

# PM<sub>10</sub> Bound Elements, Ions, Carbon and PAHs during Festive Biomass Burning over the Brahmaputra Valley

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**Abstract:** Episodic *meji* burning (a festive biomass burning) is a significant local contributor of PM<sub>10</sub> in the mid-Brahmaputra Valley. The PM<sub>10</sub> samples of *meji* burning period were chemically characterized for elements, ions, carbon and the USEPA's 16 priority Polycyclic Aromatic Hydrocarbons (PAHs). This paper presents the concentrations of various attributes of PM<sub>10</sub> during *meji* festivities. As such, linear relationship between EC, OC and other chemical species, and diagnostic ratios of selected PAHs indicated strong influence of biomass burning on PM<sub>10</sub> load and its properties.

**Key words:** *Meji*, biomass burning, PM<sub>10</sub> characterization, PAHs.

## Introduction

Biomass burning during festivals is often not taken into the inventories of air pollution contributors. The traditional *meji* burning during the *Bhogali Bihu* festival of the Brahmaputra valley is a unique event that releases significant volume of particulates and shows incremental effects on wintertime PM<sub>10</sub> in the region (Deka and Hoque, 2014). This non-conventional source of atmospheric PM<sub>10</sub> emanating from an annual socio-cultural practice of burning a straw and wood vertical festive fabrication (*meji*), is a significant local contributor to the already prevalent biomass burning in the region.

Conventionally, biomass burning in the form of shifting agriculture, forest fire and household burning for cooking and heating, agricultural residue burning, etc. is taken as a major contributor of atmospheric PM<sub>10</sub> load (e.g., Yamasoe et al., 2000; Reddy and Venkataraman,

2002; Pio et al., 2003; Badarinath et al., 2004; Wang et al., 2015; Zhao et al., 2015) releasing different chemical species in to the atmosphere (Shen et al., 2011; Saud et al., 2013). At the global scale, the major source of primary carbonaceous aerosols arises from biomass burning and its widespread use as fuel for cooking and heating homes (Zheng et al., 2002; Sapkota et al., 2005; Ward and Smith, 2005). Previous studies (e.g. Crutzen and Andreae, 1990; Andreae and Merlet, 2001) have reported that biomass burning is associated with large volume of particulates and chemical oxidants into the atmosphere. Global emission flux estimates from savanna and tropical forest biomass burning showed that biomass burning could be an important source of heavy metals and black carbon to the atmosphere (Yamasoe et al., 2000). Airborne particulate trace metals are health hazards since they can be absorbed into human lung tissues during breathing (Finlayson-Pitts and Pitts, 2000; Quiterio et al., 2004). Metals such as Fe, Zn

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and Cu may release free radicals in lung fluid via the Fenton reaction, and are hypothesized to cause cellular inflammation (Birmili et al., 2006).

The present study is a maiden attempt to chemically characterize and understand the characteristics of  $PM_{10}$ , in terms of the elements, ions, carbon and PAHs, during the festive *meji* burning in the Brahmaputra Valley. This paper presents monitoring and assessment of *meji* burning periods for two consecutive years.

## Materials and Methods

### Experimental Site

The study was conducted at Tezpur University campus (Figure 1), which is a rural and residential/institutional area. The campus is sylvan, surrounded by villages and agricultural fields. It is geographically positioned around  $26^{\circ}37' N$  and  $92^{\circ}50' E$ , close to the foothills of Eastern Himalayas to the North with an average

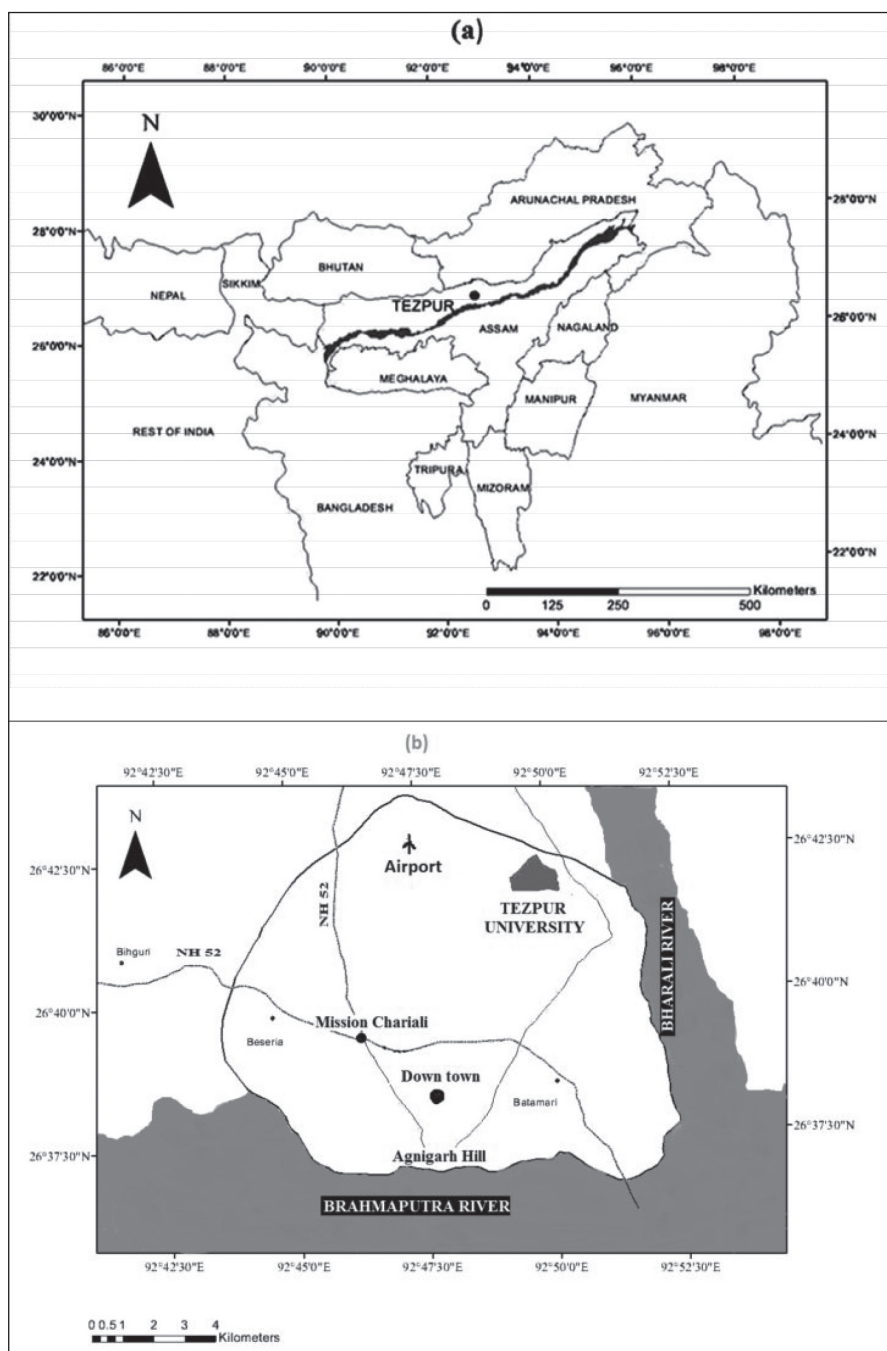


Figure 1: Map of study area: (A) Tezpur, Northeast India and adjoining regions; (B) Sampling station – Tezpur University – in Tezpur.

elevation of about 48 metres above mean sea level. The climate of Tezpur is of monsoon type with muggy summer and cool winter. According to the 2011 Census of Government of India, Sonitpur has a population of 1.7 million, 89% of which live in villages. The state of Assam has a population of 31.17 million spread over an area of 78,438 sq. km and has 25,590 villages housing over 86% of the state's population. Barring some communities, nearly entire population of the state is engaged in the *meji* burning festivities.

### Sampling

Sampling of PM<sub>10</sub> was carried out by a high volume sampler (Envirotech APM 460 BL) using Whatman Quartz microfibre filters (20.3 × 25.4 cm). Care was taken to get accurate measurements. Filters were preheated and then carefully desiccated prior and after sampling to ascertain correct measurements. The gravimetric estimation of PM<sub>10</sub> mass was done by an Analytical Balance (Sartorius make:CP225D). Sensitivity of the balance was 0.01 mg. The sampling height was 10 metres above ground.

The first sample was collected two days prior to the *Meji* burning day, which has been considered as the background sample. The 2<sup>nd</sup> sample covered the *Meji* burning episode and the 3<sup>rd</sup> and the 4<sup>th</sup> samples were collected in two days that immediately followed the episode, as muted celebrations linger in this period. The 5<sup>th</sup> and the 6<sup>th</sup> samples were collected one and two weeks after the *Meji* burning day respectively. Sampling was carried out in the cycle of 8 a.m.-8 a.m. Six 24-hr samples were collected during celebrations of each year of 2012 and 2013; Analyses of 12 samples have been presented in this paper.

### Chemical Analysis

#### Elements

Aliquot of PM<sub>10</sub> sample was digested in 9 ml HNO<sub>3</sub> in a Teflon bomb at an oven temperature of 100°C for 8 hrs. The final volume of the extract was made up to 20 ml and stored in pre-washed polyethylene bottles maintaining a pH of ~2. Elements were analyzed in an ICP-OES (PerkinElmer, OPTIMA-2100 DV).

#### Ions

For water soluble ions, an aliquot of sample was soaked in ultra-pure water (Milli-Q Simplicity system Millipore make. Resistivity = 18.2 MΩ @ 25°C) and ultrasonicated (>20 KHz) for 20 minutes in a sonicator bath (Bandelin Sonorex make) and filtered. The filtrate volume was adjusted to 20 ml in pre-washed

polyethylene bottles and kept at 4°C until analysis. Ions were analyzed in an ion chromatograph (Metrohm 882 Professional IC make). A detail of the analysis of ions is given in Deka and Hoque (2014b).

#### Carbon

Carbon was analyzed in a TOC analyzer (Analytikjena multi N/C 2100 attached with HT 1300 solid module). The particulate samples were incinerated to form CO<sub>2</sub>, which was channelized through non-dispersive infrared (NDIR) detector for measurement. Total Carbon (TC) was determined directly by feeding the sample in to the heating furnace. To estimate elemental carbon (EC), samples were preheated to remove OC and then fed into the heating chamber (Lin and Tai, 2001; Deka and Hoque, 2015). OC was obtained by subtracting EC from TC. There is a possible source of uncertainty in thermal separation of EC and OC. Humic-like substances (HULIS), typically detected in biomass smoke, are thermo-stable and, therefore, could be interfering with EC leading to slight overestimation (Reisinger et al., 2008).

Ultrapure water extract was fed in to multi N/C 2100 TOC/TNb Analyzer and Water-soluble Carbon (WSTC) and water-soluble inorganic carbon (WSIC) was obtained. The Water Soluble Organic Carbon (WSOC) was obtained by subtracting WSIC from WSTC.

#### PAHs

PAHs from aliquot of aerosol samples were extracted in dichloromethane by ultrasonic agitation (Bandelin Sonorex) at 20 KHz for 20 min. The extracts in triplicate were subsequently mixed, filtered and concentrated to 0.5-1 ml in a rotary evaporator and kept for clean-up. Samples were then cleaned up in a silica gel column (USEPA method 3630C), concentrated to 0.5-1 ml and solvent was exchanged with acetonitrile for further chromatographic analysis.

High Performance Liquid Chromatography (HPLC) system equipped with tunable absorbance UV detector and Waters PAH C18 column (Symmetry C<sup>18</sup>, 4.6 × 250 mm) was used for PAHs analysis. The mobile phase was a gradient of acetonitrile and water. Identification and quantification of the PAHs were done by calibration with PAH standard (EPA method 610 Polynuclear Aromatic Hydrocarbon 16 solution in acetonitrile).

We analyzed USEPA's 16 priority PAHs viz., naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz[a]anthracene (BaA), chrysene (CHR),

benz[b]fluoranthene (BbF), benz[k]fluoranthene (BkF), benz[a]pyrene (BaP), dibenz[a,h] anthracene (DBA), benz[ghi]perylene (BPER), and indeno[1,2,3-cd]pyrene (IND).

#### Quality Control

The ICP-OES was calibrated using ICP-Multi element standard solution VIII (Merck) and 2% HNO<sub>3</sub> (Solution-Optima Blank-Perkin Elmer Pure Plus) was used as reagent blank. To see the recovery of elements NIST SRM for Urban Particulate Matter (1648a) was used, which showed a recovery within 82 to 110% for the target elements. For estimation of anions Multielement Ion Chromatography Anion Standard Solution (Fluka Analytical) was PUT used to calibrate the IC. Cation standards were prepared in the laboratory with high purity reagents. Field blanks were taken and incorporated in the measurements. All dilutions were made in ultrapure water (18.2 grade). All the reagents used in PAHs analysis were of HPLC grade. The analytical uncertainties of duplicate analysis were within  $\pm 10\%$ . Care was taken to prevent contamination of the sample. At no point sample containers were touched with bare hands to avert contamination.

## Results and Discussion

### PM<sub>10</sub> Loading

The mass concentrations of PM<sub>10</sub> during two consecutive *meji* burning periods in 2012 and 2013 varied greatly (Figure 2). Though the maximum 24-hr PM<sub>10</sub> was found on the *meji* burning day (2<sup>nd</sup> sample) in both the years but the trends varied greatly. The 24-hour average Indian National Air Quality Standard of PM<sub>10</sub> is 100  $\mu\text{g m}^{-3}$  (CPCB, 2009) and the concentrations appeared to be much above this value.

There was an abrupt drop in the PM<sub>10</sub> concentration on the day following *meji* event in 2012, which again

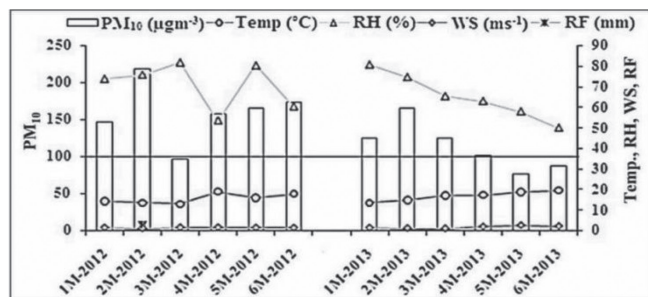


Figure 2: PM<sub>10</sub> concentration and meteorological parameters over the study period. 2M is the sample of *meji* burning. The solid line indicates 24-hr national standard of PM<sub>10</sub>.

built up on the next day and continued to remain high till the last sample was taken two weeks later. This fluctuation was not seen in the data for the same period in 2013. The former abrupt drop in PM<sub>10</sub> concentration could be attributed to rain scavenging of the aerosol on the day following *meji* burning event in 2012. The study station experienced a light rainfall (2.47 mm) and there after a long spell of dry period prevailed.

### Bulk Composition of PM<sub>10</sub>

The relative bulk composition of PM<sub>10</sub> has been shown in Figure 3. During *meji* burning period of two years ions and carbons were comparable between the years. Yet when we see the composition of *meji* day PM<sub>10</sub>, the abundance of carbon (43% in 2012 and 57% in 2013) and ions (29% in 2012 and 23% in 2013) appear to be much higher than the average conditions. Ram and Sarin (2011) had reported carbon contribution to PM<sub>10</sub> mass to be 37 and 49% for day and night samples respectively in Kanpur in the Indo-Gangetic Plain of India where biomass burning is the dominant source of PM<sub>10</sub>.

It is also reported that there is enhanced emission of ions such as K<sup>+</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> from biomass burning (Deka and Hoque, 2015). The ionic abundance in the *meji* day PM<sub>10</sub> was found too much above the average condition.

### Elemental Concentrations

The samples were chemically analyzed for eleven elements. A definite trend of elemental concentrations was missing. Generally, all elements were on the

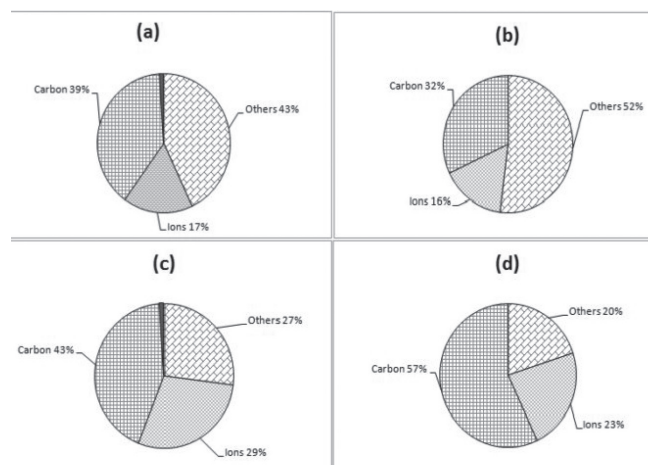


Figure 3: Relative contribution of elements, ions and carbon to PM<sub>10</sub> during (a) composition of PM<sub>10</sub> – average of *meji* burning period of 2012; (b) composition of PM<sub>10</sub> – average of *meji* burning period of 2013; (c) composition of PM<sub>10</sub> – *meji* burning day of 2012 and (d) composition of PM<sub>10</sub> – *meji* burning day of 2013.



higher side during the *meji* period of 2012 than of 2013 (Figure 4a, b). Pb and Ni have been notified as criteria air pollutants by Government of India in 2009. The national standard of 1  $\mu\text{gm}^{-3}$  (24-hr average) for Pb and 20  $\text{ngm}^{-3}$  (annual average concentration) for Ni have been set up by Government of India (CPCB, 2009). The concentration of Pb was found to be below the national standard in both the years; however, Ni was found to be two times higher ( $43.70 \pm 75 \text{ ng m}^{-3}$ ) than the national standard during the *meji* burning period of 2012. This comparison is inequitable because we are comparing a short term average Ni with the standard which is annual average. Yet, this high Ni concentration could not be overlooked. In a study (Deka and Hoque, 2015) on biomass fuel smoke particles of rural kitchens, Ca, K, Fe and Mg were the major contributing elements.

Concentrations of Na, Mn, Zn, Cr, Co, Ni and Cu were low.

Enrichment factors (EF) of the elements were calculated to assess extent of anthropogenic contribution to the elemental composition of the aerosol. Earlier studies (Reheis and Kihl, 1995; Nriagu and Pacyna, 1988; Rasmussen, 1998) suggest that an EF of an element of more than unity can be attributed to anthropogenic activities. Yet, some researchers keep a limit EF of 5 (e.g. Wu et al., 2007) or 10 (e.g. Yongjie et al., 2009; Kothai et al., 2011) to see the non-crustal or anthropogenic inputs; though EF is largely a function of particle size (Samara et al., 1990; Foltescu et al., 1996; Dietl et al., 1997).

EFs were calculated as per equation (1).

$$EF = (E/R)_{\text{PM}_{10}} / (E/R)_{\text{crust}} \quad (1)$$

where  $E$  is the elemental concentration and  $R$  is the reference element (Fe) in PM<sub>10</sub> and crust.

It was found that elements were more enriched in PM<sub>10</sub> during *meji* burning period of 2013 than 2012 (Table 1). The EFs of Zn and Pb were 145 and 656, respectively in 2013. The EF of Cd was found to be maximum—401 and 1505 during 2012 and 2013 respectively. Low enrichment of Co and Mn showed their crustal origin and there would have been anthropogenic inputs of Cr, Cu, Ni, Cd and Pb during the study period. Ielpo et al. (2016) found that the EF of K, Ca, Na, Zn and Cu was much bigger than 5 for outdoor and indoor samples of biomass fuel particulate deposits and ashes with outdoor values lower than indoor ones.

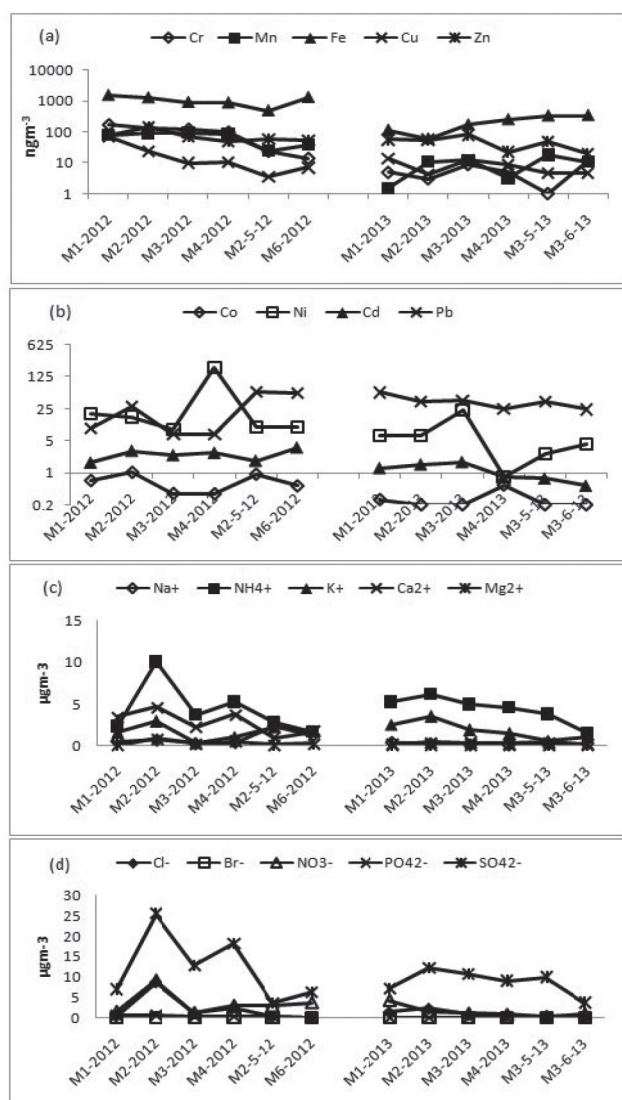


Figure 4: Elemental and ionic concentration over the study period. M2 is the sample of *meji* burning.

Table 1: Enrichment factors (EFs) of elements during 2012 and 2013 *meji* burning period

	2012	2013
Cd	400.96	1505.27
Co	0.63	1.19
Cr	49.51	49.92
Cu	30.46	38.96
Mn	2.37	1.27
Ni	10.28	27.11
Pb	37.36	656.36
Zn	39.43	145.32

### Water Soluble Ions

Ion chromatographic analyses of water extract of PM<sub>10</sub> samples were targeted to 12 major cations and anions viz. Li<sup>+</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> and F<sup>-</sup>, Cl<sup>-</sup>, Br<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup> and SO<sub>4</sub><sup>2-</sup> respectively. The levels of Li<sup>+</sup> and F<sup>-</sup> were found to be below detection limit in all the samples. The ratios of  $\sum$ anions to  $\sum$ cations were found to be much above unity and similar during both the *Meji* burning periods—1.87 (2012) and 1.68 (2013)—which would mean cation deficient conditions prevailed during both the years. Higher abundance of anions versus cations has been reported by other researchers from India (e.g. Singh et al., 2010; Ram and Sarin, 2011; Deshmukh et al., 2011; Sharma et al., 2012) and abroad (e.g. Lee and Hieu, 2013; Galindo et al., 2013).

Most of the measured ions were showing higher concentrations during *Meji* burning period of 2012 than 2013 (Figures 4c and 4d). Among the ions, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> were found to dominate in the *Meji* day samples. In another study (Deka and Hoque, 2015) on biomass fuel smoke particles of rural kitchens, the trend of average concentration of anions was found to be SO<sub>4</sub><sup>2-</sup> > Cl<sup>-</sup> > PO<sub>4</sub><sup>3-</sup> > NO<sub>3</sub><sup>-</sup> > F<sup>-</sup> > Br<sup>-</sup>. While in a study (Ielpo et al., 2016) on biomass fuel, particulate deposits and ashes in households of Mt. Everest region (Nepal), the measured ions were K<sup>+</sup>, Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>. Previous studies have put forward five major ions including K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> to be emitted by all types of biomass burning (e.g. Sillapapiromsuk et al., 2013; Yamasoe et al., 2000; Niemi et al., 2004; Park et al., 2004; Ryu et al., 2007). Ammonium and NO<sub>3</sub><sup>-</sup> may also originate from fertilizer use, fossil fuel combustion and animal waste (Coelho et al., 2011).

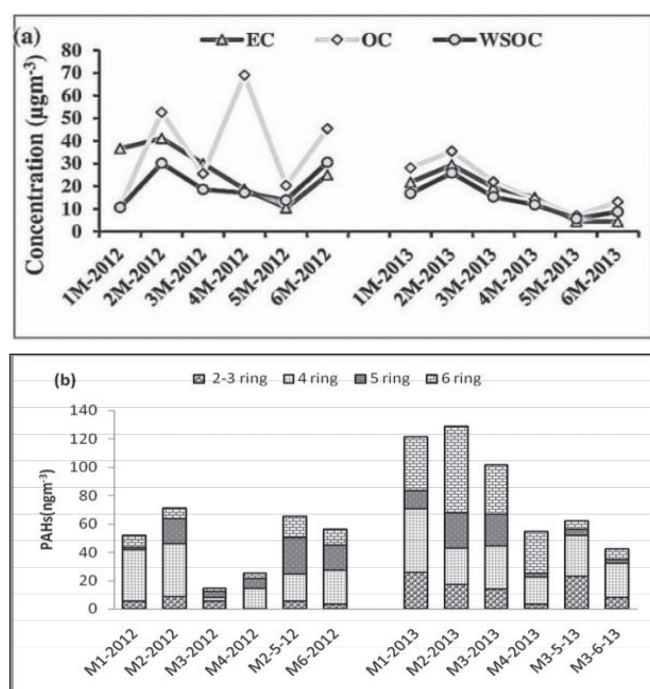
### Carbon

**Total Carbon:** In the present study we had analyzed both total and water soluble carbon of PM<sub>10</sub>. Concentrations of EC and OC with the ratio OC/EC have been put up in Table 2. The trends of EC, OC and WSOC found in the aerosol samples are illustrated in Figure 5a. The trends of EC and WSOC appear to be similar in both the years with the concentration maximum on the *Meji* burning day. The trend of OC varied between the years. In 2013 the trend of OC matched well with the trends of EC and WSOC, which was not the case in 2012. The OC in 2012 peaked abruptly after the *Meji* burning day. In fact, there was a light local rainfall in the study area during the experiment in 2012 and the sample following the rainfall exhibited higher OC. Would this mean that the light rainfall induced smoldering leading to more emission of OC (e.g. Mayol-Bracero et al., 2002; Andreae et al., 1996; Novakov and Corrigan, 1996)?

Submicron carbonaceous particles have an atmospheric residence time of 3-7 days (Cooke and Wilson, 1996; Cooke et al., 2002) and therefore are able to long range transport of hundreds to thousands of kilometres (Guazzotti et al., 2003). Guazzotti et al. (2003) had found predominance of submicron, chemically mixed particles containing abundant carbon and potassium, implying a biomass burning origin of aerosols in India. As mentioned earlier carbon shares 39 and 32% of PM<sub>10</sub> mass during *Meji* burning period of 2012 and 2013. Venkataraman et al. (2005) had estimated the relative contributions of fossil fuel combustion, open biomass burning and biofuel combustion to BC in India to be 25, 33 and 42%, respectively, whereas those to OC to be 13, 43

**Table 2: Concentration of different fractions of particulate carbon of PM<sub>10</sub> along with their ratios and a comparative account with studies elsewhere**

Location	Sampling period	TC	EC	OC	OC/EC	EC/TC	References
Tezpur, India	<i>Meji</i> , 2012 (Jan)	68.90±25	27.33±12	41.57±22	1.59±1	0.40±0.11	Present study
Tezpur, India	<i>Meji</i> , 2013 (Jan)	35.73±20	16.19±10	19.54±11	1.51±1	0.43±0.10	Present study
Chennai, India	Jan-Feb, 2007		6.5±3	9.1±4	1.5±0.47	0.42±0.08	Pavuluri et al., 2011
Kanpur, India	Jan 2007-March 2008		3.8±2	25.8±16	7.4±4		Ram et al., 2010
Uji, Japan	Nov-Dec, 1998	18	5.2	12.8	2.5		Holler et al., 2002
Beijing, China	Sept-Nov	30.1	8.9	21.2	2.4		Zhang et al., 2007
Hangzhou, China	Sept 2001-Aug 2002	25.47	4.06	21.41	5.27		Cao et al., 2009
Morogoro, Tanzania	March-Apr 2006		0.52	4.5	8.65		Mkoma et al., 2010



**Figure 5: (a) Carbonaceous species in PM<sub>10</sub> over the study period. (b) Ring-wise distribution (different coloured area) and  $\sum_{16}$ PAH concentration (column heights). 2M is the sample of *meji* burning.**

and 44%, respectively. Gustafsson et al. (2009) found yet larger contribution (46–68%) of different fractions of black carbon from biomass burning in South Asia based on radiocarbon analyses. Similarly, Ancelet et al. (2013) had found that carbonaceous species made up to 47% of PM<sub>10</sub> in a study conducted in Masterton, New Zealand, in a wood burning rural community. In another study, Stone et al. (2010) had also estimated a higher contribution from biomass burning (21%) than fossil fuel (4%) to OC in the Himalayas.

**Water Soluble Carbon:** Water soluble carbon contributed a major share to the total carbon (TC) of aerosols, which were  $32.63 \pm 12\%$  and  $42.40 \pm 12\%$ , respectively during 2012 and 2013 *meji* burning periods. Again, a high percentage of OC was water soluble. In 2012 and 2013 *meji* burning period, WSOC/OC percent ratio was found to be  $58.44 \pm 25\%$  and  $69.64 \pm 14\%$  respectively. Mkoma et al. (2010) had reported 31% WSOC/OC percent ratio of PM<sub>10</sub> in a wet season campaign in Morogoro, a rural site of Tanzania. They had also concluded that the particulate OC emerged from biofuel and charcoal burning. In another study, Mayol-Bracero et al. (2002) reported 56% of TC to be WSOC. They had also found that the dominant fraction of the carbonaceous aerosol derived from biomass burning (smoldering) is organic, and a significant fraction of

it is water soluble and can therefore contribute to the CCN activity of biomass smoke particles. The high water-soluble fraction suggests an aerosol produced mainly by smoldering processes (Andreae et al., 1996; Novakov and Corrigan, 1996). Novakov and Corrigan (1996) provided strong evidence that water-soluble organic species in smoke particles from smoldering combustion could be responsible for their CCN activity.

The WSOC/OC ratios were  $0.50 \pm 0.25$  and  $0.70 \pm 0.14$ , respectively during 2012 and 2013 *meji* burning periods. Yang et al. (2005) had reported this ratio to be 0.45 at a high altitude site (Purple Mountain Observatory) in China. Again, Ram and Sarin (2010) had reported a value of  $\sim 0.5$  at a high altitude site (Manora peak) which emphasizes the significance of secondary organic aerosols. Pathak et al. (2013) had found dominance of secondary organic aerosols in OC concentrations in the upper Brahmaputra Valley of Assam which besides from biomass burning and anthropogenic sources may also result from the large biogenic volatile organic carbon emissions from the tea gardens and vegetation surrounding the study location.

The WSIC contributed very small portion to WSTC having mass concentration of  $1.41 \pm 1 \mu\text{g m}^{-3}$  and  $0.67 \pm 0.13 \mu\text{g m}^{-3}$ , respectively during 2012 and 2013 *Meji* burning periods. Generally, the main constituents of WSIC are carbonate and hydrogen carbonate, which are formed by crustal species and emission of industries (Wang et al., 2002), which could have been in low levels in aerosols of *Meji* burning periods.

#### PAHs

The PAHs of the present study constituted  $\sim 0.11\%$  ( $182.74 \pm 86 \text{ ng m}^{-3}$ ) and  $0.27\%$  ( $311.55 \pm 122 \text{ ng m}^{-3}$ ), respectively during 2012 and 2013 *Meji* burning periods. Earlier, Ancelet et al. (2013) reported average PAHs (16 USEPA priority PAHs and Retene) of  $38.9 \pm 26 \text{ ng m}^{-3}$ , accounting for 0.3% of the PM<sub>2.5</sub> in a study conducted among wood burning rural community of Masterton, New Zealand. They found FLA and PYR as the most abundant particle-phase PAHs, with concentrations ranging from  $13.5\text{--}0.6 \text{ ng m}^{-3}$  and  $12.6\text{--}0.5 \text{ ng m}^{-3}$ , respectively. In our study CHR was maximum in both the years, with a mean of  $54.53 \pm 44$  and  $68.57 \pm 28 \text{ ng m}^{-3}$ , respectively during 2012 and 2013 *Meji* burning periods. The average indoor concentration of chrysene is substantially higher than the ash and outdoors ones in a study (Ielpo et al., 2016) on biomass fuel particulate deposits and ashes in households of Mt. Everest region (Nepal).



Out of 16 USEPA priority PAHs, seven members have been considered as potential carcinogens viz., BaA, BaP, BbF, BkF, CHR, DBA and IND. These carcinogenic PAHs (CPAHs) contributed a very high percentage of total PAHs in both years. Carcinogenic PAHs were contributing  $68.24 \pm 16$  and  $56.17 \pm 13\%$  of  $\sum_{16}$ PAHs during 2012 and 2013 *meji* burning periods. Strong correlations were observed between  $\sum$ CPAHs and  $\sum_{16}$ PAHs ( $R^2 = 0.98$  and  $0.80$ , respectively). This suggests that  $\sum_{16}$ PAHs distribution was strongly influenced by the presence of  $\sum$ CPAHs. The profile of the PAHs by the ring sizes varied between the years. The ring-wise distribution was in the order  $4- > 5- > 6- > 2\&3$  ring PAHs in 2012 and  $6- \sim 4- > 2\&3- > 5$  ring PAHs during 2013 *meji* burning periods (Figure 5b).

The Government of India too regulates BaP and has set up ambient air standard of  $1 \text{ ngm}^{-3}$  (annual average) (CPCB, 2009). The BaP concentrations in the present study were found to be  $11.62 \pm 11$  and  $0.73 \pm 0.50 \text{ ngm}^{-3}$  in 2012 and 2013 periods respectively.

For better parameterization of carcinogenicity BaP-equivalents ( $B[a]P_{eq}$ ) were calculated as per equation (2).

$$\text{Total } B[a]P_{eq} = \sum i (C_i \times TEF_i) \quad (2)$$

The  $C_i$  is the concentration of an individual PAHs and  $TEF_i$  is the corresponding Toxic Equivalence Factor. TEF was taken from Nisbet and LaGoy (1992). The Total  $B[a]P_{eqs}$  were found to be  $29.51 \text{ ngm}^{-3}$  and  $59.85 \text{ ngm}^{-3}$ , respectively for 2012 and 2013 *Meji* burning periods.

## Source Signatures

### Mass Concentration Ratio of EC/TC and OC/EC

The EC/TC ratio gives clues on the sources of the carbonaceous aerosol. Low EC/TC ratios may indicate that the carbonaceous aerosol originates mainly from biogenic aerosols and/or biomass burning (Maenhaut and Claeys, 2007).

The EC/TC ratios were found to be  $0.40 \pm 0.11$  and  $0.43 \pm 0.10$ , respectively during 2012 and 2013 *Meji* burning periods (Table 2). Ferek et al. (1998) had reported BC/TC (here EC/TC) ratio of  $0.10 \pm 0.03$  in a biomass burning episode from Brazil. Again, this ratio was reported as  $0.12 \pm 0.07$  by Cachier (1991) in Ivory coast during a biomass burning episode. However, the EC/TC ratio obtained in this study is similar to that of Pavuluri et al. (2011) (Table 1) who found biomass burning a major source of atmospheric aerosols in South and Southeast Asia.

The ratio of particulate OC/EC is also an important indicator that reflects source type and source strength (Blando and Turpin, 2000). Other researchers had also used the ratio of OC to EC for source assessment (e.g., Ram et al., 2010; Pavuluri et al., 2011). Consistent patterns of OC/EC ratio can be more easily found partly because the ratio of two chemicals tend to be less sensitive to atmospheric processing (e.g., McKeen and Liu, 1993; Wang and Zeng, 2004). Long-range transport of fire emitted OC and EC, for example, do not have a large impact on the absolute levels of OC due to dilution but can significantly alter the OC/EC ratio in remote regions. OC/EC ratio in this study viz.,  $1.59 \pm 0.77$  and  $1.51 \pm 0.77$ , respectively were less than reported studies (Table 2) except that of Pavuluri et al. (2011).

The EC/TC and OC/EC ratios of the present study suggest high concentration of EC compared to typical biomass burning study. EC emission from biomass burning depends on burn rate ( $\text{kg h}^{-1}$ ) and type of biomass (Habib et al., 2008; Stone et al., 2010). Stone et al. (2010) reported different EC/OC ratio at high and low burn rates. For mango wood, the reported ratio was 1.6 for the low burn rate compared to 0.2 for the high burn rate, whereas acacia wood at low and high burn rates were 0.3-0.4. Researchers had concluded that EC/OC ratios alone cannot be used to differentiate between combustion sources (Habib et al., 2008; Venkataraman et al., 2005).

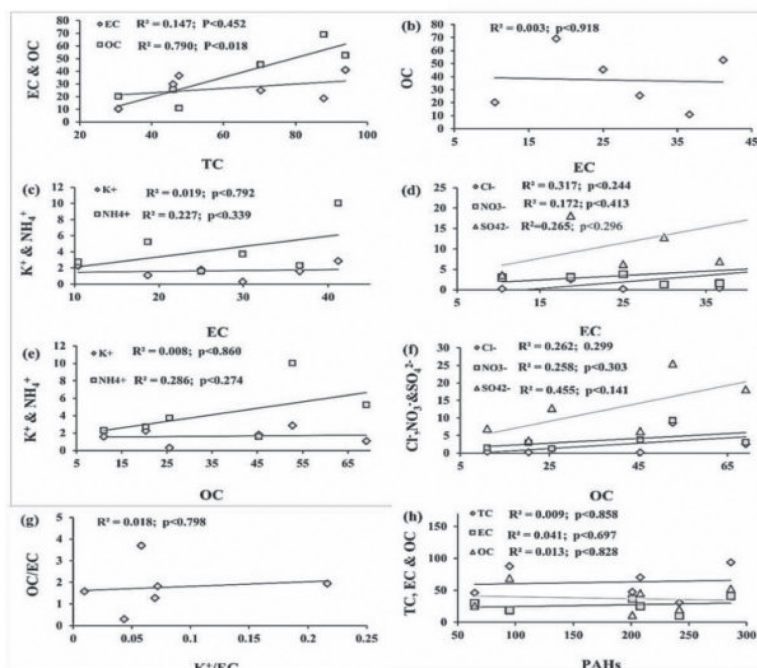
### Relationships of EC and OC with Other Biomass Burning Marker Species

Linear correlations were established between different carbonaceous components and marker species of biomass burning (Figures 6-I and 6-II). Pavuluri et al. (2011) had concluded biofuel/biomass burning as a major source of atmospheric aerosols in South and Southeast Asia on the basis of mass concentration ratios of selected components and relations of EC and OC to marker species.

Positive correlations were observed between OC and TC [Figures 6-I (a) and 6-II (a)], EC and TC [Figures 6-I (a) and 6-II (a)], OC and EC [Figures 6-I (b) and 6-II (b)], OC/EC with  $K^+/EC$  [Figures 6-I (g) and 6-II (g)] suggesting cogenic emission. Different ionic ratios were compared with sea water ratios which were not significant. Therefore, non sea-salt (nss) fractions of individual ions were not calculated to correlate the linear relations between ions and carbon. Ions emitted from biomass burning show positive relations with EC and OC. These ions, especially  $NH_4^+$ ,  $K^+$  and  $Cl^-$ , showed good correlations with OC and EC in the present study



(I) 2012 *meji* burning event



(II) 2013 *meji* burning event

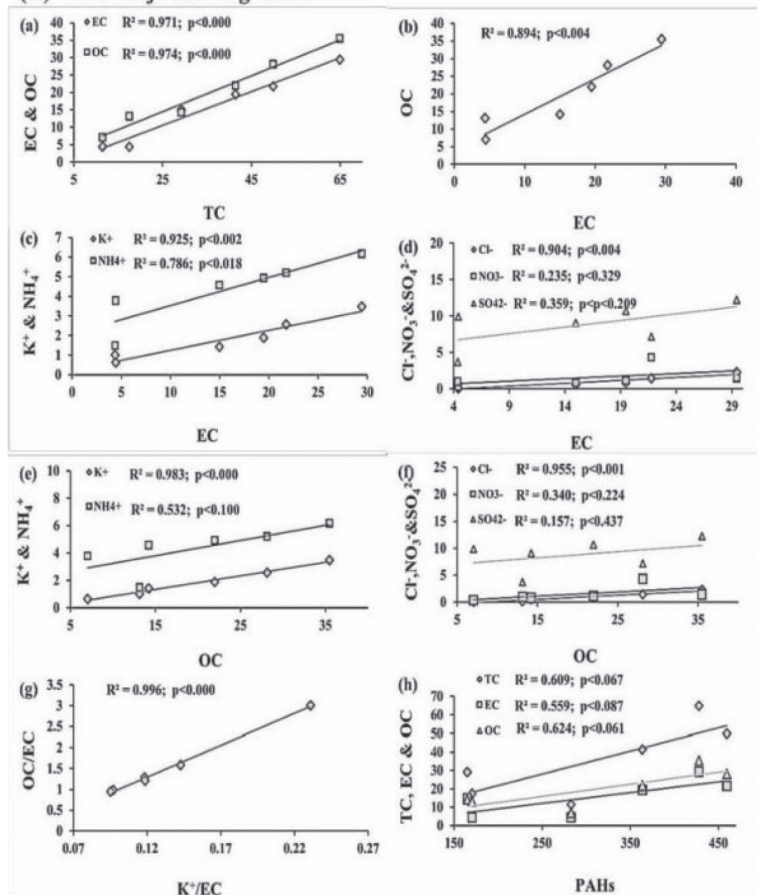


Figure 6: Linear relationships between different chemical species – (I) *Meji* burning period of 2012 and (II) *Meji* burning period of 2013.

[Figures 6-I (c), (d) and 6-II (c), (d)]. Correlations between OC, EC and  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were weaker than other ions [Figures 6-I (d) and 6-II (d)] indicating contribution of some other sources to  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  loading of  $\text{PM}_{10}$ .

Most of the combustion-derived PAHs are associated with particles such as BC and soot (Lima et al., 2005).  $\Sigma_{16}$ PAHs showed positive correlation with carbon over the study period indicating predominance of combustion derived PAHs [Figures 6-I (h) and 6-II (h)]. It is interesting to note that the correlation of these species was positive but relatively weaker in 2012 in comparison to 2013 *Meji* burning period. Most of the studied correlations were very good during 2013. Occurrence of rain in between the experimental period in 2012 could have resulted in washout of submicron particles from the atmosphere and, therefore, could be the most probable reason for weaker correlation between different chemical species of 2012 *Meji* burning period.

#### Molecular Diagnostic Ratio of PAHs

Molecular diagnostic ratios are used by the researchers to apportion the sources of atmospheric PAHs. The accurate use of diagnostic ratios depends primarily on the uniqueness of the fingerprint of the sources (Lima et al., 2005). Molecular markers are organic

species present in the ambient atmosphere that come from specific aerosol source categories, are stable in the atmosphere, and can be used quantitatively in source apportionment (Schauer and Cass, 2000). Some diagnostic ratios were calculated and compared with studies elsewhere on different types of biomass burning (Table 3). The calculated ratios were almost similar to other biomass burning studies with some exceptions. These exceptions might be due to the difference in other contributing sources of  $\text{PM}_{10}$ .

### Conclusions

The effect of *meji* biomass burning of the  $\text{PM}_{10}$  is experienced for a short period, which tends to faint within a week. Except PAHs, elements, ions and carbon were higher during 2012 than that of 2013 *meji* burning period. Most of the water soluble ions and carbon were maximum on *meji* burning day. Sulphate was contributing maximum to analyzed ions followed by ammonium in both years. High WSOC emphasize on CCN activity of organic aerosols along with the presence of smoldering phase during *meji* burning period. WSOC/OC ratio revealed dominance of secondary organic aerosols in the study region.

**Table 3: Comparative account of the molecular diagnostic ratios of the present study with different biomass burning studies**

	2012 <i>Meji</i> <sup>1a</sup>	2013 <i>Meji</i> <sup>1b</sup>	Paddy residue- ambient <sup>2a</sup>	Paddy residue- chamber <sup>2b</sup>	Crop residue/ stove <sup>3</sup>	Crop residue/ stove <sup>4a</sup>	Crop residue/ open fire <sup>4b</sup>	Wood/ stove <sup>4c</sup>
Phe/Ant	7.47±4.64	2.57±4.86						
ANT/ (ANT+PHE)	0.08±0.05	0.28±0.30	0.15±0.03	0.17±0.01	0.12±0.01	0.2	0.17-0.25	0.10-0.30
FLA/PYR	1.88±1.81	1.17±1.06	0.84±0.04	0.97±0.21				
FLA/ (FLA+PYR)	0.06±0.03	0.47±0.18	0.46±0.01	0.49±0.05	0.53±0.03	0.51-0.80	0.34-0.53	0.43-0.74
IND/ (IND+BPER)	0.58±0.29	0.84±0.08	0.49±0.03		0.54±0.02	0.31-0.50	0.39-0.94	0.16-0.69
Bap/BaP+BPER	0.70±0.16	0.12±0.09			0.60±0.05	0.23-0.67	0.43-0.98	0.38-0.78
BbF/BbF+BkF	0.63±0.45	0.61±0.39			0.55 ± 0.03	0.50-0.65	0.35-0.80	0.35-0.51
BaA/BaA+CHR	0.43±0.16	0.26±0.21			0.48 ± 0.02	0.46	0.39-0.50	0.39-0.56

1 – present study

2 – Rajput et al., 2011

3 – Shen et al., 2011

4 – Shen et al. (2011) and references therein

Linear relationship between EC, OC and other chemical species along with molecular diagnostic ratios of selected PAHs indicates strong influence of biomass burning on PM<sub>10</sub> load.  $\Sigma$ CPAHs were contributing a major portion of  $\Sigma_{16}$ PAHs and their strong correlation with  $\Sigma_{16}$ PAHs suggests that PAHs distribution was strongly influenced by the presence of  $\Sigma$ CPAHs. The results strongly support our contention that in the mid-Brahmaputra Valley the annual socio-cultural practice of festive biomass burning contributes significantly to atmospheric PM<sub>10</sub>. Chemical characterization of aerosols further endorsed the strong influence of *Meji* burning on PM<sub>10</sub> load and its properties.

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