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Black carbon emissions from biomass and fossil fuels in rural India

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Abstract. Black carbon (BC) emission from biofuel cooking in South Asia and its radiative forcing is a significant source of uncertainty for health and climate impact studies. Quantification of BC emissions in the published literature is either based on laboratory or remote field observations far away from the source. For the first time under Project *Surya*, we use field measurements taken simultaneously inside rural households, ambient air and vehicular emissions from highways in a rural area in the Indo-Gangetic-Plains region of India to establish the role of both solid biomass based cooking in traditional stoves and diesel vehicles in contributing to high BC and organic carbon (OC), and solar absorption. The major finding of this study is that BC concentrations during cooking hours, both indoors and outdoors, have anomalously large twice-daily peak concentrations reaching 60 $\mu g\,m^{-3}$ (median 15-min average value) for indoor and 30 $\mu g\,m^{-3}$ (median 15-min average value) for outdoor during the early morning (05:00 to 08:00) and early evening (17:00 to 19:00) hours coinciding with the morning and evening cooking hours. The BC during the non-cooking hours were also large, in the range of 2 to 30 $\mu g\,m^{-3}$. The peak indoor BC concentrations reached as high as 1000 $\mu g\,m^{-3}$. The large diurnal peaks seen in this study lead to the conclusion that satellite based aerosol studies that rely on once- daily daytime measurements may severely underestimate the BC loading of the atmosphere. The concentration of OC was a factor of 5 larger than BC and furthermore optical data show that absorbing brown carbon was a major component of the OC. The imprint of the cooking hour peaks were seen in the outdoor BC both in the village as well as in the highway. The results have significant implications for climate and epidemiological studies.

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1 Introduction

BC, a fine particulate matter, is a result of incomplete combustion of fossil fuels and biomass fuels. BC though short lived is also the strongest absorber of solar radiation in the atmosphere. It contributes significantly to global warming after long-lived greenhouse gases (Forster et al., 2007; Ramanathan and Carmichael, 2008; Jacobson, 2010). In addition, in South and East Asia, BC is estimated to contribute to the disruption of the monsoon in South Asia (Ramanathan et al., 2001, 2005; Lau et al., 2008) as well as East Asia (Menon et al., 2002) and heating of the elevated regions of the Himalayan-Tibetan region (Ramanathan et al., 2007; Flanner et al., 2009; Menon et al., 2010) thus potentially having a large impact on the food and water security of the region (see Lawrence and Lelieveld, 2010 for a detailed review). Due to its short life with a residential time of a maximum of two weeks as against CO_2 that can stay in the atmosphere for centuries, reduction in BC emissions has been increasingly proposed as one of the mitigation measures for limiting climate warming (Ramanathan and Carmichael, 2008; Grieshop et al., 2009; Kopp and Mauzerall, 2010).

About 3 billion human beings in developing countries, who subsist on a daily income of less than $2 a day (IEA, 2007) rely on solid biomass fuels for cooking and space heating, contributing to about 25 % of the global emissions of BC and about 50 % of the anthropogenic emissions of BC (Bond et al., 2007). In South Asia, BC emission from residential biofuels (wood, crop residue, dung) cooking is the largest source of atmospheric BC concentrations (Venkataraman et al., 2005). In India alone about 80 % of 160 million rural and 58 million urban households use solid biofuels (Venkataraman et al., 2010). Several studies have demonstrated the adverse impact on health from inhalation of fine particulate emissions containing BC from fossil and solid biomass fuels

(Pope et al., 1995; Ezzati and Kamen, 2002; Sauvain et al., 2003; Forastiere, 2004; Penn et al., 2005; Rom and Samet, 2006; Schwarze et al., 2006; Dockery and Stone, 2007). It is estimated that globally inhalation of smoke from indoor cooking using biomass fuels leads to 1.6 million deaths annually (WHO, 2002). In addition, the high levels of BC emissions from biomass fuels (Gustafsson et al., 2009; Venkataraman et al., 2005; Bond et al., 2007) can also significantly impact climate forcing from local to global scales (Forster et al., 2007; Ramanathan and Carmichael, 2008).

Indo-Gangetic Plains (IGP) region in South Asia is one of the most densely populated regions in the world and also a major source of BC emissions from cooking with biomass fuels. Large uncertainties exist in BC emission data from this region, which in turn induce large errors in estimating its radiative forcing. To reduce these uncertainties, as well as to document the role of biomass BC on health, regional climate change and Himalayan retreat, field data sets are needed. These data sets will improve the urgently needed ground measurements of BC for validation of climate and epidemiological models and will also help to guide the regulatory bodies to outline BC mitigation policies. Action taken to reduce or mitigate BC emissions from biomass fuels can save millions of lives, especially among women and children, and presents an opportunity to reduce warming on short-time scale (Grieshop et al., 2009). Thus, BC connects poverty and health with rural development and climate mitigation. Towards this goal, Project *Surya* was conceived as an international research effort and is being implemented in the IGP region in northern India (Ramanathan and Balakrishnan, 2007; Ramanathan et al., 2010).

Project Surya is a scientific intervention field experiment. Unlike earlier studies which reported BC concentrations in emission plume close to wood-burning cook stoves (e.g., Roden at al., 2006, 2009) or BC concentration from wood in laboratory conditions (e.g., Gonçalves et al., 2010), the aim of Project Surya is to simultaneously measure both indoor and outdoor BC concentration in rural village; first from traditional mud-based biomass burning cook stoves, and then after replacing them with more energy efficient cooking technologies, so as to document the impact of BC mitigation on health and climate.

The first (or pilot) phase of Surya was started in October of 2009 in a village located in the IGP region. The real-time simultaneous measurements of BC were recorded from mud stoves both indoor and outdoor and also at highway (∼3 km away) for baseline data collection. In the second (or demonstration) phase, the mud stoves were replaced with improved cook stoves and measurements were again recorded. This is the second in a series of 4 papers on the pilot phase study. The first paper (Ramanathan et al., 2011) deals with a cell-phone based BC monitoring system for large scale (e.g. 100–300 households) measurements. This is the second in this series documenting the village scale measurements with traditional mud stoves. The third paper (Praveen et al., 2011) explores the link between the local scales and regional scale radiative properties and forcing of BC. The last paper (Kar et al., 2011), reports on how replacement of the mud stoves with highly efficient cook stoves impact indoor BC concentrations, as well as compares various new cook-stove technologies on BC emissions.

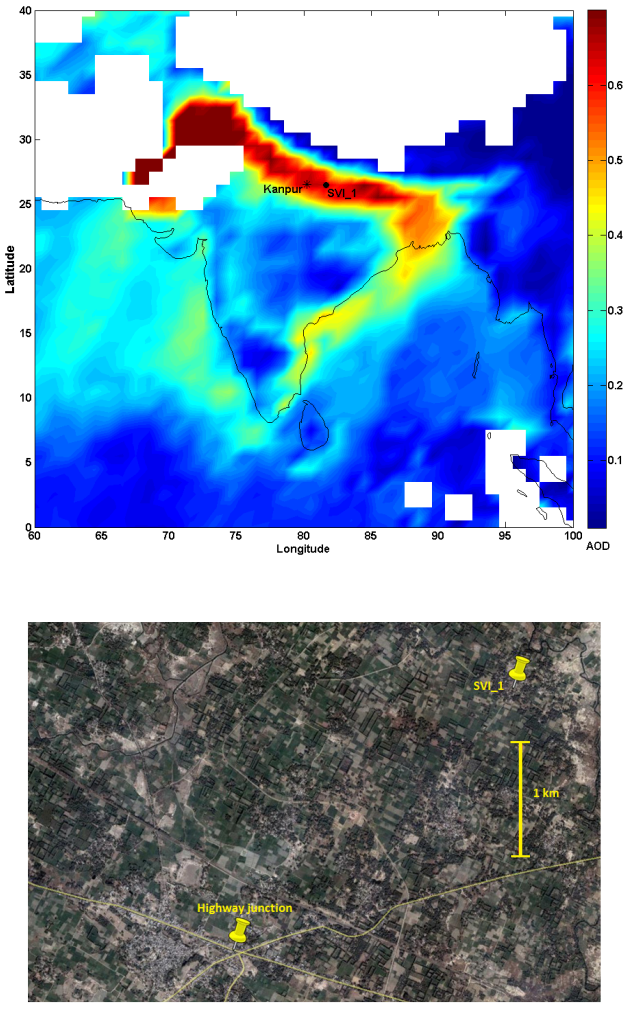

Fig. 1. (above) MODIS TERRA monthly mean aerosol optical depth (AOD) over India for November 2009. Also shown is our sampling site Surya village (SVI_1) located in the IGP region along with adjacent urban center Kanpur (26.30° N, 80.31° E). (below) Local map showing locations of SVI_1 and highway junction.

2 Experimental setup

2.1 Sampling location

Project Surya site is located in and around a village (26° N, 81° E) in the IGP region in northern India. Figure 1 shows the MODIS TERRA monthly mean aerosol optical depth (AOD) over India for November 2009 along with the location of our

sampling site. Clearly high AOD values can easily be seen over the IGP region. The Surya village (hereafter denoted as SVI_1) has around 485 households in seven close by hamlets that burn biomass fuel for their daily domestic needs of heating and cooking. For sampling we selected the largest hamlet that has around 200 households compared to other hamlets. The nearest traffic road is the Uttar Pradesh State Highway-15 (SH-15) located around 2 km to the south of the SVI_1 village. The nearest town center is located around 3 km southwest of the SVI_1 village, and where both SH-15 and National Highway-56 (NH-56) intersect (Fig. 1).

2.2 Indoor sampling

Concentrations of BC and elemental carbon (EC) were measured in the kitchen microenvironment close to mud cook stoves in the selected households. Indoor BC sampling was conducted in two periods, first between 27 September to 29 November 2009 in randomly selected 35 households, and again during 1–9 September 2010 in 18 of 35 households selected during the first period. Real-time BC concentration in indoor air was measured using a microAeth Model AE-51 (Magee Scientific, Berkeley, CA). AE-51 is a portable battery operated instrument, based on widely used Aethalometer technology (Hansen et al., 1984), and designed specifically to monitor personal exposure of BC. AE-51 draws ambient air on quartz filter based strip which then measures BC using single 880 nm LED. The length of the inlet tubing in AE-51 was 0.15 m and the instrument was placed such that it represents breathing position of cook stove emission, i.e. 1 m away and 0.6 m above the ground. The flow rate of AE-51 was set at 50 mL/min with measurement frequency of 1 min. Due to very high BC loading in cook stove emission plume; the filter strip of AE-51 was changed periodically to prevent attenuation saturation level from exceeding 120. From field test it was found that the AE-51 saturation level was reached in 30 min operating at 50 mL/min during cooking hours.

In addition to AE-51 BC measurement, we also collected 24 h indoor BC samples using our new cell-phone based BC monitoring system (BC_CBM) operating simultaneously with AE-51. The details of the BC_CBM system are described in Ramanathan et al. (2011), which is the first in series of 4 papers of this pilot phase study. Briefly, Miniaturized Aerosol filter Sampler (MAS) in BC_CBM system draws air at flow rate of 0.57 L/min and deposit particulate matter on 25 mm quartz filter (Pall Life Sciences). Some of these filters collected were analyzed for EC and organic carbon (OC) concentrations using thermal-optical EC/OC analyzer (Sunset Laboratory Inc., Forest Grove, OR) employing NIOSH TOT protocol (Schauer et al., 2003). For quality assurance, 25 mm quartz field blanks filters were also collected periodically using MAS during the sampling campaign and analyzed for EC/OC fraction using the same thermal-optical technique for blank filter correction. The term EC or BC is defined operationally in the literature and refers to the same light absorbing component (i.e. dark-colored "soot") in carbonaceous aerosols. Destructive analytical techniques (such as thermal-optical method) that utilize the thermal resistance, oxidative resistant and chemical inert nature of highly polymerized graphitic-like fraction of soot termed it as EC. Whereas non-destructive analytical techniques (such as optical method) that utilize the light absorbing characteristic of soot termed it as BC. The definition of BC includes EC as well as all components in the soot that absorbs light such as some OC fractions (known as brown carbon) that show strong absorption in near-UV region of solar spectrum. The readers are referred to Andreae and Gelencser (2006) for detail discussion on the definition of EC and BC. In this study the term EC will be used for measurement of soot by thermal-optical method (NIOSH TOT) and BC will be used for measurement of soot by optical method (AE-51) at 880-nm.

2.3 Outdoor sampling

Real-time outdoor BC concentrations were continuously measured from November 2009 using Aethalometer Model AE42 at two locations in the SVI_1 village; one at the center of the SVI_1 village, and in a less dense area on the northeast corner of the SVI_1 village. In addition, BC concentration was also measured at a highway traffic junction of SH-15 and NH-56 (3 km southeast of SVI_1 village) between 19 and 27 November 2009. Highway measurements were restricted to eight days to compare with the BC data from the SVI_1 village center. Details of outdoor measurements, data quality and analysis were given in the third paper of this series (Praveen et al., 2011). In this paper, the outdoor BC data from SVI_1 village center is compared with the indoor cook stove BC data only for the period of indoor sampling campaign. Households selected for indoor sampling were located within 100 m of SVI_1 village center.

3 Results

3.1 The large scale context of the observations

It has been demonstrated clearly (e.g., Ramana et al., 2004; Di Girolamo et al., 2004; Ramanathan and Ramana, 2005; see Lawrence and Lelieveld, 2010 for a review of the South Asian pollution problem) that the entire IGP region is subject to dense layer of atmospheric brown clouds (ABCs; consisting of BC, OC, sulfates and other aerosols) whose concentration varies with the season, reaching peak values during the dry season (November to March).

Beginning in April, dust becomes a major constituent and by July, the wet season (summer monsoon) begins and lasts until September. The monsoon rains are effective in removing the aerosols. This does not however imply that the aerosol pollution is negligible during the wet season, because aerosol emission at the surface (e.g., through cooking and transportation) continues throughout the year, and it does not

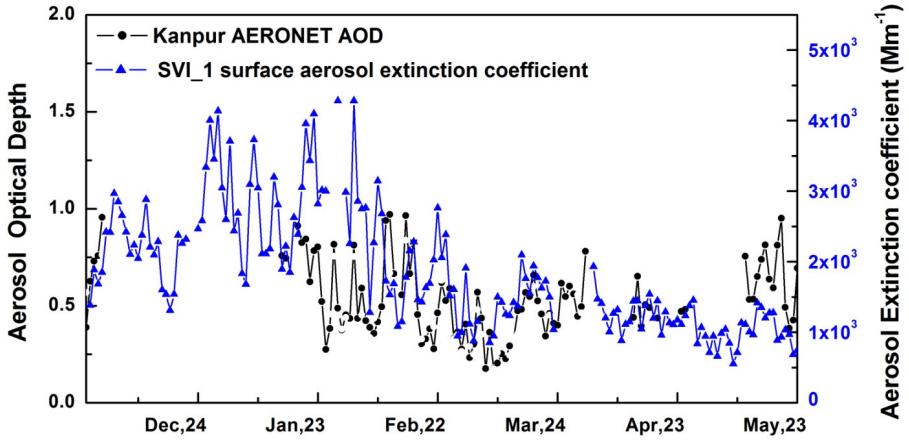

Fig. 2. Comparison of surface aerosol extinction coefficient at Surya village (SVI_1) with the column AOD measured by AERONET at neighboring urban center, Kanpur.

rain every day during the monsoon season. As a result, in between rainy days, the aerosol concentration builds up. For example, dust concentrations reach peak values during the monsoon season (Zhu et al., 2007) because dust transport into India reaches a peak during the wet season.

The primary data discussed in this study were collected during the dry season (November of 2009). The monthly mean satellite AOD shown in Fig. 1 for November 2009 clearly reveals near homogeneous layer of high AODs (>0.4) in the entire IGP region extending from Pakistan on the western side of IGP into Nepal and Bangladesh on the eastern side. Over 600 million people inhabit the IGP, with biomass cooking a major source for BC. Fossil fuel combustion (by the transport sector and local tractor emissions during agriculture activities, brick kilns and industry) is the second major source of BC. The experimental site at SVI_1 village is typical of the IGP region and the data collected in the SVI_1 site should be representative of the IGP. We are not implying that the SVI_1 data is identical to data over other regions of the IGP, but that, what we are observing at SVI_1 is part of the larger scale regional problem dealing with biomass cooking and fossil fuel combustion. This point is further illustrated in Fig. 2, where we compare the extinction coefficient, Ke, measured at SVI_1 with the column AOD measured by AERONET at a neighboring city, Kanpur (26.30° N, 80.31° E). Ke is the sum of absorption by BC and scattering by all aerosols including BC. The similarity between the monthly variation in Ke (a surface measurement) and the AOD (a column measurement) supports our claim that measurements at SVI_1 are part of the larger scale widespread ABCs. This issue is analyzed and described in greater detail in the third paper of this series (Praveen et al., 2011).

3.2 Link between indoor and outdoor BC concentrations

The 15-min median indoor (SVI_1 village kitchen) and ambient (outdoor at the SVI_1 village center) BC diurnal variation is shown in Fig. 3c for November 2009 (the dry season). The major qualitative features of the results in Fig. 3c are listed below:

1. The most striking feature of the data in Fig. 3c is the twice daily maximum in the BC concentrations. Focusing on the indoor first, peak values of about 60 $\mu g\,m^{-3}$ are reached between 05:00 and 08:00 in the morning and between 17:00 and 19:00 in the evening, closely matching the morning and evening cooking hours in the village. During the cooking hours, the outdoor BC values closely follow the diurnal peaks seen in the indoor values. We can understand this behavior by noting that the outdoor values are influenced by ventilation of cooking smoke from all of the homes of the entire SVI_1 village (and likely neighboring villages).

2. The slow exponential decay of the BC after the cooking hours is significantly different between the day light hours and the night values. The day time minima ($<10\,\mu g\,m^{-3}$) are much lower than the night time minima ($>10\,\mu g\,m^{-3}$) both indoors and outdoors. The most likely explanation is the fact that the boundary layer is deeper during mid-day than the night time values (when inversion sets in) – and as a result emission at the surface is ventilated to the free troposphere more efficiently during the day time. The role of the transportation (fossil fuel BC emission from trucks and other vehicles playing the highway several kms away) is discussed in the next section.

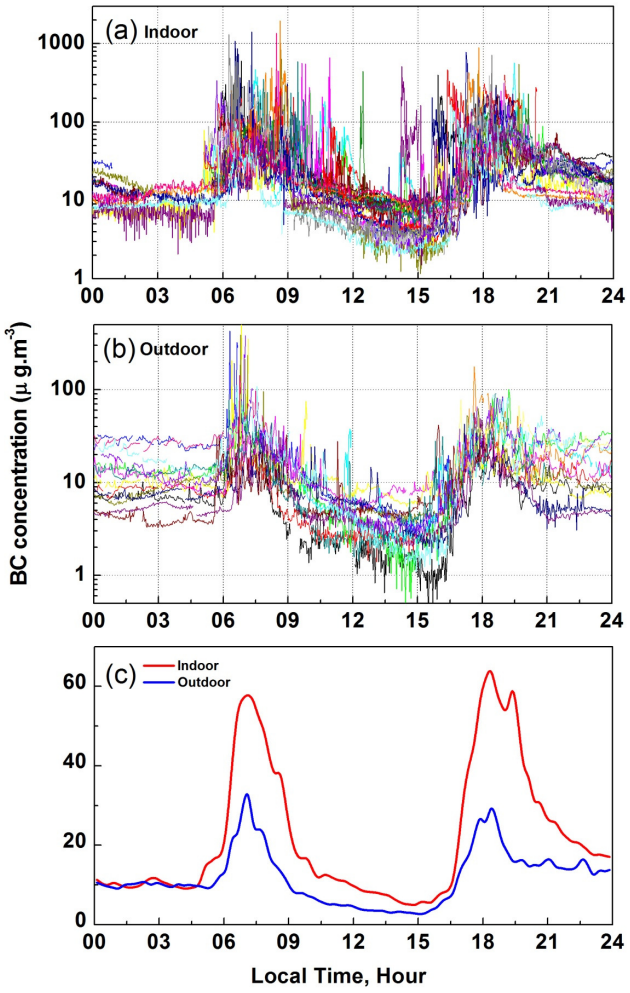

Fig. 3. Daily diurnal variation of BC concentration for the period 6-26 November 2009 in **(a)** indoor kitchen of 26 households in Surya village (SVI_1) and **(b)** at outdoor (ambient) SVI_1 village center **(c)** Comparison of 15-min median variation of BC concentration in indoor and outdoor for the same period at SVI_1 village.

3. It is also interesting to note that the non-cooking period BC indoors during the day time is larger than the outdoor BC, whereas it is smaller than the outdoor values from midnight to the beginning of cooking hour in the morning. This suggests to us that indoor cooking BC drives outdoor concentrations during the daytime until the cooking hours in the evenings, whereas, the outdoor BC (likely due to BC emissions from trucks and other vehicles) becomes more important during the night time hours.

Focusing on the quantitative values, Fig. 3a shows the indoor diurnal variation of BC concentration in 26 households. Out of 35 households sampled initially for indoor BC measurement, the data from 9 households were not included due to instrument malfunctioning or lack of battery power during the sampling period because of which diurnal variation for these households could not be recorded. During morning cooking hours (05:00 to 08:00) BC concentration varied from ∼3 to 1970 $\mu g\,m^{-3}$ with a mean value of $54\pm73\,\mu g\,m^{-3}$. Similarly, during evening cooking hours (17:00 to 19:00) BC concentration varied from ∼3 to 1070 $\mu g\,m^{-3}$ with a mean value of $62\pm61\,\mu g\,m^{-3}$. It is important to note that the mean values of BC concentration during cooking hours are derived from the data collected in 26 households where measurements were not recorded simultaneously, and therefore these values are representative of the mean exposure of SVI_1 households rather than any specific household. The large variation in the BC concentration observed during cooking hours could be attributed to the fact that that not all households cook at the same time; some households start cooking early around 05:00 while other start at a little later time. Similar reasoning also explains occasionally sharp peaks in BC concentration ($>100\,\mu g\,m^{-3}$) during the morning and evening cooking hours.

Incense burning for rituals and religious practices are common in Indian households, and could be a possible source of interference in our measurement (Jetter et al., 2002). However we did not observe any incense burning during our study period and therefore no correction was needed.

Figure 3b shows the daily diurnal variation of ambient BC concentration at SVI_1 village center. The data was recorded simultaneously with the indoor measurements. During morning cooking hour's outdoor BC concentration varied from 3 to 390 $\mu g\,m^{-3}$ with a mean value of $24\pm39\,\mu g\,m^{-3}$. Similarly, during evening cooking hour outdoor BC concentration varied from 3 to 180 $\mu g\,m^{-3}$ with a mean value of $26\pm18\,\mu g\,m^{-3}$. The ambient BC diurnal variation had similar trend to indoor BC mass concentrations with the peaks following closely the morning and evening cooking cycle suggesting strong influence of indoor cooking on outdoor BC concentration. Table 1 summarizes the mean BC concentration for indoor (kitchen) and outdoor (ambient) in Surya village (SVI_1) for November 2009.

3.3 Verification of the cooking link to BC

We used the twice daily maximum in indoor and outdoor BC to conclude that cooking was the major source for the observed diurnal variations. There was a unique opportunity to verify this conclusion, during the Ramadan religious festival (11 August to 12 September 2010) observed only by the Muslim religious community, when cooking patterns compared to other months are quite different. As the SVI_1 village population has dominant Muslim community, most of the household were observing fasting. Majority of the houses cooked the major morning meal before sunrise (03:00 to 05:00) instead of the usual 05:00 to 08:00 with in-between milk simmering during 06:00 to 07:00. The evening cooking was done before sunset (15:00 to 18.00), instead of the 17:00 to 20:00. The BC indoor measurements shown in

Table 1. Mean BC concentration for indoor (kitchen) and outdoor (ambient) in Surya village (SVI_1). The values in bracket represent the range of BC concentration.

	BC ($\mu g\,m^{-3}$)		Sampling period
	Morning cooking	Evening cooking	
Indoor	54±73 (~3–1970)	62±61 (~3–1070)	6–26 November, 2009
Outdoor	24±39 (~3–390)	26±18 (~3–180)	6–26 November, 2009

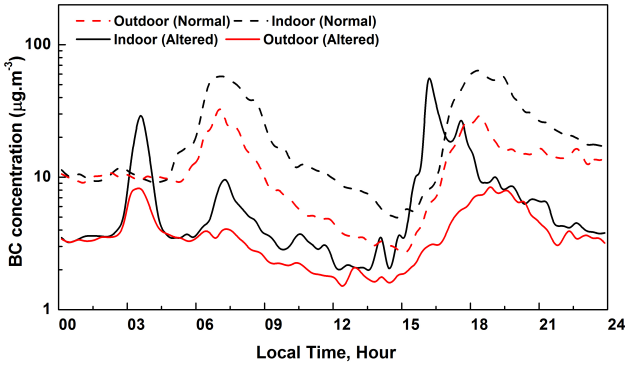

Fig. 4. 15-min median variation of BC concentration during altered cooking pattern during 1–9 September 2010 period for indoor (black solid line) and outdoor (red solid line) at Surya village (SVI_1). For comparison the 15-min median variation of BC concentration during normal cooking pattern for indoor (dotted black line) and outdoor (dotted red line) are also shown.

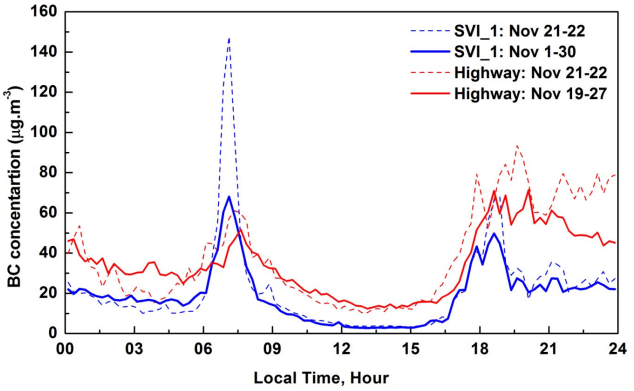

Fig. 5. Comparison of diurnal variation of BC concentration at Surya village (SVI_1) center with highway measurement (locates around 3 km to the southeast of SVI_1).

Fig. 4 clearly reveal the altered cooking pattern, with the major peaks during 03:00 to 05:00 (compared with the 05:00 to 08:00 in Fig. 3 for normal cooking pattern) and during 15:00 to 17:00 in the late afternoon hours, with a secondary peak during the milk simmering hours of 06:00 to 07:00. Ambient altered BC (shown as solid red line in Fig. 4), while it reveals the new peak during 03:00 to 05:00, is not similar to the ambient normal BC diurnal pattern shown in Fig. 3. We have to note that the large difference in transport between the dry season (when large scale transport is weak) data shown in Fig. 3 and the monsoon season (when large scale transport effects are dominant) data shown in Fig. 4 is one contributing factor. In summary, the Ramadan data for indoor BC supports the conclusion drawn from Fig. 3 that, diurnal variation and peaks in indoor and ambient BC mass concentration are primarily governed by the morning and evening cooking and can mainly attributed to burning solid biomass in inefficient biomass stoves, the dominant mode of cooking in SVI_1 village.

The ambient BC concentrations measured in SVI_1 village also compare well with the earlier studies involving field measurements of BC in IGP region. Beegum et al. (2009) reported mean BC mass concentrations of 27 $\mu g\,m^{-3}$ and 19 $\mu g\,m^{-3}$ for January and February 2006, respectively, during field observation in Delhi. Similarly, Tripathi et al. (2005) reported ambient BC mass concentration for the month of December ranging from 6 to 20 $\mu g\,m^{-3}$ over Kanpur city.

3.4 Relative importance of biomass and fossil-fuel combustion

The SVI_1 village was adjacent to a major national highway (NH-56) about 3 km southwest of SVI_1. We recorded BC measurements from 19–27 November 2009 at a traffic junction intersected by the highway. Traffic was dominated by diesel driven transport trucks and passenger buses. Figure 5 compares mean of 8-days of available data for highway with the SVI_1 village center data for November 2009. It also compares the two data sets for the 2 days (21–22 November) when simultaneous data were available for both the sites. We first note that the diurnal pattern of the difference between highway and SVI_1 for 21–22 November is similar to that for the longer period. This gives us confidence to interpret the longer-period averages in Fig. 5. The following key features are of importance for the present purposes:

1. The diurnal patterns are similar between the two sites, with the highway site peaks lagging behind the SVI_1 (village site) BC by about half hour. One possible explanation for the lag is that the sources for the peaks are the biomass cooking in neighboring villages. But, the

amplitude of the variations is much smaller in the highway site. Again, this suggests the villages as the main source for the strong diurnal variations.

2. The BC concentrations at the highway location during the non-cooking hours is a factor of 3 to 5 larger than the SVI_1 village center BC. The BC concentrations ranged from 20 to 50 $\mu g\,m^{-3}$ on the highway crossing whereas BC ranged from 3 to 15 $\mu g\,m^{-3}$ in the SVI_1 village. The significantly larger concentration in the highway site is suggestive of the importance of fossil fuel combustion source for the IGP region. Again, similar to the SVI_1 village observations, the day time minimum in the highway BC is much lower than the night time minimum. There are two factors that are contributing to this difference in the minimum BC. First is the diurnal variation in the boundary layer thickness. The second possible factor is the larger night-time traffic in trucks carrying cargo. We do not have sufficient traffic data to quantify the latter factor.

We next use two independent measurement techniques to examine the relative importance of biomass cooking and fossil fuel combustion in the observed indoor and outdoor BC in the village site.

3.4.1 Optical technique

Figure 6 compares the normalized light absorption (measured by AE42) as a function of wavelength for ambient aerosols from our two sites (SVI_1 and highway) with the previously published results from Kirchstetter et al. (2004). As shown by Kirchstetter et al. (2004) wavelength dependence of absorption is another way to understand the relative importance of biomass and fossil fuel contribution to BC. Thus far, we focused on BC. But this is not the only light absorbing aerosol produced by biomass or fossil fuel combustion. These combustion processes also produce organic aerosols, some of which strongly absorb in the near-UV region of the solar spectrum, and they are referred to as Brown Carbon (BrC) (Andreae and Gelencser, 2006). The BrC absorption increases with decreasing wavelength as approximately λ^{-2} from near infrared to UV region (Kirchstetter et al., 2004) and can go as high as approximately λ^{-7} in presence of humic-like substance in biomass aerosols (Hoffer et al., 2005; Chakrabarty et al., 2010). Whereas BC absorption increases as approximately λ^{-1} from near infrared to UV region. The Kirchstetter curves were obtained in a laboratory in California. Biomass burning produces significantly more (factor of 3 to 6 more) organic aerosols (and hence BrC) than fossil fuel combustion and as a result the spectral dependence of the absorption coefficient is much steeper than that of fossil fuel BC and OC (see the two Kirchstetter et al curves in Fig. 6). Figure 6 shows that for both locations absorption dependence on wavelength is in between biomass and fossil fuel curves. As expected, the SVI_1 village curve is slightly

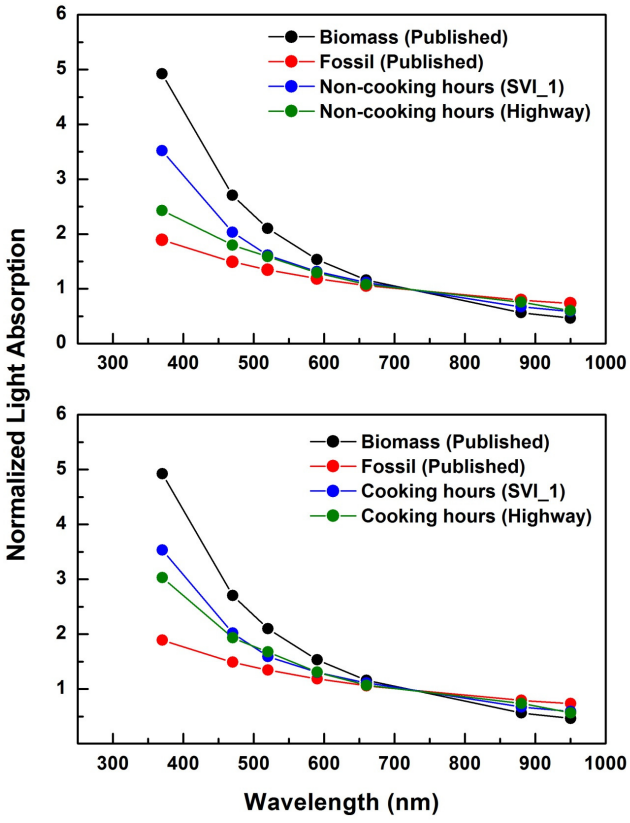

Fig. 6. Comparison of normalized light absorption in aerosol samples collected during cooking and non-cooking hours at Surya village (SVI_1) center and highway, with the previous published (Kirchstetter et al., 2004) study.

closer to the Kirchstetter biomass curve, whereas the highway curve is slightly closer to the fossil fuel curve. The data shown in Fig. 6 is yet another indication that both biomass cooking and fossil fuel contributes to the observed BC in both the SVI_1 village and the highway site.

3.4.2 Filter based mass analyses

During February-June 2010, 24 h samples were collected using BC_CBM system alternately in indoor (one selected house) and outdoor (at village center). Out of these, 30 samples from indoor and 27 samples from outdoor were analyzed for EC and OC (consisting of BrC and other organics). The sum of EC and OC is referred to as total carbon (TC). For quality control, 6 field blank (randomly selected) samples were also analyzed for EC and OC. No EC was detected in the field blanks; therefore no blank correction was needed. Figure 7 shows the OC/EC ratio as a function of EC concentration in indoor and outdoor samples. The OC/EC ratio in indoor samples varied from 2.9 to 8.4 (mean value of 5.3±1.6) after excluding two data points with ratio above 10; the corresponding EC concentration varied from 14 to

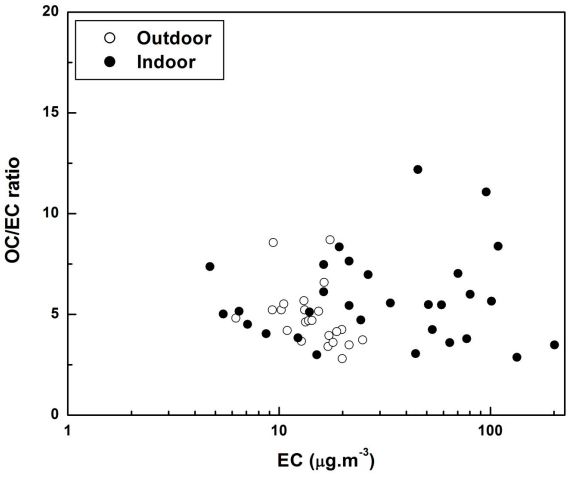

Fig. 7. Comparison between OC/EC ratio vs. EC concentration for indoor (SVI_1 village kitchen) and outdoor (SVI_1 village center) samples.

200 $\mu g\,m^{-3}$. Similarly, the OC/EC ratio in outdoor samples varied from 2.8 to 8.7 (mean value of 4.9±1.5) in close agreement with the indoor OC/EC ratio; the corresponding EC concentration varied from 6.3 to 25 $\mu g\,m^{-3}$. It is interesting to note that even though the OC/EC ratio in indoor samples is from one household, the similar ratio is found in the outdoor samples where concentrations are influenced by around 200 households, strongly suggesting that the outdoor OC/EC ratio are dominated by biomass fuel burning in indoor cook stoves.

The OC/EC ratio is characteristic of the type of fuel burned (higher for biomass fuel and lower for fossil fuel) and can be helpful in differentiating between combustion aerosol sources (Novakov et al., 2000). Typically mean OC/EC ratios for fossil fuel dominated aerosol in urban cities (range from about 1 to 3, whereas in regions dominated by biomass burning, the OC/EC ratio are about 8 (range from about 4 to 13) (Novakov et al., 2000). In our study the observed ratio of OC/EC ratio for both outdoor (ambient) and indoor (kitchen) represents aerosol from typical biomass sources. Roden at al. (2006, 2009) reported OC/EC ratio between 0.5 and 13 (mean value of 2.3) during field measurements from traditional Honduran wood-fired cookstove. The authors argued that the large amount of EC emitted during cook stove combustion may not be able to distinguish between fossil and biofuel combustion aerosol when cooking with biofuel is dominant. Schmidl et al. (2008) reported OC/EC ratio between 2.7–3.3 for hardwood and 2.6–5.7 for softwood typical burned in Austria and mid-European Alpine region for space heating during winter in domestic tiled stove. Gonçalves et al. (2010) reported OC/EC ratio from burning test in chimney-type woodstove on four common types of wood (three hardwoods and one softwood) prevalent in Portuguese. The authors found lower OC/EC in softwood (ratio of 0.9) compared to hardwood (ratio ranged between 1 and 4.4). The OC/EC ratio found in our study agrees very well with the ratio found in earlier studies. However, some of the differences is attributed to the type of fuel burned and cook stove design. We also compared the OC/BC ratio observed in this study with the gridded biomass burning BC and OC emission data (Lamarque et al., 2010) used in current climate models for the grid (26–27° N, 81–82° E). This grid includes our sampling location SVI_1. For the year 2005, the monthly gridded biomass burning OC/BC ratios in Lamarque et al. (2011) ranged from 5 to 10, which is in close agreement with the ratio observed in this study.

4 Implications

4.1 Air quality and health

The WHO recommended level for 24 h PM exposure is 25 $\mu g\,m^{-3}$. The daily BC concentration from mud stoves alone exceeded this level by factor of ∼5. Furthermore the concentration of OC was found to be factor of ∼5 higher than BC both indoor and outdoor. Thus the concentration of TC alone exceeded WHO recommended level by a factor of ∼25. Inhalation of such high concentration of PM can significantly impact the health of females (and young children) who spend most of their cooking time sitting next to mud cook stove.

4.2 Climate implications

The large regional and global climate effects of the South Asian ABCs were highlighted with data collected over the North Indian Ocean. The near-surface BC concentrations over the North Indian ocean were typically of the order of 0.5 to 1 $\mu g\,m^{-3}$. This should be contrasted with the concentrations of 10 to >100 $\mu g\,m^{-3}$ found in this study (Fig. 3) for the IGP. Furthermore, as shown in Fig. 2, the pollution persists throughout the year. The column AOD are in the range of 0.5 to 1.5 compared with peak AOD values of 0.3 over the North Indian Ocean (Ramanathan et al., 2007). Lastly, our data shows the single scattering albedo of the aerosols in the village are typically around 0.8 to 0.9, indicating highly absorbing aerosols (explained elsewhere in Praveen et al., 2011). Thus the atmospheric solar heating over the IGP by the biomass BC should be factor of 2 to 5 larger than that over the North Indian Ocean (see also Satheesh et al. (2002) for values over the Bay of Bengal). Such large solar heating by BC over the IGP, should have significant impacts on the monsoon system on meso-scales as well as regional scales. As discussed elsewhere (Ramanathan et al., 2007; Lau et al., 2008), the heating of the air by BC, would be transported to the elevated regions of the Himalayas, where it can amplify the greenhouse warming. The atmospheric heating by BC, will be accompanied by large dimming at the surface, which

can reduce crop productivity due to less sunlight for photosynthesis (Auffhammer et al., 2006) and perturb surface energy budget and the hydrological cycle.

Another important finding of climate relevance is the large increase in solar absorption in the visible wavelengths by the BrC, as shown in Fig. 6. This absorption will amplify the BC solar heating. The results obtained in this field study demonstrate the importance of biomass cooking related BC to regional climate. Lastly, most satellite observations of aerosols are collected during the day light hours (typically during 09:00 to 04:00); whereas our data shows the BC and OC reach peak concentrations during early morning (before 08:00) or late afternoon (after 17:00). It is likely satellite data of aerosols may have a large bias over regions where cooking dominates BC sources.

5 Conclusions

The fundamental insight gained from this field study is that cooking with solid biomass fuels is a major source of ambient BC over the IGP region. The second major finding is that peak values of BC are reached during early morning hours (05:00 to 08:00 hours) and early evening hours (17:00 to 19:00), and the peak values of about 100 $\mu g\,m^{-3}$ are a factor of 10 to 30 larger than the day time values. Indoor BC values during the peak hours are much larger than outdoor values. The OC concentrations are a factor of 5 larger than the BC concentrations. However, the OC had significant absorbing BrC, as revealed by the spectral absorption in wavelengths ranging from 350 nm to 600 nm. Even in the rural area sampled in this study, fossil fuel combustion from the transportation sector had a significant influence in the background BC concentrations during the non-cooking hours.

The results have significant implications to health impact studies as well as climate impact studies. On the epidemiological side, studies that rely on mean day time values will underestimate the cooking-hour exposure by factors ranging from 5 to 20. Likewise, climate impact studies that rely on satellite data or the ground based AERONET network will severely underestimate the effects of BC and other aerosols, since satellites and AERONET rely on day time data. With respects to regional climate impacts, the observed BC concentrations, the column AOD and the solar absorption efficiency of the aerosols are significantly larger than the values reported over the North Indian Ocean. This implies that the atmospheric solar heating as well as the surface dimming over the IGP region due to BC, OC and other particles from cook stove emissions should be larger by factors ranging from 2 or more. Clearly, our understanding of BC effects on monsoon and Himalayan glaciers needs to undergo a major revision with new modeling studies. Therefore, the climate forcing of BC when viewed with the already studied impacts on health of fine particulate matter emissions from biofuels necessitates the need for concerted effort to focus on finding clean energy solutions for rural household in conjunction with the measures to reduce emissions from fossil fuels particularly diesel driven vehicles. Considering the long-term actions required for mitigation of CO_2 and related pollutants the most effective strategy that would yield dividends both for environment and health in a short time span is to focus on short-lived carbonaceous pollutants such as BC. The strategy would be particularly relevant for developing countries where the reduction in short lived pollutants by introduction of efficient technologies for burning biomass for cooking is a development imperative. Given the fact that a large number of people using biomass based cook stove usually live below the poverty line it is also desirable to evolve methodologies that systematically capture the reduction in BC emissions and make it feasible for rural households to get financial benefits from the existing carbon economy. The process would facilitate and incentives the adoption of improved cooking devices in rural areas. Further the findings from this work clearly establish the need to focus on scientifically assessing the transport of black carbon emitted from biomass burning and diesel exhaust to glaciers and its potential warming impact that may lead to accelerated melting.

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