Residential Biomass Burning Emissions over Northwestern Himalayan Region of India: Chemical Characterization and Budget Estimation

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ABSTRACT

In the present study, we have determined the emission factors (EF) and estimated the emission of particulate matter (PM), organic carbon (OC), elemental carbon (EC), polycyclic aromatic hydrocarbons (PAHs), water soluble inorganic constituents (WSIC) and trace gases such as SO_2, NO and NO_2 from the combustion of biomass fuels (FW: fuel wood and DC: dung cake) used in rural sectors for cooking over Himachal Pradesh (HP), representing the Northwestern Himalayan region of India. The average EFs of PM estimated from FW and DC were 3.44 ± 2.38 and 11.43 ± 1.13 g kg^{-1}, respectively. OC and EC emission ranged from 0.106 to 3.55 g kg^{-1} and 0.07 to 0.90 g kg^{-1}, respectively for variety of biomass fuels. Total emission of PAHs from DC (44.37 mg kg^{-1}) and FW (43.25 mg kg^{-1}) noted in this study was almost similar. Similarly, the average EFs of NO_x from FW and DC were 0.59 ± 0.49 g kg^{-1} and 0.34 ± 0.18 g kg^{-1}, respectively. FWs have comparatively higher SO_2 emission (average: 0.43 ± 0.38 g kg^{-1}) than from DC (average: 0.23 ± 0.15 g kg^{-1}). Among anionic inorganic constituents emitted from FW, maximum EF was noted for Cl^- (0.30 ± 0.26 g kg^{-1}). Similarly for cations, highest EF was noted of K^+ (0.20 ± 0.09 g kg^{-1}). Ca^{2+} and Na^+ were the major cationic species identified in plumes of DC burning. Utilizing total annual consumption of biomass fuels and EFs of particulates and trace gases determined in the present study over HP and in the past study (Saud *et al.*, 2011, 2012) over Uttarakhand, budget estimates of PM, OC, EC, TC, PAHs, SO_2 and NO_x have been determined over the Northwestern Himalayan region. Total annual emission estimated over Northwestern Himalayan region are as: PM (18.32 ± 9.53 Gg), OC (4.38 ± 2.31 Gg), EC (1.39 ± 0.55 Gg) and trace gases (SO_2: 1.47 ± 1.0 Gg; NO_x: 1.77 ± 1.31 Gg).

Keywords: Particulate matter; Trace gases; Emission factor; Budget estimation; Northwestern Himalayan region.

INTRODUCTION

Rural areas of developing nations largely depend on unprocessed biomass fuels (fuel wood, dung cake and crop residues) for cooking and other household work. Use of such biomass in unvented stoves and poorly designed kitchens resulted in the emission of particulates and trace gases into the atmosphere (Bond *et al.*, 2004; Yang *et al.*, 2008), that greatly influence not only the local but the distant atmosphere as well by long range transport. Biomass burning emissions (including aerosols and trace gases) have been the focus of the study for many researchers as they not only influence Earth's radiation balance via scattering and absorption of incoming solar radiation, but also emissions like polycyclic aromatic hydrocarbons (PAHs) have a severe deteriorating

impact on human health (Davidson *et al.*, 2005; Ramanathan and Carmichael, 2008). Besides these direct effects, biomass burning emissions can also indirectly affect climate by modifying cloud microphysical properties (Novakov and Corrigan 1996; Markus *et al.*, 2009).

Particulates in the biomass burning smoke mainly composed of carbonaceous aerosols, consist of organic carbon (OC) which scatters most of the solar radiation resulting in a net negative radiative forcing and elemental carbon (EC) which absorbs solar radiation and is primarily responsible for net positive radiative forcing (Andreae *et al.*, 2004; Bond and Bergstrom, 2006; Ramanathan and Carmichael, 2008). Volatile organic compounds (VOCs) emitted during the combustion of biomass is converted into secondary organic aerosol (SOA). Sudheer *et al.* (2015), studied the formation of SOA over the semi-arid environment of Western India, the water soluble organic compound (WSOC) and SOA calculated by EC tracer method exhibit a strong linear correlation suggesting the considerable amount of SOA over the region. Similarly, Rastogi *et al.* (2015) studied the role of sources and meteorological conditions in SOA formation

over Indo Gangetic Plain, their study suggested the day time oxidation of VOCs to SOA from the oxidants like OH^-, O_3, whereas night time oxidation is governed by NO_3^- radical. Study by Rastogi *et al.* (2015) also revealed that the contribution of biomass burning emissions to carbonaceous aerosols is relatively more than those from fossil fuel burning. Certain types of carbonaceous aerosols such as volatile organic compounds (VOCs) and PAHs are harmful to human health (Nel, 2005). PAHs are a class of organic compounds that are semi volatile in nature and contain two or more aromatic rings. They are highly carcinogenic, especially benzo[a]pyrene, which can cause immune suppression in both animals and humans (Hall, 1989). Besides carbonaceous aerosols, water soluble inorganic cations and anions like ammonium chloride and potassium chloride (Alves *et al.*, 2010) act as cloud condensation nuclei, thereby causing negative radiative forcing (Cachier and Ducret 1991; Penner *et al.*, 1992). Alongside of particulates, trace gases like SO_2 and NO_x (NO + NO_2) are also emitted from biomass combustion which generally depends on the burning properties of the biomass fuel (pyrolysis, flaming and smoldering). SO_2 and NO_x are primarily the sources of secondary aerosol (sulphate and nitrate) in the atmosphere which are formed by gas to particle conversion (Xu and Penner, 2012; Sharma *et al.*, 2014a). SO_2 and NO_x owing to its gas to particle conversion under optimum conditions have been studied widely for different categories of biomass (Reddy and Venkataraman, 2002; Gadi *et al.*, 2003; Saud *et al.*, 2011).

Kuniyal *et al.* (2015) studied the particulate concentration in the Kullu valley (Himalayan region) and reported that coal, wood and straw burning could be the reason for the particulate concentration over the region. The major source of fluoride and potassium in the Himalayan region is possibly due to burning of wood and straw. Sharma *et al.* (2014b) studied $PM_{2.5}$ and gaseous pollutants (NH_3, NO, NO_2, and SO_2) over a wide area of the northwestern Himalayan region (Palampur, Kullu, Shimla, Solan and Nahan) of India, study reveals that the region is mainly influenced by the local activities, i.e., tourism activities, agricultural activities, biomass burning and vehicular emission. These pollutants along with the valley winds gets transported to the even higher altitudes that eventually coincides with glaciated terrain, results in increasing melting rates (Mayewski and Jeschke, 1979; Gautam *et al.*, 2009b).

Quantification of different chemical species from the combustion sources is imperative at the regional as well as on global scale to successfully perceive its atmospheric impacts. To estimate the emissions of aerosol and trace gases from biomass combustion, so far, we have calculated the emission factor (EF) (amount of target species emitted per amount of dry fuel consumed in g kg^{-1}) for the different chemical species over Indo Gangetic Plain (IGP), India (Saud *et al.*, 2011, 2013), a region characterized by high agricultural activities with a variety of crop production and cattle proportion involved in dairy production. In another study, Sen *et al.* (2014) from our group reported the emission estimate of particulates and trace gases emitted during the burning of biomass collected from the western India, region with great geographical variability ranging from the

arid deserts of Rajasthan to the coastal areas of Gujarat and Maharashtra of India.

Probably for the first time present study has been taken to estimate the emission of PM, OC, EC, PAHs, WSIC, SO_2 and NO_x from the combustion of FWs and DC, collected from the rural areas of Himachal Pradesh. Using EF_s values of particulates and trace gases from our earlier study (Saud *et al.*, 2011, 2012; Singh *et al.*, 2013) over Uttarakhand along with the EF_s values estimated in the present study for the above mentioned chemical species over Himachal Pradesh, we have also estimated the total emission of the aforesaid chemical species over the Northwestern Himalayan region of India.

METHODOLOGY

Description of Sampling Site and Collection of Biomass

Himachal Pradesh (HP) is a state of northern India situated in the North western region of Himalayas between 30°12'40"–33° 12'4"N latitudes and 75°47'55"–79°04'22"E longitudes. It is bordered by Jammu and Kashmir in the north, Punjab on the east, Haryana on the South, Uttarakhand on the Southeast and Tibet on the East. HP is the least urbanized state with 90% population living in the rural areas and widely depends on biomass fuels as a source of energy. Biomass samples comprising of fuel wood (all types of woods used as an energy source) and (ii) dung cakes (faeces of domestic animals) were collected from 20 different rural sites of HP covering almost entire residential part of HP (see Fig. 1 and Table S1). Biomass sampling sites were selected considering the fact that these sites (i) represent villages with population >1000 (ii) 50–70% of the inhabitants of the village dependent on biomass as their primary source of energy to meet their household requirements, (iii) the two sites from each districts must be away from the district head quarters by 10–15 km. A total of fifty-six samples comprising of 28 different types of FW (Table S2) and 03 samples of DC were collected and burned in the biomass burning setup as discussed in the subsequent section.

Biomass Burning Set-up

Biomass fuels collected were dried in a natural environment, the way the rural population using the biomass does (i.e., collecting the biomass fuels and then storing it in the sheds to be used in due course). The fuel woods collected were cut into smaller pieces of approximately 3–5 cm diameter and 15–20 cm length and burned in the biomass burning set up. Biomass burning set-up used for burning experiment was designed following the methodology of Venkataraman and Rao (2001). Detailed description regarding the biomass burning set up (Fig. S1) had been reported in our earlier paper by Saud *et al.* (2011). However, a brief description is mentioned in the supplementary material (S1). During the smoldering phase, generally high CO/CO_2 ratio is expected, whereas, the flaming phase reports the lower values of CO/CO_2 ratio. Impact of decrease in pressure of oxygen on these ratios hence on burning condition was studied by Philips *et al.* (1969) they found that the overall decrease in pressure of oxygen have not much effect on the
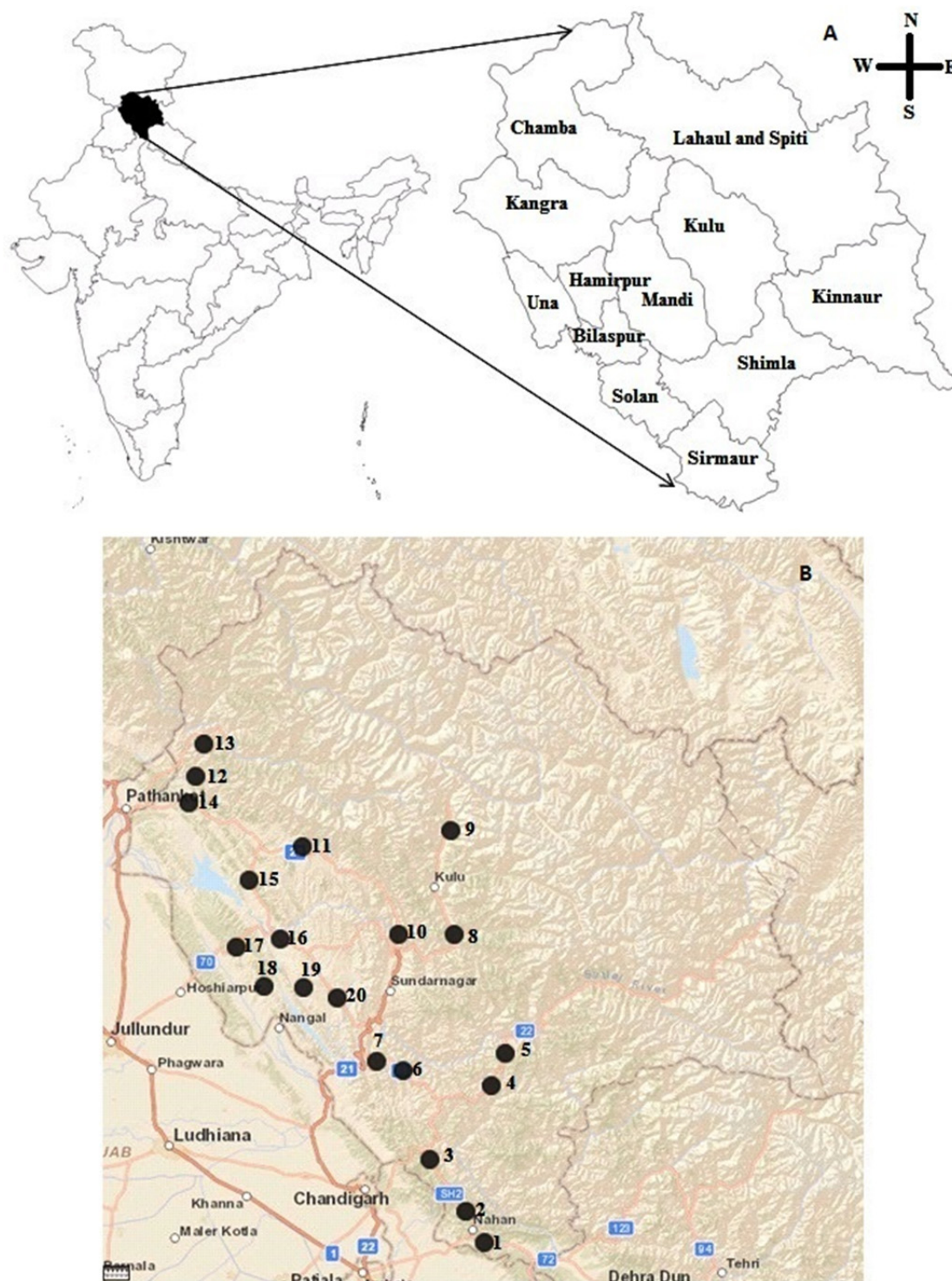
Fig. 1. Northwestern Himalayan region (HP) of India (A) and Sampling locations (B).

ratio of CO/CO_2 emission. Therefore in the present study influence of decrease in the partial pressure of oxygen in the atmosphere on burning of biomass fuel at higher altitude is not addressed.

Measurement of Particulate Matter (PM)

Prior to burning of biomass, quartz micro fiber filter (QM-A) papers were baked in a muffle furnace at 550°C

for 6 h to remove any traces of organic impurities and were kept in desiccators for 24 h to remove the moisture. Based on the mass difference of the filter paper before and after the burning, amount of PM emitted was determined with a micro balance (M/s. Sartorius, resolution ± 10 µg). The mass concentration was then calculated by dividing the amount deposited on QM-A filter with total volume passed during the burning of known amount of biomass sample.

Determination of OC and EC

OC and EC fraction of PM were analyzed on thermal optical OC/EC Analyzer (model: DRI 2001A; Make: Atmoslytic Inc., Calabasas, CA, USA) using IMPROVE TOR protocol (Chow *et al.*, 1993, 2001; Fung *et al.*, 2002). Detail description about the principle and working of the instrument has been reported in our earlier work (Saud *et al.*, 2012). Briefly, carbon fractions were heated at different temperature in pure helium to volatilized organic carbon from the sample deposit in a non oxidizing atmosphere and then in a 98% helium and 2% oxygen for the complete combustion of elemental carbon. Rau (1986) estimated the uncertainty of 4–6% in the measurements that constitutes a total of 13% uncertainty in OC and EC after considering the percentage error associated with burning system.

Analysis of PAH

QMA filter and polyurethane foam (PUF) plugs were used for the collection of particulate phase and gaseous phase PAHs, respectively. Prior to collection, QMA filters were baked at 550°C for 5 hr, similarly each PUF was cleaned three times (30min per cycle) with acetone using ultrasonication to minimize PAH blank. Details for the sample collection, storage and extraction can be noted from our earlier work (Singh *et al.*, 2013; Saxena *et al.*, 2014). Detail analysis of PAH using Gas Chromatography (GC) is described in supplementary material (S2).

Analysis of Water-Soluble Ionic Component (WSIC) and Trace Gases SO_2 and NO_X (NO and NO_2)

Technical details of the extraction process, sample preparation and analysis of WSIC have been discussed in Saud *et al.* (2013) and Sen *et al.* (2014). Similarly, trace gases emitted during the combustion process were estimated by the chemical wet method. Briefly, procedures for the analysis of WSIC and trace gases are described in supplementary material (S3 and S4).

RESULTS AND DISCUSSION

Distribution of Biomass Fuels Used in Rural Sector

According to rural energy data base of TERI (The Energy Research Institute) (TEDDY 2013) firewood used as the primary energy source for cooking in 70% of rural households in HP. Total consumption of different categories of biomass fuels (FW and DC) is nearly 2.1 Mt yr^{-1}. There is no published data about the consumption of individual biomass fuel in different areas of Himachal Pradesh. In present study percentage consumption of different FWs in rural areas for cooking and heating purpose has been determined by surveying several villages of Himachal Pradesh. 29 different types of biomass fuels (FW and DC) were collected from the 20 different rural sites of HP. It is important to mention here that each sampling site represents the rural area from where the varieties of biomass fuels were collected from different households. Fig. S2 clearly depicts that *Pinus sp.* and *Eucalyptus* were found at 20–30% of the rural sites from the 20 different rural sites. Similarly, use of *Cedrela sp.*, *Morus sp.* and *Cedrus sp.* for

cooking purpose were noted at around 15% of the rural sites while the other biomass fuels had ranged between 5–10%. Similarly, Fig. S3 illustrates the percentage variability in the types of biomass fuel used in the different districts of HP for households work. Rural households of Kangra and Hamirpur districts of HP were observed to have maximum variability (~20%) followed by Sirmur and Bilaspur (10–20%) and Kullu, Chamba, Una, Shimla, Mandi and Solan (> 10%).

Determination of Emission Factor

Experimentally determined EF (g kg^{-1}) of different chemical species from the combustion of FW and DC are described in details in the subsequent section. To assure statistical significance of data, burning experiments were done in duplicate for each biomass fuel collected from different mountainous region of Himachal Pradesh. Briefly, emission factor is defined as total mass of pollutant (M_x in g) emitted on burning of dry biomass (My in kg). Present study uses dilution method where the air gets well mixed in a definite ratio with the fuel extraction in the duct (Fig. S1). Method of obtaining M_x is proposed by Dhammapala *et al* (2006) as:

$$M_x = \int_{to}^{t} \Delta C_x Q_{duct}\, dt \tag{1}$$

$$EF_x \text{ (g kg}^{-1}) = M_x/M_y \tag{2}$$

where in Eq. (1), ΔC_x is the concentration of pollutant measured within the stack, Q_{duct} is the volumetric flow rate through the duct ($m^3\ s^{-1}$), t_0 is the time of ignition(s), and t is the time at the conclusion of smoldering, EF_x is the emission factor of pollutant x.

Particulate Matter (PM)

Table 1 summarizes the emission factor of PM for two types of biomass (FW and DC) collected from different sites of HP. Table 1 also, represents the emission estimates over Northwestern region of Himalayas (discussed in subsequent section) from the reported emission values of particulates over Uttarakhand and emission values estimated in the present study over HP. In the present study, particulate emission factor for DC was almost 3.3 times higher than of FW. Average emission factors of PM for FW and DC were 3.44 ± 2.38 and 11.43 ± 1.13 g kg^{-1}, respectively over HP. Among the FW collected, *Mallotus sp.* emits considerably higher PM (8.45 ± 6.99 g kg^{-1}) followed by *Gondla* (5.50 ± 4.33 g kg^{-1}) and *Malus sp.* (5.79 ± 3.42 g kg^{-1}). Spatial variation of emission factor of PM for biomass fuels was also observed among the districts of HP; FW collected from Kullu emits higher concentration of PM (6.50 ± 4.41 g kg^{-1}) followed by Solan (4.92 ± 2.23 g kg^{-1}) and Shimla (4.05 ± 1.86 g kg^{-1}). Spatial variation of emission factor of PM for widely used fuel woods such as *Pinus sp.* and *Eucalyptus sp.* were also studied. PM emission was highest from the *Pinus sp.* collected from Solan (4.92 g kg^{-1}) followed by Bilaspur (3.10 g kg^{-1}). Similarly, *Eucalyptus sp.* emits higher concentration of PM collected from the district Mandi (3.80 g kg^{-1}) followed by Chamba (2.08 g kg^{-1}).

Table 1. Average emission factor (g kg^{-1}) of PM, OC, EC, PAHs SO$_2$, NO and NO$_2$ from the burning of different biomass fuels used for domestic purpose in the rural areas of HP and Northwestern region of Himalayas.

a) EFs Particulates and carbonaceous (g kg^{-1})

States	FW				DC			
	PM	OC	EC	TC	PM	OC	EC	TC
Himachal Pradesh	3.44 ± 2.38	0.61 ± 0.44	0.31 ± 0.16	0.91 ± 0.60	11.43 ± 1.13	3.20 ± 0.34	0.26 ± 0.02	3.46 ± 0.36
Uttarakhand[a]	3.19 ± 1.68	0.92 ± 0.54	0.27 ± 0.08	1.19 ± 0.62	-	-	-	
Northwestern Himalayan region	3.31 ± 2.03	0.76 ± 0.49	0.29 ± 0.12	1.05 ± 0.61	11.43 ± 1.13	3.20 ± 0.34	0.26 ± 0.02	3.46 ± 0.36

b) EFs trace gases (g kg^{-1})

States	FW				DC			
	SO$_2$	NO	NO$_2$	NO$_x$	SO$_2$	NO	NO$_2$	NO$_x$
Himachal Pradesh	0.43 ± 0.38	0.40 ± 0.32	0.19 ± 0.17	0.59 ± 0.49	0.23 ± 0.15	0.23 ± 0.12	0.11 ± 0.06	0.34 ± 0.18
Uttarakhand[a]	0.17 ± 0.04	0.08 ± 0.05	0.07 ± 0.03	0.15 ± 0.08	-	-	-	
Northwestern Himalayan region	0.30 ± 0.21	0.24 ± 0.18	0.13 ± 0.10	0.37 ± 0.28	0.23 ± 0.15	0.23 ± 0.12	0.11 ± 0.06	0.34 ± 0.18

c) PAH EF (mg kg^{-1})

States	FW			DC		
	Gaseous	Particulate	Total	Gaseous	Particulate	Total
Himachal Pradesh	6.70	36.54	43.25	7.80	36.57	44.37
Uttarakhand[b]	11.6	53.7	65.3	-		
Northwestern Himalayan region	9.15	45.12	54.27	7.80	36.57	44.37

[a] Saud *et al.* (2011, 2012); [b] Singh *et al.* (2013).

On comparing the present results with our earlier studies over IGP, India (Saud *et al.*, 2011) and Western India (Sen *et al.*, 2014), it has been noted that the average EF of PM for FW in present study is almost comparable with the average EF from the IGP (4.34 ± 1.06 g kg^{-1}) but twice on comparing with the results of Western India (1.69 ± 0.98 g kg^{-1}). Similarly, emission factor of PM from DC in the present study is lower than IGP (16.26 ± 2.29 g kg^{-1}) but substantially higher from the DC of Western India (5.37 ± 3.90 g kg^{-1}). Determination of EF for FWs based on similar study conducted by Li *et al.* (2009) and Habib *et al.* (2008), were almost comparable with our present results, however the present EF was recorded ~2.5 times lower when compared with the study carried by Roden *et al.* (2006) (Table 2). Similarly, EF of PM from DC was found substantially higher than the EF reported by Habib *et al.* (2008). Earlier studies (Parashar *et al.*, 2005; Saud *et al.*, 2012; Sen *et al.*, 2014) concluded that the particulate EF of PM for DC is higher than that for FW (Table 2). Our results in the present study concurred with earlier results and may be attributed to the fact that DC normally burns under smoldering conditions.

Organic Carbon and Elemental Carbon (OC & EC)
Emission of carbonaceous aerosol (organic and elemental carbon) during the combustion process is highly dependent on the characteristics of biomass fuel. Biomass fuel with low burning efficiency led to the increased emission of carbonaceous aerosol (Saud *et al.*, 2012). Also, biomass fuels with high moisture when burns are responsible for higher contribution of OC to the particulates (Rajput *et al.*, 2014). EF of OC from FW (av. 0.60 ± 0.44 g kg^{-1}) was notably lower than EF of OC from DC (3.20 ± 0.34 g kg^{-1})

(Table 1). Among the FWs collected, *Mallotus sp.* (1.35 g kg^{-1}), *Tectona sp.* (1.25 g kg^{-1}) and *Malus sp.* (1.25 g kg^{-1}) were found to have higher OC emission. Similarly, EF of EC was higher for *Tectona sp.* (0.66 g kg^{-1}), *Strychnos sp.* (0.55 g kg^{-1}) and *Malus sp.* (0.54 g kg^{-1}). Average EF of EC from DC was noted lower (0.26 ± 0.02 g kg^{-1}) than the average EC emission from FW (0.31 ± 0.16 g kg^{-1}).

Contribution of OC to particulates was almost 1.6 times higher in the emissions from DC samples than from FW samples (Fig. S4). Possible reason of higher emission factor of OC could be the heat of combustion that was found lower (18.72 MJ kg^{-1}) for DC than FW (average 21.5 MJ kg^{-1}). Also, DC samples had higher moisture content than FW. As a consequence, contribution of EC emissions to the particulates was higher from FW (Fig. S4). The average EFs of OC and EC from the biomass fuels (FW and DC) showed spatial variation. Average EF of OC from the biomass fuels collected from the rural areas of Kullu (1.44 g kg^{-1}) and Hamirpur (1.03 g kg^{-1}) was observed to have higher values, however average EF of EC noted to vary from 0.18 to 0.72 g kg^{-1} with maximum value recorded in Kullu (0.72 ± 0.25g kg^{-1}). Spatial variability of OC and EC emission from the most commonly used biomass fuel (*Pinus sp.* and *Eucalyptus sp.*) was also studied. OC and EC emission factor from *Pinus sp.* were noted maximum from Solan (OC: 0.82 g kg^{-1} and EC: 0.41) and minimum from Sirmur (OC: 0.14 g kg^{-1} & EC: 0.09 g kg^{-1}). However, OC emission from *Eucalyptus sp.* collected from the district Mandi (0.51 g kg^{-1}) was highest and from Hamirpur (0.19 g kg^{-1}) was noted lowest. Similarly, EC emissions from *Pinus sp.* and *Eucalyptus sp.* were recorded maximum from Solan (0.41 g kg^{-1}) and Chamba (0.42 g kg^{-1}) and minimum from Sirmur (0.09 g kg^{-1}) and Bilaspur (0.15 g kg^{-1}), respectively.

Table 2. Comparative studies of emission factor for particulates and trace gases of FWs and DC.

Fuel type	PM	OC	EC	SO_2	NO_x	Location	Reference
Fuel wood	1.6 ± 0.98					Western India	Sen *et al.* (2014)
Fuel wood		0.95 ± 0.27	0.35 ± 0.07			IGP, India	Saud *et al.* (2012)
Fuel wood[F]	2.2 ± 1.2	0.62 ± 0.64	0.83 ± 0.69			China (rural)	Guofeng *et al.* (2012)
Fuel wood	4.34 ± 1.06			0.26 ± 0.10	0.76	IGP, India	Saud *et al.* (2011)
Various fuel wood				(0.1–1.17)	(0.8–8.1)	Southeastern US	McMeeking *et al.* (2009)
Fuel wood[F]	3.08 ± 0.82[*]	1.14 ± 0.40	1.49 ± 0.69			China	Li *et al.* (2009)
Fuel wood	3.2 ± 2.0		0.60 ± 0.15			South Asia	Habib *et al.* (2008)
Fuel wood[F]	8.5 ± 1.6	4.0 ± 0.9	1.5 ± 0.3			Honduras	Roden *et al.* (2006)
Fuel wood		4.4	1			India	Parashar *et al.* (2005)
Fuel wood		3.5 ± 1.9	1.1 ± 0.5			South Asia	Venkatraman *et al.* (2005)
Wood				0.7 ± 0.6		India	Gadi *et al.* (2003)
Fuel wood				0.48		India	Reddy *et al.* (2002)
Fuel wood				0.8	2.0	India	Garg *et al.* (2001)
Fuel wood	3.44 ± 2.38	0.60 ± 0.44	0.31 ± 0.16	0.43 ± 0.38	0.59 ± 0.49		Present Study
Dung cake	5.37 ± 3.90					Western India	Sen *et al.* (2014)
Dung cake		3.87 ± 1.09	0.49 ± 0.25			IGP, India	Saud *et al.* (2012)
Dung cake	16.26 ± 2.29			0.28 ± 0.09	0.58	IGP, India	Saud *et al.* (2011)
Dung cake	3.0 ± 1.9		0.12			South Asia	Habib *et al.* (2008)
Dung cake		12.6 ± 4.5	4.4 ± 2.2			India	Parashar *et al.* (2005)
Dung cake		0.25	(0.12–0.17)			South Asia	Venkatraman *et al.* (2005)
Dung cake				1.4 ± 0.9		India	Gadi *et al.* (2003)
Dung cake				0.84		India	Reddy *et al.* (2002)
Dung cake				0.6	0.86	India	Garg *et al.* (2001)
Dung cake	11.43 ± 1.13	3.20 ± 0.34	0.26 ± 0.02	0.23 ± 0.12	0.34 ± 0.18		Present Study

Variability observed in the emission factor of PM for the *Eucalyptus* and *Pinus* species collected from the different areas of HP could be due to the different geographical and climatic conditions in which they grow. Unlike *Eucalyptus sp* that normally found at the foot hills, *Pinus sp* grows at the higher altitude and therefore exposed to severe cold conditions. As illustrated above particulate and carbonaceous emission is minimum from the *Eucalyptus sp* collected from Hamirpur and maximum from the district Mandi. Geographical and climatic conditions of these two districts are different. Although, Hamirpur is situated at higher altitude (785 m, above sea level) therefore the winters are cold and summers are hot with adequate sunshine. Contrary, Mandi is situated on the bank of Beas river and has an elevation of 1044 m. Climatic conditions varies from hot sub humid tropical in the southern tract to the cold alpine and glacial in the northern and eastern mountain at the higher altitude. Comparatively, dry summers in Hamirpur and sub humid tropical conditions in Mandi and cold conditions at the higher altitude districts might be responsible for the variability in the emissions from the biomass fuels.

In the present study OC emission values from FW concur with the values reported by Guofeng *et al.* (2012), whereas OC and EC EFs recorded during field based measurements from the combustion of FW (Parashar *et al.*, 2005; Venkataraman *et al.*, 2005; Roden *et al.*, 2006; Li *et al.*, 2009) were higher than those reported in the present study. Similarly, OC and EC emission factor generated in the present study from DC were lower compared to those reported by Parashar *et al.* (2005) (Table 2).

Polycyclic Aromatic Hydrocarbons (PAHs)

Average emission factors of particulate phase and gaseous phase PAHs were determined experimentally following the protocol opted by Singh *et al.* (2013). Results from simulated burning experiments are given in Tables 1 and 3. Depending on the geographical area, a degree of natural variability in fuel types was noticed that has led to a range of emission factors. The average emission factor of total PAHs from DC (44.37 mg kg^{-1}) evaluated in this study was almost comparable with that of FW (43.25 mg kg^{-1}). In the case of FWs, maximum emission of total PAHs was noted from *Dalbergia sp.* (74.68 mg kg^{-1}) and minimum from *Magnolia sp.* (22.54 mg kg^{-1}). Verifying the profile of total PAHs (gaseous phase and particulate phase) in Fig. S5 from burning of FW and DC, it has been noted that emission of 3–4 rings PAHs (Ace, Acy, Flu, Anth, Phen, Pyr, Flt, Chry and BaA) was higher from FW (33.39 mg kg^{-1}) than from DC (28.0 mg kg^{-1}). Contrary, EFs for 5–6 ring PAHs (BkF, BbF, BaP, DahA, BghiP and IcdP) was higher from DC (16.37 mg kg^{-1}) than from FWs (9.86 mg kg^{-1}), most likely due to physical adsorption of these PAHs in particulate phase only. This observation resembles with the results reported by Singh *et al.* (2013) and Zou *et al.* (2003). In the present study, emission of particulate phase PAHs was found to be higher from DC than from FW, this could be due to the fact that DC burns mainly in smoldering phase. Also, during smoldering, the conditions become favorable for the condensation/adsorption of PAHs on suspended particles present in air and contributed an additional 70% of the total PAH load from DC and FW (Jenkins *et al.*, 1996).

Table 3. Average emission factor (mg kg^{-1}) of gaseous and particulate PAHs from the combustion of FW and DC over HP.

	FW (mg kg^{-1})			DC (mg kg^{-1})		
	Gaseous	Particulate	Total	Gaseous	Particulate	Total
Acy	2.61	-	2.61	-	-	-
Ace	2.89	-	2.89	2.74	-	2.74
Flu	0.66	-	0.66	4.18	-	4.18
Phen	0.55	0.92	1.47	0.88	1.18	2.06
Anth	-	4.73	4.73	-	1.70	1.70
Flt	-	3.94	3.94	-	5.09	5.09
Pyr	-	8.98	8.98	-	4.56	4.56
BaA	-	4.57	4.57	-	4.31	4.31
Chry	-	3.54	3.54	-	3.37	3.37
BbF	-	2.59	2.59	-	7.42	7.42
BkF	-	2.10	2.10	-	0.68	0.68
BaP	-	0.82	0.82	-	1.40	1.40
DahA	-	1.25	1.25	-	2.99	2.99
BghiP	-	2.74	2.74	-	3.45	3.45
IcdP	-	0.36	0.36	-	0.43	0.43
$\sum_{PAHs}$	6.71	36.54	43.25	7.80	36.57	44.37

Acenapthylene (Acy), Acenapthene (Ace), Fluorine (Flu), Phenanthrene (Phen), Anthracene (Anth), Fluoranthene (Fla), Pyrene (Pyr), Benzo (a) anthracene (BaA), Chrysene (Chy), Benzo (b) fluranthrene (BbF), Benzo(k)fluoranthene (BkF), Benzo (a) pyrene (BaP), Dibenzo (a, h) anthracene (DahA), Benzo (ghi) perylene (BghiP), Indeno (1,2,3-cd) pyrene (IcdP).

Normally, the extent of emission from biomass fuel depends on its calorific value (CV) i.e., the biomass fuel that has high burn rate leads to the lesser emission or reduction in the release of major products of incomplete combustion, vice versa of it causes significant loading of particulates (Singh *et al.*, 2013). Fuel burn rate (average: 0.52 ± 0.13 kg h^{-1}) observed in the present study for fuel woods ranges from 0.25 kg h^{-1} to 0.75 kg h^{-1}. Values noted in the present study were in the range when compared with the values reported by Gupta *et al.* (1998) and Singh *et al.* (2013). Fuel burn rate for DC (0.45 kg h^{-1}) recorded lower than the FW. Also, calorific values of biomass fuels are negatively correlated with the particulate emission. Calorific value of fuel woods noted higher (average: 21.50 MJ kg^{-1}) than the DC (average: 18.72 MJ kg^{-1}). This could be the reason for higher gaseous and particulate PAHs emission from the DC. Sen *et al.* (2014) also recorded the negatively correlation between EF and CV and observed that emission of particulates and trace gases are higher from residential biomass fuels with a lower CV across western India.

Emission factors of PAHs for variety of FWs were evaluated in past by different researchers globally (Table 4). Gadi *et al.* (2012), estimated emission factor of particulate phase PAHs from biomass fuels used in the rural areas of Delhi, India were almost consistent with the present study. Among all the toxic PAHs, listed by US EPA, BaP has the highest carcinogenic potency with long term persistency in the environment. In the present study value of BaP ranged from 0.48 to 2.22 mg kg^{-1} (average: 0.82 ± 0.67mg kg^{-1}) for FW. On comparing our mean BaP values with those reported by Singh *et al.* (2013) for IGP of India and Gupta *et al.* (1998) for India, Brazil, Mexico, Zimbabwe and USA, It has been noted that the values of BaP were in the range of the values previously reported for India, however the values of BaP were considerably at the higher side when compared with

Zimbabwe (0.77 mg kg^{-1}) and at lower side for Brazil (2.61 mg kg^{-1}), Mexico (2.14 mg kg^{-1}) and USA (2.25 mg kg^{-1}).

Water Soluble Inorganic Constituents (WSIC)

EFs of cations (Na$^+$, NH$_4^+$, K$^+$, Mg^{2+}, Ca^{2+}) and anions (F$^-$, Cl$^-$, SO$_4^{2-}$, NO$_3^-$, PO$_4^{3-}$) from FW and DC over HP are summarized in Table 5. Average EF estimated of K$^+$ (0.20 ± 0.09g kg^{-1}) and Na$^+$ (0.17 ± 0.10g kg^{-1}) were highest among the FWs. Similarly, Na$^+$ (0.058 ± 0.007g kg^{-1}) and Ca^{2+} (0.074 ± 0.003g kg^{-1}) were the dominant species in the emissions from DC. Among the FW samples, *Strychnos sp.* emits highest amount of K$^+$ (0.92g kg^{-1}) whereas emission from *Magnolia sp.* was found to be the least (0.036 g kg^{-1}). Among anionic species, average EF of Cl$^-$ (0.30 ± 0.26g kg^{-1}) was highest followed by SO$_4^{2-}$ (0.22 ± 0.19g kg^{-1}) and PO$_4^{3-}$ (0.13 ± 0.03g kg^{-1}) from FW, whereas, emission of Cl$^-$ (0.13 g kg^{-1}) and SO$_4^{2-}$ (0.14 g kg^{-1}) from DC were almost of same magnitude. Among the FWs, Cl$^-$ emission was maximum from *Dalbergia sp.* (0.70g kg^{-1}) and *Tectona sp.* (0.67 g kg^{-1}), whereas, SO$_4^{2-}$ EF was highest in the emissions from *Darindu* (0.52 g kg^{-1}) and *Dalbergia sp.* (0.62 g kg^{-1}). Cl$^-$ and NO$_3^-$ emission from FW was noted to exhibit significant positive correlation ($p < 0.05$) with Na$^+$, Mg^{2+} and Ca^{2+}. Similarly, SO$_4^{2-}$ emission was found to have significant positive correlation with Na$^+$ and Ca^{2+}, respectively (Table S3).

The spatial variability in the emission of individual anions and cations emanating from FW collected from the rural areas of Himachal Pradesh has been also studied. Highest emission of Na$^+$ was seen for FW from Solan (0.66 g kg^{-1}) and Sirmur (0.425 ± 0.169 g kg^{-1}). Similarly, high EF for NH$_4^+$ was noted for FW from Kangra (0.32 ± 0.28g kg^{-1}). K$^+$ emission was almost consistent from the FW of Kullu, Solan and Kangra; however the EFs value was marginally on the higher side for the FW collected from Hamirpur (0.36 g kg^{-1}). Among the anions, no significant spatial variation

Table 4. PAHs emission from fuel woods and dung cake: Comparative study.

	PAH (mg kg^{-1})			References
	Gaseous phase	Particulate phase	Total	
Fuel wood			5–683	Jenkins *et al.* (1996)
Fuel wood	104.6	5.2	109.7	Oanh *et al.* (2005)
Fire place/Soft wood		39.8		McDonald *et al.* (2000)
Fire wood		1.6–8.2		Kakareka *et al.* (2005)
Fuel wood	2.2–3.7	40.9		Gadi *et al.* (2012)
Fuel wood	7.24	45.28	52.52	Singh *et al.* (2013)
Fuel wood	6.87	36.91	43.78	Present study
Dung Cake		3.1–5.5		Venkataraman *et al.* (2002)
Dung Cake				Gadi *et al.* (2012)
Dung Cake	3.08	56.46	59.54	Singh *et al.* (2013)
Dung Cake	7.80	42.61	50.41	Present study

Table 5. Average emission factors (g kg^{-1}) and budget estimation (Mg yr^{-1}) of inorganic constituents from the burning of different biomass fuels used in the rural areas of HP.

a) EF (g kg^{-1})

	Anions				
	F$^-$	Cl$^-$	SO$_4^{2-}$	NO$_3^-$	PO$_4^{3-}$
FW	0.03 ± 0.01	0.30 ± 0.26	0.22 ± 0.19	0.10 ± 0.05	0.13 ± 0.03
DC	0.035 ± 0.003	0.13 ± 0.014	0.143 ± 0.004	0.058 ± 0.011	nd
	Cations				
	Na$^+$	NH$_4^+$	K$^+$	Mg^{2+}	Ca^{2+}
FW	0.17 ± 0.10	0.16 ± 0.12	0.20 ± 0.09	0.55 ± 0.01	0.95 ± 0.02
DC	0.058 ± 0.007	0.018 ± 0.009	0.033 ± 0.001	0.015 ± 0.002	0.074 ± 0.003

b) Budget estimation (Mg yr^{-1})[a]

	Anions				
	F$^-$	Cl$^-$	SO$_4^{2-}$	NO$_3^-$	PO$_4^{2-}$
FW	50.0 ± 10.0	550.0 ± 480.0	400.0 ± 340.0	170.0 ± 100.0	240.0 ± 60.0
DC	10.0 ± 1.0	40.0 ± 4.0	42.0 ± 1.0	17.0 ± 3.0	nd
	Cations				
	Na$^+$	NH$_4^+$	K$^+$	Mg^{2+}	Ca^{2+}
FW	300.0 ± 180.0	290.0 ± 210.0	360.0 ± 160.0	100.0 ± 10.0	170.0 ± 30.0
DC	17.0 ± 2.0	5.0 ± 2.0	10.0 ± 3.0	4.5 ± 0.6	22.0 ± 1.0

FW = Fuel Wood, DC = Dung Cake; [a]Consumption of FW (1.8 Mt yr^{-1}) and DC (0.3 Mt yr^{-1}): Rural energy database of TERI (TEDDY, 2012–13); nd: not detected.

was observed for F$^-$. However, large scale spatial variability in the emission of Cl$^-$ and SO$_4^{2-}$ was noted. The highest Cl$^-$ emission from FW was observed for the area of Hamirpur (0.62 ± 0.45 g kg^{-1}), followed by Sirmur (0.42 ± 0.23 g kg^{-1}) and Solan (0.43 ± 0.30 g kg^{-1}). SO$_4^{2-}$ emission factor was noted to be the highest for the FW collected from the Hamirpur (0.43 ± 0.22 g kg^{-1}) followed by Una, Kullu and Mandi areas of Himachal Pradesh.

SO$_2$ and NO$_x$

Average emission factors of NO$_x$ (NO + NO$_2$) and SO$_2$ from FW and DC based on annual consumption of the biomass fuels are summarized in Table 1. Variability in the EF of NO$_x$ (NO + NO$_2$) was observed with the type of biomass fuels (FW & DC). Average emission factors of NO$_x$ were estimated to be 0.59 ± 0.49 g kg^{-1} and 0.34 ± 0.18 g kg^{-1} for FW and DC, respectively. Among FW collected, higher emission of NO$_x$ was observed from *Dalbergia sp.* (1.46 ± 0.50 g kg^{-1}), *Cedrela sp.* (0.90 ± 0.06 g kg^{-1}),

Mallotus sp. (0.80 ± 0.06 g kg^{-1}) and *Gondla* (0.69 ± 0.33 g kg^{-1}). Comparatively, higher EFs of NO$_x$ from FW could be attributed to the fact that FW burns (flaming condition) mainly in high temperature zone (600–900°C) that leads to the reaction between atmospheric nitrogen and atmospheric oxygen which finally resulted in the thermal formation of NO (Kituyi *et al.*, 2001). In the present study percentage contributions of NO and NO$_2$ to NO$_x$ were 68% and 32%, respectively for FWs. EFs of NO and NO$_2$ noted for FW (0.40 ± 0.32 g kg^{-1} and 0.19 ± 0.17 g kg^{-1}) were higher than DC (0.23 ± 0.12 g kg^{-1} and 0.11 ± 0.06 g kg^{-1}).

Spatial variability in the EFs of NO$_x$ was also observed for FW collected from the different areas of HP. EF of NO$_x$ for the FW collected from the districts Kullu (0.70 ± 0.83 g kg^{-1}), Unna (0.75 ± 0.66 g kg^{-1}) and Hamirpur (0.77 ± 0.32 g kg^{-1}) were found to be quite similar, followed by Kangra (0.45 ± 0.29 g kg^{-1}) and Sirmur (0.57 ± 0.26 g kg^{-1}). Spatial variability of NO$_x$ from the biomass fuels (*Pinus sp* and *Eucalyptus sp.*) collected from the different district of

HP was also done. NO_x emission from *Pinus sp.* collected from Una district was comparatively high followed by the districts Bilaspur (0.48 g kg^{-1}) and Hamirpur (0.45 g kg^{-1}). Similarly, NO_x emission from the burning of *Eucalyptus sp.* collected from the district Chamba (0.75 g kg^{-1}) was highest whereas *Eucalyptus sp.* collected from the district Bilaspur (0.02 g kg^{-1}) was noted to have the lowest NO_x emission. *Pinus sp.* and *Eucalyptus sp* from Una and Chamba possibly burns for longer time in flaming than in smoldering phase and therefore could be the reason of higher NO_x emission from these areas.

Similarly, average emission factor of sulphur dioxide (SO_2) from FWs (0.43 ± 0.38 g kg^{-1}) was considerably higher than from DC (0.23 ± 0.15 g kg^{-1}) (Table 1). Highest emitter of SO_2 was *Gondla* (1.03 SO_2) followed by *Borassus sp.* (0.81 g kg^{-1}) and *Cedrela sp.* (0.71 g kg^{-1}). Spatially, highest emission of SO_2 was noted to be from the FWs collected from the district Hamirpur (0.73 ± 0.57 g kg^{-1}), followed by Kullu (0.50 ± 0.34 g kg^{-1}), Bilaspur (0.41 ± 0.32 g kg^{-1}) and Kangra (0.40 ± 0.32 g kg^{-1}). Spatial variability is noticed in the emission factor of SO_2 from the burning of the *Pinus sp* and *Eucalyptus sp*. *Pinus sp* collected from Hamirpur (0.40 g kg^{-1}) emits higher SO_2 followed by Shimla (0.34 g kg^{-1}). This suggests that wood collected from the Shimla are probably more smoldering than those collected from Hamirpur. Similarly, *Eucalyptus sp.* from the district Mandi (0.54 g kg^{-1}) noted to have the maximum emission of SO_2 than *Eucalyptus sp.* from Bilaspur (0.51 g kg^{-1}) and therefore could be inferred that woods from Mandi have comparatively longer flaming phase. Also, from the above discussion it was observed that the woods from Bilaspur have lower SO_x, NO_x and EC emission; this could be probably attributed to

the longer smoldering stage during the course of burning.

Emission Estimates of PM, OC, EC, PAHs and Trace Gases over Himachal Pradesh

The budget of emission of PM, SO_2, NO_x, OC, EC, PAHs and water soluble cations and anions from residential biomass fuels have been calculated and summarized in Tables 5 and 6, using their EFs and the annual consumption data (TEDDY, 2013) for FW and DC for the state of Himachal Pradesh. Because of the higher consumption of FW (1.8 Mt yr^{-1}) compare to DC (0.3 Mt yr^{-1}), the annual budgets of particulates and gases from FW (PM: 6.19 ± 4.28 Gg yr^{-1}, SO_2: 0.77 ± 0.68 Gg yr^{-1} and NO_x: 1.06 ± 0.88 Gg yr^{-1}) were higher than from DC (PM: 3.44 ± 0.39 Gg yr^{-1}, SO_2: 0.07 ± 0.04 Gg yr^{-1} and NO_x: 0.10 ± 0.05 Gg yr^{-1}). The annual budget estimate points towards the fact that though the particulates EF of DC was higher than FW but the overall emission relies on the consumption of the biomass fuel.

Similarly, budget of emission of OC and EC was also estimated (Table 6). Annual emission of OC from FW and DC was almost consistent, whereas, emission of EC from FW (0.55 ± 0.28 Gg yr^{-1}) was almost seven times higher than that observed for DC (0.078 ± 0.006 Gg yr^{-1}). Contribution of total carbon (OC + EC) to the particulates from the burning of both FW and DC were 26% and 30%, respectively. Emission estimates of OC and EC for FW and DC in the present study were considerably lower than those reported by Sen *et al.* (2014) over Western region of India.

Using the laboratory generated emission factor of PAHs from the residential biomass burning and consumption of biomass fuel (FW and DC), the budgets of total PAHs emission for the state of Himachal Pradesh were estimated

Table 6. Budget estimates (Gg yr^{-1}) of PM, OC, EC, PAHs, SO_2, NO and NO_2 from the burning of different biomass fuels used for domestic purpose in the rural areas of HP and Northwestern region of India.

a) Particulates and carbonaceous emission (Gg yr^{-1})[a]

States	FW				DC			
	PM	OC	EC	TC	PM	OC	EC	TC
Himachal Pradesh	6.19 ± 4.28	1.08 ± 0.79	0.55 ± 0.28	1.63 ± 0.99	3.44 ± 0.39	0.96 ± 0.10	0.08 ± 0.01	1.04 ± 0.11
Uttarakhand[b]	8.61 ± 4.52	2.48 ± 1.46	0.73 ± 0.23	3.21 ± 2.69	-	-	-	-
Northwestern Himalayan region	14.90 ± 9.14	3.42 ± 2.21	1.31 ± 0.54	4.73 ± 2.75	3.44 ± 0.39	0.96 ± 0.10	0.08 ± 0.01	1.04 ± 0.11

b) Trace gases emission (Gg yr^{-1})[a]

States	FW				DC			
	SO_2	NO	NO_2	NO_x	SO_2	NO	NO_2	NO_x
Himachal Pradesh	0.77 ± 0.68	0.72 ± 0.57	0.34 ± 0.30	1.06 ± 0.88	0.07 ± 0.05	0.07 ± 0.04	0.03 ± 0.0.02	0.10 ± 0.05
Uttarakhand[a]	0.46 ± 0.11	0.22 ± 0.13	0.19 ± 0.18	0.41 ± 0.31	-	-	-	-
Northwestern Himalayan region	1.40 ± 0.95	1.08 ± 0.81	0.59 ± 0.45	1.67 ± 1.26	0.07 ± 0.05	0.07 ± 0.04	0.03 ± 0.02	0.10 ± 0.05

c) PAH (Mg yr^{-1})[a]

States	FW	DC
Himachal Pradesh	78.0 ± 65.0	13.2 ± 3.0
Uttarakhand[c]	180.0 ± 60.0	-
Northwestern Himalayan region	243.0 ± 120.0	13.2 ± 3.0

[a] Consumption of FW (HP: 1.8 Mt yr^{-1}; Uttarakhand: 2.7 Mt yr^{-1}; Northwestern Himalayan region: 4.5 Mt yr^{-1}) and DC (HP: 0.3 Mt yr^{-1}; Uttarakhand: not used) from rural energy database (TEDDY 2012–2013), [b] Saud *et al.* (2011, 2012); [c] Singh *et al.* (2013).

(Table 6). Budget estimation of total annual PAHs from DC in the present study was 13.2 ± 3.0 Mg yr^{-1}. Total annual PAHs emission from FW (78.0 ± 65.0 Mg yr^{-1}) estimated in this study was comparatively higher than annual emission from DC. Singh *et al.* (2013) estimated the total PAHs (FW: 3.13 ± 1.08 Gg yr^{-1}; DC: 2.95 ± 0.98 Gg yr^{-1}) for IGP, India. Values in the present study were considerably lower, however the values for individual states of IGP such as: Haryana (FW: 80.0 ± 30.0 Mg yr^{-1}), Punjab (FW: 90.0 ± 40.0 Mg yr^{-1}) and Delhi (FW: 10.0 ± 0.4 Mg yr^{-1}) were consistent with our results.

Consumption data and emission factors of cations and anions emitted from the combustion of biomass were used to estimate the annual emission of the aforementioned species. Cations have the variable annual emission from the FW and DC, respectively. Amongst the cationic species emitted from the FWs, K$^+$ (360.0 ± 160.0 Mg yr^{-1}) had the highest annual emission followed by Na$^+$ (300.0 ± 180.0 Mg yr^{-1}) and NH$_4^+$ (290.0 ± 210.0 Mg yr^{-1}). Similarly Na$^+$ (17.0 ± 2.0 Mg yr^{-1}) and Ca^{2+} (22.0 ± 1.0 Mg yr^{-1}) were observed to be the top emitted species from the burning of DC. Anionic emissions from the FW were noted to be in the following order: Cl$^-$ (550.0 ± 480.0 Mg yr^{-1}) > SO$_4^{2-}$ (400.0 ± 340.0 Mg yr^{-1}) > PO$_4^{3-}$ (240.0 ± 60.0 Mg yr^{-1}) > NO$_3^-$ (170.0 ± 100.0 Mg yr^{-1}) > F$^-$ (50.0 ± 10.0 Mg yr^{-1}). Total annual cationic and anionic emission from the biomass fuels of HP were 1.27 ± 0.77 Gg and 1.51 ± 0.99 Gg, respectively (Table 5).

Emission Estimates of PM, OC, EC, PAHs and Trace Gases over Northwestern Himalayan Region

Saud *et al.* (2011, 2012) and Singh *et al.* (2013) reported the budget estimation of particulates and trace gases from the burning of biomass fuels collected from the rural areas of Uttarakhand, a mountainous state in the IGP region of India. EFs calculated for the emissions in the present study over Himachal Pradesh and the one over Uttarakhand were used to estimate the annual emissions of PM, OC, EC, PAHs and trace gases (SO$_x$ and NO$_x$) from biomass fuels (FW + DC) over Northwestern Himalayan region (HP and Uttarakhand). Average particulate emission factor calculated over Northwestern Himalayan region for FW and DC was 3.31 ± 2.03 g kg^{-1} and 11.43 ± 1.13 g kg^{-1}, respectively (Table 1). PM emissions noted in this study over Northwestern Himalayan region from FW (14.90 ± 9.14 Gg yr^{-1}) was approximately four times higher than from DC (3.44 ± 0.39 Gg yr^{-1}) (Table 6).

Similarly, average emission factors of OC, EC and total carbon (TC) were 0.76 ± 0.49 0.29 ± 0.12 g kg^{-1} and 1.05 ± 0.61 g kg^{-1}, respectively from FW. Average OC emission observed in the present study from DC (3.2 ± 0.34 g kg^{-1}) was considerably higher than those noted for FW (0.76 ± 0.49 g kg^{-1}) (Table 1). Annual emission budget estimates of OC, EC and TC from FW and DC are as follows: OC$_{FW}$ (3.42 ± 2.21 Gg yr^{-1}), EC$_{FW}$ (1.31 ± 0.54 Gg yr^{-1}), TC$_{FW}$ (4.73 ± 2.75 Gg yr^{-1}), OC$_{DC}$ (0.96 ± 0.10 Gg yr^{-1}), EC$_{DC}$ (0.08 ± 0.01 Gg yr^{-1}) and TC$_{DC}$ (1.04 ± 0.11 Gg yr^{-1}) (Table 6). Similarly, total average PAHs emission over Northwestern Himalayan region from FW (54.27 mg kg^{-1}) was higher compared to PAHs emission from DC (44.37 mg kg^{-1}) (Table 1). Particulate PAHs contribution to total PAHs was considerably higher than gaseous PAHs emission from both FW and DC. Annual emission of total PAHs estimated over Northwestern Himalayan region from FW and DC was 243.0 Mg and 13.2 Mg, respectively (Table 6).

EFs of trace gases (SO$_2$ and NO$_x$) were also estimated over Northwestern Himalayan region for FW and DC (Table 1). Average SO$_2$ emission (0.30 ± 0.21 g kg^{-1}) was almost consistent with NO$_x$ (0.37 ± 0.28 g kg^{-1}) emission from FW. However, average NO contributions to NO$_x$ was approximately two times higher than NO$_2$. SO$_2$ emission factor (0.23 ± 0.15 g kg^{-1}) was noted slightly less as compared to NO$_x$ (0.34 ± 0.18 g kg^{-1}) for DC. Estimate of the annual emissions of SO$_2$ and NO$_x$ from FW (SO$_2$: 1.40 ± 0.95 Gg yr^{-1}; NO$_x$: 1.67 ± 1.26 Gg yr^{-1}) were considerably higher than from DC (SO$_2$: 0.07 ± 0.05 Gg yr^{-1}; NO$_x$: 0.10 ± 0.05 Gg yr^{-1}) (Table 6).

Comparison of Emission of Pollutants from Northwestern Himalayan Region with Western and IGP Region of India
Emission Factor

Present study over Northwestern Himalayan region is compared with the emission of different chemical species from FW and DC over Western and the IGP region of India (Table 7). The particulate emission in the present study from FW and DC was considerably high as compared to the average EFs estimated for FW (1.69 ± 0.98 mg kg^{-1}) and DC (5.37 ± 3.90 g kg^{-1}) over Western India by Sen *et al.* (2014). Contrary, particulate EF noted for FW and DC by Saud *et al.* (2011) over IGP region was noted high as compared to the emission values evaluated over Northwestern Himalayan region (Table 7). Similarly, values of OC and EC from the burning of FWs were slightly higher in present study than observed over Western India (OC: 0.43 ± 0.29 mg kg^{-1}: EC: 0.25 ± 0.16 g kg^{-1}) (Sen *et al.*, 2014). Contrary, EF of OC over the IGP region (OC: 0.95 ± 0.27 g kg^{-1}) were at higher side whereas EF of EC (0.35 ± 0.07 g kg^{-1}) was comparable with present value over Northwestern Himalayan region. EF of SO$_2$ for FW over Northwestern Himalayan region (0.30 ± 0.21 g kg^{-1}) was higher than reported in Saud *et al.* (2011) work over IGP region (0.26 ± 0.10 g kg^{-1}), however, the average values for DC in present study (0.23 ± 0.15 g kg^{-1}) were in the range (0.28 ± 0.09 g kg^{-1}). SO$_2$ EF was substantial lower in the present study than those reported by Sen *et al.* (2014) over Western India. Similarly, EF of NO$_x$ was observed to be lower for FW and DC over Northwestern Himalayan region than those over Western (FW: 1.60 ± 0.74 g kg^{-1}; DC: 1.41 ± 0.68 g kg^{-1}) and IGP region of India (FW: 0.76 g kg^{-1}; DC: 0.58 g kg^{-1}). Emission values corresponding to average sum of anions (F, Cl$^-$, NO$_3^-$, SO$_4^{2-}$, PO$_4^{3-}$) and cations (Na$^+$, NH$_4^+$, K$^+$, Mg^{2+} and Ca^{2+}) over the study region were almost parallel with the values observed over IGP (anion: 0.59 ± 0.13 g kg^{-1}; cation: 0.30 ± 0.03 g kg^{-1}), however the same values were found lower over Western India when compared with the values noted over Northwestern Himalayan region (anion: 0.57 ± 0.28 g kg^{-1}; cation: 0.43 ± 0.18 g kg^{-1}).

Table 7. Inter comparison of average emission factor, budget estimation and ratios for various chemical constituents from the burning of residential biomass (FW + DC) fuels used in the different regions of India.

	IGP region of India[a]		Western region of India[b]		Northwestern Himalayan region	
	EF (g kg^{-1})	Budget estimates (Gg yr^{-1})[c]	EF (g kg^{-1})	Budget estimates (Gg yr^{-1})[c]	EF (g kg^{-1})	Budget estimates (Gg yr^{-1})[c]
PM	FW:4.34 ± 1.06 DC:16.26 ± 2.29	1330 ± 557	FW:1.69 ± 0.98 DC:5.37 ± 3.90	123.66 ± 71.23	FW:3.31 ± 2.03 DC:11.43 ± 1.13	18.32 ± 9.53
Carbonaceous aerosol						
OC	FW:0.95 ± 0.27 DC:3.87 ± 1.09	321.24 ± 138.96	FW: 0.43 ± 0.29 DC:1.14 ± 0.67	28.66 ± 16.72	FW:0.76 ± 0.49 DC:3.20 ± 0.34	4.38 ± 2.31
EC	FW:0.35 ± 0.07 DC:0.49 ± 0.25	47.83 ± 22.4	FW: 0.25 ± 0.16 DC: 0.14 ± 0.08	11.39 ± 6.52	FW:0.29 ± 0.12 DC:0.26 ± 0.02	1.39 ± 0.55
TC (OC +EC)	FW:1.30 ± 0.34 DC:4.36 ± 1.34	369.0 ± 161.3	FW: 0.68 ± 0.45 DC:1.28 ± 0.75	39.9 ± 23.2	FW:1.05 ± 0.61 DC:3.46 ± 0.36	5.77 ± 2.86
WSIC						
∑Anions	0.59 ± 0.13	80.0 ± 29.33	0.31 ± 0.24	15.68 ± 12.14	0.57 ± 0.28	1.51 ± 0.99
∑Cations	0.30 ± 0.03	39.68 ± 9.51	0.11 ± 0.09	5.56 ± 0.50	0.43 ± 0.18	1.27 ± 0.60
Trace gases						
SO_2	FW:0.26 ± 0.10 DC:0.28 ± 0.09	25.09 ± 11.84	FW: 0.66 ± 0.37 DC: 0.74 ± 0.48	34.81 ± 19.87	FW:0.30 ± 0.21 DC:0.23 ± 0.15	1.47 ± 1.00
NO_x (NO +NO_2)	FW:0.76 ± 0.53 DC:0.58 ± 0.44	52.76 ± 37.10	FW: 1.60 ± 0.74 DC: 1.41 ± 0.68	121.02 ± 49.08	FW:0.32 ± 0.28 DC:0.34 ± 0.08	1.77 ± 1.31
Ratios						
	OC/EC	OC/TC	TC/PM	K^+/EC	Cl^-/EC	SO_4^{2-}
IGP region of India	FW: 2.71 DC: 7.89	FW: 0.79 DC: 0.88	FW: 0.31 DC: 0.30	FW: 0.37 DC: 0.20	FW: 0.94 DC: 0.79	FW: 0.18 DC: 0.14
Western region of India	FW: 1.72 DC: 8.14	FW: 0.63 DC: 0.89	FW: 0.40 DC: 0.24	FW: 0.11 DC: 0.12	FW: 0.38 DC: 1.12	FW: 0.092 DC: 0.107
Northwestern Himalayan region of India	FW: 2.62 DC: 12.30	FW: 0.72 DC: 0.92	FW: 0.31 DC: 0.30	FW: 0.64 DC: 0.12	FW: 0.96 DC: 0.50	FW: 0.70 DC: 0.55

[a] Saud *et al.* (2011, 2012, 2013), [b] Sen *et al.* (2014), [c] Based on total consumption of biomass fuels (FW + DC).

Budget Estimation

We also report here the comparative study of total annual emission budget estimates of particulates and trace gases from the biomass fuels (FW+DC) in three different regions of India (Table 7). Total annual emission of PM from biomass fuels (FW +DC) of IGP, India (1330 ± 557 Gg yr^{-1}) was much higher followed by Western India (123.66 ± 71.23 Gg yr^{-1}) and Northwestern Himalayan region of India (18.32 ± 9.53 Gg yr^{-1}). This is due to the fact that in IGP alone total annual consumption of biomass fuel (FW+DC) is approximate 135 Mt followed by Western India (50.6 Mt). In Northwestern Himalayan region (HP + Uttarakhand) total consumption of biomass fuel is significantly less (4.8 Mt). Similarly, carbonaceous emissions from IGP (OC: 321.24 ± 138.96 Gg yr^{-1}; EC: 47.83 ± 22.4 Gg yr^{-1}) were also found to have highest followed by Western India (OC: 28.66 ± 16.72 Gg yr^{-1}; EC: 11.39 ± 6.52 Gg yr^{-1}) and Northwestern Himalayan region of India (OC: 4.38 ± 2.31 Gg yr^{-1}; EC: 1.39 ± 0.55 Gg yr^{-1}). Total budget estimates of trace gases noted in three different regions are as follows: SO_2 (Northwestern Himalayan region of India: 1.47 ± 1.0 Gg yr^{-1}; Western India: 34.81 ± 19.87 Gg yr^{-1}; IGP, India: 25.09 ± 11.84 Gg yr^{-1}). Like SO_2, NO_x annual budget estimation follows the same pattern (Table 7). Here it is significant to note that despite of less consumption of biomass fuel in Western India as compared to IGP; annual emission of SO_2 and NO_x was comparatively higher in Western India from the burning of biomass fuels.

In the present study we have also estimated emission per square kilometer of particulates and trace gases over different regions of India (Fig. 2). PM emission observed per km^2 over the IGP (0.52 ± 0.21 Mg km^{-2}) and Western India (0.23 ± 0.13 Mg km^{-2}) was almost 3 and 1.3 times higher than over Northwestern Himalayan region (0.16 ± 0.08 Mg km^{-2}). Similarly OC and EC emissions were 3 and 1.5 times and 1.2 and 1.7 higher over IGP (OC: 0.12 ± 0.05 Mg km^{-2}; EC: 0.019 ± 0.008 Mg km^{-2}) and Western India (OC: 0.05 ± 0.03 Mg km^{-2}; EC: 0.21 ± 0.01 Mg km^{-2}), respectively than Northwestern Himalayas (OC: 0.04 ± 0.02 Mg km^{-1}; EC: 0.012 ± 0.005 Mg km^{-2}). Emissions of SO_2 and NO_x were notably high over Western India (SO_2: 0.06 ± 0.03 Mg km^{-2}; NO_x: 0.22 ± 0.09 Mg km^{-2}) which was approximately 3.5 and 17.0 times higher than over Northwestern Himalayan region (SO_2: 0.018 ± 0.007 Mg km^{-2}; NO_x: 0.013 ± 0.008 Mg km^{-2}). Per km^2 emission of anion and cation were found almost 4 times high over IGP and 2.5 and 1.5 times higher over Western India when compared with Northwestern Himalayan region.
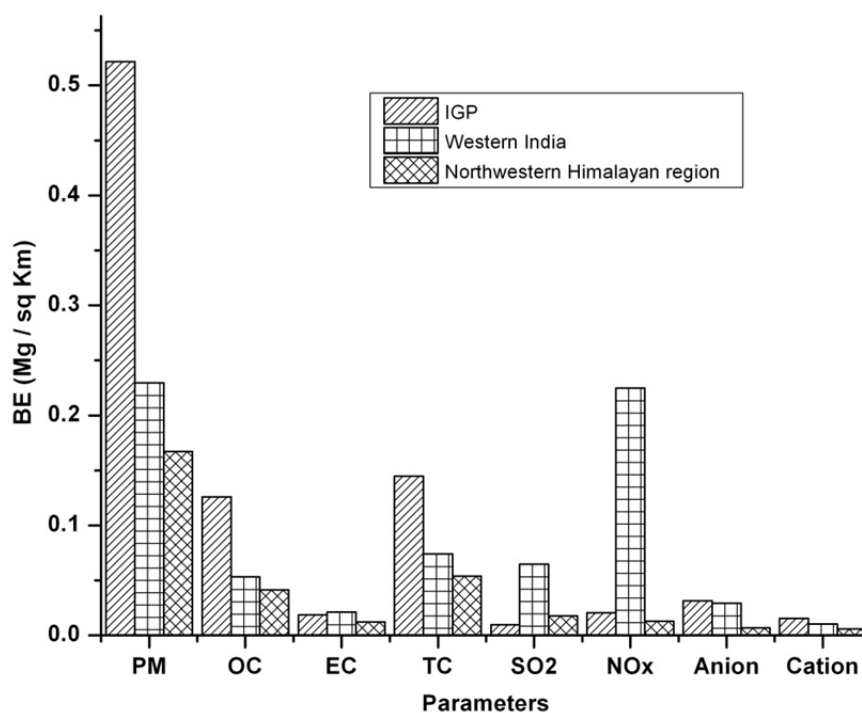
Fig. 2. Budget estimation (Mg km^{-2}) of chemical species over different environment of India: Comparative study.

Significant Ratios

Ratios of OC, EC and TC to PM in the flue gases from the combustion sources have been used as the tracer in the source apportionment studies. Higher ratio of OC to EC is the indicative of biomass burning; conversely, it alludes towards the presence of fossil fuel combustion sources (Novakov *et al.*, 2000). In earlier studies, Saud *et al.* (2012) and Sen *et al.* (2014) have reported OC/EC, OC/TC, TC/PM, K$^+$/EC and Cl$^-$/EC ratios over the IGP and Western India from. In the present study, we have also calculated the above ratios for both the fuel types (FW and DC). Average OC/EC ratio was highest for DC (12.30) followed by FW (2.62). Similarly, average OC/TC ratios for FW (0.72) and DC (0.92) varied marginally. OC/EC ratio calculated over IGP and Western regions of India for biomass fuels was >1.0 and OC/TC ratio in the present study for FW and DC was consistent with the ratios reported for same biomass fuels over the IGP and Western region of India. Similarly, Cl$^-$/EC ratio reported in biomass burning emission was 0.15–0.44 (Ferek *et al.*, 1998; Yamasoe *et al.*, 2000), average Cl$^-$/EC ratio in the present study (0.96) was comparatively higher but resembles with the values reported over IGP (0.94) for FW. The constituents of K$^+$ and Cl$^-$ rich particles present in the vegetation volatilized during the combustion process and afterwards converted into the particulate phase by nucleation or condensation (Alves *et al.*, 2010). Emissions from fossil fuel combustion do not have K$^+$ and therefore K$^+$/EC ratio is most likely to be zero or negligible (George *et al.*, 2011). In the present study the average K$^+$/EC ratio calculated (FW: 0.64 and DC: 0.12) were in the range of the ratios calculated by Andreae, (1983) and Ram *et al.* (2010) for biomass burning emission.

CONCLUSIONS

Emission factors and budgets of chemical species (OC, EC, PAHs, SO$_2$, NO$_x$, Na$^+$, K$^+$ Mg^{2+}, Ca^{2+}, F$^-$, Cl$^-$, SO$_4^{2-}$, NO$_3^-$ and PO$_4^{3-}$) were determined in the laboratory through simulation of burning for the two categories of biomass fuels (FW and DC) collected from the rural areas of Himachal Pradesh. In addition, EFs of chemical species from biomass fuels over Uttarakhand were used to estimate the annual emission of particulate and trace gases over Northwestern Himalayan region.

Important features in the present study are discussed below:

1) EFs of PM from FWs (3.44 ± 2.38 g kg^{-1}) over HP were comparatively lower than that from DC (11.43 ± 1.13 g kg^{-1}). Average emissions of SO$_2$ and NO$_x$ from FW were 0.43 ± 0.38 g kg^{-1} and 0.59 ± 0.49 g kg^{-1}, respectively. Similarly, EFs of SO$_2$ and NO$_x$ from DC were 0.23 ± 0.15 g kg^{-1} and 0.34 ± 0.18 g kg^{-1}.

2) Emission of carbonaceous aerosols (OC and EC) constitutes ~26% and ~30% of the PM from FWs and DC, respectively. It was observed that OC emissions over HP from FW (0.60 ± 0.44 g kg^{-1}) were lower than from DC (3.20 ± 0.34 g kg^{-1}). Annual emission of OC from both FW (1.08 ± 0.79 Gg yr^{-1}) and DC (0.96 ± 0.10 Gg yr^{-1}) were comparable despite of the fact that the consumption value of DC was significantly less. EF of EC for FW (av. 0.31 ± 0.16 g kg^{-1}) and DC (av. 0.26 ± 0.02 g kg^{-1}) was nearly same. Total annual carbonaceous aerosol (OC +EC) emission observed from the biomass fuels over HP was 2.66 ± 1.0 Gg yr^{-1}.

3) The average emission factors of total PAHs from FW and DC over HP was $\sum_{PAHs}$ 43.25 mg kg^{-1} and $\sum_{PAHs}$

44.71 mg kg^{-1}, respectively. Based on consumption of biomass fuels, the budget estimates of emission of total PAHs were 77.85 Mg yr^{-1} and 13.31 Mg yr^{-1} for FW and DC, respectively.

4) Emission factors of anions from the burning of biomass fuels over HP were higher than those of cations. Among anions, maximum EF was noted for Cl$^-$ (FW: 0.30 ± 0.26 g kg^{-1}; DC: 0.13 ± 0.01 g kg^{-1}) and SO$_4^{2-}$ (FW: 0.22 ± 0.19 g kg^{-1}; DC: 0.143 ± 0.004 g kg^{-1}) for both biomass fuels (FW and DC). Similarly, for cations highest EF was noted for K$^+$ (0.20 ± 0.09 g kg^{-1}) and Na$^+$ (0.17 ± 0.10 g kg^{-1}) for FW. Ca^{2+} (0.074 ± 0.003 g kg^{-1}) and Na$^+$ (0.058 ± 0.007 g kg^{-1}) were the dominant species in plumes of DC burning.

5) PM emissions noted in this study over Northwestern Himalayan region from FW and DC are 14.90 ± 9.14 Gg yr^{-1} and 3.44 ± 0.39 Gg yr^{-1}, respectively. Annual emission budget estimates of OC and EC from FW and DC are as follows: OC$_{FW}$ (3.42 ± 2.21 Gg yr^{-1}), EC$_{FW}$ (1.31 ± 0.54 Gg yr^{-1}), OC$_{DC}$ (0.96 ± 0.10 Gg yr^{-1}) and EC$_{DC}$ (0.08 ± 0.01 Gg yr^{-1}). Total PAHs estimated over Northwestern Himalayan region from FW and DC was 243.0 ± 120.0 Mg yr^{-1} and 13.2 ± 3.0 Mg yr^{-1}, respectively. Total annual ionic emission estimated from the burning of biomass fuels of the North western region of Himalaya was 2.78 ± 1.76 Gg yr^{-1}. Annual emission estimate of SO$_2$ and NO$_x$ from FW (SO$_2$: 1.40 ± 0.95 Gg yr^{-1}; NO$_x$: 1.67 ± 1.26 Gg yr^{-1}) were considerably higher than from DC (SO$_2$: 0.07 ± 0.05 Gg yr^{-1}; NO$_x$: 0.10 ± 0.05 Gg yr^{-1})

ACKNOWLEDGEMENTS

SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

REFERENCES

Alves, C.A., Gonçalves, C., Pio, C.A., Mirante, F., Caseiro, A., Tarelho, L., Freitas, M.C. and Viegas, D.X. (2010). Smoke Emissions from Biomass Burning in a Mediterranean Shrubland. *Atmos. Environ.* 44: 3024–3033.

Andreae, M.O. (1983). Soot Carbon and Excess Fine Potassium: Long-range Transport of Combustion-derived Aerosols. *Science* 220: 1148–1151.

Andreae, M.O., Rosenfeld, D., Artaxo, P., Costa, A.A., Frank, G.P., Longo, K.M. and Silva-Dias, M.A.F. (2004). Smoking Rain Clouds over the Amazon. *Science* 303: 1337–1342.

Bond, T.C., Streets, D.G., Yarber, K.F., Nelson, S.M., Woo, J.H. and Klimont, Z. (2004). A Technology-based Global Inventory of Black and Organic Carbon Emissions from Combustion. *J. Geophys. Res.* 109: D14203.

Bond, T.C. and Bergstrom, R.W. (2006). Light Absorption by Carbonaceous Particles: An Investigative Review. *Aerosol Sci. Technol.* 40: 27–67.

Cachier, H. and Ducret, J. (1991). Influence of Biomass Burning on Equatorial African Rains. *Nature* 336: 464–466.

Chow, J.C., Watson, J.G., Pritchett, L.C., Pierson, W.R., Frazier, C.A. and Purcell, R.G. (1993). The DRI Thermal/Optical Reflectance Carbon Analysis System: Description, Evaluation and Applications in US Air Quality Studies. *Atmos. Environ.* 27A: 1185–1201.

Davidson, C.I., Phalen, R.F. and Solomon, P.A. (2005). Airborne Particulate Matter and Human Health: A Review. *Aerosol Sci. Technol.* 39: 737–749.

Dhammapala, R., Claiborn, C., Jimenez, J., Corkill, J., Gullett, B., Simpson, C. and Paulsen, M. (2007). Emission Factors of PAHs, Methoxyphenols, Levoglucosan, Elemental Carbon and Organic Carbon from Simulated Wheat and Kentucky Bluegrass Stubble Burns. *Atmos. Environ.* 41: 2660–2669

Ferek, R.J., Reid, J.S. and Hobbs, P.V. (1998). Emission Factors of Hydrocarbons, Halocarbons, Trace Gases and Particles from Biomass Burning in Brazil. *J. Geophys. Res.* 103: 32107–32118.

Fung, K.K., Chow, J.C., and Watson, J.G. (2002). Evaluation of OC/EC Speciation by Thermal Manganese Dioxide Oxidation and the IMPROVE Method. *J. Air Waste Manage. Assoc.* 52: 1333–1341.

Gadi, R., Kulshestha, U.C., Sarkar, A.K., Garg, S.C. and Parashar, D.C. (2003). Emissions of SO$_2$ and NO$_x$ from Biofuels in India. *Tellus Ser B* 55: 787–795.

Gadi, R., Singh, D.P., Saud, T., Mandal, T.K. and Saxena, M. (2012). Emission Estimates of Particulate PAHs from Biomass Fuels Used in Delhi, India. *Hum. Ecol. Risk Assess.* 18: 871–887.

Garg, A., Shukla, P.R., Bhattacharya, S. and Dadhwal, V.K. (2001). Sub-region (District) and Sector Level SO$_2$ and NO$_x$ Emissions for India: Assessment of Inventories and Mitigation Flexibility. *Atmos. Environ.* 35: 703–713.

Gautam, R., Liu, Z., Singh, R.P. and Hsu, N.C. (2009). Two Contrasting Dust-dominant Periods over India Observed from MODIS and CALIPSO. *Geophys. Res. Lett.* 36: L06813, doi: 10.1029/2008GL036967.

George, S.K., Nair, P.R., Parameswaran, K. and Jacob, S. (2011). Wintertime Chemical Composition of Aerosols at a Rural Location in the Indo-Gangetic Plains. *J. Atmos. Solar Terr. Phys.* 13: 1798–1809.

Guofeng, S., Siye, W., Wen, W., Yanyan, Z., Yujia, M., Bin, W., Rong, W., Wei, L., Ye, S.H.H., Yifeng, Y., Wei, W., Xilong, W., Xuejun, W. and Shu, T. (2012). Emission Factors, Size Distributions, and Emission Inventories of Carbonaceous Particulate Matter from Residential Wood Combustion in Rural China. *Environ. Sci. Technol.* 46:

4207–4214.

Gupta, S., Saksena, S., Shankar, V.R. and Joshi, V. (1998). Emission Factors and Thermal Efficienciesof Cookingbiofuels Fromfive Countries. *Biomass Bioenergy* 14: 547–559.

Habib, G., Venkataraman, C., Bond, T.C. and Schauer, J.J. (2008). Chemical, Microphysical and Optical Properties of Primary Particles from the Combustion of Biomass Fuels. *Environ. Sci. Technol.* 42: 8829–8834.

Hall, M. (1989). Relative Contribution of Various forms of Cytochrome P450 to the Metabolism of Benzo(a)pyrene, by Human Liver Microsomes. *Carcinogenesis* 10: 1815–1821.

Jenkins, B.M., Jones, A.D., Turn, S.Q. and Williams, R.B. (1996). Emission Factors for Polycyclic Aromatic Hydrocarbons from biomass Burning. *Environ. Sci. Technol.* 30: 2462–2469.

Kituyi, E., Marufu, L., Wandiga, S.O., Jumba, I.O., Andreae, M.O. and Helas, G. (2001). Carbon Monoxide and Nitric Oxide from Biofuels fires in Kenya. *Energy Convers. Manage.* 42: 1517–1542.

Kuniyal, J.C., Sharma, M., Chand, K. and Mathela, C.S. (2015). Water Soluble Ionic Components in Particulate Matter (PM_{10}) during High Pollution Episode Days at Mohal and Kothi in the North-Western Himalaya, India. *Aerosol Air Qual. Res.* 15: 529–543.

Lee, S., Kim, H.K., Yan, B., Cobb, C.E., Hennigan, C., Nichols, S., Chamber, M., Edgerton, E., Jansen, J.J., Hu, Y., Zheng, M., Weber, R.J. and Russell, A.G. (2008). Diagnosis of Aged Prescribed Burning Plumes Impacting an Urban area. *Environ. Sci. Technol.* 42: 1438–1444.

Li, X., Wang, S., Duan, L., Hao, J. and Nie, Y. (2009). Carbonaceous Aerosol Emissions from Household Biofuels Combustion in China. *Environ. Sci. Technol.* 43: 6076–6081.

Mayewski, P.A. and Jeschke, P.A. (1979). Himalayan and Trans-Himalayan Glacier Fluctuations Since AD 1812. *Arct. Alp. Res.* 11: 267–287.

McMeeking, G.R., Kreidenweis, S.M., Baker, S., Carrico, C.M., Chow, J.C., Collett, J.L., Hao, W.M., Holden, A.S., Kirchstetter, T.W., Malm, W.C., Moosmuller, H., Sullivan, A.P. and Wold, C.E. (2009). Emissions of Trace Gases and Aerosols during the Open Combustion of Biomass in the Laboratory. *J. Geophys. Res.* 114: D19210, doi: 10.1029/2009JD011836.

Nel, A. (2005). Air Pollution–Related illness: Effects of Particles. *Science* 308: 804–806.

Novakov, T. and Corrigan, C.E. (1996). Cloud Condensation Nucleus Activity of the Organic Component of Biomass Smoke Particles. *Geophys. Res. Lett.* 23: 2141–2144.

Novakov, T., Andreae, M.O., Gabriel, R., Kirchstetter, T.W., Mayol-Bracero, O.L. and Ramanathan, V. (2000). Origin of Carbonaceous Aerosols over the Tropical Indian Ocean: Biomass Burning or Fossil Fuels? *Geophys. Res. Lett.* 27: 4061–4064.

Parashar, D.C., Gadi, R., Mandal, T.K. and Mitra, A.P. (2005). Carbonaceous Aerosol Emissions from India. *Atmos. Environ.* 39: 7861–7871.

Penner, J.E., Dickinson, R.E. and O'Neill, C.A. (1992). Effects of Aerosol from Biomass Burning on the Global Radiation Budget. *Science* 256: 1432–1434.

Philips, R., Vastola, F.J. and Walker, P.L. (1969). The Effect of Oxygen Pressure and Carbon Burn Off on the Product Ratio of the Carbon-oxygen Reaction. *Carbon* 7:479–485.

Rajput. P., Sarin, M.M., Sharma, D. and Singh, D. (2014). Characteristics and Emission Budget of Carbonaceous Species from Post-harvest Agricultural-waste Burning in Source Region of the Indo-Gangetic Plain. *Tellus Ser. B* 66: 21026, doi: 10.3402/tellusb.v66.21026.

Ram, K., Sarin, M.M. and Tripathi, S.N. (2010). One-year Record of Carbonaceous Aerosols from an Urban Location (Kanpur) in the Indo-Gangetic Plain: Characterization, Sources and Temporal Variability. *J. Geophys. Res.* 115: D24313–24327.

Ramanathan, V. and Carmichael, G. (2008). Global and Regional Climate Changes Due to Black Carbon. *Nat. Geosci.* 1: 221–227.

Rastogi, N., Patel, A., Singh, A. and Singh, D. (2015). Diurnal Variability in Secondary Organic Aerosol Formation over the Indo-Gangetic Plain during Winter Using Online Measurement of Water-Soluble Organic Carbon. *Aerosol Air Qual. Res.* 15: 2225–2231, doi: 10.4209/aaqr.2015.02.0097.

Rau, J.A. (1986). Residential Wood Combustion Aerosol Characterization as a Function of Size and Source Apportionment Using Chemical Mass Balance Modeling (Ph.D. Dissertation). Oregon Graduate Center, Beaverton, OR, USA.

Reddy, M.S. and Venkataraman, C. (2002). Inventory of Aerosol and Sulphur Dioxide Emissions from India, II, Biomass Combustion. *Atmos. Environ.* 36: 697–710.

Roden, C.A., Bond, T.C., Conway, S. and Pinel, A.B.O. (2006). Emission Factors and Realtime Optical Properties of Particles Emitted from traditional Wood Burning Cookstoves. *Environ. Sci. Technol.* 40: 6750–6757.

Saud, T., Mandal, T.K., Gadi, R., Singh, D.P., Sharma, S.K., Saxena, M. and Mukherjee, A. (2011). Emission Estimates of Particulate Matter (PM) and Trace Gases (SO_2, NO and NO_2) from Biomass Fuels Used in Rural Sector of Indo-Gangetic Plain, India. *Atmos. Environ.* 45: 5913–5923.

Saud, T., Gautam, R., Mandal, T.K., Gadi, R., Singh, D.P., Sharma, S.K., Dahiya, M. and Saxena, M. (2012). Emission Estimates of Organic and Elemental Carbon from Household Biomass Fuel Used over the Indo-Gangetic-Plain (IGP), India. *Atmos. Environ.* 61: 212–220.

Saud, T., Saxena, M., Singh, D.P., Dahiya, M., Sharma, S.K., Datta, A., Gadi, R. and Mandal, T.K. (2013). Spatial Variation of Chemical Constituents from the Burning of Commonly Used Biomass Fuels in Rural Areas of the Indo-Gangetic Plain (IGP), India. *Atmos. Environ.* 71: 158–169.

Saxena, M., Singh, D.P., Saud, T., Gadi, R., Singh, S., Sharma, S.K. and Mandal, T.K. (2014). Study on Particulate Polycyclic Aromatic Hydrocarbons over Bay of Bengal in Winter Season. *Atmos. Res.* 145–146: 205–213.

Sen, A., Mandal, T.K., Sharma, S.K., Saxena, M., Gupta, N.C., Gautam, R., Gupta, A., Gill, T., Rani, S., Saud, T., Singh, D.P. and Gadi, R. (2014). Chemical Properties of Emission from Biomass Fuels Used in the Rural Sector of the Western Region of India. *Atmos. Environ.* 99: 411–424.

Sharma, S.K., Mandal, T.K., Saxena, M., Sharma, A., Datta, A. and Saud, T. (2014a). Variation of OC, EC, WSIC and Trace Metals of PM_{10} in Delhi, India. *J. Atmos. Solar Terr. Phys.* 113: 10–22.

Sharma, S.K., Mandal, T.K., Sharma, C., Kuniyal, J.C., Joshi, R., Dhyani, P.P., Rohtash, Sen, A., Ghayas,H., Gupta, N.V., Sharma, P., Saxena, M., Sharma, A., Arya, B.C. and Kumari, A. (2014b). Measurements of Particulate ($PM_{2.5}$), BC and Trace Gases Over the Northwestern Himalayan Region of India. *Mapan-J. Metrol. Soc. I.* 29: 243–253.

Singh, D.P., Gadi, R., Mandal, T.K., Saud, T., Saxena, M. and Sharma, S.K. (2013). Emissions Estimates of PAH from Biomass Fuels Used in Rural Sector of Indo-Gangetic Plains of India. *Atmos. Environ.* 68: 120–126

Sudheer, A.K., Rengarajan, R. and Sheel, A. (2015). Secondary Organic Aerosol over an Urban Environment in a Semi–arid Region of Western India. *Atmos. Pollut. Res*. 6: 11–20.

TEDDY: TERI Energy Data Directory Year book (2012-13). The Energy Research Institute, New Delhi, India.

Venkataraman, C. and Rao, G.U.M. (2001). Emission Factors of Carbon Monoxide and Size Resolved Aerosols from Biofuel Combustion. *Environ. Sci. Technol.* 35: 2100–2107.

Venkataraman, C., Habib, G., Eiguren-Fernandez, A., Miguel, A.H. and Friedlander, S.K. (2005). Residential Biofuels in South Asia: Carbonaceous Aerosol Emissions and Climate Impacts. *Science* 37: 1454–1456.

Xu, L., and Penner, J.E. (2012). Global Simulations of Nitrate and Ammonium Aerosols and their Radiative Effects. *Atmos. Chem. Phys.* 12: 9479–9504.

Yang, S., He, H., Lu, S., Chen, D. and Zhu, J. (2008). Quantification of Crop Residue Burning in the Field and its Influence on Ambient Air Quality in Suqian, China. *Atmos. Environ.* 42: 1961–1969.

Zou, L.Y., Zhang, W. and Atkinson, S. (2003). The Characterization of Polycyclic Aromatic Hydrocarbons Emissions from Burning of Different Firewood Species in Australia. *Environ. Pollut.* 124: 283–289.