

Evaluation of PM_{2.5} forecast using chemical data assimilation in the WRF-Chem model: a novel initiative under the Ministry of Earth Sciences Air Quality Early Warning System for Delhi, India

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Air quality has become one of the most important environmental concerns for Delhi, India. In this perspective, we have developed a high-resolution air quality prediction system for Delhi based on chemical data assimilation in the chemical transport model – Weather Research and Forecasting with Chemistry (WRF-Chem). The data assimilation system was applied to improve the PM_{2.5} forecast via assimilation of MODIS aerosol optical depth retrievals using three-dimensional variational data analysis scheme. Near real-time MODIS fire count data were applied simultaneously to adjust the fire-emission inputs of chemical species before the assimilation cycle. Carbon monoxide (CO) emissions from biomass burning, anthropogenic emissions, and CO inflow from the domain boundaries were tagged to understand the contribution of local and non-local emission sources. We achieved significant improvements for surface PM_{2.5} forecast with joint adjustment of initial conditions and fire emissions.

Keywords: Air quality, particulate matter, chemical data assimilation, aerosol optical depth, fire emissions.

THE National Capital Region (NCR) of India, especially Delhi, encounters severe air pollution episodes during post-monsoon, which continue during the whole winter season, putting the public at high risk. Managing air

quality levels in Delhi has emerged as a complex task in recent years. It is now a matter of serious concern for the regulatory authorities as well as scientific and academic institutions. In recent years, the Government of India (GoI) has been seeking credible scientific studies to develop meaningful policy options to mitigate the risk that acute air pollution episodes pose to public health in the NCR. In order to address this, the Ministry of Environment, Forest and Climate Change (MoEFCC), GoI, has recently launched the National Clean Air Programme (NCAP). The Government has also notified a Graded Response Action Plan (GRAP) to impose temporary controls on industries, power generation, transportation and construction activities in and around Delhi to avert severe air pollution episodes. For this effort, the Ministry of Earth Sciences (MoES), GoI, has been identified as the nodal ministry responsible for developing and deploying an air quality early warning system for Delhi.

Emissions of chemical compounds and aerosols have increased in Delhi as a result of extensive use of fossil fuels, biomass burning and intense agricultural practices in the surrounding regions^{1,2}. Delhi ranks high among the list of regions extremely vulnerable to adverse impacts of air pollution. While the adverse health impact of PM_{2.5} pollution was long known, a recent research report indicates that exposure to outdoor PM_{2.5} pollution in Delhi severely impacts human health³. Unfavourable meteorological conditions coupled with a variety of emission sources in the region are thought to be responsible

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for extreme air pollution episodes occurring in the NCR⁴⁻⁷. In recent years, severe pollution episodes with a very high level of PM_{2.5} concentrations are found to occur more frequently during the winter season that affect daily life in Delhi on heavy pollution days⁸⁻¹⁰. Such extreme events underscore the need for an operational air quality early warning system in order to timely forecast and delineate appropriate and effective air quality management plans for extreme air pollution events. This has drawn significant research interest in predicting PM_{2.5} levels using numerical prediction models^{7,11,12}.

However, simulating and predicting extreme air pollution episodes, particularly high PM_{2.5} concentration in the highly polluted NCR, is a challenging task for the numerical weather prediction models^{7,13}. Large uncertainties are involved in the prediction of atmospheric aerosols because predictions using chemical transport models suffer from errors in emission inventories¹⁴, inadequate understanding of some of the processes (e.g. secondary organic aerosol formation), inaccuracies in the initialization of chemical and physical atmospheric state, and systematic and random errors due to numerical approximations. To reduce the impact of these errors on short-term (1–3 days) forecasts, we need to bring the modelled initial state as close to the observations as possible. This can be achieved by chemical data assimilation, which combines observations and model simulations. Recent developments have revealed that the assimilation of atmospheric chemical composition observations improves the air quality forecast by minimizing the uncertainties of both the chemical initialization of targeted chemical compounds and emissions in chemistry transport models¹⁵⁻¹⁹. Studies have demonstrated that significant improvement in forecasting PM_{2.5} could be achieved through assimilating satellite retrievals of aerosol optical depths (AOD) in chemistry transport models^{19,20}.

In order to improve the accuracy of PM_{2.5} prediction, in this study we develop a novel approach to constrain near real-time fire emissions and improve aerosol initial conditions for PM_{2.5} through assimilating MODIS AOD retrievals. The first version of the prediction system was developed jointly by scientists of MoES institutions and the National Centre for Atmospheric Research (NCAR), USA. This modelling framework consists of a high-resolution fully coupled state-of-the-science. Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) and three-dimensional variational (3DVAR) framework of the community gridpoint statistical interpolation (GSI) system. The 3DVAR-GSI system assimilates data from satellites on AOD, which is related to the emissions from various anthropogenic and natural sources, including dust and stubble burning. The model also takes into account the long-range transport of dust from dust storms and particulate matter from stubble burning, and provides prediction with a lead time of 72 h. The modelling framework also consists of tracers for carbon

monoxide (CO) emissions from stubble burning, anthropogenic emissions within the model domain and transport of CO from outside the domain to help determine the relative importance of Delhi and non-Delhi emission sources. We implemented the CO tracers following the tagged-tracer approach described in our previous work^{13,21}. The advanced Air Quality Early Warning System (AQEWS) was made operational during October 2018–January 2019. The AQEWS was meant to issue alerts on large-scale air pollution events that may occur over the Delhi region.

This study provides an overview of the AQEWS framework, including a description of the air quality forecasting model based on WRF-Chem, a brief description of the data assimilation system, and *in situ* observations. Finally, it highlights the impact of assimilation results on the accuracy of the forecasts. MoES is continuing attempts to assimilate more data from various observational platforms and other criteria pollutants to further improve the accuracy of PM_{2.5} predictions.

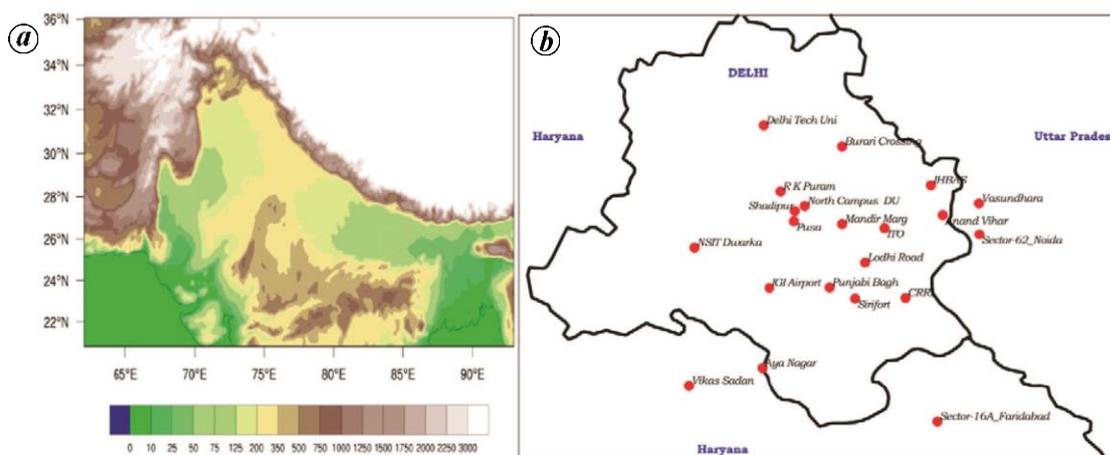
Materials and methods

WRF-Chem model

To simulate the transport of aerosols and other chemical species in the Delhi region, WRF-Chem version 3.9.1 has been identified as one of the several air quality models for use in AQEWS. Table 1 lists the physical and chemical parameterization schemes of model configuration. The outer computational domain of the model includes the entire northern region of India, also covering Pakistan on the western side (Figure 1a). The outer domain was defined at a horizontal grid spacing of 10 km in both the latitudinal and longitudinal directions, while the inner domain was defined at 2 km resolution covering the NCR. The model top was set to 10 hPa and included 47 vertical levels. Prior anthropogenic emissions of aerosols and trace gases were taken from the EDGAR-HTAP for the year 2010 at 0.1° grid resolution and scaled to 2018 using scaling factors as given in Venkatraman *et al.*²². No diurnal variation was added to the emissions. For the inner domain, the original emissions were processed using a mass-conserving emission preprocessor to match the model inner grid spacing of 2 km. It can be seen that re-gridding emissions from 10 to 2 km do not resolve the true variability in emissions at 2 km resolution ([Supplementary Figure 1](#)). However, the total mass emitted is the same before and after re-gridding for a given region. The model uses MOZART-4 gas-phase chemistry linked to the GOCART aerosol scheme (MOZCART). The GOCART aerosol model simulates five major types of aerosols, namely sulphate, black carbon (BC), organic carbon (OC), dust and sea salt. Nitrate and secondary organic aerosols are missing in the MOZCART scheme.

Table 1. Physical and chemical schemes used in the model set-up

Atmospheric process	Schemes
Cloud microphysics	Thompson <i>et al.</i> ³⁶
Longwave radiation	Rapid radiative transfer model developed for general circulation models (GCMs) ³⁷
Shortwave radiation	Rapid radiative transfer model developed for GCMs ³⁷
Surface layer	Monin–Obukhov (Janjic Eta) scheme ³⁸
Land surface model	Unified Noah land-surface model ³⁹
Planetary boundary layer	Bougeault and Lacarrere (BouLac) TKE ⁴⁰
Cumulus	Grell–Freitas ensemble scheme ⁴¹
Gas phase chemistry	Model for ozone and related tracers chemical mechanism ²⁴
Aerosol processes	Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport Model ⁴²

**Figure 1.** *a*, Model outer domain (10 km) with topographic contours. *b*, Model innermost domain (2 km); red dots represent the location of air quality monitoring stations in the National Capital Region of India.

Biogenic emissions were calculated on-line from the Model of Emissions of Gases and Aerosols from Nature (MEGAN)²³. The static geographical fields such as soil properties, vegetation fraction, land-use pattern, erodible fraction and terrain height were taken from the MODIS data. Spatially and temporally (six hourly) varying chemical boundary conditions were provided by global model simulations from the Model for Ozone and Related Chemical Tracers (MOZART-4) 10-year climatology²⁴. After March 2019, AQEWS was driven by the analysis and forecast product (Ensemble-Kalman filtering) produced by the Indian Institute of Tropical Meteorology-Global Forecasting System (IITM-GFS, T1534) spectral model initial and boundary conditions at 12.5 km grid resolution.

Data assimilation algorithm

The chemical data assimilation algorithm employed in WRF-Chem for assimilation of MODIS AOD retrievals is based on a 3DVAR scheme of the GSI system (version 3.5), similar to Kumar *et al.*¹⁹. The GSI-3DVAR scheme combines information from the MODIS AOD and model

background AOD to find optimal analysis state by minimizing the following cost function

$$J = \frac{1}{2} x^T B^{-1} x + \frac{1}{2} (Hx - o)^T O^{-1} (Hx - o), \quad (1)$$

where x represents the state vector that consists of aerosol chemical composition and meteorological variables used in AOD calculations. B is commonly referred to as background error covariance (BEC) matrix which determines how much of the difference between observed and modelled values actually contributes to the analysis increment, and also spreads observations horizontally and vertically. In eq. (1), H represents the forward operator that transforms the GOCART aerosol chemical composition to AOD using the community radiative transfer model (RTM) in the GSI framework. For the minimization of the cost function J , iterative conjugate gradient method was used, which is described in detail in DTC²⁶. The maximum number of iterations was set to 50 to reach the 10^{-9} convergence threshold for the GSI solution. During the assimilation cycle, when the background error was larger than the observation error, the model aerosol concentrations were adjusted to push the model AOD

towards the MODIS AOD. On the other hand, when background error was smaller than observation error, the model aerosol concentrations were not changed.

Sixteen GOCART aerosol species, temperature, pressure, relative humidity and grid thickness variables in WRF-Chem are included as the state variables in GSI. The composition of GOCART aerosol module includes fine unspiciated aerosol contribution (P_{25}), organic carbon (hydrophobic OC_1 and hydrophilic OC_2), organic black carbon (hydrophobic BC_1 and hydrophilic BC_2), sulphate (SO_4), dust of different sizes (D_1, D_2, D_3, D_4 and D_5 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 with effective radii of 0.3, 1.0, 3.25 and 5 μm respectively). All aerosol components are defined as the control variables and are adjusted directly by the AOD assimilation.

BEC statistical parameters for all the aerosols have been modelled using the community Generalized Background Error (GEN_BE) code, which utilizes the difference between two forecasts initialized by different meteorology and emissions. In this study, the first WRF-Chem forecast was initialized with GFS forecasts using default EDGAR-HTAP and FINN emissions. The second forecast was initialized using ERA-Interim and EDGAR-HTAP with FINN emissions enhanced by 100%. This uncertainty of 100% in both anthropogenic and biomass burning emissions is assumed based on the uncertainties among different emission inventories by Granier *et al.*²⁷, Wiedinmyer *et al.*²⁸ and Jena *et al.*¹⁴. The rationale behind these perturbations was to capture uncertainties in air quality simulations due to errors and biases in transport and emissions. BEC statistical parameters were then calculated for the WRF-Chem forecast valid at the assimilation time, i.e. 0900 UTC. Other sources of uncertainties in the state variables such as those resulting from chemical mechanisms, boundary conditions, dry and wet depositions, etc. were not considered. Figure 2 shows an example of the estimated vertical distribution of background error variance for BC. It can be seen that variance is highest well within the planetary boundary layer (PBL) and decreases vertically above the PBL height. This indicates that larger changes in aerosol mass concentration after AOD assimilation will take place within the PBL.

In eq. (1), o represents the MODIS AOD retrievals, and O is the observation error covariance matrix. In the present AQEWS WRF-Chem set-up, we have used level-2 MODIS 550-nm AOD from NASA near real-time (NRT) retrievals at 10 km resolution from both Aqua and Terra satellite platforms. MODIS overpass corresponding to 10:30 and 1:30 local time over India was assimilated into WRF-Chem during the 0900 UTC assimilation cycle. Following Remer *et al.*²⁵, observational errors for MODIS retrievals were specified as $(0.05 + 0.15 \times \text{AOD})$ over the land area.

Experimental design

We have performed two sets of simulations to examine the impact of assimilation on short-term $PM_{2.5}$ prediction. The first experiment was performed for the period 1–20 November 2017, covering the worst air quality events that occurred on 7 and 13 November. Long-range transport of PM (biomass burning in the northwestern region of NCR), local emissions in the NCR and stable meteorological conditions were the main factors for extremely poor air quality during these events. This experiment was performed in hind-cast mode to examine how the assimilation of MODIS AOD retrievals and daily fire emissions improve the prediction of the $PM_{2.5}$ system for the above-mentioned events. In this experiment, biomass burning emissions of aerosols and trace gases were updated daily based on Fire INventory from NCAR (FINN) version 1.5. The model was driven by the National Centers for Environmental Prediction Global Forecast System (GFS) as provided by NCAR (<http://rda.ucar.edu/datasets/ds084.1>). In the second experiment, the model was run during 15 October 2018–28 January 2019 to provide forecast for the next 72 h. Due to the non-availability of real-time fire emissions, the model was updated with one-day-old fire emissions and corrected with the latest fire count data from MODIS before the assimilation cycle. The same fire emissions were continued for 72 h in the forecast. In both the experiments, WRF-Chem simulations were initialized at 0900 UTC, and aerosol initial conditions updated through assimilation of both MODIS Aqua and Terra AOD. While Terra and Aqua AOD are available around 0600 UTC and 0900 UTC, they are assumed to be available for assimilation at 0900 UTC,

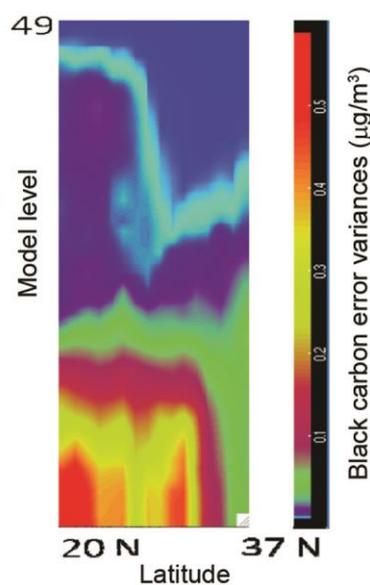


Figure 2. Vertical distribution of error variance for black carbon (BC) mass concentration over Delhi.

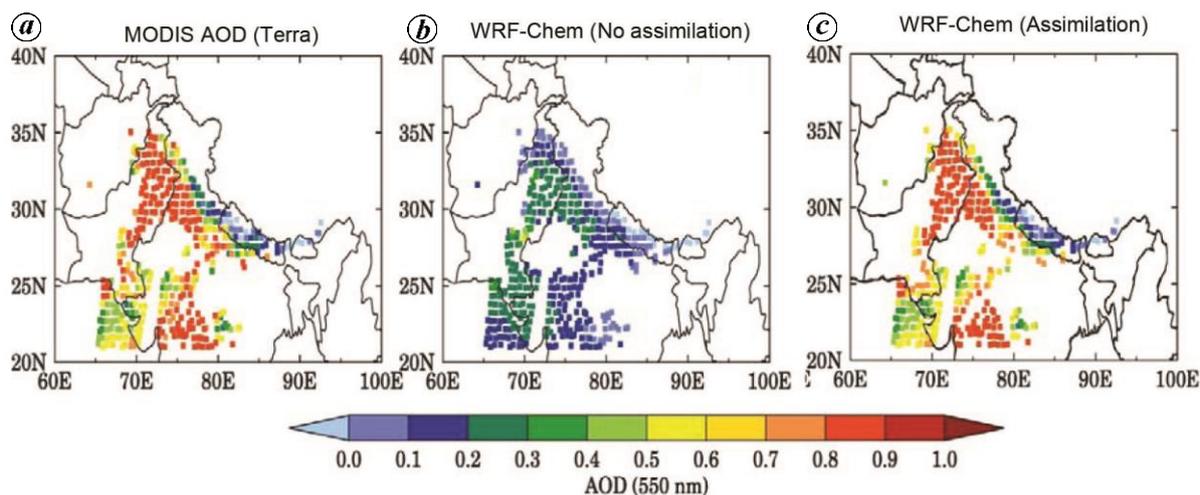


Figure 3. Spatial distribution of (a) MODIS aerosol optical depth (AOD), (b) model AOD without data assimilation and (c) model AOD with data assimilation on 7 November 2017 at 0900 UTC.

which is a common assumption in the 3DVAR framework and is essential to avoid frequent stops in the forecast cycles. The WRF-Chem initial state updated through assimilation of MODIS AOD was then integrated forward to provide 72 h forecast. Every day, chemical fields were initialized from the previous WRF-Chem forecast at 0900 UTC, aerosol initialization updated through assimilation, and meteorology refreshed using the GFS forecast.

Results and discussion

Impact of AOD assimilation on surface $PM_{2.5}$ in the hind-cast experiment

To examine the robustness of improvement in surface $PM_{2.5}$ due to MODIS AOD assimilation, we performed two simulations for the period 1–20 November 2017. The first experiment (control experiment) was conducted without AOD assimilation, but fire emissions were updated every day. The second experiment was conducted with assimilated MODIS AOD every day at 0900 UTC. Figure 3 a–c shows the spatial distribution of MODIS AOD and AOD simulated by the model with and without data assimilation respectively, for a typical case of 7 November 2017 at 0900 UTC. It can be seen that AOD simulated by the model in the control experiment (Figure 3 b, without assimilation) has been significantly underestimated over the entire region compared to AOD retrieved by MODIS (Figure 3 a). Maximum AOD values in the range 0.3–0.5 were observed in the control experiment compared to the values (0.8–1.0) observed from the satellite. After assimilation of the MODIS AOD, simulated AOD showed significant improvement and much better agreement with the observations (Figure 3 c). As expected, after data assimilation, a large enhancement in

AOD was noticed over the northwestern States of India, which brought the WRF-Chem simulated AOD closer to the MODIS AOD. Higher values of AOD observed over this region were a result of large-scale crop residue burning activities that occurred on 5 and 6 November 2017. The increment in AOD values in the analysis (at $t = 0$, assimilation cycle 0900 UTC) due to assimilation was about 1.5 to 2.0 times more than that of AOD simulated without assimilation. This increment in AOD indicates that the data assimilation system mostly leads to a positive increment in aerosol mass concentration, and it is able to adjust chemical initial conditions for aerosols efficiently.

In order to examine the performance of the assimilation system for air quality applications, time series data of hourly $PM_{2.5}$ concentrations averaged over Delhi from the control and data assimilation runs for the first day of the forecast were compared with the independent surface $PM_{2.5}$ observations in the Delhi region (Figure 4 a and b). The black line in Figure 4 shows $PM_{2.5}$ concentration averaged from the network of 21 air quality monitoring stations in the NCR operated by the Central Pollution Control Board (CPCB), IITM and IMD (Figure 1 b). Two worst air quality episodes, the first during 7 and 8 November 2017 and the second during 12 and 13 November 2017, occurred in Delhi during the simulation period. Hourly mean surface $PM_{2.5}$ concentration reached up to $770 \mu\text{g}/\text{m}^3$ during these events (daily mean $650 \mu\text{g}/\text{m}^3$). The performance statistics for the control experiment and data assimilation experiment for normal days, severe pollution days and overall simulation period was analysed. It can be seen that the control run (red colour time series) did not perform well, although it was able to capture the diurnal and day-to-day variations associated with the synoptic-scale variability. Similar to AOD, the WRF-Chem simulations without assimilation

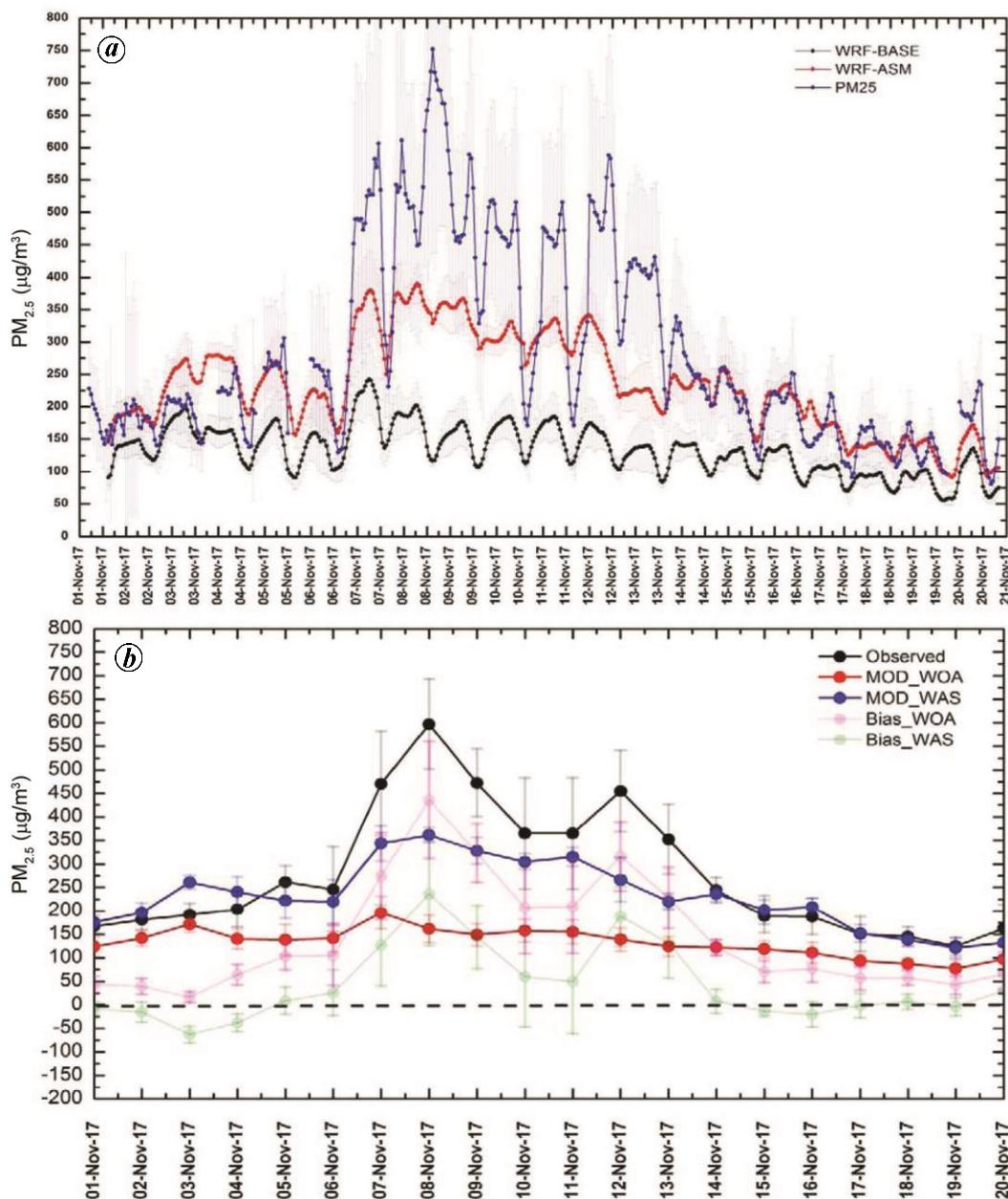


Figure 4. *a*, Hourly time series of observed $PM_{2.5}$ (black), simulated $PM_{2.5}$ without assimilation (red) and simulated $PM_{2.5}$ with assimilation (blue) during 1–20 November 2018 for Delhi. *b*, Daily mean time series of observed $PM_{2.5}$ (black), simulated $PM_{2.5}$ without assimilation (red) and simulated $PM_{2.5}$ with assimilation (blue). Green and pink lines show that biases between observed and simulated $PM_{2.5}$ daily mean concentration without and with assimilation respectively.

showed significant underestimation of the observed $PM_{2.5}$ mass concentration. The statistics showed that, in control simulations, there were larger biases of about $-150 \mu\text{g}/\text{m}^3$ for the entire simulation period, and about $-68 \mu\text{g}/\text{m}^3$ for normal days (excluding the 7–13 November 2017 pollution event). In particular, for severe pollution events (7 and 8 November 2017, 12 and 13 November 2017), when daily mean observed $PM_{2.5}$ concentration was larger than $600 \mu\text{g}/\text{m}^3$ (8 November), the daily mean bias was as high as $435 \mu\text{g}/\text{m}^3$ (Figure 4 *b*). While for the 7–13 November

2017 pollution event, the mean bias was $285 \mu\text{g}/\text{m}^3$, suggesting a significant error in simulations. After assimilation of the MODIS AOD, the time series of the hourly $PM_{2.5}$ concentration from the analysis showed much better agreement in comparison to that from the experiment without assimilation. The magnitude of bias and RMSEs decreased, and correlation increased for the surface $PM_{2.5}$ mass concentration. Bias for the first day of forecast decreased to $-48 \mu\text{g}/\text{m}^3$ for the entire simulation period and $+5 \mu\text{g}/\text{m}^3$ for normal days (excluding the 7–13 November

2017 pollution event). For the 8 November event, the bias decreased to $236 \mu\text{g}/\text{m}^3$; whereas for the entire pollution event (7–13 November 2017), the bias decreased to $133 \mu\text{g}/\text{m}^3$. This indicates that the assimilated values are closer to observations vis-à-vis control estimates. For the second day and third day of the forecast, the bias decreased to -56 and $76 \mu\text{g}/\text{m}^3$ respectively, for the entire simulation period (Supplementary Figure 2). This bias indicates that the performance statistics is better for the second day than the third day of the forecast with data assimilation. In comparison with the control experiment, WRF-Chem with assimilation was able to capture the pollution event on 7 and 8 November as well as 12 and 13 November 2017, although peak $\text{PM}_{2.5}$ concentration was still underestimated in the assimilation experiment. Enhancements of about $250 \mu\text{g}/\text{m}^3$ in surface $\text{PM}_{2.5}$ mass concentration were seen during these events when

compared to the control experiment. In contrast, WRF-Chem control runs completely failed to capture these severe pollution events. A significant increase in surface $\text{PM}_{2.5}$ during these pollution events indicated that air quality forecast could benefit substantially from the MODIS AOD assimilation.

Impact of AOD assimilation on $\text{PM}_{2.5}$ forecast and forecast verification

The improvements observed in WRF-Chem simulated $\text{PM}_{2.5}$ mass concentration due to the assimilation of MODIS AOD during 2017 simulations encouraged us to launch this system in forecasting mode from October 2018. In addition to daily forecasts, we also setup an evaluation system to examine day-to-day accuracy of our forecasts. Time series data of hourly and daily mean $\text{PM}_{2.5}$ concentrations from the first day of the forecast were compared with the air quality observations averaged from the 21 air quality monitoring stations in the NCR (Figure 5a and b). Figure 1b shows the geographical location of individual monitoring stations in Delhi. The blue lines in Figure 5a and b show averaged $\text{PM}_{2.5}$ concentration from the air quality monitoring stations, and the red lines show the simulated $\text{PM}_{2.5}$ concentration averaged over the grids covering the NCR. We have also compared the time series of hourly air quality index (AQI; red) with that forecasted (teal) by the model (Figure 5c). Hourly AQI for $\text{PM}_{2.5}$ was calculated based on National Ambient Air Quality Standard (NAAQS), and break-point concentration suggested in the CPCB notification. It can be seen that on most of the days (during the study period), average air quality in Delhi was in the ‘poor’ to a ‘very poor’ category, except for a few days when AQI was in the ‘moderate’ range. On the other hand, on 5 and 7 November 2018, 22 and 23 December 2018, and 12, 18 and 22 January 2019, exceptionally high values of mean $\text{PM}_{2.5}$ concentration (above $500 \mu\text{g}/\text{m}^3$) were observed in Delhi, with AQI in the ‘severe’ category.

It can be seen that overall the model with data assimilation performed quite well from 14 October to 23 November 2018. It should be noted that this period coincides with the active stubble-burning season in the surrounding States of Delhi, particularly Punjab and Haryana. In our simulation, emissions were adjusted based on MODIS fire data, as outlined earlier. Few previous studies have shown that winter-time fire in these States influences the air quality in Delhi under the conducive synoptic-scale conditions^{2,5}. Figure 6 shows the total MODIS fire count in the northwestern region of Delhi, the percentage contribution of CO tagged to stubble burning in the NW region to the total concentration of CO from all sources in Delhi, and total windblown dust concentration ($\mu\text{g}/\text{m}^3$) respectively. It is apparent from Figure 6a that most of the crop residue burning took place

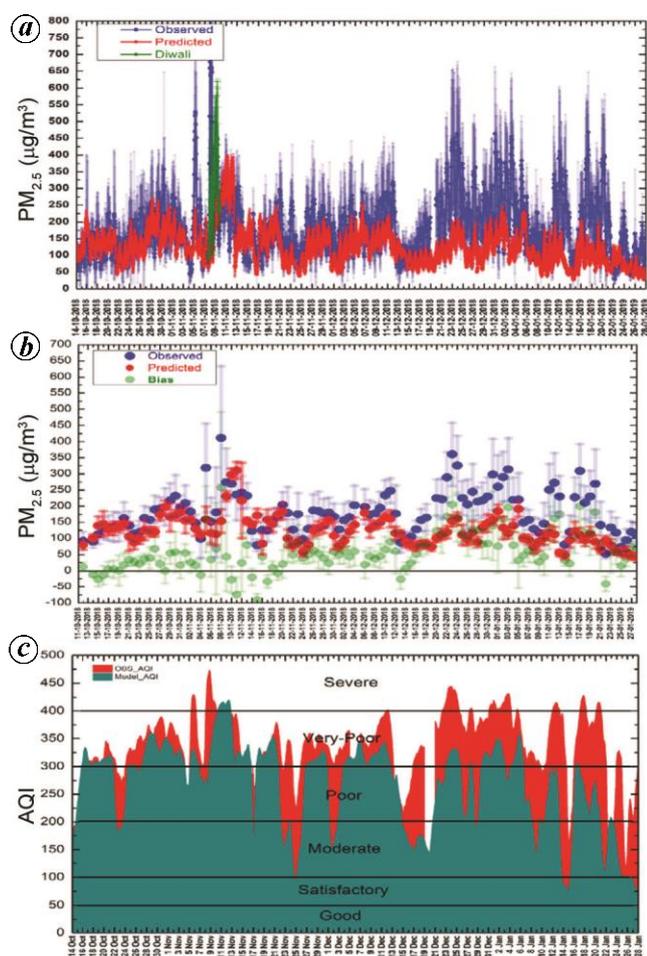


Figure 5. a, Hourly time series of observed mean surface $\text{PM}_{2.5}$ concentration (blue) and simulated mean surface $\text{PM}_{2.5}$ concentration (red) for Delhi. Green over-plot shows the simulated $\text{PM}_{2.5}$ with Diwali emissions. b, Time series of observed daily mean surface $\text{PM}_{2.5}$ concentration (blue) and simulated daily mean surface $\text{PM}_{2.5}$ concentration (red), and their difference (green) for Delhi. c, Hourly time series of observed mean air quality index (AQI; red) and simulated (teal) AQI for Delhi.

from 10 October to 23 November 2018 in the northwestern region of India. The percentage contribution of CO from stubble burning showed that fires in the NW region contributed 25–65% to air pollution in Delhi, depending upon the fire activity and prevailing synoptic-scale conditions. Larger impact (>30%) of crop residue burning on air quality in Delhi was seen from 28 October to 1 November, 6 to 11 November, and 17 to 20 November 2018. The contribution of windblown dust to total PM_{2.5} concentration was more between 14 and 26 October as well as 21 and 25 November 2018. Forecasts with data assimilation captured day-to-day variability in PM_{2.5} fairly well during the period dominated by crop residue burning.

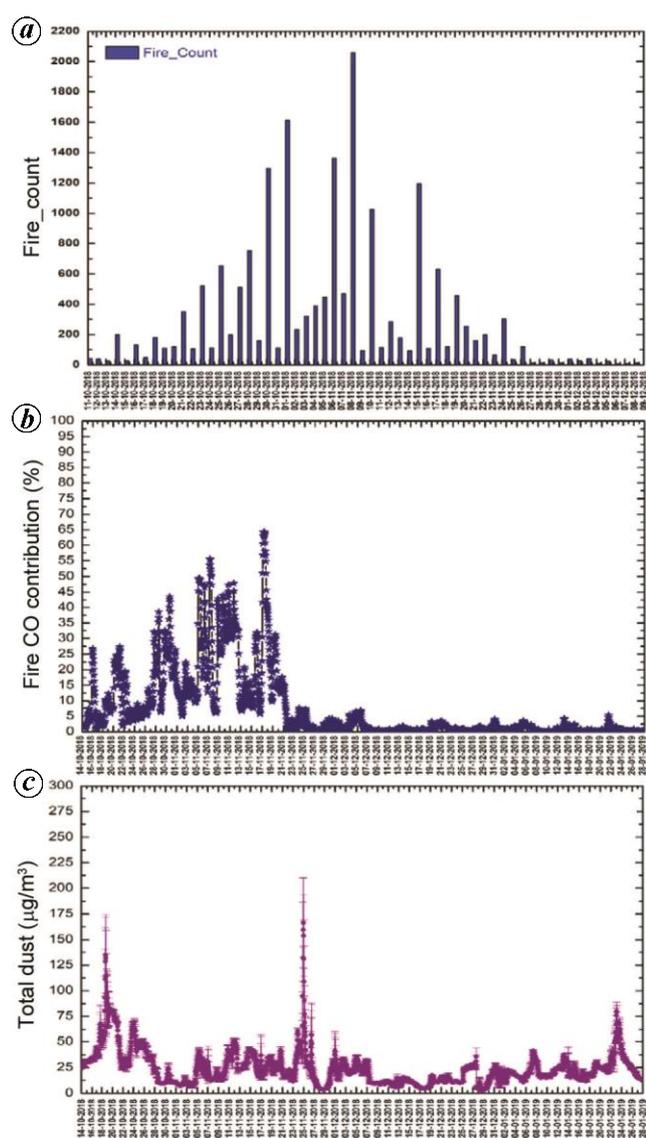


Figure 6. *a*, Time series of daily MODIS (Aqua + Terra) fire counts over the northwestern (NW) region of India for 11 October–12 December 2018. *b*, Time series of percentage contribution of CO tagged to stubble burning in the NW region to the total concentration of CO from all sources in Delhi. *c*, Time series contribution of windblown dust concentration ($\mu\text{g}/\text{m}^3$).

However, on 5 and 8 November 2018, the data assimilation system pushed the PM_{2.5} concentration towards the observations, and the model significantly underestimated the mean PM_{2.5} concentration by 160 and 258 $\mu\text{g}/\text{m}^3$ respectively (Figure 5 *b*).

On 5 November 2018, the NCR experienced widespread traffic jam from evening hours to the late-night hours that resulted in a large amount of PM_{2.5} emissions. Similarly, on 7 November 2018, there was burning of a large amount of crackers in the NCR during early night hours on the day of Diwali that contributed substantially to increased PM_{2.5} emissions. Earlier studies have also shown exceptionally high concentration of PM_{2.5} following Diwali in Delhi^{8,10}. Again with data assimilation, the PM_{2.5} concentration was close to the observations, but the model failed to predict high concentration of PM_{2.5} during Diwali festival. The mean model bias during Diwali was $-170 \mu\text{g}/\text{m}^3$ (RMSE = 270) – about 51% of the corresponding observed concentration. In order to examine the impact of firecracker emissions, we carried out an additional sensitivity experiment by doubling PM_{2.5} concentration in the NCR and all the surrounding states (Punjab, Haryana, Uttar Pradesh, Madhya Pradesh, Uttarakhand, Himachal Pradesh and Rajasthan) on the day of Diwali (green line in Figure 5 *a*). After doubling the emissions on Diwali day, the magnitude of the bias and RMSE decreased to $-32 \mu\text{g}/\text{m}^3$ and 194 respectively.

Further, it is interesting to note that the mean bias on 9 and 10 November 2018 was about $50 \mu\text{g}/\text{m}^3$, indicating that the model over predicted PM_{2.5} concentration. The AQI immediately after Diwali was forecasted in the very poor to severe category (Figure 6). In order to avert these potential air pollution episodes, decision-making authorities in the CPCB issued a public notification banning construction activities and entry of heavy vehicles (diesel vehicles, mostly trucks) into Delhi for three days. Since modelled emissions were not reduced to scale with this kind of transient activity, the forecast showed higher PM_{2.5} concentration than the observations.

Model evaluation statistics against the observations for the entire study period (14 October 2018 to 28 January 2019) showed that the magnitude of the mean model bias was $\sim 57 \mu\text{g}/\text{m}^3$ (RMSE = 93) for the first day of forecast. This bias was about 32% of the corresponding observed concentration, indicating that data assimilation was able to adjust aerosol initial conditions in a reasonable way. Model evaluation statistics for the second day of forecast showed that the mean model bias was about $\sim 61 \mu\text{g}/\text{m}^3$ (RMSE = 99). This bias indicates that the same performance persisted for the second day of the forecast (Supplementary Figure 3). For the active fire season (14 October–23 November), magnitude of the mean model bias was $\sim 23 \mu\text{g}/\text{m}^3$ (RMSE = 62), about 14% of the corresponding observed concentration. While from 26 November 2018 to 28 January 2019 (winter period), magnitude of the mean model was $\sim 79 \mu\text{g}/\text{m}^3$ (RMSE = 108),

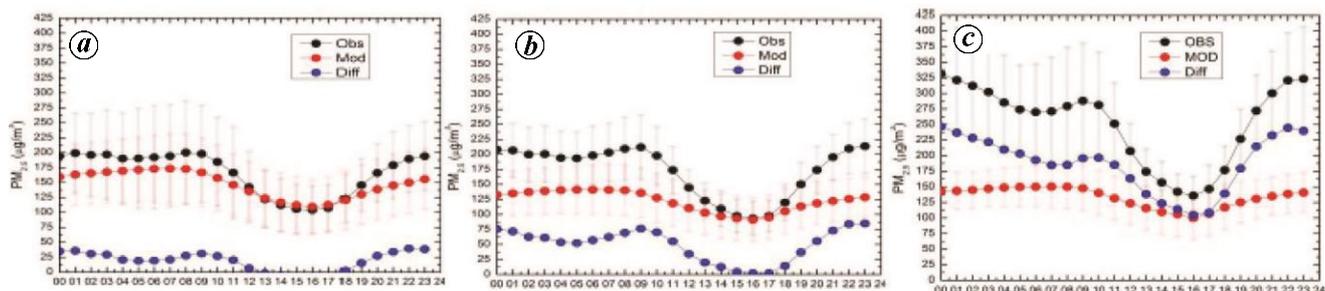


Figure 7. *a*, Mean diurnal variation of $PM_{2.5}$ for the biomass burning period (12 October–19 November 2018). *b*, Mean diurnal variation of $PM_{2.5}$ at the start of winter season (20 November–11 December 2018). *c*, Mean diurnal variation of $PM_{2.5}$ for peak winter season (12 December 2018–27 January 2019). Black colour represents the observed $PM_{2.5}$, blue the simulated $PM_{2.5}$ and red colour represents their difference.

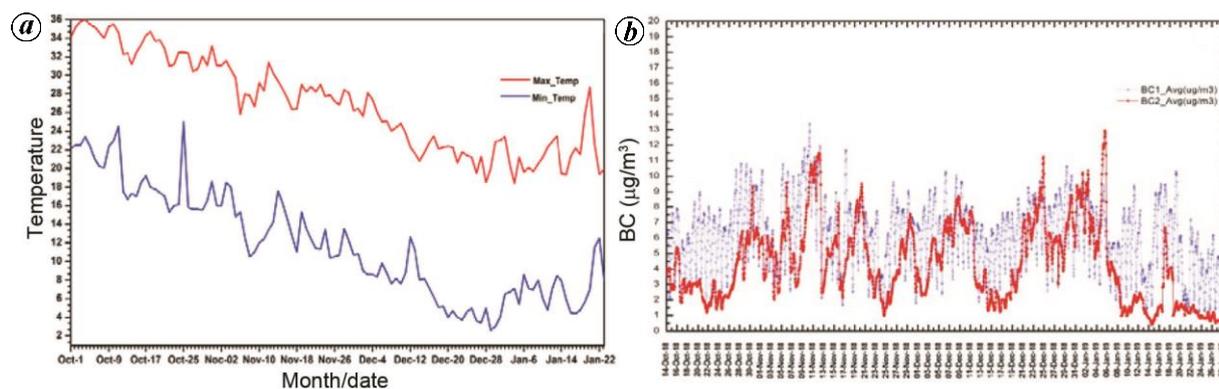


Figure 8. *a*, Time series of daily maximum and minimum temperature for Delhi (1 October 2018–22 January 2019). *b*, Time series of model-simulated BC1 (black) and BC2 (red, aged black carbon) concentrations averaged over Delhi.

about 41% of the corresponding observed concentration, indicating a factor of three increase in model bias during the post biomass burning season. In particular, model bias was substantial ($\sim 130 \mu\text{g}/\text{m}^3$) from 22 December 2018 to 2 January 2019 and from 13 to 21 January 2019, even after AOD data assimilation. The statistics indicates that the model with fire emissions and data assimilation system forecasted $PM_{2.5}$ concentration much accurately for the biomass burning season compared to the post burning season. During the winter season (December and January), systematic underestimation by the WRF-Chem forecast was seen (Figure 5a) at night-time and early morning hours in particular.

Figure 7a–c shows the mean diurnal variation of observed (black) $PM_{2.5}$, simulated (red) $PM_{2.5}$ and their difference (blue) for 12 October–19 November 2018 (biomass burning season), 20 November–11 December 2018 (start of winter season), and 21 December 2018–15 January 2019 (deep winter season) period respectively. Similarly, Figure 8a and b shows the temporal evolution of daily maximum (red) and minimum (blue) temperature and model-simulated BC concentration respectively. Model BC1 (black) and BC2 (red) represent the aged black carbon concentrations, which can be used as a proxy for the stagnant conditions or recirculation of the

same air mass from where it had originated. Higher the BC2 concentration, lesser would be the introduction of air masses from outside and this would indicate more stagnant conditions which are favourable for accumulating $PM_{2.5}$ concentration. During active stubble burning period, mean hourly $PM_{2.5}$ concentration from assimilation showed much better agreement with observations. The magnitude of bias was less than $30 \mu\text{g}/\text{m}^3$ during night-time and early morning hours, and it was close to $5 \mu\text{g}/\text{m}^3$ during afternoon hours (1200–1800 local time), indicating that the forecasting experiment benefitted substantially from data assimilation and fire emissions from stubble burning. However, simulated diurnal variation of $PM_{2.5}$ concentration deviated considerably from the observations at the beginning of the winter season, and the bias became even larger during the peak winter season. During the start of the winter season (20 November–11 December 2018), night-time temperature showed a consistent drop from about 13° to 8° . The magnitude of bias was $\sim 60 \mu\text{g}/\text{m}^3$ during night-time and early morning hours. Although forecast still performed better during afternoon hours, a bias of $\sim 10 \mu\text{g}/\text{m}^3$ indicated that the data assimilation was not able to adjust the initial conditions as efficiently during this period. One of the reasons for this could be the use of a climatological background

error in the data assimilation that would also include the effects of the crop residue burning period. When the minimum temperature dropped below 6°, the magnitude of bias increased to ~220 $\mu\text{g}/\text{m}^3$ during night-time and early morning hours, and to ~150 $\mu\text{g}/\text{m}^3$ (1200–1800 h) in the afternoon during the peak winter season. This bias indicates that the data assimilation system performed poorly during the peak winter period. To address this issue, efforts are now being made to include monthly and diurnal emissions and monthly varying background errors in the present operational forecasting system.

The increase in mean bias during the winter season, particularly during night-time and early morning hours, may be associated with the local biofuel burning during winter. During cold winter nights, open biomass burning on the streets/housing societies occurs at numerous locations in and around the NCR. People burn wood, leaf litter, garbage, plastics, tyres, etc. as these are available almost free-of-cost compared to clean energy sources (for which one would need to pay), to keep themselves warm in extremely cold conditions. Earlier studies have also shown significant enhancement in chloride (chemical tracer for garbage, plastics and tyre burning) during the peak winter season^{29–31}. Benzene and acetonitrile (chemical tracer for biomass burning) were also associated with the large increase in biomass burning during low-temperature periods³². A large drop in temperature (from 13° to 4°) during the winter months could trigger an increase in local biomass burning activity and cause a spike in air pollutant emissions. For example, spike observed in mean $\text{PM}_{2.5}$ concentration during 21 December 2018–2 January 2019 (Figure 5 b) may have been triggered by increased local biomass burning activity due to the sudden drop in temperature (from 8° to 4°; Figure 8 a), and stable and stagnant weather conditions that did not allow recirculation of air masses (Figure 8 b). The emission inventory used in this study does not include information about these sources. Therefore, the model grossly underestimates the surface $\text{PM}_{2.5}$ concentration during winter period, even after data assimilation. Another source of error during the winter season could be the availability of a fewer AOD observations that go into data assimilation. Due to frequent hazy, foggy and cloudy conditions (because of western disturbances) in the peak winter season, often AOD data are missing over the northwestern region of the IGP. These missing data limit the efficient assimilation of AOD and the quality of initial conditions. GOCART chemical mechanism does not account for nitrate and secondary organic aerosols. Ghude *et al.*²⁹ showed about 28% contribution of SOA in total $\text{PM}_{2.5}$ concentration during peak winter months. This also adds a source of uncertainty in the $\text{PM}_{2.5}$ estimate. Previous studies over India showed that WRF overestimates the boundary layer height during winter season¹⁹. The deeper boundary layer dilutes the surface $\text{PM}_{2.5}$ concentration and is an additional source of uncertainty that could

impact the forecast even after data assimilation. While the estimation of these uncertainties is important, it is outside the scope of this study.

However, in order to evaluate the impact of emissions from fugitive sources, we carried out an additional set of sensitivity experiments for two typical days during the peak winter season. Instead of using flat hourly emissions, we provided diurnal variability of emissions to the model. We first calculated the hourly ratio between observed and modelled mean $\text{PM}_{2.5}$ concentration (Figure 7 c). This ratio was then applied to $\text{PM}_{2.5}$, OC, BC, NH_3 , and SO_2 emission inventory to account for local biomass burning emissions in the forecast. The results showed significant improvement in diurnal variation of surface $\text{PM}_{2.5}$ after accounting for local emissions in the assimilation system (Figure 9). Mean model bias without local emission was 134 (± 60) $\mu\text{g}/\text{m}^3$, which reduced to 60 (± 67) $\mu\text{g}/\text{m}^3$ after accounting for local emissions. This improvement indicates that the joint adjustment of $\text{PM}_{2.5}$ initial conditions from AOD assimilation and emissions scaling is an efficient way to improve $\text{PM}_{2.5}$ forecast. Previous work also showed improvement in the short-term forecast with joint adjustment of initial conditions and emissions after the assimilation cycle^{18,20}.

Dissemination of air quality alerts

A website (<https://ews.tropmet.res.in>) was developed for archiving all the observational and prediction products, and to provide 72 h forecast to the public and officials of the Environmental Pollution Control Authority, CPCB, Delhi Pollution Control Committee and the Prime Minister's Office for taking necessary steps depending upon the requirements. This early warning system will issue alerts on large-scale air pollution events that may occur over the Delhi region (Figure 10). The air quality early

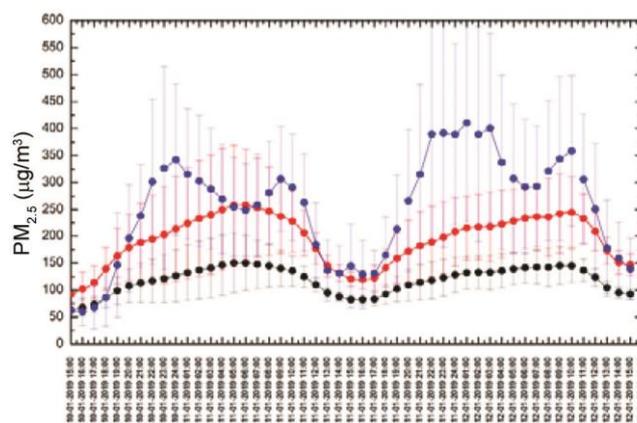


Figure 9. Diurnal variation of mean surface $\text{PM}_{2.5}$ concentration averaged over Delhi for 10–12 January 2019. Black colour represents the simulated $\text{PM}_{2.5}$, blue the observed $\text{PM}_{2.5}$ and red colour represents the simulated $\text{PM}_{2.5}$ after increasing emission.

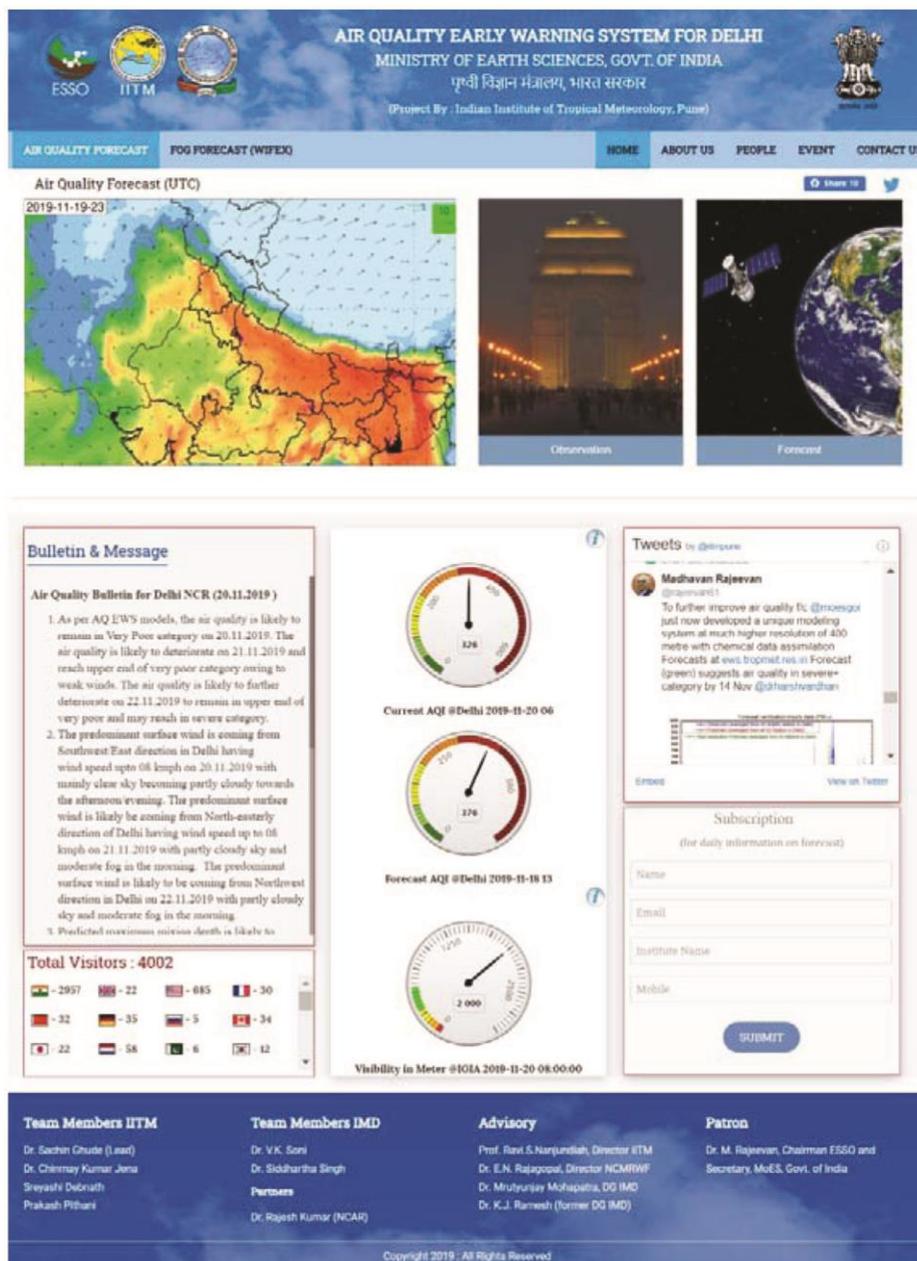


Figure 10. Snapshot of AQEWS website (ews.tropmet.res.in) developed for the dissemination of air quality information.

warning system was formally launched by the Minister of Science and Technology, Ministry of Earth Sciences, and Ministry of Environment, Forest and Climate Change on 14 October 2018. The website provides spatial animation of 72 h WRF-Chem high-resolution forecast for $PM_{2.5}$ and CO from fire emissions for the northern region (Figure 1) of India as well as for Delhi region based on data assimilation, forecast of coarse resolution $PM_{2.5}$ and CO based on Finnish Meteorological Institute (FMI) SILAM model, $PM_{2.5}$ and PM_{10} forecast based on Copernicus Atmospheric Monitoring Service product, as well as dust and AOD forecast based on the NCMRWF NCUM

model. The website also issues bulletins and warning messages for air quality conditions in Delhi. Additionally, the website provides real-time information on $PM_{2.5}$ concentration and AQI at locations in Delhi (AQMS network), latest satellite maps of fire count and AOD (MODIS), real-time boundary layer height (from Ceilometer), and 10-day forecast of ventilation index, boundary layer height and surface wind fields (products from IITM GEFS T1534 ensemble forecast system). The website also provides verification of daily air quality forecast based on the data assimilation system as described in detail in this study.

Conclusion

Here we report the development of a 3DVAR approach to assimilate retrievals of AOD from MODIS to improve the initial conditions of WRF-Chem, which was used for AQEWS at MoES, GoI, to provide operational air quality forecast for Delhi. The system was applied to take near real-time fire emission from stubble burning based on FINN v1.5 inventory. The system was designed to account for the uncertainties from meteorology and from anthropogenic emissions in the BEC matrix in the assimilation framework. In order to evaluate the impact of the inclusion of fire emissions and AOD assimilation, verification experiments were performed in hind-cast mode to model extreme pollution events that occurred during 1–20 November 2017 over Delhi. The hind-cast experiment showed that the forecast performed much better with the AOD assimilation than the forecast without assimilation (but with real-time fire emissions) for surface PM_{2.5} and AOD. For the extreme pollution events (7 and 13 November), data assimilation pushed modelled surface PM_{2.5} towards the observations, but WRF-Chem still significantly underestimated observed surface PM_{2.5} concentration. Previous studies also showed the same behaviour of underestimating PM_{2.5} while simulating extreme pollution events with MODIS AOD assimilation³³. The same assimilation system was run in a forecast mode to provide a 72 h operational air quality forecast for the NCR from 14 October 2018 onwards. The verification results showed that the forecast performance was better for PM_{2.5} for the active biomass burning period (mean bias ~23 µg/m³) than that of post biomass burning (winter) period (mean bias ~79 µg/m³). Although data assimilation during the post biomass burning period pushes modelled surface PM_{2.5} toward observations, the mean diurnal variation still showed significant bias throughout the diurnal cycle.

The present study summarizes the ongoing efforts initiated by MoES, GoI, to build a prototype chemical data assimilation in high-resolution chemistry transport model for air quality early warning system for Delhi. It has been shown that with the satellite AOD and fire (crop residue burning) data assimilation forecasted surface PM_{2.5} concentration matched better with the measurements. Although AOD assimilation had improved PM_{2.5} forecasts, the correction of emission biases may also be an important area of development for improving forecast skill. Such an application signifies the important role of chemical data assimilation in operational air quality prediction using the numerical model. Spatial and temporal variation of observed PM_{2.5} is very large in Delhi and forecast with data assimilation could not capture this variability accurately. Therefore, a very high-resolution emission inventory is necessary to be implemented in the forecasting system to capture the observed spatial variability. In addition, meteorology plays a significant role in

the accuracy of PM_{2.5} forecast, and estimated PM_{2.5} is subject to several sources of uncertainty, particularly, uncertainties in the model parameterization for PBL height³⁴, surface wind speed and choice of chemical schemes³⁵. Therefore, future studies should explore the sensitivity of these parameters to the accuracy of location-specific PM_{2.5} forecast. The MoES, GoI, will make further attempts to assimilate more observations of other pollutants from satellites and from the air quality monitoring network to improve the accuracy of predictions.

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