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Supplement of

Mitigation of $PM_{2.5}$ and ozone pollution in Delhi: a sensitivity study during the pre-monsoon period

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S1. Comparisons between observations and model results of domain-03 and domain-04

The model (driven by ECMWF) results of domain-03 (D03, 5 km) and domain-04 (D04, 1.67 km) are compared with observations, as shown in Fig. S9. One can see that the model performance is not improved with higher resolution in D04. The median and mean values of $PM_{2.5}$ and ozone from D03 simulation agree well with observations, although there is slightly overestimation of NOx. The $PM_{2.5}$ and NOx, which are mainly primary pollutants, are even more overestimated by D04 than by D03. The secondary pollutant ozone is therefore more underestimated by D04, due to depleted by too much NOx. These may imply an overestimation of NOx emission in the inventory and/or an underestimation of horizontal mixing efficiency in the WRF-Chem model with high resolution simulations.

S2. Comparison between simulations driven by ECMWF and NCEP datasets

The model performance of meteorology simulation is validated by the measurements in Delhi as shown in Fig. S10 (temperature-T and relative humidity-RH) and Fig. S11 (wind pattern). Both simulations driven by ECMWF and NCEP datasets reproduce the T very well with averaged factor around 1 and R=0.9 compared with measurements, although some underestimations can be found in the results driven by ECMWF when T is less than 35°C. The model results driven by ECMWF reproduce RH fairly well (R=0.7), and much better than the NCEP one (R=0.4). The model results driven by NCEP under-predict RH by 20-40%, despite an underestimation in high RH regime (RH>50%) can also be observed in the results driven by ECMWF. These findings are consistent with a recent study (Chatani and Sharma, 2018), which shows the WRF-Chem driven by ECMWF can reproduce much better meteorological conditions compared with observations over India than the driven by NCEP. They also reported that this is a general situation over the whole year (2010) of India and North Pakistan simulation, but the pre-monsoon (April-May) possibly experiences the largest underestimation of RH by more than 20% over Delhi in the results driven by NCEP. The observed wind pattern, dominated by the West-North wind direction, is reasonably captured by simulations driven by both ECMWF and NCEP (Fig. S11). Simulation driven by NCEP produces slightly better wind direction than the one driven by ECMWF, but with a slight overestimation of wind speed can be observed as indicated by less blue colour regions in Fig. S11b.

The model driven by NCEP data predicts slightly lower $PM_{2.5}$ (Fig. S1-S2) and very close O_3 (Fig. S5-S6) concentrations compared to the ECMWF driven one, although a large difference in relative humidity can be found. The lower $PM_{2.5}$ values from NCEP driven results possibly due to the higher height of PBL, which can approach ~3500 meter during afternoon in contrast of ~2500 meter of the ECMWF driven one. The deeper PBL dilutes the fresh emitted $PM_{2.5}$ in the surface layer. This can be especially important in Delhi, where primary particles are the major contributor to $PM_{2.5}$ during pre-monsoon (see section 3.1), and secondary inorganic aerosol (SIA), including sulphate, nitrate and ammonium, only contributes 20-25% of $PM_{2.5}$ loading in both ECMWF and NCEP results. It is worth noting that the difference in relative humidity results between model driven by ECMWF and NCEP may have a larger impact on $PM_{2.5}$ loading and SIA formation during winter period in Delhi when the atmosphere is more humid.

In general, the model driven by ECMWF can produce better meteorological conditions and $PM_{2.5}$ results than the NCEP driven one, while similar O_3 results are found. In this study, our baseline simulation is driven by ECMWF dataset.

S3 Regional Influence of the Delhi Urban Plume

The pollution plume from local emissions in Delhi can also influence downwind regions, particularly to the southeast of Delhi in this season due to the prevailing northwest wind. Fig. S12 shows the spatial distribution of SIs corresponding to traffic emissions for $PM_{2.5}$ and O_3 over Delhi and nearby regions. We consider only the local traffic sector (TRA) here, since it is the governing factor for both $PM_{2.5}$ and O_3 in Delhi, and the major contributor of primary $PM_{2.5}$ and NOx. In this study, we use O_3 peak hour (15:00 LT) with the fully developed PBL to represent the influence of plume in daytime. And we use the early morning before PBL development (05:00 LT) to represent the influence in night, which shows a strong regional interaction indicated by the highest sensitivity of $PM_{2.5}$ to the emissions from NCR emissions (Fig. 4a). In general, the Delhi urban plume has a broader influence at night, possibly facilitated by favourable meteorological conditions of strong regional interactions. The NOx-rich urban plume depletes O_3 in downwind regions during the night with sensitivity larger than 70%, in contrast of a negligible sensitivity (<10%) for $PM_{2.5}$. This indicates that Delhi urban plume has a larger and broader impact on O_3 than on $PM_{2.5}$ in the downwind regions.

Table S1. SAFAR network measurements in Delhi.

No.	Station Name	Short Name	Latitude	Longitude	$PM_{2.5}$	O_3	NOx	Meteorology	Environment Describe
1	C V Raman	CVR	28.72	77.20	Yes	--	--	--	Downtown
2	Delhi University	DEU	28.69	77.21	Yes	--	--	--	Highly populated Residential
3	Airport T3	AIR	28.56	77.10	--	Yes	Yes	--	Airport city side
4	Ayanagar	AYA	28.48	77.13	--	Yes	--	Yes	Suburban background
5	NCMRWF	NCM	28.62	77.36	--	Yes	--	Yes	Industrial, Upwind Entry
6	Pusa	PUS	28.64	77.17	--	--	--	Yes	Background

Table S2. Design of training runs for building Gaussian process emulator.

Training Runs No.	Factors for each emission sector			
	DOM (area source)	TRA (line source)	POW+IND (point source)	NCR[*] (regional transport)
1	1.2958	0.87408	1.0316	0.33741
2	0.75507	1.556	1.8606	0.45469
3	0.48991	0.95171	0.22896	1.416
4	1.4326	1.779	0.63716	1.3508
5	1.3191	0.40663	0.59954	1.1988
6	0.067129	0.023068	1.1011	0.50473
7	0.92064	1.83	0.19348	0.06012
8	0.1336	0.19012	0.38896	0.87948
9	0.37848	1.449	0.90053	1.0461
10	1.6056	0.51501	0.013731	0.75497
11	0.51618	1.2396	1.7039	1.208
12	1.12	0.62141	1.3866	0.96124
13	0.84394	0.20906	0.4144	1.8251
14	0.60487	0.3878	1.6648	1.7574
15	1.5254	1.991	1.4452	1.5008
16	1.784	1.629	0.87087	0.23874
17	1.8007	1.0225	1.2664	1.6046
18	1.0119	1.1866	1.5495	1.9241
19	0.26926	0.73029	0.79889	0.17177
20	1.9168	1.3829	1.9492	0.66879

[*] *Emissions in the National Capital Region surrounding Delhi (domain-03 as shown in Fig. 1), representing the influence of regional transport from surrounding Delhi.*

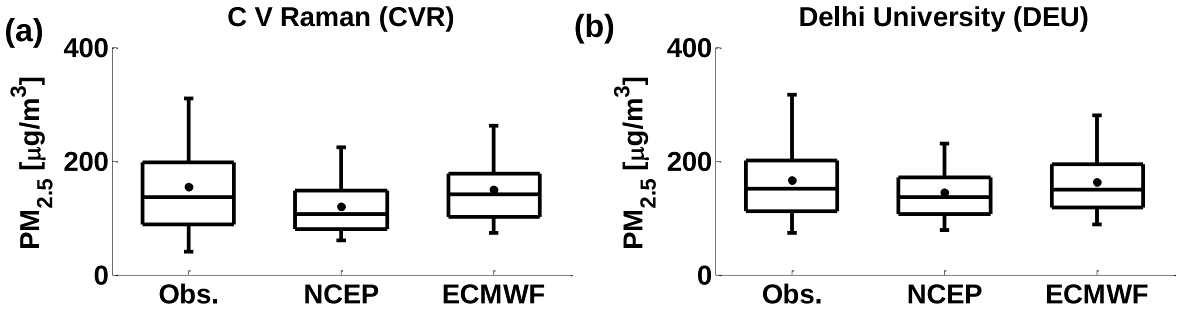

Figure S1. Comparison of the frequency distributions of observed and modelled (driven by NCEP and ECMWF datasets) hourly $PM_{2.5}$ concentrations. (**a**) CVR; (**b**) DEU. The boxplots show the median, mean (black dot), 25% percentile, 75% percentile, 95% percentile and 5% percentile values.

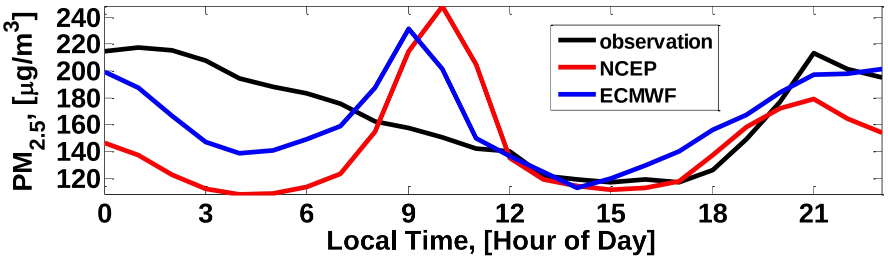

Figure S2. Diurnal patterns of $PM_{2.5}$ at DEU site (marked in Fig. 2). The results are averaged during 02-15 May 2015.

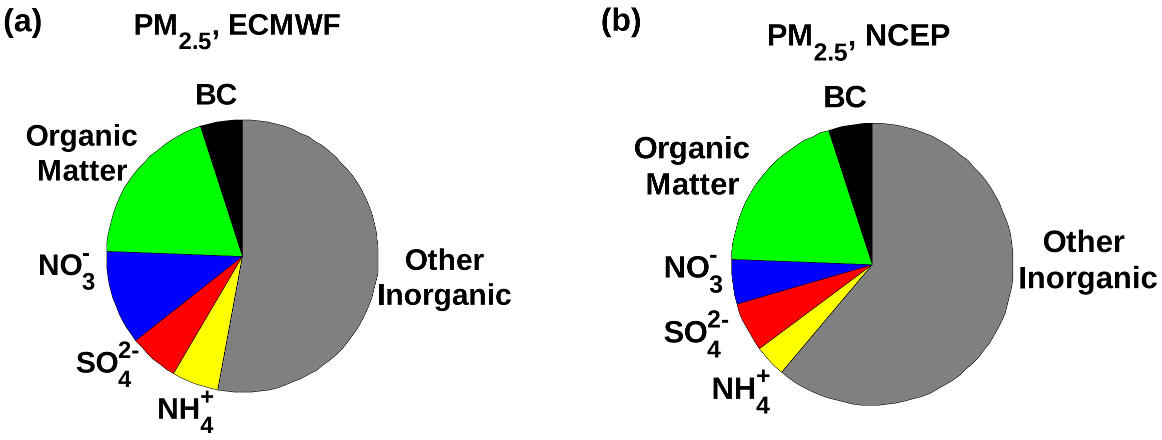

Figure S3. The simulated compositions of $PM_{2.5}$ at Delhi city background site (PUS). The modelled masses of each compounds are averaged during 02-15 May 2015. (**a**) drive by ECMWF data; (**b**) drive by NCEP data.

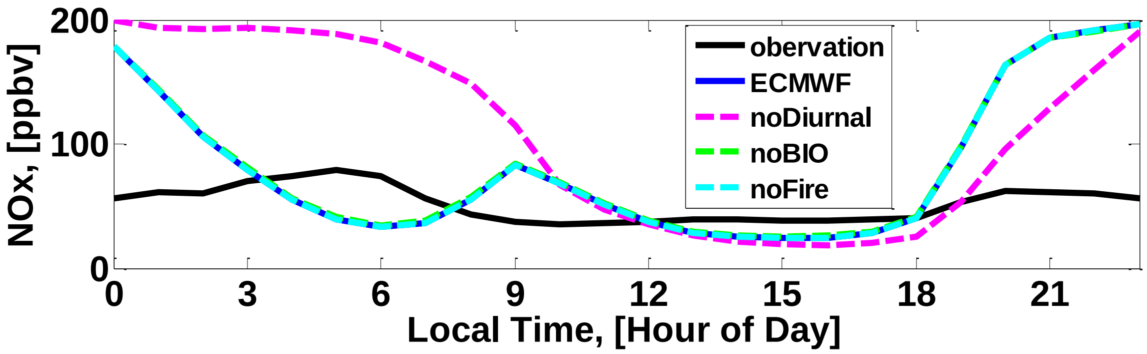

Figure S4. Diurnal patterns of NOx concentration from WRF-Chem model and observational results at AIR site (marked in Fig. 2). The results are averaged during 02-15 May 2015. Note that 'ECMWF' indicates the model results driven by ECMWF reanalysis data.

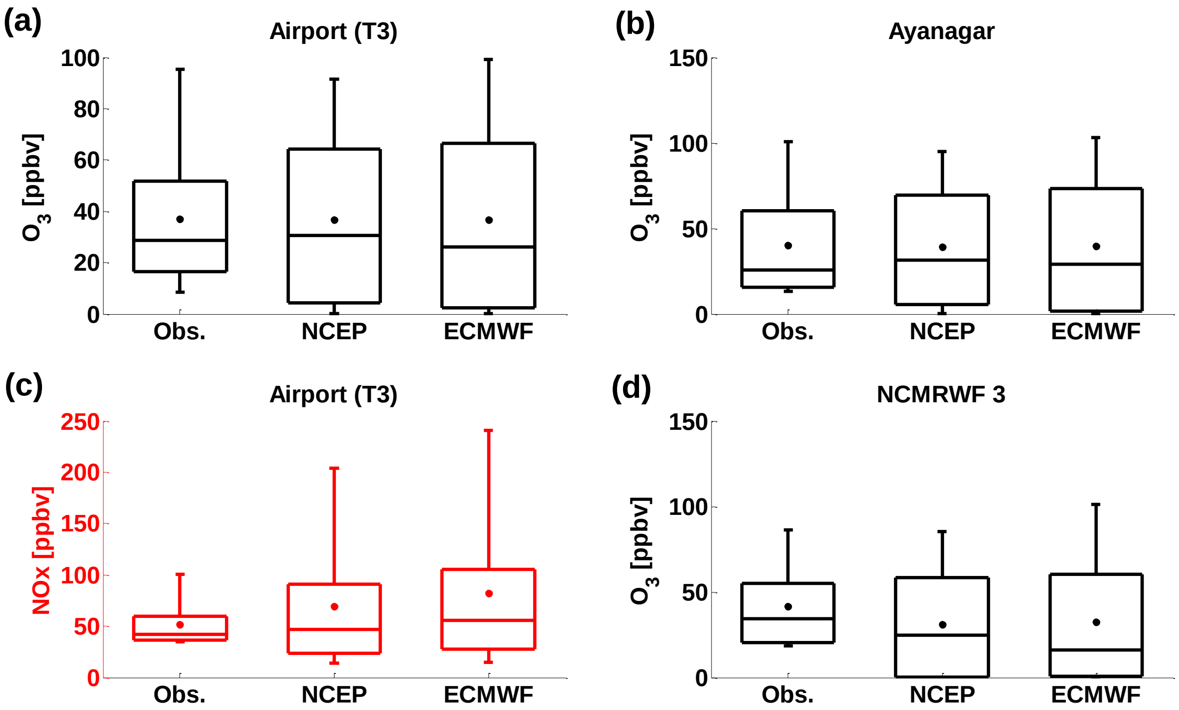

Figure S5. Comparison of the frequency distributions of observed and modelled hourly results (driven by NCEP and ECMWF datasets). (**a**) O_3 at AIR; (**b**) O_3 at AYA; (**c**) NOx at AIR; (**d**) O_3 at NCM. The boxplots show the median, mean (black dot), 25% percentile, 75% percentile, 95% and 5% values.

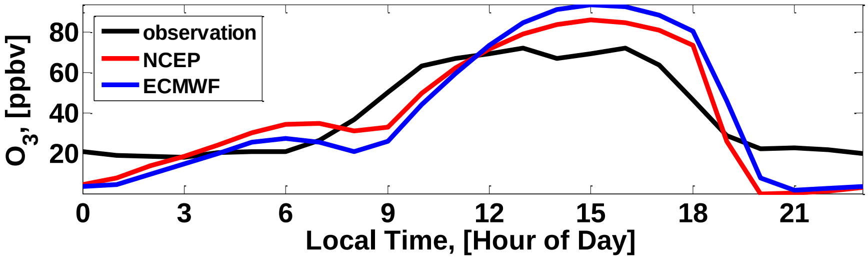

Figure S6. Diurnal patterns of O_3 at AYA, similar as Fig. S2. The 'NCEP' and 'ECMWF' indicate the model results driven by NCEP and ECMWF datasets, respectively.

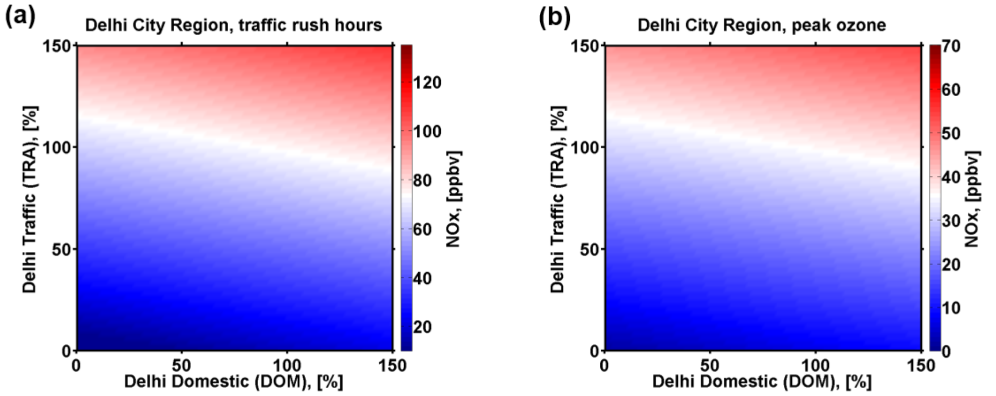

Figure S7. Response surfaces for NOx concentrations over Delhi City Region as a function of local traffic and domestic emissions in Delhi, during average rush hour (**a**) and ozone peak period (**b**).

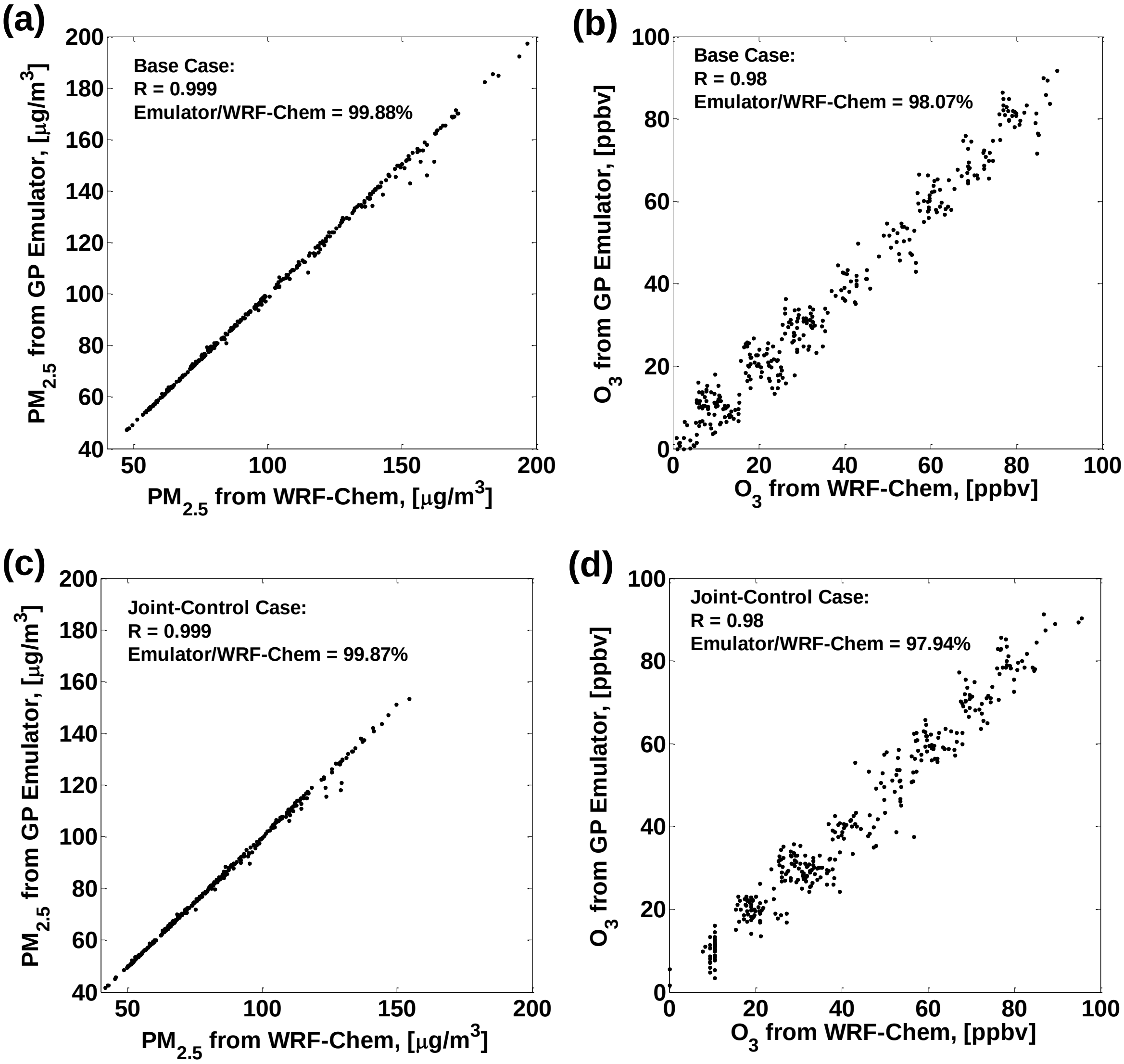

Figure S8. Extra validation of Gaussian process emulator results in the mitigation strategy according to Fig. 7. The accuracy of the emulator for reproducing current conditions of PM$_{2.5}$ **(a)** and O$_3$ **(b)**, i.e. base case without changing emissions. The accuracy of the emulator for reproducing regional joint coordination conditions of PM$_{2.5}$ **(c)** and O$_3$ **(d)**, i.e. NCR joint control case with local traffic emissions reduced by 50% and regional emissions reduced by 30%. All the results are averaged over Delhi City Region, with hourly resolution during the simulation period.

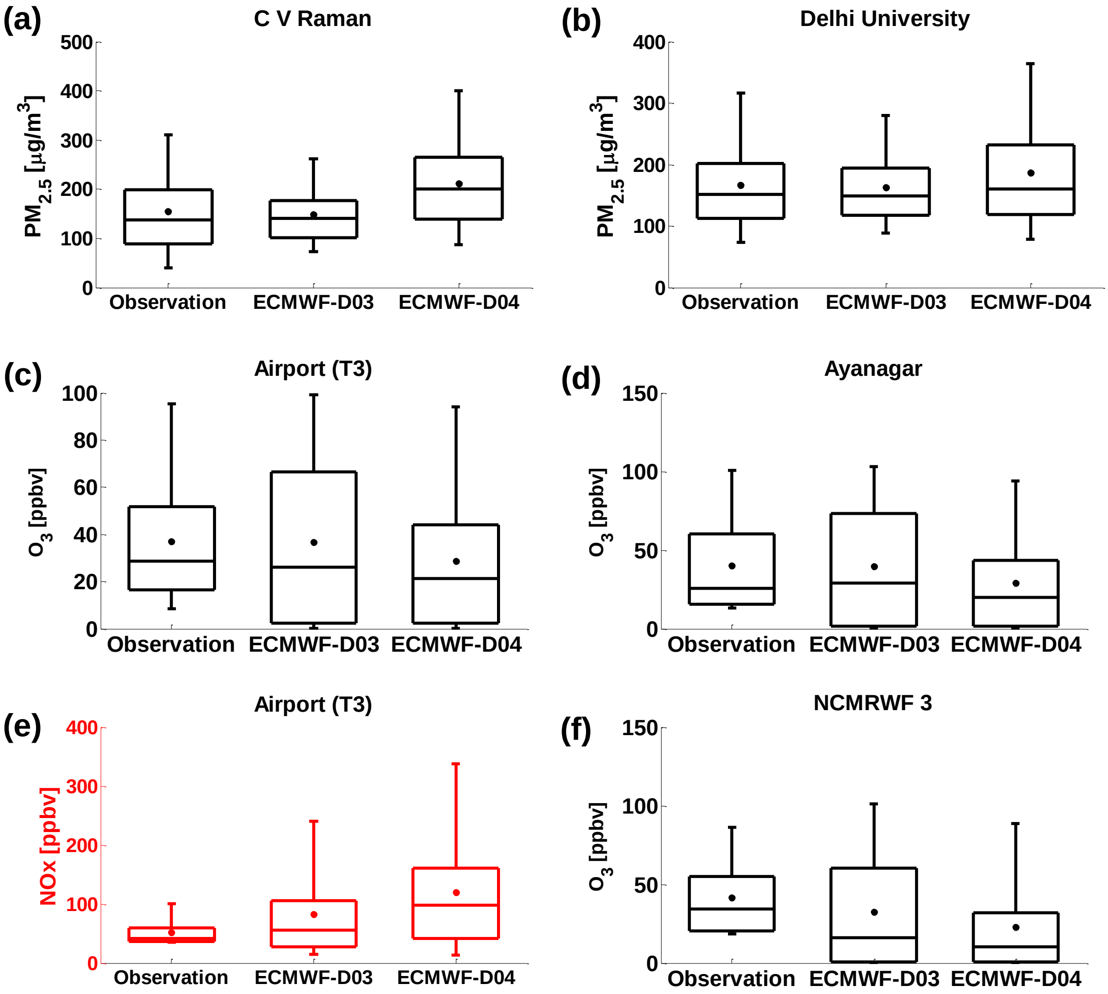

Figure S9. Comparisons of frequency distributions between observations and model results of domain-03 and domain-04. (**a**) PM$_{2.5}$ at CVR; (**b**) PM$_{2.5}$ at DEU; (**c**) O_3 at AIR; (**d**) O_3 at AYA; (**e**) NOx at AIR; (**f**) O_3 at NCM. The WRF-Chem model was driven by ECMWF dataset.

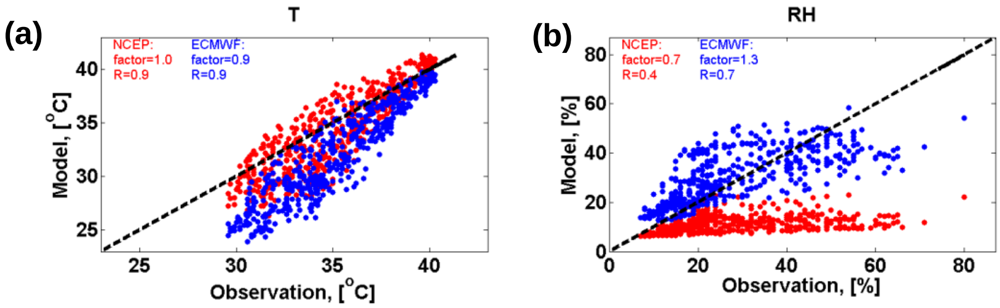

Figure S10. Comparisons of modelled meteorological conditions with all measurements over Delhi. (**a**) temperature (T); (**b**) RH. The red dots indicate the results of WRF-Chem driven by NCEP reanalysis data, blue dots indicate the results of WRF-Chem driven by ECMWF reanalysis data, and the black dashed line indicates the 1:1 line. The measurement sites are given in Table S1, and the corresponding model results are extracted.

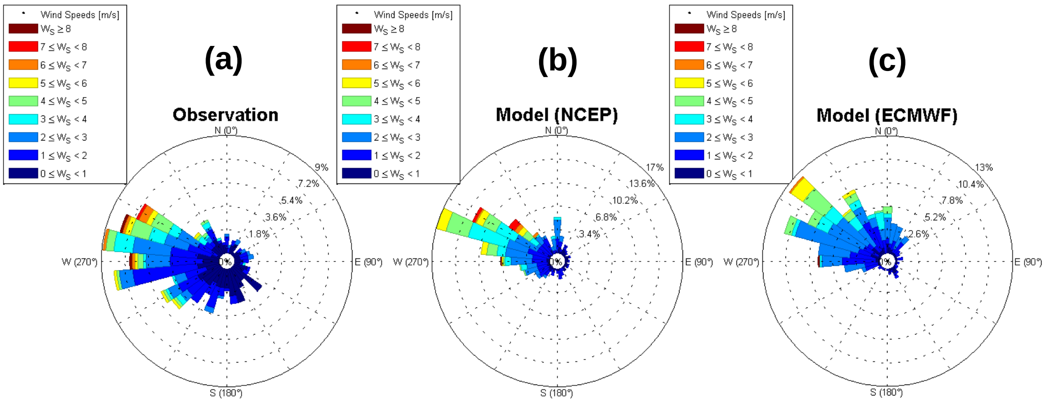

Figure S11. Wind rose pattern of measurements and modelled wind pattern over Delhi. The results from all sites are shown. (**a**) observations; (**b**) model driven by NCEP; (**c**) model driven by ECMWF. The measurement sites are given in Table S1, and the corresponding model results are extracted.

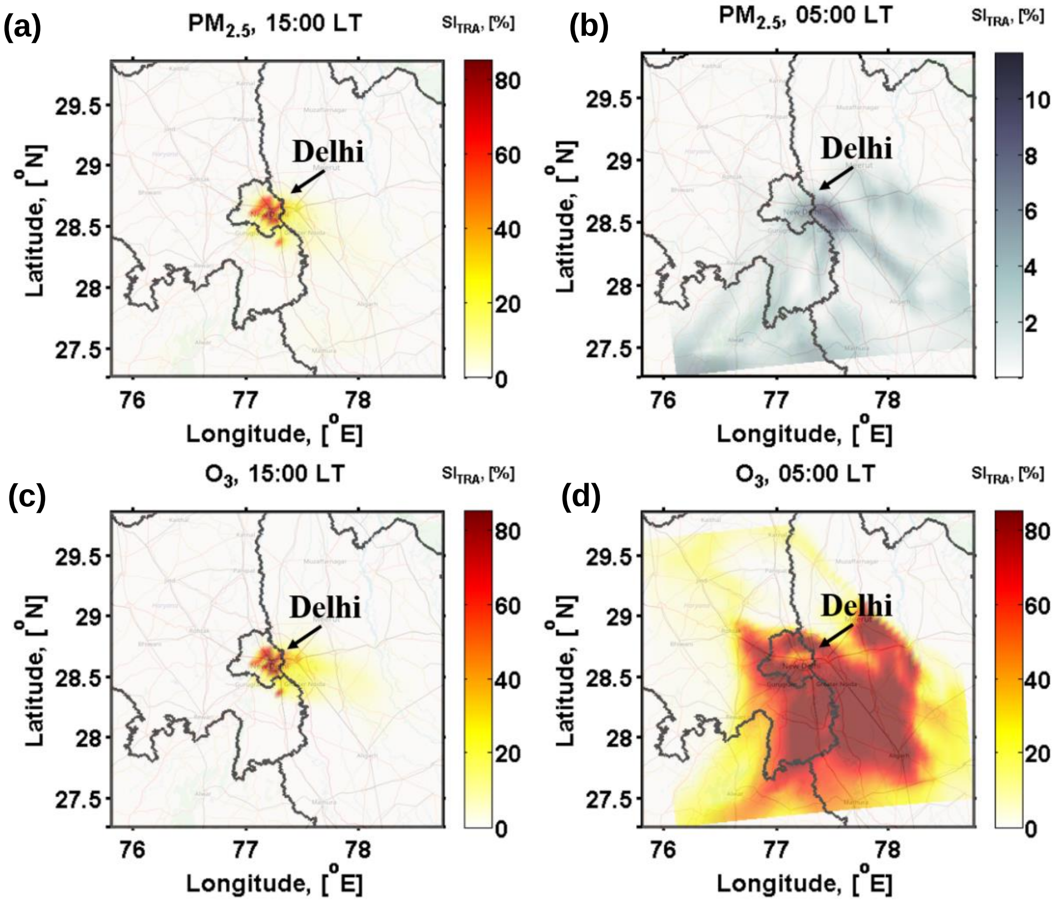

Figure S12. Horizontal distribution of sensitivity index for local traffic emissions in Delhi (SI_{TRA}). The model results are averaged over 02-15 May 2015. Sensitivity indices are shown for: (**a**) $PM_{2.5}$ during ozone peak hour (15:00 LT), (**b**) $PM_{2.5}$ before PBL developed (05:00 LT), (**c**) O_3 at 15:00 LT, and (**d**) O_3 at 05:00 LT. Noting that the scale of colorbar in panel (b) is different from the others.

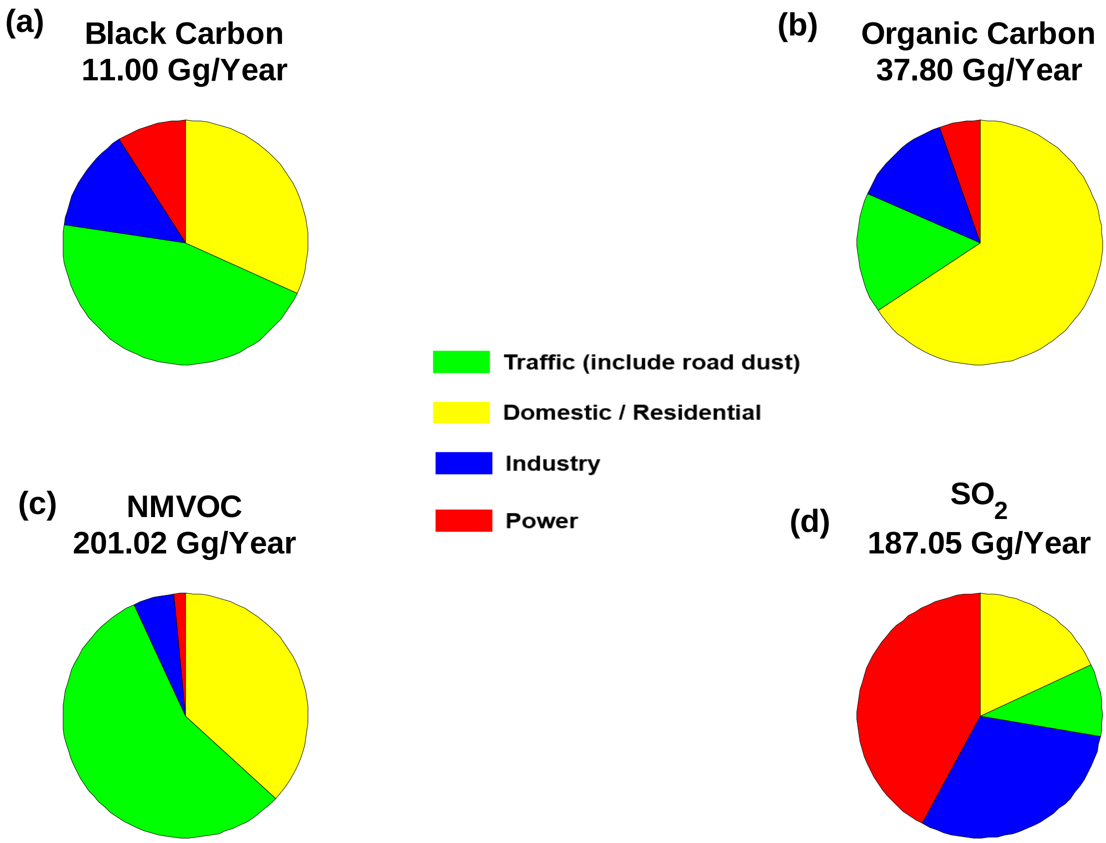

Figure S13. Annual emission of different sectors in Delhi from SAFAR inventory. (a) black carbon; (b) organic carbon; (c) non-methane VOC and (d) SO_2.

Supplementary References:

Chatani, S., and Sharma, S.: Uncertainties Caused by Major Meteorological Analysis Data Sets in Simulating Air Quality Over India, Journal of Geophysical Research: Atmospheres, 123, 6230-6247, doi:10.1029/2017JD027502, 2018.