Simulation of aerosol fields over South Asia using CHIMERE – part-II: performance evaluation

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In this paper, we evaluate the performance of the chemical transport model 'CHIMERE' over large Indian region (4-37.5°N; 67-88.5°E) for multiple vears (2006, 2007 and 2008) by comparing the model simulations with concurrent aerosol measurements from different locations. Model simulated nearsurface black carbon mass concentrations agreed satisfactorily with measurements at various locations (oceanic, inland and island sites), in general, except during monsoon months, when the model underpredicted the measurements. Similar results were obtained when model simulated column integrated PM₁₀ mass concentrations were correlated with MODIS-derived aerosol optical depth (AOD), using AOD as a proxy for aerosol loading. The underperformance of the model during monsoon arises, at least partly, due to the model-simulated rainfall being higher than the actual rainfall over the Indian domain, during the monsoon season. Notwithstanding these, the general performance of the CHIMERE model to simulate aerosol loading over Indian domain during dry months is, in general, found to be satisfactory.

Keywords: Aerosols, black carbon, chemistry transport model.

Introduction

THE potential advantages of chemical transport models in simulating aerosol fields regionally/globally for climate impact assessment have been detailed in part-1 of the this two-part paper. Currently, chemistry transport models can perform simulations with a spatial resolution ranging from 1 to 200 km. Despite their limited spatial coverage, ground-based measurements provide accurate data against which the model simulations could be compared and validated at different time scales. Once validated, the models are specially suited for application over large spatial domains, where it is not feasible to maintain dense observational sites.

Seigneur¹ scrutinized the ability of different mathematical models in simulating pollution episodes and found that several models are able to reasonably (within 15–70%) capture the processes^{2–4}. Bessagnet *et al.*⁵ estimated the concentrations of PM₁₀ at various locations in France using the CHIMERE model and found that modelled values are closer to observations during winter than in summer. Inconsistency involved in the simulation of heterogeneous and aqueous phase processes was considered to be the chief factors contributing to the model deficiencies. Examining the signature of natural sources in PM₁₀ concentrations over Europe using the CHIMERE model, Vautard *et al.*⁶ found that the model underestimated by as much as 30-50%. Evaluating the performance of GOCART and CHIMERE models over Indian landmass for the year 2006, Moorthy *et al.*⁷ reported that, though both the models under-predicted the concentrations, CHIMERE performed better in simulating shorter scale (spatial and temporal) variations. They also pointed out the need for improvement in the boundary layer parameterization schemes to improve the predictions. Chin et al.8, simulated the concentration of aerosols with GOCART model over North America for the year 2001 and compared those obtained from 135 sites of the IMPROVE network of observational sites. They reported that the model reproduced the spatial and temporal variations of the observed sulphate accurately, but overestimated the dust and carbonaceous aerosols. They also concluded that Asian dust has a larger impact potential than African dust and is transported more efficiently than sulphate because of its elevated plume and low loss during transport. However, most of the above efforts used only very short database.

In this paper, we evaluate the performance of CHIMERE chemical transport model in simulating aerosols over India employing an extended period database of 3 years (2006–2008) and compared the results with

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concurrent ground-based measurements and satellite data. We have simulated the spatio-temporal distribution of the particulate matter mass concentration (PM_{10}), black carbon (BC) mass concentration, aerosol optical depth and the ratio of organic carbon (OC) and black carbon (OC/BC) over Indian sub-continent at several locations. The rationale of choosing this model has been its better ability to simulate shorter scale variations (as reported by Moorthy *et al.*⁷) and the wide use of this model as an air pollution forecast model and is a part of the national air pollution forecasting system⁹⁻¹⁴.

Over Indian domain we have carried out the following validation experiments: (a) Comparison of CHIMEREmodelled BC concentration with measured BC at different inland stations; Bangalore (12.97°N; 77.6°E), Tiruvananthapuram (8.5°N; 77°E) and Kharagpur (22.52°N, 87.52°E); (b) Comparison of CHIMERE-modelled BC concentration with BC concentration measurements over the oceanic regions such as the Bay of Bengal, the Arabian Sea and an island station, Minicoy; (c) Correlation between CHIMERE-modelled columnar PM₁₀ concentration (used as a proxy for AOD) and MODIS aerosol optical depth (AOD).

Model specifications and simulation domain

The specifications of the 3D-chemistry transport model CHIMERE version Chimere 2008c have been used in this study^{5,9} and the domain of its application has already been stated in part-1 of this paper. A detailed description of the model and boundary condition is available in the literature^{5,15–17}. In our present simulation studies, the MM5 model with grid resolution of $\frac{1}{2} \times \frac{1}{2}$ degree has been used to generate meteorological input files and AVN/NCEP FNL data have been used to force the MM5 model. We used a single domain simulation over India and the region of interest ranged from (3.25-38.75°N; 64.75-97.25°E) with a central grid at (21°N, 81°E) in MM5 simulations. The boundary conditions are obtained from MOZARD and/or LMDz-INCA models and aerosol boundary conditions are specified based on GOCART global simulations^{18,19}. The Global Land Cover Facility (GLCF) database has been utilized for land use²⁰ and the biogenic emissions are based on the land cover. Eight vertical levels have been used, extending from surface to 500 hPa.

The horizontal transport of chemical species is treated using the Godunov scheme²¹ and vertical transport is integrated in the model using the first order UPWIND scheme. In model calculations horizontal mixing is not taken into account and vertical turbulent mixing is considered in the boundary layer^{22,23}. Aerosols are represented depending on their size distribution and compositions, following Gelbard and Seinfeld²⁴. Dry deposition and wet deposition as well as secondary aerosol formation are considered^{25–27}. An interface with EDGAR 3.2 fast Track 2000 dataset, which incorporates anthropogenic emission of Kyoto Protocol greenhouse gases (CO₂, CH₄, N₂O and F-gases HFCs, PFCs and SF10) and air pollutants (CO, NMVOC, NO_x, SO₂) for the year 2000 on global scale, has been used to represent the anthropogenic emissions.

Observational data

The datasets used to evaluate the model simulations included those from ground-based fixed sites, from field campaigns and also from satellite (MODIS) based measurements. These stations are part of the ARFINET chain²⁸. For BC, the continuous measurements have been carried out using inter-compared Aethalometers (Magee Scientific Ins, USA) following a common measurement protocol at the chosen location. For ambient PM_{10} , we chose Bengaluru (12.97°N; 77.6°E, 920 m amsl), which is one of the fastest growing urban conglomerates in the southern central part of peninsular India. It has a population of ~8.4 million (census 2011) and more than 3.3 million vehicles plying on its roads²⁹. We have used data collected by the Karnataka State Pollution Control Board (KSPCB) from 2006 to 2008 at six monitoring stations with a frequency of 3 observations per day. A brief discussion is given below.

Ground-based measurements (ARFINET)

Regular measurements of near-surface BC mass concentration have been made from a few fixed locations representing different geographical regions (as listed below) for evaluating the model performance. These stations formed the components of the ARFINET chain²⁸. At these stations, BC mass concentrations have been measured using inter-compared Aethalometers, following a common measurement protocol. The details of these are available elsewhere^{8,30} and hence only a brief outline is provided below. The locations used in this study are described below:

Bengaluru (12.97°N, 77.6°E; 920 m amsl) is one of the fastest growing urban conglomerates in the central part of southern peninsular India, and has a population of \sim 8.4 million (census 2011) and more than 3.3 million vehicles plying on its roads²⁹.

Minicoy (8.3°N, 73.04°E; 1 m amsl) is a remote tiny island (area nearly 4.4 sq. km) station, part of the Lakshadweep archipelago, and situated in southern Arabian Sea, about 400 km off the west coast of India³¹. It represents typical oceanic location, far away from major anthropogenic emission (except for those from the fishing boats and from the households of its small population). The tiny size and low population make it representative of a rather clean marine location. BC concentration data at this location have been collected from February to December 2006. This station is also used to provide ground-based observational support for ICARB field experiments during February to May 2006 (ref. 32). The details of measurements at the above two sites are available in Vinoj *et al.*³³.

Thiruvananthapuram (8.5°N, 77°E; 3 m amsl) is a coastal semi-urban station situated at southern tip of the peninsula, very close to the Arabian Sea. The observation site is free from any major industrial or urban impact as there are no source regions in the neighbourhood. The urban area, located approximately 10 km southeast of sampling site, contributes, to a certain extent, to the BC concentrations when land breeze (offshore) prevails^{34,35}.

Kharagpur (22.52°N, 87.52°E) is a small town in the eastern part of the Indo-Gangetic Plain, ~80 km off the east coast of India and ~100 km southwest of Kolkata. The measurements have been done from the campus of the Indian Institute of Technology at Kharagpur³⁶.

ICARB measurements over the oceans

'Integrated Campaign for Aerosols, Gases and Radiation Budget' (ICARB) has been a multi-instrumented, multiplatform field experiment, conducted from March to May 2006 (ref. 37). In the ocean segment of ICARB, extensive measurements of aerosols, including BC, were carried out over Arabian Sea and Bay of Bengal from a specially configured laboratory aboard the oceanographic research vessel *Sagar Kanya*³⁸ and these data have been used in this study.

PM₁₀ and aerosol optical depth

For ambient PM_{10} , we have used the data collected by KSPCB from 2006 to 2008 at six monitoring stations with a frequency of 3 observations per day. Besides, MODIS AOD (level 3 data with $1^{\circ} \times 1^{\circ}$ resolution at 0.55 µm) from 2006 to 2008 have been used as a proxy to columnar PM_{10} as AOD measured from satellite gives the total effect of aerosol present from surface to top.

Results

*Diurnal and seasonal variation of BC/PM*₁₀ over *Bengaluru*

The monthly mean diurnal variation of BC (for the period 2006–2008) is shown in Figure 1. Although the patterns are similar, the absolute magnitudes differed significantly. The peak observed in the morning time is related to the fumigation effect^{39–41} and also to the increase in vehicular emissions, and the afternoon low is mostly attributed to increased ventilation due to deepening of the ABL^{30,36}.

The late afternoon (~15 h IST) is the hottest time of the day, and at this time the boundary layer height and turbulence are high^{41,42}. The evening peak occurs due to the nocturnal boundary layer. At night, the presence of limited turbulence activities and a shallow nocturnal boundary layer cause a nocturnal peak. Several studies have been carried out over different locations of India (Thiruvananthapuram, Kanpur, Port Blair, Delhi, Pune, Kharagpur, Ahmedabad, Visakhapatnam) and these studies have also shown a similar pattern of variation in the diurnal variation of BC, which indicates that model simulations are valid^{35,36,43,44}.

The seasonality with winter high and monsoon low also was seen in both the measurements and model. Similar to the observed BC concentrations, the modelled BC concentration reached $\sim 10 \ \mu g \ m^{-3}$ during the primary peak and $\sim 8 \ \mu g \ m^{-3}$ during the secondary peak and the diurnal low was $\sim 3 \ \mu g \ m^{-3}$ during in the winter months. During May to August, BC mass concentration decreased drastically. During these months, both peaks of the day showed more or less the same concentration. For May, the highest concentration was $\sim 2 \ \mu g \ m^{-3}$ while the diurnal low was $\sim 1 \ \mu g \ m^{-3}$. Subsequently decrease in BC concentration in both modelled and observed was seen in monsoon months where the highest concentration was lower than $1 \ \mu g \ m^{-3}$ while the diurnal low concentration was $\sim 0.5 \ \mu g \ m^{-3}$. It is important to note that model simulations were able to reproduce observed features.

Temporal variation of modelled PM₁₀ concentration over Bengaluru was similar to the observed values (Figure 2). The highest concentrations were observed in the winter months and the lowest in the summer months. As a one-to-one comparison of modelled and observed PM10 was not possible and PM₁₀ measurements were available only three times a day, the daily averaged modelled and observed PM₁₀ concentrations are compared in Figure 3. It emerges that the simulated PM₁₀ concentration is underestimated by a factor of 2 and in Figure 3, an offset value was added to compare the modelled and observed PM_{10} daily variation pattern. Despite large day-to-day variability in the PM₁₀ concentrations, the general features of the temporal variation of modelled PM₁₀ were similar to the observed PM₁₀ concentrations. Other studies have also reported that the CHIMERE model underestimates the PM_{10} concentration by approximately 30–50% (refs 5, 6).

Model versus observations

Figure 4a shows a scatter plot of the modelled daily mean BC concentration at Bengaluru, against the corresponding daily mean values from measurement and the line shows the least square fit. The general agreement is fairly good, with a correlation coefficient of 0.51 and slope of 0.61, indicating that the simulations are underestimate (by a mean value of ~1.6), despite the general



Figure 1. Diurnal variation of model simulated monthly BC over Bengaluru: a, modelled BC; b, observed BC (ng m⁻³).



Figure 2. Diurnal variation of model simulated monthly PM_{10} over Bengaluru ($\mu g m^{-3}$).

agreement in the nature of variations. In view of the distinct seasonality of the meteorology and advection pathways, we separated the data for the dry (October-May) and wet (June-September) seasons and made separate scatter plots in Figure 4b and c. This resulted in a better agreement for the dry months (slope of 0.77 and correlation coefficient of 0.63) showing the improved performance of the model, while the association became very poor in the wet season with a correlation coefficient of 0.34 and slope 0.044 showing a poor performance of the simulation during monsoon season. This has been corroborated by repeating similar analysis for Kharagpur, where we notice a fairly tight correlation (as compared to Bengaluru) with a correlation coefficient of 0.75 and slope of 0.68 when whole year data were compared (Figure 5a). Figure 5b and c showed separately the data for dry and wet season.

At the semi-urban coastal station Thiruvananthapuram, Figure 6 *a* shows a still better performance by the model with a correlation coefficient of 0.77 and slope of 0.8. Figure 6 *b* and *c* shows the seasonally separated data for dry and wet seasons, the correlation for dry season was observed as ~0.7 (slope decreased to 0.6) and for wet season 0.4, though the simulations were largely underestimate (slope 0.13), much similar to those seen at Kharagpur.



Figure 3. Inter-annual variation of daily concentration of modelled PM₁₀ over Bengaluru: *a*, 2006; *b*, 2007; *c*, 2008.

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Moving over to the island location, Minicoy, Figure 7 *a* shows that though the model simulated the variations fairly well with an overall correlation of 0.61, the simulated values were largely over-estimated (slope ~1.4), which is in sharp contrast to those seen over the mainland. Even after separating into seasons (Figure 7 *b* and *c*), the correlation remained the same for dry months (0.65), the overestimation increased (slope 1.8), while during wet season, the model totally failed to reproduce either the values or the variations (Figure 7 *c*).

Examining the seasonality it emerges that, while the model performed fairly well at all the stations during dry months, its performance was unsatisfactory during the wet season; both in reproducing the variabilities and the magnitudes. Composite plots of the correlations between



Figure 4. Comparison of modelled BC and measured BC (smoothed 3 day running mean) over Bengaluru for 2007-08: a, all months; b, January–May, October–December; c, June–September.

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simulated and observed BC concentration (3-day running average) over these stations are shown in Figure 8. Figure 8 *a* gives the correlation values over various stations throughout the year, Figure 8 *b* shows the same for months other than monsoon and Figure 8 *c* gives correlation for monsoon months. We can notice from this figure that Kharagpur and Thiruvananthapuram show high correlation (~0.75) even for the whole dataset. Even at Minicoy (~0.6) and Bengaluru (~0.5), the relation is fairly good, considering that the modelled BC concentrations values were over a grid of $0.5^{\circ} \times 0.5^{\circ}$, while measurements were at point-locations. A scatter plot (not shown here) of these variables revealed clustering of points and a careful examination of this revealed that data for monsoon months formed a separate cluster. Hence we separated data pertaining to the monsoon months from the total to see the impact on monsoon months in the total correlation. Removal of data of monsoon months resulted in an improvement in the correlations of Bengaluru and Minicoy are ~0.65 while for Thruvananthapuram ~0.7 and Kharagpur ~0.65 correlations, though reduced slightly, remained high. However the correlation was very low for the monsoon months over all the stations, being as low as ~0.24 over Minicoy. Considering the instrumental uncertainty of ~10% and the limitation of point measurement versus grid measurements, this comparison indicates a reasonably good validation of model performance over land and ocean, especially during the dry seasons. In the monsoon months modelled BC concentration was very





Figure 5. Comparison of modelled BC and measured BC (smoothed 3 day running mean) over Kharagpur for 2007–08: a, whole year; b, January–May, October–December; c, June–September.

Figure 6. Comparison of modelled BC and measured BC (smoothed 3 day running mean) over Thiruvananthapuram for 2006–07: a, whole year; b, January–April, October–December; c, May–September.

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low compared to measured BC concentration consistently. One of the possible causes could be that the model simulated rainfall was higher than the actual leading to higher washout. This underestimation of pollutants (BC and PM_{10}) in model simulation is a drawback of the model in the present form. A detailed study should be performed with the meteorological scheme to explore more about the rainfall pattern and its comparison with actual rainfall.

Ship-borne measurements

After evaluating the performance of CHIMERE over fixed locations, we compare the simulations with the campaign data, i.e. ship-borne measurements from ICARB.



Figure 7. Comparison of modelled BC and measured BC (smoothed 3 day running mean) over Minicoy for 2006: *a*, whole year; *b*, February–April, November–December; *c*, May–October.

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There are some issues with this comparison which could be neglected at fixed site. These are (i) varying location of observational location as the ship moves, (ii) temporal variation is superposed with spatial variation. Simulation results had grid resolution of 55 km and temporal resolution of 1 h. Hence to compare both datasets, we averaged cruise data falling in the model grid and in a particular hour. Figure 9 shows a fairly good correlation (with coefficient 0.48 and slope 0.63) between measured and simulated BC concentrations; however there is a lot of scatter. Separating the data into the two oceanic regions – Bay of



Figure 8. Correlation between modelled and observed BC concentration over various stations: a, whole year; b, whole year except monsoon month; c, monsoon months.



Figure 9. Scatter plot of black carbon concentration observed during ICARB experiment over Oceanic region near Indian continent and CHIMERE simulated black carbon concentration for the same location and time.



Figure 10. Scatter plot of black carbon concentration observed during ICARB experiment and CHIMERE simulated black carbon concentration for the same location and time, over (a) Bay of Bengal, (b) Arabian Sea.



Figure 11. Scatter plot of MODIS AOD and CHIMERE simulated PM10 concentration over Indian region (4–37.5°N; 67–88.5°E) for 2006–2008: *a*, January; *b*, February; *c*, March; *d*, April; *e*, May; *f*, June; *g*, July; *h*, August; *i*, September; *j*, October; *k*, November; *l*, December.



Figure 12. Correlation between MODIS AOD and CHIMERE simulated PM10 concentration over Indian region (4–37.5°N; 67–88.5°E) for 2006–2008.

Bengal and Arabian Sea respectively, in Figure 10 a and b, it is interesting to note that the correlation and slope increased for Bay of Bengal (correlation coefficient 0.55; slope 0.84), revealing a better performance of the model than over Arabian Sea (correlation coefficient 0.44; slope 0.24), where though the model captured the variations fairly well, the values are too much under-estimated.

Columnar PM₁₀ mass: model versus MODIS AOD

To evaluate the performance of CHIMERE over the study region in simulating columnar PM₁₀ concentration (aerosol loading), we used spaced-based measurements of AOD from MODIS as a proxy for PM₁₀. For this comparison we used level 3 MODIS derived AOD. Though AOD is an optical property, it could be used as a measure of aerosol column mass loading (with a reasonable assumption of vertical homogeneity in aerosol type). Monthly datasets of PM₁₀ and MODIS AOD have been compared for the three years (2006–2008) (Figure 11) and correlation coefficients corresponding to each month (average for all three years) have been shown in Figure 12. This figure indicates reasonably good agreement during the dry months (similar to the case with BC) while for monsoon months the agreement becomes poor. Except for the monsoon months, correlation between MODIS AOD and columnar PM_{10} concentration was higher than 0.5. Highest correlation ~0.7 was observed during premonsoon (April-May) and lowest correlation was observed during the peak monsoon (July).

Conclusions

We have evaluated the performance of a chemical transport model 'CHIMERE' over Indian domain by studying the spatial and temporal heterogeneity of PM_{10} and BC mass concentration over large Indian region (4–37.5°N; 67–88.5°E). Model simulations are made for three consecutive years, 2006, 2007 and 2008. Reliability of simu-

lation results has been studied by validating it with various observed data. The major findings of the study are listed below:

- Comparison of modelled BC with measured BC at various locations (oceanic, inland and island sites) showed that except monsoon months, the performance of the model was satisfactory. The possible cause for the underestimation of BC concentration during monsoon months is because the model rainfall is more than the actual rainfall over the Indian domain.
- Comparison of PM₁₀ particulate mass concentrations simulated by the model versus those measured showed good agreement.
- Correlation analysis between column PM₁₀ particulate mass concentrations simulated by the model versus MODIS AOD showed good correlation throughout the year except for the monsoon months.
- This study indicates the ability of CHIMERE model to simulate aerosol loading over Indian domain. However, it appears that improvement is required in the model in order to capture the aerosol concentration during the monsoon months.
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