

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

Ying Chen^{1,2*}, Oliver Wild^{1,2}, Edmund Ryan^{1,9}, Saroj Kumar Sahu⁴, Douglas Lowe⁵,
Scott Archer-Nicholls⁶, Yu Wang⁵, Gordon McFiggans⁵, Tabish Ansari¹, Vikas Singh⁷,
5 Ranjeet S. Sokhi⁸, Alex Archibald⁶, Gufran Beig³

¹Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK

²Data Science Institute, Lancaster University, Lancaster, LA1 4YW, UK

³Indian Institute of Tropical Meteorology, Pune, India

⁴Environmental Science, Dept. of Botany, Utkal University, Bhubaneswar, India

10 ⁵Centre for Atmospheric Sciences, School of Earth, Atmospheric and Environmental
Sciences, University of Manchester, Manchester, UK

⁶NCAS-Climate, Department of Chemistry, University of Cambridge, Cambridge, UK

⁷National Atmospheric Research Laboratory, Gadanki, AP, India

15 ⁸Centre for Atmospheric and Climate Physics Research, University of Hertfordshire,
Hatfield, Hertfordshire, UK

⁹School of Mathematics, University of Manchester, Manchester, UK

Correspondence to: Ying Chen (y.chen65@lancaster.ac.uk)

Abstract:

Fine particulate matter (PM_{2.5}) and surface ozone (O₃) 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 O₃. Here, we demonstrate a numerical framework for investigating the impacts of mitigation strategies on both PM_{2.5} and O₃ 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 O₃ 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 O₃ in Delhi. They together govern the O₃ 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 O₃. 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 O₃ increase. This study provides scientific evidence to support the need for joint coordination of controls on local and regional scales to achieve effective reduction in PM_{2.5} whilst minimizing the risk of O₃ increase in Delhi.

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, 2016a, b). In addition to the local impacts, the Indian monsoon can
45 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 and it has interactions with climate and ecosystems world-wide (Menon et al., 2002;Gao et al., 2019).

PM_{2.5} (particulate matter with an aerodynamic diameter of less than 2.5 µm) is a major air
50 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
55 annual mortalities across India (Conibear et al., 2018). Previous studies reported an annual averaged PM_{2.5} loading of 110-140 µg/m³ in Delhi during 2015-2018, leading to ~10,000 premature deaths per year in the city (Chen et al., 2019; Chowdhury and Dey, 2016; WHO, 2016a). 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
60 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 (O₃), 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

65 cardiovascular diseases are increased from short-term exposure to high ambient O₃ 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 (NO_x) is
the main source of surface ozone. Rapid economic development in India has greatly increased
the emissions of these O₃ precursors (Duncan et al., 2016), leading to significant increases in
70 O₃ especially during the pre-monsoon period (Ghude et al., 2008). Hourly maximum O₃
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
75 dimming effect of aerosols and removal of hydroperoxy radicals are reduced, facilitating O₃
production (Huang et al., 2018b;Li et al., 2018;Hollaway et al., 2019). Furthermore, co-
reduction of NO_x and PM_{2.5} emissions may increase O₃ in cities where O₃ 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 (Silver et al., 2018),
80 Beijing (Wu et al., 2015;Liu et al., 2017;Chen et al., 2018) 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 O₃ as a side-effect of emission controls focused on PM_{2.5}. Therefore, studies
of mitigation strategies that target both PM_{2.5} and O₃ are urgently needed (Chen et al., 2018),
particularly as urban air pollution in India has been much less well studied than in many other
85 countries.

To investigate the impacts of mitigation strategies with respect to both PM_{2.5} and O₃, we
demonstrate a framework for generating emission-sector based pollutant response surfaces
using Gaussian process emulation (O'Hagan and West, 2009;O'Hagan, 2006). The response

surfaces describe that how the pollutants, i.e., PM_{2.5} and O₃, will respond to the changes in
90 emissions from different sectors. We conduct global sensitivity analysis to identify the
dominant emission sectors controlling PM_{2.5} and O₃, and then generate sector based response
surfaces to quantify the impacts on PM_{2.5} and O₃ 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.,
95 1999). Conventionally, chemical transport models (CTMs) 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) or computational efficient
100 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.,
105 2016; Lee et al., 2012). Here, we use surrogate model to explore the sensitivity of PM_{2.5} and O₃
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
110 impacts of mitigation scenarios on PM_{2.5} and O₃ 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 O₃ increase.

2. Materials and Methods

115 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
120 reduction in crop yields (Ghude et al., 2014) from the exposure to PM_{2.5} and O₃ over India.

In this study, we focus on the hot and dry pre-monsoon period in Delhi, when average temperatures are around 32 °C and relative humidity (RH) is about 35% (Ojha et al., 2012). O₃ 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
125 significantly to particulate matter in Delhi (Kumar et al., 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), when Delhi was not significantly influenced by dust storms 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-
130 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
135 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. No precipitation was recorded in Delhi during the simulation period.

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 ECMWF reanalysis dataset (ERA-Interim) assimilates observations with a number of nearly 10^7 per day (Dee et al., 2011), and is used for grid nudging, initial and boundary conditions for WRF-Chem at horizontal and temporal resolutions of $0.75^\circ \times 0.75^\circ$ and 6 hours, respectively. 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 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

160 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 $0.1^\circ \times 0.1^\circ$ resolution is used to represent anthropogenic emissions
apart from over Delhi, where they are represented by a high-resolution monthly inventory for
165 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 PM₁₀, PM_{2.5}, black carbon, organic
carbon, NO_x, CO, SO₂ and NMVOC (non-methane volatile organic compounds) from five
170 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
175 (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
180 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
carbons (OLET), internal olefin carbons (OLEI) and PAR on a molar ratio of 1:1:4 following

185 (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

190 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}, O₃ and NO_x 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).
195 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

200 We perform global sensitivity analysis (GSA) (Iooss and Lemaître, 2015) to quantify the sensitivity of modelled outputs (PM_{2.5} and O₃ for this study) to changes in the model inputs, which for this study are emissions from the different emission sectors. One-at-a-time sensitivity analysis is a common way of calculating model sensitivity and involves varying a single model input while the other inputs are fixed at nominal values, e.g., Wild (2007). While one-at-a-time
205 approach is relatively easy to implement, it assumes that the model response to different inputs is independent and this can lead to biased results (Saltelli et al., 1999;Pisoni et al., 2018;Carslaw et al., 2013). GSA overcomes the problems of the one-at-a-time approach by averaging over the other inputs rather than fixing them at specific values. This allows

calculation of first-order sensitivity indices (SIs) for each variable, corresponding to the i^{th} input variable and the j^{th} output point, is given by the Eq. 1 (Ryan et al., 2018).

$$SI_{i,j} = \frac{\text{Var}[E(y_j | x_i)]}{\text{Var}(y_j)} \times 100\% \quad (1)$$

where x_i is the i^{th} element of the input; and y_j is the j^{th} element of the output. The ‘ $E(\bullet)$ ’ and ‘ $\text{Var}(\bullet)$ ’ denote the mathematical function that calculate the expectation and variance, respectively. The simplest way of computing $SI_{i,j}$ is by brute force, but this is also the most computationally intensive (Ryan et al., 2018).

The extended Fourier Amplitude Sensitivity Test (eFAST), first developed by Saltelli et al. (1999), is a commonly used approach to perform GSA and calculate SIs and is adopted in this study because of its high efficiency. A basic overview and detailed equations of the eFAST method are given in the section 2.2.2 of Ryan et al. (2018). A challenge to using eFAST is that it typically requires thousands of model runs. To overcome this, we employ a computationally cheaper surrogate model in place of our expensive simulation model WRF-Chem. A surrogate model is a simple model (usually statistical) which can map the inputs to the outputs of the simulation model with sufficiently good accuracy given the same inputs. In this study, we choose a type of surrogate model called a Gaussian process emulator, which works like a function for multi-dimensional interpolation and has been used extensively in many areas of applied science (Carslaw et al., 2013; Koehler and Owen, 1996; Queipo et al., 2005; Vanuytrecht and Willems, 2014; vu et al., 2015; Degroote et al., 2012) and uncertainty assessment of atmospheric models (Lee et al., 2016; Lee et al., 2012; Lee et al., 2011). Gaussian process emulators typically require a relatively small number of runs of the computational-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. For a basic overview

of a Gaussian process emulator see O’Hagan (2006), detailed introduction and equations are also given in the section 2.3 of Ryan et al. (2018). Before using the emulator in place of the WRF-Chem model to carry out the thousands of model runs required for GSA, we train the
235 emulator using a relatively small number of WRF-Chem model runs. Following previous studies (Carslaw et al., 2013; Lee et al., 2016), a Maximin Latin hypercube space-filling design is employed to select 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
240 Latin hypercube random designs to find the optimal one where the parameter space is filled most effectively. This ensures that the sets of inputs chosen cover as large a fraction of the input space as possible. Full details (including R codes) of how to generate the Gaussian process emulator, eFAST method and GSA can be found in Ryan et al. (2018).

In this study, we focus on a limited number of the emission sectors to demonstrate the
245 effectiveness 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,
250 3.1 and 8.5 Gg/month in May 2015, respectively. The Gaussian process emulator is trained using 20 executions of the WRF-Chem model, 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 O₃ 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
255 cluster. The accuracy of the emulator as a surrogate of WRF-Chem model is evaluated using a ‘leave-one-out’ cross-validation (Bastos and O’Hagan, 2009). This involves training the

emulator using 19 out of the 20 sets of inputs/outputs from the WRF-Chem model runs and then evaluating the emulator against the 20th simulation. This process is carried out for each of the 20 sets of inputs/outputs. Given that the output space is multi-dimensional (i.e. modelled O₃ and PM_{2.5} varied spatially and in time), the validation of the emulator by comparing 10,000 (random-samples varied spatially and in time) of emulator output values against the corresponding output values of the WRF-Chem model. The emulator validation plot is shown in Fig. 3. Modelled and emulated O₃ and PM_{2.5} lie very close to the 1:1 line with R² values of more than 95% as shown in Fig. 3, indicating that the emulation provides an accurate representation of the input-output relationship of the WRF-Chem 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, 2006b, a; Zhao et al., 2017; Xing et al., 2017). Here, we analyse the responses of PM_{2.5} and O₃ to changes in emissions from each sector of between 0% 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 O₃ 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 O₃ in Delhi. We then conduct a global sensitivity analysis, using the eFAST method (see section 2.3) along with Gaussian process emulation, to determine the sensitivity of modelled O₃ 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 O₃ increase.

3. Results and Discussion

285 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%,
290 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
295 (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 information. Detailed studies of traffic emissions and their variation in Delhi would help improve these model simulations.

300 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

305 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).

310 The model well captures the peak O₃ with a bias of less than 5%, although it underestimates O₃ during night-time (Fig. 4b). In general, the diurnal pattern and magnitude of O₃ 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 NO_x is overestimated by a factor of 2-3 at night (Fig. S4), and the
315 excess NO depletes O₃. This is indicated by the frequency distribution of O₃ and NO_x (Fig. S5), where the median values of observed O₃ and NO_x are matched well by the model. However, the higher peaks of modelled NO_x concentration lower the modelled O₃ levels, indicating that Delhi is in VOC-limited photochemical regime. Similar results are found at AYA (Fig. S6). The larger underestimation of O₃ at NCM (Fig. S5d, industrial environment
320 site) suggests that NO_x 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 O₃ in Delhi during simulation period, we investigate the contributions from other factors (biomass burning and biogenic emissions). We turn off these sources in the WRF-Chem
325 simulation and find that there is a negligible contribution from 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 represented well 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-
330 monsoon results; as described above, 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 O₃. However, there is a
335 15-20% contribution to O₃ from biogenic emission of VOCs, highlighting that O₃ 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 O₃ over Delhi, we test the effect of removing the diurnal
340 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,
345 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 NO_x in the early morning (Fig.
S4), which leads to greater depletion of O₃ (Fig. 4d). However, the concentration of O₃ is about
350 20-25% higher during the ozone peak hour in the afternoon in the 'noDiurnal' case, as the
daytime NO_x emissions are less (Fig. S4). This sensitivity test also highlights the VOC-limited
nature of O₃ production in Delhi.

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 variation of 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), when O_3 and traffic emissions are anti-correlated. Traffic contributes ~75% of total NO_x emission in Delhi (Fig.

6b), and the shallow PBL during the night traps the NO_x. This removes O₃ through chemical reaction in the absence of solar radiation. As the PBL develops in the morning, the sensitivity of O₃ 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 developed PBL could be the reason for this. The O₃ 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 O₃ in Delhi. NO_x, mainly originating from traffic emissions, is underestimated by ~30% during the O₃ peak period (Fig. S4). This uncertainty can propagate into the Gaussian process emulator and could lead to underestimation of the influence of traffic on peak O₃, but is not expected to change the nature of our conclusion about the predominance of regional transport and local traffic emissions. In addition, it is noteworthy that the NO_x-rich urban plume from Delhi has a substantial influence on O₃ 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 O₃ 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 O₃ 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 O₃ 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 NO_x , contributing only 15% of that from traffic (Fig. 6). A 10-20% reduction in NO_x is expected when reduce local domestic emissions by 50%; but a 35-45% reduction is seen for a 50% reduction in local traffic emissions (Fig. S7). In addition, VOC is reduced more than NO_x when controlling domestic emissions, as the VOC/ NO_x 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%. We note that our model may underestimate the influence of traffic emissions on O_3 to some extent as described above (section 3.4), suggesting that the ozone increase could be stronger than we predict. 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 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). We test this by performing an additional run with WRF-Chem using emission reductions of 50% and 30% for sectors of local traffic and the surrounding NCR region, respectively. We compare the WRF-Chem results of the additional run and the base case (without change of emissions) against the corresponding results from Gaussian process emulator (Fig. S8). We find that the $PM_{2.5}$ and O_3 results from the model runs lie within 5% of those estimated with the emulator and with R^2 higher than 95%, demonstrating the high quality of the emulation approach adopted here and underlining its deeper value for identifying

mitigation approaches. 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).

430

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 O₃ 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 O₃ 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 O₃ in Delhi. They co-dominate the O₃ peak and PM_{2.5} in Delhi during daytime, while the regional transport governs PM_{2.5} during the night, in line with a recent study showing that ~60% of PM_{2.5} in Delhi originates from outside (Amann et al., 2017). Controlling local traffic emissions in Delhi would have the notable side effect of increasing O₃, at least in the pre-monsoon/summer period (peak O₃ season) that we consider here. This is in line with recent increases in O₃ seen in China (Silver et al., 2018;Li et

450 al., 2018). The Chinese experience suggests that regional joint coordination is required to
effectively mitigate PM_{2.5} pollution in Beijing (Liu et al., 2016). Our pollutant response
surfaces go one step further and suggest that joint coordinated emission controls with the NCR
region surrounding Delhi would be required to not only achieve a more ambitious reduction of
PM_{2.5} but also to minimize the risk of O₃ increases. In the regional joint coordination,
455 residential energy use could be a dominant emission sector over a large region in India
(Conibear et al., 2018).

5. Discussion

The experiences of developed countries (Dooley, 2002; EPA, 2011) and recently in China
460 (Huang et al., 2018a; Wang et al., 2019) 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
465 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 O₃ 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
470 decision makers, and the framework we have demonstrated here would provide a strong
foundation for this.

Author contributions

OW and YC conceived the study. YC performed the simulations and emulation, and processed and
475 interpreted the results with help from YW. ER designed and built the Gaussian Process emulator. GB and
SKS provided the observations and SAFAR emission inventory. DL., AA, SAN. and GM help pre-process
the emission data and develop the emission toolkit. VS and RSS provided useful discussion on the emission
inventory. RSS led the development of the PROMOTE project. YC and OW wrote the manuscript with
inputs from all co-authors.

480 Notes

The authors declare no competing financial interest.

Acknowledgments

This work was supported by the NERC/MOES/Newton Fund supported PROMOTE project (grant
number NE/P016405/1 and NE/P016480/1). The work of E. Ryan was supported by the NERC (grant
485 number NE/N003411/1). The Indian Institute of Tropical Meteorology, Pune, is supported by the Ministry
of Earth Science, Government of India. The observations and high-resolution emission inventory are
provided by the SAFAR project under MoES (<http://safar.tropmet.res.in>). The authors appreciate the efforts
of the entire team involved in PROMOTE and SAFAR projects. The paper is based on interpretation of
scientific results and in no way reflect the viewpoint of the funding agency.

490 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
495 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://github.com/douglowe/PROMOTE-emissions/releases/tag/v1.0).

References:

- Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., and Winiwarter, W.: Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, *Environmental Modelling & Software*, 26, 1489-1501, <https://doi.org/10.1016/j.envsoft.2011.07.012>, 2011.
- Amann, M., Purohit, P., Bhanarkar, A. D., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Kiesewetter, G., Klimont, Z., Liu, J., Majumdar, D., Nguyen, B., Rafaj, P., Rao, P. S., Sander, R., Schöpp, W., Srivastava, A., and Vardhan, B. H.: Managing future air quality in megacities: A case study for Delhi, *Atmospheric Environment*, 161, 99-111, <https://doi.org/10.1016/j.atmosenv.2017.04.041>, 2017.
- Ashworth, K., Wild, O., and Hewitt, C. N.: Impacts of biofuel cultivation on mortality and crop yields, *Nature Clim. Change*, 3, 492-496, 10.1038/nclimate1788, 2013.
- Aslam, M. Y., Krishna, K. R., Beig, G., Tinmaker, M. I. R., and Chate, D. M.: Seasonal Variation of Urban Heat Island and Its Impact on Air-Quality Using SAFAR Observations at Delhi, India, *American Journal of Climate Change*, Vol.06No.02, 12, 10.4236/ajcc.2017.62015, 2017.
- Avnery, S., Mauzerall, D. L., Liu, J., and Horowitz, L. W.: Global crop yield reductions due to surface ozone exposure: 2. Year 2030 potential crop production losses and economic damage under two scenarios of O₃ pollution, *Atmospheric Environment*, 45, 2297-2309, <https://doi.org/10.1016/j.atmosenv.2011.01.002>, 2011.
- Bastos, L. S., and O'Hagan, A.: Diagnostics for Gaussian Process Emulators, *Technometrics*, 51, 425-438, 10.1198/TECH.2009.08019, 2009.
- Beig, G., Chate, D. M., Ghude, S. D., Ali, K., Satpute, T., Sahu, S. K., Parkhi, N., and Trimbake, H. K.: Evaluating population exposure to environmental pollutants during Deepavali fireworks displays using air quality measurements of the SAFAR network, *Chemosphere*, 92, 116-124, <http://dx.doi.org/10.1016/j.chemosphere.2013.02.043>, 2013.
- Carlsaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., Mann, G. W., Spracklen, D. V., Woodhouse, M. T., Regayre, L. A., and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect forcing, *Nature*, 503, 67, 10.1038/nature12674, 2013.
- 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.
- Chen, L., Guo, B., Huang, J., He, J., Wang, H., Zhang, S., and Chen, S. X.: Assessing air-quality in Beijing-Tianjin-Hebei region: The method and mixed tales of PM_{2.5} and O₃, *Atmospheric Environment*, <https://doi.org/10.1016/j.atmosenv.2018.08.047>, 2018.
- Chen, Y., Cheng, Y., Ma, N., Wolke, R., Nordmann, S., Schüttauf, S., Ran, L., Wehner, B., Birmili, W., van der Gon, H. A. C. D., Mu, Q., Barthel, S., Spindler, G., Stieger, B., Müller, K., Zheng, G. J., Pöschl, U., Su, H., and Wiedensohler, A.: Sea salt emission, transport and influence on size-segregated nitrate simulation: a case study in northwestern Europe by WRF-Chem, *Atmos. Chem. Phys.*, 16, 12081-12097, 10.5194/acp-16-12081-2016, 2016a.
- Chen, Y., Cheng, Y. F., Nordmann, S., Birmili, W., Denier van der Gon, H. A. C., Ma, N., Wolke, R., Wehner, B., Sun, J., Spindler, G., Mu, Q., Pöschl, U., Su, H., and Wiedensohler, A.: Evaluation of the size segregation of elemental carbon (EC) emission in Europe: influence on the simulation of EC long-range transportation, *Atmos. Chem. Phys.*, 16, 1823-1835, 10.5194/acp-16-1823-2016, 2016b.
- Chen Y, Wild O., Conibear L., Ran L., He J., Wang L., Wang Y.: Local Characteristics of and Exposure to Fine Particulate Matter (PM_{2.5}) in Four Indian Megacities, *Atmospheric Environment*, 10.1016/j.aeaoa.2019.100052, 2019
- Chowdhury, S., and Dey, S.: Cause-specific premature death from ambient PM_{2.5} exposure in India: Estimate adjusted for baseline mortality, *Environment International*, 91, 283-290, <https://doi.org/10.1016/j.envint.2016.03.004>, 2016.
- Chowdhury, S., Dey, S., Tripathi, S. N., Beig, G., Mishra, A. K., and Sharma, S.: "Traffic intervention" policy fails to mitigate air pollution in megacity Delhi, *Environmental Science & Policy*, 74, 8-13, <http://dx.doi.org/10.1016/j.envsci.2017.04.018>, 2017.
- Conibear, L., Butt, E. W., Knote, C., Arnold, S. R., and Spracklen, D. V.: Residential energy use emissions dominate health impacts from exposure to ambient particulate matter in India, *Nature Communications*, 9, 617, 10.1038/s41467-018-02986-7, 2018.
- Cusworth, D. H., Mickley, L. J., Sulprizio, M. P., Liu, T., Marlier, M. E., DeFries, R. S., Guttikunda, S. K., and Gupta, P.: Quantifying the influence of agricultural fires in northwest India on urban air pollution in Delhi, India, *Environmental Research Letters*, 13, 044018, 10.1088/1748-9326/aab303, 2018.
- Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol,

- C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Quarterly Journal of the Royal Meteorological Society*, 137, 553-597, 10.1002/qj.828, 2011.
- Degroote, J., Couckuyt, I., Vierendeels, J., Segers, P., and Dhaene, T.: Inverse modelling of an aneurysm's stiffness using surrogate-based optimization and fluid-structure interaction simulations, *Structural and Multidisciplinary Optimization*, 46, 457-469, 10.1007/s00158-011-0751-7, 2012.
- Denier van der Gon, H. A. C., Hendriks, C., Kuenen, J., Segers, A., and Visschedijk, A.: TNO Report: Description of current temporal emission patterns and sensitivity of predicted AQ for temporal emission patterns 2011.
- Dooley, E.: Clearing the Air over the London Fog, *Environmental Health Perspectives*, 110, A748-A749, 2002.
- Duncan, B. N., Lamsal, L. N., Thompson, A. M., Yoshida, Y., Lu, Z., Streets, D. G., Hurwitz, M. M., and Pickering, K. E.: A space-based, high-resolution view of notable changes in urban NO_x pollution around the world (2005–2014), *Journal of Geophysical Research: Atmospheres*, 121, 976-996, doi:10.1002/2015JD024121, 2016.
- EPA: Technical support document for the proposed mobile source air toxics rule: ozone modeling, Office of Air Quality Planning and Standards, US Environmental Protection Agency, Research Triangle Park, NC, US, 49, (last access: 01 Oct. 2018), 2006a.
- EPA: Technical support document for the proposed PM NAAQS rule: Response Surface Modeling, Office of Air Quality Planning and Standards, US Environmental Protection Agency, Research Triangle Park, NC, US, 48, (last access: 01 Oct. 2018), 2006b.
- EPA: Benefits and costs of the Clean Air Act 1990-2020, the second prospective study, <https://www.epa.gov/clean-air-act-overview/benefitsand-costs-clean-air-act-1990-2020-second-prospective-study>, (last access: 20 Aug. 2018). 2011.
- Fleming, Z. L., Doherty, R. M., Von Schneidmesser, E., Malley, C. S., Cooper, O. R., Pinto, J. P., Colette, A., Xu, X., Simpson, D., Schultz, M. G., Lefohn, A. S., Hamad, S., Moolla, R., Solberg, S., and Feng, Z.: Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health, *Elem Sci. Anth*, 6, 12, 2018.
- Gao, M., Guttikunda, S. K., Carmichael, G. R., Wang, Y., Liu, Z., Stanier, C. O., Saide, P. E., and Yu, M.: Health impacts and economic losses assessment of the 2013 severe haze event in Beijing area, *Science of The Total Environment*, 511, 553-561, <https://doi.org/10.1016/j.scitotenv.2015.01.005>, 2015.
- Gao, M., Beig, G., Song, S., Zhang, H., Hu, J., Ying, Q., Liang, F., Liu, Y., Wang, H., Lu, X., Zhu, T., Carmichael, G. R., Nielsen, C. P., and McElroy, M. B.: The impact of power generation emissions on ambient PM_{2.5} pollution and human health in China and India, *Environment International*, 121, 250-259, <https://doi.org/10.1016/j.envint.2018.09.015>, 2018.
- Gao, M., Sherman, P., Song, S., Yu, Y., Wu, Z., and McElroy, M. B.: Seasonal prediction of Indian wintertime aerosol pollution using the ocean memory effect, *Science Advances*, 5, eaav4157, 10.1126/sciadv.aav4157, 2019.
- Ghude, S. D., Jain, S. L., Arya, B. C., Beig, G., Ahammed, Y. N., Kumar, A., and Tyagi, B.: Ozone in ambient air at a tropical megacity, Delhi: characteristics, trends and cumulative ozone exposure indices, *J Atmos Chem*, 60, 237-252, 10.1007/s10874-009-9119-4, 2008.
- Ghude, S. D., Jena, C., Chate, D. M., Beig, G., Pfister, G. G., Kumar, R., and Ramanathan, V.: Reductions in India's crop yield due to ozone, *Geophysical Research Letters*, 41, 5685-5691, 10.1002/2014GL060930, 2014.
- Ghude, S. D., Chate, D. M., Jena, C., Beig, G., Kumar, R., Barth, M. C., Pfister, G. G., Fadnavis, S., and Pithani, P.: Premature mortality in India due to PM_{2.5} and ozone exposure, *Geophysical Research Letters*, 43, 4650-4658, 10.1002/2016GL068949, 2016.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled "online" chemistry within the WRF model, *Atmospheric Environment*, 39, 6957-6975, <http://dx.doi.org/10.1016/j.atmosenv.2005.04.027>, 2005.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181-3210, 10.5194/acp-6-3181-2006, 2006.
- Gupta, M., and Mohan, M.: Validation of WRF/Chem model and sensitivity of chemical mechanisms to ozone simulation over megacity Delhi, *Atmospheric Environment*, 122, 220-229, <http://dx.doi.org/10.1016/j.atmosenv.2015.09.039>, 2015.
- Hollaway, M., Wild, O., Yang, T., Sun, Y., Xu, W., Xie, C., Whalley, L., Slater, E., Heard, D., and Liu, D.: Photochemical impacts of haze pollution in an urban environment, *Atmos. Chem. Phys. Discuss.*, 2019, 1-26, 10.5194/acp-2019-29, 2019.

- Huang, G., Brook, R., Crippa, M., Janssens-Maenhout, G., Schieberle, C., Dore, C., Guizzardi, D., Muntean, M., Schaaf, E., and Friedrich, R.: Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970–2012, *Atmos. Chem. Phys.*, 17, 7683-7701, 10.5194/acp-17-7683-2017, 2017.
- Huang, J., Pan, X., Guo, X., and Li, G.: Health impact of China's Air Pollution Prevention and Control Action Plan: an analysis of national air quality monitoring and mortality data, *The Lancet Planetary Health*, 2, e313-e323, 10.1016/S2542-5196(18)30141-4, 2018a.
- Huang, X., Wang, Z., and Ding, A.: Impact of Aerosol-PBL Interaction on Haze Pollution: Multiyear Observational Evidences in North China, *Geophysical Research Letters*, 45, 8596-8603, 10.1029/2018GL079239, 2018b.
- Iooss, B., and Lemaître, P.: A review on global sensitivity analysis methods. In: Meloni, C., Dellino, G. (Eds.), *Uncertainty Management in Simulation Optimization of Complex Systems: Algorithms and Applications*. Springer, 2015.
- Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, *Atmos. Chem. Phys.*, 15, 11411-11432, 10.5194/acp-15-11411-2015, 2015.
- Jethva, H., Chand, D., Torres, O., Gupta, P., Lyapustin, A., and Patadia, F.: Agricultural Burning and Air Quality over Northern India: A Synergistic Analysis using NASA's A-train Satellite Data and Ground Measurements, *Aerosol and Air Quality Research*, 18, 1756-1773, 10.4209/aaqr.2017.12.0583, 2018.
- Khare, M., Gargava, P., and Khan, A. A.: Effect of PM_{2.5} chemical constituents on atmospheric visibility impairment AU - Khanna, Isha, *Journal of the Air & Waste Management Association*, 68, 430-437, 10.1080/10962247.2018.1425772, 2018.
- Koehler, J. R., and Owen, A. B.: 9 Computer experiments, in: *Handbook of Statistics*, Elsevier, 261-308, 1996.
- Kumar, R., Barth, M. C., Madronich, S., Naja, M., Carmichael, G. R., Pfister, G. G., Knote, C., Brasseur, G. P., Ojha, N., and Sarangi, T.: Effects of dust aerosols on tropospheric chemistry during a typical pre-monsoon season dust storm in northern India, *Atmos. Chem. Phys.*, 14, 6813-6834, 10.5194/acp-14-6813-2014, 2014a.
- Kumar, R., Barth, M. C., Pfister, G. G., Naja, M., and Brasseur, G. P.: WRF-Chem simulations of a typical pre-monsoon dust storm in northern India: influences on aerosol optical properties and radiation budget, *Atmos. Chem. Phys.*, 14, 2431-2446, 10.5194/acp-14-2431-2014, 2014b.
- Kumar, R., Barth, M. C., Pfister, G. G., Delle Monache, L., Lamarque, J. F., Archer-Nicholls, S., Tilmes, S., Ghude, S. D., Wiedinmyer, C., Naja, M., and Walters, S.: How Will Air Quality Change in South Asia by 2050?, *Journal of Geophysical Research: Atmospheres*, 123, 1840-1864, doi:10.1002/2017JD027357, 2018.
- Lee, L. A., Carslaw, K. S., Pringle, K. J., Mann, G. W., and Spracklen, D. V.: Emulation of a complex global aerosol model to quantify sensitivity to uncertain parameters, *Atmos. Chem. Phys.*, 11, 12253-12273, 10.5194/acp-11-12253-2011, 2011.
- Lee, L. A., Carslaw, K. S., Pringle, K. J., and Mann, G. W.: Mapping the uncertainty in global CCN using emulation, *Atmos. Chem. Phys.*, 12, 9739-9751, 10.5194/acp-12-9739-2012, 2012.
- Lee, L. A., Reddington, C. L., and Carslaw, K. S.: On the relationship between aerosol model uncertainty and radiative forcing uncertainty, *Proceedings of the National Academy of Sciences*, 113, 5820-5827, 10.1073/pnas.1507050113, 2016.
- Lelieveld, J., Bourtsoukidis, E., Brühl, C., Fischer, H., Fuchs, H., Harder, H., Hofzumahaus, A., Holland, F., Marno, D., Neumaier, M., Pozzer, A., Schlager, H., Williams, J., Zahn, A., and Ziereis, H.: The South Asian monsoon—Pollution pump and purifier, *Science*, 10.1126/science.aar2501, 2018.
- Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q., and Bates, K. H.: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China, *Proceedings of the National Academy of Sciences*, 201812168, 10.1073/pnas.1812168116, 2018.
- Liu, H., Wang, X. M., Pang, J. M., and He, K. B.: Feasibility and difficulties of China's new air quality standard compliance: PRD case of PM_{2.5} and ozone from 2010 to 2025, *Atmos. Chem. Phys.*, 13, 12013-12027, 10.5194/acp-13-12013-2013, 2013.
- Liu, J., Mauzerall, D. L., Chen, Q., Zhang, Q., Song, Y., Peng, W., Klimont, Z., Qiu, X., Zhang, S., Hu, M., Lin, W., Smith, K. R., and Zhu, T.: Air pollutant emissions from Chinese households: A major and underappreciated ambient pollution source, *Proceedings of the National Academy of Sciences*, 113, 7756-7761, 10.1073/pnas.1604537113, 2016.
- Liu, J., Xiang, S., Yi, K., and Tao, W.: Co-Mitigation of Ozone and PM_{2.5} Pollution over the Beijing-Tianjin-Hebei Region, 2017 AGU Fall Meeting, New Orleans, 2017AGUFM.A53F2327L, 2017.

- Lu, X., Hong, J., Zhang, L., Cooper, O. R., Schultz, M. G., Xu, X., Wang, T., Gao, M., Zhao, Y., and Zhang, Y.: Severe Surface Ozone Pollution in China: A Global Perspective, *Environmental Science & Technology Letters*, 10.1021/acs.estlett.8b00366, 2018.
- Marrapu, P., Cheng, Y., Beig, G., Sahu, S., Srinivas, R., and Carmichael, G. R.: Air quality in Delhi during the Commonwealth Games, *Atmos. Chem. Phys.*, 14, 10619-10630, 10.5194/acp-14-10619-2014, 2014.
- Menon, S., Hansen, J., Nazarenko, L., and Luo, Y.: Climate Effects of Black Carbon Aerosols in China and India, *Science*, 297, 2250-2253, 10.1126/science.1075159, 2002.
- Mohan, M., and Bhati, S.: Analysis of WRF Model Performance over Subtropical Region of Delhi, India, *Advances in Meteorology*, 2011, 10.1155/2011/621235, 2011.
- Mohan, M., and Gupta, M.: Sensitivity of PBL parameterizations on PM10 and ozone simulation using chemical transport model WRF-Chem over a sub-tropical urban airshed in India, *Atmospheric Environment*, 185, 53-63, <https://doi.org/10.1016/j.atmosenv.2018.04.054>, 2018.
- Mukherjee, A., and Toohy, D. W.: A study of aerosol properties based on observations of particulate matter from the U.S. Embassy in Beijing, China, *Earth's Future*, 4, 381-395, doi:10.1002/2016EF000367, 2016.
- Murrells, T. P., Passant, N. R., Thistlethwaite, G., Wagner, A., Li, Y., Bush, T., Norris, J., Walker, C., Stewart, R. A., Tsagatakis, I., Whiting, R., Conolly, C., Okamura, S., Peirce, M., Sneddon, S., Webb, J., Thomas, J., MacCarthy, J., Choudrie, S., Webb, N., and Mould, R.: UK Emissions of Air Pollutants 1970 to 2009, Available: https://uk-air.defra.gov.uk/assets/documents/reports/cat07/1401131501_NAEI_Annual_Report_2009.pdf, (last access: 12 Nov. 2018), 2009.
- O'Hagan, A.: Bayesian analysis of computer code outputs: A tutorial, *Reliability Engineering & System Safety*, 91, 1290-1300, <https://doi.org/10.1016/j.ress.2005.11.025>, 2006.
- O'Hagan, A., and West, M.: *Handbook of applied Bayesian analysis*, Oxford University Press, New York, 2009.
- Ojha, N., Naja, M., Singh, K. P., Sarangi, T., Kumar, R., Lal, S., Lawrence, M. G., Butler, T. M., and Chandola, H. C.: Variabilities in ozone at a semi-urban site in the Indo-Gangetic Plain region: Association with the meteorology and regional processes, *Journal of Geophysical Research: Atmospheres*, 117, doi:10.1029/2012JD017716, 2012.
- Pisoni, E., Clappier, A., Degrauwe, B., and Thunis, P.: Adding spatial flexibility to source-receptor relationships for air quality modeling, *Environmental Modelling & Software*, 90, 68-77, <https://doi.org/10.1016/j.envsoft.2017.01.001>, 2017.
- Pisoni, E., Albrecht, D., Mara, T. A., Rosati, R., Tarantola, S., and Thunis, P.: Application of uncertainty and sensitivity analysis to the air quality SHERPA modelling tool, *Atmospheric Environment*, 183, 84-93, <https://doi.org/10.1016/j.atmosenv.2018.04.006>, 2018.
- Pope, C. A., Ezzati, M., and Dockery, D. W.: Fine-Particulate Air Pollution and Life Expectancy in the United States, *New England Journal of Medicine*, 360, 376-386, 10.1056/NEJMsa0805646, 2009.
- Queipo, N. V., Haftka, R. T., Shyy, W., Goel, T., Vaidyanathan, R., and Kevin Tucker, P.: Surrogate-based analysis and optimization, *Progress in Aerospace Sciences*, 41, 1-28, <https://doi.org/10.1016/j.paerosci.2005.02.001>, 2005.
- Ran, L., Zhao, C., Geng, F., Tie, X., Tang, X., Peng, L., Zhou, G., Yu, Q., Xu, J., and Guenther, A.: Ozone photochemical production in urban Shanghai, China: Analysis based on ground level observations, *Journal of Geophysical Research: Atmospheres*, 114, D15301, 10.1029/2008JD010752, 2009.
- Ryan, E., Wild, O., Voulgarakis, A., and Lee, L.: Fast sensitivity analysis methods for computationally expensive models with multi-dimensional output, *Geosci. Model Dev.*, 11, 3131-3146, 10.5194/gmd-11-3131-2018, 2018.
- Sahu, S. K., Beig, G., and Parkhi, N. S.: Emissions inventory of anthropogenic PM2.5 and PM10 in Delhi during Commonwealth Games 2010, *Atmospheric Environment*, 45, 6180-6190, <http://dx.doi.org/10.1016/j.atmosenv.2011.08.014>, 2011.
- Sahu, S. K., Beig, G., and Parkhi, N.: High Resolution Emission Inventory of NOx and CO for Mega City Delhi, India, *Aerosol and Air Quality Research*, 15, 1137-1144, 10.4209/aaqr.2014.07.0132, 2015.
- Saltelli, A., Tarantola, S., and Chan, K. P. S.: A Quantitative Model-Independent Method for Global Sensitivity Analysis of Model Output, *Technometrics*, 41, 39-56, 10.1080/00401706.1999.10485594, 1999.
- Sharma, A., Ojha, N., Pozzer, A., Mar, K. A., Beig, G., Lelieveld, J., and Gunthe, S. S.: WRF-Chem simulated surface ozone over south Asia during the pre-monsoon: effects of emission inventories and chemical mechanisms, *Atmos. Chem. Phys.*, 17, 14393-14413, 10.5194/acp-17-14393-2017, 2017.
- Sharma, S. K., Mandal, T. K., Sharma, A., Jain, S., and Saraswati: Carbonaceous Species of PM2.5 in Megacity Delhi, India During 2012–2016, *Bulletin of Environmental Contamination and Toxicology*, 100, 695-701, 10.1007/s00128-018-2313-9, 2018.
- Shields, M. D., and Zhang, J.: The generalization of Latin hypercube sampling, *Reliability Engineering & System Safety*, 148, 96-108, <https://doi.org/10.1016/j.ress.2015.12.002>, 2016.

- Silver, B., Reddington, C. L., Arnold, S. R., and Spracklen, D. V.: Substantial changes in air pollution across China during 2015–2017, *Environmental Research Letters*, 13, 114012, 10.1088/1748-9326/aae718, 2018.
- Stafoggia, M., Bellander, T., Bucci, S., Davoli, M., de Hoogh, K., de' Donato, F., Gariazzo, C., Lyapustin, A., Michelozzi, P., Renzi, M., Scortichini, M., Shtein, A., Viegi, G., Kloog, I., and Schwartz, J.: Estimation of daily PM₁₀ and PM_{2.5} concentrations in Italy, 2013–2015, using a spatiotemporal land-use random-forest model, *Environment International*, 124, 170-179, <https://doi.org/10.1016/j.envint.2019.01.016>, 2019.
- Thunis, P., Degraeuwe, B., Pisoni, E., Ferrari, F., and Clappier, A.: On the design and assessment of regional air quality plans: The SHERPA approach, *Journal of Environmental Management*, 183, 952-958, <https://doi.org/10.1016/j.jenvman.2016.09.049>, 2016.
- Turner, M. C., Jerrett, M., Pope, C. A., 3rd, Krewski, D., Gapstur, S. M., Diver, W. R., Beckerman, B. S., Marshall, J. D., Su, J., Crouse, D. L., and Burnett, R. T.: Long-Term Ozone Exposure and Mortality in a Large Prospective Study, *American journal of respiratory and critical care medicine*, 193, 1134-1142, 10.1164/rccm.201508-1633OC, 2016.
- Vanuytrecht, E., and Willems, P.: Global sensitivity analysis of yield output from the water productivity model, *Environmental Modelling and Software*, 51, 323-332, 10.1016/j.envsoft.2013.10.017, 2014.
- vu, N., Rafiee, R., Zhuang, X., Lahmer, T., and Rabczuk, T.: Uncertainty quantification for multiscale modeling of polymer nanocomposites with correlated parameters, *Composites Part B: Engineering*, 68, 446-464, 10.1016/j.compositesb.2014.09.008, 2015.
- Wang, T., Xue, L., Brimblecombe, P., Lam, Y. F., Li, L., and Zhang, L.: Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects, *Science of The Total Environment*, 575, 1582-1596, <https://doi.org/10.1016/j.scitotenv.2016.10.081>, 2017.
- Wang, Y., and Chen, Y.: Significant Climate Impact of Highly Hygroscopic Atmospheric Aerosols in Delhi, India, *Geophysical Research Letters*, 0, 10.1029/2019GL082339, 2019.
- Wang, Y., Li, W., Gao, W., Liu, Z., Tian, S., Shen, R., Ji, D., Wang, S., Wang, L., Tang, G., Song, T., Cheng, M., Wang, G., Gong, Z., Hao, J., and Zhang, Y.: Trends in particulate matter and its chemical compositions in China from 2013–2017, *Science China Earth Sciences*, 10.1007/s11430-018-9373-1, 2019.
- WHO: Review of evidence on health aspects of air pollution - REVIHAAP final technical report, World Health Organization: Geneva, 2013.
- WHO: Neurological syndrome and congenital anomalies, Zika Situation Report, 1-7, 2016a.
- WHO: WHO Global Urban Ambient Air Pollution Database (update 2016), Available: <http://www.who.int/airpollution/data/cities-2016/en/>, (last access: 08 Nov. 2018), 2016b.
- Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, *Geosci. Model Dev.*, 4, 625-641, 10.5194/gmd-4-625-2011, 2011.
- Wild, O.: Modelling the global tropospheric ozone budget: exploring the variability in current models, *Atmos. Chem. Phys.*, 7, 2643-2660, 10.5194/acp-7-2643-2007, 2007.
- Willmott, C. J., Robeson, S. M., and Matsuura, K.: A refined index of model performance, *International Journal of Climatology*, 32, 2088-2094, doi:10.1002/joc.2419, 2012.
- Wu, J., Xu, Y., and Zhang, B.: Projection of PM_{2.5} and Ozone Concentration Changes over the Jing-Jin-Ji Region in China, 143-146 pp., 2015.
- Xing, J., Wang, S., Zhao, B., Wu, W., Ding, D., Jang, C., Zhu, Y., Chang, X., Wang, J., Zhang, F., and Hao, J.: Quantifying Nonlinear Multiregional Contributions to Ozone and Fine Particles Using an Updated Response Surface Modeling Technique, *Environmental Science & Technology*, 51, 11788-11798, 10.1021/acs.est.7b01975, 2017.
- Xing, J., Ding, D., Wang, S., Zhao, B., Jang, C., Wu, W., Zhang, F., Zhu, Y., and Hao, J.: Quantification of the enhanced effectiveness of NO_x control from simultaneous reductions of VOC and NH₃ for reducing air pollution in the Beijing–Tianjin–Hebei region, China, *Atmos. Chem. Phys.*, 18, 7799-7814, 10.5194/acp-18-7799-2018, 2018.
- Zaveri, R. A., and Peters, L. K.: A new lumped structure photochemical mechanism for large-scale applications, *J. Geophys. Res.*, 104, 30387-30415, 1999.
- Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), *Journal of Geophysical Research: Atmospheres*, 113, 10.1029/2007JD008782, 2008.
- Zhao, B., Wu, W., Wang, S., Xing, J., Chang, X., Liou, K. N., Jiang, J. H., Gu, Y., Jang, C., Fu, J. S., Zhu, Y., Wang, J., Lin, Y., and Hao, J.: A modeling study of the nonlinear response of fine particles to air pollutant emissions in the Beijing–Tianjin–Hebei region, *Atmos. Chem. Phys.*, 17, 12031-12050, 10.5194/acp-17-12031-2017, 2017.