Year-long simulation of gaseous and particulate air pollutants in India

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ABSTRACT

Severe pollution events occur frequently in India but few studies have investigated the characteristics, sources, and control strategies for the whole country. A year-long simulation was carried out in India to provide detailed information of spatial and temporal distribution of gas species and particulate matter (PM). The concentrations of O3, NO2, SO2, CO, as well as PM2.5 and its components in 2015 were predicted using Weather Research Forecasting (WRF) and the Community Multiscale Air Quality (CMAQ) models. Model performance was validated against available observations from ground based national ambient air quality monitoring stations in major cities. Model performance of O3 does not always meet the criteria suggested by the US Environmental Protection Agency (EPA) but that of PM2.5 meets suggested criteria by previous studies. The performance of model was better on days with high O3 and PM2.5 levels. Concentrations of PM2.5, NO2, CO and SO2 were highest in the Indo-Gangetic region, including northern and eastern India. PM2.5 concentrations were higher during winter and lower during monsoon season. Winter nitrate concentrations were 160–230% higher than yearly average. In contrast, the fraction of sulfate in total PM2.5 was maximum in monsoon and least in winter, due to decrease in temperature and solar radiation intensity in winter. Except in southern India, where sulfate was the major component of PM2.5, primary organic aerosol (POA) fraction in PM2.5 was highest in all regions of the country. Fractions of secondary components were higher on bad days than on good days in these cities, indicating the importance of control of precursors for secondary pollutants in India. Capsule abstract: Predicted gaseous and particulate air pollutants in India using WRF/CMAQ in 2015 were validated and the variations were analyzed for future source and health analysis.

1. Introduction

Ever increasing population coupled with rapid growth of industries and urbanization has led to significant air pollution in the world. The situation is more alarming in developing Asian countries like India and China, which together house 36.5% of the world’s population (UN, 2015). In comparison to China, while studies are limited, air quality is worse in India. For example, according to World Health Organization (WHO)’s reports, 15, 21 and 18 Indian cities featured in top 50 most polluted cities with PM10 in 2011, 2014 and 2016, while China had 5, 1 and 5 for the same years, respectively (WHO, 2011, 2014, 2016). Such high concentrations of PM led to enormous pre-mature mortality in India (Chhabra et al., 2001; Dholakia et al., 2014; Maji et al., 2017; Sahu and Kota, 2017). Although people spend most of their time in enclosed rooms (Klepeis et al., 2001), previous studies (Chithra and Nagendra, 2013; Suryawanshi et al., 2016; Tanja et al., 2008) in India have shown that outdoor air pollution significantly affects indoor air quality. Thus, understanding the ambient concentrations of air...
pollutants in different parts of the country will aid in assessing overall mortality associated with pollution exposure in future.

Studies have been conducted to understand the severity of air pollution, the benefits of regulations, and potential control methodologies in India by analyzing ground based measurements (Aneja et al., 2001; Datta et al., 2010; Foster and Kumar, 2011; Gogikar and Tyagi, 2016; Mohan and Kandya, 2007; Ravindra et al., 2006). For example, despite the implementation of compressed natural gas as primary fuel for public transport in Delhi since April 2001, Ravindra et al. (2006) observed a decrease in CO, SO2 and PAHs, but an increase in PM10 and NOx concentrations, in Delhi from 2000 to 2003, due to increase in number of vehicles and ineffective catalytic converters. Beig et al. (2013) showed that NOx, PM and ozone (O3) levels were higher than the WHO approved levels, even though control measures were taken during the Common Wealth Games (CWG) in 2010. Moreover, there were instances where these levels were higher than before and after games period. Satellite retrieved data was also used to study the air quality in India (Badarinath et al., 2009b; Gautam et al., 2009; Ghude et al., 2013; Gupta et al., 2006). Ghude et al. (2008) estimated NOx hot spots in the country using European Remote Sensing Satellite (ERS2) and Environmental Satellite (Envisat). Badarinath et al. (2009a) used National Aeronautics Space Administration (NASA)’s Moderate Resolution Imaging Spectroradiometer (MODIS) Aerosol Optical Depth (AOD) data to study the impact of agricultural burning in Indo-Gangetic plane on the Arabian Sea. Anu Rani et al. (2010) observed higher MODIS AOD in Indo-Gangetic plane coinciding with crop residue burning season.

Even though these studies give insight into the status of air quality, the analysis is often confined to the observation site and is costly. Regional chemical transport models (CTMs) provide prediction of air pollutants with high resolution of temporal and spatial distributions. Gupta and Mohan (2013) predicted PM10 concentrations in New Delhi for a month using emissions obtained by Emissions Database for Global Atmospheric Research (EDGAR) in Weather Research and Forecasting Model with Chemistry (WRF-Chem) model. Marrapu et al. (2014) used WRF-Chem model to predict speciated PM and gaseous pollutants during the CWG using the emission inventories developed during System of Air Quality Forecast and Research (SAFAR) project for Delhi and Intercontinental Chemical Transport Experiment-B (INTEX-B) (Zhang et al., 2009) for other regions. Roy et al. (2008) used a regional chemistry transport model (CTM) to study seasonal variation of O3 and its precursors using emissions from Beig and Brasseur (2006). Jena et al. (2015) studied the influence of biomass burning on springtime O3 using WRF-Chem and fire emissions from national center for atmospheric research (NCAR). Sarkar et al. (2016) predicted gaseous pollutants for three months using WRF-CAMx and emissions estimated from Pandey et al. (2014). Gupta and Mohan (2015) studied the sensitivity of different chemical mechanisms used in WRF-Chem in predicting O3 in New Delhi. Table S1 summarizes modeling studies across India, which tried to understand the seasonal variation of air pollutants. Most of these studies have either been carried out for few weeks or concentrated in a single region or pollutant. To the best of authors’ knowledge, no studies in past have concentrated on understanding the seasonal variations and model performance of all criteria pollutants in different parts of the country. It is imperative to carry out a long-term simulation to understand the seasonal variations of different pollutants at different regions in the country to help design effective control measures in the country. Ghude et al. (2016) predicted PM2.5 and O3 in the country using a year-long 36 km horizontal resolution WRF-Chem model simulation with EDGAR emissions recently. However, as the goal of that study was to estimate premature mortality due to pollutants, the seasonal variation of those pollutants in different regions of the country wasn’t discussed.

The goal of this study is to carry out a one-year long simulation to predict concentrations of gaseous pollutants as well as PM2.5 and its components, whose observations are rarely available in India. This is of the first study that helps to understand the seasonal variation of criteria air pollutants in all regions of India. This would aid in validating the available emission inventories and to better design control strategies in future. The government of India came up with an official air quality index in 2014 to inform the public about the status of air quality in the country. To support this, the concentrations of regulated air pollutants are regularly monitored and reported at different locations in the country by the central pollution control board (CPCB). This study validates the model at different regions of the country with the available observation data. This helps in identifying problems existing in simulating air pollutants in India, which helps future studies to explore right places for improvements. The predicted concentrations in this study would be used subsequently in other studies to understand the dominant sources sectors and regions in the country and assess the potential health risk (Guo et al., 2017).

2. Methodology

The Community Multi-scale Air Quality Model (CMAQ) (Byun and Schere, 2006) version 5.0.2 was used in this study with SAPRC-11 photochemical mechanism (Carter, 2011) and AERO6 aerosol chemistry module (Binkowski and Roselle, 2003). Changes made to better predict the secondary organic and inorganic components of PM2.5 were discussed in detail in Hu et al. (2016) and are only briefly summarized here: (i) heterogeneous chemistry pathways to estimate the formation of sulfate and nitrate from SO2 and NOx in the gas phase (Ying et al., 2014), (ii) more detailed treatment of isoprene oxidation chemistry (Ying et al., 2015), (iii) SOA yields were corrected for vapor wall-loss (Zhang et al., 2014), and (iv) improvement in predicting secondary organic aerosol (SOA) by adding surface controlled reactive uptake of dicarbonyls, isoprene epoxydiol and methacrylic acid epoxide (Li et al., 2015; Ying et al., 2015). All the hours in 2015 were simulated using the CMAQ model with a horizontal grid resolution of 36 km (117 X 117 grids) covering India and parts of the surrounding countries in Asia as shown in Fig. 1. The resolution was determined by considering computing capacity, resolution of available inputs, and the scientific problems. There were 18 layers in the model with surface layer thickness of 35 m and the overall model height of 20 km. Annual anthropogenic emissions of CO, NOx, SO2, ammonia, non-methane volatile organic compounds (VOCs), PM2.5, PM10, elemental carbon (EC) and organic carbon (OC) were generated from EDGAR, version 4.3 (http://edgar.jrc.ec.europa.eu/terms_of_use.php). Yearly and seasonal averaged emissions of PM2.5, its primary components i.e. OC and EC, and gaseous pollutants CO, NOx, SO2, and VOCs are shown in Figure S1. All the pollutants had maximum emissions in Indo-Gangetic plain. The emissions of all the pollutants were more in winter compared to other seasons. Winter PM2.5 emissions were 400, 500, 150 and 100 g/s in north, east, west and south Indian cities, respectively. EC and OC emissions had similar spatial distribution as PM2.5. NOx emissions were high in north India (6–8 mol/s) and south India along the coast (2–4 mol/s), with central and east India being low. VOC emissions were maximum in the Indo-Gangetic plain (20 mol/s) and minimum in south India (5 mol/s). SO2 emissions in north India were much higher than other regions and were mainly from point sources. CO emissions were 10–20 mol/s in north and south India, and comparatively lower in other parts of the country.

The US EPA’s SPECIATE 4.3 source profiles were used to estimate emissions for different VOCs and PM components (Wang et al., 2014a). The re-gridded emissions of individual species were mapped to model species needed by the SAPRC photochemical mechanism and the AERO6 aerosol module. An in-house preprocessor was used to generate hourly emissions based on monthly, weekly and diurnal temporal allocation profiles as mentioned in Wang et al. (2014a) and references within. The base year of EDGAR v4.3 is 2010, and source specific scaling factors listed in Tables S2-S4 were used to adjust the emissions to 2015. All the species of emissions were cataloged into PM, VOCs, SO2,
and NOX. Energy emissions of all states in 2010 were multiplied by the factors listed in Table S2, which were based on statewide power plant reports of coal consumption increase and emission controls from 2010 to 2015 (http://cea.nic.in/monthlyarchive.html). On-road and off-road factors listed in Table S3 were based on statewide transportation report of petroleum products consumption increase and emission control in 2010 and 2015 (http://www.petroleum.nic.in/docs/pngstat.pdf). Agriculture, industry and residential emissions were adjusted based on the nationwide scaling factors of emissions increase from 2010 to 2015 (http://www.indiaenvironmentportal.org.in/files/file/pngstat.pdf) in Table S4.

Model for Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012) was used to generate biogenic emissions, with the plant function types based on the Global Community Land Model (CLM 3.0) files and 8 day MODIS leaf area index (LAI) data. Additionally, the fire inventory from National Center for Atmospheric Research (NCAR) (Wiedinmyer et al., 2011) was used for open biomass burning emissions. Dust and sea salt emissions were generated in line during the CMAQ simulations as in Hu et al. (2015). Initial and boundary conditions used for the simulation were based on default data provided by the CMAQ model for clean continental conditions. The results of first five days were excluded in the analysis to minimize the impact of initial conditions (Zhang et al., 2012).

Meteorological inputs were generated using WRF version 3.6.1 (Skamarock et al., 2008) with initial and boundary conditions from FNL (Final) Operational Global Analysis data on 1.0 × 1.0° grids from National Center for Atmospheric Research for every 6 h (http://dss.ucar.edu/datasets/ds083.2/). The WRF model has identical horizontal resolution as CMAQ model, but has 29 vertical layers. The first eight layers from the surface were the same in both CMAQ and WRF models. Similar approach has been used in Zhang et al. (2012) and Hu et al. (2016).

Fig. 1. India map with the locations of nine cities selected for analysis. The color of each dot on the city shows the averaged predicted PM$_{2.5}$ concentrations in that city. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
gested by Emery et al. (2001) was based on the topography of India could be a reason for this as the benchmark suggested by the model not considering the feedbacks of aerosols, which could be significant in India. RH is generally under-predicted except in January. Similar model performance was observed in other Asian studies (Hu et al., 2015, 2016; Wang et al., 2014b). Figures S2–S5 show daily variation of observed and predicted meteorological parameters at the nine major cities shown in Fig. 1. Under-estimations of temperature were observed during monsoon at Mumbai, Hyderabad and Chennai. Few observed wind speed peaks from April to June at Kolkata were not fully captured. Bengaluru experiences constant southwest winds from May to October, but the model predicts large variations during this period, although it predicts generally constant values from southwest to northwest. It is very likely due to the synoptic influence that the WRF model misses. At Chennai, relative humidity was under-predicted. Despite these miss-predictions, overall WRF captured majority of trends and peaks in observations. Generally, WRF model performance is reliable based the comparison with previous studies mentioned above.

### Table 1

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### 3. Results

#### 3.1. Model performance of meteorological parameters

Meteorology plays an important role in transformation, emission, deposition and transport of air pollutants. In this study, wind speed (WS), wind direction (WD), temperature (T) and relative humidity (RH) predicted by the WRF model was validated using data from the National Climate Data Center (NCDC) in the simulation domain. Table 1 shows the model performance using mean bias (MB), gross error (GE) and root mean squared error (RMSE), along with mean observation and prediction of the meteorological parameters for all the months in 2015. Table S5 shows the formulae used to estimate the statistical metrics used in this study. The performance of the model for different parameters were compared with the criteria suggested by Emery et al. (2001) for a model with grid sizes of 4–12 km. Mean bias and gross error of the predicted temperature, except one month, do not fall under benchmark. The model does a good job in predicting WS, which is evident from 8, 11 and 11 months falling under suggested criteria for MB, GE and RMSE, respectively. Except two months and one month for RMSE, all the months do not fall under benchmark for WD. In addition to uncertainties of the model itself, resolution of 36 km and the topography of India could be a reason for this as the benchmark suggested by Emery et al. (2001) was based on finer simulations (4 or 12 km resolution) in U.S. Additionally, it also could be due to WRF model not considering the feedbacks of aerosols, which could be significant in India. RH is generally under-predicted except in January. Similar model performance was also observed in other Asian studies (Hu et al., 2015, 2016; Wang et al., 2014b). Figures S2–S5 show daily variation of observed and predicted meteorological parameters at the nine major cities shown in Fig. 1. Under-estimations of temperature were observed during monsoon at Mumbai, Hyderabad and Chennai. Few observed wind speed peaks from April to June at Kolkata were not fully captured. Bengaluru experiences constant southwest winds from May to October, but the model predicts large variations during this period, although it predicts generally constant values from southwest to northwest. It is very likely due to the synoptic influence that the WRF model misses. At Chennai, relative humidity was under-predicted. Despite these miss-predictions, overall WRF captured majority of trends and peaks in observations. Generally, WRF model performance is reliable based the comparison with previous studies mentioned above.

#### 3.2. Model performance of gaseous species and PM$_{2.5}$

The nine cities with observations include Delhi and Lucknow in northern-India, Patna and Kolkata in Eastern-India, Hyderabad, Chennai and Bengaluru in southern-India, and Mumbai and Ahmedabad in western-India. Except Kolkata, whose data was downloaded from the monitoring station operated by the U.S. consulate (http://photos.state.gov/libraries/india/231771/PDFs/jan-dec_2015.pdf), data from monitoring stations operated by CPCB (http://cpcb.nic.in/RealTimeAirQualityData.php) was used for analysis. If a city had data in multiple locations, the averaged data from all the locations was used for analysis. Table 2 shows the model performance metrics for criteria pollutants O$_3$, CO, SO$_2$ and NO$_2$, and PM$_{2.5}$ at nine different cities in India. Mean observation, mean prediction, mean fractional bias (MFB), mean fractional error (MFE), normalized mean bias (NMB) and normalized mean error (NME) were used as performance metrics. Unlike Ahmadabad where the model slightly under-predicted O$_3$, over-prediction was observed in other three cities Patna, Delhi and Mumbai. Model did not satisfy the NMB and NME criteria levels set by the EPA (USEPA, 2007). This bias in the model performance could be due to uncertainties associated with emission inventory (Lu et al., 2011), unknown atmospheric processes (Zhang et al., 2011), and meteorological conditions. For example, Sharma and Khare (2017) suggested that errors in emissions of volatile organic compounds and NOx and meteorology can have significant effect in predicted O$_3$ at Delhi. This could also be the reason for the slight over-prediction of O$_3$ in this study. Except in Kolkata, Mumbai and Hyderabad, the model slightly under-predicted the concentrations of PM$_{2.5}$. The MFB and MFE values in all cities lie in the criteria level of ± 0.6 and 0.75 suggested by the EPA (USEPA, 2007). This indicates that the model performance is acceptable and the base case model can be used for regulatory applications for PM$_{2.5}$. Similar model performance i.e. lower biases in predicted PM$_{2.5}$, but higher O$_3$ biases was observed in Ghude et al. (2016). The model under-predicted CO in all cities. This is evident from MFB ranging from −0.63 in Delhi to −1.33 in Chennai. CO emissions seem to be better in northern cities (MFB = −0.8) than southern cities (MFB = −0.99). Yarragunta et al. (2017) suggested that CO is mainly due to vehicles in northern India and coal fired power plants and biomass burning in southern India. Unlike vehicular traffic which is concentrated in a city, residential and biomass burning are distributed. Thus, coarser grid used in this study could be the main reason for this under-prediction,
Overall, as observed from Table 2, model predicted PM$_{2.5}$ better than O$_3$.Figures S6 and S7 show monthly variations of observed and predicted concentrations of 1-h peak O$_3$ and daily PM$_{2.5}$ at different cities in India, where observations were available. O$_3$ concentrations peaked during October and November in all the four cities. The peak concentrations could be due to burning of agricultural residues, which release high amounts of O$_3$ precursors (Sahu et al., 2015). High concentrations follow trend similar to observations in all the stations, i.e. higher concentrations in colder months due to lower solar radiation and wind speed resulting in lesser vertical transport. Moreover, in most of the months, prediction met NMB and NME criteria standards for the highest cutoff range of 60 ppb. To study the performance of the model in predicting higher concentrations, four different cut-off ranges, 30, 60, 90 and 120 μg/m$^3$ were used for PM$_{2.5}$. Similar to O$_3$, the performance of the model indicates that the model predicted well in predicting higher concentration events. MFB and MFE of PM$_{2.5}$ met the criteria limits for all the months, for all the cutoffs. Overall, as observed from Table 2, model predicted PM$_{2.5}$ better than O$_3$.Fig. 2 shows the monthly changes in MFB, MFE, NMB and NME of different cut-off ranges, 30, 40, 50 and 60 ppb were used for O$_3$. Generally, the results indicate that model performance got better with higher cutoff, indicating that the model performs well in predicting higher concentration events. Model performance was better during monsoon and pre-monsoon compared to post-monsoon and winter. In seven months, model prediction met NMB and NME criteria standards for the highest cutoff range of 60 ppb. To especially in regions with higher vehicular traffic. Neither the predictions nor the observations exceeded the daily Indian national air quality standard of 80 μg/m$^3$ for SO$_2$. However, except Ahmadabad and Bengaluru, model over-predicted SO$_2$ in all cities. The model significantly over-predicts SO$_2$ in northern India. One reason could be due to slight over-estimation of SO$_2$ by the emission inventory used in this region, which might not have considered the recent shifting of coal to gas based power plants (Sharma and Khare, 2017). Further studies are required in future to address this issue. Except in Mumbai, model predicted NO$_2$ reasonably well in all cities. The average MFE is 0.63, 0.76, 0.64 and 0.89 in northern, eastern, southern and western cities, respectively. Hu et al. (2016) carried out similar analysis in the north-western Chinese cities which fall in this domain. The performance of this model, for all pollutants but O$_3$ and NO$_2$, is better than the north western Chinese cities.

Fig. 2 shows the monthly changes in MFB, MFE, NMB and NME of PM$_{2.5}$ and O$_3$ in India. Data from all the cities were used for analysis. Previous studies used different cutoff for O$_3$ to analyze the performance of the models. While, the US EPA suggests O$_3$ cutoff of 40–60 ppb (USEPA, 2005), some studies opt for lower cutoff based on the study domain (Hu et al., 2016). In this study, four different cut offs 30, 40, 50 and 60 ppb were used for O$_3$. Overall, the results indicate that model performance got better with higher cutoff, indicating that the model performs well in predicting higher concentration events. Model performance was better during monsoon and pre-monsoon compared to post-monsoon and winter. In seven months, model prediction met NMB and NME criteria standards for the highest cutoff range of 60 ppb. To
Figure S8 shows the diurnal change in model’s performance of O3, PM2.5, CO, SO2, and NO2. Except for SO2, the model performs slightly better in night compared to day time hours. NME for O3, PM2.5, CO and NO2 during night was 1, 15, 5, 12 and −23% less than day, respectively. Figure S9 shows the comparison between modelled compositions of PM2.5 in this study with observed compositions from limited studies in literature. It should be noted that the studies do not have same study episodes. Generally, predicted and observed PM2.5 compositions agree with each other. For example, fraction of SO2 was more in southern cities compared to other cities in India. OC in PM2.5 was highest in all cities in observations, and all but southern cities in predictions.

This study generally reproduces the observed monthly variations of pollutants and shows similar composition of PM2.5 although uncertainties exist. The model results are good to analyze the characteristics as well as spatial and temporal variations in India and the overall evaluation of model performance indicates that more studies in several directions.

3.3. Seasonal variation of pollutants

Fig. 3 shows the seasonal changes in gaseous criteria pollutants, O3, CO, SO2 and NO2. The year was divided into four seasons, winter (December to February), pre-monsoon (March to May), monsoon (June to August) and post-monsoon (September to November). NO2...
Concentrations in winter and post-monsoon were higher than pre-monsoon and monsoon. Moreover, NO₂, SO₂ and CO reached as high as 65 ppb, 70 and 1.6 ppm at Indo-Gangetic plain, which includes Punjab, Delhi, Uttar Pradesh, Bihar and West Bengal, and houses many industries and coal-fired power plants.

Figure 4 and Figure 5 shows the seasonal change in concentrations of total PM₂.₅ and its components, respectively. Higher PM₂.₅ concentrations were observed in the Indo-Gangetic plain and peaked in winter and post-monsoon. Greater emissions (Figure S1) aided by topography results in high PM₂.₅ concentrations in this region. Primary components of PM₂.₅, elemental carbon and primary organic aerosol (POA) were higher in winter due to increase in emissions from house hold wood burning and agricultural activities (Behera and Sharma, 2015).

Maximum SOA concentrations predicted in post monsoon, winter, pre-monsoon and monsoon in the country were 12.8, 10, 7.5 and 2.8 μg/m³, respectively. This could be due to greater anthropogenic emissions of SOA precursors and acidity of aerosols in colder months (Fu et al., 2016; Rengarajan et al., 2011). SO₄, NO₃ and NH₄ peaked in winter and post-monsoon and were least during monsoon. Figure S10 shows the relative difference between concentrations of POA, EC, SO₄ and NO₃ in winter and yearly average. The observed increase of these secondary inorganic components could be associated with higher emissions of SO₂ and NO₂ coupled with greater PM₂.₅ which provides more surface area for heterogeneous transformation of SO₂ and NO₂ to form SO₄ and NO₃ in winter (Zheng et al., 2015). Increase of NO₃ in winter is likely due to the fact that gas-to-particle partition is shifted to the particle phase even

Fig. 3. Seasonal changes in predicted concentrations of O₃, NO₂, SO₂ and CO in India.
though HNO₃ formation is reduced in winter. For sulfate, there is no such competition and a reduction in photochemical formation of SO₄ in winter due to lower solar radiation and temperature in winter is expected (Hu et al., 2016). This could be the reason for relatively greater NO₃ increase than SO₄ despite higher increase in SO₂ (30–80%) than NO₂ (10–45%).

Fig. 6 shows the monthly changes in the fraction of primary, organic and inorganic components of PM₂.₅ at different cities in India. PM₂.₅ concentrations exceeded the annual Indian national air quality standard (INAAQS) of 40 μg/m³ in northern and eastern cities, Delhi, Lucknow, Patna, Kolkata and western city, Mumbai during the post-monsoon and winter seasons. The average PM₂.₅ concentrations in other cities rarely exceed INAAQS in any month. However, their concentrations peaked in winter. Primary organic aerosol (POA) followed by SO₄ fraction in total PM₂.₅ were maximum in north and eastern cities in India. POA was maximum in October and minimum in June or July. For example, in Delhi, POA in PM₂.₅ was 31% in October and 23% in July. SO₄ fraction was maximum in monsoon and minimum in winter. The ratios of fractions of SO₄ in monsoon to winter were 1.6, 1.8, 2.1 and 1.9 in Delhi, Lucknow, Patna and Kolkata, respectively. Unlike SO₄, NO₃ was maximum in winter and minimum in monsoon. The ratios of winter to monsoon fractions of NO₃ in northern and eastern cities were 16.1 and 9.7, respectively. Similar conclusions were achieved in previous studies in north-India (Behera and Sharma, 2010; Saxena et al., 2017). In western cities, POA dominated in all seasons except monsoon. In southern cities, unlike other parts of the country, SO₄ dominated in all seasons except monsoon. Average fraction of SO₄ in southern cities was 1.6 times of other cities. However, similar to northern cities, fraction of NO₂ was highest in winter in these cities. Overall, the fraction of NH₄ was highest in southern cities (7.5%) compared to other cities (4.8%). NH₄ peaked during winter in all cities, contributing to 10, 5 and 4.6% of total PM₂.₅ in southern, northern and eastern cities, and western cities, respectively. This indicates that more efforts have to be put in southern cities to reduce concentrations of precursors to secondary inorganic components.

Average OC to EC ratio in all cities during post monsoon, winter, pre-monsoon and monsoon was 3.2, 3.4, 3.1 and 2.8, respectively, as shown in Table S6. The predicted values in this study at different cities were in general agreement with the observations from various studies.

3.4. Comparison of PM₂.₅ components in good and bad days

To design an effective PM₂.₅ control strategy, it is imperative to determine whether the increase in concentrations was only due to unfavorable weather conditions. Fig. 7 shows the changes in fractional contribution of different primary and secondary components of PM₂.₅ on good days, i.e. concentrations less than the 24-h INAAQS standard of 60 μg/m³, and poor days, i.e. concentrations exceeding INAAQS. As southern cities and Ahmadabad rarely exceed INAAQS, they were not shown. The ratio of averaged PM₂.₅ concentrations on poor and good days in different seasons varied from 1.6 to 1.9, 1.7 to 2.6, 1.6 to 1.7, 2 to 3.8 and 1.9 to 2.5 in Delhi, Patna, Lucknow, Kolkata and Mumbai, respectively. Overall, the maximum difference between averaged PM₂.₅ concentrations in good and bad days occurred in post-monsoon and winter, compared to pre-monsoon and monsoon. During winter in all cities, except Kolkata, the fraction of primary component of PM₂.₅ was higher on good compared to poor days. For example, in Delhi during winter, differences in fraction of primary and secondary components of PM₂.₅ in good and bad days was 3.9% and –5.2%, respectively. In Mumbai, the fraction of secondary PM components was higher in bad days than in good days in all seasons. Kolkata and Lucknow had the maximum increases in secondary PM₂.₅ on poor days compared to good days by 9 and 5%, respectively. Among all the secondary components, SO₄ dominated in all the cities on both good and bad days. Thus, even though the higher concentrations predicted during winter and post-monsoon are due to increase in emissions of primary components and unfavorable weather conditions, the importance of secondary PM cannot be neglected. Special attention needs to be taken to control the precursors of secondary PM components in the country to reduce PM₂.₅ concentrations.
Fig. 5. Seasonal variation in predicted PM$_{2.5}$ components (sulfate (SO$_4$), nitrate (NO$_3$), ammonium (NH$_4$), elemental carbon (EC), primary organic aerosol (POA), secondary organic aerosol (SOA), and “Other” components) in India in 2015.
4. Conclusion

Gaseous pollutants and particulate matter were simulated in the whole year 2015 in India using CMAQ model with WRF generated meteorology and EDGAR based emission inventories. Model performance in predicting PM$_{2.5}$, O$_3$, SO$_2$, CO and NO$_2$ at nine different cities falling in different regions of the country was studied. Model performance of PM$_{2.5}$ and NO$_2$ is reliable, although the model slightly overpredicts O$_3$ and SO$_2$ and under-predicts CO in most of the cities. Further analysis revealed that model does a decent job on hours with high O$_3$ and PM$_{2.5}$ concentrations. In addition to errors in predicted meteorological fields, the chief reasons for biases observed in the model performance could be the uncertainties in the top-down estimations in EDGAR emission inventory and the scaling factors used. Thus, future studies should concentrate on carrying out finer resolution modeling using emission inventories developed using bottom-up approaches at least in mega cities in India. Also, source-oriented air quality modeling studies are to be carried out to estimate possible uncertainties in the emissions and model processes.

NO$_2$, SO$_2$ and CO peaked during winter and were least during monsoon. Moreover, these pollutants had maximum concentrations at Indo-Gangetic plain. Similarly, PM$_{2.5}$ and its components peaked in winter, with average ratios of winter to monsoon concentrations of SO$_4$, NO$_3$ and NH$_4$ were 1.6, 8 and 2.6, respectively. Fraction of NH$_4$ in PM$_{2.5}$ in southern cities was higher than other parts of the country. Fraction of NO$_3$ in PM$_{2.5}$ was higher in winter and lower in monsoon. Fraction of SO$_4$ in PM$_{2.5}$ was higher in monsoon and lower in winter. In southern cities, SO$_4$ dominated all other components of PM$_{2.5}$ unlike other parts of the country where POA fractions were highest. Comparisons of PM$_{2.5}$ components on good and poor days indicate that it is necessary to control precursors of secondary inorganic PM in the country for effective control strategies.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.03.003.

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