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Research Paper

Quantification of Organic Carbon from Biomass Versus Non-Biomass Burning Emissions to Fine Aerosol

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In the present study, size-segregated aerosol samples (in five size ranges) were collected from Patiala (30.33°N, 76.40°E; 250m amsl), located in the northwestern Indo-Gangetic Plain (IGP) during October 2012 to September 2013 to quantify the contribution of biomass burning (BB) to ambient organic carbon (OC) in fine aerosol. All of these samples were analyzed for major inorganic ions, whereas, particulate matter smaller than 1 µm aerodynamic diameter (PM_1) were also analyzed for carbonaceous aerosol (OC, elemental carbon (EC), and water-soluble organic carbon (WSOC)). The contribution of BB derived potassium (K^+_{BB}) to ambient fine K^+ is found to be dominant (77±24%) over an annual cycle. Further, regression parameters of OC and K^+_{BB} linear relationships are used to quantify the contribution of BB to ambient OC in PM_1. The contribution of BB derived OC (OC_{BB}) to fine OC was significantly higher during autumn (69%), winter (54%) and summer (40%) seasons in comparison to that during spring (26%) and wet (2%) seasons with the annual average of 41±30%. Emissions from agricultural-waste burning during autumn and summer, and from bio-fuel (Babool, Cowdung cake, Eucalyptus, Jujube and Shisham) burning during winter are the major sources of ambient OC over the study region. The contribution of BB derived water-soluble organic carbon ($WSOC_{BB}$) to fine WSOC exhibited similar seasonal variation as that of OC_{BB} to fine OC. It was 75%, 55%, 14%, 44% and 5% during autumn, winter, spring, summer and wet seasons respectively, with the annual average of 43±32%. These results have implication in climate modelling, health impact assessment and planning effective mitigation strategies.

Keywords: Potassium; Tracer; OC; WSOC; Agriculture-waste Burning, India

Introduction

Biomass burning (BB) is among the major sources of atmospheric pollution with direct short- and long-term climate implications over many regions of the world (Kaskaoutis *et al.,* 2014 and references therein). It is important to quantify BB contribution to aerosol loading over different geographical regions. On the global scale, forest fires and crop-residue burning account for nearly ~90% of the total wild land fires (Barnaba *et al.,* 2011). In Asia, the emissions from forest fire and crop-residue burning are big contributors (~45% and ~34%) to the total BB emissions (Streets *et al.,* 2003). India is the second highest crop-residue burning (~84 Tg yr^{-1}) country in Asia (Streets *et al.,* 2003), and crop-residue burning in northern India [northwestern Indo-Gangetic Plain (IGP)] contributes nearly ~20% to the total global emission budget of carbonaceous aerosol (Rajput *et al.,* 2014a). However, the role of BB in the mitigation policies is often overlooked due to lack of reliable data on their source strength. Several studies over the IGP suggest that BB is the largest source of carbonaceous aerosol (Rengarajan *et al.,* 2007; Ram *et al.,* 2012; Rastogi *et al.,* 2014; Singh and Kaskaoutis, 2014; Choudhary *et al.,* 2017; Satish *et al.,* 2017; Kumar *et al.,* 2018; Sharma *et al.,* 2018); however, studies on the quantification of contribution of BB to ambient organic carbon (OC), and water-soluble OC (WSOC) over the northwestern IGP are scarce. On the global scale, a number of studies had quantified the contribution of BB to ambient OC using

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radiocarbon, levoglucosan and biomass burning derived K^+ (K^+_{BB}) (Gelencser *et al.,* 2007; Gilardoni *et al.,* 2009; Gustafsson *et al.,* 2009). Quantification by radiocarbon is well established technique however; its measurement is very tedious, time consuming, and costly as compared to the measurements of levoglucosan and potassium. Levoglucosan can be used as a unique tracer for BB (Jordan *et al.,* 2006), but it has some limitations because of being unstable in the atmosphere for longer time (Kuo *et al.,* 2008), and relatively difficult to measure in comparison to K^+. Although measurement of K^+ is relatively easy but it is a debatable tracer for identification of BB because there could be multiple sources of K^+ in the atmosphere such as BB, mineral dust, sea-salts etc. (Pachon *et al.,* 2013 and references therein). Fine fraction of water-soluble K^+ can be used as a biomass burning tracer but one should be cautions when applying to coarse particulates (Reche *et al.,* 2012). However, K^+ has been established as a tracer of BB over the IGP by several studies (Ram and Sarin, 2011; Rajput *et al.,* 2014b; Singh *et al.,* 2014; Rastogi *et al.,* 2016).

Major objectives of the present study are to assess the contribution of BB to ambient fine K^+ (unlike any of the reported study from this region), and to estimate the contribution of BB to loading of OC and WSOC in PM_1 (hereafter also referred as fine aerosol).

Experimental Approach

The present study has been carried out over Patiala (30.33°N, 76.40°E; 250 m above mean sea level; Fig. 1), which is located in the northwestern part of the IGP during October-2012 to September 2013. Study region experiences two distinct types of large scale agricultural-waste burning episodes i.e., paddy-residue burning during October-November and wheat-residue burning in the month of May (Singh *et al.,* 2016a). Red dots in Fig. 1 (captured with Moderate Resolution Imaging Spectroradiometer (MODIS) onboard polar orbiting NASA's satellite during end of the October month) denote the fire counts, which indicates that high BB activities were present over the study region during that time. For complete description about the sampling site and prevailing emission sources, reference is made to our earlier publications (Rastogi *et al.,* 2015; Singh *et al.,* 2016a; Rastogi *et al.,* 2016).

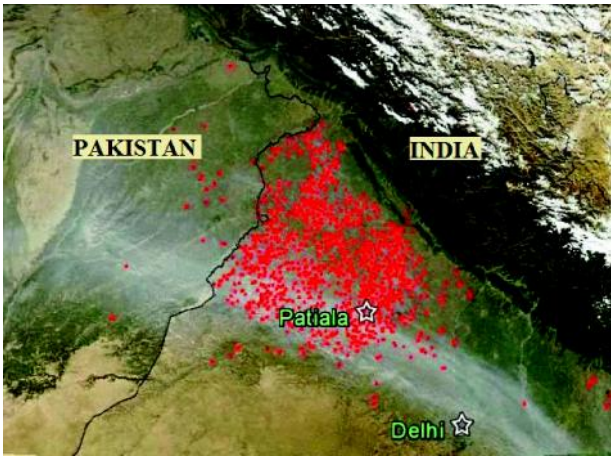

Fig. 1: Map showing sampling location Patiala (Source: Google Erath &MODIS NASA)

Aerosol samples were collected using 4-stage cascade impactor attached with high volume air sampler (Staplex, USA; Model: 234) at the air flow rate of 1.1 m^3 min^{-1} with sampling time from 09:00-18:00 hrs Indian Standard Time (IST). A total of 52 samples have been collected over an annual cycle and each sample have 5 filters corresponding to the particle collection at five different size ranges i.e., $PM_{<0.95}$ (~PM_1), $PM_{0.95\text{-}1.5}$, $PM_{1.5\text{-}3.0}$, $PM_{3.0\text{-}7.2}$ and $PM_{>7.2}$. Therefore, the number of analyzed samples were 260 (52x5). Depending upon the prevailing meteorological conditions over the year, data sets have been classified into five seasons named as autumn (October–November), winter (December–February), spring (March–April), summer (May–June) and wet (July–September), as explained in our earlier publication (Rastogi *et al.,* 2016). The concentrations of K^+ along with other cations (Na^+, NH_4^+, Ca^{2+} and Mg^{2+}) and anions (Cl^-, SO_4^{2-} and NO_3^-) were measured in all the five size ranges with dual channel ion chromatograph (Dionex, USA; Model: ICS-5000). In PM_1, mass concentrations of elemental carbon (EC) and OC were measured on EC-OC analyzer (Sunset Laboratory, USA; Model: 2000), and that of WSOC on TOC analyzer (Shimadzu, TOC-5000A). Based on replicate analyses of samples, the precision of measurements for water-soluble ionic species, OC and WSOC was found to be 5%, 8% and 4%, respectively. For complete description about the size-segregated aerosol sampling and their chemical analysis reference is made to our earlier publications (Singh *et al.,* 2016a and 2016b). The average values

of meteorological parameters, measured by automatic weather station of Indian Meteorological Department (IMD) observatory, Patiala are summarized in Table 1.

Results and Discussion

During observation period (October 2012 to September 2013), mass concentration of total suspended particulates (TSP; derived by summation of aerosol mass in different size ranges) varied from 88 to 387 $\mu g\ m^{-3}$ with higher concentration (average±sd.;±1σ) during autumn (267±69), summer (227±77), and winter (179±55) seasons and relatively lower concentration during spring (137±38), and wet (130±62) seasons. Size-segregated aerosol mass distribution analysis showed that aerosol mass was most abundant (~55%) in fine mode (PM_1), followed by much lower contribution in $PM_{3.0\text{-}7.2}$ (16%), and $PM_{>7.2}$ (13%), and least in $PM_{0.95\text{-}1.3}$ (8%) and $PM_{1.5\text{-}3.0}$ (8%) over the year. High abundance of fine aerosol broadly reflects relatively higher contribution of anthropogenic combustion sources to ambient aerosol loading, as aerosol produced by combustion sources are relatively small in size as compared to natural aerosol produced by physical mechanism (Seinfeld and Pandis, 2006). During autumn, emissions from large scale paddy-residue burning in the open fields are the major source of aerosol loading while during winter, remnant of crop-residue burning and emissions from thermal power plants, bio-fuel (such as Cowdung cake, wood sticks from Babool, Eucalyptus, Jujube and Shisham) and fossil fuel combustion coupled with shallow boundary layer lead to high aerosol loading (Rajput *et al.,* 2011; Singh *et al.,* 2016c). Occurrence of occasional dust storms along with high speed winds and dry atmospheric conditions may cause high concentration of particulates during spring and summer season (Singh *et al.,* 2015). Additionally, emissions from wheat-residue burning during the first half of May (summer) are additional sources of aerosol loading during summer (Singh *et al.,* 2016b). These observations indicate that both anthropogenic as well as natural sources are responsible for high aerosol loading over the site, as reported in our earlier publication (Singh *et al.,* 2015).

Use of Potassium as A Biomass Burning Tracer

Overall, total mass concentration of K^+ (sum of concentrations in all the five sizes) varied from 0.06 to 5.3 $\mu g\ m^{-3}$ with highest concentration (average ± sd.; ±1σ) during autumn (3.0±1.2), followed by a decreasing trend from winter (1.6±0.74), summer (0.64±0.40), spring (0.22±0.14) to wet (0.19±0.14) season. The K^+ exhibited high concentration during those seasons when the emissions from agricultural-waste burning (paddy-residue and wheat-residue during autumn and summer, respectively) and bio-fuel burning (during winter) were dominant.In PM_1, a strong linear correlation between K^+ and OC (r=0.93; p<0.0001; Fig. not shown) suggests that the emissions from BB is a dominant source of fine K^+. The K^+/OC ratio in PM_1 variedfrom 0.01 to 0.11 (0.05±0.03). The K^+/OC ratio can be used as characteristics of species emitted from different type of biomass burning such as agricultural-waste and bio-fuel. These ratios are comparable to earlier reported ratio of 0.06 over different locations of the IGP (Ram and Sarin, 2011; Rastogi *et al.,* 2014) and tropical forest (0.06, Andreae and Merlet, 2001). However, this ratio is higher than that reported for bio-fuel burning (0.01) and lower than that reported for charcoal burning (0.08) (Andreae and Merlet, 2001). In addition, earlier studies have suggested relatively high OC/EC ratio for carbonaceous aerosol coming from BB emissions as compared to those from vehicular emissions (fossil fuel emissions) (Sandradewi *et al.,* 2008). In the present study, the temporal variability of fine mode K^+ and total concentration of K^+ has shown same trend as exhibited by OC/EC mass ratio, which further confirms the hypothesis that ambient K^+ over the study region is mainly associated with the BB emissions.

The study region is surrounded by semi-arid region and thus, dust could also be a possible source of ambient K^+ along with BB emissions as there is reasonably significant correlation (r=0.65; p<0.0001) between Ca^{2+} and K^+ (Fig. not shown). Pio *et al.* (2008) proposed a method to distinguish the signal of K^+ from BB emissions and dust. They suggested that the enrichment of K^+ in the $PM_{2.5}$ is an indicator of dominant contribution of K^+ from BB emissions. Thus, to assess the potential source (BB or dust) of ambient K^+, fraction of fine mode K^+ (in PM_1) in total K^+ was investigated. The temporal variation in the mass ratio of fine K^+ to total K^+ is shown in Fig. 2A, which depicted that around 80% (period average) of the total concentration is in fine mode. This observation reveals the dominance of BB derived K^+ in comparison to that from natural source (dust) over the study region.

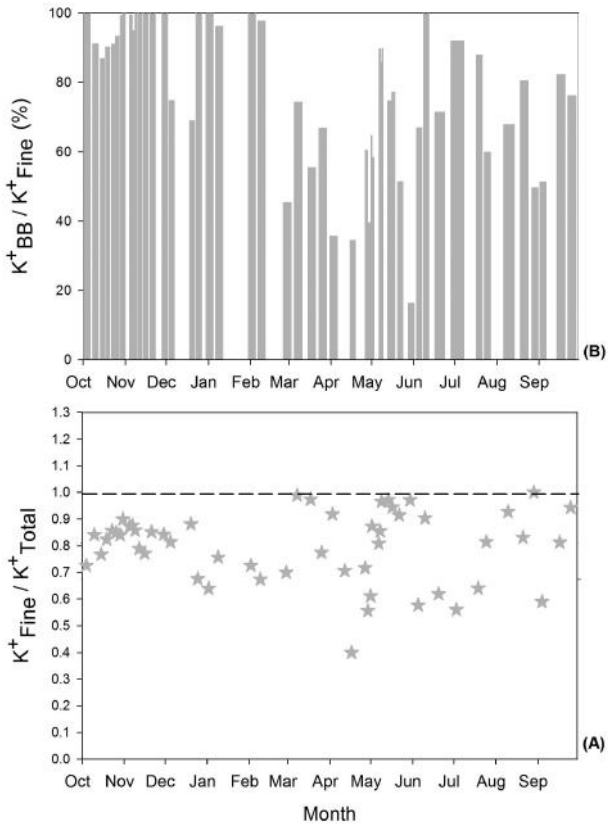

Fig. 2: Temporal variability in the mass ratio of (A) fine potassium in total potassium and (B) biomass burning derived potassium (K^+_{BB}) in fine potassium (K^+_{Fine})

The concentration of K^+ corrected for contributions from the dust and sea-salts, has been used as a tracer for BB emissions (Pio *et al.*, 2008). The relationship between Ca^{2+} and K^+ in fine mode (PM_1) has been explored to estimate the concentration of K^+ not related with mineral dust (Fig. is not shown). Here, the lowest ratio 0.60 is considered as the K^+/Ca^{2+} ratio in dust. Anything above this ratio reflect the contribution of K^+ from non-dust sources. Furthermore, this K^+/Ca^{2+} mass ratio has been used in equation no. (1) to derive the contribution of K^+_{BB} to fine K^+.

$$(K^+_{BB})_{Fine} = K^+_{nss} - 0.60(Ca^{2+}_{nss} - Ca^{2+}_{BB}) \quad (1)$$

Here, K^+_{BB}, K^+_{nss}, Ca^{2+}_{nss} and Ca^{2+}_{BB} refer to BB derived potassium, non-sea salt (nss) potassium, nss-calcium and BB derived calcium, respectively in PM_1. Sea-salt contribution to fine aerosol loading is insignificant over the study region as it is very far from sea shore, and same is reflected in trace amount of measured Na^+ (annual average=0.15 µg m^{-3}). So, nss fraction of potassium and calcium are practically represented by the total measured concentration of potassium and calcium in PM_1. Temporal variation in the mass ratio of K^+_{BB} to the concentration of fine K^+ is shown in Fig. 2B. All the samples collected during paddy-residue burning period (October-November) showed K^+_{BB}/K^+ ratio well above 0.9, and variable fraction has been observed during other months of study. Seasonally, the contribution of to fine K^+ was high during autumn (96%), winter (85%), wet (72%) and summer (70%) seasons, and ~46% during spring season. Significant contribution of to fine K^+ during autumn and summer is attributed to paddy- and wheat-residue burning in the open fields; whereas during winter, it is mainly due to bio-fuel burning and remnant from the paddy-residue burning. Lowest ratios during spring are ascribed to long-range transported dust from the Thar and Arabian deserts. On annual scale, the contribution ofto fine K^+ is found to be dominant (77 ± 24%) over the study region. Earlier study by Pachon *et al.* (2013) in rural area of the Atlanta, a site mainly influenced by BB emissions, also showed that significant fraction (82%) of K^+ comes from BB. Present study indicates that the dominant source of K^+ is BB emissions, and it is reasonable to use fine K^+ as BB tracer over the northwestern part of the IGP.

Quantification of Organic Carbon from BB Emissions

The mass concentrations of OC and WSOC in PM_1 ranged from 2.8 to 60 (17±13) and 1.9 to 33 µg m^{-3} (10±8), respectively over an annual cycle. Concentrations of both OC and WSOC are found to be highest during autumn, followed by winter, summer, spring and minimum during wet season (Table 1). Detailed description of temporal trend in the mass concentrations of OC and WSOC are provided in our earlier publication (Singh *et al.*, 2016a). The concentration of in PM_1 has been used to quantify the BB derived OC (OC_{BB}) in PM_1 by literature-based method (Gilardoni *et al.*, 2009). A linear relationship between OC and K^+_{BB} has been investigated (Fig. 3A), and it was found that the intercept of linear fit is statistically non-zero (7.4 µg m^{-3}), indicating the additional source of OC that does not belong to BB. Therefore, the difference between ambient OC and

Table 1: Seasonal mean values of temperature (Temp), relative humidity (RH), organic carbon (OC) and water-soluble organic carbon (WSOC) in PM_1

Seasons	Temp (°C) (Avg ± 1σ)	RH (%) (Avg ± 1σ)	OC (μg m^{-3})	WSOC (μg m^{-3})
Autumn	27 ± 3	56 ± 10	31 ± 15	20 ± 7.6
Winter	17 ± 4	71 ± 15	18 ± 5.5	9.8 ± 2.6
Spring	30 ± 3	49 ± 11	10 ± 1.7	4.7 ± 1.2
Summer	38 ± 3	54 ± 13	14 ± 6.2	8.3 ± 3.1
Wet	34 ± 2	76 ± 09	5 ± 1.8	3.3 ± 1.2

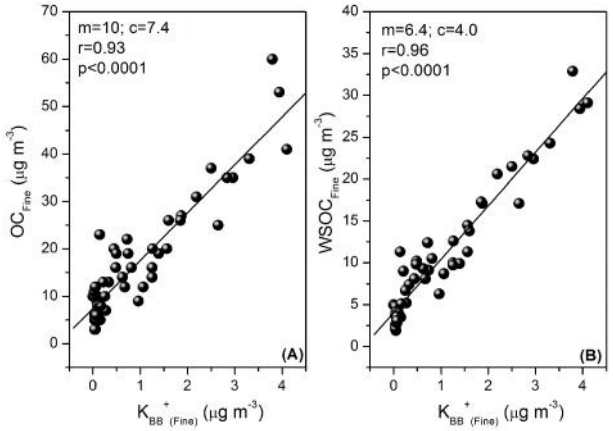

Fig. 3: Relationship between biomass burning derived fine potassium (K^+_{BB}) and (A) mass concentration of fine organic carbon (OC_{Fine}), and (B) fine water-soluble organic carbon ($WSOC_{Fine}$)

intercept would give the OC_{BB}. Here, it is relevant to state that accurate estimation of OC_{BB} using the current approach is difficult because of several assumptions involved, as also suggested by Pio *et al.* (2008). Furthermore, intercept (in μg m^{-3}) is subtracted from all the data points to estimate OC_{BB} which may lead to relatively large error for samples with low concentrations of OC, usually observed during wet season.

Estimated BB contribution to ambient fine OC varied between 0 and 88% (annual average: 41±30%) (Fig. 4A) with higher contribution during autumn (69%), winter (54%) and summer (40%) seasons, and relatively lower contribution during spring (26%) and wet (2%) seasons, suggesting BB is a significant source of ambient fine OC over the study region especially during autumn, winter and summer seasons. The emissions from large scale paddy-residue burning during autumn and the remnant of crop-residue burning along with emissions from bio-fuel burning during winter are the major factors for high contribution of BB to ambient OC during these seasons. During summer (in May), it is due to wheat-residue burning (Rastogi *et al.*, 2016; Singh *et al.*, 2016a). Relatively lower contribution of BB to ambient OC during spring and wet seasons is mainly due to absence of any large scale agricultural-waste burning emissions. Over an annual scale, fraction of K^+_{BB} to fine K^+ is 77% but it is not reflected in OC, as contribution of OC_{BB} to total OC is 41% over an annual cycle. It is due to use of primary tracer in quantification of OC_{BB}, where OC_{BB} mainly represents the fraction of primary organics from BB. However, the actual contribution of BB to ambient OC may be high if secondary OC from BB is also included (Singh *et al.*, 2016b). A study by Jung *et al.* (2014) over Daejeon, Korea, has reported relatively lower contribution of BB to OC (45%) as compared to present study (73%) during the paddy-residue burning period (from mid of October to mid of November).

Recently, Srinivas *et al.* (2016) had reported $\Delta^{14}C$ based BB fraction ($f_{BB\text{-}TOC}$) to total OC in TSP as 0.81±0.03 (range: 0.76 to 0.87) over Hisar (at a distance of ~180 km in the southwest of study region) during December month. Hisar also represents a typical semi-urban atmosphere surrounded by agricultural fields like the present study region and situated downwind of sampling site during winter. Present study estimates the contribution of BB to OC varying from 0.40 to 0.71 (avg: 0.54 ± 0.18) in PM_1 during winter (December to February), which is lower than that reported by Srinivas *et al.* (2016) in TSP (0.81±0.03). Quantification in present study is based on the PM_1samplescollected only during daytime, unlike Srinivas *et al.* (2016). Relatively high contribution of BB to OC abundances had been reported for samples collected during nighttime in comparison to those collected during daytime, because local inhabitants generally burn biomass (bonfire) in the late evening/night during winter to keep themselves and their livestock warm (Rastogi *et al.*, 2014). This could also be one of the possible reasons for lower value of BB contribution to OC in present study than that reported by Srinivas *et al.* (2016). Another possible reason can be the size of particulates under

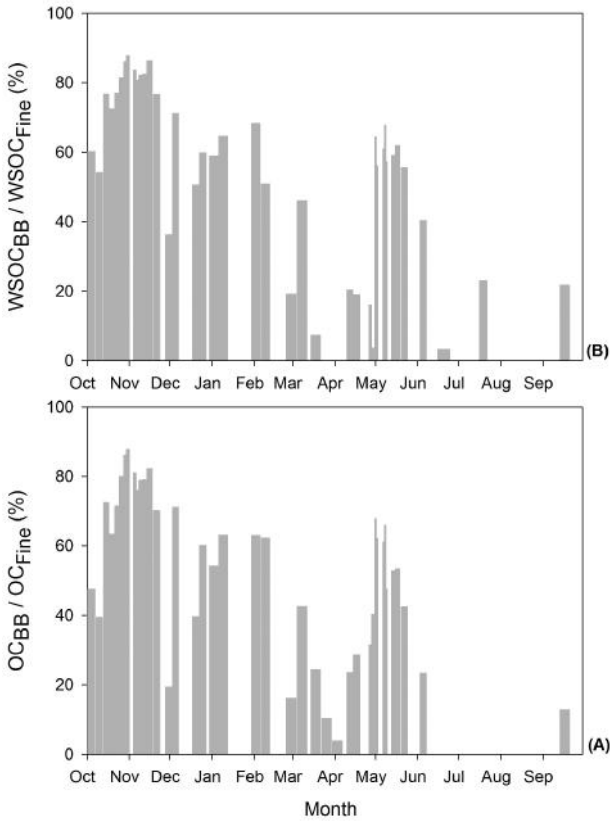

Fig. 4: Fraction of biomass (BB) derived (A) organic carbon (OC) and (B) water-soluble organic carbon (WSOC) in fine aerosol

consideration as aged and other primary organics are generally found in coarse mode (>2.5 micron) (Chalbot *et al.*, 2014). Further, present estimation is based on the primary tracer and represent primary OC_{BB}, as mentioned earlier. This is well established that estimation using isotopic signature involved relatively less error in comparison to K^+ based method, however, $\Delta^{14}C$ measurements are difficult, time consuming, and costly in comparison to the measurement of K^+. Hence, present study suggests that one can assess the contribution of BB to ambient OC using K^+.

The contribution of BB to fine WSOC has also been quantified by employing the same method as used in the case of OC. The intercept in this case is 4 $\mu g\ m^{-3}$ (Fig. 3B), which is subtracted from ambient WSOC to estimate BB derived WSOC. The contribution of BB to ambient fine WSOC ranged from 0 to 88% (annual average: 43±32%) over an annual cycle (Fig. 4B). $WSOC_{BB}$ follows the similar temporal trend as exhibited by OC_{BB} with contribution of 75%, 55% and 44% during autumn, winter and summer seasons respectively, whereas it is found to be 14% for spring and 5% for wet season attesting that same sources contribute to $WSOC_{BB}$ and OC_{BB}. In nutshell, present study demonstrates that BB is significant source of ambient fine OC and quantify the contribution of BB to ambient OC, which has important implications in climate models and air quality regulation.

Summary and Conclusions

In the present study, the impact of BB on ambient K^+, OC and WSOC in PM_1 have been examined over an annual cycle over Patiala, northwest part of the IGP. BB is found to be the major source of ambient fine K^+ over the study region with the average contribution of 77±24% to total fine K^+ over an annual cycle. Further, BB derived fine mode K^+ is used to quantify the contribution of BB to ambient fine OC. On annual scale, OC_{BB} contributes around 41±30% to the ambient fine OC with higher contribution during autumn (69%), winter (54%) and summer (40%) seasons, and relatively lower contribution during spring (26%) and wet (2%) seasons. Similarly, the fraction of $WSOC_{BB}$ to ambient fine WSOC is found to be significant (43±32%) with seasonal contribution of 75%, 55%, 14%, 44% and 5% during autumn, winter, spring, summer and wet, respectively. Agricultural-waste burning emissions (paddy- and wheat-residue) during autumn and summer, and bio-fuel (such as Babool, Cowdung cake, Eucalyptus, Jujube and Shisham) burning during winter are the major sources of OC_{BB}. Quantification of BB contribution to atmospheric OC load is important in climate and air quality models, and may help policy makers in designing the mitigation strategies.This study also provides the baseline data of biomass burning derived OC for the northwestern IGP where biomass burning have a significant impact on the regional and global climate.

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