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Performance evaluation of chemistry transport models over India

K. Krishna Moorthy [a], S. Naseema Beegum [a,b], N. Srivastava [c,1], S.K. Satheesh [c,d,*], Mian Chin [e], Nadege Blond [f], S. Suresh Babu [a], S. Singh [b]

[a] Space Physics Laboratory, Vikram Sarabhai Space Centre, Thiruvananthapuram 695022, India
[b] Radio & Atmospheric Sciences Division, National Physical Laboratory, CSIR, New Delhi 110012, India
[c] Centre for Atmospheric and Oceanic Sciences, Indian Institute of Science, Bangalore 560012, India
[d] Divecha Centre for Climate Change, Indian Institute of Science, Bangalore 560012, India
[e] NASA Goddard Space Flight Center, Greenbelt, MD 20771, USA
[f] Laboratoire Image Ville Environnement, ERL 7230, Faculté de géographie et d'aménagement, 3, rue de l'Argonne, 67000 Strasbourg, France

HIGHLIGHTS

- A space-time synthesis of aerosol black carbon is generated over India.
- The strong seasonal variations observed, with a winter high and summer low.
- Highest BC concentration was observed over the Indo-Gangetic Plain.
- Chemistry transport model simulations deviates from the measurements.
- An improvement in the ABL parameterization scheme might improve model predictions.

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ABSTRACT

Using continuous and near-real time measurements of the mass concentrations of black carbon (BC) aerosols near the surface, for a period of 1 year (from January to December 2006) from a network of eight observatories spread over different environments of India, a space-time synthesis is generated. The strong seasonal variations observed, with a winter high and summer low, are attributed to the combined effects of changes in synoptic air mass types, modulated strongly by the atmospheric boundary layer dynamics. Spatial distribution shows much higher BC concentration over the Indo-Gangetic Plain (IGP) than the peninsular Indian stations. These were examined against the simulations using two chemical transport models, GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport) and CHIMERE for the first time over Indian region. Both the model simulations significantly deviated from the measurements at all the stations; more so during the winter and pre-monsoon seasons and over mega cities. However, the CHIMERE model simulations show better agreement compared with the measurements. Notwithstanding this, both the models captured the temporal variations; at seasonal and sub-seasonal timescales and the natural variabilities (intra-seasonal oscillations) fairly well, especially at the off-equatorial stations. It is hypothesized that an improvement in the atmospheric boundary layer (ABL) parameterization scheme for tropical environment might lead to better results with GOCART.

1. Introduction

Black carbon (BC) aerosols are emitted from the incomplete combustion of carbonaceous materials. They strongly absorb solar radiation over a wide spectral band thereby contributing significantly to global warming (Jacobson, 2000; Hansen et al., 2000; Ramanathan and Carmichael, 2008; Bergstrom et al., 2010), besides dimming the Earth's surface (Satheesh and Ramanathan, 2000). This would affect the atmospheric stability and the hydrological cycle (Flanner et al., 2007; Koch et al., 2009; Ramanathan and Carmichael, 2008). The heterogeneous source distribution of the species has varying regional significance (Shindell and Faluvegi, 2009). The estimated radiative forcing of BC is +0.34 W m^{-2}, which is 20% of that of carbon dioxide (+1.66 Wm^{-2}) (IPCC, 2007). However, the uncertainties in its radiative forcing estimates remain still high due to the mixing of BC with other species, which

* Corresponding author. Centre for Atmospheric and Oceanic Sciences, Indian Institute of Science, Bangalore 560012, India. Tel.: +91 80 22933070; fax: +91 80 23600865.
E-mail addresses: satheesh@caos.iisc.ernet.in, profsks@gmail.com (S.K. Satheesh).
[1] Tel.: +91 80 22933070; fax: +91 80 23600865.

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increases the absorption by 50% (Schnaiter et al., 2005; Moffet and Prather, 2009), high spatial heterogeneity in the emission rates of the species (Koch et al., 2009), and the factors such as rainout rates and vertical distribution of BC relative to clouds (Haywood and Ramaswamy, 1998; Bond, 2010; Babu et al., 2010). Including the mixing, the atmospheric radiative forcing due to BC is estimated to be about +0.46 W m^{-2} considering an emission rate of 8.2 million tons. The uncertainties in aerosol radiative forcing estimation are the key parameters for the large errors in the accurate prediction of its climate impact (IPCC, 2007).

BC assumes importance owing to its chemically inert nature, fine size and longer residence time (Gelencser, 2004; Babu and Moorthy, 2001). During this atmospheric lifetime of about a week, BC particles can get transported (by the synoptic winds) thousands of kilometres away from their source (Wolf, 1984; Moorthy et al., 2004), reaching as far as Arctic and Antarctic (Konig-LangloKing and Pettré, 1998; Chaubey et al., 2010). Besides affecting the radiation balance, weather and climate, these aerosols cause degradation of air quality, health hazards (Seinfeld and Pandis, 1998; Bowler and Brimblecombe, 2000; Schwartz et al., 1996), and affect even the crop yield (Chameides et al., 1999). All these indicate the need for accurate knowledge of the spatio-temporal distribution of BC (including the altitude profile). However, extensive measurements leading to an accurate spatio-temporal synthesis still remains at large on many regions including Asia. A synergistic approach of measurements and modelling only can lead to such improved understanding (Remer et al., 2009). In assimilating measurements (either point measurements over a long time from ground-based observatories or images from space with adequate spatio-temporal resolution) into regional aerosol models, suitable for incorporating in climate models, chemical transport models have special importance. These models provide regional/global information on the optical, chemical and physical properties of different aerosol species by combining available measurements and meteorology of different scales using a set of physics-based equations and this provide air quality management plans to improve and maintain ambient air quality (Fujita, 2006). Such models are very useful in simulating atmospheric conditions including aerosols over a larger spatial domain where the ground based measurements are sparse and examining their impacts on radiative and climate forcing. In addition, these models are essential tools to forecast the air quality. Since these model results largely rely on the set of assumptions (e.g. emission inventories, meteorological fields and so on), it is desirable to compare the model simulations with measurements as frequently as possible in space and time to validate the applicability of a given model for a particular region and/period. It also would provide new information, which could be used to fine tune/improve the model. Further, retrieval of BC concentration from satellite remote sensing algorithms remains a challenge. The ground-based measurements, therefore, are essential to evaluate both model results and satellite-based retrievals.

Despite a number of studies on BC over India (Babu and Moorthy, 2002; Babu et al., 2002, 2004; Latha et al., 2004; Sumanth et al., 2004; Moorthy et al., 2004; Tripathi et al., 2005; Satheesh et al., 2006; Moorthy and Babu, 2006; Pant et al., 2006; Moorthy et al., 2007; Nair et al., 2007; Bano et al., 2011; Srivastava et al., 2012), most of these have remained location and/or season-specific and of isolated nature. Moreover, there have not been efforts to synthesize BC over the subcontinent and to examine the spatio-temporal heterogeneity and make comparison with model simulations. This calls for a synergy of well-focused, co-ordinated and simultaneous measurements of aerosol BC from different environments and models. The ARFI (Aerosol Radiative Forcing over India) project of the Indian Space Research Organization's Geosphere Biosphere Program (ISRO-GBP) is specifically tuned to the above objective through the field campaign ICARB (Integrated Campaign for Aerosols gases and Radiation Budget), conducted during March–May 2006. As a first step, a synthesis of BC mass concentration on a regional scale using the database (for the spring season), generated from the network of eight observatories, was attempted in an earlier work (Beegum et al., 2009). In this paper, we report the details of a space-time synthesis of BC over the region based on the continuous measurements from spaced network stations and its comparison with values simulated using two widely used models, GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport) and CHIMERE. The results from observation and models are examined and possible reasons for the agreement/mismatch are discussed. Such an attempt to compare quantitatively between the measurements and model simulated values and delineating the natural variabilities in the respective time series data are being done for the first time over India.

2. Observational data

Near-real time measurements of the mass concentrations (M_B) of aerosol black carbon (BC) were carried out from eight aerosol observatories; namely, Minicoy (8.3° N, 73.0° E, 1 m msl, MCY), Trivandrum (8.55° N, 76.9° E, 3 m msl, TVM), Port Blair (11.6° N, 92.7° E, 60 m msl, PBR), Hyderabad (17.5° N, 78.4° E, 545 m msl, HYD), Pune (18.5° N, 73.9° E, 559 m msl, PUN), Kharagpur (22.5° N, 87.5° E, 28 m msl, KGP), Delhi (28.6° N, 77.2° E, 260 m msl, DEL) and Nainital (29.2° N, 79.3° E, 1950 m msl, NTL), covering Indian mainland and adjacent oceanic regions as shown in Fig. 1. Here, DEL and KGP represent urban and semi-urban locations in the Indo-Gangetic Plain (IGP); HYD and PUN represent urban locations. TVM is a semi-urban coastal station in the south India; NTL is a high altitude location in the central Himalayas, and MCY and PBR are two island locations representing respectively the Arabian Sea (AS) and Bay of Bengal (BoB). The measurements are made using Aethalometers (Magee Scientific), which were inter-compared following a common protocol prior to the ICARB campaign. While most of these sites were away from the traffic or industrial activities, Delhi and Hyderabad are exceptions, which were well within the urban centres. More details are given in Beegum et al. (2009).

The details of the Aethalometer, and its application for continuous and near-real time field measurements of BC have been discussed in several papers (Hansen et al., 1984; Babu and Moorthy, 2002; Beegum et al., 2009). The instrument samples ambient air at a preset flow rate (that varied between 3 and 4 L per minute (LPM), across the stations, depending on the typical concentrations encountered at each station; the higher flow rates were used at stations having lower BC concentration. The time base (sampling rate) was uniformly kept at 5 min. BC mass concentrations are estimated by measuring the change in transmittance (incremental attenuation, ΔATN) of the quartz fibre filter tape of the instrument, upon which the particles in the sampled air get deposited and M_B is estimated from ΔATN using the effective specific mass absorption cross-section of the black carbon (16.6 m^2 g^{-1}, for 880 nm channel), area of the sample spot, and the flow rate. The effective specific absorption cross-section accounts for the amplification of the absorption due to multiple scattering in the filter fibre matrix and the shadowing effects (respectively the 'C' factor and 'R' factor, (Weingartner et al., 2003; Arnott et al., 2005; Schmid et al., 2006)). The details are given in several earlier papers (for example, Beegum et al., 2009 and references there in). A 1 μm sharp cut cyclone inlet was used at each station to avoid coarse particles such as dust entering the instrument. For the configuration used in this study, the uncertainty in M_B was in the range 40–50 ng m^{-3}.

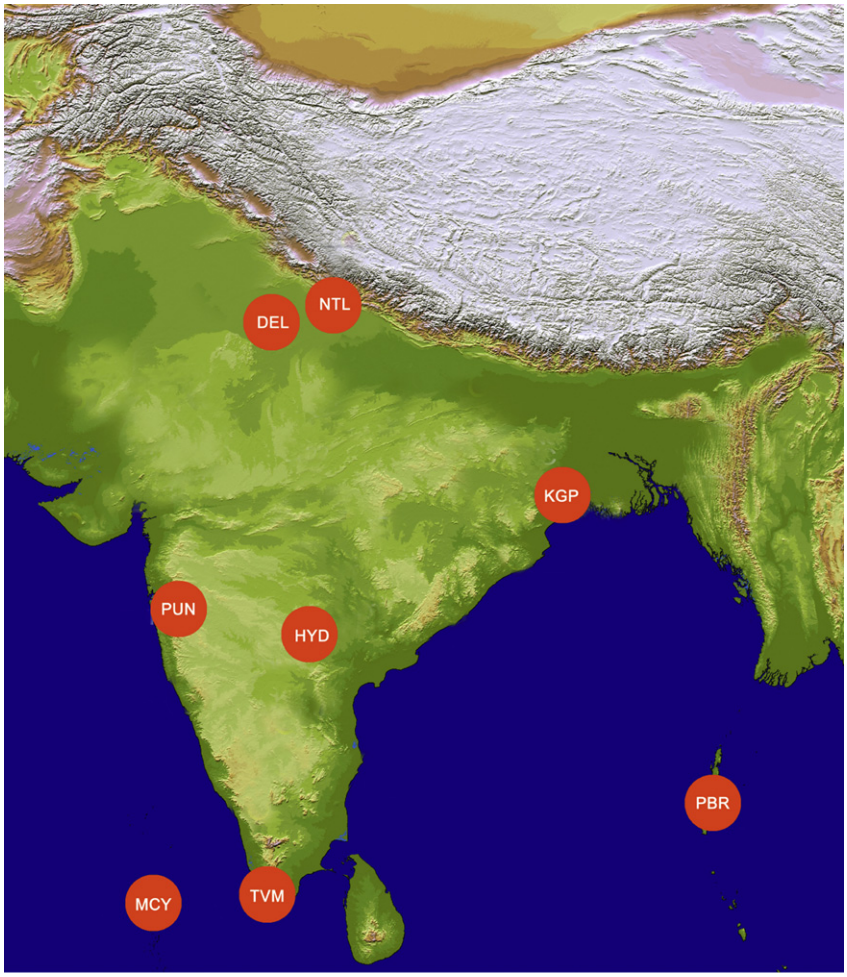

Fig. 1. The spatial distribution of the network stations having measurements of BC mass concentrations.

3. General meteorology

The prevailing wind vectors, mostly above the Atmospheric Boundary Layer (ABL), have a great significance in the efficient advection of aerosols. As such, the monthly mean wind vectors derived from NCEP (National Centre for Environmental Prediction) at 850 hPa (except for Nainital, where we used 700 hPa values) from January to December, 2006 is shown in Fig. 2. The figure clearly reveals the well-known seasonality in wind pattern, changing from easterlies or northeasterlies to westerlies or northwesterlies from winter (December to February) to pre-monsoon (March to May) to summer monsoon season (June to September). The anticyclonic circulation that prevailed over the Central India during January resulted in strong northeasterlies over the peninsula and northwesterlies over the IGP. Towards February, the anticyclone weakened and stronger northwesterlies got established over IGP and central India; and easterlies/northeasterlies, over the Peninsula, AS, as well as over BoB. During April, the wind speeds increased, and a stronger anti-cyclonic circulation appeared over the northwest AS, which drove strong northwesterlies over the AS and western coastal India. Towards June, the winds become stronger ($>$10 m s^{-1}) over the entire region with stronger south-westerlies over the oceanic regions of BoB and AS and westerlies over the Indian peninsula. This wind pattern persisted through the summer, though the speeds gradually decreased. By October, the pattern changed dramatically with very weak ($<$2 m s^{-1}) northeasterlies over the subcontinent. The wind speed depicted a considerable increase towards November/December with easterlies/northeasterlies over the BoB, as well as in the peninsular region.

4. Results and discussions

4.1. Monthly variations of BC mass concentrations

With a view to examining the nature of the mean temporal changes throughout the year as the season changes, the monthly mean BC concentration at each station have been examined and the resulting spatial distributions are shown in Fig. 3, where the diameter of each circle is proportional to the (monthly mean) value of M_B at the particular location (the mean value in μg m^{-3} is also written beside each circle). The station DEL has been given a different colour to differentiate it from the nearby high altitude station NTL, which has a drastically low value.

Significant spatio-temporal heterogeneity is easily discernible from the figure. Extremely high values are observed at the urban centres of HYD and DEL during Winter Monsoon Season (WMS, December to February) with a peak value of ~27 μg m^{-3} during January for DEL. These are attributed partly to the forest fire/biomass activities in the vicinity of the measurement location (Beegum et al., 2009). All the stations, where the continuous measurements were available throughout the year, including urban stations, showed a decreasing trend in the concentrations from

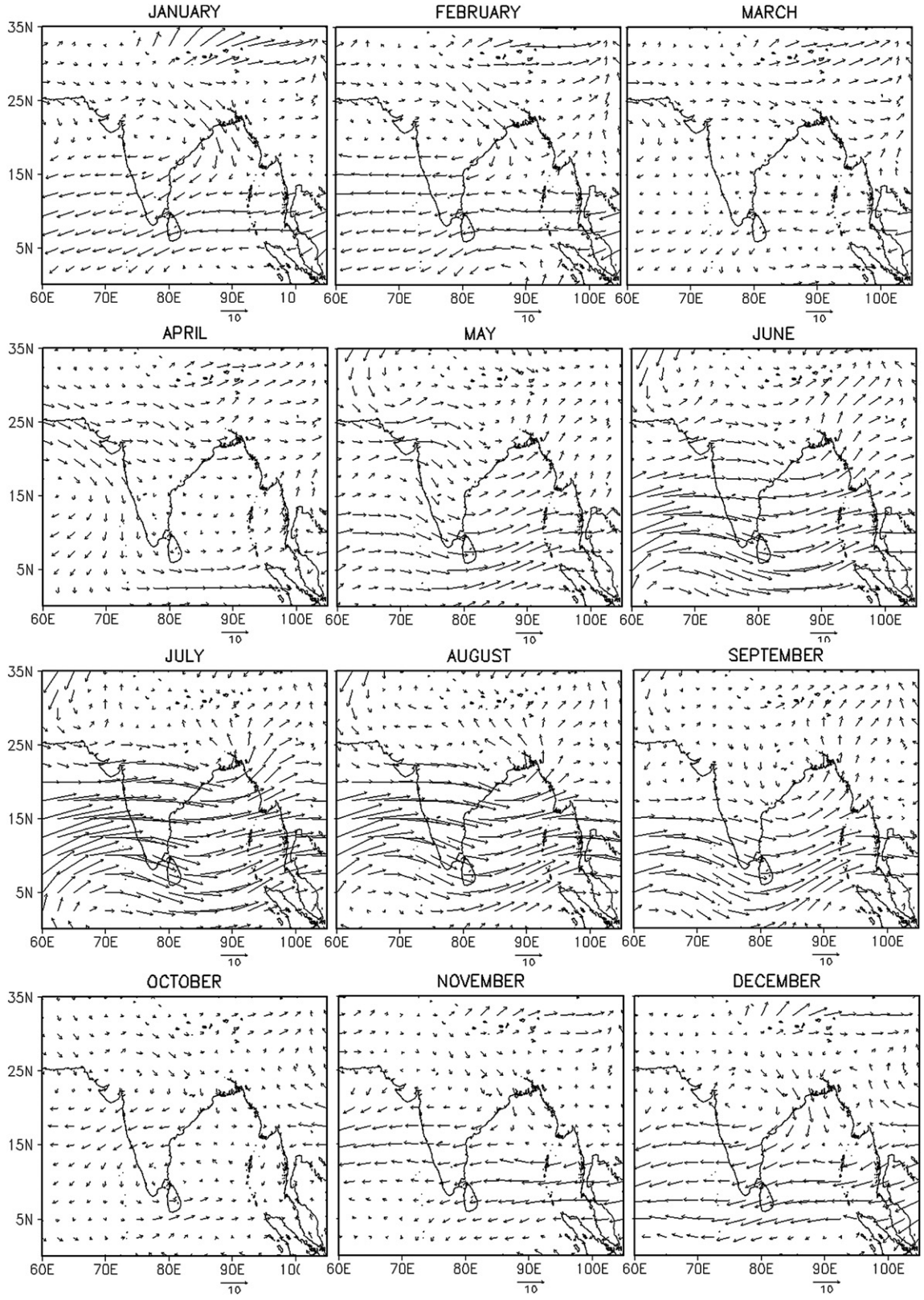

Fig. 2. The NCEP derived prevailing monthly mean wind vectors at 850 from January to December 2006.

WMS to Summer Monsoon Season (SMS, June to September). However the values of M_B observed at the urban centre HYD during SMS were much higher than those observed for other stations. This could be attributed, at least partly, to the location of the sampling site being well within the urban centre. The stations in the peninsular region registered low values in comparison with the station in the Indo Gangetic Plain (KGP) irrespective of the seasons, except the urban centre HYD. The coastal station TVM showed

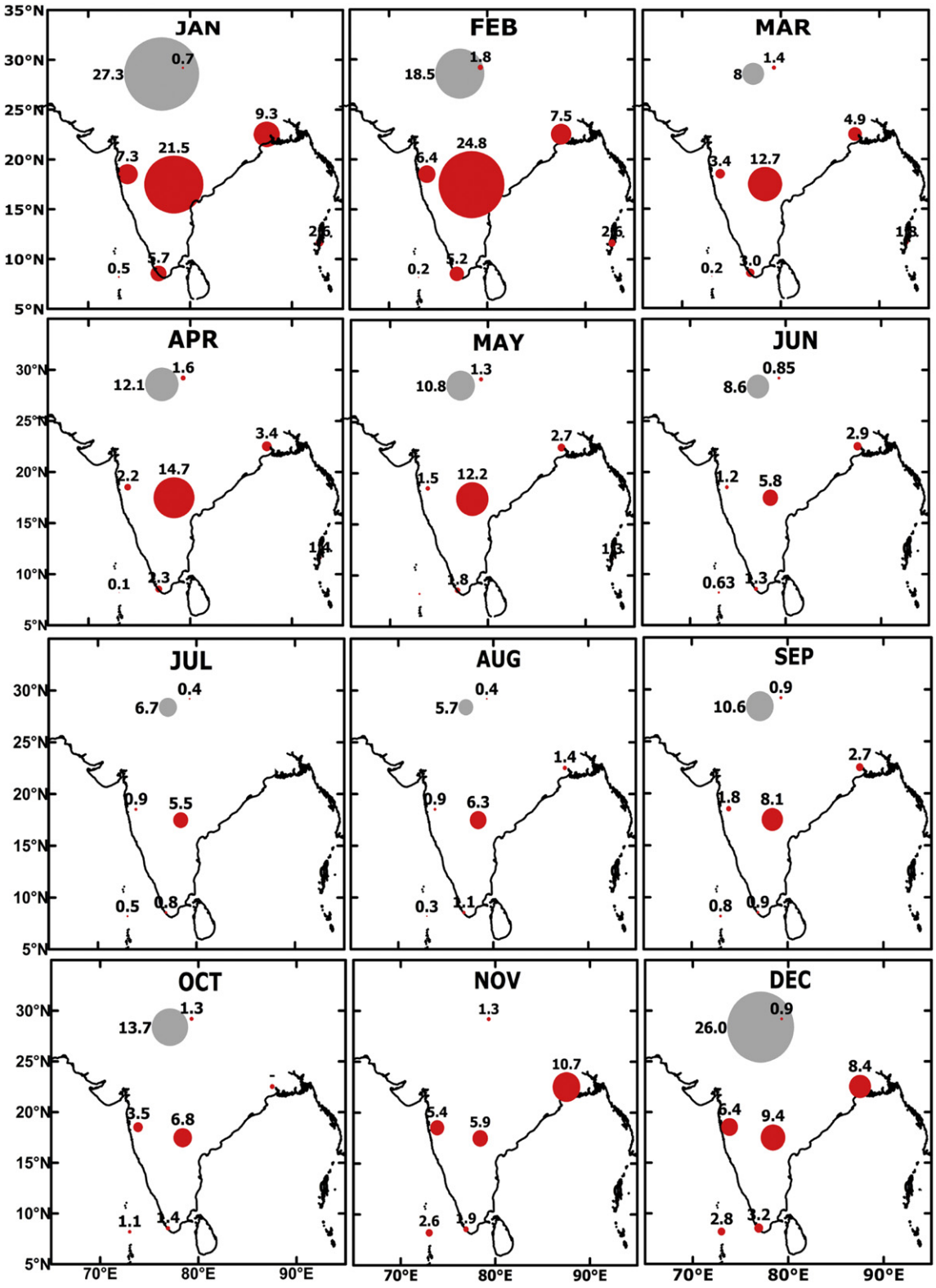

Fig. 3. The spatial pattern of M_B showing the monthly mean variations of BC mass concentrations for all the eight stations from January to December 2006. The diameter of the circles is proportional to the BC mass concentrations at the station.

lower values of M_B than those observed at similar semi-urban inland locations such as PUN and KGP. A large decrease in M_B is observed from WMS to Pre-Monsoon Season (PrMS, March, April and May) at all the stations (by a factor of 2). Possible reasons are the increase in the ventilation coefficient (V_c) from winter to summer and the change in airmass type. Ventilation coefficient, defined as the product of the boundary layer height and the transport wind, indicates the efficiency of the atmospheric

boundary layer to flush out the pollutants (Stull, 1988; Ramana et al., 2004; Nair et al., 2007). Based on extensive measurements over the IGP, Nair et al. (2007) have shown the strong correlation between changes in the BC concentrations and the ventilation coefficient. As the ABL, in general, and daytime, in particular, deepens from winter to pre-monsoon period over entire Indian landmass (due to increased solar heating) and winds also generally increases (Fig. 2), there could a steady increase in V_c from WMS to PrMS at all the stations. This would lead to a general reduction in M_B, except at the urban centres, where the local source impacts such as automobile exhaust, emissions from industrial and household activities would continue to dominate (Beegum et al., 2009). The further decrease in M_B in SMS is ascribable to the scavenging of aerosols by extensive precipitation during summer monsoon. As the removal mechanisms become weak as the season shifts to Post Monsoon Season (PoMS, October and November), M_B gradually increases. In addition to the dynamics of the ABL, emissions from the local traffic, burning of fuels (both fossil fuels and biofuels) for cooking/household activities and biomass burning/forest fire activities in the vicinity would also be contributing to the diurnal, day-to-day, monthly and seasonal variability of all the time-scales at each of the stations. However, the contribution from each of the sources varies widely depending upon the geographical location of the measurement site. For e.g, the effluence from the burning of crop residues in the vast agricultural fields of north-India could have influence on the observed high concentrations of BC during the post monsoon months at NTL and DEL.

The temporal variations are modulated by different time scale dynamical processes in the atmosphere. Among these, the well-known feature is the seasonal changes in M_B associated with the changes in the prevailing synoptic air mass types. At all the mainland stations over the Peninsula, the dry continental air mass (with northeasterly or northwesterly winds) prevailing during January/March period (Fig. 2), gradually shift to north-westerlies by April, conducive for bringing in air mass from the Arabian Sea, west Asia/western coastal India; and towards May, strong northwesterly (northeasterly over IGP) marine airmass gets established. The marine airmass prevails throughout the SMS and this contributes significantly to the low values of M_B, the shift in prevailing air mass from continental to marine, also causes a reduction in the values of M_B at all stations. Additional removal of BC takes place during this season by the extensive rainfall associated with the Indian summer monsoon, though the intensity and quantum of rainfall varies across the mainland.

Among the island locations, BC concentrations are significantly higher at PBR than MCY. This arises due to two reasons: (i) PBR is part of a longer island chain than MCY and is much more anthropogenically influenced and somewhat urbanized than MCY. It has an airport, a harbour and quite a few automobiles and is a tourist destination. (ii) Besides these local factors, significant long-range transport of accumulation mode aerosol occurs (within the boundary layer, as well as above) from East Asia, South China and East/Central India (Moorthy et al., 2003; Moorthy and Babu, 2006; Sreekanth et al., 2011), as seen from ground-based and airborne measurements. The extensive ship-borne measurements of BC during ICARB have also shown a longitudinal gradient in M_B with higher values of BC in the eastern BoB than close to the eastern coast of India (Nair et al., 2008). On the other hand, MCY is a tiny island with a land area of $\sim$10 km^2 with no major human activities other than household and fishing. Even though the values of M_B remained low over MCY throughout the year, seasonal pattern associated with

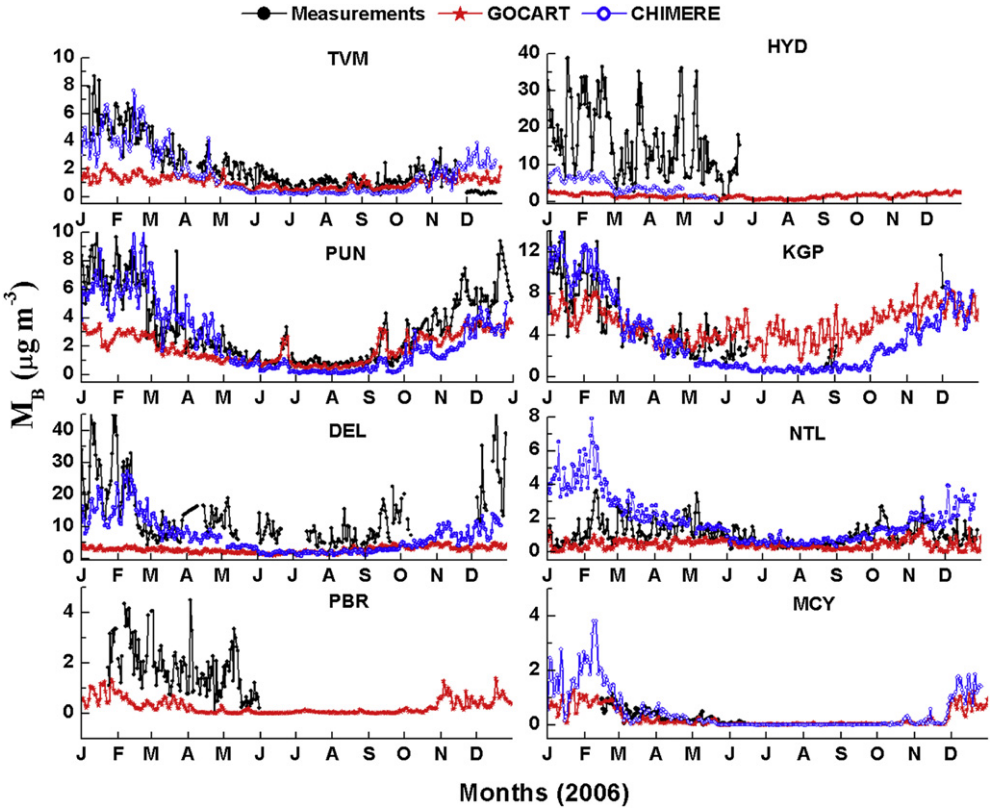

Fig. 4. Comparison between the measured BC mass concentrations and the simulations using GOCART and CHIMERE. The black curves show the measurements, the blue curve and the red curve show the model values with the CHIMERE and GOCART respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
The adjustment factor (AF) applied for different air mass period for GOCART and CHIMERE.

Stations	Continental air mass period (Nov–Mar)		Marine air mass period (Apr–Oct)	
	GOCART	CHIMERE	GOCART	CHIMERE
TVM	2.4	1.0	1.7	3.0
HYD	9.5	3.7	9.0	7.0
PUN	2.0	1.4	1.3	2.8
KGP	1.3	0.9	0.7	2.0
DEL	5.9	1.5	4.0	3.4
NTL	5.5	0.4	2.5	0.9
PBR	5.5	–	9	–
MCY	2.5	1.4	2.0	3.2

changes in airmass types is clearly discernible. Based on measurements from Maldives, in the Arabian Sea, Corrigan et al. (2006) have reported that there is an order of magnitude increase in BC mass concentrations as the prevailing airmass shifts from marine to continental.

The highland station NTL shows low values of M_B primarily due to the high altitude and remoteness of its environment. Nevertheless, the concentration of BC over NTL is significantly influenced by effluents from IGP. Consequently, the BC mass concentrations at NTL are higher than that of the island station MCY and even comparable to that of mainland stations of TVM and KGP (during May/June); despite being a high altitude station. Thus, it appears that the spatial distribution of BC over India is the combined effect of local production, modulated by the atmospheric dynamics added with the advected component from source regions. It may also be noted that the decrease in BC concentration from WMS to PrMS is limited only to the surface, resulting from the enhanced dispersion in the case of inland stations. This decrease is not necessarily due to reduction in the emissions. The BC particles near the surface that are carried by the deep convection (during pre-monsoon and summer) to higher atmospheric levels would lead to a higher BC concentration there. Airborne measurements have revealed presence of significantly higher concentrations of BC above the ABL at 2–5 km altitude (Babu et al., 2008, 2010, 2011) during pre-monsoon season. Corrigan et al. (2008) have reported the existence of high altitude BC plume, ~ at 1500 m msl, over the rather pristine Indian Ocean region during the same period (PrMS of 2006). Recently using high altitude balloon measurements (Babu et al., 2011) have shown prevalence of aerosol layers with large BC concentration in the free-troposphere, at altitude as high as 4–6 km and above.

4.2. Comparison with chemical transport Models

With a view to examining the near surface measurements over the Indian region with the values simulated by the widely used

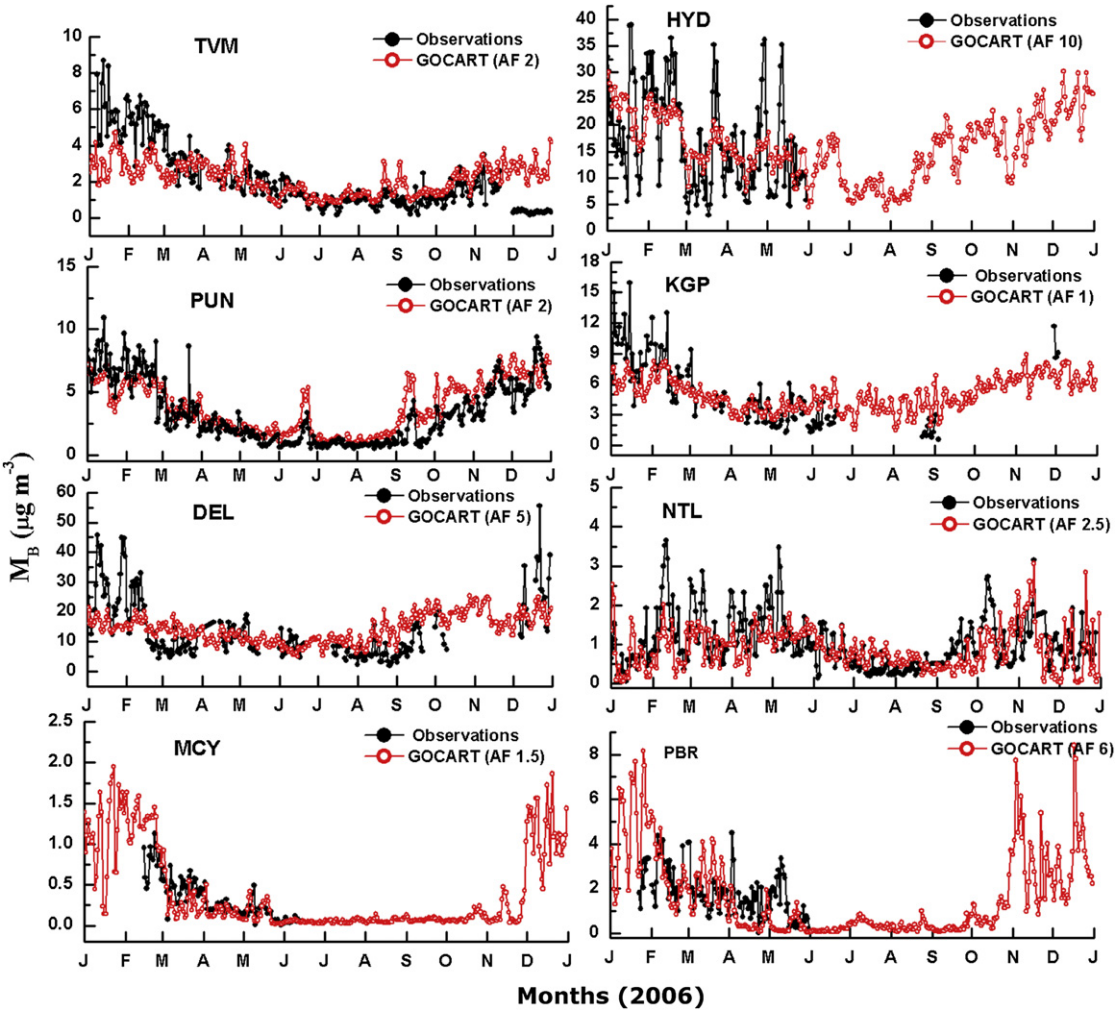

Fig. 5. Comparison between the measured BC mass concentrations and the GOCART model simulated values using the adjustment factor.

chemical transport models, we considered the GOCART and CHIMERE models.

The Goddard Chemistry Aerosol Radiation and Transport (GOCART) model is a global model that simulates major tropospheric aerosol types, including sulphate, dust, Organic Carbon (OC), BC and sea-salt aerosols (Chin et al., 2002, 2009). The emission inventories employed in GOCART is based on the compilation of inventories of black carbon (BC), primary organic carbon (OC), and SO_2 emissions from land-based anthropogenic sources, ocean-going vessels, air traffic, biomass burning, and volcanoes for the period 1980–2000 by Diehl et al. (2012). They have compiled a gridded inventory in a resolution of $1.0^\circ \times 1.0^\circ$ of land-based anthropogenic emissions of black carbon (BC), primary organic carbon (OC), and SO_2 as annual emissions from 1980 to 2007. For BC and OC, the basis for this inventory is a gridded inventory for the year 1996 from Bond et al. (2004) and yearly global emission trends for 17 regions from Streets et al. (2006, 2008, 2009). GOCART is driven by the assimilated meteorological fields from the NASA Goddard Earth Observing

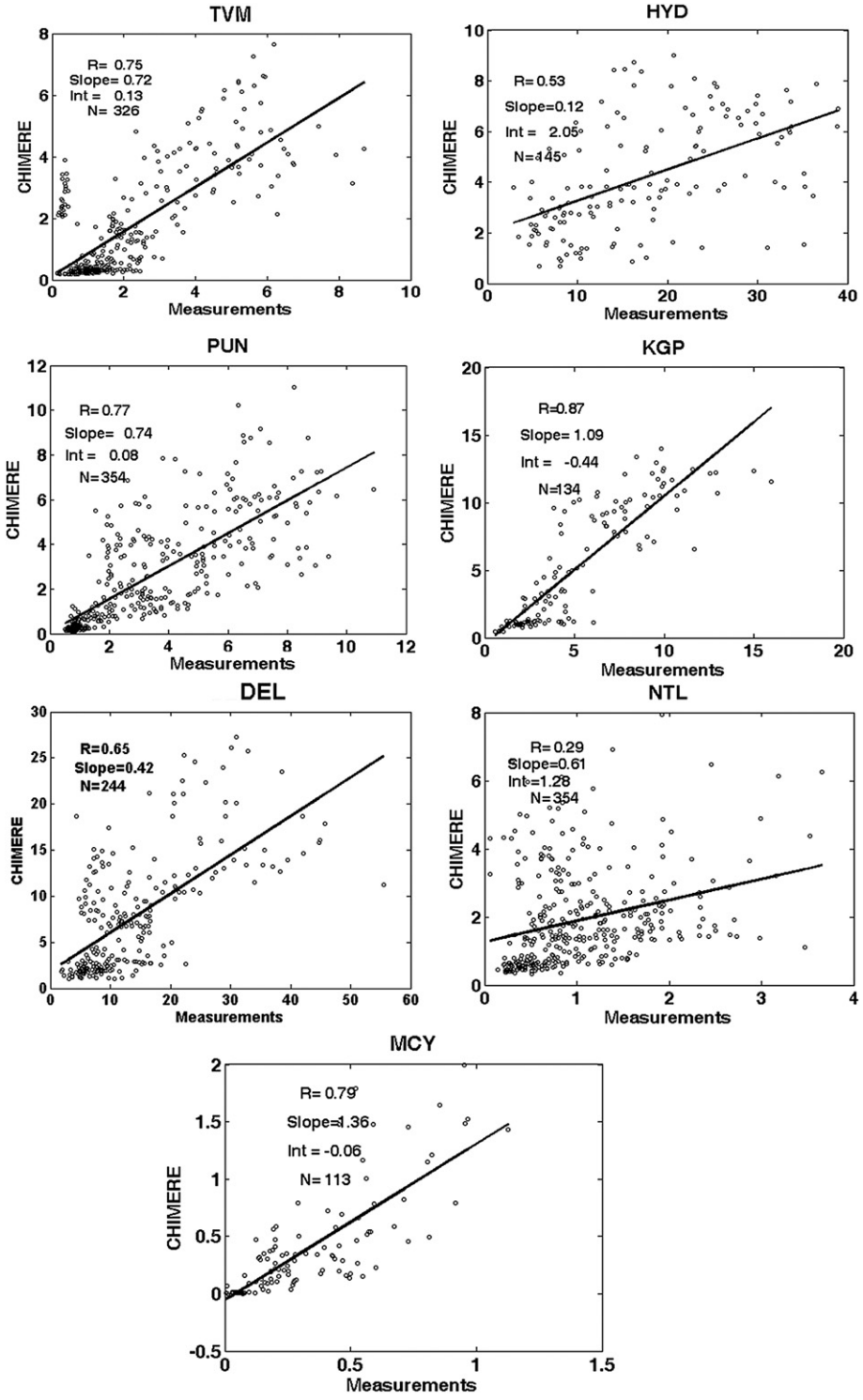

Fig. 6. Scatter plots between the measurements and CHIMERE generated values of BC mass concentrations for all the stations. Note that *x*-axis and *y*-axis range is not the same.

System Data Assimilation System (GEOS-DAS). In this work, the fields from the Modern-Era Retrospective Analysis for Research and Applications (MERRA) produced with the GEOS-DAS system (Rienecker et al., 2011) are used with a GOCART configuration at a horizontal resolution of 1.25° longitude by 1° latitude and 72 vertical levels from surface to 0.01 hPa. Processes in GOCART include emission of aerosols and precursor gases, chemistry (prescribed OH, H_2O_2 and NO_3 fields for gaseous sulphur oxidations), advection, boundary layer turbulent mixing, moist convection, gravitational settling, surface deposition, and wet scavenging. Details of GOCART model can be found in several earlier publications (Chin et al., 2000, 2002, 2009; Ginoux et al., 2001, 2004). In this study, anthropogenic emissions of BC are from the A2-ACCMIP global inventory described in Diehl et al. (2012) with emission from China and India replaced by a recent study (Lu et al., 2011). Daily biomass burning emissions are from the Global Fire Emission Dataset Version 3 (GFED, Van der Werf et al., 2010). The multi-scale model CHIMERE is designed for the simulation for aerosols, ozone and other pollutant species. Anthropogenic dataset over study region has been generated using Emission Dataset for Global Atmospheric Research (EDGAR 3.2 Fast Track, 2000 dataset). EDGAR dataset gives global estimate of emission at country level. CHIMERE can run over a range of spatial scales from the regional scale (several thousand kilometres) to the urban scale (100–200 km) with resolutions from 1–2 km–100 km. In the present study, the simulations have been done using the spatial resolution of 100–200 km, so that both the model resolutions may approximately match. The model is off-line and has to be forced for meteorology and boundary conditions. Meteorology data is not provided directly, but a CHIMERE interface with meso-scale model, WRF, has been used. For the present study, the model CHIMERE has been run from January to December 2006 over the Indian region with the forcing of meteorology, emissions, boundary and initial conditions as described by Hodzic et al. (2009, 2010), at 50 km horizontal resolution and 8 vertical levels extending from the surface up to 500 hPa. For aerosols, a set of boundary conditions is proposed based on GOCART global simulations (Ginoux et al., 2001; Valari and Menut, 2008). CHIMERE needs input on land use information, as well as biogenic emission potentials, based on land cover. The GLCF database is used for land use (http://glcf.umiacs.umd.edu/). Emissions come from local, nonpublic databases. Among the physical processes in the models, the chemical mechanism is adapted from the EMEP, photolytic rates are attenuated using liquid water or relative humidity and aerosol thermodynamic equilibrium is attained using the ISORROPIA model. Three advection schemes such as the Parabolic Piecewise Method (PPM, a three order horizontal scheme (Colella and Woodward, 1984), the Godunov scheme (Van Leer, 1979) and the simple upwind first-order scheme are implemented. Boundary layer turbulence is represented as the diffusion process (Troen and Mahrt, 1986); dry deposition is as in Wesely (1989); vertical wind is through a bottom-up mass balance scheme. Wet deposition and secondary organic aerosol formation are also considered. Six aerosol sizes are represented as "bins" in the model. As a comparative study, the time series of daily mean M_B of both the model-simulated daily mean values are shown in Fig. 4, along with the corresponding measurements for the period January to December 2006. It is very interesting to see that, at all stations, the model simulations using CHIMERE are much closer to the observations than those from the GOCART-simulated values, which are significantly lower except during summer monsoon season. However, the episodic fluctuations in the measurements at the urban centres of DEL and HYD are absent in both the model simulations. The ratio between the measurements and GOCART model simulations (termed as the adjustment factor (AF) hereafter), however, varied from 1 (for KGP) to 10 (for HYD) with lower values for remote locations without the proximity to any point sources and higher values for urban locations where the local sources are important.

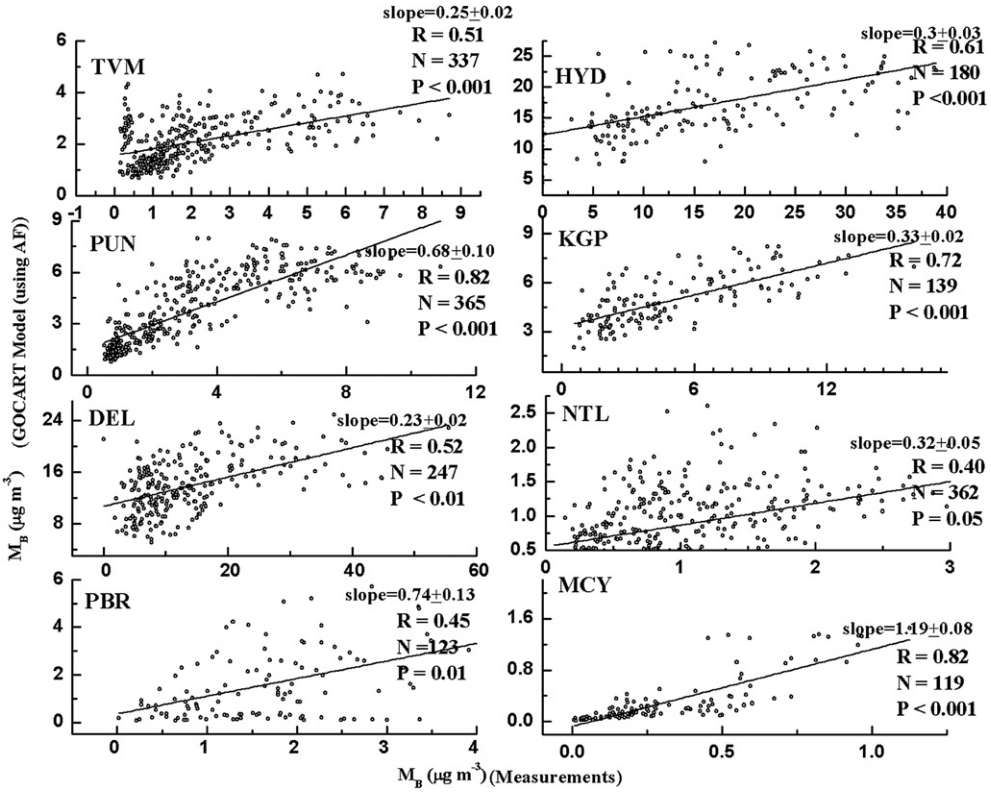

Fig. 7. Scatter plots between the measurements and GOCART generated values (multiplied by the AF) of BC mass concentrations for all the stations. Note that *x*-axis and *y*-axis range is not the same.

Whereas for CHIMERE, on an average, the AF were much lower (between 0.8 and 2.5), except at HYD, where the ratio was 5.0. This high ratio appears to be arising from the speciality of the sampling site, being surrounding by highways having extensive traffic always. At KGP, the GOCART model values are closer to the measurements even without the AF. For the urban centre HYD, all the measured values are found to be in the range of 5–35 μg m^{-3}, with very rapid fluctuations; whereas the GOCART simulations are as low as 2–3 μg m^{-3} with weak fluctuations throughout the period. The AF used for various stations are given in Table 1.

From the table above, it emerges that;

- In general, CHIMERE model agrees with the measurements better throughout. Both the models show very high deviation for the sites located within the urban centres of megacities (DEL, HYD).
- Examining seasonally, the deviations (as quantified by the adjustment factor, AF) for GOCART model are very high during winter season, when continental air mass also prevails. This might be similar to the case with RegCM 4.0 simulations of BC which also showed significant deviations from measurements during winter. Examining this, Nair et al. (2012) have attributed this high deviation to the non-local atmospheric boundary layer parameterization scheme, that tend to over-estimate the vertical distribution in the convective ABL, as the model fails to capture the stable ABL during winter, which actually confines the aerosols within the a shallow region (typically 500 to 800 m) as compared to the summer time values of 1.5–2 km. This would be more conspicuous in the continental station like DEL, HYD, NTL and PUN. Over the stations like MCY, TVM, PBR etc, where the annual variation of ABL height is not as large as the inland stations, the continental air mass leads to increased advection of BC aerosols from the regions where they are confined. In this aspect, the CHIMERE model appears to perform better as the adjustment factors are very low. Even with the adjustment factor the model simulations are not able to capture the episodic variations at HYD, especially during the winter season.
- During marine air mass period, when the monsoon dominates, the BC concentrations per se decrease over all stations. The ABL

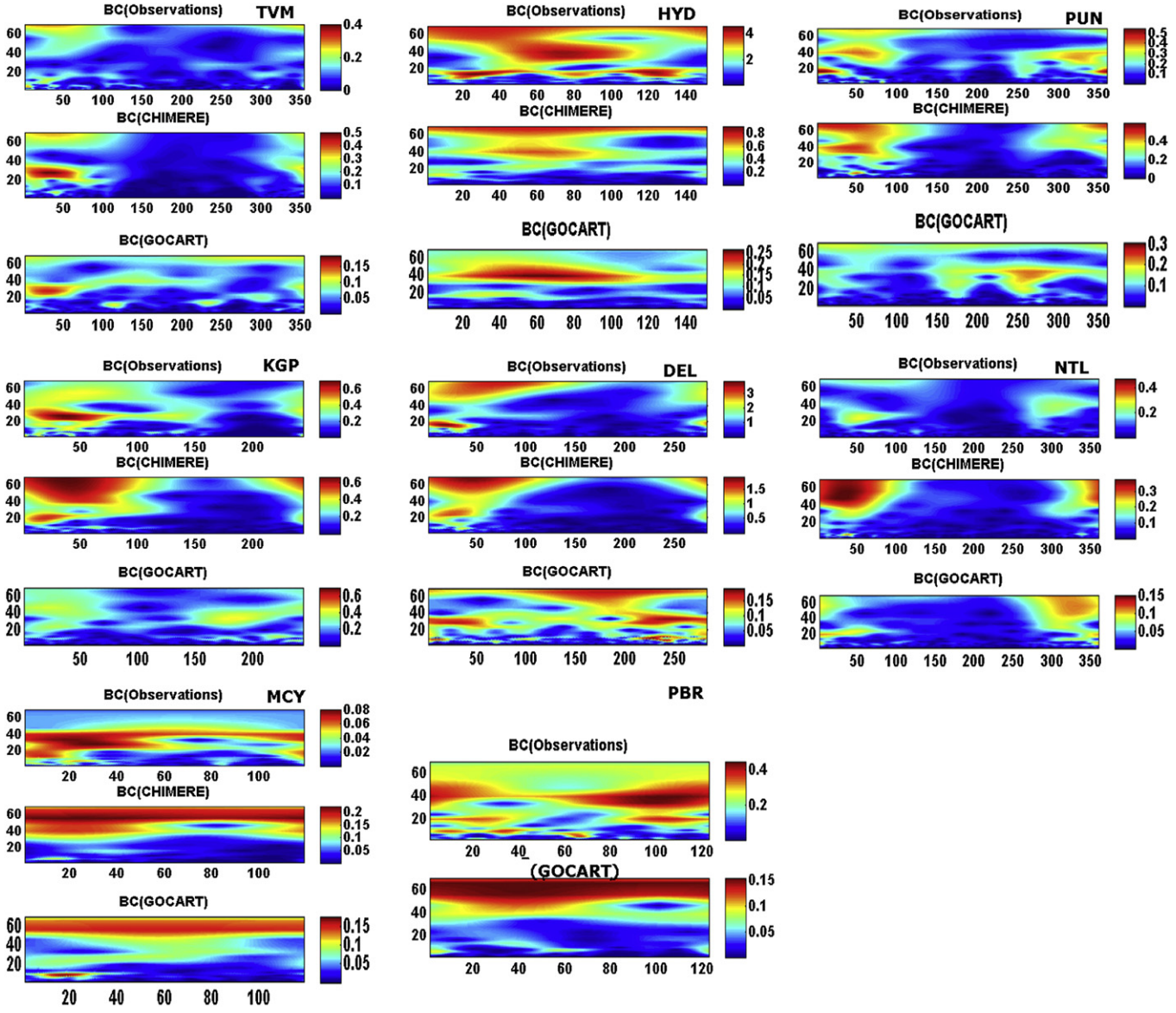

Fig. 8. Wavelet spectra of the time series of daily mean M_B at all the stations for observation and the model simulations of GOCART and CHIMERE.

has very little role to play in modulating the concentrations, as the thermal convections are highly subdued by the extensive monsoon activity throughout the country. In such conditions, the Table shows that the GOCART simulations are better representative of the measurements than the CHIMERE. However, the AFs for CHIMERE during this season, though are larger than those for GOCART, still are not as high as the GOCART factors in winter.

- As the BC concentrations are very small during monsoon season (almost 1/5 of the winter values) the contributions of this to annual pattern is small and the net result is that CHIMERE tends to simulate the regional BC better than GOCART.
- It is hypothesized that an improvement in the ABL parameterization scheme for tropical environment might lead to better results with GOCART.

Even though the simulations using CHIMERE remained low ($<10\ \mu g\ m^{-3}$), the values were found to be significantly higher than the GOCART simulated ones. Surprisingly, at other mainland stations of PUN, KGP, DEL and TVM, CHIMERE simulated values show very good comparison with the corresponding measurements, including the seasonal patterns. This observation is highly significant, as even for the urban centre DEL, the model was able to simulate the BC values realistically except a few spikes attributable by highly localized activities. As the GOCART model simulations showed significant deviation from the measured values, the temporal variations in the time series data has been examined after removing the mean shift in the simulated values by multiplying with appropriate AF for each of the stations and the resulting pattern is shown in Fig. 5. The figure clearly evidences that the model simulated values are able to capture the temporal variations at all the stations except at the urban locations.

With a view to examining how well the simulations reproduce the relative changes in the measurements, scatter plots between the measurements and models have been generated for all the stations and the results are shown in Fig. 6 and Fig. 7 respectively for CHIMERE and GOCART (using AF). The corresponding correlation coefficients (R), slopes, intercepts (Int), the total number of points (N) and the statistical significance of the fit, the 'p' values are also given in the figures (As the p values were <0.001 for all the stations, implying highly significant correlation coefficient with 99% confidence level, these values are not mentioned in the figures). Fig. 6 reveals very good correlation between the CHIMERE-simulations and measurements, except at the high altitude station NTL. Even at the urban centres of DEL and HYD, the correlations coefficients were >0.50. Except at the urban centres as well as at the high altitude station NTL, the slopes of the scatter plots were close to 1.0. The high intercepts (2.0) for the urban centres indicated, to a certain extent, the high degree of prevailing local activities, which is rather difficult for any model to capture. Similarly, the scatter plots for the GOCART simulations, multiplied by the AF, (Fig. 7) also revealed very good correlation between the measurements and model at all the stations, especially at PUN, KGP and MCY. These are stations were local source activities are generally weak or only moderate and transport plays a major role in causing the changes to BC concentration. Another interesting observation is that the correlation coefficients for the GOCART model were higher than that for CHIMERE at PUN, MCY and NTL. Notwithstanding these, the slopes of the graphs were also improved. In general, for the urban centres with significant

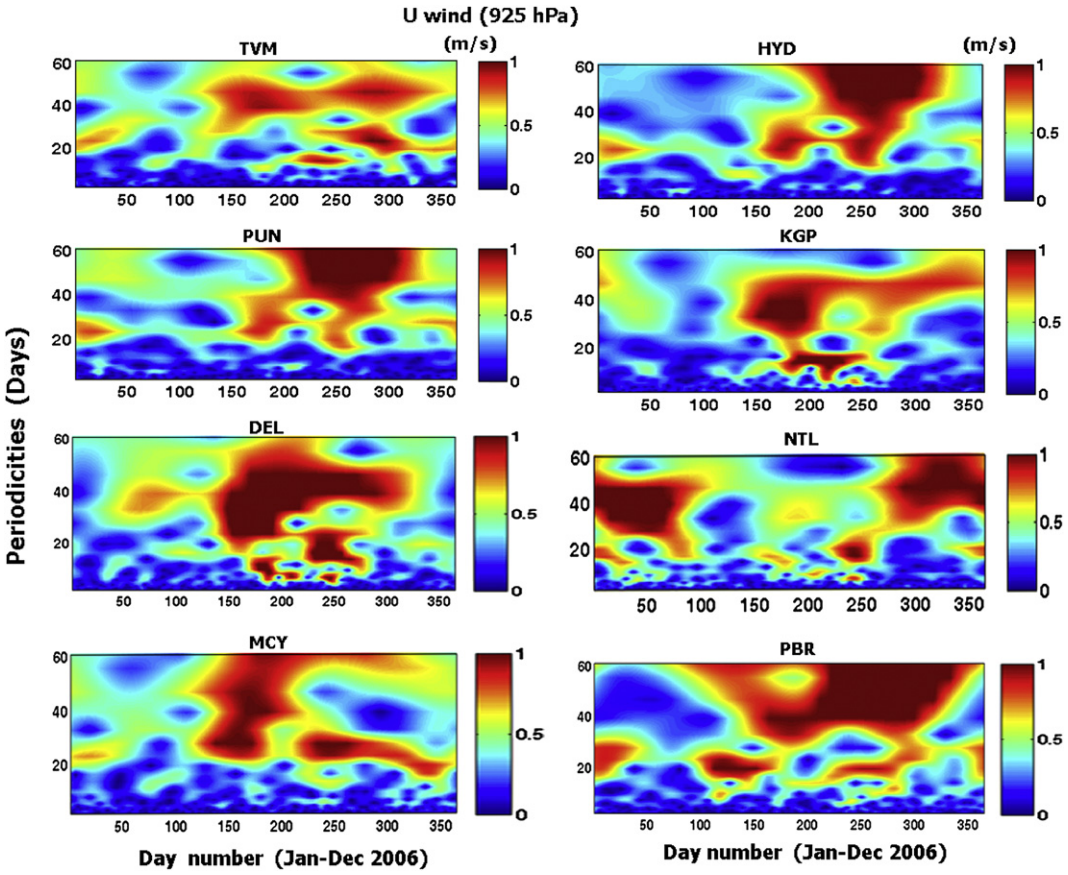

Fig. 9. Spectra obtained from time series of the daily mean zonal wind vectors from January–December 2006 at 925 hPa (NCEP derived) at all the stations, except at NTL (where the winds at 700 hPa was taken).

local anthropogenic activities and the Himalayan station NTL, the correlation coefficients were low for both models suggesting that the models does not capture even the temporal trends and did not follow the seasonal/intra-seasonal fluctuations. This was true even at the station PBR.

In general, the significant deviations between the two model simulations (Fig. 4) and GOCART with the measurements, might arise either from the difference in the parameterization of the advection processes, boundary layer schemes, or inappropriate emission inventories or a combination of all. As the GOCART model simulations have been improved significantly after incorporating the adjustment factor, it emerges that the emission inventories used in the models might be the prime factor for the large deviations. Another point to be noted here is that the spatial resolution of the two models does not match. GOCART being a coarse resolution model in comparison with the CHIMERE, the improper emission inventories that go into it might be an important factor for the much larger departure for the model. Unlike the General Circulation Models, as the chemical transport Models solve only for the continuity equation, large difference between the model simulations might arise from the difference in schemes used for the advection and diffusion processes. As such, the models may lack wave parameterization schemes and hence the modulations produced by these.

With a view to examining this, the dominant frequency components present in the model simulations as well as in the observed M_B values have been estimated by applying wavelet analysis using a Morlet wavelet (Torrence and Compo, 1998; Beegum et al., 2009a). Morlet is a complex wavelet, which can be decomposed into real and imaginary parts and the mother wavelet of the morlet is a plane wave modulated by a Gaussian of the form, $\Psi(x) = C\exp(-x^2/2)\exp(i\omega x)$, where ω is the angular frequency. The resulting wavelet spectra are given in Fig. 8 for CHIMERE and GOCART models, along with those for the corresponding measurements. The analysis revealed the presence of three dominant periodicities, 30–50 days; 13–22 days, and 9–11 days periodicity at all the stations with varying amplitudes, both in the observations and in models. All the periodicities mentioned above are within the cone of confidence (not shown in figure). In addition, shorter periodicity of the order 1 week is also observed in significant amplitudes in the measurements especially at the urban centres, which might be associated with either the episodic events such as forest fires/biomass burning activities and reduced traffic on weekends, or due to the removal of BC from the atmosphere or combination of all. The periodicities observed in the surface mass concentrations in M_B are examined in details. The analysis of the meteorological parameters within the ABL revealed the prevalence of similar periodicities in the atmosphere. Analysis of the wavelet spectra of the time series of NCEP wind vectors at 925 hPa and subsequent phase propagation estimation led to the identification of 30–50 day Madden Julian Oscillation and planetary waves of periodicities of 13–22 days (Quasi 16 day) and 9–11 days (Quasi 10 day) in the atmosphere. These periodic fluctuations in the meteorological parameters can cause either convergence (or advection) or divergence (or dispersion) and produce subsequent signatures on aerosol concentrations (Beegum et al., 2012).

With a view to examining the periodicities in the concurrent meteorological parameters, the time series of the daily mean zonal wind vectors from January–December 2006 at 925 hPa (NCEP derived) at all the stations, except at NTL (where the winds at 700 hPa was taken) have been examined and the resulting spectra is shown in Fig. 9. The analysis revealed the presence of similar periodicities such as 30–60 days, 13–22 days and 9–11 days in significant amplitudes at all the four seasons. Further analysis of the phase propagation of the periodicities as a function of longitude led to the identification of 30–50 day Madden Julian Oscillation and planetary waves of periodicities of 13–22 days (Quasi 16 day) and 9–11 days (Quasi 10 day) in the atmosphere.

It is interesting to observe that the temporal patterns of amplitudes of all the periodicities are similar for both observations and model simulations at most of the stations. At all stations, except the urban centres, the magnitudes of the periodicities in the model simulated values were higher than those of the corresponding measurements. Even though all the three periodicities were present in the wavelet spectra of both the model simulations too, the amplitudes of the periodicities were much lower than those in the measurements. As the absolute magnitudes of the M_B in the model were order of magnitudes lesser than that of the corresponding observations, a quantitative comparison between the two is difficult.

As such, the time series data at all the stations have been subjected to Fourier Transform (FFT) analysis (Cooley and Tukey, 1965). From the corresponding amplitudes of the resulting FFT spectra, the percentage contributions of the periodicities to the seasonal mean BC mass concentration (PSM) have been estimated for the measurements and models (GOCART and CHIMERE) for all the stations during the pre-monsoon season and these are shown in Fig. 10. CHIMERE domain we have used in our study is [3.25°N–38.75°N; 64.75°–97.25°E]. Since PBR falls outside the domain, simulations for PBR are not included. Despite the significant differences in the values between the stations, the PSM for all the three periodicities

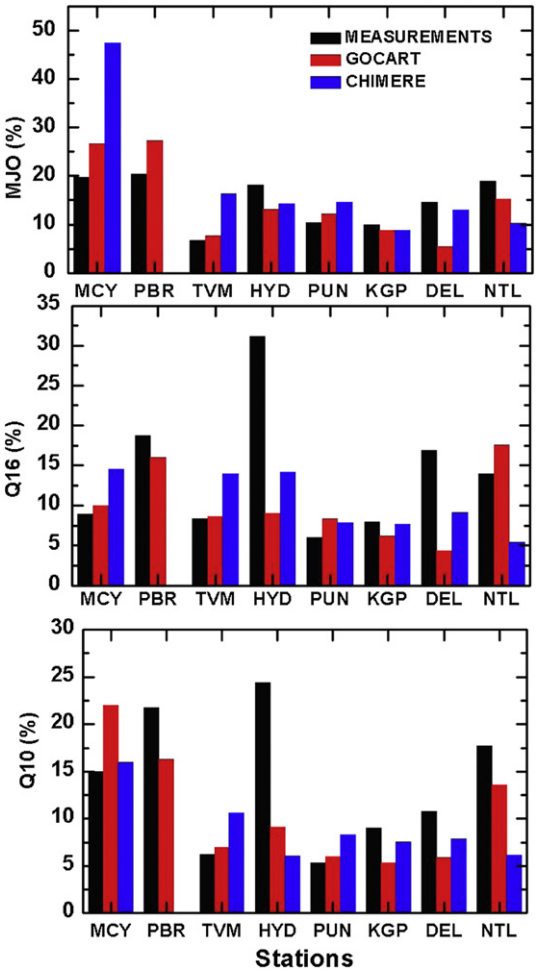

Fig. 10. The percentage contribution of the wave amplitudes to the seasonal mean BC mass concentrations (PSM) for the periodicities in the time series data at all the eight stations.

were comparatively higher at the near equatorial stations compared with the off-equatorial stations for both the models. At most of the stations, the estimated PSM values did not agree with both the model values. Comparatively closer agreement is observed for the stations KGP, PUN, TVM and MCY. At PUN and MCY, GOCART was closer to the measurements, whereas at KGP and TVM, CHIMERE values were better. As the data span is for a complete year covering all the four seasons, the seasonal pattern of the PSM values were estimated and are shown in Fig. 11 along with those from the measurements. It is interesting to observe that the PSMs were most close during the pre-monsoon season, gradually differing through the summer monsoon season, and the highest difference were noted during winter monsoon season. Even the absolute values of both the model simulations were agreeing well with the measurements during PrMs and SMS. Even though the wave activities are stronger during winter, the effects of local sources are dominating over the advection and boundary layer turbulent mixing processes during winter. Whereas during pre-monsoon and summer monsoon seasons, the atmosphere is more conducive for the efficient dispersals of aerosols (both horizontal and vertical) even within the boundary layer. At the stations, where statistically significant correlations were observed between the observations and model simulations, the discrepancy in PSM values was low, at least for two of the three periodicities.

The discrepancy between the PSM values for the two models might be due to the difference in the various parameterization schemes employed in the models. The large discrepancy in the PSM values between the measurements and models for shorter timescale periodicities at the urban centres like DEL and HYD might be either due the inadequate data on the episodic events that go into the model or due to the coarser resolution of the model. In GOCART, the advection, the key parameter, is estimated by a flux-form semi-Lagrangian method (Lin and Rood, 1996). The parameterization has been given for moist convection and boundary layer turbulent mixing (Chin et al., 2000). As the GEOS DAS (Goddard Earth Observing System Data Assimilation System) meteorological data includes both prognostic and diagnostic fields such as wind vectors, pressure, temperature, specific humidity, cloud mass flux, turbulent diffusion coefficient, boundary layer depth, and cloud fraction etc, the wave activities observed in all these fields would eventually reflect in the modelled aerosol parameters also. Earlier comparative studies have also pointed out the discrepancy between the GOCART model simulated values and observations, which in turn called for model improvement on aerosol size distributions, the refractive indices of dust and black carbon aerosols, and biomass burning emissions (Chin et al., 2007, 2009). The advection parameterization used in CHIMERE for the present study is the first order Van Leer scheme, which is found to provide good accuracy for transport of high concentrations in plumes. This model also includes the "urban parameterization", which is not included in most large-scale weather models, which is for correcting the wind speed in the surface layer (due to increased roughness) in cities. This correction has effects on urban versions of the model, and mostly on primary pollutants like BC (http://www.lmd.polytechnique.fr/chimere/). These provisions in the model might be the reason for the more realistic temporal fluctuations in the species concentrations.

However, the natural variabilities, the modulations produced due to the natural atmospheric dynamics, are not reproduced accurately in either of these models, which might arise due to the improper advection and boundary layer parameterization schemes in the chemical transport models or inadequate measurements over the Indian regions going into the reanalysis data. Since both

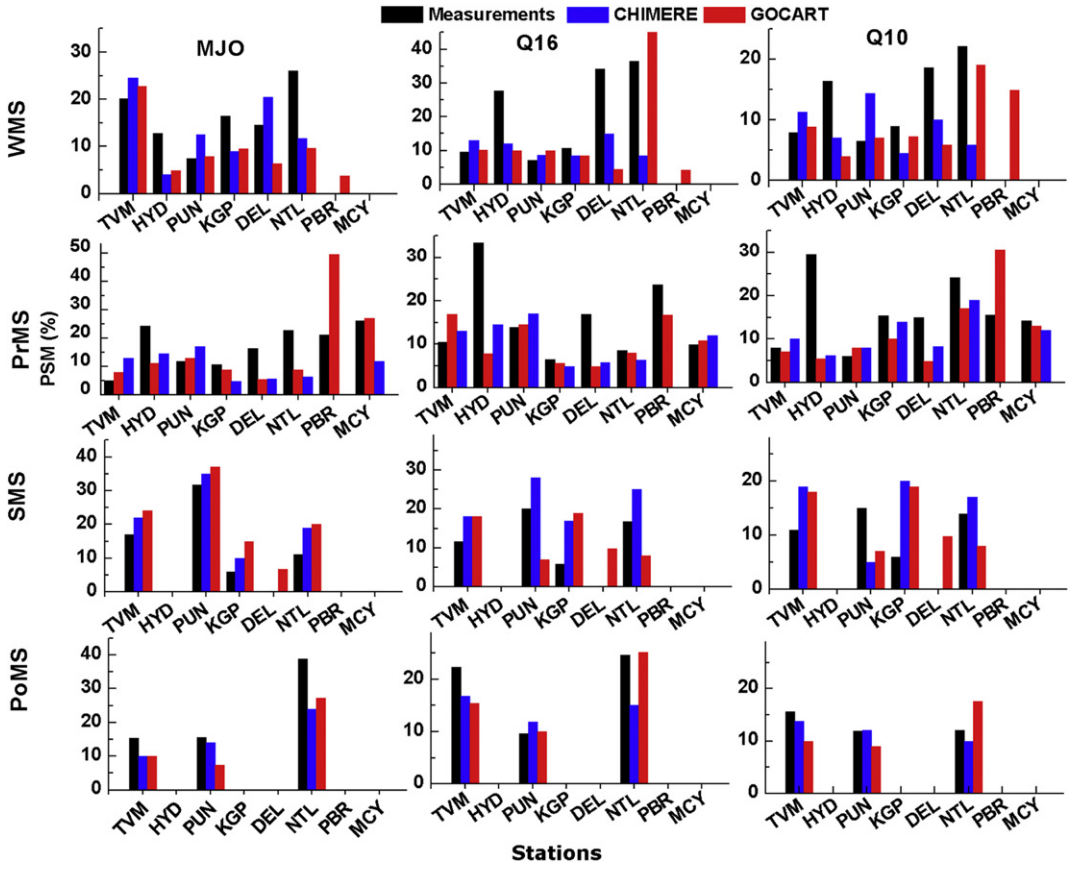

Fig. 11. The PSM for the periodicities in the time series of model simulations along with the corresponding measurements for all the seasons.

the model simulations were agreeing well with the measurements during the seasons with deeper boundary layer and the maximum mismatch has been observed during the winter, when the boundary layer processes are weak, the parameterization schemes employed for the boundary layer in the models might not be able to represent the seasonal changes in the ABL process well. Another import fact is that unlike the General Circulation Models, there are no wave parameterization schemes in the chemical transport Models. It has been reported in the literature that even in the general circulation models, the simulation of Madden Julian Oscillation (MJO) fails and this has been considered as a critical test for the GCMs' ability to simulate it at the tropics (Sperber, 2004). All these point to the need for improving the chemical transport models to simulate BC concentrations especially over Indian region and calls for more sustained observations and if possible an inverse parameterization.

In addition to the above discussed issues, the emission modelling in the models utilized only satellite-based information, on a monthly variability of observations, while the emission retrieval approach allows daily variability of the emissions. However, based on the comparative study between GOCART model simulations and observations from 173 AERONET stations across the globe, Chin et al. (2009) have reported that the obtained poor correlation between Aerosol Optical Depth and concentration of absorbing aerosols, especially over Asia, might be due to incorrect optical properties for dust and carbonaceous aerosols assumed in the model. In this context it might be kept in mind that based on ground based and satellite data Moorthy et al. (2007) have shown that the absorption efficiency of Asian dust is considerably higher than those of the African dust. Another main limitation of these models is the coarse spatial resolution, which fails to reproduce local details of aerosol dynamics. All these uncertainties limit the accuracy of aerosol modelling, especially over the Indian subcontinent as demonstrated in the present study.

5. Summary and conclusions

A space-time synthesis of the surface mass concentrations of BC has been carried out, for the first time, using measurements, for a period of 1 year, from eight network stations spread over Indian mainland and two islands in Bay of Bengal and Arabian Sea. The measurements were also compared with the globally accepted emission inventory models, CHIMERE and GOCART, and the time series data have been subjected to spectrum analysis to delineate the natural variabilities. The major findings are listed below.

- The surface mass concentrations of aerosol BC exhibits clear seasonal variations with the highest values in winter, followed by the continuous decrease towards the end of the summer monsoon and again increases towards post monsoon season.
- The seasonal variations are attributed to the changes in air mass types from continental to marine added with the local ABL dynamics. Spatially the Indo-Gangetic plain experienced much larger BC concentration than the peninsular stations. Higher BC concentrations at PBR than MCY is attributed to advection from East Asian countries.
- The comparative study between the measured BC mass concentration and the corresponding simulations using chemistry transport models revealed that the GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport) showed large deviation from the measurements whereas CHIMERE showed reasonably good agreement with the measurements. However, both the models were able to capture the seasonal/sub-seasonal fluctuations at most of the stations.
- It is hypothesized that an improvement in the ABL parameterization scheme for tropical environment might lead to better results with GOCART.
- Detailed analysis of the time series data revealed that the models could reproduce the natural variabilities in reasonably good accuracy especially over off-equatorial stations

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