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Atmospheric Chemistry and Physics Discussions

Mitigation of $PM_{2.5}$ and Ozone Pollution in Delhi: A Sensitivity Study during the Pre-monsoon period

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Atmospheric Chemistry and Physics Discussions

Abstract:

Fine particulate matter ($PM_{2.5}$) and surface ozone (O_3) are major air pollutants in megacities such as Delhi, but the design of suitable mitigation strategies is challenging. Some strategies for reducing $PM_{2.5}$ may have the notable side-effect of increasing O_3. Here, we demonstrate a numerical framework for investigating the impacts of mitigation strategies on both $PM_{2.5}$ and O_3 in Delhi. We use Gaussian process emulation to generate a computationally efficient surrogate for a regional air quality model (WRF-Chem). This allows us to perform global sensitivity analysis to identify the major sources of air pollution, and to generate emission-sector based pollutant response surfaces to inform mitigation policy development. Based on more than 100,000 emulation runs during the pre-monsoon period (peak O_3 season), our global sensitivity analysis shows that local traffic emissions from Delhi city region and regional transport of pollutions emitted from the National Capital Region surrounding Delhi (NCR) are dominant factors influencing $PM_{2.5}$ and O_3 in Delhi. They together govern the O_3 peak and $PM_{2.5}$ concentration during daytime. Regional transport contributes about 80% of the $PM_{2.5}$ variation during the night. Reducing traffic emissions in Delhi alone (e.g., by 50%) would reduce $PM_{2.5}$ by 15-20% but lead to a 20-25% increase in O_3. However, we show that reducing NCR regional emissions by 25-30% at the same time would further reduce $PM_{2.5}$ by 5-10% in Delhi and avoid the O_3 increase. This study provides scientific evidence to support the need for joint coordination of controls on local and regional scales to achieve effective reduction on $PM_{2.5}$ whilst minimize the risk of O_3 increase in Delhi.

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

1. Introduction

Exposure to air pollutants increases morbidity and mortality (Huang et al., 2018a;WHO, 2013). The urban air quality in India, especially in Delhi, is currently among the poorest in the world (WHO, 2013, 2016b, a). In addition to the local impacts, the Indian monsoon can transport air pollutants to remote oceanic regions, inject them into the stratosphere and redistribute them globally (Lelieveld et al., 2018). This makes the impact of Indian air pollution wide ranging regionally and globally as well as having interactions with climate and ecosystems world-wide.

$PM_{2.5}$ (particulate matter with an aerodynamic diameter of less than 2.5 μm) is a major air pollutant, causing increases in disease (Pope et al., 2009;Gao et al., 2015;Stafoggia et al., 2019) and reduced visibility (Mukherjee and Toohey, 2016;Wang and Chen, 2019;Khare et al., 2018). The population of India experiences high $PM_{2.5}$ exposure, and this is responsible for ~1 million premature deaths per year (Conibear et al., 2018;Gao et al., 2018). Residential emissions are estimated to contribute ~50% of $PM_{2.5}$ concentrations and to cause more than 0.5 million annual mortalities across India (Conibear et al., 2018). The World Health Organization (WHO) reported an annual averaged $PM_{2.5}$ loading of ~140 $\mu g/m^3$ in Delhi in 2016 (WHO, 2016b), leading to ~11,000 premature deaths per year in the city (Chowdhury and Dey, 2016). In Delhi, the traffic sector (~50%) and the domestic sector (~20%) are the major local contributors to $PM_{2.5}$ (Marrapu et al., 2014). Efforts to control traffic emissions in Delhi in recent years by introducing an alternating 'odd-even' licence plate policy have led to reductions in $PM_{2.5}$ of less than 10% (Chowdhury et al., 2017). This indicates that there is an urgent need for a coordinated plan to mitigate $PM_{2.5}$ pollution (Chowdhury et al., 2017).

Surface ozone (O_3), another major air pollutant, is damaging to health and reduces crop yields (Ashworth et al., 2013;Lu et al., 2018;Kumar et al., 2018). The risks of respiratory and cardiovascular diseases are increased from short-term exposure to high ambient O_3 and from long-term exposure at low levels (WHO, 2013;Turner et al., 2016;Fleming et al., 2018). Oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NOx) is the main source of surface ozone. Rapid economic development in India has greatly increased the emissions of these O_3 precursors (Duncan et al., 2016),

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

leading to significant increases in O_3 especially during the pre-monsoon period (Ghude et al., 2008). Hourly maximum O_3 reaches as much as 140 ppbv during the pre-monsoon season in Delhi (Ghude et al., 2008), comparable to the most polluted regions in China (150 ppbv, Wang et al., 2017) and higher than the most polluted areas in the U.S. (110 ppbv, Lu et al., 2018).

Mitigation of $PM_{2.5}$ pollution may lead to an increase in surface ozone, because the dimming effect of aerosols and removal of hydroperoxy radicals are reduced, facilitating O_3 production (Huang et al., 2018b;Li et al., 2018;Hollaway et al., 2019). Furthermore, co-reduction of NOx and $PM_{2.5}$ emissions may increase O_3 in cities where O_3 production is in a VOC-limited photochemical regime (Ran et al., 2009;Xing et al., 2018;Xing et al., 2017). This has recently been reported in a number of Asian megacities, e.g. Shanghai (Ran et al., 2009), Beijing (Wu et al., 2015;Liu et al., 2017;Chen et al., 2018a) and Guangzhou (Liu et al., 2013). Delhi and coastal cities in India, which are known to be VOC-limited (Sharma et al., 2017), may face increased O_3 as a side-effect of emission controls focused on $PM_{2.5}$. Therefore, studies of mitigation strategies that target both $PM_{2.5}$ and O_3 are urgently needed (Chen et al., 2018a), particularly as urban air pollution in India has been much less well studied than in many other countries.

To investigate the impacts of mitigation strategies with respect to both $PM_{2.5}$ and O_3, we demonstrate a framework for generating emission-sector based pollutant response surfaces using Gaussian process emulation (O'Hagan and West, 2009;O'Hagan, 2006). We conduct global sensitivity analysis to identify the dominant emission sectors controlling $PM_{2.5}$ and O_3, and then generate sector based response surfaces to quantify the impacts on $PM_{2.5}$ and O_3 of emission reductions. In contrast to simple sensitivity analysis varying one input at a time, this allows full exploration of the entire input space, accounting for the interactions between different inputs (Pisoni et al., 2018;Saltelli et al., 1999). Conventionally, chemical transport models (CTMs, e.g. WRF-Chem) are used to calculate the impacts on pollutants concentrations of different mitigation scenarios. However, the computational expensive of CTMs makes them unsuitable for performing global sensitivity analysis or generating response surfaces, which usually require thousands of model runs. To overcome this difficulty, source-receptor relationships (Amann et al., 2011)

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

or computational efficient surrogate models, trained on a limited number of CTM simulations, are used to replace the expensive CTM. These approaches have been used to perform sensitivity and uncertainty analysis of regional air quality models (Pisoni et al., 2018), assessment of regional air quality plans (Zhao et al., 2017;Xing et al., 2017;Pisoni et al., 2017;Thunis et al., 2016) and sensitivity and uncertainty analysis of global and climate simulations (Ryan et al., 2018;Lee et al., 2016;Lee et al., 2012). Here, we use surrogate model to explore the sensitivity of $PM_{2.5}$ and O_3 on sector based emission controls in Delhi, for developing a mitigation strategy addressing both pollutants.

In this study, we demonstrate the value of such a framework for supporting decision makers in determining better mitigation strategies. We give examples of its use in investigating impacts of mitigation scenarios on $PM_{2.5}$ and O_3 pollutions in Delhi, and demonstrate that regional joint coordination of emission controls over National Capital Region (NCR) of Delhi is essential for an effective reduction of $PM_{2.5}$ whilst minimizing the risk of O_3 increase.

2. Materials and Methods

2.1 WRF-Chem Model Baseline Simulation

WRF-Chem (v3.9.1) – an online, fully coupled chemistry transport model (Grell et al., 2005) – has been widely used in previous studies of air quality across India (Marrapu et al., 2014;Mohan and Gupta, 2018;Gupta and Mohan, 2015;Mohan and Bhati, 2011). The model has also been used to estimate the health burden (Conibear et al., 2018;Ghude et al., 2016) and reduction in crop yields (Ghude et al., 2014) from the exposure to $PM_{2.5}$ and O_3 over India.

In this study, we focus on the hot and dry pre-monsoon period in Delhi, when average temperatures are around 32 ^{o}C and relative humidity (RH) is about 35% (Ojha et al., 2012). O_3 approaches its annual peak in pre-monsoon due to strong solar radiation (Ghude et al., 2008;Ojha et al., 2012). During the pre-monsoon period, desert dust can contribute significantly to particulate matter in Delhi (Kumar et al.,

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

2014b;Kumar et al., 2014a). Here, we perform WRF-Chem simulation for the period of 2–15 May 2015 (with two additional days for spin-up), which was not significantly influenced by dust storm in Delhi according to MODIS observations (https://earthdata.nasa.gov/earth-observation-data/near-real-time/hazards-and-disasters/dust-storms). Strong dust storms started to influence the Indo Gangetic Plain on 21-24 April and 19 May 2015, respectively. This minimizes the uncertainties resulting from dust storm simulation and permits a stronger focus on anthropogenic emissions. Resuspended dust from road traffic is also a major contributor to $PM_{2.5}$ in Delhi, and this is estimated and included in the emission inventory as described below.

The model configuration follows the study of Marrapu et al. (2014), and the parameterizations used are listed in Table 1. Three nested domains are used, with coverage of South Asia (45 km resolution), the Indo Gangetic Plain (15 km resolution), and the National Capital Region (5 km resolution), see Fig. 1. A test simulation with a fourth domain over Delhi at 1.67 km resolution suggests that a further increase in resolution does not substantially improve model performance (details in Text S1), and this is in line with results from a previous study (Mohan and Bhati, 2011). The Carbon Bond Mechanism version Z (CBMZ, Zaveri and Peters, 1999) coupled with the MOSAIC (Zaveri et al., 2008) aerosol module with four size bins is used to represent gaseous chemical reaction and aerosol chemical and dynamical processes. We neglect wet scavenging and cloud chemistry processes here, as the impact of these is likely to be negligible during the dry pre-monsoon period over India.

The initial and boundary conditions for chemical species are provided from MOZART-4 global results (https://www.acom.ucar.edu/wrf-chem/mozart.shtml). Our baseline simulation is driven by European Centre for Medium-Range Weather Forecasts (ECMWF) meteorological data, as we find that this reproduces regional meteorology better than that from the National Centers for Environmental Prediction (NCEP) over India, consistent with a recent study (Chatani and Sharma, 2018). The wind pattern and temperature over Delhi in May 2015 is generally captured well in simulations driven by either meteorological dataset, but the model captures the variation in relative humidity much better (R=0.7) with

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

ECMWF data than with NCEP data (R=0.4, negative bias of 20-40%). More detailed discussion is provided in Text S2.

The high-resolution Fire Inventory from NCAR (FINN, Wiedinmyer et al., 2011) is adopted to provide biomass burning emissions. Interactive biogenic emissions are included using the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006). The global Emission Database for Global Atmospheric Research with Task Force on Hemispheric Transport of Air Pollution (EDGAR-HTAP, Janssens-Maenhout et al., 2015) version 2.2 (year 2010) at 0.1° × 0.1° resolution is used to represent anthropogenic emissions apart from over Delhi, where they are represented by a high-resolution monthly inventory for 2015 developed under the System of Air Quality Forecasting and Research (SAFAR) project (Sahu et al., 2011;Sahu et al., 2015). In the absence of a diurnal variation in emissions specific to Delhi, we adopt diurnal variations from Europe in this study (Denier van der Gon et al., 2011). The SAFAR inventory provides emission fluxes of PM_{10}, $PM_{2.5}$, black carbon, organic carbon, NOx, CO, SO_2 and NMVOC (non-methane volatile organic compounds) from five sectors, including power (POW), industry (IND), domestic or residential (DOM), traffic (TRA) and wind blow dust from roads (WBD). Wind blow dust includes dust resuspended from vehicle movement on paved and unpaved roads (Sahu et al., 2011), and is therefore closely related to traffic emissions, and we combine this into the traffic sector for our study.

The NMVOC emissions are speciated according to the EDGAR (v4.3.2) global inventory (Huang et al., 2017), and are then lumped for the CBMZ chemistry scheme. The speciation mapping is detailed in Table 2 and described below, and a toolkit has been developed to perform this mapping. Emissions of alcohols and ethers are split 20%:80% between methanol and ethanol by mass and then converted to molar emissions with a fractionation based on (Murrells et al., 2009). Emissions of paraffin carbon (PAR) are calculated by converting mass emissions from each VOC group to molar emissions and then multiplying by the number of paraffin carbons in order to conserve carbon. Hexanes and higher alkanes are converted to molar emissions of hexane and then multiplied by six to give PAR emissions. Other alkenes are mapped to molar emissions of butane, and this is then apportioned between terminal olefin

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

carbons (OLET), internal olefin carbons (OLEI) and PAR on a molar ratio of 1:1:4 following (Zaveri and Peters, 1999). Ketones are split 60%:40% by mass between acetone (KET) and methyl-ethyl ketone (MEK), then converted to molar emissions with fractions based on (Murrells et al., 2009). As MEK is not included in the CBMZ mechanism, we apportion molar emissions of MEK equally between KET and PAR.

2.2 Observational Network

Air quality and meteorological monitoring networks are operated in Delhi under the SAFAR project coordinated by IITM (Ministry of Earth Sciences, Government of India). Measurements of $PM_{2.5}$, O_3 and NOx during the May 2015 simulation period are available from six monitoring stations in Delhi: C V Raman (CVR), Delhi University (DEU), Indira Ghandi International Airport Terminal-3 (AIR), Ayanagar (AYA), NCMRWF (NCM) and Pusa (PUS). The instruments are calibrated and measurements are quality controlled in the SAFAR project (http://safar.tropmet.res.in); more details are given in previous studies (Sahu et al., 2011;Beig et al., 2013;Aslam et al., 2017). Site locations are shown in Fig. 2 and measured variables are given in Table S1.

2.3 Global Sensitivity Analysis of Urban Air Pollution

We perform global sensitivity analysis (Iooss and Lemaître, 2015) to quantify the sensitivity of modelled $PM_{2.5}$ and O_3 to each emission sector. Global sensitivity analysis has major advantages over a simple one-at-a-time sensitivity analysis, where a single input is varied while the other inputs are fixed at nominal values, as the latter approach can lead to underestimation of the true sensitivity (Saltelli et al., 1999;Pisoni et al., 2018). The extended Fourier Amplitude Sensitivity Test (eFAST, Saltelli et al., 1999) is a commonly used approach to perform GSA, and is adopted in this study. Since eFAST typically requires thousands of model runs, we employ a computationally cheaper surrogate model in place of WRF-Chem. In this study, we use Gaussian process emulation to create the surrogate model (O'Hagan, 2006), since a Gaussian process emulator typically requires a relatively small number of runs of the computationally-expensive model to generate. This is in contrast to other surrogate modelling approaches, such as neural networks, which typically require thousands of model runs to train them. Gaussian process

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

emulators have been used previously in the uncertainty assessment of atmospheric models (Lee et al., 2016;Lee et al., 2012;Lee et al., 2011). Following these studies, a Latin hypercube space-filling approach is employed to provide the designs of training runs for WRF-Chem. Latin hypercube sampling is a statistical method for generating a near-random sample of parameter values from a multidimensional distribution (Shields and Zhang, 2016). Here, we search through 100,000 Latin hypercube random designs to find the optimal one where the parameter space is filled most effectively. More details of these approaches are described in a previous study (Ryan et al., 2018).

In this study, we focus on a limited number of the emission sectors to demonstrate the effective of the approach: domestic/residential emissions in Delhi (DOM), traffic emissions in Delhi (TRA, including WBD), power and industry in Delhi (POW+IND) and total emissions in the National Capital Region outside Delhi (NCR). NCR represents the contribution of regional transport to pollution in Delhi. According to the SAFAR emission inventory, the total $PM_{2.5}$ emissions of DOM, TRA, POW+IND and NCR are about 1.8, 6.1, 3.1 and 8.5 Gg/month in May 2015, respectively. Gaussian process emulators are built based on 20 training runs of WRF-Chem, with emission scaling drawn from a variation range of 0-200% for each of the four specified sectors (Table S2). Emulation of the impacts of mitigation scenarios on $PM_{2.5}$ and O_3 can be performed in minutes on a laptop, in contrast to simulations with WRF-Chem which require a few days on a high-performance computing cluster. We perform 'leave-one-out' cross-validation (O'Hagan and West, 2009;Wang et al., 2011) with 10,000 random samples to check that the Gaussian process emulator can fully represent the results of WRF-Chem. Modelled and emulated O_3 and $PM_{2.5}$ lie very close to the 1:1 line with R values of more than 0.99 as shown in Fig. 3, suggesting that the emulation provides a good representation of the model.

2.4 Response Surfaces

Response surfaces are useful for investigating the relationship between model inputs and outputs, in this case between sectoral emissions and modelled pollutant concentrations. They have been widely applied for air quality studies and policy making (EPA, 2006a, b;Zhao et al., 2017;Xing et al., 2017). Here, we analyse the responses of $PM_{2.5}$ and O_3 to changes in emissions from each sector of between 0%

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

and 200%. The computationally efficient Gaussian process emulation enables us to generate response surfaces without the computational burden of a large number of runs of the WRF-chem model.

2.5 Outline of Analysis

We use the WRF-Chem model to simulate the hourly concentrations of O_3 and $PM_{2.5}$ over the Delhi region during 2-15 May 2015 and evaluate the results against observations. We perform a simple sensitivity analysis to investigate the contributions of biomass burning and biogenic emissions to $PM_{2.5}$ and O_3 in Delhi. We then conduct a global sensitivity analysis, using Gaussian process emulation, to determine the sensitivity of modelled O_3 and $PM_{2.5}$ concentrations to changes in the dominant anthropogenic emission sectors. Finally, we generate response surfaces to identify appropriate mitigation strategies for reducing $PM_{2.5}$ while minimizing the risks from O_3 increase.

3. Results and Discussion

3.1 Model Performance

The WRF-Chem model captures the general magnitude and variation in $PM_{2.5}$ well (Fig. 4a), with mean bias and error of about -3.5% and 11%, respectively, and an index of agreement (Willmott et al., 2012) of 75%. The frequency distributions of modelled $PM_{2.5}$ are also similar to the observations, with differences in mean and median concentrations of less than 10%, although high concentration spikes are missed by the model (Fig. S1). The modelled $PM_{2.5}$ peaks around 09:00 local time (LT) because the rush hour enhances traffic emissions before the planetary boundary layer (PBL) height has increased (Fig. 4a). This is also seen in the modelled results at DEU (Fig. S2), which is closer to a motorway and shows a more intense $PM_{2.5}$ peak in the morning rush hour. $PM_{2.5}$ is overestimated during the morning rush hour (around 09:00 am) and underestimated during the early morning (03:00-05:00 LT, Fig. 4a and Fig. S2). This may suggest that there is an earlier rush hour or more traffic activity at night in Delhi than in European cities, since we adopted European diurnal emission patterns in this study in the absence of local

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

information. Detailed studies of traffic emissions and their variation in Delhi would help improve these model simulations.

The modelled chemical composition of $PM_{2.5}$ is shown in Fig. S3. Secondary inorganic aerosol (SIA), including sulphate, nitrate and ammonium, only contributes ~25% of aerosol mass in our simulation. In the absence of particle inorganic composition measurements during the simulation period, we compare our results with a previous modelling study of Delhi during the post-monsoon season (Marrapu et al., 2014), which also shows a ~25% contribution of SIA to $PM_{2.5}$ loading, in line with our results. Furthermore, our results are also consistent with an observational study, which reported the mass fraction of organic matter (usually calculated as 1.4 times OC) and elemental carbon (usually equivalent to black carbon in modelling studies, (Chen et al., 2016b) in $PM_{2.5}$ of ~20% and ~6% in Delhi during May 2015, respectively (Sharma et al., 2018).

The model well captures the peak O_3 with a bias of less than 5%, although it underestimates O_3 during night-time (Fig. 4b). In general, the diurnal pattern and magnitude of O_3 are captured by WRF-Chem (Fig. 4b), with normalized mean bias and error of about -20% and 35%, respectively, and an index of agreement of 65%. The underestimation during night-time is likely to be because NOx is overestimated by a factor of 2-3 at night (Fig. S4), and the excess NO depletes O_3. This is indicated by the frequency distribution of O_3 and NOx (Fig. S5), where the median values of observed O_3 and NOx are matched well by the model. However, the higher peaks of modelled NOx concentration lower the modelled O_3 levels, indicating that Delhi is in VOC-limited photochemical regime. Similar results are found at AYA (Fig. S6). The larger underestimation of O_3 at NCM (Fig. S5d, industrial environment site) suggests that NOx emission from the industry sector may be overestimated.

3.2 Impacts of Biogenic and Biomass Burning Emissions

Before exploring the importance of the four selected anthropogenic emission sectors on $PM_{2.5}$ and O_3 in Delhi during simulation period, we investigate the contributions from other factors (biomass burning and biogenic emissions). We remove these sources and find that there is a negligible contribution from

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

biogenic emissions to $PM_{2.5}$ concentrations over Delhi in this season (Fig. 4c and 4d). It is worth noting that biogenic emissions may contribute to secondary organic aerosol (SOA) in Delhi, but the formation of SOA is not well represented by the CBMZ-MOSAIC chemistry-aerosol mechanisms used in this study. However, this weakness is not expected to have a major influence on our pre-monsoon results; as described before, the difference of organic matter fraction between simulation and observation (Sharma et al., 2018) in May 2015 is less than 5%. About 10% of $PM_{2.5}$ in Delhi is derived from biomass burning during the simulation period. Crop burning in Haryana and Punjab states is a major source of this (Jethva et al., 2018;Cusworth et al., 2018). In contrast, there is a negligible contribution from biomass burning to O_3. However, there is a 15-20% contribution to O_3 from biogenic emission of VOCs, highlighting that O_3 production in Delhi is strongly VOC-limited.

3.3 Effect of the Diurnal Variation in Emissions

In order to investigate the competing influences of meteorology and human activities on the diurnal patterns of $PM_{2.5}$ and O_3 over Delhi, we test the effect of removing the diurnal variation in anthropogenic emissions ('noDiurnal', see Fig. 4c and 4d). Modelled $PM_{2.5}$ concentrations are very similar to the 'baseline' run during daytime when the PBL is well developed, with differences of less than 5%. This suggests that meteorological processes such as vertical mixing, advection and transport are the dominant factors controlling $PM_{2.5}$ in the daytime. In contrast, freshly emitted pollutants are trapped at night when the PBL is shallow, and concentrations are very sensitive to the emission flux, so that the diurnal pattern of emissions is the dominant factor at night. The $PM_{2.5}$ concentration is almost doubled in the early morning (03:00-09:00 LT, Fig. 4c) when the PBL is shallow and emissions in the 'noDiurnal' case are higher. There is also a large increase in NOx in the early morning (Fig. S4), which leads to greater depletion of O_3 (Fig. 4d). However, the concentration of O_3 is about 20-25% higher during the ozone peak hour in the afternoon in the 'noDiurnal' case, as the daytime NOx emissions are less (Fig. S4). This sensitivity test also highlights the VOC-limited nature of O_3 production in Delhi.

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

3.4 Sensitivity Analysis of Pollutants in Delhi

The importance of each anthropogenic emission sector to pollutant concentrations in Delhi is investigated using global sensitivity analysis and indicated by global sensitivity indices (SIs), as shown in Fig. 5. The sensitivity index is a measure of the contribution of the variation in pollutants from one emission sector to the total variation across all four sectors considered here. A larger SI indicates a larger influence from the corresponding sector to the modelled average surface $PM_{2.5}$ or O_3 over Delhi City Region (marked in Fig. 2) in this study.

The $PM_{2.5}$ concentration is most sensitive to emissions from the NCR region surrounding Delhi, with a sensitivity index higher than 50% most of time (Fig. 5a) and reaching 80-90% and ~60% during 03:00-07:00 LT and 12:00-17:00 LT, respectively. During the rush hours in the morning and evening, the sensitivity to NCR emissions is lower, while the sensitivity to Delhi traffic emissions increases by ~30%. Around 10:00 LT, local traffic emissions and emissions from NCR have a similar effect on $PM_{2.5}$. In contrast, local traffic emissions dominate the $PM_{2.5}$ in Delhi around 20:00 LT, with a sensitivity contribution of up to ~80%. This is caused by the collapse of the PBL in the evening rush hour at around 20:00 LT which enhances the sensitivity to fresh local emissions. Local traffic emissions contribute ~60% of primary $PM_{2.5}$ emission in Delhi (Fig. 6a), which remains concentrated in the PBL during rush hours. In contrast, the fully developed PBL in the daytime mixes air down from the free troposphere (Chen et al., 2016a), where regional transport of pollutants from NCR can be important. This could explain the second peak in the sensitivity to NCR emissions (50-60%) during the afternoon (Fig. 5a).

The O_3 in Delhi City Region is overwhelmingly dominated by local traffic emissions with a sensitivity index higher than 80% at night-time (Fig. 5b). Traffic contributes ~75% of total NOx emission in Delhi (Fig. 6b), and the shallow PBL during the night traps the NOx. This removes O_3 through chemical reaction in the absence of solar radiation. As the PBL develops in the morning, the sensitivity of O_3 to traffic decreases and the sensitivity to NCR emissions increases. The sensitivity to NCR emissions reaches its highest point (70%) when the PBL is fully developed around 15:00 LT. As discussed above, the downward mixing of air from the free troposphere and dilution of local emissions in the fully

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

developed PBL could be the reason for this. The O_3 peak coincides with the highest PBL at this time because photolysis and development of the PBL are both driven by solar radiation. The development of the PBL increases the contribution from regional transport, and precursors emitted from the NCR are one of the dominant contributors to the peak of O_3 in Delhi. In addition, it is noteworthy that the NOx-rich urban plume from Delhi has a substantial influence on O_3 in downwind regions across the NCR as well, as discussed in Text S3.

3.5 Mitigation Strategies

To demonstrate a framework for developing better mitigation strategies for addressing both $PM_{2.5}$ and O_3 pollution in Delhi, emission-sector based pollutant response surfaces are generated using Gaussian process emulation (Fig. 7). For local emissions in Delhi, we focus mainly on traffic and residential sectors here, because we find that power and industrial emissions have a more limited influence on $PM_{2.5}$ and O_3 concentrations in Delhi (Fig. 5). A range of different mitigation strategies are analysed, aiming at mitigating $PM_{2.5}$ pollution whilst minimizing the risk of O_3 increase.

We find that the responses of $PM_{2.5}$ and O_3 to each emission sector are nearly linear in Delhi. The response surfaces show that reducing local traffic emissions in Delhi leads to an efficient decrease in $PM_{2.5}$ loading (Fig. 7a) but increases O_3 greatly (Fig. 7b). Reducing local domestic emissions decreases $PM_{2.5}$ loading less than reducing traffic but without increasing O_3. The small impact on O_3 may be because domestic emissions are not a major source of NOx, contributing only 15% of that from traffic (Fig. 6). A 10-20% reduction in NOx is expected when reduce local domestic emissions by 50%; while, 35-45% reduction in NOx can be expected by a 50% reduction in local traffic emissions (Fig. S7). In addition, VOC is reduced more than NOx when controlling domestic emissions, as the VOC/NOx emission ratio (kg/kg) is 1.8 in contrast to a ratio of 0.4 for traffic emissions. Greater reduction of VOC suppresses the increase of O_3 in Delhi, which is a VOC-limited environment. A reduction in local traffic emissions alone of 50% could decrease Delhi $PM_{2.5}$ loading by 15-20%, but this would also increase O_3 by 20-25%. To prevent the side-effect of increasing O_3 by controls on traffic emissions, regional cooperation would be required to reduce emissions in the NCR region surrounding Delhi by 25-30%, which also permits a

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019


Atmospheric Chemistry and Physics Discussions

further reduction of $PM_{2.5}$ by 5-10% (Fig. 7c and 7d). This is consistent with a recent study showing that ~60% of $PM_{2.5}$ in Delhi originates from outside (Amann et al., 2017). The suggested regional joint mitigation with NCR surrounding Delhi is in line with a recent study for mitigating $PM_{2.5}$ in Beijing, which showed that regional coordination over the North China Plain could lead to a reduction in $PM_{2.5}$ of up to 40% in winter (Liu et al., 2016).

4. Summary

Previous studies have shown that emission controls focusing on mitigation of $PM_{2.5}$ may result in substantial increases of surface ozone over urban areas that are in VOC-limited photochemical environment. Comprehensive studies of mitigation strategies with respect to both $PM_{2.5}$ and O_3 are urgently required, but are limited in India. In this study, we demonstrate a numerical framework for informing emission-sector based mitigation strategies in Delhi that account for multiple pollutants.

By using Gaussian process emulation with an air quality model (WRF-Chem), we generate a computational efficient surrogate model for performing global sensitivity analysis and calculating emission-sector based pollutant response surfaces. These enable us to exhaustively investigate the impacts of different mitigation scenarios on $PM_{2.5}$ and O_3 in Delhi, which help decision makers choose better mitigation strategies. Global sensitivity analysis shows that pollutants originating from the National Capital Region (NCR) surrounding Delhi and local traffic emissions are the major contributors of $PM_{2.5}$ and O_3 in Delhi. They co-dominate the O_3 peak and $PM_{2.5}$ in Delhi during daytime, while the regional transport governs $PM_{2.5}$ during the night. Controlling local traffic emissions in Delhi would have the notable side effect of O_3 increases, at least in the pre-monsoon/summer (peak O_3 season) that we consider here. Our pollutant response surfaces suggest that joint coordinated emission controls with the NCR region surrounding Delhi would be required to minimize the risk of O_3 increases and to achieve a more ambitious reduction of $PM_{2.5}$. In the regional joint coordination, residential energy use could be a dominant emission sector over a large region in India (Conibear et al., 2018).

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

5. Discussion

The experiences of developed countries (Dooley, 2002;EPA, 2011) and recently in China (Huang et al., 2018a) show that regional joint coordination can be achieved by changing energy infrastructure (e.g., replacing fossil fuel by renewable energy and natural gas), desulphurisation and denitrification technologies, popularization of new energy vehicles, strict control of vehicle exhaust and reducing road and construction dust. Further studies with more detailed information on specific emission sectors and strategies for clean-technology development and popularization would permit deeper insight into air pollution mitigation approaches suitable for Delhi. These are needed to address both $PM_{2.5}$ which has a higher impact on public health (e.g., Huang et al., 2018a), and O_3 which greatly impacts regional ecology and agriculture (e.g., Avnery et al., 2011). A more comprehensive evaluation of the health and economic benefits of different mitigation strategies would greatly help Indian decision makers, and the framework we have demonstrated here would provide a strong foundation for this.

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

Author contributions

O. W. and Y. C. conceived the study. Y. C. performed the simulations and emulation, and processed and interpreted the results with help from Y. W. E. R. designed and built the Gaussian Process emulator. G. B. and S. K. S. provided the observations and SAFAR emission inventory. D. L., A. A., S. A.-N. and G. M. help pre-process the emission data and develop the emission toolkit. V. S. and R. S. S. provided useful discussion on the emission inventory. R. S. S. led the development of the PROMOTE project. Y. C. and O. W. wrote the manuscript with inputs from all co-authors.

Notes

The authors declare no competing financial interest.

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Data availability

NCEP FNL operational model global tropospheric analyses (ds083.2) were downloaded from https://rda.ucar.edu/data/ds083.2/, and sea surface temperature data were downloaded from http://polar.ncep.noaa.gov/sst/. ECMWF interim reanalyses (ERA-Interim) were downloaded from http://apps.ecmwf.int/datasets/data/interim-full-daily. MOZART-4 global model results are downloaded from http://www.acd.ucar.edu/wrf-chem/mozart.shtml. FINN biomass burning emissions dataset is downloaded from http://bai.acom.ucar.edu/Data/fire/. Toolkits for emission processing are available from https://github.com/douglowe/WRF_UoM_EMIT/releases/tag/v1.0 and https://github.com/douglowe/PROMOTE-emissions/releases/tag/v1.0 .

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

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Preprint. Discussion started: 1 August 2019

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Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

Table 1. Configuration of WRF-Chem

Physics	**WRF option**
Micro physics	Lin scheme (Lin, 1983)
Surface Layer	MM5 similarity
Boundary layer	YSU (Hong, 2006)
Cumulus	Grell 3D
Urban	3-category UCM
Shortwave radiation	Goddard shortwave (Chou, 1998)
Longwave radiation	Rapid Radiative Transfer Model
Chemistry and Aerosol	**Chem option**
Gas-phase mechanism	CBMZ (Zaveri and Peters, 1999)
Aerosol module	MOSAIC with 4 bins (~40 nm to 10 µm) (Zaveri et al., 2008)
Photolysis rate	Fast-J photolysis scheme (Wild et al., 2000)
Emissions Inventories	
Anthropogenic Emissions	SAFAR-2015 Delhi and EDGAR-HTAP v2.2
Biogenic Emissions	MEGAN (Guenther et al., 2006)
Biomass Burning Emissions	FINN (Wiedinmyer et al., 2011)

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

Table 2. Map of NMVOC from EDGAR emission to CBMZ scheme.

EDGAR Name	Description	CBMZ [mol]
VOC1	Alcohols	20% CH_3OH
		80% C_2H_5OH
VOC2	Ethane	C_2H_6
VOC3	Propane	PAR*3
VOC4	Butane	PAR*4
VOC5	Pentane	PAR*5
VOC6	Hexanes + other Alkanes	PAR*6
VOC7	Ethene	ETH
VOC8	Propene	OLET+PAR
VOC9	Ethyne	PAR*2
VOC10	Isoprene	ISOP
VOC11	Monoterpenes	ISOP*2
VOC12	Other Alkenes	OLEI*0.5+OLET*0.5+PAR*2
VOC13	Benzene	TOL-PAR
VOC14	Toluene	TOL
VOC15	Xylenes	XYL
VOC16	Trimethylbenzenes	XYL+PAR
VOC17	Other Aromatics	XYL+PAR
VOC18	Esters	RCOOH
VOC19	Ethers	20% CH_3OH
		80% C_2H_5OH
VOC21	Formaldehyde	HCHO
VOC22	Other Aldehydes	ALD2
VOC23	Ketones	60% KET
		40% KET+PAR
VOC24	Alkanoic Acids	RCOOH

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

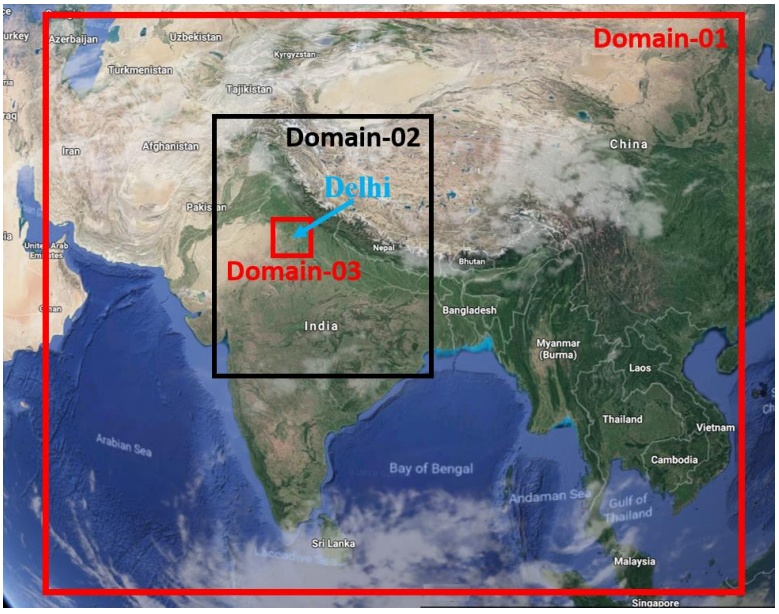


Figure 1. Map of simulation domains, modified from © Google Earth.

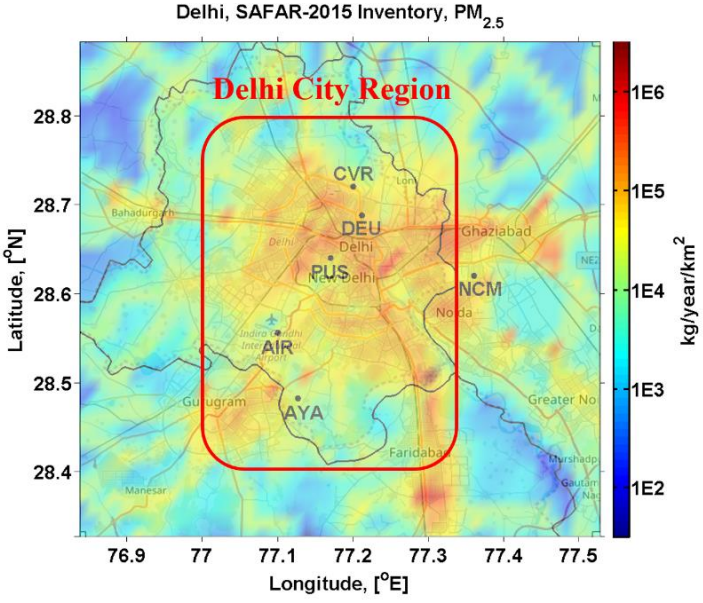


Figure 2. SAFAR inventory of total $PM_{2.5}$ emission. The locations of measurement sites over Delhi are marked by black dots, and the Delhi City Region is marked by a red box.

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

Atmospheric Chemistry and Physics Discussions

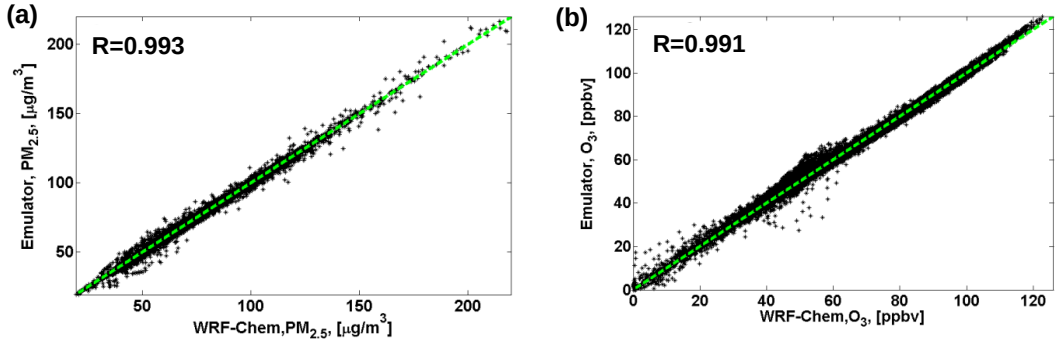


Figure 3. Validation of Gaussian process emulator with WRF-Chem model. (**a**) $PM_{2.5}$; (**b**) O_3. The green dashed line indicates the 1:1 line.

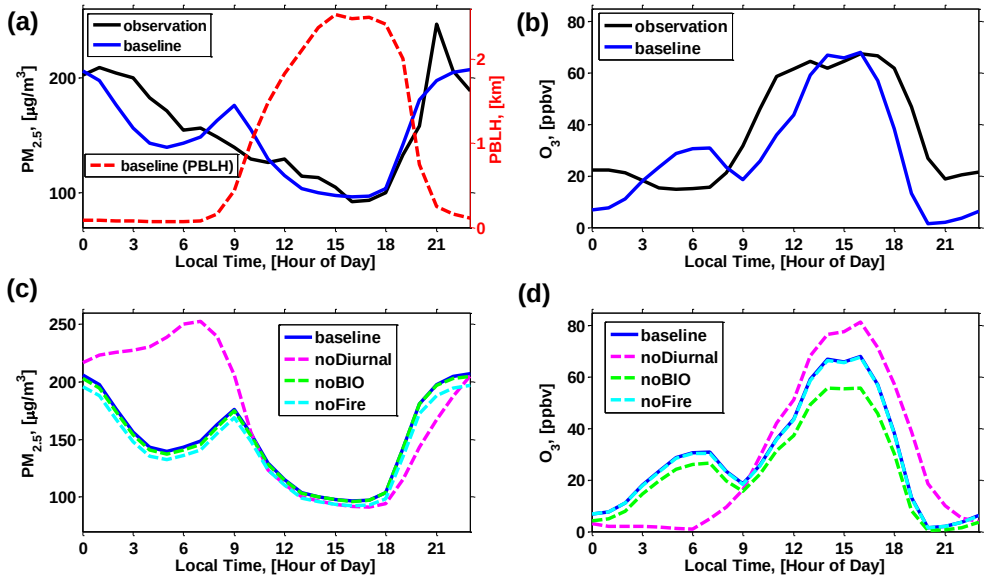
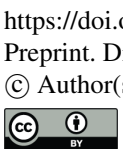


Figure 4. Average diurnal patterns of pollutants during the 2-15 May 2015 simulation period. (**a**) Modelled and observed $PM_{2.5}$ and model PBL height (PBLH); (**b**) O_3; (**c**) results of sensitivity studies for $PM_{2.5}$; (**d**) results of sensitivity studies for O_3. The left panels (a, c) are for site CVR, and the right panels (b, d) are for site AIR (marked in Fig. 2). The sensitivity runs 'noFire' and 'noBIO' show model results without biomass burning and biogenic emissions, respectively; and 'noDiurnal' show model results with constant anthropogenic emissions rates throughout the day.

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019



Atmospheric Chemistry and Physics Discussions

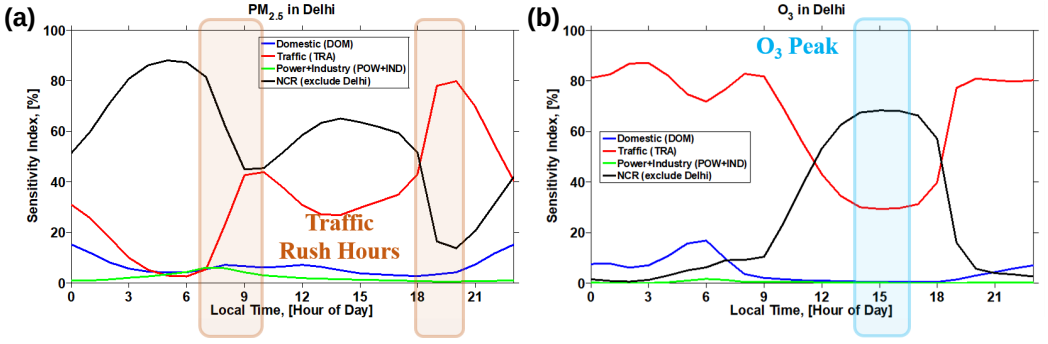


Figure 5. Averaged diurnal pattern of global sensitivity indices during the 2-15 May simulation period. (**a**) $PM_{2.5}$; (**b**) O_3. The $PM_{2.5}$ and O_3 results are averaged over Delhi City Region (marked with red box in Fig. 2). The morning and evening rush hours and the period of peak ozone are marked with the boxes to highlight the notable changes in contribution from each emission sector.

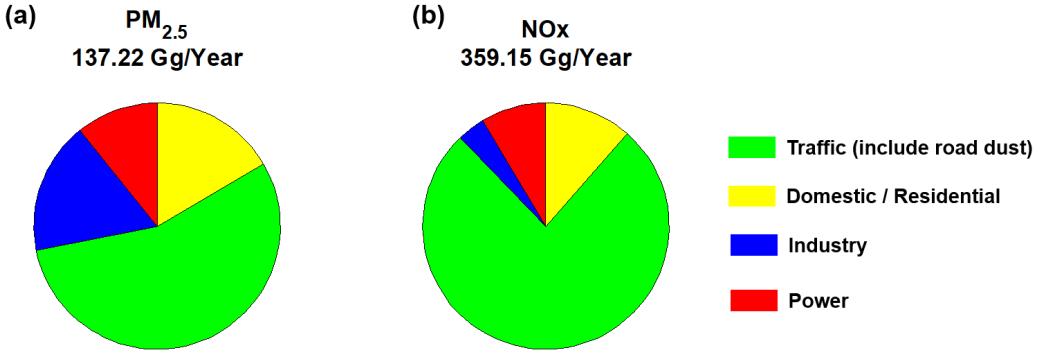


Figure 6. Annual emission of different sectors in Delhi from SAFAR inventory. (a) $PM_{2.5}$; (b) NOx.

https://doi.org/10.5194/acp-2019-618
Preprint. Discussion started: 1 August 2019

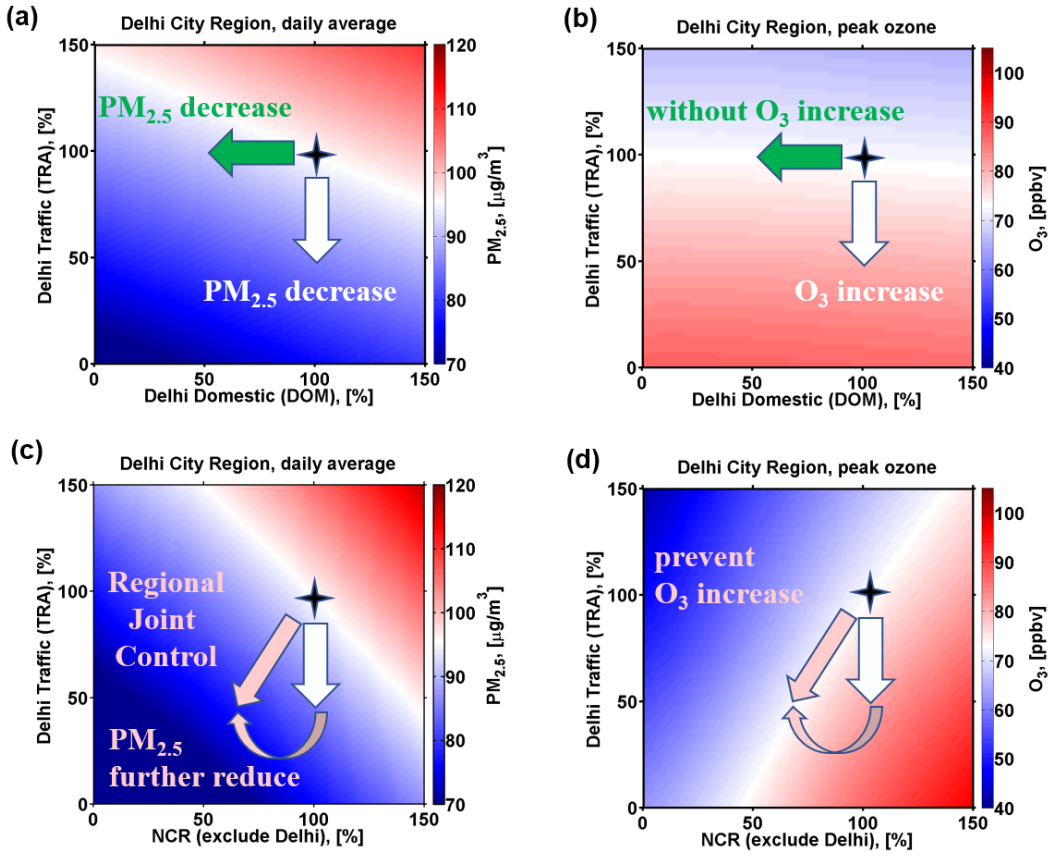


Figure 7. Response surfaces for $PM_{2.5}$ and ozone concentrations over Delhi City Region, averaged over 2-15 May 2018. (**a**) Daily average of $PM_{2.5}$ concentrations as a function of local traffic and domestic emissions in Delhi; (**b**) peak hourly ozone concentrations as a function of local traffic and domestic emissions in Delhi; (**c**) daily average of $PM_{2.5}$ concentrations as a function of local traffic emissions in Delhi and emissions in NCR region surrounding Delhi; and (**d**) peak hourly ozone concentrations as a function of local traffic emissions in Delhi and emissions in NCR region surrounding Delhi. The star indicates current conditions and the arrows show the effect of possible emission controls.