Significant cooling effect on the surface due to soot particles over Brahmaputra River Valley region, India: An impact on regional climate

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Abstract

Black carbon (BC) is an important atmospheric aerosol constituent that affects the climate by absorbing (directly) the sunlight and modifying cloud characteristics (indirectly). Here, we present first time yearlong measurements of BC and carbon monoxide (CO) from an urban location of Guwahati located in the Brahmaputra River valley (BRV) in the northeast region of India from 1st July 2013 to 30th June 2014. Daily BC concentrations varied within the range of 2.86 to 11.56 $\mu\text{g m}^{-3}$ with an annual average of $7.17 \pm 1.89 \mu\text{g m}^{-3}$, while, CO varied from 0.19 to 1.20 ppm with a mean value of 0.51 ± 0.19 ppm during the study period. The concentrations of BC ($8.37 \mu\text{g m}^{-3}$) and CO (0.67 ppm) were ~39% and ~55% higher during the dry months (October to March) than the wet months (April to September) suggesting that seasonal changes in meteorology and emission sources play an important role in controlling these species. The seasonal $\Delta\text{BC}/\Delta\text{CO}$ ratios were highest (lowest) in the pre-monsoon (winter) $18.1 \pm 1.4 \mu\text{g m}^{-3} \text{ ppmv}^{-1}$ ($12.6 \pm 2.2 \mu\text{g m}^{-3} \text{ ppmv}^{-1}$) which indicate the combustion of biofuel/biomass as well as direct emissions from fossil fuel during the pre-monsoon season. The annual BC emission was estimated to be 2.72 Gg in and around Guwahati which is about 44% lower than the mega city 'Delhi' (4.86 Gg). During the study period, the annual mean radiative forcing (RF) at the top of the atmosphere (TOA) for clear skies of BC was $+9.5 \text{ Wm}^{-2}$, however, the RF value at the surface (SFC) was -21.1 Wm^{-2} which indicates the net warming and cooling effects, respectively. The highest RF at SFC was in the month of

April (-30 Wm^{-2}) which is coincident with the highest BC mass level. The BC atmospheric radiative forcing (ARF) was $+30.16$ (annual mean) Wm^{-2} varying from $+23.1$ to $+43.8 \text{ Wm}^{-2}$. The annual mean atmospheric heating rate (AHR) due to the BC aerosols was 0.86 K day^{-1} indicates the enhancement in radiation effect over the study region. The Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) captured the seasonal cycle of observed BC fairly well but underestimated the observed BC during the month of May–August. Model results show that BC at Guwahati is controlled mainly by anthropogenic emissions except during the pre-monsoon season when open biomass burning also makes a similar contribution.

Keywords: Black carbon, Carbon monoxide, Radiative impact, WRF-Chem

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1. Introduction

Black carbon (BC) and carbon monoxide (CO) are key atmospheric trace species that are primarily emitted into the atmosphere due to incomplete combustion of fossil fuels (diesel and coal) and biomass/biofuels (stubbles, dung and crop residues). BC plays a unique role in the Earth's climate system directly by absorbing the short-wave solar radiation, reducing the reflectivity (albedo) of snow and ice via deposition, and indirectly by modifying the cloud characteristics (Ramanathan and Carmichael, 2008; Koch and Del Genio, 2010; Das and Jayaraman, 2011; Kumar et al., 2011; Ramanathan et al. 2011; Sand et al., 2013). In view of this, BC has become a frontline concern all over the world for climate change strategies. The global mean positive radiative forcing recently estimated due to BC, independent of co-emitted species, is 1.1 Wm^{-2} (Bond et al., 2013), which is larger than that of methane and is around two third of the CO_2 radiative forcing. The mixing of BC with other aerosols forms atmospheric brown cloud over different parts of the world including Southeast Asia where BC is shown to exert a strong influence on regional climate, weaken the Indian summer monsoon circulation and intensify the tropical cyclones (Jacobson, 2001; Menon et al., 2002; Ramanathan et al., 2005; Chen et al., 2010; Lack et al., 2012). Through these mechanisms, it has been linked to various climate impacts, including increased temperatures and accelerated

ice and snow melting (IPCC, 2007). BC can be transported far away from emission sources due to sub-micron size range and longer atmospheric lifetime (few days to weeks). The transport of BC particle on regional scale leads to visibility impairment and even air pollution (formation of haze) in downwind regions (Gebhart et al., 2001; Tiwari et al., 2014) and detrimental impact on human health due to their fine particle size range which allows them to easily penetrate into the human respiratory system (Pope et al., 2002; Herich et al., 2011). Bergin et al., (2001) reported that BC also reduces plant productivity after depositing on leaves and Chameides et al., (1999) found that crops productivity reduced up to ~10 to 20% over India and China due to the high concentration of soot particles.

Carbon monoxide (CO) also affects the climate and human health with a positive indirect radiative forcing of 0.23 Wm^{-2} and dissipates a large fraction of OH radical in the troposphere (Seinfeld and Pandis, 2006; IPCC, 2013). It could be transported even further than BC due to its longer lifetime as compared to BC in the troposphere / boundary layer (Dickerson et al., 2002). A significant correlation between CO and BC is indicative of similarity in their sources (Pan et al., 2011 and references therein). Generally, their ratios ($\Delta\text{BC}/\Delta\text{CO}$) are used to estimate the BC emissions and to disassemble the source characteristics as well as for validating the emission inventories (Spackman et al., 2008; Subramanian et al., 2010; Girach et al., 2014; Pathak et al., 2016). Over the Indian region, the studies investigating near surface CO have been limited (Sahu et al., 2006; Lal et al., 2012; Sarangi et al., 2014; and references therein).

India, due to rapid urbanization and industrialization as well as tremendous population growth for the last three decades, is regarded as the second largest emitter of BC (soot particle) after China in the world (Ramanathan and Carmichael, 2008; Wang et al., 2011; Bond et al., 2013). At present, the global annual emissions of BC is $\sim 8 \text{ Tg yr}^{-1}$ and its source emission contributions are mainly from open biomass burning ($\sim 42\%$), fossil fuels combustion (38%) and biofuels burning ($\sim 20\%$) (Bond et al., 2004). In another study, Ramanathan and Carmichael, (2008) reported different source contribution and the highest BC emission contribution is from fossil fuel combustion ($\sim 40\%$) and rest of 60% are from both combined biomass and biofuels burning. In another regional study over South Asia, Kumar et al., (2015) reported the contribution of BC from anthropogenic (60%) and biomass burning (37%) sources over South Asia highlighting large regional variability in BC emission sources.

In India, the Government has taken a keen interest in regular monitoring of BC mass concentrations and under different network programs such as aerosol radiative forcing network (33 monitoring sites), system for air quality forecasting and research, and other several individual monitoring station through research organization in India. These measurements cover nearly the whole spatial extent of India and have been reported in several studies (Tripathi et al., 2005; Das et al., 2009; Chatterjee et al., 2012; Tiwari et al., 2013a; 2013b; 2014; Sarkar et al., 2014; Singh et al., 2015; Talukdar et al., 2015; reference therein). The vast majority of these studies were focused on measuring BC over the Indo-Gangetic Basin (IGB), urban, suburban and rural sites located in the northern and southern regions of India. However, only a few sites with short-term measurements (only two weeks at Guwahati) have been conducted over the Brahmaputra River Valley (BRV) that lies in between the southeast Tibetan Plateau and the Assam Hills in the eastern part of India (Pathak et al., 2010; Chakrabarty et al., 2012; Sand et al., 2013; Rahul et al., 2014). These studies provide important information about season-specific BC levels in the BRV but do not provide a comprehensive picture of temporal distributions of BC as well as CO in this region where annual mean daily temperature is reported to rise at 0.04 K/decade (Assam Science Technology and Environment Council: ASTEC, 2011). The reported warming trends in recent years have been accompanied by unpredictable and erratic precipitation characterized by both the drought-like periods and extreme spells of rain. Detailed information on the mass concentrations and distribution of BC and CO, which warm the atmosphere, over BRV region are therefore crucial for the improvement of regional air quality and to evaluate the ability of state-of-the-art chemistry transport models in simulating BC over this region.

In light of the above conditions, this study presents continuous measurements of near surface BC and CO (yearlong) concentrations from an urban site Guwahati in the BRV region. These measurements are used to examine (i) seasonal variability of BC and CO mass concentrations and their relation to meteorological parameters, (ii) BC emission estimation and identification of possible source location, and (iii) estimation of local scale radiative impact of BC at the top of atmosphere, surface and in the atmosphere and (iv) examine the ability of Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) in simulating the seasonal cycle of BC at Guwahati.

2. Measurements and instrument

2.1 Observational site

The sampling site for the monitoring of BC and CO is located in the Department of Environmental Science Division in Guwahati University, Guwahati (26°11'N, 91°44'E; 55 meters above sea level), the capital city of the Assam State, which is situated on the bank of the BR (Fig. 1). The city lies between the BRV and the Shillong Plateau covering an area of ~216 km² surrounded by the Nilachal, Chitrachal, and Narakasur hills, to the west, north and south, respectively. The total population of the city is about 968,549 in 2011 (<http://www.census2011.co.in/census/city/191-guwahati.html>). The surrounding of the city is also covered with thick green tea plants along with tropical rain forests in the hilly areas. The region experiences mild sub-tropical climate with warm and dry periods (pre-monsoon: March to late May), a heavy rainfall period (monsoon: June to September) and charming with dry winter (December to February) and transition period / post-monsoon (October to November). The annual mean rainfall is about 1800 mm and ~ 90% of this rainfall occurs between May and September, with maximum in July–August (Sharma et al., 2009). A major road leads toward Shillong, Meghalaya (50 meters away) near the sampling site. According to ASTEC, 2011 more than 400,000 vehicles get around the Guwahati's roads every day, and approximately 70% of these vehicles don't have emission clearance certificates, thus emitting large quantities of carbonaceous aerosols, oxides of nitrogen and sulfur (ASTEC, 2011). The instruments for the monitoring of BC, CO, and metrological parameters were placed on the top of the department building at a height of ~10 meters for the period from 1st July 2013 to 30th June 2014.

The BC mass concentration was continuously measured using a seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm) Aethalometer (Magee Scientific Co., Berkeley, California, USA, Model AE-31) during the study period. The atmospheric air was pumped through an inlet at the desired flow rate (4 L/min) which impinges on a quartz microfiber strip. Due to strong light absorption by BC at near-infrared wavelengths, the mass concentrations of BC was determined by converting light attenuation (measured at 880nm) using calibration factor suggested by Weingartner et al., (2003). The attenuation was obtained by calculating the difference between light transmission through the particle-free reference spot in the filter and the particle-laden sample spot. The methods of calibration, operation, and maintenance of AE-31 are widely described (Tiwari et al., 2013a; 2014; 2015; Safai et al., 2014; Singh et al.,

2015; Joshi et al., 2016). An absorption efficiency value of $16.6 \text{ m}^2/\text{g}$ was used to determine the BC mass concentrations. The BC concentration is calculated from the rate of change of attenuation (ATN), however, it was observed that as the filter tape advances, the BC concentrations obtained using an Aethalometer apparently rise (Hansen et al., 2007). Therefore, as the ATN increases, the measured BC concentrations are underestimated. This “Loading Effect” was taken into account when using empirical correction algorithms. In order to account for this effect, we have used the correction algorithm (Weingartner et al., 2003; Virkkula et al., 2007). The carbon monoxide (CO) was simultaneously measured with BC by CO analyzer of Ecotech Pty Ltd. Australia (Serinus 30: NDIR – GFC: auto-ranging from 0-200 ppm with high resolution (5 minutes) and 1000 cc/min flow rate. The zero measurements and span calibration of CO instrument were performed regularly using the standard calibration mixture of INOX, India (Yadav et al., 2014).

The meteorological parameters such as wind speed (WS), wind direction (WD), temperature (Temp), relative humidity (RH) (Envirotech Instruments Private Limited, New Delhi : as WS10 : Anemometer; WD10 : Potentiometric; Temp : RT10-Thermistor; RH : RT10-Capacitance) and precipitation (RF) were measured by automatic weather station which is parallel installed on similar height of BC and CO sampler over monitoring site. During the study period, four seasons: winter (December, January, and February), pre-monsoon (March, April and May), monsoon (June, July, August and September), and retreating monsoon/post-monsoon (October and November) were considered on the basis of local meteorological conditions (Gogoi et al., 2009). The seasonal diurnal mean values standard deviation of meteorological parameters such WS, WD, Temp, and RH were plotted (Fig. 2). In the present study, the annual mean Temp was $24.7 \pm 4.6 \text{ }^\circ\text{C}$, however, seasonally, it was lowest during the winter ($18.2 \pm 3.1 \text{ }^\circ\text{C}$) and highest in the monsoon ($29.1 \pm 2.1 \text{ }^\circ\text{C}$). The RH also showed a distinct seasonal variability with the annual mean of $82 \pm 6\%$ characterized by highest values in the monsoon (85%) and lowest in the pre-monsoon (77%). Surface the wind, which plays a crucial role in controlling accumulation and dispersion of pollutants, was found to be on average very low ($1.1 \pm 0.7 \text{ m/s}$) with the seasonal highest in the pre-monsoon ($1.9 \pm 1.0 \text{ m/s}$) and lowest in the winter (0.6 m/s). The seasonal mean directions were 190° (S), 206° (SW), 221° (SWW) and 204° (SW) during the monsoon, post-monsoon, winter and pre-monsoon seasons respectively. The mean rainfall was 1395 mm with the highest (436 mm) in July and the lowest was in February (13mm), however, no rainfall was observed in December and January (Fig. 3 and Table 1).

3. Results and discussion

3. 1. Variability in concentrations of BC and CO

Fig. 3 shows the day-to-day and monthly variability of BC and CO concentrations along with its running mean (24 points) during 1st July 2013 to 30th June 2014 over Guwahati. The average (daily mean) mass concentration of BC was $7.17 \pm 1.89 \mu\text{g m}^{-3}$ that varied between 2.86 (13th August 2013) and 11.56 (19th March 2014) $\mu\text{g m}^{-3}$. The mean concentration of BC gradually build-up from the October and remained high until April (dry period) after which it started decreasing. CO mixing ratios varied from 0.19 to 1.20 ppm with mean (0.51 ± 0.19 ppm) and was the highest in January (0.79 ppm) which is approximately 54% higher than the total mean value of CO. Lowest CO levels were observed in August (0.32 ppm). The mean CO concentration was much lower than the National ambient air quality standard (2 ppm) which are stipulated by the Central pollution control board of India (<http://www.cpcb.nic.in>), however, there is no standard made for BC till now. The changes in the concentrations of BC and CO daily and monthly are driven by variations in emission sources (anthropogenic and biomass burning) and variability in meteorological conditions which play a crucial role in accumulation and deposition. Fig. 3 also shows the rainfall data which indicates that more than 87% of rainfall occurred during the monsoon season during the study period which leads to strong wet removal of atmospheric aerosols in comparison to the dry period. The concentrations of BC ($8.37 \mu\text{g m}^{-3}$) and CO (0.67 ppm) were ~39% and ~55% higher during the dry months (October to March) than the wet months (April to September) which suggests that the rainfall is an effective scavenger of atmospheric aerosols as resulted in considerable amount of BC concentrations decreased over the study region (Tiwari et al., 2013a). Lee and Kim (2010) reported that a higher rainfall rate resulted in a higher aerosols deposition rate of aerosols within a precipitating cloud, whereas, in insignificant rainfall, a steady increase in the mean mass BC concentration, especially over the low-altitude locations coupled with uninterrupted generation of incompletely combusted carbon material as well as longer lifetime is expected during the dry period. They also suggested that during the boreal spring period (pre-monsoon) over the Southeast Asian region, the accumulation of BC increases when the local emissions of aerosols are prominently associated with anthropogenic sources (fossil fuel and biomass combustion). The shallower mixed layer depths and low ventilation coefficient (VC) also cause increase in the concentrations of atmospheric aerosols during the winter period over low-altitude regions as well as the stronger vertical mixing of aerosol during the pre-monsoon and monsoon seasons which play a dominant role in the monthly as

well as seasonal variability in dilution of concentration of atmospheric aerosols as well as CO. The VC, a product of two important meteorological parameters (mixed layer depth and wind speed), signalize the assimilative of ventilation and potential capacity of the atmosphere, was estimated. It was around seven times higher (mean: $1439 \text{ m}^2/\text{s}$) during the pre-monsoon (March, April, and May) as compared to other months, however, the higher concentrations of BC over study region (north-eastern India) during the pre-monsoon season may be appearance of enhanced biomass burning as an additional sources of BC as well as the impact of local topography of the observation site which is surrounded by the BRV and hills. Frequency distributions of hourly BC mass concentrations were separated into three segments (< 4 , $4-8$ and $> 8 \mu\text{g m}^{-3}$) in all four seasons during the study period over Guwahati (see sup. Fig. 1). It was found that the highest contribution (79%) was in monsoon below $4 \mu\text{g m}^{-3}$, however, during the post-monsoon, winter and pre-monsoon seasons, it was highest (17, 33 and 25%) in the segment greater than $8 \mu\text{g m}^{-3}$. During the pre-monsoon period, the concentrations were mostly greater than $7 \mu\text{g m}^{-3}$ (82%) indicating a serious atmospheric pollution induced by BC over Guwahati during this period. Also, in overall the study period, about 54% of the BC data exceeded the mean BC concentrations which indicate the presence of higher carbonaceous aerosols loadings over the BRV region. Higher loadings of BC might be contributing to the surface temperature increase over the BRV region. Earlier studies suggested the significant increase of temperature (at a rate of 0.03 K/year) from 1980 to 2005 and also drastically increase in the summer monsoon mean rainfall (at the rate of 4 mm/year) (Chakrawarty et al., 2012). In another study, Rahul et al. (2014) reported a vertical profile of BC loadings in the BRV region and found that the significantly higher (three-fold) concentrations of BC in between 2.5 to 6 km heights as compared to surface level during the monsoon period with higher heating rates of $\sim 2 \text{ K/day}$ due to BC which affects atmospheric stability leading the precipitation pattern over this region.

In Table 2, we have compared the observed annual mean BC mass concentrations of Guwahati with different geographic environments such as urban, rural, industrial and high altitude located in the Indo-Gangetic Basin (IGB), the BRV and Himalayan foothills of northern and north-eastern India located in between (20.26° N , 85.83° E) and (30.73° N , 76.78° E). From the table, it was observed that the north eastern part of India (BRV: Agartala, Dibrugarh, and Shillong) show concentrations of BC similar to the IGB. The high altitude locations show much lower BC concentrations than the BRV region. BC concentrations in the BRV are much higher than the southern and western parts of India (Bangalore: Aswathy et

al., 2010; Pune: Safai et al., 2014, Ooty: Udayasoorian et al., 2014; Trivandrum: Babu and Moorthy, 2002 and Udaipur: Vyas et al., 2010). Sharma et al., (2012) reported much higher BC concentrations in the neighboring country at Kathmandu, Nepal ($8.4 \pm 5.1 \mu\text{g m}^{-3}$ varied from 1.9 to $39.9 \mu\text{g m}^{-3}$). Other neighboring countries like Pakistan (Lahore: $21.7 \mu\text{g m}^{-3}$: Husain et al., 2007) and Bangladesh (Dhaka: $4\text{--}48 \mu\text{g m}^{-3}$: Begum et al., 2012) also exhibit higher soot particles concentrations. In addition, the East Asia (in China) also reported very high concentrations (BC) ranging from $9.3\text{--}14.2 \mu\text{g m}^{-3}$ in 14 cities (Zhang et al., 2008). The concentrations of BC was also much higher ($4\text{--}10 \mu\text{g m}^{-3}$) over a megacity, Istanbul, Turkey (Ozdemir et al., 2014). The concentrations of BC over the European and US countries are much lower than the BRV region [France ($1.5 \mu\text{g m}^{-3}$): Saha and Despiau, 2009; Finland ($0.25 \mu\text{g m}^{-3}$): Hyvärinen et al., 2011; Portugal ($1.3 \mu\text{g m}^{-3}$): Pereira et al., 2012; Italy (213 ng m^{-3}): Cristofanelli et al., 2013; Northeastern US ($10\text{--}900 \text{ ng m}^{-3}$): Ahmed et al., 2014; Texas, USA ($0.23\text{--}2.48 \mu\text{g m}^{-3}$): Raysoni et al., 2011; Finnish Arctic ($0\text{--}120 \text{ ng m}^{-3}$): Raatikainen et al., 2015; Spain ($0.03\text{ to }10.9 \mu\text{g m}^{-3}$): Milford et al., 2016]. This comparison clearly shows that BC levels in the BRV region are much higher compared to several parts of the world and emission mitigation strategies should focus on reducing man-made BC emissions in the BRV region.

3.2 Diurnal variations of BC, CO, and $\Delta\text{BC}/\Delta\text{CO}$

Diurnal variations of BC and CO and $\Delta\text{BC}/\Delta\text{CO}$ are very much important in understanding the role of mesoscale atmospheric processes as well as the effect of local and regional human activities. In the present study, the seasonal diurnal variability of BC and CO and $\Delta\text{BC}/\Delta\text{CO}$ at Guwahati were sorted into four major seasons (monsoon, post-monsoon, winter, and pre-monsoon) and its diurnal characteristics were plotted (Fig. 4A & B). Annually BC showed a gradual increase from 07:00 to 10:00 Local Standard Time (LST), lower concentrations from 12:00 to 17:00 LST, and second maxima during 20:00 to 23:00 LST (see sup. Fig. 2). During the study period, the significant variability in meteorological parameters was observed, however, there are no any pronounced distributions in BC diurnal variability during the different seasons. The post-monsoon season shows a unimodal distribution with the lowest values during day time (12:00 to 16:00 LST) and higher (20:00 to 09:00 LST) in the morning, evening as well as night time, however, in the winter and pre-monsoon, there was less variability between day and night time BC levels owing to the development of the boundary planetary boundary layer and pollution lifting with the valley breeze during the day (Nair et al., 2007) because of the topography of Guwahati and low wind speed. This pattern is totally

different from that observed in the other urban areas of the Indian subcontinent where BC concentrations reach to the maximum in the early morning and evening due to the traffic emissions as well as the shallow planetary boundary layer. The monsoon season is characterized by pronounced variability with a bimodal distribution (BC peaks between 06:00 to 10:00 LST and 20:00 to 23:00 LST) which clearly indicates the impact of traffic emission in the morning and evening hours. Seasonally, BC mass concentrations were highest in the pre-monsoon ($8.24 \pm 1.72 \mu\text{g m}^{-3}$) followed by winter ($8.19 \pm 1.39 \mu\text{g m}^{-3}$), post-monsoon ($7.29 \pm 21.60 \mu\text{g m}^{-3}$), and monsoon ($5.58 \pm 1.30 \mu\text{g m}^{-3}$) seasons which is different from the earlier studies (Safai et al., 2014; Tiwari et al., 2014; Singh et al., 2015; references therein) with an annual amplitude (ratio of max / min) of ~ 1.5 (see sup. Fig. 3). Similar but much higher concentrations in the pre-monsoon and lower concentrations in the monsoon period have been reported for the Hindu-Kush- Himalayan region (Gerter et al., 2016). Generally, BC concentrations at different Indian sites vary greatly between day and night (Tiwari et al., 2013a; Safai et al., 2014) but we don't see such a large variability in the present study (Fig. 4).

In the case of CO concentrations, the fluctuations in their diurnal variability in the pre-monsoon, post monsoon and monsoon seasons were weaker compared to the winter season. The seasonally mean mixing ratios of CO were 700 ± 170 , 510 ± 90 , 500 ± 160 and 380 ± 130 ppbv during the winter, post-monsoon, pre-monsoon and monsoon seasons, respectively. The relatively higher CO atmospheric loadings during the winter period over the study region might be due to large-scale fossil fuels and biomass burning (Pathak et al., 2016). The highest concentrations were in between 09:00 to 11:00 (733 ppbv) LST and 20:00 to 23:00 (827 ppbv) LST indicating the impact of the huge amount of fossil fuels burning during traffic movement. In the pre-monsoon, the stable and lower CO concentrations loading during day and night time was likely related to the mixing of air masses within a deeper boundary layer that leads to a higher VC as compared to winter (Goyal and Chalapati, 2007). In the post-monsoon, the CO concentration increased significantly (28%) in night-time from 19:00 to 05:00 LST as compared to day time (06:00 to 18:00 LST) which is likely due to the impact of trapping of anthropogenic emissions in a shallower boundary layer. In the monsoon season, both BC and CO were observed lower because it is lifted to middle and upper troposphere by deep convection associated with the Indian summer monsoon as demonstrated by satellite-based observations and washout/rainout effect (Girach et al., 2014; Rosenfeld et al., 2014). Overall, the night time concentrations of BC was observed slightly higher (4%) as compared

to day time, however, in the monthly analysis, it was lower by ~16% (August,13) and 8% (December,13) during the night time as compared to day-time respectively. In three months (September: 12%, October: 27% and November: 21%) the concentrations of BC were found higher, while, there were no clear variations are seen in the months of July, January, March, May and June during the study period between day and night time which is indicating less impact of boundary layer conditions in this months.

The ratio (BC/CO) depends on a number of factors including the carbon/oxygen ratio of fuel getting burned over, measurement-location, events, type of air masses sampled and type of fuel getting burned at the source location. Some part of CO may also produce by photo-oxidation of volatile organic compounds (VOCs). During aging and transport, the soot particles are removed by deposition process either wet or dry, whereas, for CO, it is removed primarily through oxidation by hydroxyl radicals. Kondo et al. (2006) suggested that the BC/CO emission ratio increases with ambient temperature. During the study period, the BC/CO ratio was $14.1 \pm 4.9 \mu\text{g m}^{-3} \text{ppmv}^{-1}$ varying from 4.6 to $35.5 \mu\text{g m}^{-3} \text{ppmv}^{-1}$. Similar ratio values have been observed over the Indian Ocean, the Arabian Sea, and an urban site Hyderabad (Dickerson et al., 2002; Badarinath et al., 2007; Girach et al., 2014). The ratio values in the range of 0.29 to $29.83 \text{ ngm}^{-3} \text{ppbv}^{-1}$ were reported over USA, Europe, South-East Asia and South Asia (Matsui et al., 2011; Cristofanelli et al., 2013). In the present study, the ratio ($\Delta\text{BC}/\Delta\text{CO}$) was also calculated by $(\text{BC}-\text{BC}_0) / (\text{CO}-\text{CO}_0)$ because it indicates the emission sources (Han et al., 2009; Pan et al., 2011). The CO_0 is baseline concentrations of CO and was determined as values of the 1.25 percentile of data for monsoon (38 ppbv), post-monsoon (59 ppbv), winter (33 ppbv) and pre-monsoon (95 ppbv), whereas, the background value of BC concentration was assumed zero as suggested by Pan et al., (2011). Diurnally seasonal $\Delta\text{BC}/\Delta\text{CO}$ ratios were plotted (Fig. 4C) which shows a bimodal distribution with higher values in morning and evening and lower in day-time and night-time except during the winter period likely due to the different boundary layer (BL) condition. In winter, the BL depth was lower as compared to other seasons which leads to trapping of pollutants in a smaller volume and higher surface concentrations. In morning and evening due to the impact of fossil fuel combustion, however, in the evening, it was due to biomass burning (Tiwari et al., 2015). The seasonal ratios are highest in the pre-monsoon ($18.1 \pm 1.4 \mu\text{g m}^{-3} \text{ppmv}^{-1}$) followed by the monsoon ($15.0 \pm 2.1 \mu\text{g m}^{-3} \text{ppmv}^{-1}$), post-monsoon ($14.2 \pm 1.9 \mu\text{g m}^{-3} \text{ppmv}^{-1}$) and winter ($12.6 \pm 2.2 \mu\text{g m}^{-3} \text{ppmv}^{-1}$). Higher $\Delta\text{BC}/\Delta\text{CO}$ ratio with non-significant relationship ($r = 0.08$) between them during the pre-monsoon season indicates the lower

production of CO due to photochemical oxidation of CH₄ during higher solar radiation as well higher O₃ level and other volatile organic compounds than direct emissions from anthropogenic emissions (Pan et al., 2011), however, higher ratio ($14.2 \mu\text{g m}^{-3} \text{ppmv}^{-1}$) with significant positive correlation (0.44) during the post-monsoon season was due to combined effect of biomass as well as fossil fuel combustion. Pathak et al. (2016) studied the ratio ($\Delta\text{BC}/\Delta\text{CO}$) at Dibrugarh which is located approximately 450 km in the northeast direction on similar height during January 2012 – December, 2013 and found the highest ratio in winter ($10.6 \pm 0.6 \text{ ngm}^{-3} \text{ppbv}^{-1}$) followed by the pre-monsoon ($8.9 \pm 0.7 \text{ ngm}^{-3} \text{ppbv}^{-1}$), monsoon ($4.7 \pm 0.5 \text{ ngm}^{-3} \text{ppbv}^{-1}$), post-monsoon ($7.2 \pm 0.8 \text{ ngm}^{-3} \text{ppbv}^{-1}$). In the pre-monsoon, the $\Delta\text{BC}/\Delta\text{CO}$ ratio at Guwahati was 60% higher than Dibrugarh indicating a difference in sources of both pollutants over the study region. In another study over the Bay of Bengal, Girach et al. (2014) found higher ratio during the winter ($20.3 \text{ ngm}^{-3} \text{ppbv}^{-1}$) and pre-monsoon ($16 \text{ ngm}^{-3} \text{ppbv}^{-1}$) seasons and suggested the role of biomass burning and fossil fuel combustion in both seasons over the region. Pan et al. (2011) reported the higher ratios (10.3 ± 0.3 and $11.6 \pm 0.5 \text{ ngm}^{-3} \text{ppbv}^{-1}$) during two biomass burning events in eastern China which confirm the impact of biomass burning in case of higher ratio. In Beijing, the ratio $\Delta\text{BC}/\Delta\text{CO}$ was $\sim 8 \text{ ngm}^{-3} \text{ppbv}^{-1}$ in early morning which was attributed due to increased emission from heavy diesel engines vehicles (Han et al., 2009). Ratios ($\Delta\text{BC}/\Delta\text{CO}$) analysis during the study period indicated the impact of biofuel/biomass burning during the pre-monsoon season.

3.3. Estimation of BC emission

The concentration of atmospheric primary aerosols is a function of atmospheric dispersion, removal processes (vertical and horizontal) and emission rates (Husain et al., 2007). The decrease in BC concentration during mid-day emerges to be strongly impacted by the increased horizontal and vertical dispersion as the vehicular emissions over the cities are assumed to remain the same throughout the day (Bond et al., 2004; Cohen and Wang, 2014; Lee et al., 2016). A mathematical model, which converts mass concentration of BC into its emissions has been used previously for estimation of BC emissions over the different regions including India (Dutkiewicz et al., 2009; Bano et al., 2011; Dumka et al. 2013) considering local/regional vertical and horizontal dispersion rates and the mean concentrations times the affected volume which can be taken as the area of the city times the boundary layer heights (Eq. 1).

$$\text{mass of BC emitted} = [BC] \cdot (\text{area of city}) \cdot MH \cdot \left[\frac{K \cdot dt}{MH} + \frac{V \cdot dt}{D_e} \right] \quad (\text{Eq. 1})$$

Where K is the rate of vertical mixing, MH is mixing height, V is the surface WS, and D_e is the effective diameter of the city. The last term is a dimensionless factor to account for vertical and horizontal dispersion which represents the number of times the volume, filled during the time period 'it' (Dutkiewicz et al., 2009). Hourly intervals MH values were extracted from the HYSPLIT model (<http://www.arl.noaa.gov/ready/hysplit4.html>). During the study period, the seasonal MH during the day (mean: 669 m) and night (mean: 271m) were 476, 1003, 547 and 451 m and 209, 394, 293 and 197 m for the winter, pre-monsoon, monsoon and post-monsoon seasons, however, the seasonal mean was 442, 698, 420 and 319 m respectively. The K value is defined under calm conditions by assuming that the volume (mean) over the city would be filled once per every 24 h [$1 = (K \cdot 24 / MH_{\text{mean}})$]. While this limitation makes the absolute value of total emissions from the model uncertain, the temporal profile should be reliable (Dutkiewicz et al., 2009). The seasonal K values were calculated varying from 13.3 (post-monsoon) to 39.0 (pre-monsoon) m/hr. The area of the city (Guwahati) was taken $\sim 255 \text{ km}^2$ with an equivalent diameter of 18 km. The wind speed was higher during the pre-monsoon (7.54 km/hr) however, the lower was in the post-monsoon (2.32 km/hr). These wind speeds are strong enough to fill the city volume in 3-9 hours and thus, BC emissions estimated with this approach will be significantly overestimated and should not be considered as representative of emissions from Guwahati city only. Further, the value of BC at a single monitoring cannot fully represent the mean value of the city, however, the averaged diurnal profiles of mass are expected to be fairly consistent over the city. In light of the above conditions, we expect this simple model to provide only a reasonable picture of temporal trends in the emissions over the study area. The monitoring site is expected to be influenced heavily by traffic emissions due to the presence of a major connecting city road within 250 meter of the observation site. Several industrial units are also located (within 8 to 12 km) in the NNE direction from the sampling site.

During the study period, the seasonal mean BC emissions in Guwahati were calculated (Fig. 5) and were in the order of 806 (pre-monsoon), 214 (winter), 138 (monsoon) and 85 (post monsoon) Kg/hr. The annual mean was 311 Kg/hr (annual total emission: 2.72 Gg). The annual emissions of BC at present study in and around Guwahati are $\sim 44\%$ lower than

megacity Delhi (4.86 Gg) (Bano et al., 2011), however, it is much lower than the other megacities as Hyderabad and Karachi (Dutkiewicz et al., 2009; Dumka et al., 2013). Large differences in between day and night BC emissions were observed with day time emissions being 4.3 (346 kg/hr), 5.1(1347 kg/hr), 3.0 (208 kg/hr) and 2.5(121 kg/hr) times higher than the night times (81, 265, 68 and 50 kg/hr) during the winter, pre-monsoon, monsoon and post-monsoon respectively. During the pre-monsoon, the highest day time BC emissions indicating the transportation of BC from other location to the receptor site. In this period, the farmers burns crops residue for the next farming cycle over IGP region, which produces enormous amount of fine particles as well as soot particles (Reddy and Venkataraman, 2002; Gustafsson et al., 2009; Kumar et al., 2011; Tiwari et al., 2013a; Kaskaoutis et al., 2014; Bisht et al., 2015).

3.4. Source identification of BC and CO

To understand the potential source regions of atmospheric pollutants, isentropic back trajectories analysis ending at 500 m over Guwahati were obtained by using the Hybrid Single-Particle Lagrangian Integrated Trajectory model on a 24-h daily basis during the study period (HYSPLIT: <http://www.arl.noaa.gov/ready/hysplit.html>) (Draxler, R. et al., 2014). A technique 'called concentration weighted trajectory (CWT) analysis' was developed by Seibert et al. (1994), which defines the concentrations at the receptor site to the respective backward trajectories was also applied in the present study to know the source region of BC and CO. Concentrated weighted backward trajectory study was conducted for BC (A) and CO (B) over Guwahati and are depicted in Fig. 6 A and B respectively. During the study period, it was observed that the three major sectors were influencing the Guwahati region as IGP region in the west direction (WD), south and southwest direction from inland sides of West Bengal (WB), Bangladesh (BA) and from the largest bay "Bay of Bengal: BoB", and south-east (SE) region for both BC and CO. The concentrations of BC were lowest when the air masses arrived (23%) to Guwahati from SE directions ($< 4 \mu\text{gm}^{-3}$), however, the highest concentrations were found when the winds were blowing from WD (35%) which is directly associated with polluted air masses from Ganges Valley (IGP) region ($> 9 \mu\text{gm}^{-3}$) (Das et al., 2015; Pathak et al., 2016) and the second largest sector was from BB, WB, BA ($< 7 \mu\text{gm}^{-3}$) direction (44%). In the case of CO, it was different than BC and was 80% higher concentrations ($> 0.9 \text{ ppm}$) of CO when the wind arrived from the IGP region to study region than the BB, WB, BA and SE ($< 0.5 \text{ ppm}$).

In addition to this, the conditional probability function (CPF) technique was applied to identify the directions contributing to the upper 25 percentile of BC and CO data (Kim et al., 2003; Kim and Hopke, 2004). The probable source directions are estimated on the basis of the probability of a given source contribution from a particular WD in exceedance of a predetermined threshold value criteria. The daily contribution is assigned to each hour of a given day to match to the same hourly wind data (wind speed and wind direction). The CPF is defined as follow:

$$CPF_{\Delta\theta} = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \quad \text{Eq. -1}$$

Where $m_{\Delta\theta}$ is the number of occurrence from wind sector $\Delta\theta$ that exceeded the threshold criterion, and $n_{\Delta\theta}$ is the total number of data from the same wind direction sector. In the present study, 24 wind sectors were selected ($\Delta\theta = 15^\circ$). The periods for which the winds were less than 0.5 m/s (mostly calm wind condition) were excluded from this analysis due to the isotropic behavior of wind vane under calm wind conditions. Generally calm wind is considered when WS is less than 1 m/s but in the present study, it was taken 0.5 m/s to maintain reasonable sample size because lower wind speed accounted for $\sim 40\%$ data observed here. A threshold criterion of the upper 25 percentile was chosen to define the directionality of the sources of the pollutants. The sources of the pollutants are likely to be located in the directions that have high conditional probability values. Fig. (7A and B) shows the CPF analysis for both BC and CO that are generally from incomplete combustion of fossil fuel, clearly indicate that the source direction for both and are different source region. BC was dominating when the wind blew from the north-northeast (NNE), whereas, CO was from west-northwest direction. In NNE direction, there are major industrial locations "Export Promotion Industrial Park (EPIP), Amingaon" are located such as Industrial Growth Centres (IGC) at Chariduar, and Matia, Seven Integrated Infrastructure Development (IID) Centres at, Dalgaon, Malinibeel, Bhomoraguri/Naltoli, Demow, Titabor, Dahudi, Silapathar etc under Assam Industrial Development Corporation Ltd and that could be one of the reasons that this sector leads to higher concentrations of BC. Higher CO concentrations may be due to combustion of fossil fuel from transport because a national highway passes from that side.

HYSPLIT Trajectory and CWT analysis identify that the major source regions for BC are from IGP as well as BoB region which are affecting to the BRV region by transportation, however, the CPF analysis indicated that the impact of local source of BC also which were produced by anthropogenic activities around the sampling site. In the case of CO, all analysis

indicated that the major sources region is from the IGP region. The overall results demonstrate that Ganges and Brahmaputra rivers valley renders as a hot-spot areas for atmospheric pollution over South Asia.

3.5. Radiative impact due to BC aerosol on regional climate

The BC induced radiative forcing (RF) and atmospheric heating rates (ARH) have been calculated over Guwahati by using the OPAC model (Hess et al., 1998) in conjunction with the SBDART model (Richiazzi et al., 1998) as explained in Panicker et al., 2013; Bisht et al., 2015. The Optical Properties of Aerosols and Clouds (OPAC) model is used to obtain BC aerosol optical properties such as aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry parameter (g). For this purpose, BC mass is converted into number density following Hess et al. (1998) and subsequently used in the continental average model. The OPAC model uses the surface number concentrations to obtain vertical profile using equation $N=N(o) e^{(-h/z)}$; where $N(o)$ is the surface number concentration and Z are scale height, which we define according to the season. This aerosol vertical profile constrained by observed surface mass concentrations is used to obtain AOD, SSA, and g. Note that BC mass alone has been used in this experiment to get BC optical properties. OPAC model provides AOD, SSA, and ASP at 60 wavelengths. The estimated values of BC SSA at 500 nm is 0.2257, and that of g is 0.353. The OPAC derived AOD, SSA and g were further used in SBDART to obtain short wave (0.3-3 μm) radiative fluxes for aerosol and no aerosol conditions at the surface (SFC) and at Top of the Atmosphere (TOA). The difference in fluxes in aerosol and without aerosol condition was calculated to estimate radiative forcing both at SFC and TOA. The atmospheric heating rate was calculated using the equation (Eq. 2; Bisht et al., 2015).

$$\frac{dT}{dt} = \frac{-1}{rC_p} \frac{dF}{dz} \quad \text{Eq. -2}$$

Where C_p is the specific heat capacity, ρ is the density of air and dF/dZ is the radiative flux divergence. Heating rates were computed as a difference between fluxes with and without BC conditions. For calculation of radiative forcing and heating rates, we used the tropical model profile of SBDART model and also used column ozone from OMI and water vapor from MODIS as additional input parameters along with aerosol optical properties. A large variability was found in the RF at TOA, SFC and atmospheric radiative forcing (ARF), ATM

during this study period (Fig. 8), due to the presence of absorbing types of particles in the atmosphere due burning of fossil fuel and biofuels/biomass. The monthly mean RFs at the TOA for clear skies of BC forcing values were varying between + 7.2 (August) to + 13.5 (April) Wm^{-2} with annual mean (+ 9.5 Wm^{-2}), however, the RFs values at the SFC were -15.9 (August) to - 30.3 (April) Wm^{-2} with mean (-21.1 Wm^{-2}) respectively. The highest forcing at the surface was in the month of April (-30 Wm^{-2}) which is attributed to the high BC mass level during this period. Seasonally, it was observed highest during the pre-monsoon (+12.0 and -25.6 Wm^{-2}) and lowest (+7.9 and -17.0 Wm^{-2}) in the monsoon periods for both at TOA and SFC, respectively, which clearly indicates the reduction of shortwave radiation at the surface exclusively due to BC absorption. The BC atmospheric radiative forcing, which is the difference between the TOA and SFC forcing, was + 30.16 (annual mean) Wm^{-2} varying from + 23.1 to + 43.8 Wm^{-2} . The mean negative RF (- 21.1 W m^{-2}) at SFC indicates the cooling effect, however, in the positive value of RF (+ 9.5 Wm^{-2}) at the TOA, indicates the net warming effects. The forcing values were found to be proportional to the BC mass and were less affected by other meteorological factors. The calculated values of monthly AHR due to BC aerosols were varying from 0.65 to 1.23 K day^{-1} with an annual mean (0.86 K day^{-1}) which indicate the enhancement in radiation effect over the study region. During the pre-monsoon period, the AHR was around 23% higher than the mean AHR value. This enhanced atmospheric heating and surface cooling induced by BC might reduce convection and cloud formations and influence cloud cover (Satheesh et al., 2000; Park et al., 2010; Koch and Del Genio, 2010).

3.6. Evaluation of WRF-Chem

In this section, we examine the ability of WRF-Chem to simulate BC aerosols observed at Guwahati. The objective of evaluating WRF-Chem is to establish the credibility of WRF-Chem so that it can be used to address a variety of open scientific questions in this region. The WRF-Chem simulations used in this study are described in detail in Kumar et al. (2015) and thus only a brief description of the important aerosol processes is provided here. The aerosol processes are represented using the Goddard Chemistry Aerosol Radiation and Transport (GOCART) bulk scheme (Chin et al., 2000; 2002), which simulates five major tropospheric aerosol types including black carbon, organic carbon, sulfate, dust, and sea salt, assuming externally mixed aerosols. BC aerosols are assumed to be present in hydrophobic and hydrophilic modes with a hydrophobic to a hydrophilic conversion e-folding lifetime of 2.5 days. All primary BC emissions are assumed to be hydrophobic. Both the hydrophobic

and hydrophilic BC components are subjected to dry deposition while only the hydrophilic BC component is removed by wet deposition. Anthropogenic emissions of BC and other aerosol are based on the Emission Database for Global Atmospheric Research developed for the assessment of Hemispheric Transport of Air Pollutants version 2 (EDGAR-HTAP2) that provides monthly varying emissions of trace species at a spatial resolution of $0.1^\circ \times 0.1^\circ$ for the year 2010 (Janssens-Maenhout et al., 2015). Daily varying biomass burning at the spatial resolution of 1 km^2 are based on the Fire Inventory from NCAR version 1 (FINN v1; Wiedinmyer et al., 2011). We also included three BC tracers on top of standard BC particles in WRF-Chem to keep track of BC emitted from different source types (anthropogenic emissions, biomass burning emissions and long-range transport from domain boundaries) and regions (North India, West India, South India and East India). The reader is referred to Kumar et al. (2015) for further details of BC tracers and other model configuration details such as physical parameterizations, and initial and boundary conditions for meteorology and chemistry.

BC simulated by this configuration of WRF-Chem has been compared with observations from 21 Indian sites and it has been shown that WRF-Chem captures the seasonal variability of BC mass concentration fairly well over most parts of India and performs better compared to other regional and global models in this region (Kumar et al., 2015). Here, we extend our previous evaluation by comparing daily mean observed and simulated BC mass concentrations over Guwahati (Fig. 9a). Like observations, WRF-Chem also shows highest BC mass concentrations during the winter and lowest values during the monsoon season. The wintertime WRF-Chem values agree better with the observations while the model underestimates the observed values during the pre-monsoon and monsoon seasons. We also examined the relative contribution of anthropogenic, biomass burning and long-range transport (inflow from the domain boundaries) to model simulated BC mass concentrations at Guwahati (Fig. 9b). Note that relative contribution of different sources are shown only from 20th January onwards because first 20 days of the model run were considered as a spin-up for the tracers (Kumar et al., 2015). The relative contributions clearly show that anthropogenic emissions are the main source of BC mass concentrations observed at Guwahati except during the pre-monsoon season when the biomass burning emissions occasionally makes a similar or even larger contribution. Long-range transport does not contribute significantly to BC mass concentrations at Guwahati. The underestimation of BC mass concentration by WRF-Chem from May-August could be associated mostly with uncertainties in

anthropogenic emissions because anthropogenic emissions dominate the BC mass loadings at Guwahati during this period (Fig. 9b).

4. Conclusions

Black carbon and carbon monoxide were measured for the first time at Guwahati, an urban location of the Brahmaputra River Valley, the northeast region of India from 1st July 2013 to 30th June 2014. Daily BC concentrations varied within the range of 2.86 to 11.56 $\mu\text{g m}^{-3}$ with an annual average of $7.17 \pm 1.89 \mu\text{g m}^{-3}$, while, CO varied from 0.19 to 1.20 ppm with a mean of 0.51 ± 0.19 ppm during the study period. The concentrations of BC ($8.37 \mu\text{g m}^{-3}$) and CO (0.67 ppm) were ~39% and ~55% higher during the dry months (October to March) than the wet months (April to September) suggesting that the seasonal changes in meteorology and emission sources play a crucial role in controlling the variability of these species at Guwahati. The seasonal $\Delta\text{BC}/\Delta\text{CO}$ ratios were highest (lowest) in the pre-monsoon (winter) $18.1 \pm 1.4 \mu\text{g m}^{-3} \text{ ppmv}^{-1}$ ($12.6 \pm 2.2 \mu\text{g m}^{-3} \text{ ppmv}^{-1}$). Higher $\Delta\text{BC}/\Delta\text{CO}$ ratio during the pre-monsoon season indicated that the combustion of biofuel /biomass as well as direct emissions from fossil fuel. The seasonal mean BC emissions were in the order of 806 (pre-monsoon), 214 (winter), 138 (monsoon) and 85 (post monsoon) Kg/hr, however, the annual mean was 311 Kg/hr (annual emission: 2.72 Gg). However, these emission estimates might be significantly overestimated due to the presence of stronger winds at Guwahati. The annual emissions of BC estimated in and around Guwahati are ~ 44% lower than those estimated using the same methodology for the megacity Delhi (4.86 Gg). The monthly mean radiative forcing (RF) at TOA for clear skies of BC forcing values were varying between +7.2 to +13.5 Wm^{-2} with annual mean (+ 9.5 Wm^{-2}), however, the RFs values for SFC were -15.9 to -30.3 Wm^{-2} with mean (-21.1 Wm^{-2}) which indicates the net warming (TOA) and cooling (SFC) effects. The ARFs due to BC, which is the difference between the TOA and SFC forcing, was + 30.16 Wm^{-2} . The annual value of AHR due to the BC aerosols was 0.86 K day⁻¹ which indicates the enhancement in radiation effect over BRV region. The WRF-Chem model captures the seasonal variability of observed BC at Guwahati fairly well but underestimated the observed values during the May-August. The source contribution analysis showed that anthropogenic emissions are the main source of BC at Guwahati except during the pre-monsoon season which is when open biomass burning makes a similar or even larger contribution. The underestimation of observed BC by WRF-Chem during the May-August is attributed mostly to uncertainties in anthropogenic emissions indicating the need for improving the seasonal cycle of anthropogenic emissions over this region.

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Legend

Figures:

Fig. 1: Map of India showing the sampling site at Guwahati University.

Fig. 2: Seasonal diurnal variations of meteorological parameters: (A) wind speed: WS, (B) wind direction: WD, (C), temperature: Temp, and (D) relative humidity: RH over Guwahati during the period from July 2013 to June 2014.

Fig.3: Day- to-day variability of BC (red circle) and CO (green circle) along with its running mean during the study period 1st July 2013 to 30th June 2014. The horizontal dot lines represent the running mean value of BC and CO. The vertical box (blue color) shows the monthly mean of BC with standard deviation. The bar graph shows the monthly rainfall over Guwahati.

Fig.4: Intra-seasonal diurnal variability of mean hourly data of BC and CO concentrations during the study period over Guwahati

Fig. 5: Diurnal plot of BC emissions during different seasons over Guwahati

Fig. 6: Concentrated weighted backward trajectory map for BC (A) and CO (B) at Guwahati

Fig.7: A: CPF analysis for BC and B: CPF analysis for CO over Guwahati

Fig.8: Estimation of radiative forcing due to black carbon from 1st July to 30th June 2014 over Guwahati

Fig. 9: [a] Daily variability of observed and WRF-Chem simulated BC mass concentration during the study period over Guwahati. [b] Percentage contributions of anthropogenic emissions, biomass burning emissions and long-range transport, i.e. inflow from the domain boundaries to WRF-Chem simulated BC mass concentrations.

Table:

Table 1: Monthly mean mass BC and CO concentrations along with monthly mean meteorological parameters (wind speed: WS, wind direction: WD, temperature: Temp, relative humidity: RH, solar radiation: SR and mixing height: MH) over Guwahati during the study period

Table 2: Average BC concentrations at various locations in IGB and BRV region along with the present study

Table 1: Monthly mean mass BC and CO concentrations along with monthly mean meteorological parameters (wind speed: WS, wind direction: WD, temperature: Temp, relative humidity: RH, solar radiation: SR and mixing height: MH) over Guwahati during the study period

Months,	Year	BC ($\mu\text{g m}^{-3}$)	CO (ppm)	WS (m/s)	WD ($^{\circ}$)	Temp ($^{\circ}\text{C}$)	RH (%)	SR (Wm^{-2})	MH (m)
July,	2013	5.6 ± 2.4	0.33	0.9	190.6	29.4	85.3	181.2	476.7
August,	2013	4.3 ± 1.9	0.32	0.8	189.1	29.5	84.4	174.7	386.7
September,	2013	5.9 ± 2.6	0.41	0.8	185.4	29.2	84.1	170.9	396.1
October,	2013	6.2 ± 2.8	0.50	0.7	193.2	26.7	85.3	150.4	335.5
November,	2013	8.4 ± 2.6	0.52	0.6	221.0	22.7	84.0	148.4	302.1
December,	2013	8.3 ± 3.7	0.65	0.5	237.9	19.5	84.7	103.5	318.7
January,	2014	8.1 ± 3.7	0.79	0.6	211.4	17.1	84.6	97.7	363.8
February,	2014	8.2 ± 3.2	0.64	0.8	213.4	18.1	80.9	119.0	411.9
March,	2014	8.3 ± 3.4	0.58	1.3	210.4	22.0	76.6	150.0	671.9
April,	2014	8.9 ± 2.9	0.54	2.2	212.4	25.5	72.6	191.3	878.5
May,	2014	7.5 ± 2.4	0.40	2.2	191.9	27.5	80.9	170.4	657.6
June ,	2014	6.5 ± 2.4	0.45	1.9	203.0	28.4	85.5	149.9	559.8

Table 2: Average BC concentrations at various locations in IGB and BRV region along with the present study

Locations	Lat	Long	Sampling Period	BC ($\mu\text{g m}^{-3}$)	Reference
Guwahati	26.11	91.44	2013-14	7.17	Present study
Guwahati	26.11	91.44	2011 (Winter)	9-41	Chakrabarty et al., 2012
Guwahati	26.11	91.44	2014(Monsoon)	6.5	Rahul et al., 2014
Dibrugarh	27.48	95.0	2012-23	4.0	Pathak et al., 2016
Agartala	23.76	91.26	2010-12	9.35	Guha et al., 2015
Shillong	25.57	91.88	2010	5.0	Kundu and Borgohain, 2010
Darjeeling	27.1	88.15	2010-11	3.45	Sarkar et al., 2015
Kharagpur	22.52	87.52	2004	2.7-9.3	Beegum et al., 2009
Kolkata	22.34	88.22	2012-13	5.27	Taludkar et al., 2015
Dhanbad	23.47	86.30	2012	6.3	Singh et al., 2015
Bhubaneswar	20.26	85.83	2007-08	5.2	Das et al., 2009
Varanasi	25.28	82.95	2009	8.92	Singh et al., 2015
Gorakhpur	26.75	83.38	2013-14	20.0	Singh and Rastogi, 2014
Surya, Village	26	81	2009-10	11.7	Praveen et al., 2012
Allahabad	25.45	81.85	2008	6.9	Ramachandran & Kedia, 2010
Kanpur	26.24	80.24	2004 (winter)	7.03	Nair et al., 2007
Agra	27.1	78.02	2004	20.6 (7.1–48.3)	Safai et al., 2007
Pant Nagar	29.0	79.5	2009-12	5.5	Joshi et al., 2016
Nainital	29.4	79.5	2007	1.34	Dumka et al., 2010
Mukteshwar	29.44	79.62	2005-07	0.81	Hyvarinen et al., 2009
Dehradun	30.32	78.03	2013	3.3	Kant et al., 2015
New Delhi	28.35	77.12	2013	6.7	Tiwari et al., 2013
Chandigarh	30.73	76.78	2001-02	3.7	Chowdhury et al., 2007
Patiala	30.34	76.38	2008 (winter)	5-15	Kharol et al., 2012

Fig. 1: Map of India showing the sampling site at Gauhati University.

Fig. 2: Seasonal diurnal variations of meteorological parameters: (A) wind speed: WS, (B) wind direction: WD, (C), temperature: Temp, and (D) relative humidity: RH over Guwahati during the period from July 2013 to June 2014.

Fig.3: Day- to-day variability of BC (red circle) and CO (green circle) along with its running mean during the study period 1st July 2013 to 30th June 2014. The horizontal dot lines represent the running mean value of BC and CO. The vertical box (blue colour) shows the monthly mean of BC with standard deviation. The bar graph shows the monthly rainfall over Guwahati.

Fig.4: Intra-seasonal diurnal variability of mean hourly data of BC and CO concentrations during the study period over Guwahati

Fig. 5: Diurnal plot of BC emissions during different seasons over Guwahati

Fig. 6: Concentrated weighted backward trajectory map for BC (A) and CO (B) at Guwahati

Fig.7: A: CPF analysis for BC and B: CPF analysis for CO over Guwahati

Fig.8: Estimation of radiative forcing due to black carbon from 1st July to 30th June 2014 over Guwahati

Fig. 9: [a] Daily variability of observed and WRF-Chem simulated BC mass concentration during the study period over Guwahati. [b] Percentage contributions of anthropogenic emissions, biomass burning emissions and long-range transport, i.e. inflow from the domain boundaries to WRF-Chem simulated BC mass concentrations.