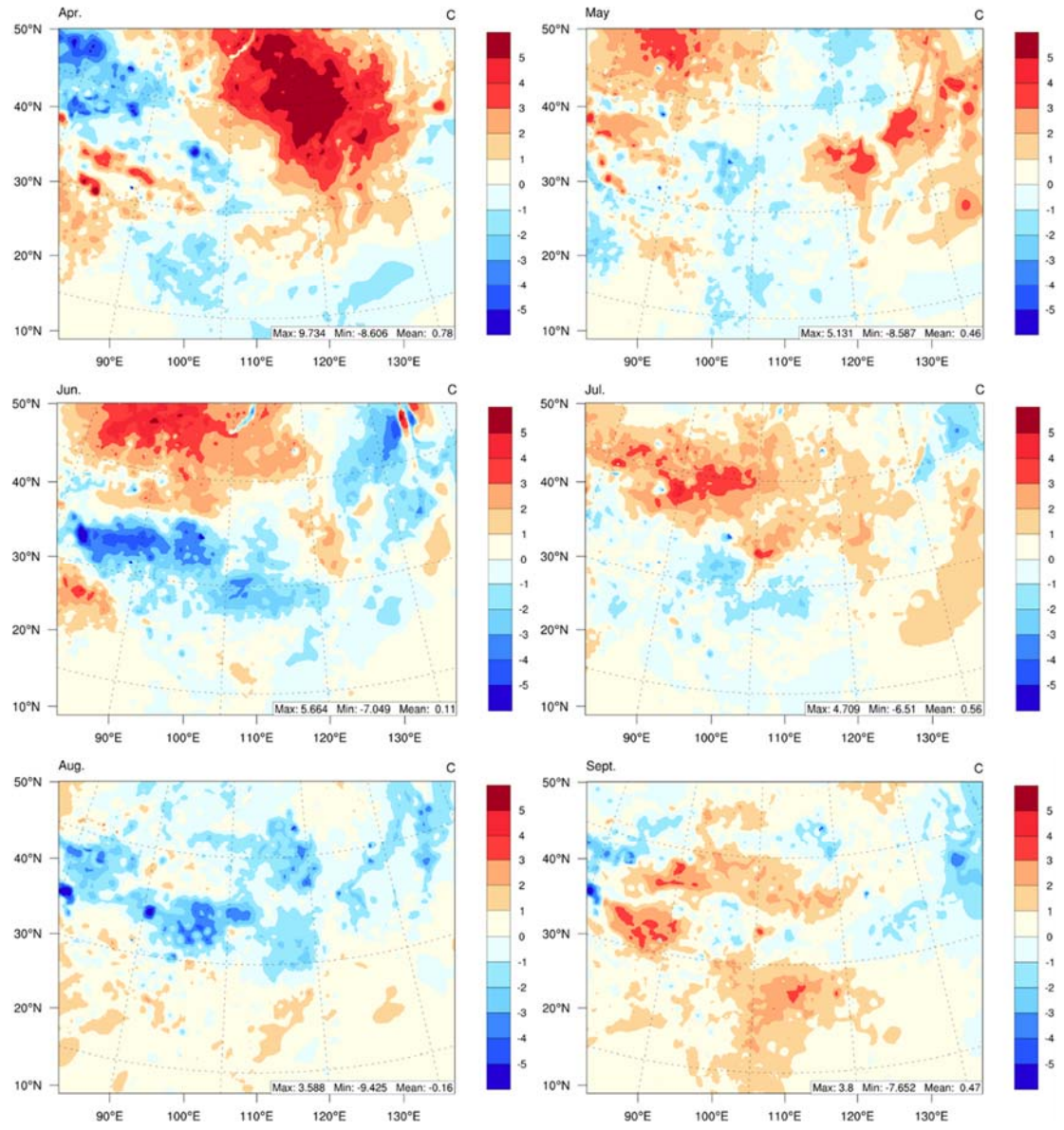


Fig. S10 The difference in temperature (°C) between 2013 and 2017 ( $= 2017 - 2013$ ) during April to September



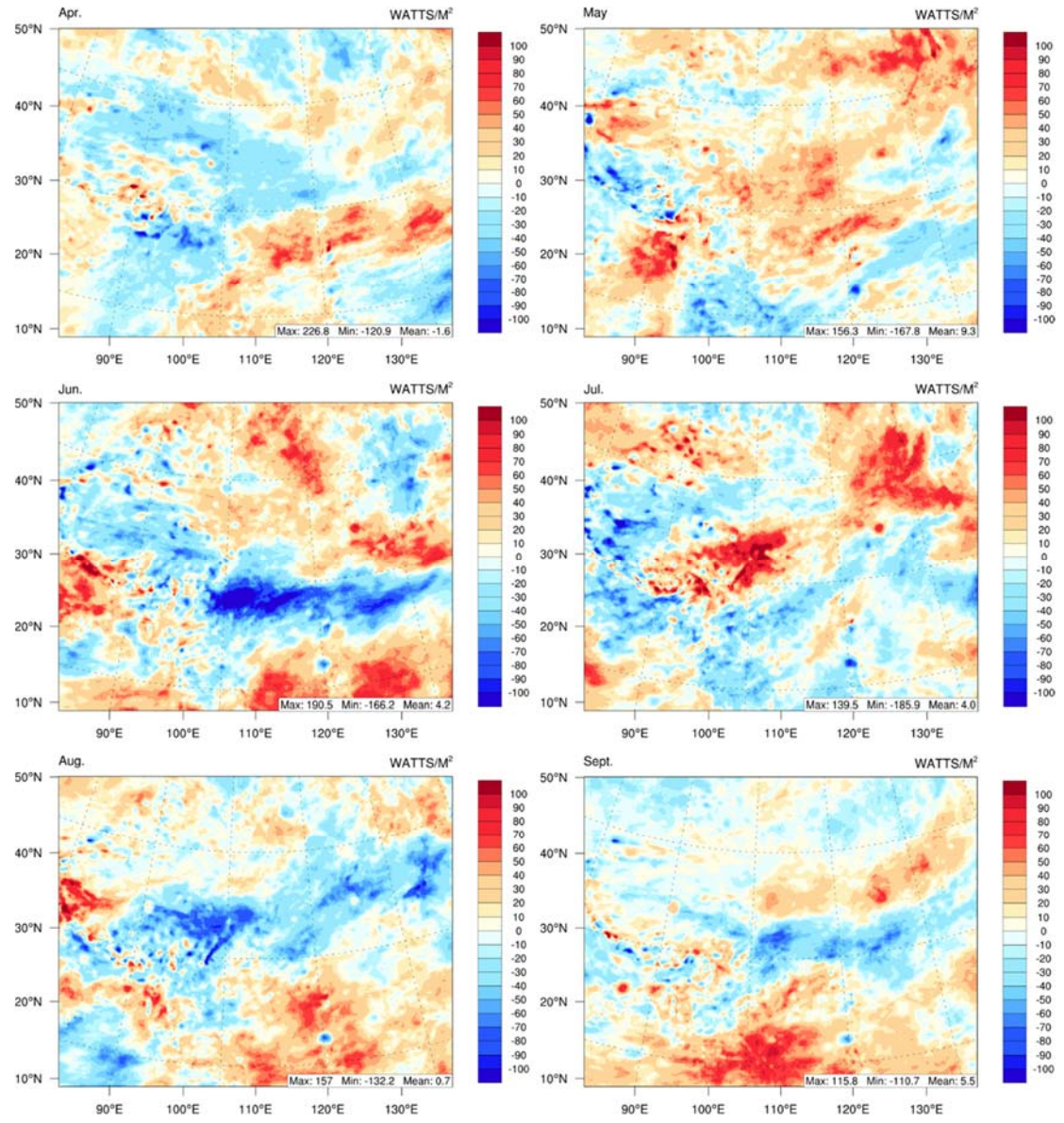


Fig. S11 The difference in RGRND ( $w/m^2$ ) between 2013 and 2017 ( $= 2017-2013$ ) during April to September



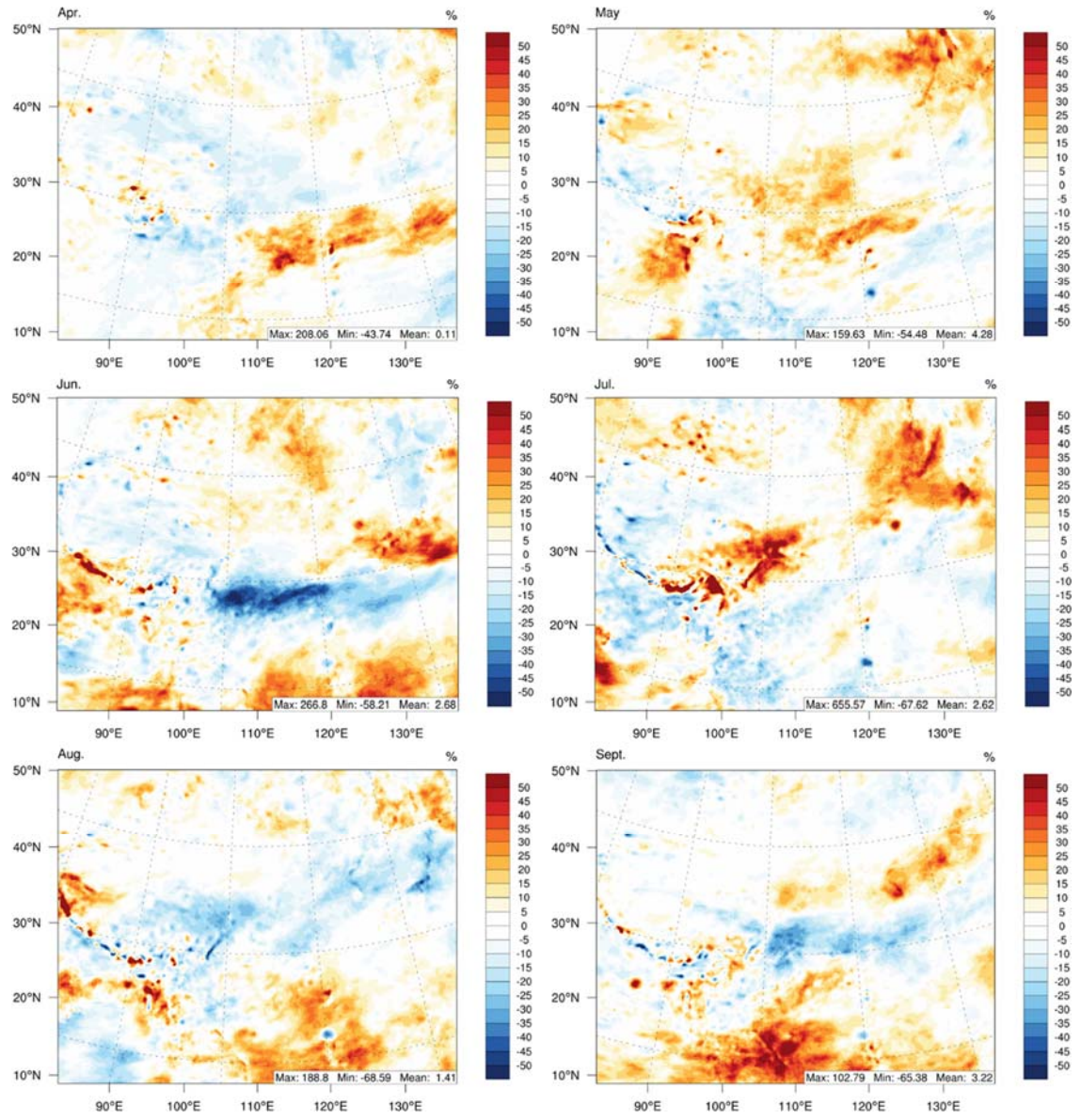


Fig. S12 The relative change (%) in RGRND between 2013 and 2017 ( $= 2017 - 2013$ ) during April to September

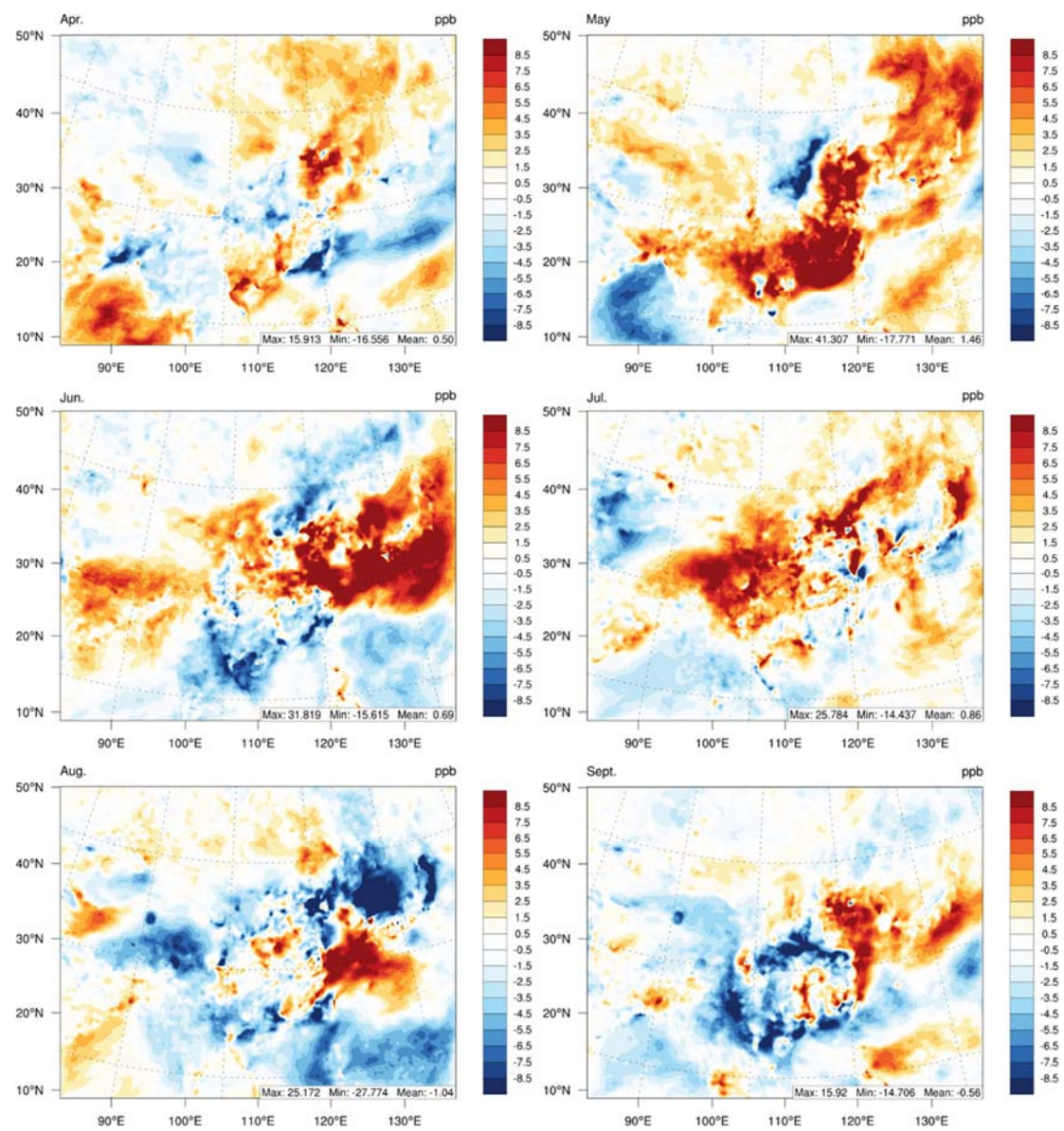


Fig. S13 The change of AvgMDA8 O<sub>3</sub> due to  $\Delta$ Met-drive in April–September

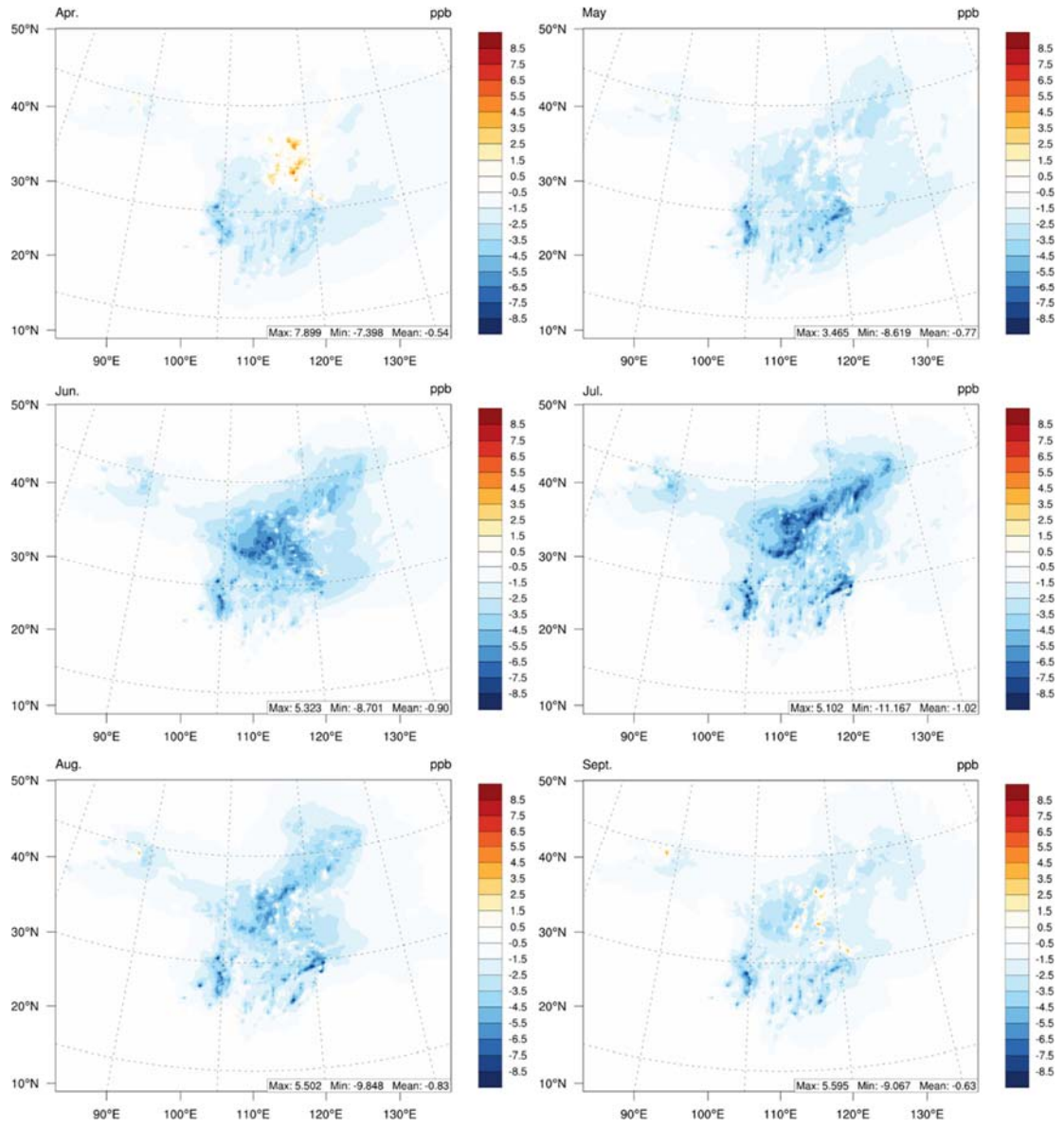


Fig. S14 The change of AvgMDA8 O<sub>3</sub> due to  $\Delta\text{Emis-drive}$  in April–September



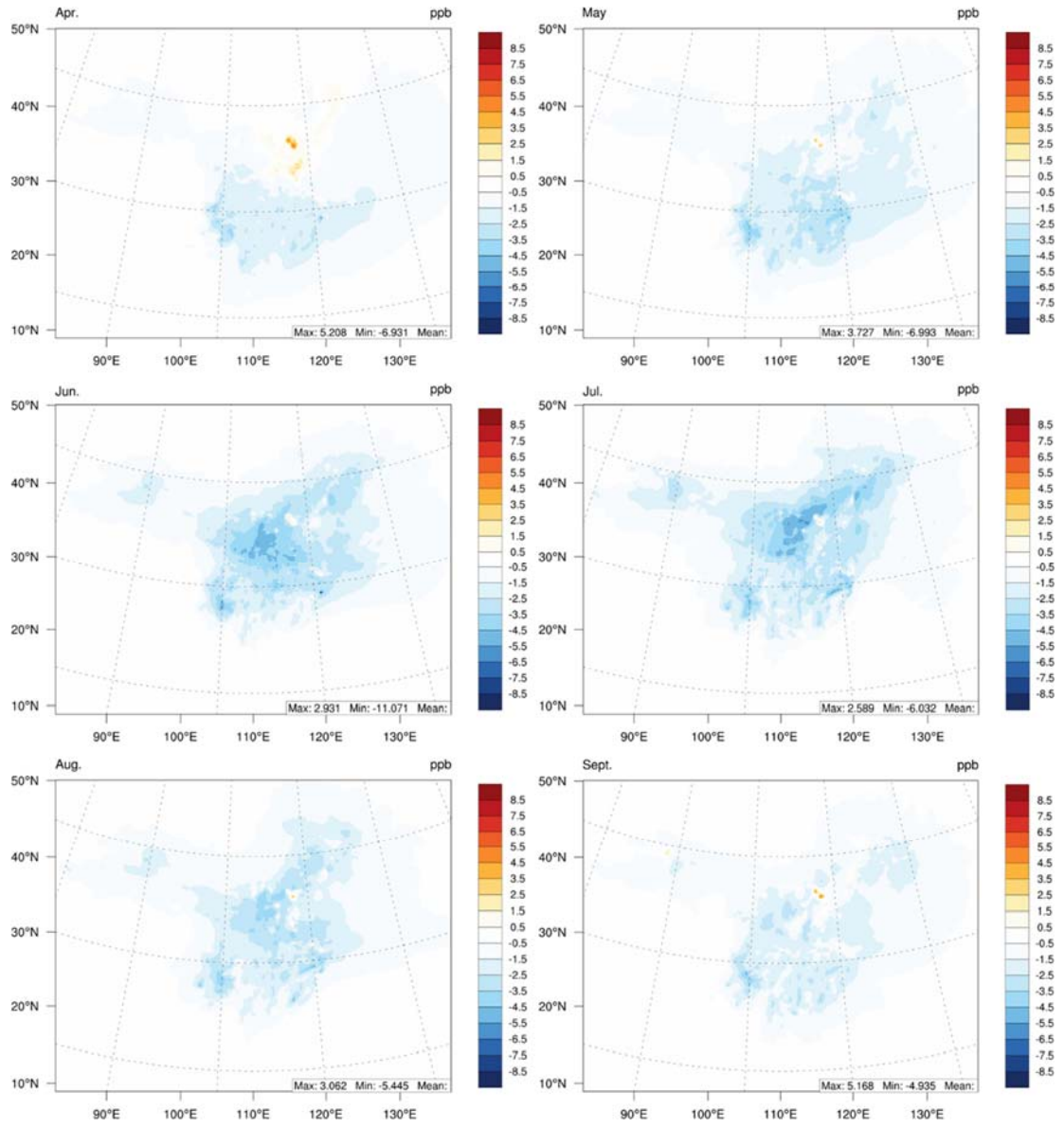


Fig. S15 The change of average  $O_3$  due to  $\Delta E_{\text{emis}}$ -drive in April–September



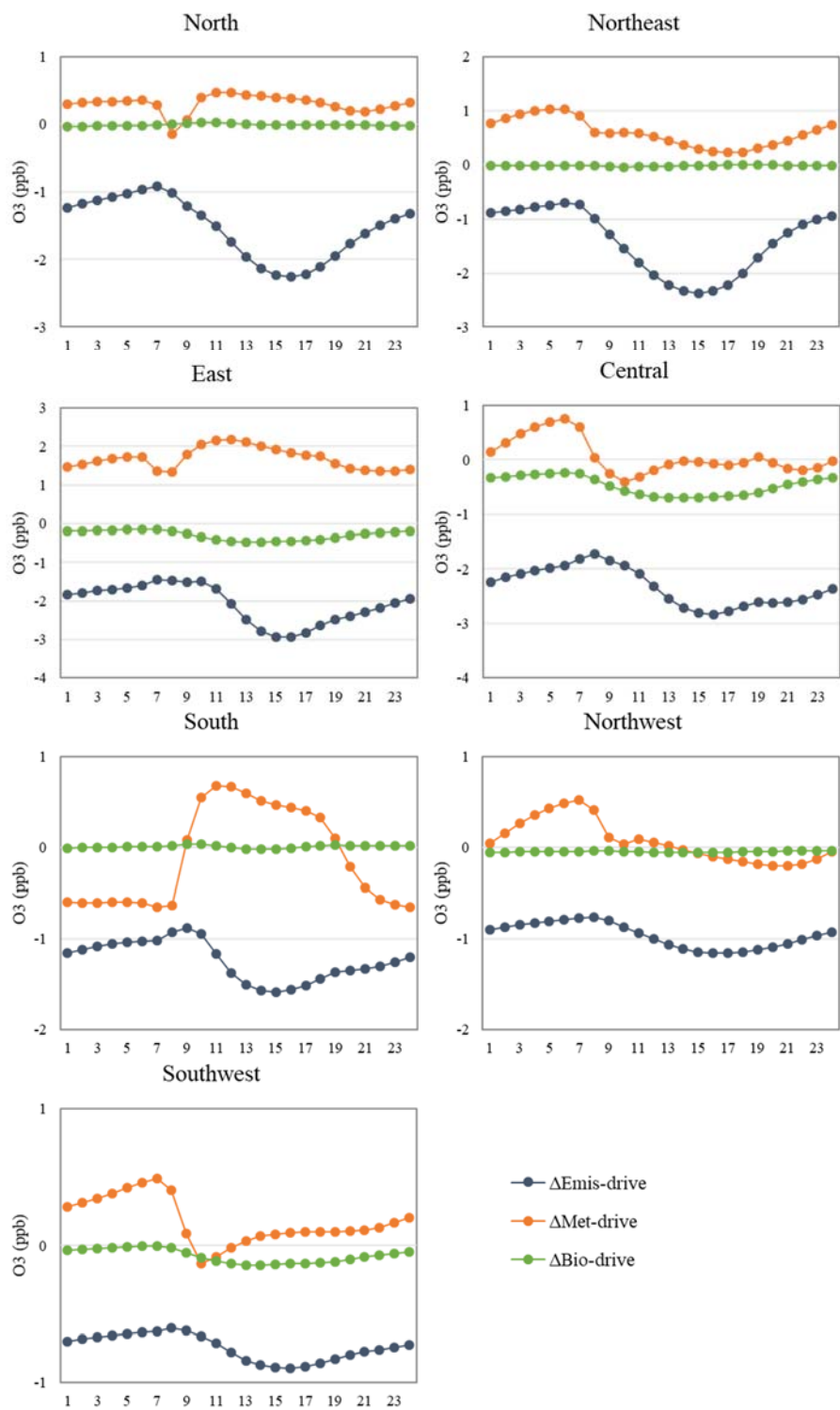


Fig. S16 The simulated diurnal variation of O<sub>3</sub> response (ppb) to the change in meteorology and emission in warm-seasons from 2013 to 2017

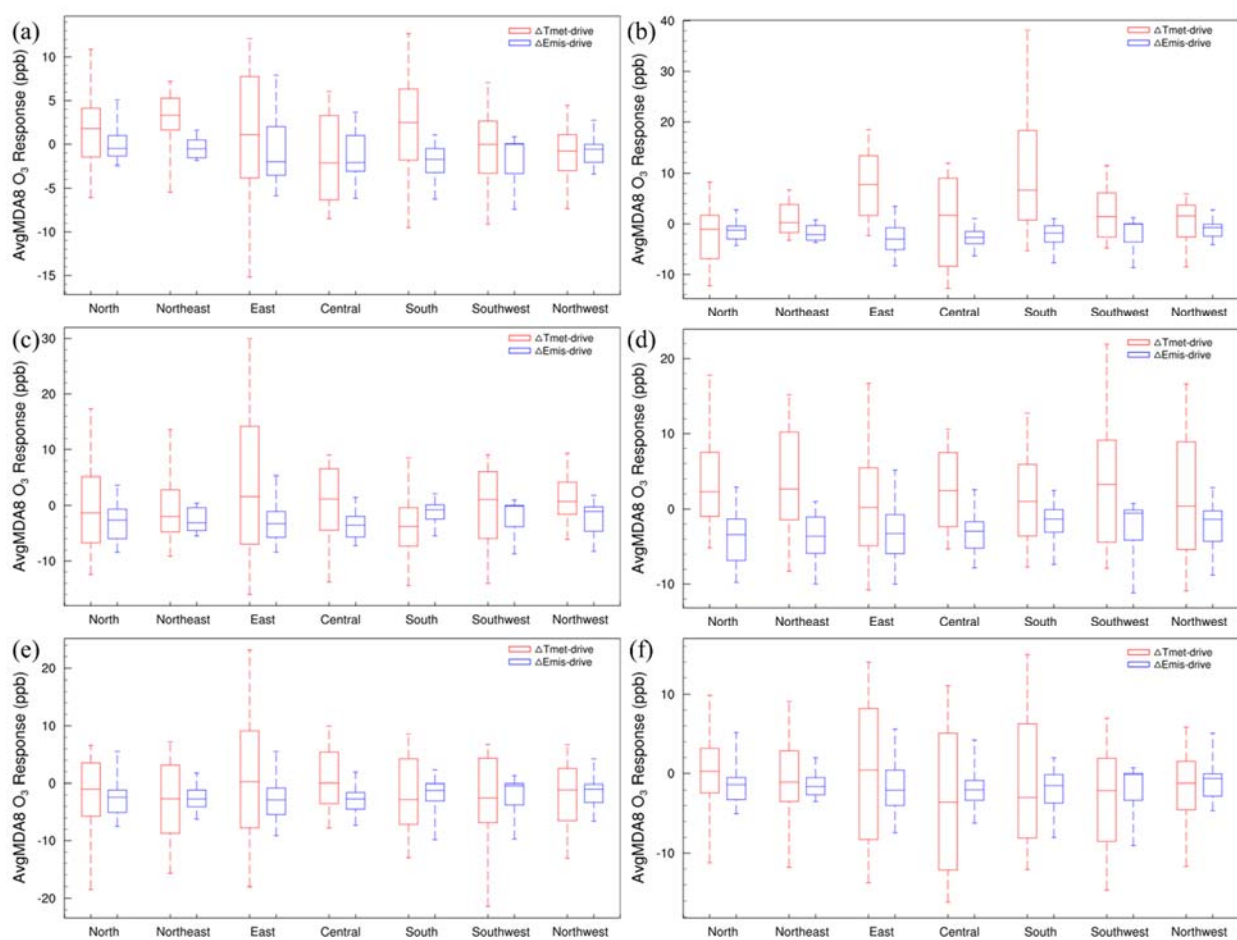


Fig. S17 The AvgMDA8 O<sub>3</sub> response to meteorology and emission in (a) April, (b) May, (c) June, (d) July, (e) August, and (f) September. Color boxes show the range of the 5th and 95th percentiles values and the median line shows the median value

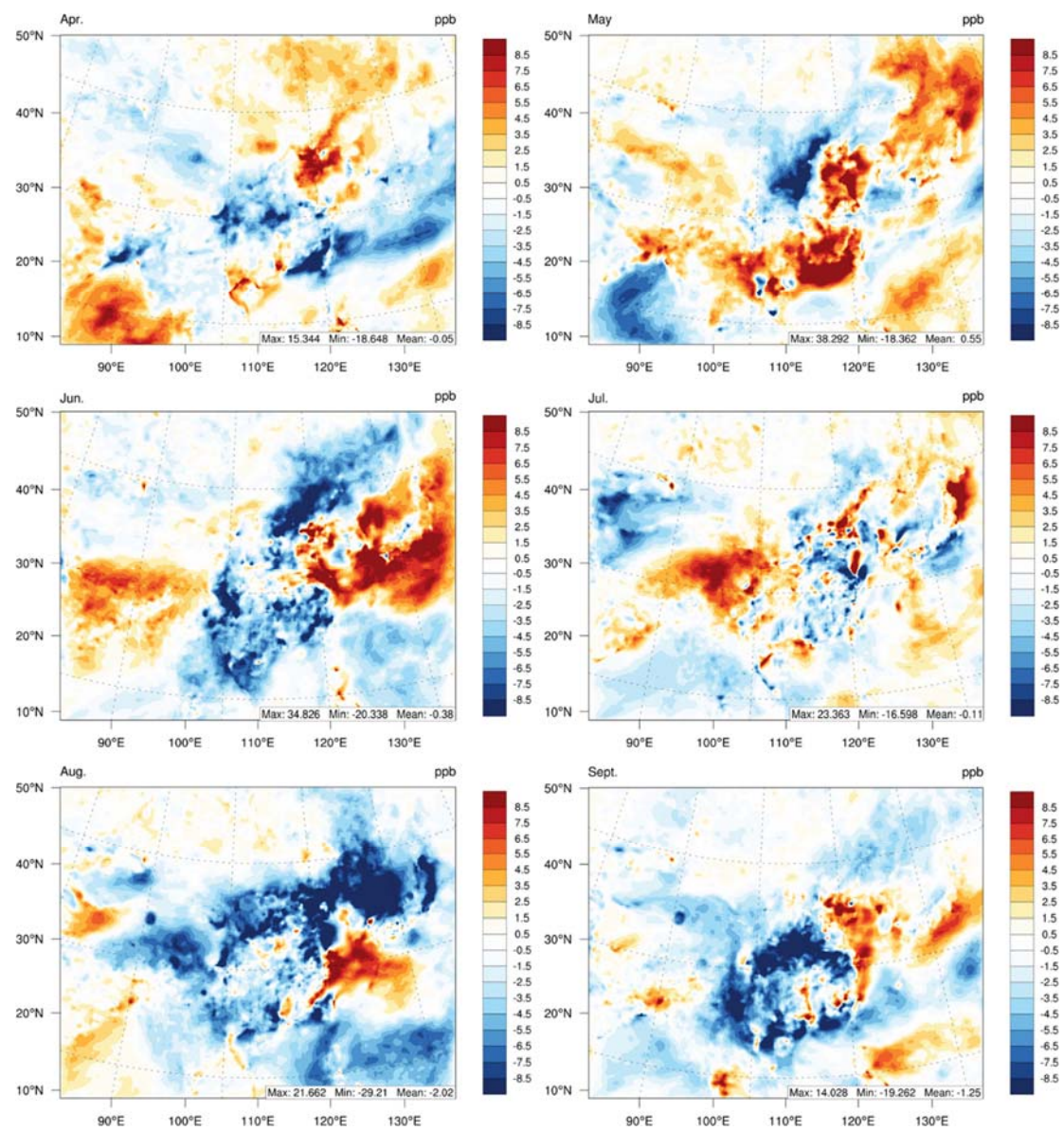


Fig. S18 The  $\Delta$ TOT change of AvgMDA8 O<sub>3</sub> in April–September

Table S1 WRF performance at simulating meteorological variables in the warm season in 2013

| Meteorological variable | Statistic  | Unit   | North  | North-east | East   | Central | South  | North-west | South-west |
|-------------------------|------------|--------|--------|------------|--------|---------|--------|------------|------------|
| Wind Speed              | MeanOBS    | (m/s)  | 2.99   | 2.91       | 3.06   | 2.34    | 2.65   | 2.67       | 2.12       |
|                         | MeanPRD    | (m/s)  | 3.10   | 3.06       | 2.86   | 2.39    | 2.61   | 3.08       | 2.54       |
|                         | Bias       | (m/s)  | 0.11   | 0.16       | −0.20  | 0.04    | −0.05  | 0.42       | 0.42       |
|                         | GrossError | (m/s)  | 1.19   | 1.05       | 1.13   | 0.97    | 1.00   | 1.50       | 1.24       |
|                         | RMSE       | (m/s)  | 1.58   | 1.33       | 1.61   | 1.34    | 1.32   | 2.03       | 1.64       |
|                         | SysRMSE    | (m/s)  | 0.93   | 0.89       | 1.17   | 1.00    | 0.90   | 1.32       | 1.04       |
|                         | UnsysRMSE  | (m/s)  | 1.26   | 0.98       | 1.10   | 0.87    | 0.95   | 1.53       | 1.26       |
|                         | IOA        |        | 0.78   | 0.78       | 0.78   | 0.69    | 0.78   | 0.67       | 0.66       |
| Temperature             | MeanOBS    | (K)    | 291.60 | 290.35     | 297.25 | 297.71  | 299.72 | 290.68     | 292.00     |
|                         | MeanPRD    | (K)    | 291.15 | 289.25     | 296.56 | 296.73  | 299.33 | 290.67     | 290.06     |
|                         | Bias       | (K)    | −0.45  | −1.10      | −0.69  | −0.97   | −0.39  | −0.01      | −1.95      |
|                         | GrossError | (K)    | 1.63   | 1.82       | 1.73   | 1.81    | 1.30   | 2.17       | 2.76       |
|                         | RMSE       | (K)    | 2.06   | 2.29       | 2.31   | 2.34    | 1.64   | 2.83       | 3.73       |
|                         | SysRMSE    | (K)    | 0.57   | 1.22       | 0.91   | 1.08    | 0.62   | 0.43       | 1.97       |
|                         | UnsysRMSE  | (K)    | 1.97   | 1.91       | 2.10   | 2.05    | 1.50   | 2.79       | 3.16       |
|                         | IOA        |        | 0.97   | 0.93       | 0.91   | 0.91    | 0.91   | 0.97       | 0.94       |
| Humidity                | MeanOBS    | (g/kg) | 8.01   | 9.67       | 15.03  | 14.18   | 18.49  | 6.92       | 11.85      |
|                         | MeanPRD    | (g/kg) | 7.99   | 9.06       | 14.95  | 14.20   | 18.12  | 6.59       | 10.77      |
|                         | Bias       | (g/kg) | −0.02  | −0.61      | −0.08  | 0.02    | −0.37  | −0.33      | −1.08      |
|                         | GrossError | (g/kg) | 1.28   | 1.19       | 1.59   | 1.58    | 1.57   | 1.57       | 2.49       |
|                         | RMSE       | (g/kg) | 1.77   | 1.51       | 2.14   | 2.08    | 2.04   | 2.16       | 3.86       |
|                         | SysRMSE    | (g/kg) | 0.67   | 0.85       | 1.04   | 1.13    | 1.19   | 0.66       | 2.80       |
|                         | UnsysRMSE  | (g/kg) | 1.61   | 1.22       | 1.83   | 1.70    | 1.62   | 2.03       | 2.60       |
|                         | IOA        |        | 0.87   | 0.77       | 0.81   | 0.74    | 0.67   | 0.85       | 0.79       |



Table S2 WRF performance at simulating meteorological variables in the warm season in 2017

| Meteorological variable | Statistic  | Unit   | North  | North-east | East   | Central | South  | North-west | South-west |
|-------------------------|------------|--------|--------|------------|--------|---------|--------|------------|------------|
| Wind Speed              | MeanOBS    | (m/s)  | 3.00   | 3.00       | 2.81   | 2.38    | 2.46   | 2.68       | 2.07       |
|                         | MeanPRD    | (m/s)  | 3.02   | 3.12       | 2.64   | 2.25    | 2.40   | 3.06       | 2.48       |
|                         | Bias       | (m/s)  | 0.01   | 0.12       | −0.18  | −0.14   | −0.06  | 0.38       | 0.41       |
|                         | GrossError | (m/s)  | 1.21   | 1.06       | 1.08   | 0.96    | 0.99   | 1.47       | 1.21       |
|                         | RMSE       | (m/s)  | 1.61   | 1.36       | 1.53   | 1.34    | 1.32   | 1.96       | 1.59       |
|                         | SysRMSE    | (m/s)  | 0.99   | 0.87       | 1.10   | 1.03    | 0.90   | 1.26       | 0.99       |
|                         | UnsysRMSE  | (m/s)  | 1.25   | 1.03       | 1.07   | 0.84    | 0.95   | 1.49       | 1.24       |
|                         | IOA        |        | 0.78   | 0.79       | 0.78   | 0.71    | 0.76   | 0.71       | 0.66       |
| Temperature             | MeanOBS    | (K)    | 292.84 | 291.16     | 297.66 | 297.42  | 300.37 | 290.95     | 292.04     |
|                         | MeanPRD    | (K)    | 292.13 | 290.14     | 296.88 | 296.49  | 299.78 | 290.55     | 289.96     |
|                         | Bias       | (K)    | −0.71  | −1.02      | −0.77  | −0.93   | −0.60  | −0.40      | −2.08      |
|                         | GrossError | (K)    | 1.76   | 1.79       | 1.74   | 1.73    | 1.39   | 2.30       | 2.83       |
|                         | RMSE       | (K)    | 2.22   | 2.26       | 2.31   | 2.27    | 1.73   | 2.98       | 3.80       |
|                         | SysRMSE    | (K)    | 0.77   | 1.12       | 0.97   | 0.99    | 0.68   | 0.64       | 2.10       |
|                         | UnsysRMSE  | (K)    | 2.06   | 1.92       | 2.08   | 2.02    | 1.57   | 2.89       | 3.16       |
|                         | IOA        |        | 0.97   | 0.95       | 0.91   | 0.91    | 0.90   | 0.97       | 0.93       |
| Humidity                | MeanOBS    | (g/kg) | 8.22   | 9.23       | 15.52  | 15.13   | 19.34  | 7.16       | 12.96      |
|                         | MeanPRD    | (g/kg) | 8.36   | 8.85       | 14.87  | 14.24   | 18.33  | 6.95       | 11.66      |
|                         | Bias       | (g/kg) | 0.14   | −0.38      | −0.66  | −0.89   | −1.01  | −0.20      | −1.30      |
|                         | GrossError | (g/kg) | 1.39   | 1.18       | 1.65   | 1.65    | 1.75   | 1.63       | 2.51       |
|                         | RMSE       | (g/kg) | 1.93   | 1.52       | 2.18   | 2.08    | 2.18   | 2.25       | 3.84       |
|                         | SysRMSE    | (g/kg) | 0.87   | 0.70       | 1.23   | 1.30    | 1.38   | 0.59       | 2.73       |
|                         | UnsysRMSE  | (g/kg) | 1.70   | 1.32       | 1.77   | 1.57    | 1.67   | 2.15       | 2.67       |
|                         | IOA        |        | 0.87   | 0.80       | 0.79   | 0.76    | 0.63   | 0.83       | 0.81       |

Table S3 Precursor emission rates of 40 scenarios used to establish the response surface

| Case | NO <sub>x</sub> | SO <sub>2</sub> | NH <sub>3</sub> | VOC   |
|------|-----------------|-----------------|-----------------|-------|
| #1   | 0.020           | 0.500           | 1.111           | 1.280 |
| #2   | 0.045           | 1.500           | 0.099           | 0.720 |
| #3   | 0.080           | 0.125           | 1.580           | 1.680 |
| #4   | 0.125           | 0.875           | 1.605           | 0.013 |
| #5   | 0.180           | 1.125           | 0.395           | 0.461 |
| #6   | 0.245           | 1.875           | 0.420           | 1.549 |
| #7   | 0.320           | 0.031           | 1.901           | 0.963 |
| #8   | 0.405           | 0.469           | 0.011           | 1.795 |
| #9   | 0.500           | 0.781           | 1.097           | 0.051 |
| #10  | 0.605           | 1.719           | 1.298           | 0.627 |
| #11  | 0.720           | 0.281           | 0.176           | 1.843 |
| #12  | 0.845           | 1.219           | 1.855           | 1.181 |
| #13  | 0.980           | 1.531           | 1.726           | 1.885 |
| #14  | 1.125           | 1.969           | 0.538           | 0.115 |
| #15  | 1.280           | 0.008           | 0.672           | 0.819 |
| #16  | 1.445           | 0.242           | 1.956           | 0.157 |
| #17  | 1.620           | 0.633           | 0.044           | 1.373 |
| #18  | 1.805           | 1.617           | 1.328           | 1.949 |
| #19  | 2.000           | 0.195           | 1.462           | 0.205 |
| #20  | 0.195           | 1.055           | 0.274           | 1.037 |
| #21  | 0.380           | 1.320           | 0.145           | 0.451 |
| #22  | 0.555           | 1.930           | 1.824           | 1.539 |
| #23  | 0.720           | 0.070           | 0.702           | 1.987 |
| #24  | 0.875           | 0.680           | 0.903           | 0.001 |
| #25  | 1.020           | 0.945           | 1.989           | 0.346 |
| #26  | 1.155           | 1.805           | 0.001           | 1.332 |
| #27  | 1.280           | 0.383           | 0.956           | 0.771 |
| #28  | 1.395           | 1.367           | 1.176           | 1.705 |
| #29  | 1.500           | 1.758           | 0.122           | 0.018 |
| #30  | 1.595           | 1.992           | 1.669           | 0.492 |
| #31  | 1.680           | 0.002           | 1.648           | 1.606 |
| #32  | 1.755           | 0.123           | 0.440           | 1.009 |
| #33  | 1.820           | 0.564           | 0.506           | 1.815 |
| #34  | 1.875           | 1.561           | 1.922           | 0.062 |
| #35  | 1.920           | 0.158           | 0.020           | 0.664 |
| #36  | 1.955           | 0.967           | 1.172           | 1.905 |
| #37  | 1.980           | 1.221           | 1.355           | 1.221 |
| #38  | 1.995           | 1.904           | 0.206           | 1.900 |
| #39  | 2.000           | 0.049           | 1.951           | 0.131 |
| #40  | 0.000           | 0.576           | 1.761           | 0.861 |

Table S4 The average variation of AvgMDA8 O<sub>3</sub> for regions due to ΔMet-drive in April–September

| Region     | AvgMDA8 O <sub>3</sub> variation (Mean ± Std. Dev. ppb) |            |            |           |            |            |
|------------|---------------------------------------------------------|------------|------------|-----------|------------|------------|
|            | April                                                   | May        | June       | July      | August     | September  |
| North      | 1.6 ± 1.8                                               | −1.6 ± 2.8 | −0.7 ± 4.0 | 2.3 ± 2.7 | −0.6 ± 2.8 | 0.3 ± 1.9  |
| North-east | 3.3 ± 1.2                                               | 0.5 ± 1.7  | −1.6 ± 2.5 | 2.8 ± 3.6 | −2.5 ± 3.4 | −0.7 ± 2.1 |
| East       | 1.4 ± 3.4                                               | 7.8 ± 3.5  | 3.5 ± 6.8  | 0.4 ± 3.0 | 0.6 ± 4.7  | 0.6 ± 4.6  |
| Central    | −1.9 ± 2.7                                              | 1.6 ± 5.5  | 2.3 ± 3.0  | 3.1 ± 2.8 | 1.0 ± 3.0  | −2.9 ± 4.9 |
| South      | 2.7 ± 2.6                                               | 8.5 ± 5.3  | −3.5 ± 2.3 | 0.9 ± 2.8 | −3.0 ± 3.1 | −2.9 ± 4.0 |
| South-west | 0.0 ± 1.7                                               | 1.7 ± 2.7  | 1.2 ± 3.3  | 2.9 ± 4.1 | −2.1 ± 3.4 | −2.6 ± 3.2 |
| North-west | −0.8 ± 1.3                                              | 1.3 ± 2.0  | 1.0 ± 1.8  | 0.9 ± 4.2 | −1.2 ± 2.7 | −1.3 ± 2.1 |

Table S5 The average variation of AvgMDA8 O<sub>3</sub> for regions due to ΔEmis-drive in April–September

| Region     | AvgMDA8 O <sub>3</sub> variation (Mean ± Std. Dev. ppb) |            |            |            |            |            |
|------------|---------------------------------------------------------|------------|------------|------------|------------|------------|
|            | April                                                   | May        | June       | July       | August     | September  |
| North      | −0.4 ± 0.8                                              | −1.4 ± 0.8 | −2.9 ± 1.6 | −3.7 ± 1.8 | −2.6 ± 1.3 | −1.5 ± 1.0 |
| North-east | −0.5 ± 0.6                                              | −2.0 ± 0.8 | −2.8 ± 1.3 | −3.5 ± 1.5 | −2.6 ± 1.0 | −1.6 ± 0.7 |
| East       | −1.4 ± 1.8                                              | −2.9 ± 1.3 | −3.3 ± 1.5 | −3.2 ± 1.6 | −2.9 ± 1.5 | −2.0 ± 1.4 |
| Central    | −1.8 ± 1.2                                              | −2.7 ± 0.8 | −3.6 ± 1.2 | −3.1 ± 1.2 | −2.8 ± 0.9 | −2.0 ± 0.9 |
| South      | −1.8 ± 0.8                                              | −1.9 ± 1.0 | −0.9 ± 0.9 | −1.4 ± 1.0 | −1.4 ± 1.1 | −1.6 ± 1.2 |
| South-west | −0.7 ± 1.2                                              | −0.8 ± 1.3 | −0.9 ± 1.4 | −1.2 ± 1.4 | −1.0 ± 1.3 | −0.8 ± 1.2 |
| North-west | −0.7 ± 0.6                                              | −0.9 ± 0.7 | −1.5 ± 1.3 | −1.7 ± 1.3 | −1.3 ± 1.0 | −0.9 ± 0.9 |