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Assessment of Indoor & Outdoor Black Carbon Emissions Rural Areas of Indo-Gangetic Plain: Seasonal Characteristics, Source Apportionment and Radiative Forcing

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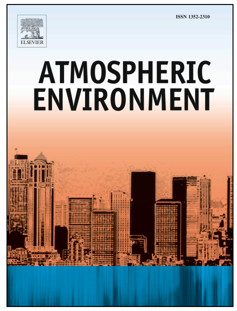

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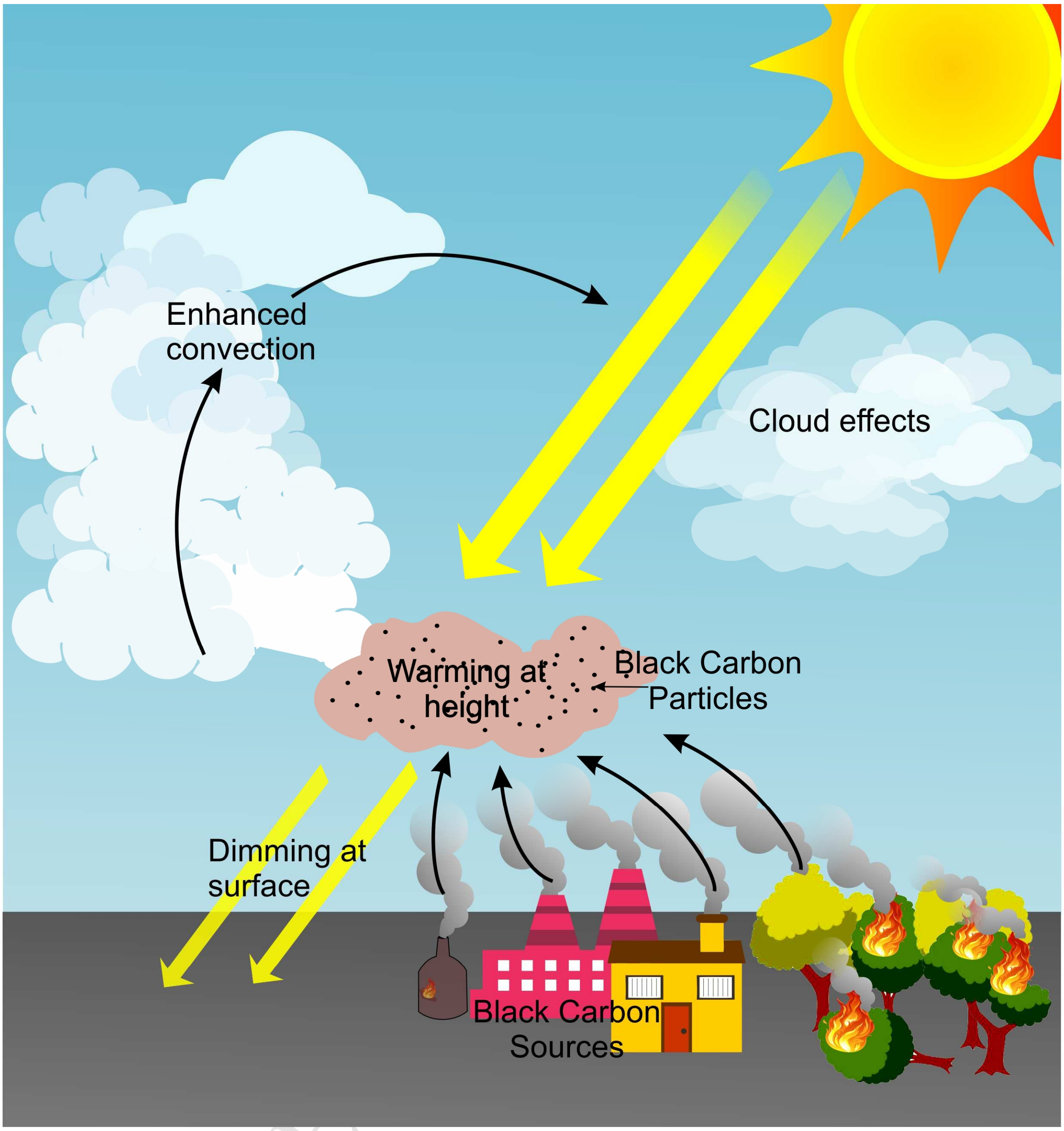
Enhanced
convection
Cloud effects
Warming at
height
Black Carbon
Particles
Dimming at
surface
Black Carbon
Sources

Assessment of Indoor & Outdoor Black Carbon emissions rural areas of Indo-Gangetic Plain: seasonal characteristics, source apportionment and radiative forcing

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Abstract

Black Carbon (BC) has been widely recognized as the second largest source of territorial and global climate change as well as a threat to human health. There has been serious concern of BC emission and its impact in Indo-Gangetic Plains (IGP) due to the use of biomass and fossil fuels for cooking, transportation and industrial activities. An attempt has been made to study indoor (Liquefied Petroleum Gas- LPG & Traditional cookstoves users households) and outdoor concentrations; seasonal characteristics; radiative forcing and source of apportionment of BC in three districts (Sitapur, Patna and Murshidabad) of IGP during January to December 2016. The seasonal concentrations of BC in LPG (traditional cookstoves) users households were 3.79 ± 0.77 μgm^{-3}(25.36 ± 5.01 μgm^{-3}) during the winter; 2.62 ± 0.60 μgm^{-3} (16.36 ± 3.68 μgm^{-3}) during the pre-monsoon; 2.02 ± 0.355 μgm^{-3} (8.92 ± 1.98 μgm^{-3}) during the monsoon and 2.19 ± 0.47 μgm^{-3} (15.17 ± 3.31 μgm^{-3}) during the post-monsoon seasons. However, the outdoor BC concentrations were 24.20 ± 4.46, 19.80 ± 4.34, 8.87 ± 1.83, and 9.14 ± 1.84 μgm^{-3} during winter, pre-monsoon, monsoon and post-monsoon seasons respectively. The negative radiative forcing (RF) at the surface suggests a cooling effect while a warming effect appears to be occurring at the top of the atmosphere. The atmospheric forcing of BC and aerosols also show a net warming effect in the selected study areas. The analysis of BC concentrations and fire episodes indicated that the emissions from biomass burning increases the pollution

concentration. The backward trajectory analysis through the HYSPLIT model also suggests an additional source of pollutants during winter and pre-monsoon seasons from the northwest and northern region in the IGP.

Keywords: Black carbon, Biomass burning, Radiative forcing, Hotspots, Health impact, Backward trajectory, Firewood, HYSPLIT

1.0 Introduction

Black carbon (BC) concentrations have continuously increasing throughout the world due to growing anthropogenic activities, directly contributes to atmospheric warming and serious threat to human health (UNEP, 2011). BC plays an important role in global climate change after CO_2 (Venkataraman et al., 2005; Bond et al., 2007, 2013; Forster et al., 2007; Gustafsson et al., 2009; Ramanathan and Carmichael, 2008). The spatial distribution of BC has affected monsoon pattern in east/south Asia (Menon et al., 2002; Ramanathan et al., 2001, 2005; Lau et al., 2008) by heating the Himalayan-Tibetan region (Ramanathan et al., 2007; Flanner et al., 2009; Menon et al., 2010). It is also responsible for the enhanced glacier melting (Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2007; Koch et al., 2009a, Menon et al., 2010). During 1999-2004, annual average melting reached to 0.85 min Lahaul/Spiti glaciers of 915 km^2 in Himalaya (Berthier et al., 2007). Such accelerated melting is threat to water supplies and food security, potentially slowing the region's socio-economic development (Lawrence and Lelieveld, 2010).

The Indo-Gangetic Plain (IGP) region is home of 900 million people and one of the highly agricultural productive regions of the world. IGP is reported as one of the largest source of BC emission due to anthropogenic activities, burning of crop residue and biomass, forest fires, vehicular emission, brick kilns and coal based power plants (Prasad et al., 2006, Ramachandran and Cherian, 2008; Rehman et al., 2011; Kharol et al., 2014, Saud, et al., 2012, Pandey and Venkataraman, 2014, Kaskoutis et al., 2014, Singh and Kaskoutis, 2014, Arif et al., 2018, Chauhan and Singh 2018, Sarkar et al., 2018). Enhancement in BC concentrations were also observed during Diwali (light) festival in Greater Noida (Singh and Sharma, 2012).This festival is very popular and celebrated all over India (rural or urban areas), candles are lighted in houses and people play with fire crackers, source of absorbing aerosols and soot particles.

About 90% of rural households of IGP are still using biomass (firewood, cow dung cake and crop residue) for cooking and has been recognized as one of the major sources of BC in the region. As a primary component of particulate matter, BC not only leads to indoor/outdoor air pollution but also have serious threat to human health (Mishra et al., 2005, Grahame and Schlesinger, 2010). Quantification of BC emissions from direct sources like households, transportation, industries and open biomass burning is poorly understood in semi-urban areas of IGP. Therefore, there is a need of national policy and mitigation measures to reduce impact of BC. The preparation of policy and management of BC emissions need data on causes, periodic concentrations, variations and meteorological characteristics of contaminants. Hence, an attempt has been made to study the pollution load and impacts in growing districts of IGP (Sitapur, Patna and Murshidabad) (**Fig. 1**) where approximately, 96% of rural families rely on biofuel cooking (firewood, crop residue, cow dung, kerosene, etc.). In these growing districts, Patna has been ranked among the top 100 air polluted cities in the world (WHO, 2014). Here, we have carried out regular indoor and outdoor BC mass concentration measurements at 120 randomly selected households (LPG and biomass users, 60 each) in the middle of village (12 locations) as well as nearby road (6 locations) in selected districts during the period of January- December 2016. To the best of our knowledge, this is the first comprehensive study on measurements of BC concentration in rural areas. The radiative forcing and indoor/outdoor seasonal BC variations will be of great importance for policy formulation and control of air pollution in IGP. This study is focused to (1) measure indoor/outdoor BC emission, (2) investigate the seasonal and diurnal variations of BC (3) evaluate the potential sources for BC and (4) analyze radiative forcing.

2. Experimental Setup

2.1 Experimental sites and general meteorology

Based on the biofuels use pattern and socio-economic conditions, three districts namely Sitapur (27.6°N, 80.18°E), Patna (25.35° N, 85.12° E) and Murshidabad (23.43º N, 87.49º E) were selected to study spatial distributions of BC concentrations and detailed analysis (**Fig. 1**). The climate of Sitapur and Patna district's is 'sub-tropical humid' and considered as 'Cwa' kind based on the Koppen Climate

Classification (Sanderson, 1999). The summer temperature vises very high upto (40-50^{0}C) in both Sitapur and Patna districts due to the intensity of a tropical sun. The district Murshidabad is tropical and categorized as 'Aw' kind of climate. The annual mean temperature of Murshidabad district is approximately 27 °C and monthly mean temperature ranges from 17-35 °C. The westerly and north-westerly winds bring air mass during the pre-monsoon season; from the west/southeast during the monsoon season and from the north/northeast during the post-monsoon/winter season (Prasad et al., 2006; Moorthy et al., 2007). Hence, these locations were ideal for long-term indoor and outdoor BC measurements to understand the dynamics of aerosols and BC concentrations over the IGP (Ramanathan et al., 2005; Nair et al., 2007) and related climatic impacts (Gautam et al., 2010).

2.2 Instrumentation and data analysis

Six revenue blocks (2 blocks per district) were selected from the above mentioned three districts for the socio-economic survey. In six blocks, 12 villages (2 villages per block) with varying socio-economic conditions were identified through reconnaissance survey for households study (**Table 1**). Total 300 households (25 households per village) were randomly selected from these villages to evaluate socio-economic conditions, fuel consumption patterns, fuel types, health issues, barriers to clean fuel energy accessibility and adaptability of households.

BC concentrations were measured in the cooking area close to traditional and LPG cookstoves in 120 randomly selected households (60 LPG and 60-biomass users). Simultaneously, BC concentration measurements were also carried out in the middle of selected villages (12 locations) as well as nearby roads (6 Nos.). Measurements were done at an interval of five minutes through portable micro-aethalometers (Model AE-42) and aethalometers (Model AE-33), Magee Scientific, USA (Hansen et al., 1984). The observations were made at 370, 470, 520, 590, 660, 880 and 950 nm wavelengths. The emissions of BC from fossil fuel provides peak at 830 nm wavelengths while other components of aerosol have irrelevant absorption peak at this wavelength, hence, 880 nm channel was considered for measurement of BC concentrations. The inlet pipe was 0.15 m and the instruments were fixed at 1 m aside and 1 m above the surface to receive uniformly diffused concentration from the cookstoves (indoor) and other sources (outdoor). The flow rate of aethalometers was set at 3 L min^{-1} because of huge emissions at these locations. Details of instrument, uncertainties and rectifications can be referred in numerous publications (Hansen et al., 1984; Babu and Moorthy, 2002; Weingartner et al., 2003; Arnott et al., 2005; Schmid et al., 2006).

2.3 Fire count analysis and transport pathway

The enhanced BC concentrations in the IGP has also been reported due to the agricultural residue burning in fields and forest fire in northwest, northeast and central states of India (Singh et al., 2014, Singh and Kaskaoutis 2014, Sarkar et al. 2018). The fire spots in agriculture field and forest were counted through National Aeronautics and Space Administration's Earth Observatory and Firms Web Fire Mapper data (Tipayarom et al., 2007). To study the effects of agricultural residue and forest fire on BC concentrations, a correlation between the fire counts from MODIS and the outdoor BC concentrations were also analyzed. The eight days backward trajectories were computed for each district by using HYSPLIT4 model (Dumka et al., 2013; Draxler and Rolph, 2014; Dumka et al., 2015; Bisht et al., 2015). The HYSPLIT4 model helped in examining the impacts of other probable sources on measured BC concentrations, local air quality and the other neighboring areas. The global reanalysis data were utilized as an input for calculating isentropic backward trajectories. In IGP, most of the farmers generally burn their agricultural straw between 18:00 to 21:00 hrs (local time) and 19:00 hrs was considered as starting time for computation of trajectories and pathways of pollutants.

2.4 Estimation of radiative forcing

Aerosols vary in their chemical compositions that control the radiative forcing and aerosol cloud interactions (Boucher et al., 2013). In Intergovernmental Panel on Climate Change, Fifth Assessment Report (AR5), total aerosol forcing associated with black carbon is estimated as -0.03 and +0.02 Wm^{-2} over the periods1990-2010 and 2000-2010 respectively (Myhreet al., 2013). We have computed radiative forcing (RF) using Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998, Prasad et al., 2007, Tiwari et al., 2016).The measured BC concentrations were used into the OPAC model (Hess et al., 1998) to calculate optical depth and single scattering albedo.

Further, the output of OPAC model was used in the SBDART model to assess the radiative forcing of both aerosol and BC. The radiative forcing of both aerosol and BC for atmospheric layers were computed at 5^o zenith interval to compute the average diurnal forcing. The average diurnal forcing at the surface (SUR) and top of the atmosphere (TOA) were estimated separately. The atmospheric forcing (ATM) was computed as difference of TOA and SUR forcing along and beyond the elements contributing to warming.

3.0 Results and Discussion

3.1 Households Biofuel Consumption

The study area considered in the present study shows 90-95% rural households use biofuel cooking (firewood, crop residue, cow dung cakes, coal, and kerosene). Firewood was the primary energy source for cooking in 44.93% households in Sitapur district followed by cow dung cake (26.63%), crop residue (24.16%), LPG (3.12 %) and other sources (1.12 %). In Patna district, cow dung cake was the primary source of fuel for cooking in 42.16% households followed by crop residue (30.85%), firewood (20.42%), LPG (4.14%) and other sources (2.36%). In the Murshidabad district, crop residue was the primary source for cooking in 52.24% households followed by cow dung cake (28.86 %), firewood (14.32%), LPG (2.10 %) and other sources (2.10 %). Figure 2 shows distribution of different sources of biofuel cooking in the considered study locations. The average monthly consumption of firewood and coal was 145-154 kg, while the average monthly consumption of LPG was 10 kg for each household. It was also found that each household spends ~Rs. 470.00 per-month (US$ 7-8/month) to access clean fuel energy (LPG) and on an average of Rs. 560 per month (US$ 8-9/month) to purchase firewood and coal. One of the interesting findings from the survey is that large numbers of households are spending money to purchase traditional fuel compared to the clean energy due to cultural preferences, fuel availability, accessibility and inadequate LPG distribution centers. The average use of LPG is upto 3.12 % in rural households in these districts that needs to be enhanced to use clean fuel (LPG) to reduce BC emissions indoors as well as in outdoors. Energy and its technologies play a key role in socio-economic development of the community to the national levels to minimize threat to human health, environmental pollution and climate impacts

The primary survey results of selected districts were also compared with Census of India's rural household fuel use data (for 2001 and 2011) to understand the fuel use pattern and validation of survey results. The decadal average percentage use of firewood in these districts has increased by 6.49% (0.649%/year) followed by LPG 2.77%, and crop residue 1.26% during 2001-11 due to increase of 5.83 lakh (0.583 million) rural houses (0.583 lakh/year) (1 lakh = 10^5). However, there is an increase of 5.19% (@0.519%/year) use of firewood and 2.77% in LPG with the increase of 121.16 Lakh (12.116 million) rural houses (12.116 Lakh/year) during the same period in IGP, which are using large percentage of traditional sources of cooking (**Table 2**).

As per the 2015 survey, crop residue (35.65%) was the primary fuel for cooking in rural households (**Fig. 3**) followed by cow dung cakes (32.56 %), firewood (26.56%) and LPG (3.12%). The 2011 Census data also indicates that crop residue was the primary energy source for cooking in 34.48% of the rural households of these districts of IGP followed by firewood (31.40%), cow-dung cakes (26.25 %) and LPG (3.73%). An increase of 6.31% use of cow dung cake (1.262%/year) and 1.17% in crop residue during 2011-15 in rural households due to increase of rural houses as well as fuel availability and accessibility, however, other kind of fuels show a declining trend. Further, an average use of clean fuel is much lower in these districts (3.12-3.73%) as compared to IGP (5.68%) and Indian average (11.4%) while higher in traditional fuel, which needs to enhance the use of clean fuel (LPG) in the region to minimize the impacts of BC.

3.2 Indoor BC emissions from households using traditional and LPG cookstoves

The type of cooking fuel is the main contributor of increased BC concentrations. High BC concentrations were observed in both indoors and outdoors environment during cooking hours. The daily BC mass concentration in the proximity of traditional Cookstoves users varies from 0.06 to 21.86 μgm^{-3} in the selected households during the measurement periods. The highest average indoor BC concentrations were found during the winter season (December to February) while lowest during the monsoon (June to September) (**Fig. 4a**) in the study area. The indoor mean BC concentration shows highest concentration (25.36 ±5.01 μgm^{-3}) during the winter and lowest (8.92 ± 1.98 μgm^{-3}) during the monsoon seasons in traditional cooking (cookstoves users). The observed BC concentrations are found to be different from earlier study by Rehman et al. (2011) in Kanpur city (it may be noted that the Kanpur is an industrial city where the economic conditions is much better than the three locations considered, many people use LPG compared to other sources of biofuel cooking). Similarly, the BC concentrations were observed maximum (3.79 ± 0.77 μgm^{-3}) during the winter season and minimum (2.02 ± 0.355 μgm^{-3}) during the monsoon season using LPG as source of cooking. The monthly average indoor and outdoor BC concentrations are summarised in **Table 3**. The seasonal concentrations in traditional cookstove user's households were 25.36 ± 5.01 μgm^{-3}during the winter season, 16.36 ± 3.68 μgm^{-3} during the pre-monsoon (March to May), 8.92 ± 1.98 μgm^{-3}during the monsoon and 15.17 ± 3.31 μgm^{-3}during the post-monsoon (October to November) (**Fig. 4b**). Similarly, the seasonal mean concentrations in LPG user's households were 3.79 ± 0.77 μgm^{-3} in the winter; 2.62 ± 0.60 μgm^{-3} in the pre-monsoon; 2.02 ± 0.355 μgm^{-3} in the monsoon and 2.19 ± 0.47 μgm^{-3} in the post-monsoon (**Fig. 4c**). In Sitapur, BC concentrations vary from 1.80 to 25.36 μgm^{-3} during the entire pre-monsoon season while, from 1.0 to 25.16 μgm^{-3} in Patna and 1.0 to 24.52 μgm^{-3} in Murshidabad districts. The BC concentration was observed to be the highest (25.36 μgm^{-3}) in cow-dung cake and crop residue user's households as compared to firewood user's households due to low calorific value (9.79 to 14.20 MJ/ kg) and thermal efficiency (8.90-17.10%) of cow-dung cake (Harshika et al., 2014). It has resulted in the wastage of biofuel and the huge BC emissions from the traditional cookstoves.

It is noted that 90-95% rural households of selected districts are using traditional cooking fuels with high emission of BC concentrations indoor environment that leads to the indoor/outdoor air pollution and high health problems in the IGP. The Government of India has introduced several programs to reduce BC emissions from residential area such as improved cookstove program, family-size biogas plants, community biogas plants and Ujjawala scheme (distribution of LPG to poor people) for rural households and compressed natural gas in transportation sector. Still the BC emissions from residential is not reducing and making a potential area for research to understand the effectiveness of these programs.

It is well known that while cooking, people inhale BC and particulate matters from burning of bio/fossil fuels. The health of women and children are more vulnerable due to closer and larger inhalation of fine particulate matter while cooking and being near high concentration zone. It is also well known that the emissions from biofuel cooking have serious health problems such as acute lower respiratory infections, lung cancer, blindness (cataract), tuberculosis (TB), asthma, and chronic obstructive pulmonary disease as well as heart disease among the women and children (Smith 2000; Smith et al. 2000a; Parikh et al. 2001). The impact is maximum in rural areas due to lack of clean fuel, improved cookstoves and separate and ventilated cooking (kitchen) place. The primary survey conducted in selected villages, where females were also interviewed particularly on the health concerns to understand the negative health impacts associated with emissions. The survey reports found similar findings, women are cognizant of higher health impacts (both short and long-term health effects). About 26% females reported eyes watering during the cooking time followed by eyes itching (25%), asthma and respiratory problems (22%), cardiovascular disease (17%) and coughing (10%). These results were further verified with the data available at nearby community health centers. It has also been noted that these effects have immediate implications for spending per household expenditure on health (Rs. 258-275/month). It may be noted that no official data about the human health suffering were available from the rural or city hospitals, so we are not able to discuss any data. However, we consulted nearby PHC/CHC doctors about the common diseases in the area and survey results were validated with the doctors.

3.2.1 Indoor Diurnal Variation of BC

The hourly mean diurnal variations of BC for 120 selected households (60 biofuels and 60 LPG users) are shown in **Fig.4b**. In the morning cooking hours (06:00 to 09:00 hrs), indoor BC mass concentrations in biofuel cooking were observed to vary from 1.80 to 22.16μgm^{-3} with an average value of 12.15 μgm^{-3} in all selected households. While in evening cooking hours (17:00 - 20:00 hrs), BC concentrations vary in the range 1.90 to 25.36μgm^{-3} with an average value of 13.6 μgm^{-3}. However, at LPG cooking loctions, indoor BC concentrations vary in the range 0.26-3.19 μgm^{-3} with an average value of 1.62 μgm^{-3}in the morning cooking hours and in the range 0.13-3.79 μgm^{-3} in the evening cooking hours with an average value of 1.45 μgm^{-3} in all selected households (**Fig.4c**). The seasonal analysis of BC concentrations shows that the biomass user's households were exposed to 25.36 ± 3.12 μgm^{-3} (3.79 ± 0.58 μgm^{-3}) during the winter; 15.15 ± 1.45 μgm^{-3} (2.38 ± 0.45 μgm^{-3}) during the pre-monsoon; 8.40 ± 0.75 μgm^{-3} (2.02 ± 0.28 μgm^{-3}) during the monsoon and 13.69±1.25 μgm^{-3} (1.99 ± 0.17 μgm^{-3}) during the post-monsoon season in the morning cooking. While during the evening cooking, the mass concentrations were about 19.31 ± 2.82 μgm^{-3} (3.24 ± 0.76 μgm^{-3}); 14.59 ± 2.29 μgm^{-3} (2.47 ± 0.40 μgm^{-3}); 8.29 ± 1.51 μgm^{-3} (1.47 ± 0.29 μgm^{-3}) and 13.45 ± 2.18 μgm^{-3} (1.98 ± 0.32 μgm^{-3}) during the winter, pre-monsoon, monsoon and post-monsoon seasons, respectively. The indoor BC concentrations in LPG user's households was ten times (90.3%) lower in LPG cooking compared to biofuels cooking during entire study periods due to high calorific value (46.1 MJ/kg) of LPG and thermal efficiency of LPG gas stove (64%). It is pertinent to mention here that only 3.12 -3.73 % households use LPG as a cooking fuel source that makes the region vulnerable to both health and climate impacts.

3.3 Outdoor BC Concentrations in the center of village

The BC measurements (outdoor) were also carried out in the center of village to compare the impact of indoor BC emissions and other sources. During morning cooking hours, outdoor BC mass concentrations varies in the range of 4.39 -24.20 μgm^{-3} with an average value of 18.51 μgm^{-3} while in the evening hours, BC concentrations vary in the range of 8.06 -21.72 μgm^{-3} with an average value of 16.07 μgm^{-3}. To analyse the variation of BC, it is essential to understand the influence of the local, regional and national emission sources. The daily BC mass concentration in the vicinity of designated sites vary in the range of 0.98-24.20 μgm^{-3} during the whole study period. BC concentrations vary in the range of 0.98-24.20 μgm^{-3} in Sitapur, 1.70-24.10 μgm^{-3} in Patna and 1.0-23.1 μgm^{-3} in Murshidabad districts throughout the monitoring period (over the year).

The highest average outdoor BC concentrations were observed during the winter season while lowest during the monsoon season (**Fig. 4a**). The mean mass concentration of BC was at its maximum (24.20 ± 4.46 μgm^{-3}) during the winter season while minimum (8.67 ± 1.83 μgm^{-3}) during the monsoon season because of rainfall. The BC concentrations show maxima peak (24.20 ± 4.46 μgm^{-3}) in the month of

January because of lower planetary boundary layer (PBL), increased biofuel burning and brick kilns activities. The seasonal outdoor BC concentrations were 24.20 ± 4.46 μgm^{-3}during the winter season, 19.80 ± 4.34 μgm^{-3} during the pre-monsoon, 8.87 ± 1.83 μgm^{-3} during the monsoon and 9.14 ± 1.84 μgm^{-3} during the post-monsoon seasons (**Fig. 5a**). Apart from the local emissions and meteorological conditions, long-range pollutants from crop residue burning, forest fires and dusts from Thar and Arabia peninsula further change the particles size, mixing of the dust and BC influencing the climatic conditions (Dey et al., 2004, Prasad and Singh, 2007, Gautam et al. 2010).

The diurnal variation of BC concentrations at local scale is important to understand the local emission variability that helps us to calculate regional and national emissions (Tiwari et al., 2013). The outdoor BC diurnal variations presented a similar pattern as indoor concentrations with the peaks in the morning and evening food cooking cycles that suggest the strong impact of indoor cooking on outdoor BC mass concentrations. BC concentrations start increasing before sunrise, with large peaks in morning hours due to high BC emissions from other local emissions. The high BC concentrations observed during evening hours as compared to the morning hours with BV values greater than 18 μgm^{-3} in outdoor environment. The ratio of BC mass concentrations were approximately 35% higher during 19:00 to 22:00 hrs in outdoor environment, showing emissions from local households and other sources. The maxima peaks are attributed mainly due to local (residential and industrial), traffic and crop residue burning that do not change in these months. The enhanced mass concentrations in the residual layer enhanced mass concentrations at the surface during burning period.

The wind speed and height of the atmospheric boundary layer show an important role in the diurnal variation of concentrations and dispersion of pollutants. To recognize the significance of deviations in the boundary layer height on diurnal concentrations, the relationship between BC and mixing layer depth (MLD) was analyzed around monitoring sites with the NOAA HYSPLIT model (https://www.ready.noaa.gov/HYSPLIT_traj.php). The mixing layer heights were calculated from the impending temperature profile by detecting the height of an eminent inversion at each point. The average mixing height was found to be lower (30%) in the evening compared to the daytime. It is important to note that BC concentrations were decreased (~2.45 μg m^{-3}) at 15:00 hrs associated with the high wind speed, temperature, allow fast dispersion horizontal and vertical direction of pollutants during the pre-monsoon season. The BC concentrations found to increase further after 16:00 hrs in the study region (**Fig. 5b**). The ratios of BC mass concentrations were approximately 35% higher during 19:00 to 22:00 hrs in outdoor environment due to reduced wind speed, low temperature, lower mixing layer and high use of traditional fuel for cooking is an indicative of high local emission from households influenced by meteorological factors. The BC values again gradually fall after 22: 00 hrs due to reduced domestic emissions. It is found that diurnal deviation of BC concentrations attributed to the higher use of biofuels for cooking and other purposes. The observed BC concentrations were similar to BC concentrations measured in Varanasi (Singh and Rai, 2014) and slightly high from Gorakhpur (Vaishya et al., 2017), Ballia (Tiwari et al., 2016), Kanpur (Kanawade et al., 2014) and Agra (Safai et al., 2008) (**Table 4)**. These findings confirm that rural and small cities are major contributors of BC emission as in the case of large cities in IGP, affecting poor air quality, warming of the region and threat to human health.

3.4 BC emissions from the transportation sector

BC emissions in the study area are not only attributed to residential area but also from other sources like transportation. Hence, the emission from transport (highway) were also measured at the NH-30 (Lucknow to Shajahanpur road at Sitapur), SH-21 (Sitapur-Lakhimpur Khiri road at Laharpur), NH-22 (at Patna), NH-431 (Patna - Fatuha Road at Fatuha), NH-114A (Behrampure - Jalangi Road at Behrampur) and NH-12 (Farakka - Malda Road at Farakka). The main findings of the monitoring of mobile sources is that BC concentrations during the morning and evening hours show similar trend as observed in the centre of the village. During morning time (05:00 to 10:00 hrs), BC concentrations were observed from 3.13 to 19.85μgm^{-3} with an average value of 9.44 μgm^{-3} while during evening hours (17:00 to 22:00 hrs), the BC emissions vary in the range of 3.48 to 20.82μgm^{-3} with an average value of 9.60 μgm^{-3}. The monthly average of BC concentrations was observed to be highest level (20.82 ± 4.76 μgm^{-3}) during winter and pre-monsoon seasons while lowest (8.92 ± 1.98 μgm^{-3}) during monsoon season (**Fig. 6**). BC concentrations are washed out due to precipitation. Higher BC concentrations in the month of January is also associated with the higher bio/fossil fuel burning for cooking as well as heating, municipal waste burning, brick kilns activities, and lower PBL depth/ mixing height.

The seasonal concentrations of BC were 20.82 ± 4.76 μgm^{-3} during winter, 14.97 ± 3.34 μgm^{-3} during pre-monsoon, 8.92 ± 1.98 μgm^{-3} during monsoon and 10.71 ± 2.38 μgm^{-3} during post-monsoon seasons. The diurnal variations show minimum BC concentration (1.68 μgm^{-3}) at NH-431 (Fatuha) and highest (19.55μgm^{-3}) at NH-30 (Sitapur) due to heavy traffic flow (550 vehicles/hrs). BC mass concentrations start to increase before sunrise and reach at the maximum level during 06:00 - 09:00 hrs and show low values around 15:00 hrs. This pattern suggests that indoor BC emissions also affect the on road BC concentrations. It is noted that the roadside BC mass concentrations were drastically reduced in mid-afternoon (15:00 hrs) due to reduced vehiclar movement, high speed, low emission and negligible cooking activities as well as the vertical and horizontal dispersion of the atmospheric BC. In evening, the mean BC concentrations increased due to increased vehicular movement and cooking activities and reduction in vertical mixing.

3.4.1 The effect of weekend on black carbon emissions

The dynamics of BC concentrations were studied on the weekdays and weekends. Earlier studies (Zhang et al., 2009; Sahu et al., 2011; Mascia et al., 2016**)** have shown that the particulates and gaseous pollutants depend upon the location due to diverse surroundings and atmospheric settings. In the large cities, offices, academic institutions and other business activities (industrial and commercial) remain close during the weekend. However, in the rural areas, activities remain same except the movement of vehicles on the highways and nearby other roads. Pronounced reduction in aerosol optical depth and mass concentration of aerosol has been observed in Bangalore city (Satheesh et al., 2011) during the weekend. To quantify the findings, the effect of change in anthropogenic events, industrial activities and traffic flow on BC concentrations were examined during weekdays and weekend near NH-30 (Sitapur), SH-21 (Laharpur), NH-22 (Patna), NH-431 (Fatuha), NH-114A (Behrampure) and NH-12 (Farakka). The average evening peak of BC concentrations were observed between 1.68 to 19.55 μgm^{-3} in weekdays and 3.91 to 5.86 μgm^{-3} in weekends. The diurnal variations show minimum concentrations on weekends, particularly on

Saturday at NH-431 (3.91 µgm^{-3}) and NH-114A (3.95 µgm^{-3}). However, high BC concentrations were observed on the weekend at NH-30 (4.78 µgm^{-3}) and NH-22 (5.86 µgm^{-3}) due to heavy traffic movement in the weekend. These roads were very close to state capital and district headquarters.

3.4 The role of agriculture biomass burning

Substantial increases in agriculture residue burning have been reported over the northwestern parts of India (Singh et al., 2014, Singh and Kaskaoutis 2014, Sarkar et al. 2018). Recent study by Sarkar et al., (2018) has shown that the crop residue burning influences greater parts of India. Chauhan and Singh, (2017) has reported that Diwali festival and crop burning severely impacted weather conditions, air quality and visibility of National Capital Region of India for a week. However, the National Green Tribunal, of India has banned agricultural biomass burning but this practice is still going on in north, north-west and central regions of India. Farmer's burn their wheat crop residue during the May-June, mid-October and mid-November after the harvesting of rice crop. During the mid-October and November, temperature is rather cool. So the severe impact of crop burning is observed over Delhi and its surrounding areas. However, burning of wheat crop is not a severe problem in summer due to the warmer weather conditions and mixing height that result into a fast dispersion of air pollutants (Singh and Kaskoutis, 2014). This crop burning is a source of large amount of carbonaceous aerosols in the IGP (Venkataraman et al., 2006; Kaskoutis et al., 2014, Singh and Kaskoutis, 2014) and a serious threat to human health like asthma, respiratory, heart and lung diseases. To understand their effects of biomass burning on BC concentrations, we have used the MODIS-derived fire products (http://modis-fire.umd.edu/index.php). The weekly number of fire counts and average BC concentrations were correlated and is shown in **Fig. 7a**. The BC mass concentrations increase up to 24.20 µgm^{-3} during winter and pre-monsoon seasons as a result of crop burning and forest fires in the study areas and surroundings. The monthly average BC mass concentrations at Sitapur and Patna are found to be high (24.20 µgm^{-3}) as compared to Murshidabad (23.10 µgm^{-3}) during the winter and pre-monsoon seasons. The BC mass concentrations during the June - October are found to be much lower (almost half of the BC concentrations during the winter and pre-monsoon seasons) with the reductions in forest fires and agricultural crop burning.

The inter-relationship between the monthly fire counts and monthly average BC concentrations are also examined (Tipayarom et al., 2007) that shows a relatively better linear relationship (R^2= 0.564). This relationship suggests that agricultural residue burning during winter and pre-monsoon season is one of the major causes for increase in BC concentrations (**Fig. 7b)** because of great pressure, rigorous photochemistry and absence of a removal procedure (Zhang and Kim Oanh, 2002).

3.5 Aerosol radiative forcing

Daily radiative forcing (RF) over IGP was also estimated for BC and composite aerosols shown though Figure 8. The seasonal TOA radiative forcing of aerosols and BC was found to be 20.6 and 18.5 Wm^{-2} respectively during winter season; 25.5 and 21.1 Wm^{-2} during pre-monsoon; 21.6 and 17.2 Wm^{-2} during monsoon and 16.8 and 6.87 Wm^{-2} during post-monsoon season. The SUR forcing due to aerosol was -50.8, -40.2, -20.7 and -10.2 Wm^{-2} and due to BC was -19.5, -22.0 -13.5 and -5.86 Wm^{-2} respectively during winter, pre-monsoon, monsoon and post-monsoon. An inconsistency was observed in the radiative forcing over study area due to the inconsistent presence of absorbent particles due to burning of fossil fuel and biofuels. The atmospheric radiative forcing for aerosols and (BC) was estimated as +75.8 (+39.7), +77.1(+42.1), +34.7 (+20.8) and +25.1 (+10.2) Wm^{-2} during the winter, pre-monsoon, monsoon and post-monsoon respectively. ATM, TOA, positive radiative forcing of BC and aerosol particles are indicative of a warming effect while the SUR radiative forcing shows a cooling effect in the study areas. These results are quite high in these semi-urban areas and large differences were observed in the radiative forcing at surface, atmosphere and top of the atmosphere due to the presence of absorbing aerosols. These radiative forcing may be high due to influence of dust particles and BC emission from western region of IGP during the winter and pre-monsoon season (Dey et al., 2004, Singh et al., 2004). Many researcher in the Indo-Gangetic plains (Prasad et al., 2007, Day and Tripathi, 2008; Gautam et al., 2010, Tiwari et al., 2016) also found similar results. The impact of high radiative forcing is also clearly visible over the Himalayas and Tibetan Plateau (Zhang et al., 2015).

3.6 Role of long-range transport of dust

The dominant westerly winds transport dust to a long range from Thar Desert and Arabia Peninsula in the Indo-Gangetic plains. The dusts are transported up to the eastern parts of the IGP depending upon the meteorological conditions and wind speed during pre-monsoon season (Dey et al., 2004, Gautam et al., 2009, Srivastava et al., 2010b) and transport to central and eastern parts of IGP. Apart from the dust from the desert, the impact of emissions from the open burning of crops/ forest fires from the north/central region of India and black smoke consists of carbon particles from coal based power plants were also observed over IGP (Ramachandran and Cherian, 2008, Prasad et al., 2009, Sarkar et al. 2018). The emissions from these sources located in the nearby areas and long-range transport of pollutants are likely to influence our BC measurements. The resident time of BC in atmosphere is ~1 week to 10 days (Reddy and Venkataraman, 1999), so 8 days isentropic backward trajectories were examined by using HYSPLIT model to know pollutants transport pathways. The NOAA HYSPILT backward trajectories show directions of the air mass reaching at different measuring locations (**Fig. 9**).

During the winter and pre-monsoon seasons, BC concentrations varies in the range 20.1-24.82 μgm^{-3} due to transport of air mass from western parts of IGP. About 90% of the backward trajectories reach Sitapur from the northwestern regions and 10% from western areas. At Patna, dominant air mass comes from north-west parts and less from southeast region (**Fig. 9**). Similarly, about 98% of the trajectories bring air mass from northwest and western parts at Murshidabad that carry dust from Thar Desert (located in the western parts of India, however dust observed in the IGP are mainly from Arabia peninsula (Dey et al., 2004). Long-range transport of dust mixes with anthropogenic emissions along the track of dust, enhancing local BC concentrations (Bhattacharjee et al., 2007). Similar findings were also reported at Peshawar (Khan et al., 2015), Iran (Shahsavani et al., 2012) and Beijing (Zhao et al., 2009). During monsoon season, the pollutants were washed out from the atmosphere as a result BC concentrations reduced upto 36% compared to other season, mainly from local indoor emissions from cooking. These kinds of emission flow patterns not only have implications on the human health but also on the crop production and local/regional climate (Auffhammer et al., 2006). The pollutants carried from the other surrounding of IGP do affect the eastern region of IGP and eastern countries.

4.0 Conclusions

BC measurements were carried out in both indoor (LPG and traditional cookstoves users) outdoor (middle of village, roadside of the village and highways) to study diurnal and seasonal characteristics; radiative forcing; source of apportionment over the three districts of IGP. Following conclusions are drawn from our present study:

- **i.** Crop residue (35.65 %) was the primary fuel for cooking in rural households followed by cow dung cakes (32.56 %), firewood (26.56 %) and LPG (3.12 %). Use of cow dung has enhanced up to 6.31% (1.262%/year) and 1.17% crop residue during 2011- 2015 in rural households while other means of cooking have declined. Each household spends ~Rs. 470.00 per-month (US$ 7-8/month) to access clean fuel energy (LPG) while an average of Rs. 560 per month (US$ 8-9/month) to purchase traditional fuel and coal. The use of LPG is restricted in these rural areas due to economic conditions and accessibility of LPG. The present Government policy to use clean energy and provide free access to LPG in rural area may reduce BC emissions.
- **ii.** The peak values of BC are observed during morning and evening hours. In the indoor environment, BC concentrations vary in the range 1.80 to 22.16 μgm^{-3} during morning hours and 1.90 to 25.36 μgm^{-3} in the evening. Similarly, use of LPG reduces indoor BC concentrations, 0.26 to 3.19 μgm^{-3} during morning hours and 0.13 to 43.79 μgm^{-3} during evening hours. Pronounced reduction in BC concentrations upto 90.3% was found with the use of LPG during both morning and evening hours in all three districts.
- **iii.** The seasonal mean mass concentration of BC were 25.36 ± 5.01, 16.36 ± 3.68, 8.92 ± 1.98 and 15.17 ± 3.31 μgm^{-3} with the biomass indoor use during winter, pre-monsoon, monsoon and post-monsoon respectively. Similarly, the seasonal mean concentrations with LPG use were 3.79 ± 0.77, 2.62 ± 0.60, 2.02 ± 0.355 and 2.19 ± 0.47 μgm^{-3} during winter, pre-monsoon, monsoon and post-monsoon respectively. LPG use are able to reduce the BC concentration by 85, 84, 77 and 86

percent during winter, pre-monsoon, monsoon and post-monsoon respectively in case of indoor BC concentration.

iv. The outdoors, concentrations vary in the range 4.39-24.20 μgm^{-3} in the morning and 8.06-21.72 μgm^{-3} in the evening hours while the seasonal mass concentrations of BC were 24.20 ± 4.46, 19.80 ± 4.34, 8.87 ± 1.83 and 9.14 ± 1.84 μgm^{-3} during winter, pre-monsoon, monsoon and post-monsoon respectively.

v. The BC concentrations was highest (~35%) during evening (19:00 to 22:00hrs) as compared to morning due to the contribution of BC emissions from agriculture biomass burning.

vi. The correlation between the weekly number of fire episodes and average BC concentrations show a linear relationship (R^2= 0.564)suggesting that the burning of agricultural residue during the winter and pre-monsoon season worsen the air quality in the IGP, and some study (Sarkar et al. 2018) reported the impacts to a greater parts of India.

vii. ATM and TOA positive radiative forcing of BC and aerosol particles show a net warming impact in the study area while the SUR radiative forcing shows a cooling effect.

viii. The backward trajectories analysis helped in understanding the source and the region of the pollution. The biomass burning in Pakistan and Afghanistan, Punjab, Haryana and Uttar Pradesh), dust aerosols from Gulf countries and Western states of India and industrial pollution from highly industrialized northern parts of India are responsible for the high BC concentrations (40-45 μgm^{-3}) during the winter and pre-monsoon.

ix. The health impacts of BC are severe and affects people to suffer with the eyes watering (26%), itchy eyes (25%), asthma and respiratory problems (22%), cardiovascular disease (17%) and coughing (10%). These impacts tend to be particularly large in rural India since households often lack ventilation in cooking areas, even in rural areas people used to sleep in the cooking place.

Our present results will be of great help to the Ministry of Environment, Forest and Climate Change, India, World Health Organization (WHO), Environmental Protection Agency (EPA) and other global and

National agencies to formulate policy to limit BC emissions and follow clean air act to save millions of lives.

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List of Tables

Table 1: Survey and BC monitoring Locations

State	District	Block	Town/Village	Geo-coordinates
Uttar Pradesh	Sitapur	Khalilabad	Sitapur Town	27° 33' 26.5" N, 80°39' 50.4"E
			Iqri	27° 24' 52.7" N, 80°34' 11.5"E
		Laharpur	Laharpur	27° 42' 46.53" N, 80° 54' 40.19"E
			Bilariya	27° 46' 59.9" N, 80° 55' 11.7"E
Bihar	Patna	Fatuha	Maksoodpur	25° 24' 42.9" N, 85° 17' 24.3"E
			Shukalpur	25° 32' 25.9" N, 85° 15' 57.0"E
		Patna Sadar	Meethapur	25° 35' 24.9" N, 85° 08' 08.9"E
			Mohanpur Kachhuara	25° 33' 21.89" N, 84° 11' 01.30"E
West Bengal	Murshidabad	Behrampur	Basudevkali	24° 04' 22.8" N, 88° 13' 19.0"E
			Harishpur	24° 09' 17.5" N, 88° 20' 49.5"E
		Farakka	Tildanga	24° 47' 28.5" N, 87° 52' 24.9"E
			Amtala	24° 43' 32.6" N, 87° 54' 04.0"E

Table 2: Average fuel used in Indian Households and IGP locations

Fuel Type	Average Percentage of Fuel used for cooking													
	Census 1991			Census 2001					Census 2011					Primary Survey, 2016
	All India	IGP	Selected districts	All India	IGP	Selected districts	Decadal Change (Decrease/Increase) IGP	Decadal Change (Decrease/Increase) Selected districts	All India	IGP	Selected districts	Decadal Change (Decrease/Increase) IGP	Decadal Change (Decrease/Increase) Selected districts	
Firewood	71.69	48.81	31.71	64.1	41.6	24.91	-7.21	-6.8	62.55	46.79	31.4	5.19	6.49	26.56
Crop residue	--	--	--	13.1	23.82	33.22	100	100	12.33	22.07	34.48	-1.75	1.26	35.65
Cow-dung	19.6	37.79	43.99	12.8	26.23	32.62	-11.56	-11.37	10.87	21.84	26.25	-4.39	-6.37	32.56
LPG	1.22	0.82	0.26	5.7	2.91	1.43	2.09	1.17	11.4	5.68	3.73	2.77	2.3	3.12
Other (Coal & Charcoal, Kerosene, Electricity, Biogas)	7.46	12.53	23.99	4.3	5.2	7.49	-7.33	-16.5	2.61	3.45	3.97	-1.75	-3.52	1.86

Table 3: Monthly average indoor, outdoor and on road BC concentrations during January, 2015 to December 2016

Month	Sitapur				Patna				Murshidabad			
	BC in Biomass Users Household (μgm^{-3})	BC in LPG Users Household (μgm^{-3})	Outdoor (Middle of Villages) BC (μgm^{-3})	BC on Highways (μgm^{-3})	BC in Biomass Users Household (μgm^{-3})	BC in LPG Users Household (μgm^{-3})	Outdoor (Middle of Villages) BC (μgm^{-3})	BC on Highways (μgm^{-3})	BC in Biomass Users Household (μgm^{-3})	BC in LPG Users Household (μgm^{-3})	Outdoor (Middle of Villages) BC (μgm^{-3})	BC on Highways (μgm^{-3})
January	21.68	1.82	16.50	16.5	23.68	2.89	20.69	16.98	21.58	1.36	18.46	15.46
February	19.19	1.96	11.57	11.57	21.19	2.70	21.25	19.71	17.49	1.87	19.46	17.18
March	14.42	1.60	11.66	11.66	15.42	1.70	12.65	24.15	14.56	1.67	14.58	21.46
April	11.88	2.27	10.63	10.63	12.88	2.20	13.50	21.16	11.15	1.87	14.69	17.46
May	6.85	1.85	6.70	6.7	8.85	1.79	9.45	16.25	7.36	1.45	8.42	14.18
June	5.85	1.81	4.02	5.14	6.85	1.85	6.15	18.45	5.32	1.85	5.16	12.16
July	5.90	2.23	3.89	5.89	5.99	2.24	7.56	13.14	4.49	1.26	4.46	6.18

August	7.91	1.94	4.17	5.17	7.99	1.68	5.24	12.15	6.87	1.87	2.16	4.46
September	9.91	2.17	5.25	7.25	10.91	1.30	6.14	11.5	11.15	1.36	3.14	5.16
October	11.72	1.82	5.98	5.98	11.10	1.86	7.89	16.34	13.58	1.75	4.69	6.87
November	8.89	2.00	7.44	9.8	9.89	2.15	12.58	17.69	16.48	1.98	7.46	9.14
December	16.74	2.47	10.87	13.45	20.74	3.01	14.26	18.45	22.15	2.01	14.15	17.58

Table 4: Measured BC mass concentrations from various locations in the IGP

Location	**Sampling Period**	**BC in Biomass Users Household (μgm^{-3})**	**BC in LPG Users Household (μgm^{-3})**	**Outdoor BC (μgm^{-3})**	**References**
Sitapur	January-December, 2016	1.80 - 25.36	0.09-3.79	2.99-23.68	Present study
Patna		1.0 - 25.16	0.10-3.10	3.25-24.20	
Murshidabad		1.0 - 24.52	0.07-2.99	1.25-21.-35	
Patna	January- to December, 2015	-	-	21.86 ±3.48	Arif et al. 2018
Gorakhpur	2013-2015	-	-	19 ± 14	Vaishya, A. et al, 2016
Balia	June- to August, 2014	-	-	4.03	Tiwari et al, 2016
Pantnagar	2009–2012	-	-	4.8 ± 3.6	Joshi, H., et al, 2015
Varanasi	October 2008 to May 2009	-	-	2.2–19.6	Singh and Rai, 2014
Kanpur	Sept. to Nov., 2009,	60.0 (in morning)		30.0 (in morning)	Rehman et al, 2011
Delhi	January 2006 to January 2007	-	-	14.75	Bano et al., 2011
Agra	December, 2004	-	-	10.5–17.4	Safai et al., 2008

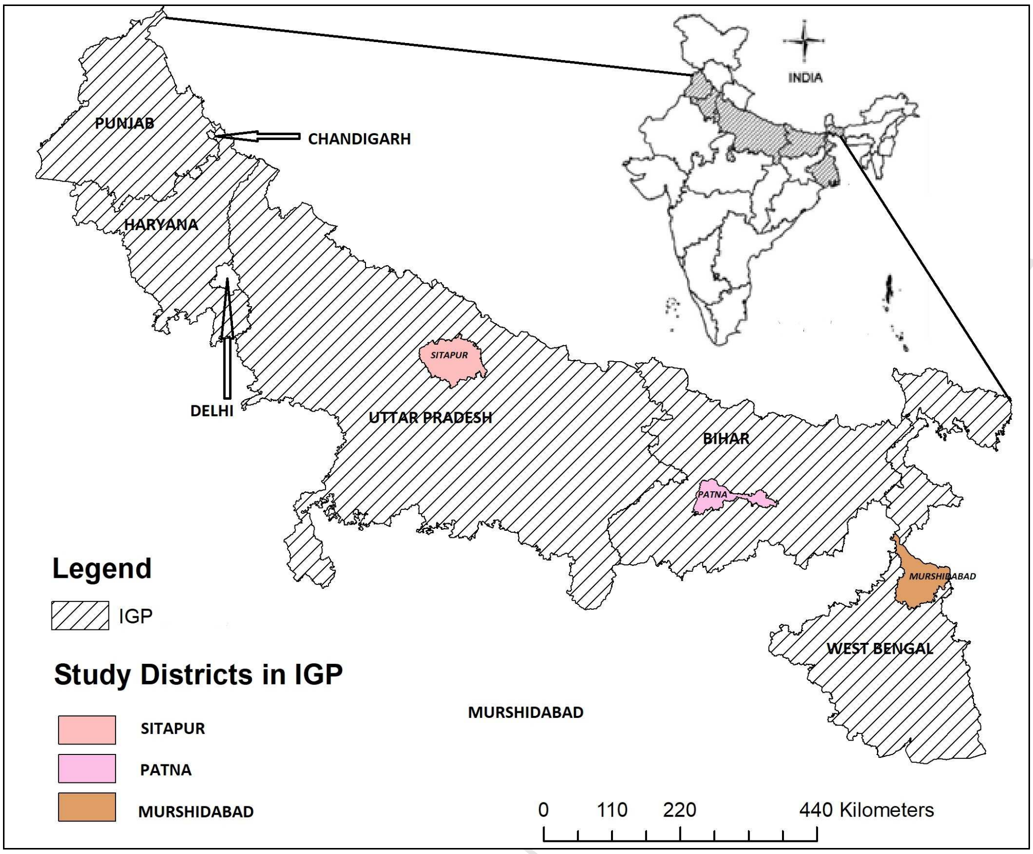

Figure 1: Black Carbon monitoring and survey locations in three districts of IGP (Indian Part)

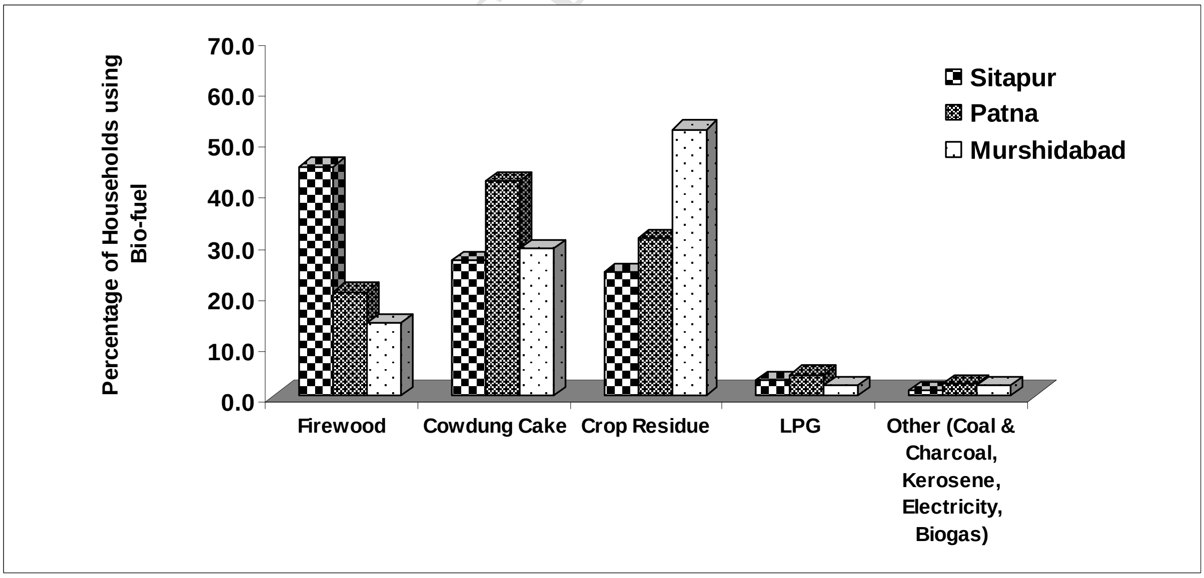

Figure 2: Different sources of biofuel used for cooking in Sitapur, Patna and Murshidabad (Primary Survey, 2015)

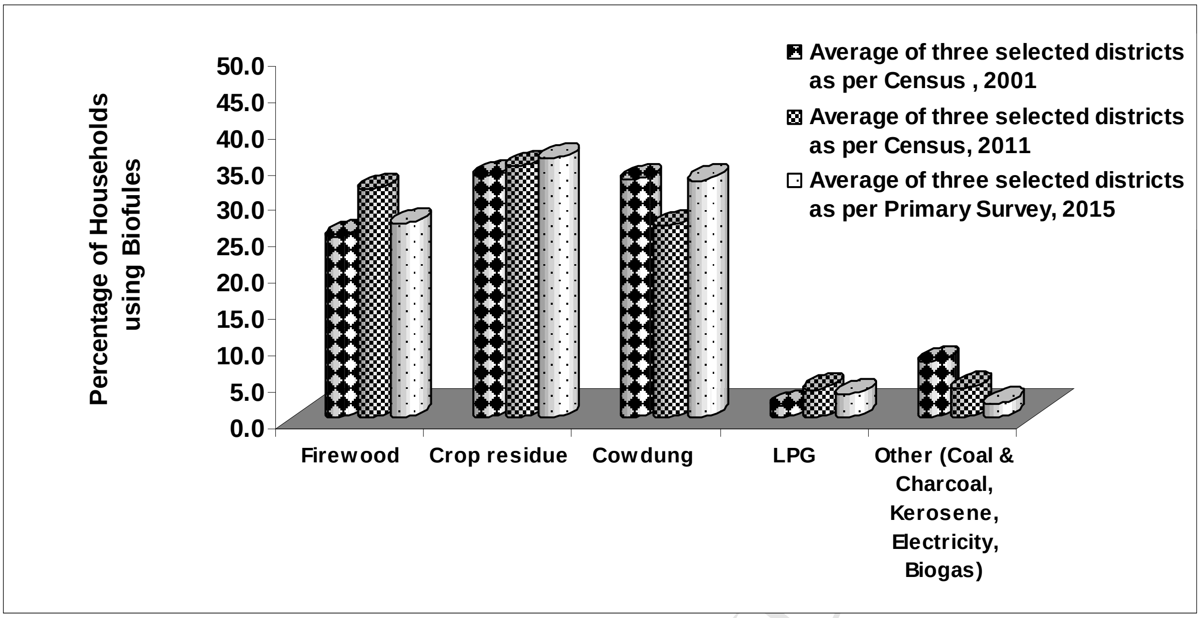

Figure 3: Variety of fuel used (in percentage) in Indian households as per the census and primary survey (2015) in representative districts of IGP

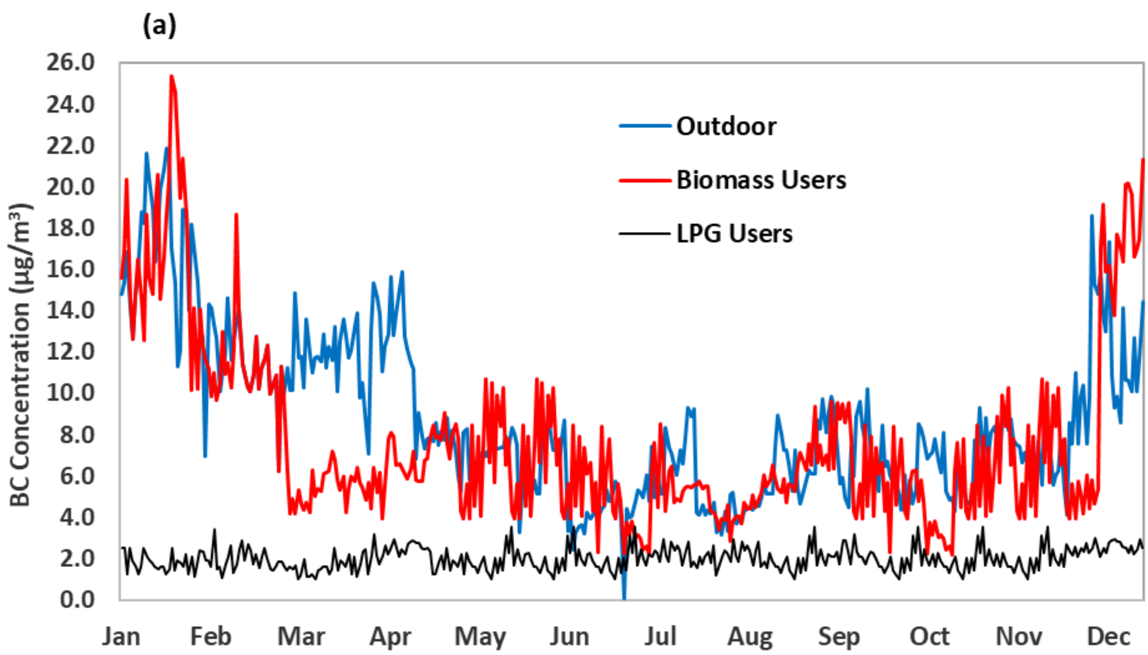

Figure 4a: The monthly average indoor (LPG & biomass users) and outdoor concentrations of BC

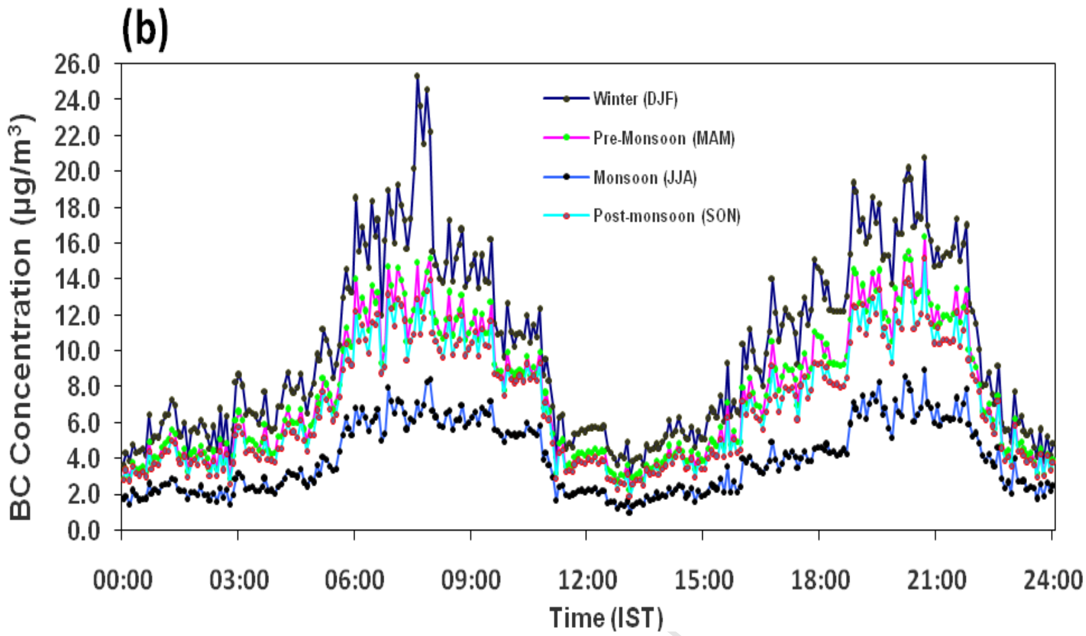

Figure 4b: Average daily seasonal indoor black carbon concentration in biofuel user's households during January to December, 2016

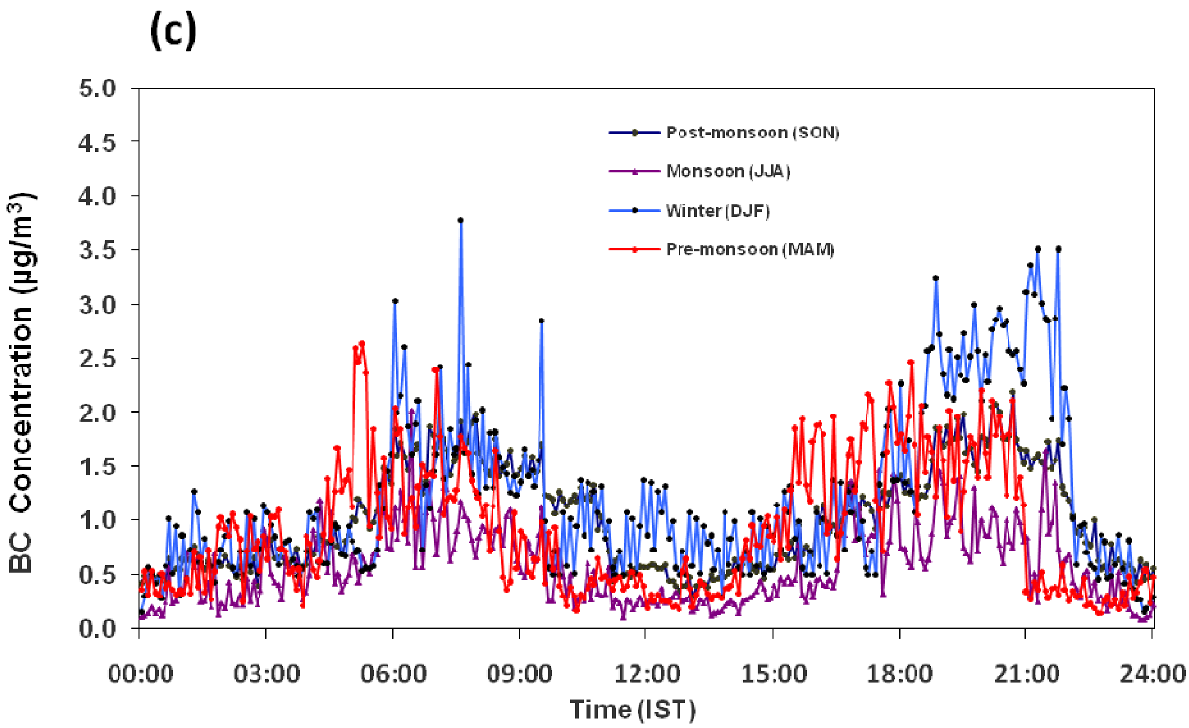

Figure 4c: Average daily seasonal indoor black carbon concentration in LPG user's households during January to December, 2016

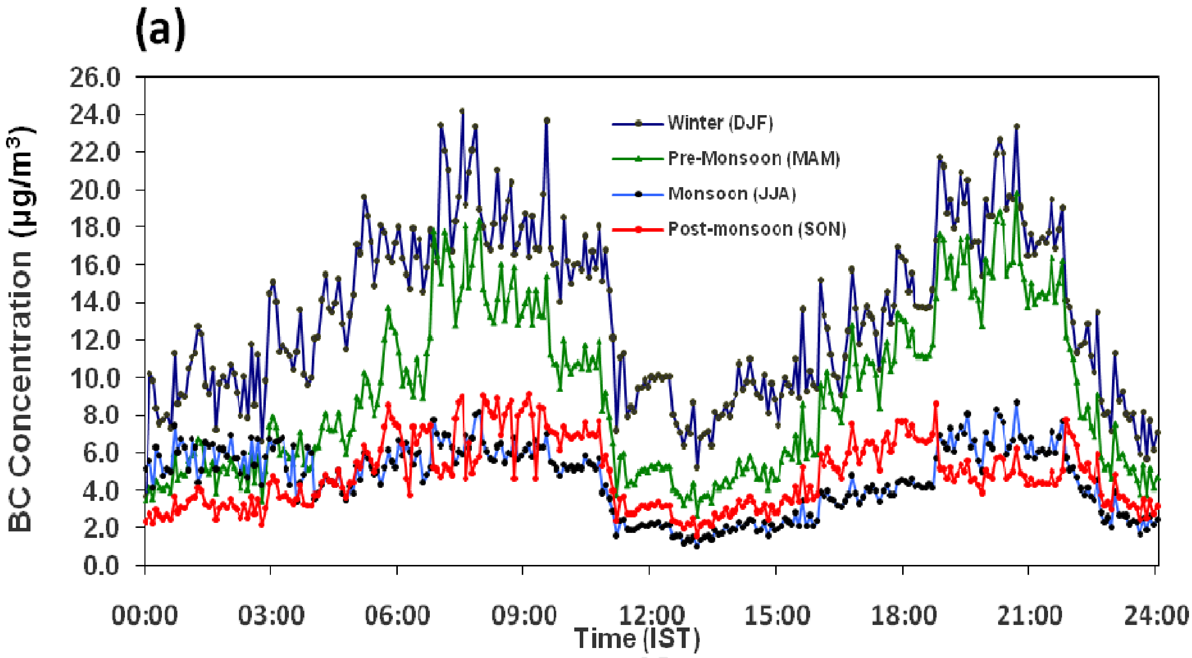

Figure 5a: Average daily seasonal outdoor black carbon concentration during January to December, 2016

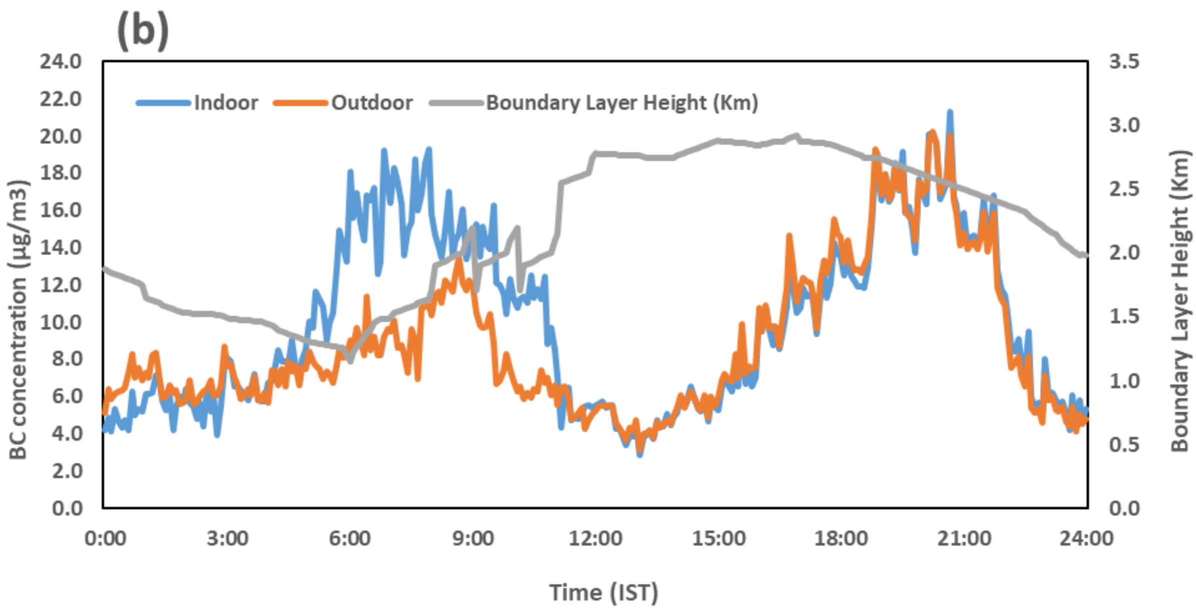

Figure 5b: Diurnal profiles of average indoor and outdoor BC concentrations during January to December, 2016

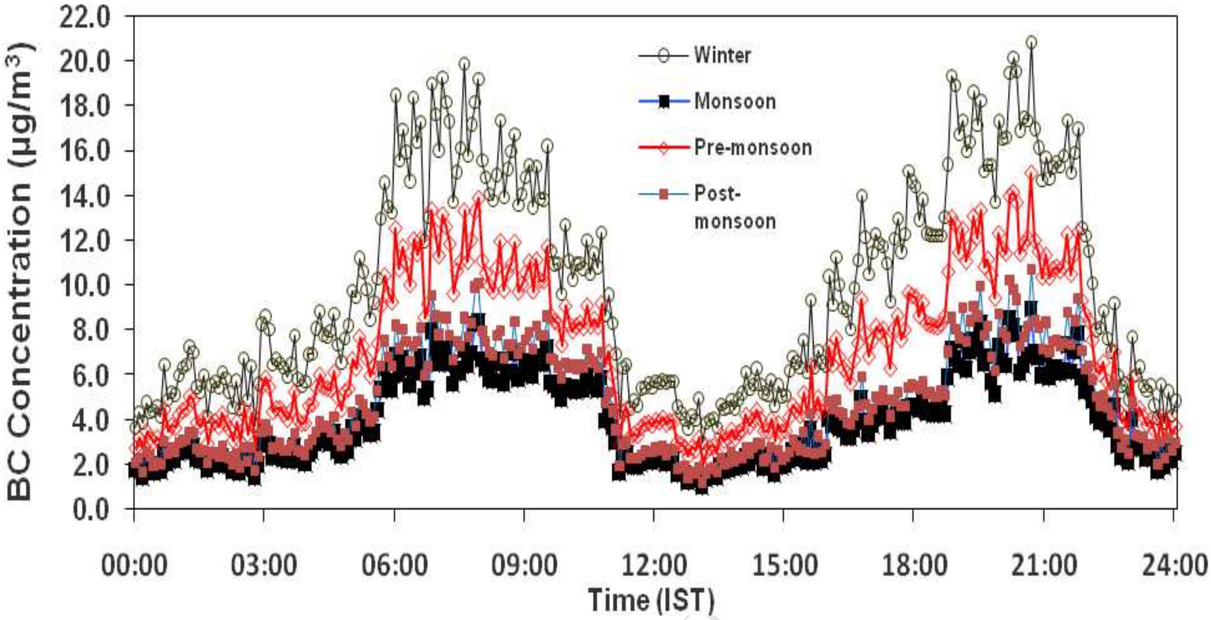

Figure 6: Seasonal averaged diurnal variation of BC concentrations at highways during January to December, 2016

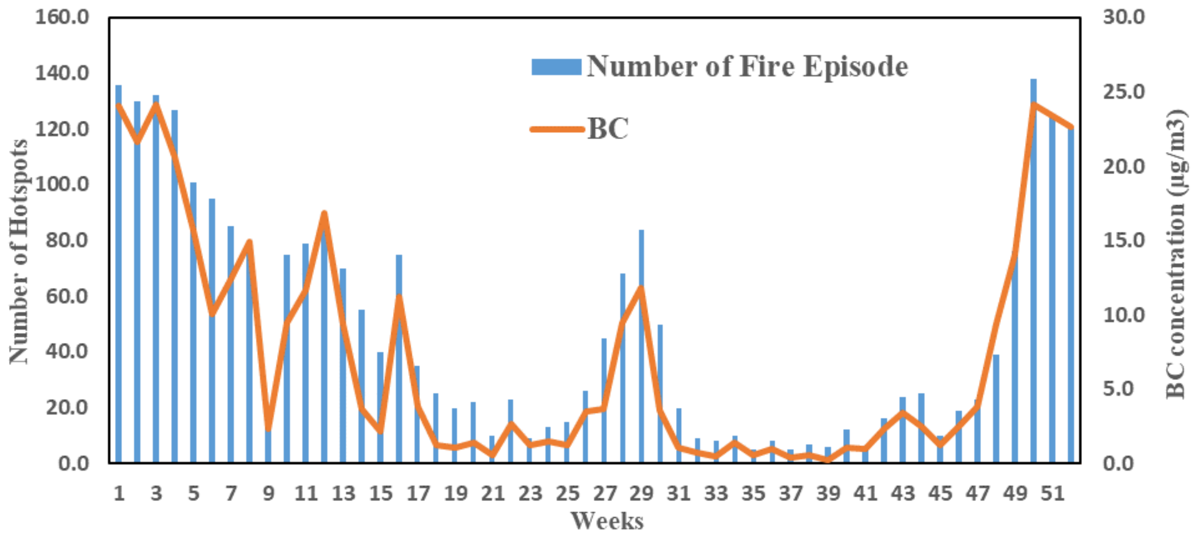

Figure 7a: weekly number of fire counts and average BC concentrations during January to December, 2016

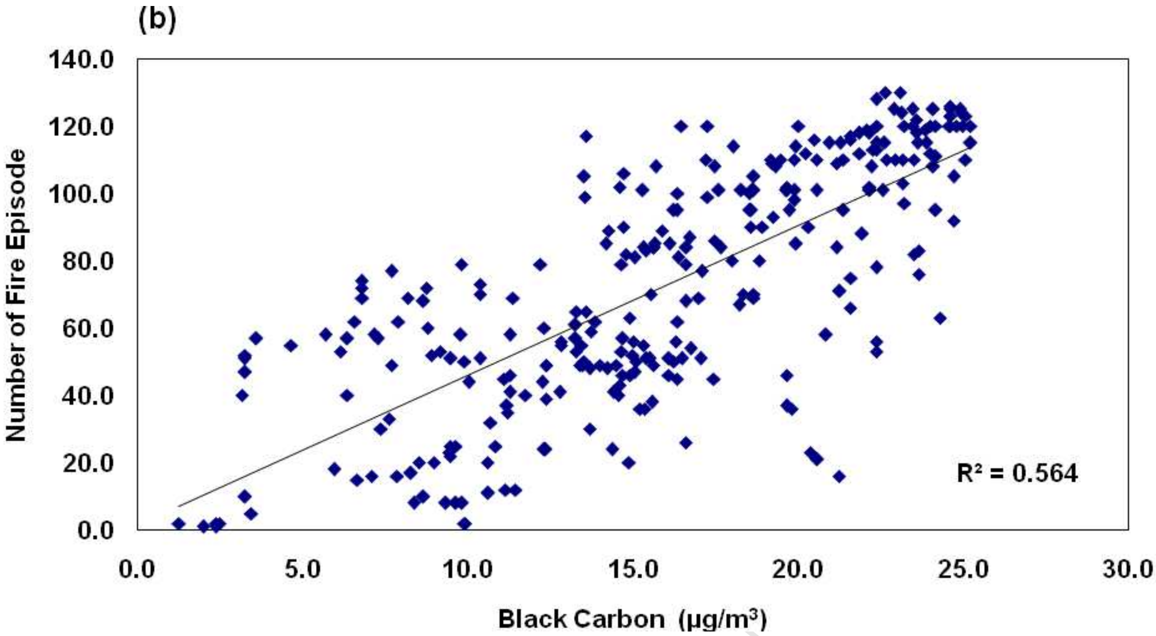

Figure 7b: A comparative plot of hotpots and daily average outdoor BC concentrations during January to December, 2016

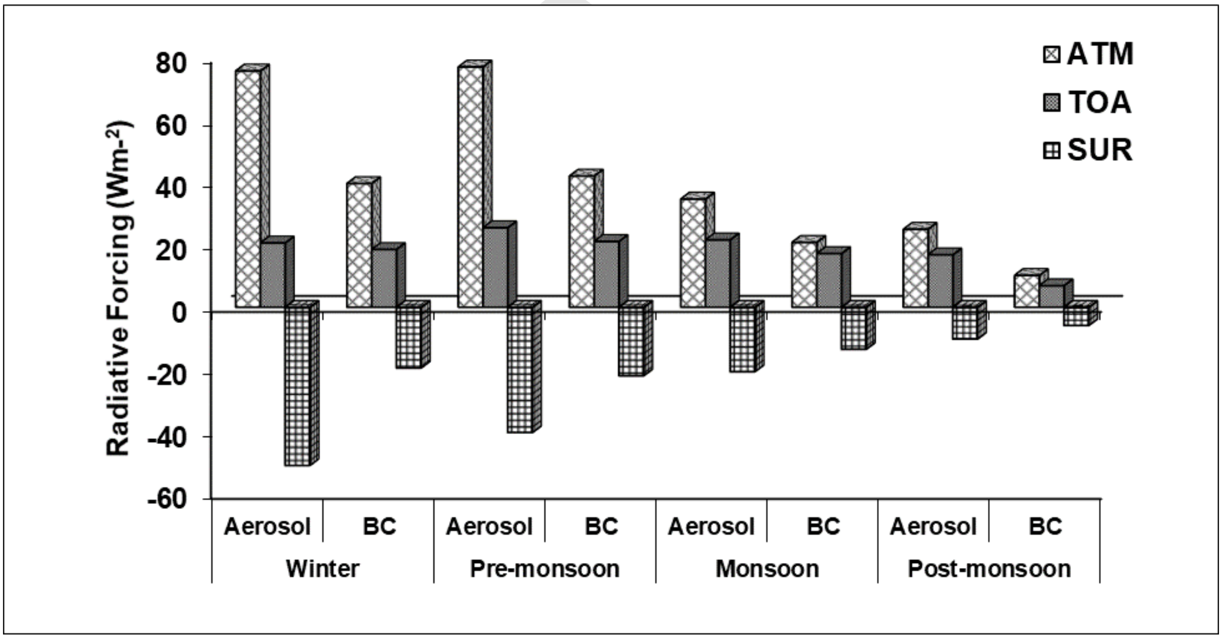

Figure 8: Composite aerosol and BC radiative forcing at the top of the atmosphere (TOA), surface (SUR) and atmosphere (ATM) over study area

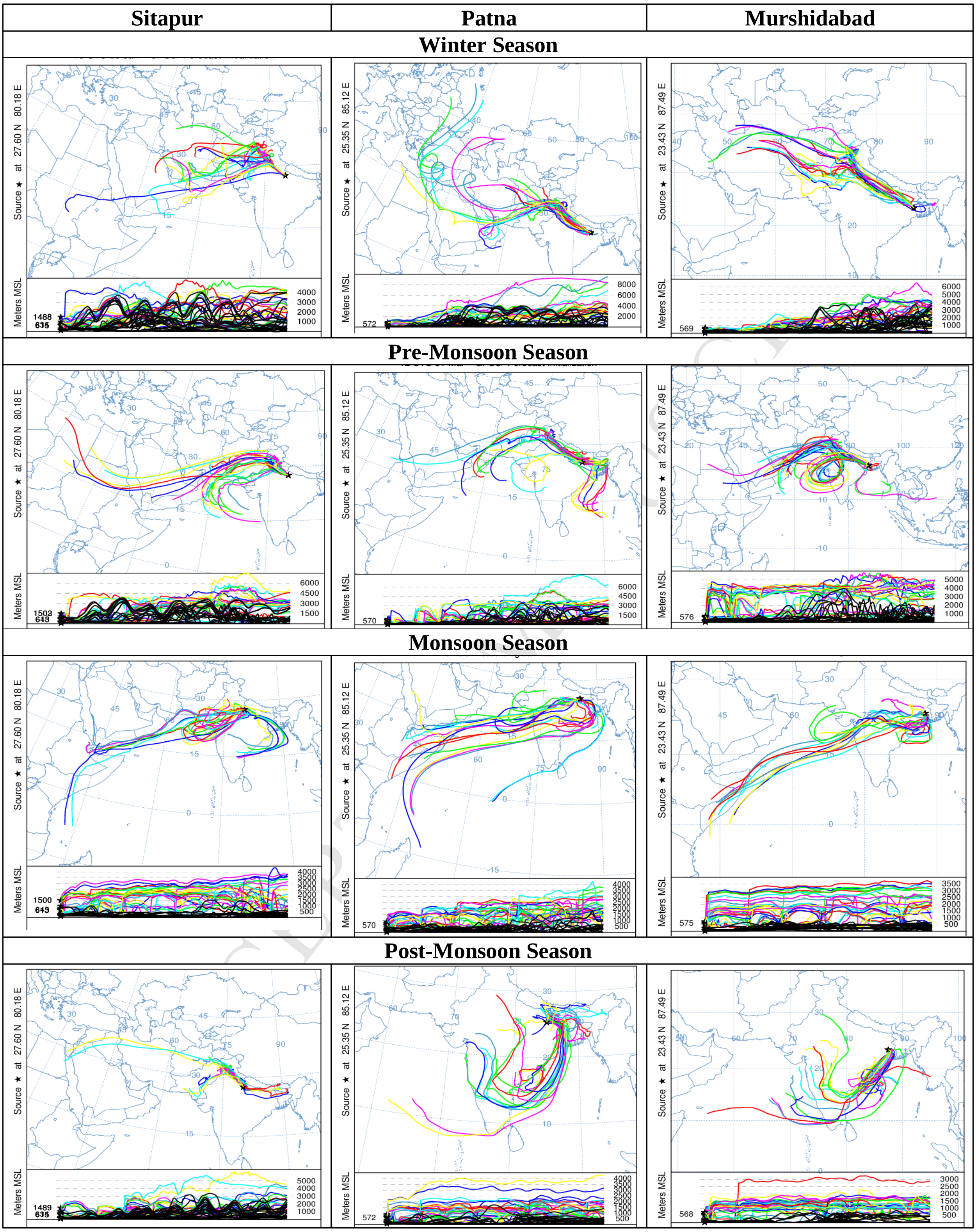

Figure 9a: 8-day's backward trajectories for four different seasons are arriving from northwest and west regions of IGP at Sitapur, Patna and Murshidabad which raise the surface BC concentrations level during the January-December, 2016

Highlights

- BC concentrations in LPG user's households was 90.3% lower than biofuels user's families.
- Diurnally the mass concentration of BC was highest (~35%) in the evening.
- Both ATM and TOA positive radiative forcing of BC and aerosol particles are showing a net warming effect on the study area while the SUR radiative forcing shows chilling effect.
- HYSPLIT modeling suggest that the smoke from biomass burning contribute significantly to air pollution levels in the cities.