



# 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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## HIGHLIGHTS

- The first time yearlong BC (mean:  $7.2 \mu\text{g m}^{-3}$ ) measurement over BRV
- Higher atmosphere forcing ( $+30.16 \text{ Wm}^{-2}$ ) & heating ( $0.86 \text{ K day}^{-1}$ ) over BRV
- Annual BC emissions are 2.72 Gg.
- 39% (BC) and 46% (CO) were higher during the dry period than wet period.
- Highest  $\Delta\text{BC}/\Delta\text{CO}$  ratio ( $18.1 \mu\text{g m}^{-3} \text{ ppmv}^{-1}$ ) in the pre-monsoon season

## GRAPHICAL ABSTRACT

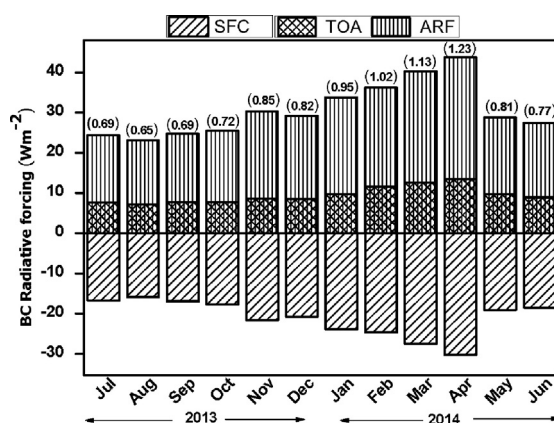


Figure: The monthly mean radiative forcing (RF) at the top of the atmosphere (TOA) due to BC aerosols for clear skies over Guwahati, northeast region of India from 1st July 2013 to 30th June 2014. The RFs values at TOA are varying between  $+7.2$  and  $+13.5 \text{ Wm}^{-2}$  with annual mean ( $+9.5 \text{ Wm}^{-2}$ ), however, at surface (SFC), the RFs values are  $-15.9$  to  $-30.3 \text{ Wm}^{-2}$  with mean ( $-21.1 \text{ Wm}^{-2}$ ), which indicate the net warming and cooling effects respectively. The Atmosphere radiative forcing (ARF) due to BC was  $+30.16 \text{ Wm}^{-2}$  indicates warming effect to the atmosphere over Guwahati.

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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  $1.20 \text{ ppm}$  with a mean value of  $0.51 \pm 0.19 \text{ ppm}$  during the study period. The concentrations of BC ( $8.37 \mu\text{g m}^{-3}$ ) and CO ( $0.67 \text{ ppm}$ ) were  $\sim 39\%$  and  $\sim 55\%$  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

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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  $-21.1 \text{ Wm}^{-2}$  which indicates the net warming and cooling effects, respectively. The highest RF at SFC was in the month of April ( $-30 \text{ Wm}^{-2}$ ) which is coincident with the highest BC mass level. The BC atmospheric radiative forcing (ARF) was  $+30.16$  (annual mean)  $\text{Wm}^{-2}$  varying from  $+23.1$  to  $+43.8 \text{ Wm}^{-2}$ . The annual mean atmospheric heating rate (AHR) due to the BC aerosols was  $0.86 \text{ K day}^{-1}$  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.

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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., 2011a,b; 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  $1.1 \text{ Wm}^{-2}$  (Bond et al., 2013), which is larger than that of methane and is around two third of the  $\text{CO}_2$  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  $\sim 10$  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  $0.23 \text{ Wm}^{-2}$  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 similarly 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 and Lal, 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  $\sim 8 \text{ Tg year}^{-1}$  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,b, 2014; Sarkar et al., 2014; Singh et al., 2015a,b; 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  $0.04 \text{ K/decade}$  (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.

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