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Evaluating the sensitivity of fine particulate matter ($PM_{2.5}$) simulations to chemical mechanism in Delhi

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Abstract

Elevated levels of fine particulate matter ($PM_{2.5}$) during winter-time have become one of the most important environmental concerns over the Indo-Gangetic Plain (IGP) region of India, and particularly for Delhi. Accurate reconstruction of $PM_{2.5}$, its optical properties, and dominant chemical components over this region is essential to evaluate the performance of the air quality models. In this study, we investigated the effect of three different aerosol mechanisms coupled with gas-phase chemical schemes on simulated $PM_{2.5}$ mass concentration in Delhi using the Weather Research and Forecasting model with the Chemistry module (WRF-Chem). The model was employed to cover the entire northern region of India at 10 km horizontal spacing. Results were compared with comprehensive filed data set on dominant $PM_{2.5}$ chemical compounds from the Winter Fog Experiment (WiFEX) at Delhi, and surface $PM_{2.5}$ observations in Delhi (17 sites), Punjab (3 sites), Haryana (4 sites), Uttar Pradesh (7 sites) and Rajasthan (17 sites). The Model for Ozone and related Chemical Tracers (MOZART-4) gas-phase chemical mechanism coupled with the Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosol scheme (MOZART-GOCART) were selected in the first experiment as it is currently employed in the operational air quality forecasting system of Ministry of Earth Sciences (MoES), Government of India. Other two simulations were performed with the MOZART-4 gas-phase chemical mechanism coupled with the Model for Simulating Aerosol Interactions and Chemistry (MOZART-MOSAIC), and Carbon Bond 5 (CB-05) gas-phase mechanism couple with the Modal Aerosol Dynamics Model for Europe/Secondary Organic Aerosol Model (CB05-MADE/SORGAM) aerosol mechanisms. The evaluation demonstrated that chemical mechanisms affect the evolution of gas-phase precursors and aerosol processes, which in turn affect the optical depth and overall performance of the model for $PM_{2.5}$. All the three coupled schemes, MOZART-GOCART, MOZART-MOSAIC, and CB05-MADE/SORGAM, underestimate the observed concentrations of major aerosol composition (NO_3^-, SO_4^{2-}, Cl^-, BC, OC, and NH_4^+) and precursor gases (HNO_3, NH_3, SO_2, NO_2, and O_3) over Delhi. Comparison with observations suggests that the simulations using MOZART-4 gas-phase chemical mechanism with MOSAIC aerosol performed better in simulating aerosols over Delhi and its optical depth over the IGP. The lowest NMB (-18.8%, MB = -27.4 µg/m^3) appeared for the simulations using MOZART-MOSAIC

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scheme, whereas the NMB was observed 32.5% (MB = -47.5 µg/m^3) for CB05-MADE/SORGAM and -53.3% (MB = -78 µg/m^3) for MOZART-GOCART scheme.

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1. Introduction

The industrial activities in India have escalated to new heights in the past three decades which consequently have led to multiple urban environmental issues, especially deteriorating air quality due to suspended particulate matter of aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) (Ghude et al., 2016; Ghude et al., 2020). Therefore, it has become a matter of serious concern for public health in India. Currently, the air quality in India, especially in the northern region of India in general and Delhi in particular, is among the poorest in the world (World Health Organization, 2018). Therefore, managing air quality levels in this region of India has emerged as a complicated task.

Recent studies indicate that the exposure to the exceptionally high level of outdoor $PM_{2.5}$ pollution in the National Capital Region (NCR) Delhi poses a serious health risk to the public in Delhi (Ghude et al., 2016; Guttikunda et al., 2013), particularly during the winter season. Diversity of emission sources (Chandra et al., 2018; Hakkim et al., 2019), larger use of fossil fuel such as transport, industries, etc. (Chen et al., 2020), and large scale intense open crop-residue burning in surrounding regions of Delhi (Jena et al., 2015a; Beig et al., 2020; Kulkarni et al., 2020) is responsible for extreme air pollution episodes in the NCR region under favourable meteorological conditions (Vadrevu et al., 2011; Gargava et al., 2015; Tiwari et al., 2018; Liu et al., 2018; Krishna et al., 2019; Chate et al., 2013; Beig et al., 2013; Parkhi et al., 2016). This has drawn significant academic and research interest in predicting high $PM_{2.5}$ levels using numerical air quality models (Guttikunda et al., 2012; Beig et al., 2013; Krishna et al., 2019; Ghude et al., 2020; Kulkarni et al., 2020). Few recent studies have tested the performance of air quality models, particularly WRF-Chem, in simulating hourly $PM_{2.5}$ concentrations in Delhi (Ojha et al.,2020; Chen et al., 2020; Ghude et al., 2020; Kulkarni et al., 2020). These studies suggested that simulating and predicting extreme air quality episodes, particularly $PM_{2.5}$ concentrations exceeding 300μg/m^3, in the NCR region is a challenging task for the air quality models (Kumar et al., 2015; Krishna et al., 2019; Bali et al., 2019). Large uncertainties are involved in the prediction of atmospheric aerosols. This is because chemical transport models predictions suffer from errors in emission inventories (Jena et al., 2015b), inadequate understanding of some of the processes (e.g., secondary organic aerosol formation) (Balzarini et al., 2015), inaccuracies in the initialization of chemical and physical atmospheric state (Ghude et al., 2020), systematic and random errors because of numerical approximations, and approaches the different chemical mechanisms use to calculate size distribution of aerosols coupled with the gas-phase chemical mechanism.

A recent study showed significant differences in simulating aerosol mass concentration over China (Chen et al., 2016, 2017), Europe (Solazzo et al., 2012; Balzarini et al. 2015; Georgiou et al. 2018), USA (Yahya et al., 2017; Hodzic et al., 2013) and Tibetan Plateau (Yang et al., 2018). Differences in the chemical mechanism (Knote et al., 2015), parameterization of heterogeneous formation of secondary inorganic aerosols (SIA), and secondary organic aerosols (SOA), which affects the aerosol process and the evolution of gas-phase precursors, are found to play a key role in the reconstruction of aerosols in above studies. For organic aerosols, the complexity of secondary formation and its aging processes and the lack of emission estimates of intermediate-volatility and semi-volatile organic compounds affect the model performance (Chen et al., 2017; Tsigaridis et al., 2014). Balzarini et al. (2015) showed that simulated total PM mass concentrations, as well as

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aerosol subcomponents, vary between the RADM2 gas-phase chemical mechanism with Modal Aerosol Dynamics Model for Europe/Secondary Organic Aerosol Model (MADE/SORGAM) and CBMZ gaseous parameterization with Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol mechanisms, and CBMZ-MOSAIC performed better in reproducing lower aerosol concentration than RADM2-MADE-SORGAM. Yang et al., (2018) also reported that RADM2-MADE/SORGAM could simulate higher surface $PM_{2.5}$ mass concentrations better than the CBMZ-MOSAIC module over the Tibetan Plateau because of the difference in aerosol compounds and distribution of computed aerosol concentrations between modes and bins. On the other hand, Georgiou et al. (2018) showed that simulated $PM_{2.5}$ by the RADM2-MADE/SORGAM mechanism exhibit lowest mean bias when compared to observations, but it overestimates the ammonium and sulfate aerosols. On the other hand, the MOSAIC aerosol mechanism overestimates $PM_{2.5}$ mass concentrations substantially over the eastern Mediterranean region. In a recent study, Curi et al., (2015) showed that magnitude of the uncertinities in AOD arrising from the assumations of aerosol mixing state (external, internal homogeneous, and internal core shell), the chemical species density, the species complex refractive index, and the hygroscopic growth factors is significant if compared with typical differences found in comparison of simulated values with AOD observations.

Most of these studies focused over the USA, Europe, or China. However, a detailed evaluation of $PM_{2.5}$ with different coupled chemcial schemes (gas-phase mechanism with aerosol schemes) over the IGP region in general and Delhi in particular with scare datasets left unclear view of WRF-Chem's ability to predict $PM_{2.5}$ over this region, a region documented to be one of the most polluted regions in South Asia. A very limited number of modelling studies have focused on evaluating the performance of the air quality models in simulating $PM_{2.5}$ mass concentration in Delhi on an hourly time scale during winter-time pollution. For example, Krishna et al., (2019); Ghude et al., (2020); Kulkarni et al., (2020) carried out WRF-Chem simulations over Delhi with MOZART-4 gas-phase chemistry and Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosol mechanism. They found that the model very well captures the temporal variation in $PM_{2.5}$ mass concentration driven by synoptic-scale meteorological variability, but shows substantial error in simulating the $PM_{2.5}$ magnitude and large model-observations differences. It is therefore important to evaluate the model capability in simulating the concentration of major $PM_{2.5}$ components and major oxidants and how different chemical mechanism affects the $PM_{2.5}$ mass concentrations over this region.

In this study, three two-month simulation experiments using Weather Research and Forecasting model with chemistry (WRF-Chem v3.9.1) were designed for the Northern region of India in general, and National Capital Region, Delhi in particular at 10 km grid resolution during winter-time. For this, we employ and inter-compare MOZART-GOCART, MOZART-MOSAIC, CB05-MADE/SORGAM coupled gas-phase chemistry and aerosol mechanisms to evaluate the simulated $PM_{2.5}$ mass concentrations with extensive ground-based observations in Delhi (17 sites), Punjab (3 sites), Haryana (4 sites), Uttar Pradesh (7 sites), and Rajasthan (17 sites). We also investigated the optical properties of aerosols, and ability of the different coupled chemical mechanism in the model to reconstruct the different aerosol components of $PM_{2.5}$ in Delhi using chemical speciation observations from the Winter Fog Experiment (WiFEX) that took place at the Indira-Gandhi International Airport, New Delhi (Ghude et al., 2017; Acharja et al., 2020). The comparison among the coupled

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aerosol schemes aims at identifying the reasons for differences in model performance. Section 2 briefly describes the gas-phase chemistry and aerosol mechanisms used for the simulations, the basic model configuration, emission data used, and data used for the model evaluation. In section 3, we present the results from the sensitivity simulations and their evaluation with surface observations. Our conclusions and suggestions for further study are given in Section 4.

2. Model setup and description

In this study, we used the Weather Research and Forecasting model coupled with chemistry WRF-Chem v3.9.1 to simulate surface $PM_{2.5}$ mass concentration during the peak winter period, starting from 1 December 2017 to 31 January 2018. Recently, the model has been widely used to simulate the air quality in Delhi (Beig et al., 2013; Gupta and Mohan 2015; Ghude et al., 2020; Kulkarni et al., 2020; Chen et al., 2020) and to estimate NO_X and $PM_{2.5}$ mass concentration over India (Ghude et al., 2013; Krishana et al., 2019; Ojha et al., 2020; Beig et al., 2020). In this study, three sets of simulations were designed using following three widely used coupled schemes (gas-phase chemical mechanisms with aerosol schemes) to simulate the $PM_{2.5}$ mass concentrations over the northern region of India.

MOZART-GOCART (MG): In the first experiment, simulation is performed with the Model for Ozone and related Chemical Tracers (MOZART-4) gas-phase chemical mechanism (Emmons et al. 2010) coupled with the Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosol scheme (Chin et al., 2000; Ginoux et al., 2001). It includes 157 gas-phase reactions, 85 gas-phase species, 39 photolysis, and 16 bulk aerosol compounds. For this experiment, the chemistry scheme is consistent with the chemistry used in the global model that provides the chemical initial and boundary conditions. The GOCART aerosol model simulates five major types of aerosols, namely, sulfate, black carbon, organic carbon, dust, and sea salt. GOCART scheme does not simulate nitrate and secondary organic aerosols. The composition of GOCART aerosol module includes fine unspeciated aerosol contribution (P_{25}), organic carbon (hydrophobic OC1 and hydrophilic OC2), organic black carbon (hydrophobic BC1 and hydrophilic BC2), sulfate (SO_4^{2-}), dust of different sizes (D_1, D_2, D_3, D_4 and D_5 representing dust with effective radii of 0.5, 1.4, 2.4, 4.5 and 8 µm respectively), and sea salt of different sizes (S_1, S_2, S_3 and S_4 representing sea salt with effective radii of 0.3, 1.0, 3.25 and 5µm respectively).

MOZART-MOSAIC (MM): In the second experiment, we used MOZART-4 gas-phase chemical mechanism coupled with the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2008) aerosol scheme. MOSIAC includes sulfate (SULF = SO_4^{2-} +HSO_4^-), methanesulfonate (CH_3SO_3), ammonium (NH_4^+), sodium (Na), calcium (Ca), nitrate (NO_3^-), chloride (Cl^-), carbonate (CO_3), black carbon (BC), primary and organic mass (OC). Other unspecified inorganic species, inert minerals, and trace metals are lumped together as OIN (other inorganic mass). MOSIAC also allowed the gas-phase to partition to the particle-phase, which include H_2SO_4, HNO_3, HCl, NH_3, and MSA (methanesulfonicacid), and also include secondary organic aerosols (SOA). MOSAIC uses a sectional aerosol bin approach for the representation of the aerosol size distribution. In the WRF-Chem model, one can choose between four and eight aerosol size bins, which are demarcated by their lower and upper dry particle diameters. In both cases, only one bin is assigned to aerosols with a diameter between 2.5 and 10 µm. Therefore, when four aerosol bins are used, three bins are assigned to

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aerosols less than 2.5 μm in diameter. When eight aerosol bins are used, seven bins are assigned to aerosols with diameters within this range. Usually, it is sufficient to use the four-bin simulation option to which the focus is on air quality and it also reduces computational complexity (Georgiou et al., 2018).

CB05-MADE/SORGAM (CMS): In the third experiment, we conducted simulations using the Carbon Bond 5 (CB-05) gas-phase mechanism (Yarwood et al., 2005,) which includes 51 chemical species and 156 reactions. Aerosol processes are represented by the Modal Aerosol Dynamics for Europe/ Secondary Organic Aerosol Model (MADE/SORGAM) (Ahmadov et al., 2012) which uses modal aerosol size distribution, and includes an advanced secondary organic aerosol (SOA) treatment based on gas-particle partitioning and gas-phase oxidation in volatility bins. The CB05-MADE/SORGAM mechanism has also been coupled to existing model treatments of various feedback processes such as the aerosol semi-direct effect on photolysis rates of major gases and the aerosol indirect effect on cloud droplet number concentration and resulting impacts on shortwave radiation (Yahya et al., 2016).

The model domain covers the entire northern region of India at a horizontal grid spacing of 10 km and 47 vertical levels stretching from the surface to 10 hPa. Prior anthropogenic emissions of aerosols and trace gases ($PM_{2.5}$, PM_{10}, OC, BC, CO, NOx, etc.) were taken from the EDGAR-HTAP (Emission Database for Global Atmos. Res. for Hemispheric Transport of Air Pollution) for the year 2010 at 0.1º x 0.1º grid resolution and scaled to 2018 using scaling factors as given in Venkatraman et al. (2018). No diurnal variation was added to emissions. Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1) (Guenther et al., 2006) and dust emissions are based on the online Atmospheric and Environmental Research Inc. and Air Force Weather Agency (AER/AFWA) scheme (Jones and Creighton, 2011). Emissions from sea salt are generated based on the scheme of Gong et al. (1997). Daily open biomass burning emissions are obtained from the Fire INventory from NCAR (FINNv1.5) (http://bai.acom. ucar.edu/Data/fire/). The chemical initial and lateral boundary conditions come from the global model simulations from the Model for Ozone and Related Chemical Tracers (MOZART-4) and the meteorological initial and lateral boundary conditions are provided by National Centers for Environmental Prediction Final Reanalysis (NCEP/FNL) dataset, which is available every 6 hours. The simulations are reinitialized monthly to constrain meteorological fields toward NCEP/FNL reanalysis data while forwarding chemistry fields from the previous day. The details configuration of physics and chemistry options used in this study, as well as their corresponding references, can be found in Table S1.

2.1. Observational datasets and evaluation protocol

The surface $PM_{2.5}$ data used in this study are taken from the air quality monitoring network operated by the Indian Institute of Tropical Meteorology (IITM) and the Central Pollution Control Board (CPCB) in Delhi (17 sites), and CPCB monitoring network in Punjab (3 sites), Haryana (4 sites), Uttar Pradesh (7 sites), and Rajasthan (9 sites). The details of these monitoring locations are given in Table S2, and the geographical locations are shown in Figure 1. These sites are representative of traffic, airport, urban, and suburban areas. The quality control and assurance method, followed by CPCB for these air quality monitoring stations, is given at https://cpcb.nic.in/quality-assurance-quality-control/. Furthermore, we take the following steps to reassure the quality of $PM_{2.5}$ observations from the CPCB network stations. For Delhi data quality, we rejected all the

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observations values below 10 µg/m^3 and above 1500 µg/m^3 at a given site if other sites in the network do not show values outside this range. The purpose of this step is to eliminate any short-term local influence that cannot be captured in the models and to retain the regional-scale variability. Second, we removed single peaks that are characterized by a change of more than 200 µg/m^3 in just one hour for all the data in CPCB monitoring stations. This step filters random fluctuations in the observations. Third, we removed some very high $PM_{2.5}$ values that appeared in the time series right after the missing values. For any given day, we removed the sites from the consideration that either experience instrument malfunction and/or appear to be very heavily influenced by strong local sources. Measurement of the inorganic aerosol composition (chemical ions) CL^-, NO_3^-, SO_4^{2-}, and NH_4^+ are made using MARGA-2S instrument during 01 December 2017 to 31 January 2018 at Delhi as a part of the WiFEX field campaign at Delhi international airport (Ghude et al., 2017; Acharja et al., 2020). The quality assurance and control process applied to the measurement of the chemical ion is given at Acharja et al., (2020). The meteorological observation data used in this study are taken from the Indian Meteorological Department (IMD).

The focus of the model evaluation was mainly to assess whether the model is able to effectively reproduce the spatial and temporal distributions of ambient total $PM_{2.5}$ mass concentrations and key $PM_{2.5}$ aerosol composition in Delhi as compared to observations. WRF-Chem is currently employed in the operational air quality forecasting system of the Ministry of Earth Sciences (MoES), Government of India. It is therefore important to examine the performance assessment of WRF-Chem for air quality simulations on a regional scale in general and over Delhi in particular during heavy winter-time pollution. Statistical evaluation metrics such as mean bias (MB), Pearson's correlation coefficient (R), normalized mean bias (NMB), normalized mean error (NME) (the definition of those measures can be found in Yu et al., 2006, and Zhang et al., 2006), and index of agreement (IOA) ranging from 0 to 1 (Yahya et al., 2016) with a value of 1 indicating a perfect agreement, is used to evaluate the perforation of different sets of the experiment. For evaluation, the observational data are paired up with the simulated data on an hourly basis for each site, and then observational data and simulated data are averaged out for all sites in Delhi, Haryana, Uttar Pradesh, and Rajasthan. The statistics are then calculated based on the state-specific data pairs.

3. Results and discussion

3.1. Meteorological evaluation

To quantitatively evaluate the model performance for basic meteorological parameters, the data for the temperature at 2m (T_{2m}), relative humidity at 2m (RH_{2m}), and wind speed at 10m (WS_{10m}) from six stations over Delhi, India is used. Statistical metrics are derived by comparing the output of the three model simulations to hourly measurements averaged over all ground stations. Table 1 shows the correlation coefficient (r), mean bias (MB), and root mean squared error (RMSE) between observed and modeled temperature at 2m (T_{2m}), relative humidity at 2m (RH_{2m}), and wind speed at 10m (WS_{10m}) over Delhi, India. Modelled T_{2m} is in good agreement with observations (NMB = 2 to 5 %) but shows higher RSME values (8.84 to 8.92^{o}C) for all three mechanisms. The statistic for RH_{2m} indicates that the model has dry bias during winter for all the three mechanisms and model over-predicts WS_{10m} by an average of ~1.2 ms^{-1} for all three mechanisms. The over prediction of wind

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speed and poor correlation could be due to the poor representation of surface drag exerted by the unresolved topography, other smaller-scale terrain features, and building morphology (Mar et al., 2016; Zhang et al., 2013).

3.2. Sensitivity simulation of different aerosol scheme

3.2.1. Fine particulate matter ($PM_{2.5}$)

Figure 2 shows the comparison for temporal variation between observed and the modeled hourly $PM_{2.5}$ mass concentrations form the sensitivity simulations with the three aerosol mechanisms from 1 December 2017 to 31 January 2018 over Delhi. Observed (black) surface $PM_{2.5}$ mass concentrations are averaged from the 17 air quality monitoring stations in Delhi, while simulated $PM_{2.5}$ are for the MG (red), MM (blue), and CMS (green) experiments are averaged from the 17 grids containing these observation sites. It can be seen that the run with the MG, MM and CMS chemical schemes did not perform well, although it could capture the temporal variation associated with the synoptic-scale variability during the study period. The mean observed $PM_{2.5}$ concentration during peak winter months was about 191 $\mu g/m^3$. Whereas, the mean modeled $PM_{2.5}$ concentration vary from 89.9 $\mu g/m^3$ with the MG mechanism to 163.8 $\mu g/m^3$ and 147.1 $\mu g/m^3$ with the MM and CMS mechanism respectively, showing a large variability in simulated $PM_{2.5}$ in Delhi among these mechanisms. All three simulations with MG, MM and CMS chemcial schemes significantly under-predict observed $PM_{2.5}$ concentration averaged over Delhi. The statistic showed a large mean bias of about -101$\mu g/m^3$ (RMSE = 146.3) for the simulation with the MG mechanism, which was about 53% of the corresponding observation. On the other hand, simulations with the CMS and MM mechanisms showed much better agreement. The performance statistic showed that the magnitude of bias decrease to -44 $\mu g/m^3$ (23%) and -27 $\mu g/m^3$ (14%) in the CMS and MM simulations, respectively. Differences between the MG, MM, and CMS simulation are more pronounced during the days when hourly $PM_{2.5}$ exceeds 250 $\mu g/m^3$, particularly on 1-7 and 25-31 December 2017, and 17-20 January 2018. Simulations with the MG mechanisms show poor ability of the model to capture these heavy pollution days, while the latter two show reasonable ability to capture hourly $PM_{2.5}$ that exceeds 250 $\mu g/m^3$. On some days, none of the simulation experiments captured the abrupt increase in $PM_{2.5}$ values observed on 18 -23 December, 26-28 December, and 8-16 January and underestimated the observed levels at the majority of the stations.

Further, we evaluated the robustness of model performance for the individual stations in NCR Delhi region. Table S3 shows the statistical performance of three MG, MM and CMS chemical schemes for seventeen stations. Again, the MG mechanism showed the poorest performance with model mean bias varying from 19% to 65% among different stations. The statistics show that for some stations, the MM mechanism performed quite well, while the CMS mechanism shows better agreement for the others (Table S3). Surface $PM_{2.5}$ concentration simulated with the MM mechanism show normalized mean bias (NMB) within ±15% for the following sites: CRRI Mathura (-1.9%), ITO (-7.3%), Lodhi Road (-4.8%), North Campus DU (-12.6%), and Shadipur (-11.5%). The sites showing the NMB within ±15% for the CMS mechanism are Burari Crossing (-11.51%), CRRI Mathura (-7.3%), ITO (-12.1%), and Lodhi Road (-2.1%). Overall, the MM mechanism shows better performance for simulating hourly $PM_{2.5}$ mass concentration over individual stations and Delhi as a whole, but

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both the MM and CMS mechanisms show significant variability in NMB among the monitoring stations. This could be because of the anthropogenic emissions of aerosols and trace gases taken from the EDGAR- HTAP at 0.1° x 0.1° grid resolution, which does not resolve the real variability in emissions in Delhi and may not accurately capture true values observed at the point of measurement. Simulations with the MG mechanism under-predicts $PM_{2.5}$ within 70% at all stations, possibly due to lack of NO_3^- and secondary organic aerosols (SOA) in the GOCART model. We find that simulated mean NO_3^- and SOA together contributed ~44 $\mu g/m^3$ with the MM mechanism, which is about 30% of total $PM_{2.5}$ mass concentration simulated during the winter period.

We also examined the model performance of MG, MM and CMS chemical schemes over the Punjab, Haryana, Uttar Pradesh, and Rajasthan (Figure 3), which are the neighboring states of Delhi and often influences the air quality in NCR region (Kumar et al., 2015; Kulkarni et al., 2020). Table S4 shows the summary of the performance statistic for the individual sites in each state. The average observed $PM_{2.5}$ concentration over Punjab was 84.21 $\mu g/m^3$ and WRF-Chem showed biases of about -24.7 $\mu g/m^3$ (RMSE = 52.1), 13.1 $\mu g/m^3$ (RMSE = 52.6) and 1.9 $\mu g/m^3$ (RMSE = 48.1) for the MG, MM, and CMS aerosol mechanisms respectively. This is about 29%, 15%, and 2% of the observed average value for the MG, MM, and CMS mechanisms, respectively. For the individual monitoring stations in Punjab, simulations with the CMS mechanism showed better performance with a bias of about 5.3% for Amritsar and 9.4% for Ludhiana, whereas the MM mechanism showed better performance with a bias of about -4.8% for Gobindgarh RIMT station.

The average observed $PM_{2.5}$ concentration over Haryana was about 138.8 $\mu g/m^3$, significantly higher (by 45%) than that of average $PM_{2.5}$ over Punjab. WRF-Chem over Haryana showed a bias of about -82.7 $\mu g/m^3$ (-59.6%), -43.7 $\mu g/m^3$ (-31.5%) and -58.9$\mu g/m^3$(-42.4%) for the MG, MM and CMS chemcial mechanisms respectively, indicating that all three aerosol mechanisms significantly under-predict $PM_{2.5}$ surface concentration. For the individual monitoring stations in Haryana, simulations with the MM mechanism showed better performance, and NMB is found to vary from -22% to 48% relative to observations. For Uttar Pradesh, the MG mechanism showed the largest bias of about -126.5 $\mu g/m^3$ (-62.2% of the observed value) while the MM and CMS showed biases of about -58.4 $\mu g/m^3$ (-28.5%) and -84.6 $\mu g/m^3$ (-42.4%) respectively, indicating a large error in simulations irrespective of the mechanism used. For the individual monitoring stations in Uttar Pradesh, NMB with simulations with the MM mechanism found to vary from -21% to 63% relative to observations. Relative to Haryana and Uttar Pradesh, performance statistics for Rajasthan show better results in terms of bias for MM mechanism (bias = -7.6$\mu g/m^3$, NMB = -8.1%). Other two chemicall mechanisms, MG and CMS, showed biases of about -43.1 $\mu g/m^3$ (-46%) and -22.3 $\mu g/m^3$ (-24%), respectively. Overall, all three MG, MM and CMS chemcial mechanisms tend to underpredict the observed $PM_{2.5}$ concentration over the majority of stations in northern India, but the MM mechanism was found to be performing better over Delhi and neighbouring states, except Punjab, where the CMS mechanism performs the best.

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310 **3.2.2. Comparison with satellite AOD**

311 We further examined how the differences between coupled chemical mechanisms translate in
312 simulating Aerosol Optical Depth (at 550 nm) over the model domain. Figure 4 shows the spatial distribution of
313 observed mean AOD (MODIS/TERRA) and simulated AOD at TERRA overpass time with three aerosol
314 mechanisms. All three mechanisms under-predict the observed AOD, and the difference between the three
315 mechanisms is more pronounced over the central and eastern regions of IGP. The mean AOD difference was the
316 highest (-58%) for the simulation with MG mechanism, while the latter two show reasonably good agreement
317 with a mean bias of about -4.3% and -6.6% for the MM and CMS mechanism. This indicates the crucial role of
318 the fine particle of aerosols, which have higher scattering efficiency, in aerosol optical depth budget (Seinfeld et
319 al., 2016; Balzarini et al. 2015; Yang et al., 2018). In spite of good agreement with mean AOD, simulations with
320 both the MM and CMS mechanisms show still large bias over the central and eastern regions of IGP compared
321 to other regions in the model domain. With the simulation with MG mechanism, the difference in magnitude
322 between observed and modelled AOD vary from -0.6 to -0.8 over this region. The observed differences between
323 simulated and observed AOD values over this region are consistent with results from previous studies (Kulkarni
324 et al., 2020 and Nair et al., 2012). These studies found that underestimation of anthropogenic emissions in the
325 IGP and errors in simulating dust emission and transport over this region are some of the reasons for differences
326 in observed and modelled AOD. However, given that emissions are constant in all the three simulation
327 experiments, the considerable differences between modelled and observed AOD might partially coming from
328 the difference in the simulation of the aerosol composition and dust scheme. In our simulation, the MG and
329 CMS use GOCART/AFWA dust scheme while MM uses the GOCART dust scheme. Some of the previous
330 studies have shown that ammonium sulfate ($(NH_4)_2SO_4$), ammonium bisulfate ($(NH_4)HSO_4$), ammonium nitrate
331 (NH_4NO_3) and ammonium chloride (NH_4Cl) scatter light more efficiently at 550 nm (Seinfeld et al., 2016),
332 while BC absorbs the light at 550 nm. The spatial pattern of mean BC concentration and concentrations of gas-
333 phase compounds that lead to secondary inorganic aerosols and distribution of secondary aerosols for the three
334 aerosol mechanisms is shown in Figure 5. We can see that the mean SO_4^{2-} (Figure 5h) was generally lower in the
335 MG experiment and highest in the CMS experiment, particularly over the IGP and northeastern India. This
336 discrepancy may be related to less chemical aqueous-phase oxidation of SO_2 by H_2O_2 in MOZART-4 gas-phase
337 scheme because all the experiment shares the same SO_2 emissions. H_2O_2 is an efficient oxidant of sulphuric
338 compounds in clouds and fog. During peak winter months, widespread fog is often detected over the IGP region
339 during early morning hours and persists till late early afternoon (Ghude et al., 2017; Jenamani et al., 2015). As
340 shown in figure 5f, simulations with the MG and MM (MOZART-4 gas-phase chemistry) mechanism showed a
341 higher concentration of H_2O_2 over IGP, suggesting inefficient oxidation of SO_2 compared to the CMS
342 experiment. Figure 5i shows the surface NO_3 concentration simulated by the MM and CMS mechanism. Since
343 the MG mechanism does not simulate nitrate aerosols, NO_3^- from the MG epxeriment is not shown here. Mean
344 NO_3^- concentration was generally higher in the MM experiment than in the CMS experiment, particularly the
345 magnitude of NO_3^- over central and eastern IGP region is larger. Similarly, as shown in Figure 5j, the magnitude
346 of mean NH_4^+ concentration was also higher in the MM experiment over central and eastern IGP. On the other
347 hand, mean HNO_3 concentration was found highest in the MG experiment, followed by the CMS experiment,
348 and the lowest was found in MM (Figure 5g) experiment. The highest HNO_3 concentration observed in the MG
349 experiment is related to the efficient photochemical conversion of NO_2 and OH to gas-phase HNO_3. However,

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the lack of aerosol thermodynamics in the MG mechanism means that HNO_3 stays in the gas-phase and does not partition to particle-phase. The main precursor for NO_3 is HNO_3, and the equilibrium between nitrate and HNO_3, and gas-phase NH_3 and HNO_3 can convert to aerosol NH_4NO_3. This indicates that the gas-particle partitioning from HNO_3 to NH_4NO_3 is more efficient in the MM experiment than in the CMS experiment. While, higher HNO_3 concentration in the CMS experiment than in the MM experiment may be related to higher surface NO_2 (Figure 5b) concentration due to an efficient O_3-NO titration process that readily transforms to HNO_3 with the photochemical reaction between NO_2 and OH, but not sufficiently converting to aerosol NH_4NO_3. The MM and CMS experiment show higher BC concentration than the MG experiment (Figure 5d), but OC concentration is higher in the MG experiment over the entire IGP region than the MM and CMS experiments (Figure 5e). Further simulated BC to OC ratios is higher in the MG experiment over the IGP, compared to the other two experiments. Few recent studies have shown the significant concentration of chloride ions (Cl^-) in the IGP region (Ghude et al., 2017) and correlation of NH_4^+ with Cl^- implied that sizeable fraction NH_4^+ with Cl^- occurred in NH_4Cl molecular form (Ali et al., 2019) through a gas-phase reaction between HCL and NH_3 (Du et al., 2010). The primary source of this chloride is winter-time biomass and trash burning that occurred widespread over the IGP region, but emissions of chloride from these sources are not provided to the model, and therefore, the MM and CMS simulations show negligible concentrations of Cl^- over the IGP (Figure not shown) region. The bias between observed and simulated AOD may partially be related to missing NH_4Cl aerosols in the simulations. Magnitude of the uncertinities in mdoel AOD also arrisie from the assumations of aerosol mixing state (external, internal homogeneous, and internal coreeshell), the chemical species density, the species complex refractive index, and the hygroscopic growth factors. Recent study show that uncertinities in mdoel AOD due to above paramter is significant if compared with typical differences found in comparison of simulated values with AOD observations (Curi et al., 2015).

3.2.3. Major $PM_{2.5}$ speciation

Table 2 shows that all three chemcial mechanisms underestimate $PM_{2.5}$ concentrations in Delhi. The lowest NMB appears for the MM mechanism (NMB = -18.8%, MB = -27.4 μg/m^3), whereas the NMB for the GM mechanism is -53.3% (MB = -78 μg/m^3) and -32.5% (MB = -47.5 μg/m^3) for the CMS mechanism. Box-whiskers' plot of observed $PM_{2.5}$ mass concentrations at IGI airport and its comparison with simulated $PM_{2.5}$ mass concentrations for the different aerosol mechanisms is given in Figure6. For observations, the 25th and 75th percentile of $PM_{2.5}$ values were observed between 75 μg/m^{3}and 190 μg/m^3, whereas 25th and 75th percentile of $PM_{2.5}$ for the MM, CMS, and MG chemcial mechanisums was observed between 75 μg/m^3 and 150 μg/m^3, 60 μg/m^3 and 120 μg/m^3, and 50 μg/m^3 and 90 μg/m^3, respectively. Among the three sensitivity experiments, the median value of $PM_{2.5}$ for MM (100 μg/m^3) simulation was found closer to observation (120 μg/m^3). Overall, $PM_{2.5}$ mass concentration simulated with the MM mechanism was found to be in better agreement with observations.

In order to understand the individual components of $PM_{2.5}$ chemical species and examine the difference in behavior by the aerosol mechanism for Delhi, we examine separately the dominant aerosol species in $PM_{2.5}$

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and gas-phase compounds that lead to secondary inorganic aerosols. Figure 6 presents the box-whiskers plot for components of $PM_{2.5}$ from the observations and simulated by the model for the different coupled aeorsol mechanisms at Delhi during the study period. It should be noted that nitrate is absent in GOCART; therefore, ammonium and nitrate are not shown in Figure 6. The MG mechanism does not simulate NH_4 but multiplies sulfate by 1.375 to account for NH_4 mass in total $PM_{2.5}$ mass concentration. Observations at Delhi during the study period suggest that the ratio of NH_4 to sulfate is about 1.545 during the winter season, which is about 11% higher. Further, simulated mean sulfate aerosols (SO_4^{2-}) concentration was largely underestimated (~ 40 - 60%) by the model in all three experiments with bias raining from 9 $\mu g/m^3$ to 14 $\mu g/m^3$ compared to the observations (Table 2). However, the gas-phase precursor (SO_2) of sulfate aerosol simulated by the model was found to be slightly overestimated by about 4 - 5 ppb in all the simulations. This implies that chemical aqueous-phase oxidation of SO_2 by H_2O_2 and a heterogeneous nucleation rate from sulphuric acid (H_2SO_4) is not efficiently simulated by all three mechanisms over Delhi during the winter period. Nitrate and sulfate interact with each other through thermodynamic equilibrium but depends upon the gas-phase ammonia concentrations. It can be seen that for NH_3, the simulations with MM and MG mechanism slightly overestimate NH_3 by about 2 – 4 ppb, respectively. On the other hand, simulations with CMS mechanism underestimate NH_3 by about 8 ppb for the same ammonia emissions. However, ammonium aerosols are underestimated by both the simulations with CMS (MB = -26.3 $\mu g/m^3$) and MM (MB = -24.8 $\mu g/m^3$) mechanisms compared to observations (~34 $\mu g/m^3$). Simulated mean nitrate concentration was generally higher in the MM (MB = -7.6 $\mu g/m^3$, NMB = ~25%) experiment than in the CMS (MB = -19.4 $\mu g/m^3$, NMB = ~62%) experiment compared to observation (28 $\mu g/m^3$) but both the experiment show nitrate is negatively biased. As discussed earlier, the main precursor for NO_3 is HNO_3, and the equilibrium between nitrate, HNO_3, and gas-phase NH_3. It can be seen that simulations with the MM and CMS mechanisms highly underestimate the HNO_3 concentration during the winter period. Overall, the gas-particle partitioning from HNO_3 to ammonium nitrate is not efficient in the MM and CMS chemical mechanism. The difference between underestimation of simulated ammonium and nitrate aerosols in the MM and CMS experiments could be due to the differences in the different treatment of the gas-to-particle partitioning from the nitric acid to ammonium nitrate as a function of humidity (Balzarini et al., 2015; Georgiou et al., 2018). Simulations with the MM and CMS simulations highly underestimate chloride aerosol concentrations (by about 0.1 $\mu g/m^3$) compared to observations (25 $\mu g/m^3$) due to the absence of anthropogenic chloride emissions. This indicates considerable uncertainty in the representation of tropospheric chloride emissions and chemistry that affects aerosol formation. Earlier studies have also shown significant enhancement in anthropogenic chloride (chemical tracer for garbage, plastics, and tires burning) during the peak winter season (Acharja et al., 2020; Ghude et al., 2017). During cold winter nights, open biomass burning occurs on the streets and numerous residential localities in the IGP. In the cold winter conditions, people burn wood, leaf litter, garbage, plastics, and tires, etc. as these are available almost free-of-cost as compared to clean energy sources for which one needs to pay. Compared to observations, organic carbon and black carbon is underestimated by all the three experiments. The lowest mean bias (~ -11 $\mu g/m^3$, ~45%) for BC is found for the simulations with MM and CMS mechanism, while the simulation with MG mechanism show ~65% bias (~ -16 $\mu g/m^3$) with respect to the observed BC concentration. For OC the lowest mean bias (~ -9 $\mu g/m^3$, ~35%) appears for the MG mechanism, whereas mean bias was ~ -12 $\mu g/m^3$, (~45%) for MM mechanism, and ~ -15 $\mu g/m^3$ (~51%) for CMS mechanism. Because all the three experiments use the same emission sources, discrepancies between the

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MG, MM, and CMS experiments could be partially attributed to differences treatment of aerosols calculations by the modal and sectional bin approach. The MOSAIC scheme in this study uses Zaveri et al. (2008) approach to divide aerosols into four bins, whereas the CMS scheme use Mozurkewich (1993) approach to divide aerosols into three modes.

4. Conclusion

In this study, we simulated atmospheric gases and aerosols using three WRF/Chem modelling configuration to investigate the effect of coupled gas-phase chemistry and aerosol mechanisms on the reproductions of aerosol concentrations and aerosol optical depth over the northern region of India for the winter period. Simulated results were compared with the air quality observational data from 17 sites in Delhi, 4 sites in Haryana, 7 sites in Uttar Pradesh, 9 sites in Rajasthan over North India. Further, the performance of MOZART-GOCART (MG), MOZART-MOSAIC (MM), and CB05-MADE/SORGAM (CMS) coupled gas-phase chemistry and aerosol mechanisms were investigated for Delhi for major $PM_{2.5}$ chemical spices observed during WiFEX field campaign at IGI Airport, Delhi. Performance of the model for basic meteorological parameters indicates that WRF-Chem could capture 2 m temperate very well but overestimate the wind speed by about 1.2 ms^{-1} at Delhi and may be related to the limited representation of the topography by the model.

Overall, all three coupled chemcial mechanisms tend to underpredict the observed $PM_{2.5}$ concentration over the majority of stations in northern India, but the MOZART-MOSAIC mechanism was found to be performing better over Delhi and neighbouring states. Surface $PM_{2.5}$ concentration simulated by the MOZART-MOZAIC and CB05-MADE/SORGAM chemical mechanism demonstrated relatively lower bias compared to MOZART-GOCART chemical mechanism. The model sufficiently captured the spatial distribution of mean AOD in all three simulations, but MOZART-GOCART highly underpredicts the observed AOD compared to the other two chemcial mechanisms. This is partly due to the difference in aerosols compounds and particularly missing nitrate and secondary organic aerosols from the MOZART-GOCART mechanism. MOZART-MOZAIC and CB05-MADE/SORGAM mechanism underestimate ammonium, nitrate, sulfate, BC, and OC aerosol mass concentrations, and anthropogenic chloride is completely missing from the simulation. These fine mode aerosols scatter/absorbed light more efficiently at 550 nm (Seinfeld et al., 2016), and underestimation of these species in simulations MOZART-MOSAIC and CB05-MADE/SORGAM mechanisms is partly related to observed-modelled bias for surface $PM_{2.5}$ and AOD over the region. Observations in Delhi show a significant contribution of chloride aerosols in SOA, and missing sources of anthropogenic chloride emission lead to large-bias between model and observed chloride concentrations. This is found to be one of the contributing factors for observed discrepancies between surface $PM_{2.5}$ and AOD in all three experiments over the northern region of India. In summary, the result suggests considerable uncertainty in MOZART-GOCART, MOZART-MOSAIC, and CB05-MADE/SORGAM chemistry in the representation of aerosol chemical species and chemistry that affects the aerosol formation. This further implies that the under-prediction of $PM_{2.5}$ concentrations in all three chemical mechanisms is partially coming from the under-prediction of major aerosol species of fine particular matter over IGI Airport, Delhi. Therefore, the selection of chemical mechanisms is a key aspect, and MOZART-

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MOSAIC mechanism could perform better in reconstructing the AOD aerosols over the northern region of India and surface $PM_{2.5}$ over Delhi and neighbouring states.

Data availability

The 0.1°× 0.1°emission grid maps can be downloaded from the EDGAR website onhttps://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=_123 per year per sector. The model data is available at Prithvi (IITM) super-computer and can be provided upon request to the corresponding author. Observational data on $PM_{2.5}$ measurements can be obtained from the CPCB website on https://app.cpcbccr.com/ccr.

Author contributions

All authors contributed to the research; CJ and SDG designed the research; CJ conducted the research; CJ and SDG wrote the paper; CJ performed the WRF/Chem model simulations; RK contributed to writing; SD, VKS, PA, SHK, MK, AJK, DMC, KA, RN, and MR formulated the research.

Competing interests

The authors declare that they have no conflict of interest.

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https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

References:

Acharja, P., Ali, K., Trivedi, D. K., Safai, P.D., Ghude, S. D., Prabhakaran, T., Rajeevan, M.: Characterization of atmospheric trace gases and water soluble inorganic chemical ions of PM_1 and $PM_{2.5}$ at Indira Gandhi International Airport, New Delhi during 2017–18 winter, Science of the Total Environment, 729, 138800, 2020.

Ali, K., Acharja, P., Trivedi, D. K., Kulkarni, R., Pithani, P., Safai, P. D., Chate, D. M., Ghude, S.D., Jenamani, R.K., Rajeevan, M.: Characterization and source identification of PM2.5 and its chemical and carbonaceous constituents during Winter Fog Experiment 2015-16 at Indira Gandhi International Airport, Delhi, Science of the Total Environment, 662,, DOI:10.1016/j.scitotenv.2019.01.285, 687-696, 2019.

Ahmadov, R., McKeen, S. A., Robinson, A. L., Bareini, R., Middlebrook,A. M., De Gouw, J. A., Meagher, J., Hsie, E.-Y.,Edgerton, E., Shaw, S., and Trainer, M.: A volatility basis set model for summertime secondary organic aerosols over the eastern United States in 2006, J. Geophys. Res. 117, D06301, https://doi.org/10.1029/2011JD016831, 2012.

Bali, K., Dey, S., Ganguly, D. and Smith, K. R.: Space-time variability of ambient $PM_{2.5}$ diurnal pattern over India from 18-years (2000-2017) of MERRA-2 reanalysis data, Atmos. Chem. Phys. Discuss.,https://doi.org/10.5194/acp-2019-731, 2019.

Balzarini, A., Pirovano, G., Honzak, L., Zabkar, R., Curci, G., Forkel, R., Hirtl, M., San José, R., Tuccella, P., and Grell, G. A.: WRF-Chem model sensitivity to chemical mechanisms choice in reconstructing aerosol optical properties, Atmos. Environ., 115, 604–619, https://doi.org/10.1016/j.atmosenv.2014.12.033, 2015.

Beig, G., Chate, D. M., Ghude, S. D., Mahajan, A.S., Srinivas, R., Ali, K., Sahu, S. K., Parkhi, N., Surendran, D., Trimbake, H. R.: Quantifying the effect of air quality control measures during the 2010 Commonwealth Games at Delhi, India, Atmospheric Environment, 80, 455-463, 2013.

Beig, G., Sahu, S. K., Singh, V., Tikle, S., Sobhana, S. B., Gargeva, P., Krishna, K. R., Rathod, A., Murthy, B.S.: Objective evaluation of stubble emission of North India and quantifying its impact on air quality of Delhi, Science of the Total Environment, 709, 136126, 2020.

Chandra, B. P., Sinha, V., Hakkim, H., Kumar, A., Pawar, H., Mishra, A. K., Sharma, G., Garg, P. S., Ghude, S. D., Chate, D. M., Pithani, P., Kulkarni, R., Jenamani, R. K., Rajeevan, M., Odd−even traffic rule implementation during winter 2016 in Delhi did not reduce traffic emissions of VOCs, carbon dioxide, methane and carbon monoxide, Curr. Sci., 114, 1318−1325, 2018.

Chate, D. M., Beig, G., Satpute, T., Sahu, S. K., Ali, K., Parkhi, N., and Ghude, S. D.: Assessments of population exposure to environmental pollutants using air quality measurements during Commonwealth Games-2010, Inhalation Toxicol., 25, 333–340, 2013.

Chen, Y., Wild, O., Ryan, E., Sahu, S. K., Lowe, D., Scott A. –N., Wang, Yu., McFiggans, G., Ansari, T., Singh, V., Sokhi, R. S., Archibald, A., and Beig, G.: Mitigation of $PM_{2.5}$ and ozone pollution in Delhi: a sensitivity study during the pre-monsoon period, Atmos. Chem. Phys., 20, 499–514, https://doi.org/10.5194/acp-20-499-2020, 2020.

https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

Chen, Q., Fu, T.-M., Hu, J., Ying, Q., and Zhang, L.: Modelling secondary organic aerosols in China, Natl. Sci. Rev., 4, 806-809, https://doi.org/10.1093/nsr/nwx143, 2017.

Chen, D., Liu, Z., Fast, J., and Ban, J.: Simulations of sulfate-nitrate-ammonium (SNA) aerosols during the extreme haze events over northern China in October 2014, Atmos. Chem. Phys., 16, 10707-10724, https://455 doi.org/10.5194/acp-16-10707-2016, 2016.

Chin, M., Savoie, D. L., Huebert, B. J., Bandy, A. R., Thornton, D. C., Bates, T. S., Quinn, P. K., Saltzman, E. S., and De Bruyn, W. J.: Atmospheric sulfur cycle simulated in the global model GOCART: Comparison with field observations and regional budgets, J. Geophys. Res.-Atmos., 105, 24689–24712, https://doi.org/10.1029/2000JD900385, 2000.

Curci, G., Hogrefe, C., Bianconi, R., Im, U., Balzarini, A., Baró, R., Brunner, D., Forkel, R., Giordano, L., Hirtl, M., Honzak, L., Jiménez-Guerrero, P., Knote, C., Langer, M., Makar, P.A., Pirovano, G., Pérez, J.L., San José, R., Syrakov, D., Tuccella, P., Werhahn, J., Wolke, R., Žabkar, R., Zhang, J., Galmarini, S.: Uncertainties of simulated aerosol optical properties induced by assumptions on aerosol physical and chemical properties: An AQMEII-2 perspective, Atmospheric Environment, 115, 541-552, https://doi.org/10.1016/j.atmosenv.2014.09.009, 2015.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, 2010.

Gargava, P., Rajagopalan, V.: Source apportionment studies in six Indian Cities drawing broad inferences for urban PM10 reductions, Air Qual., Atmosphere Health, 1–11, 2015.

Georgiou, George K., TheodorosChristoudias, YiannisProestos, JonildaKushta, PanosHadjinicolaou and Jos Lelieveld: Air quality modelling in the summer over the eastern Mediterranean using WRF-Chem: chemistry and aerosol mechanism intercomparison, Atmos. Chem. Phys., 18, 1555–1571, https://doi.org/10.5194/acp-18-1555-2018, 2018.

GhudeS. D.,Pfister,G. G., Jena, C., van der A, R.J., Emmons,L. K., and Kumar,R.: Satellite constraints of nitrogen oxide (NOx) emissions from India based on OMI observations and WRF- Chem simulations, Geophys. Res. Lett., vol. 40, 423–428, doi:10.1029/2012GL053926, 2013.

Ghude, S. D., Chate, D. M., Jena, C., Beig, G., Kumar, R., Barth,M. C., Pfister, G. G., Fadnavis, S., Rao, P.: Premature mortality in India due to $PM_{2.5}$ and ozone exposure., Geophys. Res. Lett., 43, 4650−4658, 2016.

Ghude, S.D., Bhat G.S., Prabha T., Jenamani R.K., Chate D.M., Safai P.D., Karipot A.K., Konwar M., Pithani P., Sinha V., Rao P.S.P., Dixit S.A., Tiwari S., Todekar K., Varpe S., Srivastava A.K., Bisht D.S., Murugavel P., Ali K., Mina U., Dharua M., Jaya Rao Y., Padmakumari B., Hazra A., Nigam N., Shende U., Lal D.M., et.al., Acharja P., Kulkarni R., Subharthi C., Balaji B., Varghese M., Bera S. and M. Rajeevan, Winter fog experiment over the Indo-Gangetic plains of India, Current Science, 112, 767-784, doi:10.18520/cs/v112/i04/767-784, 2017.

Ghude, S. D., Kumar, R., Jena, C., Debnath, S., Rachana G. Kulkarni, Stefano Alessandrini, MrinalBiswas, SantoshKulkrani, PrakashPithani, SaurabKelkar, VeereshSajjan, D. M. Chate, V. K. Soni, Siddhartha Singh, Ravi S. Nanjundiah, and M. Rajeevan: Evaluation of PM2.5 forecast using chemical data

https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

assimilation in the WRF-Chem model: a novel initiative under the Ministry of Earth Sciences Air Quality Early Warning System for Delhi, India, Current science (Accepted), 2020.

Ginoux, P., Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O., and Lin, S. J.: Sources and distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res.-Atmos., 106, 20255–20273, https://doi.org/10.1029/2000JD000053, 2001.

Gong, S., Barrie, L. A., and Blanchet, J. P.: Modeling sea salt aerosols in the atmosphere: 1. Model development, J. Geophys.Res., 102, 3805–3818, https://doi.org/10.1029/96JD02953, 1997.

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, https://doi.org/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, Atmos. Environ., 122, 220–229, https://doi.org/10.1016/j.atmosenv.2015.09.039, 2015.

Guttikunda, S., Gurjar, B.: Role of meteorology in seasonality of air pollution in megacity Delhi, India, Environmental Monitoring and Assessment, 184, 3199–3211, 2012.

Guttikunda, S. K.; Goel, R.: Health impacts of particulate pollution in a megacity-Delhi, India, Environ. Dev., 6, 8−20, 2013.

Hakkim, H., Sinha, V., Chandra, B. P., Kumar, A., Mishra, A., Sinha, B., Sharma, G., Pawar, H., Sohpaul, B., Ghude, S. D., Pithani, P., Kulkarni, R., Jenamani, R. K., and Rajeevan, M.: Volatile organic compound measurements point to fog-induced biomass burning feedback to air quality in the megacity of Delhi, Science of the total environment, 295-304, https://doi.org/10.1016/j.scitotenv.2019.06.438, 2019.

Hodzic, A.,Madronich, S., Aumont, B., Lee-Taylor, J., Karl, T., Camredon, M., Mouchel-Vallon, C.: Limited influence of dry deposition of semi-volatile organic vapors on secondary organic aerosol formation in the urban plume, Geophys. Res. Lett., 40, 3302-3307, doi:10.1002/grl.50611, 2013.

Jena, C., Ghude, S. D., Pfister, G.G., Chate, D.M., Kumar, R., Beig, G., Surendran, D. E., Fadnavis, S., and Lal, D.M.: Influence of springtime biomass burning in South Asia on regional ozone (O3): A model based case study, Atmospheric Environment, 100, 37-47, 2014.

Jena, C., Ghude, S. D., Beig, G., Chate, D. M., Kumar, R., Pfister, G. G., Lal, D.M., Surendran, D. E., Fadnavis, S., van der A, R. J.: Inter-comparison of different NO_X emission inventories and associated variation in simulated surface ozone in Indian region, Atmospheric Environment, 117, 61-73, 2015.

Jenamani, R. K. and Rathod, L. S.: Physical processes from data analysis, real time fog forecast system, performances of various indigenous fog models and benchmarking. Technical Report (FDP-FOG 2008-2015), Meteorological Watch Office, IGI Airport, Delhi, 2015.

Jones, S. and Creighton, G.: AFWA dust emission scheme for WRF/Chem-GOCART, 2011 WRF workshop, 20–24 June 2011, Boulder, CO, USA, 2011.

Knote, C., Tuccella, P., Curci, G., Emmons, L., Orlando, J. J., SMadronich, S., Baró, R., Jiménez-Guerrero, P., Luecken, D., Hogrefe, C., Forkel, R., Werhahn, J., Hirtl, M., Pérez, J. L., San José, R., Giordano, L., Brunner, D., Yahya, K., Zhang, Y.: Influence of the choice of gas-phase mechanism on predictions of key gaseous pollutants during the AQMEII phase-2 intercomparison, Atmospheric Environment, 115, 553-568, ISSN 1352-2310, https://doi.org/10.1016/j.atmosenv.2014.11.066, 2015.

https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

615 Krishna, R. K., Ghude, S. D., Kumar, R., Beig, G., Kulkarni, R., Nivdange, S., Chate, D. M.: Surface $PM_{2.5}$
616 estimate using satellite-derived Aerosol Optical Depth over India., Aerosol Air Qual. Res., 19, 25−37,
617 2019.
618 Kulkarni, S. H., Ghude, S. D., Jena, C., Karumuri, R. K., Sinha, B., Sinha, V., Kumar, R., Soni, V. K., and
619 Khare, M.: How Much Does Large-Scale Crop Residue Burning Affect the Air Quality in Delhi?,
620 Environ. Sci. Technol., 54, 8,4790-4799, 2020.
621 Kumar, R., Barth, M. C., Pfister, G. G., Nair, V. S., Ghude, S. D., and Ojha, N.: What controls the seasonal
622 cycle of black carbon aerosols in India?, J. Geophys. Res. Atmos., 120, doi:10.1002/2015JD023298,
623 2015.
624 Liu, L., Shawki, D., Voulgarakis, A., Kasoar, M., Samset, B.H., Myhre, G., Forster, P.M., Hodnebrog, Ø.,
625 Sillmann, J., Aalbergsjø, S.G., Boucher, O., Faluvegi, G., Iversen, T., Kirkevåg, A., Lamarque, J.-F.,
626 Olivié, D., Richardson, T., Shindell, D., and Takemura, T.: A PDRMIP multi-model study on the
627 impacts of regional aerosol forcings on global and regional precipitation, J. Climate, 31, no. 11, 4429-
628 4447, doi:10.1175/JCLI-D-17-0439.1, 2018.
629 Mar, K. A., Ojha, N., Pozzer, A., and Butler, T. M.: Ozone air quality simulations with WRF-Chem (v3.5.1)
630 over Europe: model evaluation and chemical mechanism comparison, Geosci. Model Dev., 9, 3699–
631 3728, https://doi.org/10.5194/gmd-9-3699-2016, 2016.
632 Mozurkewich, M.: The dissociation constant of ammonium nitrate and its dependence on temperature, relative
633 humidity and particle size, Atmos. Environ. A-Gen., 27, 261–270, https://doi.org/10.1016/0960-
634 1686(93)90356-4, 1993.
635 Nair, V. S., Solmon, F., Giorgi, F., Mariotti, L., Babu, S. S., Moorthy, K. K.: Simulation of South Asian
636 aerosols for regional climate studies, J. Geophys. Res. Atmos., 117, No. D04209,
637 https://doi.org/10.1029/2011JD016711, 2012.
638 Ojha, N., Sharma, A., Kumar, M., Girach, I., Ansari, T. U., Sharma, S. K., Singh, N., Pozzer, A., and Gunthe, S.
639 S.: On the widespread enhancement in fine particulate matter across the Indo-Gangetic Plain towards
640 winter, Scientific Reports, 10:5862, 2020.
641 Parkhi, N., Chate, D. M., Ghude, S. D., Peshin, S., Mahajan, A., Srinivas, R., Surendran, D., Ali, K., Singh, S.,
642 Trimbake, H., Beig, G.: Large inter annual variation in air quality during the annual festival 'Diwali' in
643 an Indian megacity, Journal of environmental sciences, 43, 265 – 272, 2016.
644 Seinfeld, J. H., Pandis, S. N., Atmospheric Chemistry and Physics: From Air Pollution to Climate Change (John
645 Wiley & Sons, 2016).
646 Solazzo, E., Bianconi, R., Pirovano, G., Matthias, V., Vautard, R., Moran, M.D., Appel, K.W., Bessagnet, B.,
647 Brandt, J., Christensen, J.H., Chemel, C., Coll, I.,Ferreira, J., Forkel, R., Francis, X.V., Grell, G.,
648 Grossi, P., Hansen, A.B., Miranda, A.I., Nopmongcol, U., Prank, M., Sartelet, K.N., Schaap, M., Silver,
649 J.D., Sokhi, R.S., Vira, J., Werhahn, J., Wolke, R., Yarwood, G., Zhang, J., Rao, S.T., Galmarini, S.,
650 Operational model evaluation for particulate matter in Europe and North America in the context of
651 AQMEII, Atmos. Environ. 53, 75-92, https://doi.org/10.1016/j.atmosenv.2012.02.045, 2012.
652 Tiwari, S., Thomas, A., Rao, P., Chate, D.M., Soni, V. K., Singh, S., Ghude, S. D., Singh, D., Hopke, P. K.:
653 Pollution concentrations in Delhi India during winter 2015–16: A case study of an odd-even vehicle
654 strategy, Atmospheric Pollution Research, https://doi.org/10.1016/j.apr.2018.04.008, 2018.

https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

655 Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., Balkanski, Y., Bauer, S.
656 E., Bellouin, N., Benedetti, A., Bergman, T., Berntsen, T. K., Beukes, J. P., Bian, H., Carslaw, K. S.,
657 Chin, M., Curci, G., Diehl, T., Easter, R. C., Ghan, S. J., Gong, S. L., Hodzic, A., Hoyle, C. R., Iversen,
658 T., Jathar, S., Jimenez, J. L., Kaiser, J. W., Kirkevåg, A., Koch, D., Kokkola, H., Lee, Y. H., Lin, G.,
659 Liu, X., Luo, G., Ma, X., Mann, G. W., Mihalopoulos, N., Morcrette, J.-J., Müller, J.-F., Myhre, G.,
660 Myriokefalitakis, S., Ng, N. L., O'Donnell, D., Penner, J. E., Pozzoli, L., Pringle, K. J., Russell, L. M.,
661 Schulz, M., Sciare, J., Seland, Ø., Shindell, D. T., Sillman, S., Skeie, R. B., Spracklen, D., Stavrakou,
662 T., Steenrod, S. D., Takemura, T., Tiitta, P., Tilmes, S., Tost, H., van Noije, T., van Zyl, P. G., von
663 Salzen, K., Yu, F., Wang, Z., Wang, Z., Zaveri, R. A., Zhang, H., Zhang, K., Zhang, Q., and Zhang, X.:
664 The AeroCom evaluation and intercomparison of organic aerosol in global models, Atmos. Chem.
665 Phys., 14, 10845–10895, https://doi.org/10.5194/acp-14-10845-2014, 2014.
666 Vadrevu, K. P., Ellicott, E., Badarinath, K. V. S., Vermote, E.: MODIS derived fire characteristics and aerosol
667 optical depth variations during the agricultural residue burning season, north India, Environ. Pollut.,
668 159, 1560−1569, 2011.
669 Venkataraman, C., Brauer, M., Tibrewal, K., Sadavarte, P., Ma, Q., Cohen, A., Chaliyakunnel, S., Frostad, J.,
670 Klimont, Z., Martin, R. V., Millet, D. B., Philip, S., Walker, K., and Wang, S.: Source influence on
671 emission pathways and ambient PM2.5 pollution over India (2015–2050), Atmos. Chem. Phys., 18,
672 8017–8039, https://doi.org/10.5194/acp-18-8017-2018, 2018.
673 Yahya, K., Wang, K., Campbell, P., Glotfelty, T., He, J., and Zhang, Y.: Decadal evaluation of regional climate,
674 air quality, and their interactions over the continental US and their interactions using WRF/Chem
675 version 3.6.1., Geoscientific Model Development Discussions, 8, 8, 6707-6756, doi: 10.5194/gmdd-8-
676 6707-2015, 2016.
677 Yahya, K., Glotfelty, T., Wang, K., Zhang, Y., and Nenes, A.: Modeling regional air quality and climate:
678 improving organic aerosol and aerosol activation processes in WRF/Chem version 3.7.1, Geosci.
679 Model Dev., 10, 2333–2363, 2017.
680 Yang, Junhua, Shichang Kang, ZhenmingJi and Deliang Chen: Modeling the Origin of Anthropogenic Black
681 Carbon and Its Climatic Effect Over the Tibetan Plateau and Surrounding Regions, Journal of
682 Geophysical Research: Atmospheres, 123, 671–692. https://doi.org/10.1002/2017JD027282, 2018.
683 Yarwood, G., Rao, S., Yocke, M., and Whitten, G. Z.: Final Report –Updates to the Carbon Bond Chemical
684 Mechanism: CB05, Rep.RT-04-00675, 246 pp., Yocke and Co., Novato, Calif., 2005.
685 Yu, S., Eder, B., Dennis, R., Chu, S.-H., and Schwartz, S.: New unbiased symmetric metrics for evaluation of
686 air quality models, Atmos. Sci. Lett., 7, 26–34, 2006.
687 Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol Interactions and
688 Chemistry (MOSAIC), Journal of Geophysical Research, 113, https://doi.org/10.1029/2007JD008782,
689 2008.
690 Zhang, Y., Liu, P., Pun, B., and Seigneur, C.: A comprehensive performance evaluation of MM5-CMAQ for
691 summer 1999 Southern Oxidants Study Episode, Part-I: Evaluation Protocols, Databases and
692 Meteorological Predictions, Atmos. Environ., 40, 4825– 4838,
693 https://doi.org/10.1016/j.atmosenv.2005.12.043, 2006.

https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

694 Zhang, Y., Sartelet, K., Wu, S.-Y., and Seigneur, C.: Application of WRF/Chem-MADRID and
695 WRF/Polyphemus in Europe – Part 1: Model description, evaluation of meteorological predictions, and
696 aerosol–meteorology interactions, Atmos. Chem. Phys., 13, 6807–6843, https://doi.org/10.5194/acp-
697 13-6807-2013, 2013.
698
699
700
701
702

https://doi.org/10.5194/acp-2020-673
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703 **FIGURE CAPTIONS:**

704 **Figure 1:** Geographical locations of air quality monitoring stations. Delhi stations are represented by blue
705 circles; red circles represent stations in the states of Punjab, Haryana, Rajasthan, and Uttar Pradesh.

706 **Figure 2:** The time series of hourly $PM_{2.5}$ concentrations from the simulation of MOZART-GOCART (red),
707 MOZART-MOSAIC (blue), and CB05-MADE/SORGAM (green) aerosol mechanism was compared with the
708 air quality observation (black) form the CPCB data in Delhi region from 1 December 2017 to 31 January 2018.

709 **Figure 3:** The time series of hourly $PM_{2.5}$ concentrations from the simulation of MOZART-GOCART (red),
710 MOZART-MOSAIC (blue), and CB05-MADE/SORGAM (green) aerosol mechanism was compared with the
711 air quality observation (black) form CPCB data of Punjab, Haryana, Uttar Pradesh, and Rajasthan.

712 **Figure4:** Spatial distribution of simulated AOD of CB05-MADE/SORGAM, MOZART-MOSAIC, MOZART-
713 GOCART, and its difference with observed mean AOD from MODIS.

714 **Figure 5:** Spatial distribution of simulated surface SO_2, NO_2, Ozone, BC, OC, H_2O_2, HNO_3, SO_4^{2-}, NO_3^-, NH_4^+
715 of CB05-MADE/SORGAM, MOZART-MOSAIC, MOZART-GOCART model.

716 **Figure 6:** Box-whisker plots of Nitrate (NO_3^-), Ammonium (NH_4^+), Chlorine (Cl), Organic Carbon (OC), Black
717 Carbon (BC), Sulfate (SO_4^{2-}), HNO_3, SO_2, NH_3, NO_2, Ozone and $PM_{2.5}$ for the MOZART-GOCART (MG),
718 MOZART-MOSAIC (MM), and CB05-MADE/SORGAM (CMS) mechanisms over IGI Airport, Delhi.

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Table1. Pearson's correlation coefficient (R), mean bias (MB), and root mean squared error (RMSE) of hourly values of temperature at 2m, relative humidity at 2m, planetary boundary layer height, and wind speed at 10m for the MOZART-GOCART (MG), MOZART-MOSAIC (MM), and CB05-MADE/SORGAM(CMS) mechanisms averaged over all stations in Delhi.

State	Variables	MOZART-GOCART			MOZART-MOSAIC			CB05-MADE/SORGAM		
		MB	**RMSE**	**R**	**MB**	**RMSE**	**R**	**MB**	**RMSE**	**R**
Delhi	T_{2m}	0.78	8.92	0.23	0.39	8.84	0.24	0.31	8.91	0.23
	RH	-36.6	41.8	0.21	-36.4	41.6	0.20	-36.5	41.7	0.17
	Wind Speed (WS_{10m})	1.2	2.0	0.26	1.2	1.9	0.25	1.1	1.9	0.27

Table2: Index of Agreement (IOA), mean bias (MB), normalized mean bias (NMB), and root mean squared error (RMSE) of hourly values of $PM_{2.5}$ for the MOZART-GOCART (MG), MOZART-MOSAIC (MM), and CB05-MADE/SORGAM (CMS) mechanisms over IGI Airport, Delhi.

State	Station	Variables	MOZART-GOCART				MOZART-MOSAIC				CB05-MADE/SORGAM			
			MB	**NMB (%)**	**RMSE**	**IOA**	**MB**	**NMB (%)**	**RMSE**	**IOA**	**MB**	**NMB (%)**	**RMSE**	**IOA**
Delhi	IGI Airport	SO_4^{2-}	-13.8	-66.7	21.2	0.36	-13.9	-66.9	19.9	0.43	-8.7	-42.3	18.9	0.48
		BC	-15.7	-68.4	20.8	0.46	-10.4	-45.1	18.2	0.52	-10.8	-47.1	18.3	0.52
		OC	-9.2	-35.3	13.1	0.51	-11.7	-44.8	15.3	0.46	-13.4	-51.3	16.5	0.44
		NH_4^+	-	-	-	-	-24.8	-73.8	30.2	0.44	-26.3	-78.1	31.5	0.43
		NO_3^-	-	-	-	-	-7.6	-25.8	18.8	0.45	-19.4	-62.4	24.7	0.41
		CL^-	-	-	-	-	-	-	-	-	-	-	-	-
		$PM_{2.5}$	-77.9	-53.3	102.7	0.53	-27.4	-18.8	81.2	0.69	-47.5	-32.5	86.5	0.66

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Figure 1:

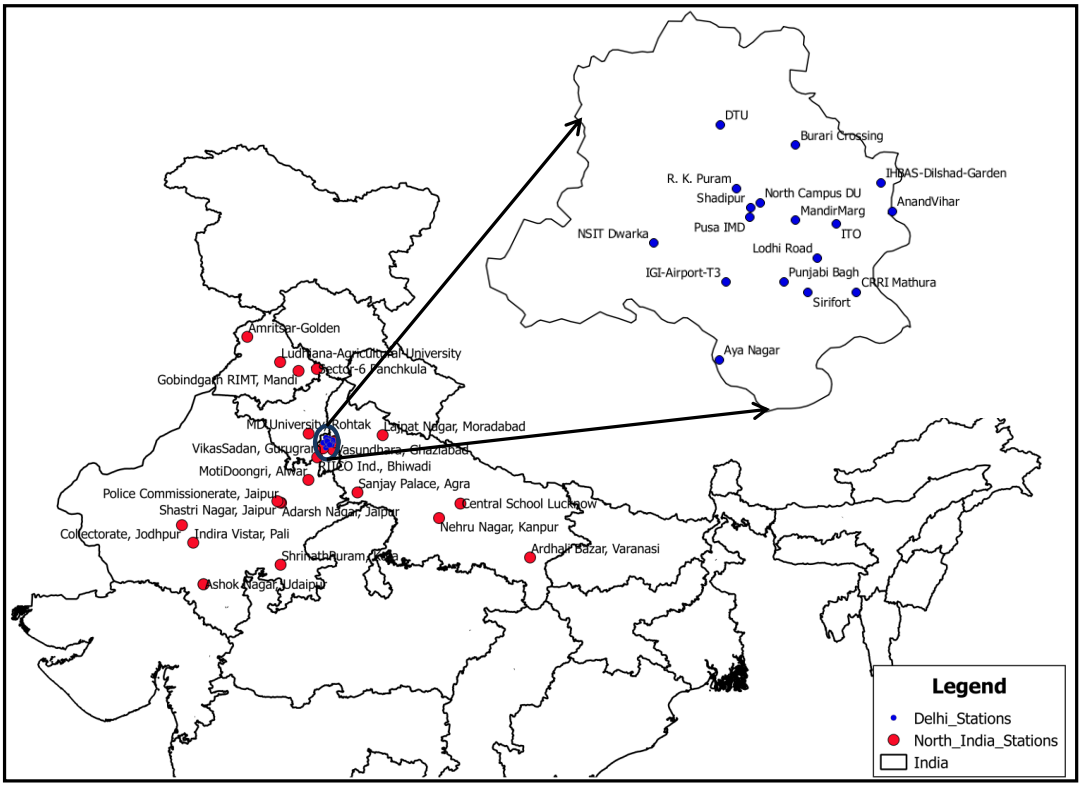

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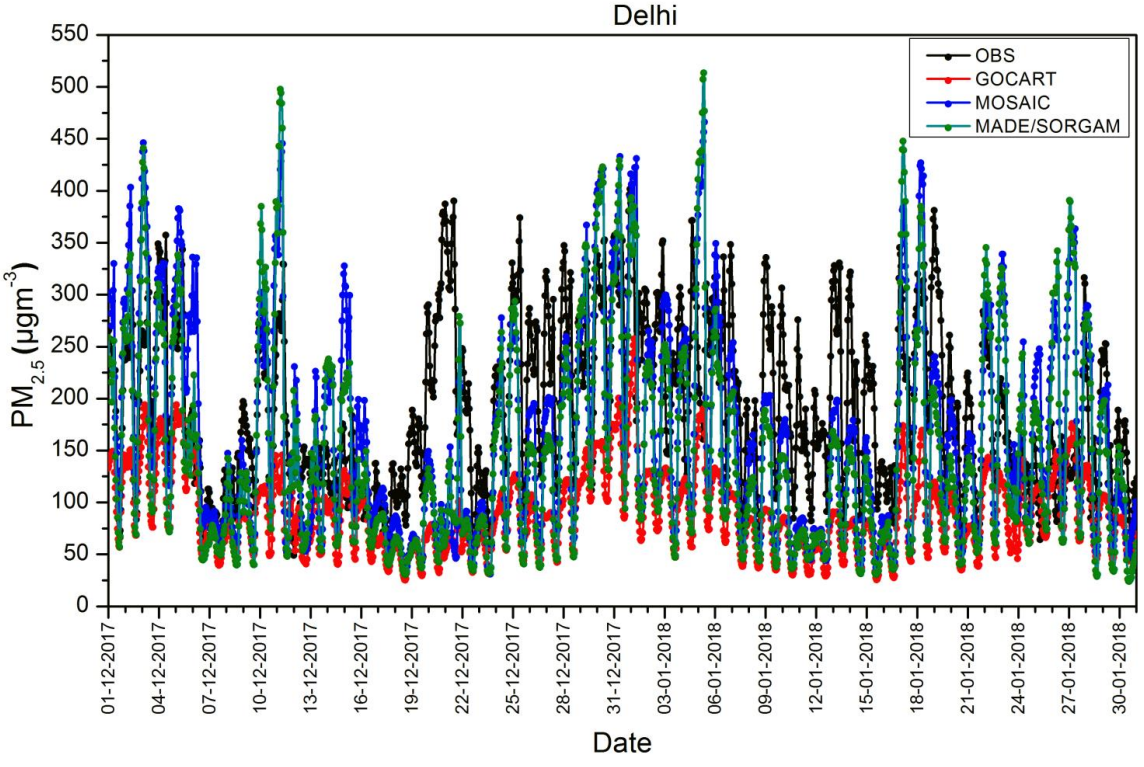
Figure 2 :



https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

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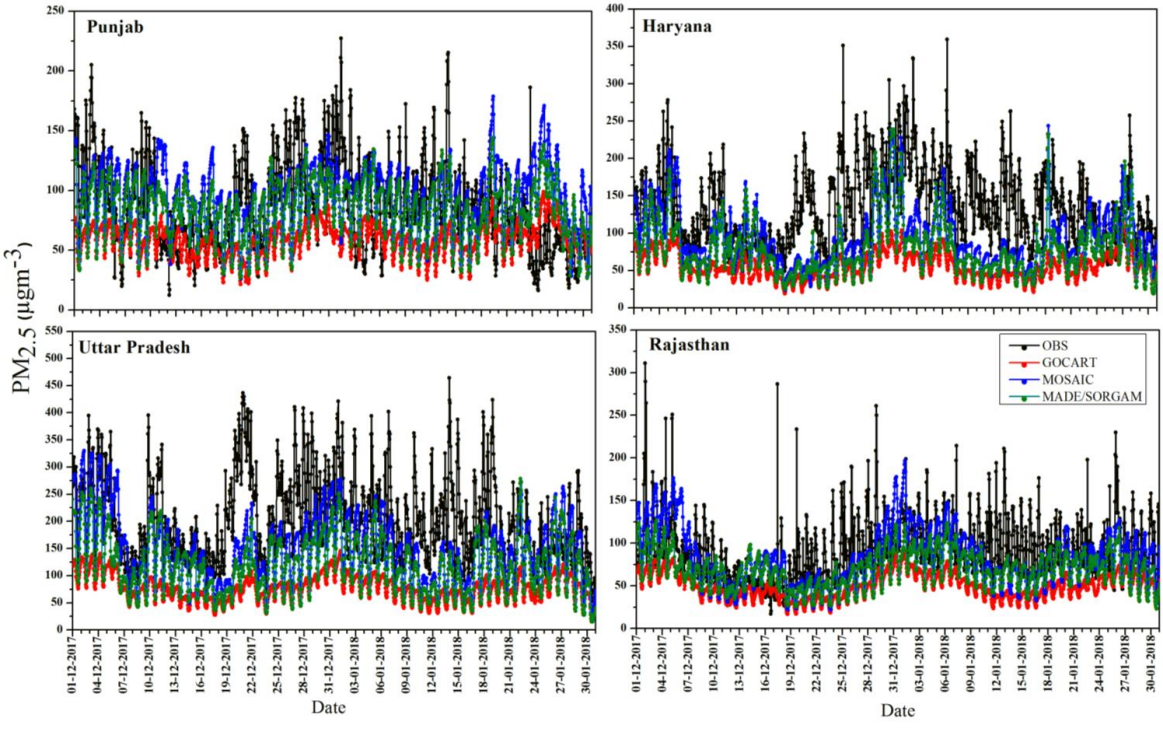
Figure 3:



https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

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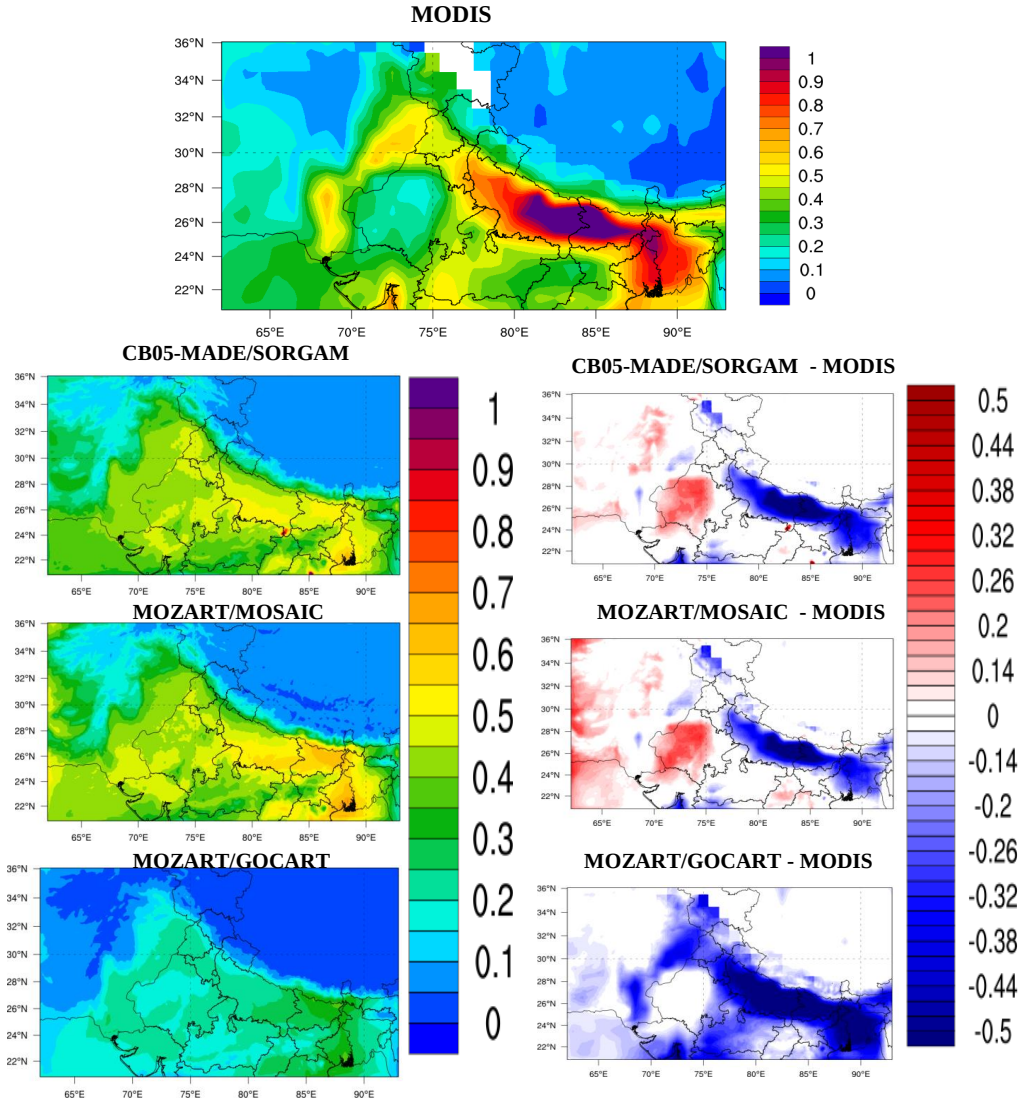
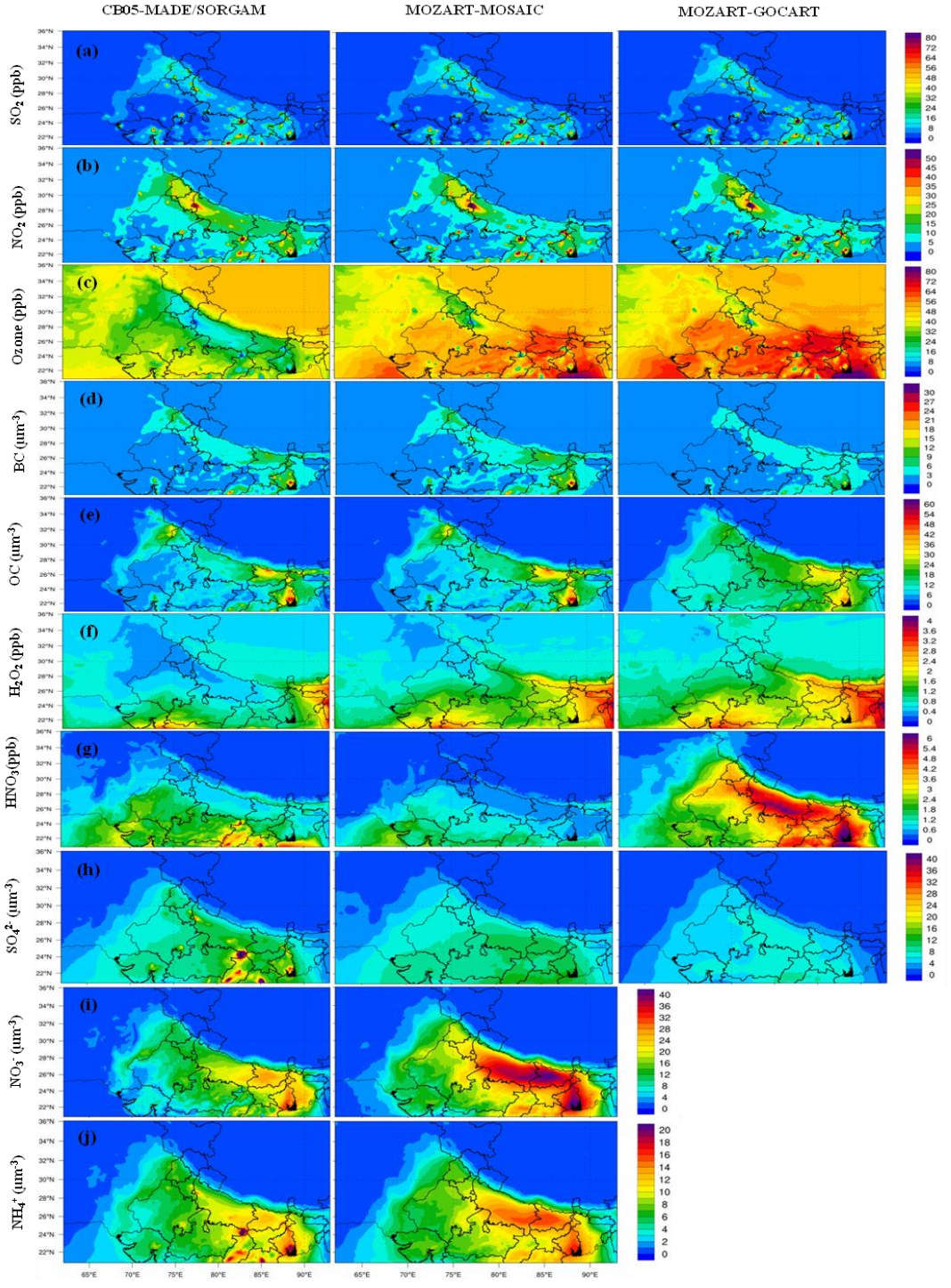
Figure 4:



https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

Atmospheric Chemistry and Physics Discussions

Figure 5:



https://doi.org/10.5194/acp-2020-673
Preprint. Discussion started: 27 August 2020

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Figure 6: