Molecular characterization of organic aerosols in the Kathmandu Valley, Nepal: insights into primary and secondary sources

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Abstract:

Organic atmospheric aerosols in the Hindu Kush-Himalayan-Tibetan Plateau region are still poorly characterized. To better understand the chemical characteristics and sources of organic aerosols in the foothill region of the central Himalaya, the atmospheric aerosol samples were collected in Bode, a suburban site of the Kathmandu Valley (KV) over a one-year period from April 2013 to April 2014. Various molecular tracers from specific sources of primary organic aerosols (POA) and secondary organic aerosols (SOA) were determined. Tracer-based estimation methods were employed to apportion contributions from each source. The concentrations of organic carbon (OC) and element carbon (EC) increased during winter with a maximum monthly average in January. Levoglucosan (a molecular tracer for biomass burning, BB) was observed as the dominant species among all the analyzed organic tracers and its annual average concentration was 788±685 ng m^{-3} (ranging from 58.8 to 3079 ng m^{-3}). Isoprene-SOA (I-SOA) represented a high concentration among biogenic-SOA tracers. For the seasonality, anhydrosugars, phenolic compounds, resin acid and aromatic SOA tracer showed similar seasonal variations with OC, and EC while monosaccharides, sugar alcohols and I-SOA tracers showed lower levels during winter. BB contributed a significant fraction to OC, averaging 24.9±10.4% during the whole year, and up to 36.3±10.4% in the post-monsoon season. On an annual average basis, anthropogenic toluene-derived secondary OC accounted for 8.8% and biogenic secondary OC contributed 6.2% to total OC. The annual contribution of fungal-spores to OC was 3.2% with the maximum during the monsoon season (5.9%). For plant debris, it accounted for 1.4% of OC during the monsoon. Therefore, OC is mainly associated with BB and other anthropogenic activity in the KV. Our findings are conducive to designing effective measures to mitigate the heavy air pollution and its impacts in the KV and surrounding area.

1. Introduction

South Asia, especially the Indo-Gangetic Plain (IGP) region, is a global air pollution hotspot. Atmospheric pollutants [e.g., organic carbon (OC), black carbon (BC), gaseous pollutants, etc.] from South Asia have been increasing during recent decades (Ramanathan et al., 2005;Muzzini and Aparicio, 2013;Lawrence and Lelieveld, 2010). While these pollutants are of concern locally near the emission sources, they can also, in short span of time, be transported to rural and remote regions over a long distance. This results in an annually recurring regional scale haze, referred to as atmospheric brown clouds (ABC), covers a large area from the Himalayan range to the Indian Ocean (Ramanathan et al., 2007). Until recently the emissions, types, levels, atmospheric transport and transformation, impacts and mitigation of various atmospheric pollutants were not well characterized in the vast mountain areas and the foothill region in South Asia. In this context, the international project of "A Sustainable Atmosphere for the Kathmandu Valley (SusKat)" was launched, aiming to comprehensively understand the causes of the severe air pollution in the region, and identifying appropriate solutions to reduce its impacts (Rupakheti et al., 2018). This paper presents analyses of samples collected as part of the SusKat field campaign.

The Kathmandu Valley (KV), the capital region of Nepal, is considered one of the most polluted regions over South Asia and the largest metropolitan region in the foothills of the Hindu Kush-Himalayas-Tibetan Plateau (HKHTP) region, facing rapid but unplanned urbanization, with the current population of approximately 4 million (Muzzini and Aparicio, 2013). Additionally, the bowl-shaped topography restrict the free flow of air, resulting in poor air quality (Pudasainee et al., 2006;Panday and Prinn, 2009). Giri et al. (2007) showed PM_{10} concentrations in Kathmandu were about 2-4 times higher than the guidelines prescribed by the World Health Organization (WHO) (PM_{10} 24-hour mean: 50 $\mu g\ m^{-3}$) (WHO, 2006). More recently, Shakya et al. (2017) reported that daily mean $PM_{2.5}$ concentrations at seven locations in the KV during 2014 were about 5 times higher than the WHO guidelines ($PM_{2.5}$ 24-hour mean: 25 $\mu g\ m^{-3}$) (WHO, 2006). Beside particulate matter, recent studies have pointed out that ground-level ozone (O_3) is also of concern (Mahata et al., 2017b;Bhardwaj et al., 2017). Ozone levels at Pakanajol in the city center exceeded the WHO's 8-hour maximum O_3 guidelines of 100 $\mu g\ m^{-3}$ on 125 days in a year (Putero et al., 2015), while Mahata et al. (2017b) reported such exceedance was for nearly 3 months at Bode (where sampling for this study was conducted) and 6 months at Nagarkot, a hilltop site downwind of the KV. The concentrations of acetonitrile and isoprene (precursor for both O_3 and secondary organic aerosol, SOA) investigated by Sarkar et al. (2016) in the KV were comparable with the highest reported elsewhere in the world. Air pollution is a clear threat to human health (leading to respiratory disease, cardiovascular disease, cancer, etc.), agricultural productivity and revenues from tourism in the KV and surrounding regions (Putero et al., 2015;Shakya et al., 2016).

Carbonaceous aerosols (OC and BC/EC, element carbon) are often a principal component of atmospheric aerosols and the atmospheric brown clouds (ABC) over South Asia (Wester et al., 2019). Sources and chemical transformations of OC are complicated, including primary emission sources (e.g. biomass/biofuel burning and fossil fuel combustion, plant debris, soil dust, etc.) and secondary formation of the oxidative products from precursor gases produced from both biogenic and anthropogenic compounds (Simoneit, 2002;Claeys et al., 2004;Fu et al., 2010). Although previous studies on organic aerosol characteristics in the KV and surrounding regions are limited, they demonstrated that OC was the main component of aerosols (Shakya et al., 2017;Kim et al., 2015). However, they have focused on a few aerosol species or a handful of organic compound classes (Chen et al., 2015;Sarkar et al., 2016). Only at a rural site, Godavari on the southern edge of the KV, analysis of organic aerosols at the molecular level has been reported (Stone et al., 2010;Stone et al., 2012). Overall, the composition and sources of OC are still poorly characterized.

Therefore, to overcome such research gaps, our study comprehensively investigates the organic molecular compositions of aerosols from the KV, including anhydrosugars, monosaccharides, sugar alcohols, phenolic compounds, resin acid, phthalic acid esters, and secondary organic aerosols produced from primary emission sources and secondary formation. We also studied the seasonal variation and molecular distribution to decipher their abundances, understand their predominant sources (primary vs. secondary), and to evaluate contributions of different sources to the carbonaceous aerosols in the suburban environment in the Himalayan foothills. Our current work enriches the database of the chemical characteristics of organic aerosols in South Asia, particularly in the HKHTP region.

2. Materials and methods

2.1. Sampling site

The KV is a round flat basin with the bottom of an elevation of approximately 1300 meters above sea level (a.s.l.) in the southern foothills of the Himalayas. It is encircled by green mountains (elevation: 1500 m to 2800 m a.s.l.) (Panday and Prinn, 2009). Our sampling was performed during April 2013 to April 2014 in Bode (27.67°N, 85.38°E, 1320 m a.s.l.), a suburban site to the east of Kathmandu city in the valley (Fig. 1). There are two major wind flows in the KV: (a) west to east, from Nagdhunga-Bhimdhunga mountain pass in the west to Nagarkot-Sanga mountain pass in the east, (b) south to north, from Bagmati River corridor to the northeast direction through the central-eastern part of the Valley. These two airflows meet around central-eastern part of the Valley and move eastward towards the Nagarkot-Sanga passes (Panday and Prinn, 2009). The Bode area receives these two air flows, and hence it is downwind of Kathmandu city and Lalitpur or Patan city located in southwest, west and northwest direction during the daytime, and Bhaktapur city located in east and southeast during nighttime (Bhardwaj et al., 2017;Mahata et al., 2017a;Rupakheti et al., 2018). In addition, it is situated in a residential area with urban buildings and houses scattered across agricultural fields with paddy, wheat, corn and vegetable farms. Some small industries (plastics, electronics, wood, fabrics, etc.) and Bhaktapur Industrial Estate are located in the south-eastern direction of Bode, as well as several brick kilns that use low quality coal during January to April (Sarkar et al., 2016). The Tribhuvan international airport in the west of Bode (~ 4 km) may have potential impacts when there is westerly wind. Approximately 1.5 and 7 km to the north, there are two reserve forests, consisting of a mix mainly broad-leaved deciduous trees and evergreen conifer trees (Department of Plant Resources, 2015). BC and O_3 measurements in the two major SusKat-ABC sites (Paknajol and Bode) in the Valley show similar levels (Putero et al., 2015;Mahata et al., 2017b). Therefore, Bode site can be taken as a representative site for the KV (Rupakheti et al., 2018).

2.2 Sample collection

The total suspended particulates (TSP) were continuously collected for 23 h (day and night time) every five days by a medium-volume sampler (model: KC-120H, Laoshan Co., China), which was installed on the rooftop of a building, approximately 20 m above ground. The flow rate was100 L min^{-1}. Overall, 82 aerosol samples were successfully obtained using 90 mm diameter quartz filters (Whatman PLC, UK). The filters were pre-baked (550 °C, 6 h) to remove all organic material and weighed by a microbalance (sensitivity: ±0.01 mg) before and after sampling. Before each weighing, they were equilibrated at constant temperature (25±3 °C) and humidity (30±5%) conditions for 24 h. Finally, the filters were preserved at 20 °C below zero until laboratory analysis. Field blanks (one blank filter each month) were also collected, briefly putting a filter onto the instrument without drawing air to assess potential contamination. There may be positive and negative artifacts during the sample handling/conditioning due to the adsorption/evaporation processes of organic aerosols (Li et al., 2018;Boreddy et al., 2017;Oanh et al., 2016). In a comparable study, Ding et al. (2013) reported the positive artifacts for OC and organic tracers were 10–20% and up to 16%, respectively.

2.3 Chemical Analysis

The aerosol samples were analyzed for major ions, OC, EC, and organic molecular tracers in the laboratory. Major ions (Ca^{2+}, Na^{+}, K^{+}, Mg^{2+}, NH_4^{+}, Cl^{-}, $SO_4^{2-}$and NO_3^{-}) were measured using an ion chromatography (Dionex, USA) with ICS-320 and ICS-1500 (Tripathee et al., 2017). The detection limit (LOD) of all the major ions was 0.01 μg m^{-3}. They denoted less than 5% of the real sample concentrations in the field blank filters (Tripathee et al., 2017). Non-sea-salt Ca^{2+} (nss-Ca^{2+}) and K^{+} (nss-K^{+}) was estimated according to the method from George et al. (2008). OC and EC were determined by a thermal/optical reflectance analyzer (Model 2001A, USA) (Wan et al., 2015). The field blank OC (0.59±0.13 μg m^{-3}) were subtracted from the filter samples. EC in the field blank sample was 0.00 μg m^{-3}.

Detailed analytical method of organic molecular tracers was described previously by Wan et al. (2017). A trace gas chromatography coupled to a PolarisQ mass spectrometry detector (GC-MS, Thermo Scientific) was used for analysis. Briefly, small filter portions (1.13-3.39 cm^2) were cut, spiked with appropriate amounts of methyl-*β*-D-xylanopyranoside (MXP, 99%, Sigma) and D_3-malic acid (DMA, CDN isotopes, 99%) as internal recovery standards. Each filter portion was then extracted three times with a mixture of 20-ml dichloromethane/methanol (2:1, *v*/*v*) at room temperature for 30 minutes. The solvent extracts in total of 60 ml were combined and successively filtered through quartz wool, concentrated, dried over ultrapure nitrogen gas and then reacted with 50 μl of 99% *N,O*-bis-(trimethylsilyl)trifluoroacetamide (BSTFA, with 1% trimethylsilyl chloride) and pyridine (*v*/*v*=2:1) at 70 °C for 3 h. n-Hexane of 150 μl was added after derivatization. A TG-5MS (30 m × 0.25 mm × 0.25 μm) was used for separation according to the GC temperature program. The oven temperature was initially held at 50 °C for 2 min, increased to 120 °C at 30 °C min^{-1}, then to 300 °C at 6 °C min^{-1} and finally held for 16 min. The MS was operated in electron ionization (EI) mode at 70 eV with a scan range of 50-650 Da.

For quantitative analysis, authentic standards processed as the samples above were used to establish the calibration curves. To quantitate the target compounds that were no available standards, surrogate compounds were used as following: erythritol for 2-methylglyceric acid (2-MGA), 2-methyltetrols (2-MTLs) and C_5-alkene triols, cis-pinonic acid (PNA) for 3-hydroxyglutaric acid (3-HGA) and 3-methyl-1,2,3-butanetricarboxylic acid (MBTCA) , pinic acid (PA) for ß-caryophyllinic acid (*ß*-CA), azelaic acid for 2,3-dihydroxy-4-oxopentanoic acid (DHOPA). Recoveries for target tracers and MXP (Table S1) were more than 75%. The exception was for malic acid (50.3%-90.5%) and cis-pinonic acid (60.2%-81.8%). The relative differences based on duplicate analysis were less than 15%. The method detection limits (MDL) were 0.04-0.13 ng m^{-3} (Table S1). Our results were not corrected for the recoveries. No target compounds were detected in the field blank filters using the same procedure with the samples.

Table 1 shows a list of chemical species (OC, EC and major ions) and molecular markers (from the sources of biomass burning (BB), fungal spores, plant debris, plastic emission, biogenic and anthropogenic secondary formation) analyzed in this study. They include anhydrosugars (levoglucosan, mannosan and galactosan), primary saccharides (sucrose, trehalose, glucose and xylose) and sugar alcohols (mannitol, arabitol, sorbitol and erythtitol), lignin and resin pyrolysis products (vanillic, syringic, *p*-hydroxybenzoic and dehydroabietic acids), phthalic acid esters, isoprene tracers (2-MGA, 2-MTLs of methylthreitol and 2-methylerythritol, C5-alkene triols of cis-2-methyl-1,3,4-trihydroxy-1-butene, 3-methy-2,3,4-trihydroxy-1-butene and trans-2-methyl-1,3,4-trihydroxy-1-butene), monoterpene tracers (PNA, PA, 3-HGAand MBTCA), *ß*-CA and DHOPA.

2.4 Estimation of measurement uncertainty

The application of surrogate standards for the quantification of most SOA tracers excluding PNA and PA) could cause additional errors to the measurements. Error in analyte measurement (EA) is propagated from the standard deviation of the field blank (EFB), error in spike recovery (ER) and error from surrogate quantification (EQ):

$$EA = \sqrt{EFB^2 + ER^2 + EQ^2}$$

EFB was 0 in this study due to the undetectable SOA tracers in the field blanks. To estimate the ER of tracers, the spike recoveries of surrogate standards within the range of 9.2% (erythritol) to 26.1% (PNA) were used. EQ was estimated by an empirical approach according to Stone et al. (2012). The relative error introduced by each carbon atom (En), oxygenated functional group (Ef) and alkenes (Ed) was estimated to be 15 %, 10% and 60%, respectively. Therefore, the EQ are calculated as:

$$EQ=En\Delta n+ Ef\Delta f+ Ed\Delta d$$

where Δn, Δf and Δd are the difference between a surrogate and an analyte of carbon atom number, oxygen-containing functional group and alkene functionality, respectively.

Currently, EQ was calculated in the range of 15% (2-MTLs) to 120% (ß-CA) and the estimation of EA ranged from17.6% to 122.4%. The estimated uncertainties for the measurement of the SOA tracers is presented in Table S2.

2.5 Meteorological parameters

The meteorological parameters (e.g., temperature (T), relative humidity (RH), etc.) used in this study were derived from Tribhuvan International Airport (www.wunderground.com), which was located west of Bode (approximately 4 km). Mixing layer height (MLH) data was measured with a Vaisala ceilometer at Bode site (Mues et al., 2017). The meteorology of KV and its surrounding regions is controlled by the South Asian monsoon circulations in the wet season (monsoon, June-September). Westerlies dominate the atmospheric circulation patterns during the dry seasons including pre-monsoon (March-May), post-monsoon (October-November) and winter (December-February) with limited precipitation (Pudasainee et al., 2006;Mues et al., 2017). Additionally, it is also influenced by local mountain valley circulation (Mues et al., 2018).

3. Results and discussion

A statistical concentration summary of major ions, OC, EC, and organic tracers identified in TSP samples collected at the Bode site is presented in Table 1. Tracers for six classes of organic compounds were detected: anhydrosugars, monosaccharides, sugar alcohols, phenolic compounds and resin acid, phthalic acid esters, and secondary organic aerosol (SOA) tracers.

3.1 Aerosol loadings

The TSP samples at Bode site exhibited daily mass concentrations from 32.0 to 723 μg m^{-3} (255±167 μg m^{-3}) during April 2013 to April 2014 (Table 1). Putero et al. (2015) reported 195±83 μg m^{-3} of online $PM_{2.5}$ concentration in Pakajol site (also one of SusKat-ABC sites), accounting for roughly 80% of TSP in our study. The TSP concentrations were comparable to those reported over other heavily polluted regions in South Asia, including Islamabad in Pakistan (Shah and Shaheen, 2008), Kolkata (Gupta et al., 2007) and Agra (Rajput and Lakhani, 2010) in India. Compared to the remote sites such as Lulang in the Tibetan Plateau (Wang et al., 2015) and Manora Peak in the central Himalaya (Ram et al., 2010), the TSP in Bode shows significantly higher mass concentrations. We found a clear seasonal variation in TSP mass concentrations (Fig. 2a), higher in pre-monsoon season (381 ± 366 μg m^{-3}) and winter (353 ± 348 μg m^{-3}) while lower in monsoon period (120 ± 107 μg m^{-3}) which was nearly half of post-monsoon season (256 ± 213 μg m^{-3}). It generally corresponded to the build-up of the atmospheric brown clouds (ABCs), which engulfed most of South Asia and the Northern Indian Ocean extending from November to May (Ramanathan et al., 2005).

Meteorological parameters may also affect the TSP concentrations. The highest TSP concentration observed during the pre-monsoon can be caused by the fugitive dust which is been blown up by strong wind and the absence of wet-precipitation (Fig S1a and c). The lower TSP concentration in the monsoon was likely related to increased precipitation (Fig S1c) after the onset of the South Asian monsoon. During this season, nearly 80% of the annual precipitation falls in the KV, which flushes out pollutants from the atmosphere (Tripathee et al., 2017;Wester et al., 2019). During winter, an inversion layer often occurs in the KV owing to its bowl-shaped topography (Pudasainee et al., 2006). The existence of an inversion layer with the lower temperature (12.0±2.41°C), wind speed (2.86±1.34 km h^{-1}), and MLH (0.34±0.08 km) (Mues et al., 2017) (Fig S1a, c and d) altogether reduced the pollution dispersion mechanism resulting in increased levels of pollutants close to the ground surface.

3.2 Major ions and OC/EC

Concentrations of eight major ions were measured in the aerosol samples from the Bode site. The total sum accounted for 17.1%±8.5% of annual average TSP mass. Sulfate ranked the highest among them (annual mean: 10.8±9.83 μg m^{-3}), followed by Ca^{2+} (7.96±6.85 μg m^{-3}), NH_4^+ (5.92±6.16 μg m^{-3}), NO_3^- (5.21±4.35 μg m^{-3}), Na^+ (3.28±1.58 μg m^{-3}), K^+ (2.43±2.82 μg m^{-3}), Cl^- (2.15±2.25 μg m^{-3}) and Mg^{2+} (0.61±0.54 μg m^{-3}). On average, the combination of SO_4^{2-}, NO_3^- and NH_4^+, i.e. the secondary inorganic aerosols, constituted more than half (51.3%) of the total ionic concentrations. The Ca^{2+} alone accounted for 22.1% of total ions.

Sulfate, ammonium and nitrate revealed a typical seasonality with the seasonally averaged concentrations ranked in the descending order of winter > pre-monsoon > post-monsoon > monsoon. This is consistent with the seasonal variation of the precursors NO_x, NO_2 and SO_2, which are mainly caused by automobile exhaust, household cooking, and emissions from brick kilns co-fired with biomass in the KV (Kiros et al., 2016;Wester et al., 2019). Currently, nearly 50% of the total motor vehicles in Nepal (approximately 2.33 million) run on the KV roads (DoTM, 2015;Mahata et al., 2017b). Diesel- or gasoline-powered generators (producing higher NO_x emissions) and garbage burning are other major pollution sources in Nepal during the sampling period, which can also emit much aerosol precursors (Stockwell et al., 2016).

Ions derived from crustal sources, such as Ca^{2+} and Mg^{2+} are related to the local fugitive dust sources such as unpaved roads and construction activities (Ram et al., 2010). Interestingly, good correlations were found for Ca^{2+} and SO_4^{2-} ($R^2 = 0.48$, $P<0.001$) , NO_3^- ($R^2 = 0.58$, $P<0.001$) and NH_4^+ ($R^2 = 0.62$, $P<0.001$), and for Mg^{2+} and SO_4^{2-} ($R^2 = 0.61$, $P<0.001$) , NO_3^- ($R^2 = 0.71$, $P<0.001$) and NH_4^+ ($R^2 = 0.69$, $P<0.001$), respectively (Table 2), which hinted that dust may co-exist with SO_4^{2-}, NO_3^- and NH_4^+ in the KV (Tripathee et al., 2017).

Carbonaceous aerosols (OC: 38.7±32.7 μg m^{-3} and EC: 9.92±5.33 μg m^{-3}) accounted for 19.2%±5.48% of TSP mass through the sampling period at the Bode site, which was higher than that of the major ions. OC alone accounted for 14.6%±4.81% of the TSP mass. During winter and pre-monsoon seasons, OC and EC showed much higher concentrations than that during wet season (Fig. 2b and c). In this study, we found that the daily OC to EC mass ratios (OC/EC) varied from 0.77 to 15.8 (annual mean: 3.78±2.73) and seasonal mean ratios of 4.44, 2.71, 3.31, and 5.86 during pre-monsoon, monsoon, post-monsoon, and winter seasons, respectively (Table 1 and Fig. 2d). The OC/EC ratios of more than 2.0 indicates the BB aerosols or the formation of secondary organic matter (Cao et al., 2007). Their influence and contribution will be discussed in the following sections. The OC/EC ratios found in this study for the KV were similar to other sites in South Asia, like Lumbini (5.16±2.09, 2.41-10.03) (Wan et al., 2017), Delhi (5.86±0.99, 2.9-9.2) (Bisht et al., 2015) and Lahore (3.9 ± 1.6,1.5-7.2) (Alam et al., 2014).

3.3 Sugar compounds

3.3.1 Anhydrosugars

Anhydrosugars of levoglucosan (1,6-anhydro-β-D-glucopyranose) and its two isomers (mannosan and galactosan) have been used as ideal molecular tracers for BB emissions (Simoneit, 2002;Bhattarai et al., 2019). They are exclusively emitted from the combustion and pyrolysis of cellulose and hemicelluloses. In the current study, the annual average concentration of levoglucosan was 788±685 ng m^{-3}, ranging from 58.8 to 3079 ng m^{-3}, which was the dominant species of the total identified tracer compounds (Table 1).

For the seasonality, levoglucosan showed significantly higher levels during winter, pre-monsoon and post-monsoon seasons (Fig. 3a). Especially, higher concentrations were recorded in winter varying from 830 to 2395 ng m^{-3} (annual mean: 1391±535 ng m^{-3}). It showed comparable levels with other sites in the world, which were badly affected by the BB emissions, e.g., New Delhi (1977 ng m^{-3}) (Li et al., 2014) and Raipur (2180 ng m^{-3}) in India (Deshmukh et al., 2016), Tasmania (4540 ± 2480 ng m^{-3}) in Australia (Reisen et al., 2013) and Lumbini (1161±1347) in Nepal (Wan et al., 2017). Our results were much higher than the aerosols (20-372 ng m^{-3}) collected at a rural Godavari site (Stone et al., 2010), located on the southern edge of the KV during 2006. Good correlations exhibited between levoglucosan and OC (R^2=0.79, P<0.001), EC (R^2=0.42, P<0.001) and nss-K^+ (R^2=0.35, P<0.01) during the sampling campaign (Fig. 4). This indicates that OC and EC in KV's aerosols are strongly related to BB source (Kim et al., 2015).

The ratio of levoglucosan to mannosan (Lev/Man) has been applied to distinguish the possible categories of biomass burnt. Previously, higher Lev/Man ratios were reported for emissions from combustion of hardwood (ranging from 12.9 to 35.4 with an average of 21.5±8.3) and agricultural residues (range from 12.7 to 55.7 with an average of 32.6±19.1) (Sang et al., 2013;Bhattarai et al., 2019). For the softwood burning, the average ratio was 4.0±1.0 (ranging from 2.5 to 5.8). In current study, the annual mean ratio of Lev/Man was 16.3±5.96 ranging from 9.13 to 33.1 with only 9 samples <10. It can be inferred that the combustion of crop residues and hardwood is likely to be one of the major sources of atmospheric pollution in this region. A previous study also reported that the combustion of wood fuel for cooking and heating is common during wintertime in Nepal while there is much more crop residue combustion during both pre- and post-monsoon seasons (Stockwell et al., 2016). This is not only a local but also a regional phenomenon; for example, Bhardwaj et al. (2017) and Wan et al. (2017) pointed out emissions from crop residue burning during the pre- and post-monsoon periods from western India and eastern Pakistan impact the air quality in Nepal. Similarly, Rupakheti et al. (2017) also showed that the combustion of agricultural residues and forest fires over the northwestern IGP region are causes of the air pollution episodes over the foothills of the central Himalayas. In addition, brick kilns mainly operated during January-April, burned substantial quantities of low-grade coal, mixed crop wastes and firewood (Kim et al., 2015;Wester et al., 2019). Such emissions may also lead to the high levels of levoglucosan observed at Bode. We must point out that the incense burning in KV may also influence the levoglucosan concentration.

3.3.2 Monosaccharides

Primary biological aerosol particle (PBAP) tracers, commonly known also as bioaerosols, were analyzed in the Bode aerosol samples, including five monosaccharides of glucose, fructose, trehalose, sucrose and xylose. PBAP is derived from fungal spores, vegetative debris, pollen, bacteria and viruses. Most of them can cause adverse effects (e.g., allergenic reaction) on human health (Bauer et al., 2008;Myriokefalitakis et al., 2017).

In current work, total monosaccharides had an annual mean concentration of 298±127 ng m^{-3}. Glucose was the predominant species among monosaccharides (124±60.0 ng m^{-3}), followed by fructose (58.2±28.3 ng m^{-3}), sucrose (48.3±27.4 ng m^{-3}), trehalose (40.8±22.0 ng m^{-3}), and xylose (26.5±18.1 ng m^{-3}) (Table 1). Except xylose, they all presented higher concentrations in pre-monsoon period while being lower in winter (Fig 3h, i, j and k). There were significant linear correlations between glucose and fructose ($R^2 = 0.77$, $p<0.001$), trehalose and glucose ($R^2 = 0.30$, $p<0.001$), trehalose and fructose ($R^2 = 0.23$, $p<0.001$), sucrose and glucose ($R^2 = 0.55$, $p<0.001$), sucrose and fructose ($R^2 = 0.55$, $p<0.001$), and sucrose and trehalose ($R^2 = 0.28$, $p<0.001$) (Table 3). Therefore, the strong correlations indicated that they were derived from common sources, e.g. from local forests in the KV during the period of high productivity of plants. In addition, the pollen produced from the flowering of local vegetation also largely contribute to glucose, fructose, trehalose, sucrose. The flowering of trees and crops peaks during the pre-monsoon season. The similar phenomenon was also reported in deciduous forests in northern Japan (Miyazaki et al., 2012).

Xylose has complex sources, including soils (Simoneit et al., 2004), microbiota (Wan and Yu, 2007), vegetation, bacteria (Cowie and Hedges, 1984) and biomass combustion (Zhu et al., 2015). It presents less abundant and only accounts for 6.90%±8.32% of the total PBAP tracers identified in the Bode aerosols. For the seasonal pattern, it is characterized by waxing in winter (47.4±24.3 ng m^{-3}) and waning in monsoon season (12.0±5.16 ng m^{-3}), which was different from the other primary monosaccharides (Table 1 and Fig. 2l). Close correlation between xylose and levoglucosan (the BB tracer) was observed in our study (Fig. S2, R^2=0.72, p<0.001), indicating that the emissions from the burning of biomass may largely contribute to xylose in Bode aerosols. A similar finding of xylose source (i.e. BB) was also proposed by Zhu et al. (2015).

3.3.3 Sugar alcohols

Total concentration of sugar alcohols (arabitol, sorbitol, erythtitol and mannitol) was 213±126 ng m^{-3}, and thus lower than that of total monosaccharides (Table 1). Mannitol (86.9±55.3 ng m^{-3}) and arabitol (68.4±39.8 ng m^{-3}) showed higher concentrations, followed by erythitol (43.1±28.8 ng m^{-3}) and sorbitol (14.2±8.02 ng m^{-3}). All of them exhibited monsoon maxima (114±61.4 ng m^{-3}, 86.6±44.5 ng m^{-3}, 56.9±33.1 ng m^{-3} and 17.9±9.31 ng m^{-3}, respectively) and winter minima (18.1±6.02 ng m^{-3}, 26.1±9.13 ng m^{-3}, 5.82±2.72 ng m^{-3}, 12.4±7.60 ng m^{-3}, respectively) (Table 1 and Fig. 3m, n, o and p). They also showed significant correlations with each other, implying their common sources (Zhu et al., 2015). Mannitol and arabitol have been mostly associated with fungal spores, along with vegetation and mature leaves and algae (Yttri et al., 2007;Myriokefalitakis et al., 2017). Recent studies proposed elevated concentrations of mannitol and arabitol were usually observed augmentation after rain events and also highly correlated with relative humidity (Yue et al., 2016;Zhu et al., 2016). Therefore, at Bode, sugar alcohols were likely emitted by plants in nearby forest and agriculture fields, especially during the monsoon with the higher relative humidity (Fig. S1b). In addition, the higher temperatures (Fig. S1a) were conducive for more active microbial activities. Notably, the levels of PBAP discussed above were much higher than other sites in the world (Zhu et al., 2015;Liang et al., 2016;Yttri et al., 2007), indicating the strong fungal spore production in the KV during the wet season.

3.4 Phenolic compounds and resin acid

Phenolic compounds (e.g., vanillic, syringic and *p*-hydroxybenzoic acids) derived from lignin pyrolysis and resin acid (e.g., dehydroabietic acid) from burning of conifer plants can be also used as biomarkers for BB. Syringic acid is prevalent in hardwood smoke while vanillic acid is dominant both in softwood and hardwood smoke (Myers-Pigg et al., 2016;Wan et al., 2019). Herbaceous plant smoke primarily contains p-anisic acid and p-anisaldehyde (e.g., *p*-hydroxybenzoic acid and *p*-hydroxybenzaldehyde). Dehydroabietic acid is a dominant compound in the total lipid material from pine wood smoke. Therefore, three pyrolysis products of lignin (vanillic, syringic and *p*-hydroxybenzoic acids) and one pyrolysis of resin acid (dehydroabietic acid) were chosen as organic markers in this study.

p-Hydroxybenzoic acid (19.8±12.3 ng m^{-3}) was the predominant species, the second was dehydroabietic acid (13.8 ±6.19 ng m^{-3}), and then vanillic (15.3±11.3 ng m^{-3}) and syringic acids (17.1±13.7 ng m^{-3}) (Table 1). They exhibited maximum concentrations during winter and pre-monsoon periods, while decreased during wet season and then increased from post-monsoon period, which was consistent with the seasonal variation of levoglucosan (Fig 3d, e, f and g). There were also significant correlations of lignin and resin pyrolysis products with levoglucosan (cellulose pyrolysis products) (Fig. S3a, *p*-hydroxybenzoic acid and levoglucosan, R^2=0.72, P<0.001; Fig. S3b, vanillic acid and levoglucosan, R^2=0.86, P<0.001; Fig. S3c, syringic acid and levoglucosan, R^2=0.83, P<0.001; Fig. S3d, levoglucosan and dehydroabietic acid, R^2=0.63, P<0.001). Such a result also shows that there are various biomass combustion sources in the valley.

The concentration ratio of syringic to vanillic acids (Syr/Van) has recently been used to further discriminate the vegetation types burned (Fujii et al., 2015;Myers-Pigg et al., 2016;Wan et al., 2019). Previous study documented the Syr/Van ratios ranged from 0.1 to 2.44 for combustion of hardwood and herbaceous angiosperm, while it varied from 0.01 to 0.24 for burning softwood (Myers-Pigg et al., 2016). Regarding the aerosol samples from KV, the Syr/Van ratio was 0.94 ± 0.18 of an annual average ranging from 0.65 to 1.31, indicating that combustion of hardwood and herbaceous plant (including crop residues) are the most likely sources of BB in the Valley. This finding agrees with the results obtained from the Lev/Man ratios discussed in Sect. 3.3.1.

Besides the information revealed by anhydrosugars discussed in section 3.3.1, lignin and resin biomarkers further confirmed that BB emissions play a significant role in contributing to organic aerosols in the KV, particularly during winter and pre-monsoon periods.

3.5 Phthalic acid esters

Phthalates or phthalic acid esters are extensively utilized as key additives in the manufacture and processing of plastic products. As they are physically rather than chemically bonded to the polymer, they can be easily released into the environment. There are potential adverse effects on ecological system and human health due to their toxicity, e.g., carcinogenicity and endocrine disruption (Fu et al., 2010;Li et al., 2016). Diethyl (DEP), di-n-butyl (DnBP) and bis-(2-ethylhexyl) (DEHP) phthalates were analyzed in current study. The annual average concentration of phthalates was 510±230 ng m^{-3} (165-1520 ng m^{-3}) (Table 1). They showed higher concentration during pre-monsoon (Fig. S4). Similar concentrations of phthalates (the total of DEP, DnBP, dimethyl, diisobutyl and di-(2-ethylhexyl) phthalates) was also investigated in an Indian urban site, with 553 ng m^{-3} (295-857 ng m^{-3}) in May and 303 ng m^{-3} (175-598 ng m^{-3}) during winter (Fu et al., 2010). In South Asia, large quantities of municipal solid wastes containing plastic products are generally disposed of in open landfills. The open burning of plastics along with other municipal solid waste is common in Nepal, and thus can also release numerous phthalate compounds into the atmosphere.

3.6 SOA tracers

Emissions of volatile organic compounds from vegetation (VOCs) into the atmosphere, especially isoprene, monoterpenes and sesquiterpenes occurs in large amounts. These biogenic VOCs (B-VOCs) are crucial precursors of biogenic SOA. Globally, the emissions of B-VOCs (1150 TgC/yr) consisting of 44% isoprene and 11% monoterpenes are much higher than emissions of anthropogenic VOCs (only 110 TgC/yr) (Guenther et al., 1995). It should be noted, besides biogenic emissions, combustion of biomass and fossil fuel also contribute to the isoprene, monoterpenes and sesquiterpenes (Jathar et al., 2014;Sarkar et al., 2016;Sarkar et al., 2017). The investigation of gaseous VOCs during winter (December 2012 to February 2013) air in the KV during SusKat-ABC campaign also showed high levels of isoprene and it was attributed (at least during high isoprene periods) mostly to biogenic emissions (Sarkar et al., 2016;Sarkar et al., 2017). It is difficult to appropriately quantify the fractions of biogenic and anthropogenic emissions of these compounds, based on ambient measurement of these species alone without measurement of BB tracers such as acetonitrile and furan. The budget of isoprene emissions (500 Tg y^{-1}) on a global scale is dominated by vegetation (Guenther et al., 2006). Therefore, in our study, we considered the oxidation products of isoprene, monoterpenes and sesquiterpenes as the tracers of biogenic emissions and attribute their main source as biogenic emissions. This may lead to some overestimation of their contributions to SOA formation.

3.6.1 Isoprene SOA tracers

Six isoprene-SOA (I-SOA) tracers were identified in the Bode aerosols: 2-MGA, two diastereoisomeric 2-methyltetrols (2-MTLs) and three C5-alkene triols. Their total concentrations ranged from 38.8-444 ng m^{-3} (174±86.2 ng m^{-3}) with the maximum (236±87.2 ng m^{-3}) in the monsoon season (Table 1). During the post-monsoon and pre-monsoon periods, their concentrations were similar, and a little lower than those during the monsoon (Fig. 5d) while being the lowest during winter. Their seasonal variation was in agreement with the ambient temperature (Fig S1a), which can influence the isoprene emissions and the photochemical processes (Shen et al., 2015;Wang et al., 2008). The annual average concentration was higher than the urban sites reported from Beijing (44.3 ng m^{-3}) and Kunming (108 ng m^{-3}) (Ding et al., 2016a), even one to two orders of magnitude higher than that from global oceans and the Arctic (Hu et al., 2013). Among I-SOA tracers, 2-MTLs were the major components (51.0%±10.5%) (Fig. 6), with an annual average of 94.4±58.9 ng m^{-3} (ranging from 10.9 to 270 ng m^{-3}). Strong correlations were exhibited between the two isomers during all the seasons (Fig. S5a), implying that they formed through the similar pathway (Shen et al., 2015;Fu et al., 2010). The daily concentration of 2-MGA ranged from 7.10-79.0 ng m^{-3} with an annual average of 34.2±14.8 ng m^{-3}. For C5-alkene triols, the average concentration was 45.0±29.4 ng m^{-3}. They positively correlated with 2-MTLs (Fig. S5b), indicating they were also the oxidation products of isoprene under low-NOx conditions.

According to the reaction chamber results from Surratt et al. (2010), the formation mechanism of 2-MGA remarkably differs from 2-MTLs. 2-MGA is formed under high-NO_x condition while 2-MTLs are mainly produced under low-NO_x or NO_x-free conditions. The formation of 2-MGA can be enhanced under lower RH condition, while it is opposite for 2-MTLs (Zhang et al., 2011). During the monsoon season, due to the conducive conditions of high temperature, high RH (>70 %) (Fig. S1b), high solar radiation and fully-grown plants, the isoprene emissions was large. In addition, NOx during this season was much lower than other seasons. Therefore, 2-MGA/2-MTLs ratios exhibited the lowest values (0.20±0.08) in the aerosol samples during this wet season (Fig. 7). In contrast, 2-MGA/2-MTLs ratios increased up to 0.95 in winter, owing to the lowest temperature and RH of the whole year (Fig. 7) and the higher NO_x concentration in the KV (Kondo et al., 2005;Kiros et al., 2016). NO_x from anthropogenic sources (industry, transportation, BB in the houses as well as in the field) and meteorological conditions with reduced mixing layer heights in winter would also favor the formation of 2-MGA and subsequently increase the 2-MGA/2-MTLs ratio.

Positive correlations were observed between 2-MGA, SO_4^{2-} and NO_3^- (Fig. 8). Budisulistiorini et al. (2017) investigated that the concentrations of B-SOA could significantly increase as the aerosol acidity enhances based on the laboratory simulations and field observations. The significant influence of I-SOA by SO_4^{2-} might be explained by the concerted nucleophilic addition to the key intermediates in the gas phase (e.g., isoprene epoxydiols), which is the rate-determining step in SOA formation (Xu et al., 2015;Li et al., 2018). Li et al. (2018) reported that SO_4^{2-} plays an important part in promoting aqueous phase oxidation of I-SOA tracers. There may be the similar effect of NO_3^- on the SOA formation that needs further research. Therefore, the increase of SO_4^{2-} and NO_3^- could effectively facilitate the ring-opening reaction of isoprene epoxydiols and the SOA formation. Thus, the higher 2-MGA in the KV may be due to the abundant SO_4^{2-} and NO_3^- during pre-monsoon season when most of the brick kilns (more than 100) are operational. Our finding demonstrated the anthropogenic pollutants (e.g., SO_2, NO_x, etc.) can be conductive to accelerating the oxidation of B-VOCs and enhancing the ambient concentrations of B-SOA.

3.6.2 Terpene SOA tracers

Besides isoprene tracers, we also measured four monoterpene oxidation products (M-SOA tracers), including PNA, PA, 3-HGA and MBTCA (Claeys et al., 2007). They are produced through the photooxidation of monoterpenes with ozone and hydroxyl radical (Iinuma et al., 2004). The annual average concentration of the total M-SOA tracers was 59.3±24.6 ng m^{-3} (Table 1). The concentration of M-SOA tracers was higher than those investigated in previous studies from the urban site in Kunming (annual average: 44.1±38.8 ng m^{-3}) (Ding et al., 2016b), three cities (Ohio, Michigan and California) in North America (summer: 30.4–60.6 ng m^{-3}) (Stone et al., 2009) and a forest site in Hyytiälä, Europe (summer: 15.1−33.3 ng m^{-3}) (Kourtchev et al., 2005).

For the seasonal variation, relatively high concentrations of M-SOA tracers occurred during pre-monsoon and post-monsoon seasons (Fig. 5e, f, g, h, and i). Interestingly, there is intensive BB in KV twice a year (forest fires and crop-residue fires during April to May, and crop-residue fires during October to November) discussed in section 3.3.1 and 3.4, which may have been associated with high concentrations of M-SOA tracers. During the fires, substantial amounts of aerosols and VOCs including isoprene and monoterpenes would generate, which can enhance the levels of B-SOA tracers (Jathar et al., 2014;Ding et al., 2013). Good correlations were obtained between the BB tracer i.e. levoglucosan and the higher generation oxidation products (e.g., 3-HGA and MBTCA, R^2=0.32 and R^2=0.53 respectively) in the Bode aerosols (Fig. S6). The forests in the KV mainly consist of broad-leaved evergreen mixed forest, oak-laurel forest and oak forest as well as the conifer tree species (Department of Plant Resources, 2015;Sarkar et al., 2016). Monoterpenes were mainly released from coniferous trees (Kang et al., 2018). Therefore, it suggested that the atmospheric aerosol compositions especially of SOA tracers over KV maybe significantly affected by BB activities.

Sesquiterpenes (e.g., *ß*-caryophyllene) are also among the biogenic SOA (B-SOA) precursors emitted from trees, which have been observed in the troposphere in a lot of studies. *ß*-CA is the tracer of *ß*-caryophyllene SOA and its concentration in the Bode aerosols was 6.31±3.86 ng m^{-3} with the range of 1.53 to 18.5 ng m^{-3}. It shared the similar seasonal variation with M-SOA tracers and positively correlated with them, indicating the possible common emission pattern.

3.6.3 Aromatic SOA tracer

Anthropogenic SOA is also an important OC source. DHOPA is a tracer of anthropogenic SOA from aromatics. In this study, DHOPA showed higher levels in winter and pre-monsoon periods while lower in monsoon season (Fig. 5). Though the major emissions of aromatics come from fossil sources, BB is also considered as an possible source in some sites of the world (Shen et al., 2015). There was a good correlation between DHOPA and levoglucosan (Fig. 9), especially during pre-monsoon with the value of R^2 at 0.73. This indicated BB emissions are also significant source of DHOPA in Bode.

3.7 Estimation of the contributions of different sources to OC

As discussed above, both the primary and secondary sources have influence on OC in the atmospheric aerosols of the KV. In this part, we will apply the tracer-based methods to evaluate the different sources' contributions to OC. It should be noted here that tracer methods can provide a reasonable estimation, but uncertainties are introduced considering the site differences and the lack of representative source profiles for the given study location. The contribution evaluated from each source to OC in the current study is still inferable.

3.7.1 BB-derived OC

Levoglucosan to OC ratios (Lev/OC) detected in source samples has been used in a wide range for quantitative estimation of the BB contribution to OC (Stone et al., 2012;Zhang et al., 2015;Wan et al., 2017), although the ratios vary among different types of biomass burnt and combustion conditions (Bhattarai et al., 2019). An average of 8.14% (8.0% to 8.2 %) for Lev/OC from the burning sites of biofuel, savanna, crop residues, tropical forests, and so on was documented in (Andreae and Merlet, 2001). Zhang et al. (2007) obtained the Lev/OC ratios ranging from 5.4%-11.8% (an average of 8.27%) from the aerosols of cereal straw (wheat, corn and rice) combustion. Sheesley et al. (2003) reported an average of 7.94% of levoglucosan from the combustion of biomass (including rice straw, biomass briquettes, dried cow-dung patties, etc.) indigenous to South Asia. However, the ratio acquired from the hardwood burning in fireplaces and stoves in the US was 14%, which was applied at the background sites in Europe (Fine et al., 2004). Stone et al. (2012) used the Lev/OC ratio of 12% ± 0.2% during the burning of acacia wood at Godavari in the KV for the CBM profile source apportionment. The mean value of Lev/OC value of BB from main biomass types was 10.1%. In this study, we choose the mostly used values of 8.14% for BB estimation (Graham et al., 2002;Fu et al., 2014;Ho et al., 2014;Sang et al., 2011;Zhu et al., 2016;Mkoma et al., 2013). In addition, the uncertainties of using different ratios were also calculated (see

Table S3). The diagnostic ratios among molecular tracers and OC (e.g., Lev/OC) from direct emissions are critical for more precise results. It's meaningful to understand the emission characteristics for individual OC emission categories, as well as in different locations, especially in South Asia.

Figures 10 and 11 present the monthly concentration variations of BB-OC and contribution of BB-OC to OC, respectively. Current estimation exhibited BB-OC contributed 24.9±10.4% (varying from 6.32% to 61.5%) to OC throughout the year in Bode aerosols (Fig. 11a). This was higher than the study in Lumbini in Nepal (19.8±19.4%) (Wan et al., 2017), and nearly twice of the BB-OC contribution to OC reported in Hong Kong (6.5%–11%) (13.1 %) (Sang et al., 2011) and the Pearl River Delta in China (Ho et al., 2014). Moreover, the contribution of BB-OC to OC in current study maximized in the post-monsoon season (36.3±10.4%), higher than that in the pre-monsoon (28.5±10.3%) and winter (27.9±8.63%) periods. These results indicate that BB severely affect the air quality in the KV, especially during the post-monsoon period. Similarly, Stone et al. (2010) reported 21±2% of OC in $PM_{2.5}$ from Godavari rural site in the outskirts of the KV during 2006, was also attributed to the primary BB sources.

3.7.2 Plant-debris-OC and fungal-spore-derived OC

Primary biological aerosol particle (PBAP) has been identified as an important source using tracers (section 3.3.2). They are likely to have a big contribution to the aerosols in Bode. In order to reveal how much they are contributing to organic aerosols, "total" plant debris was calculated based on glucose following the equation (Zhu et al., 2016;Puxbaum and Tenze-Kunit, 2003):

Cellulose (μg) = D-glucose (μg) × GF × (1/SY);

Plant debris=2 × cellulose.

where GF (0.90) is the glucose/cellulose weight conversion factor and SY (0.717) is the saccharification yield.

For OC fraction derived from fungal spores, it was estimated using mannitol levels according to the studies from Bauer et al. (2008) and Holden et al. (2011), i.e., there was 1.7 pg mannitol and 13 pg OC per spore.

As shown in Fig. 11a, fungal spore-derived OC and plant-debris-OC annually contribute to 3.15±2.86% and 1.42±1.03% of OC, respectively. The contributions were both higher in the monsoon season, with 5.85±2.50% for fungal spore-derived OC and 2.29±0.79% for plant-debris-OC to OC, respectively (Fig. 11c). During winter, the contributions were the lowest due to the inactive vegetation. There are also some similar results from the literatures. For example, Zhu et al. (2016) reported the plant debris contribution to OC was 5.6% and 4.6% during nighttime and daytime respectively from aerosols in a mid-latitudinal forest. Szidat et al. (2006) reported the plant debris contributed to 3.2% of OC during summer in urban aerosols collected in Zurich, Switzerland. The contributions of fungal aerosol to OC was 8% in the aerosols from a Brazil urban site (Emygdio et al., 2018). Liang et al. (2017) reported the fungal aerosol contributions of 3.5 ± 3.7% in aerosols from a rural site in Beijing, China. In marine aerosols, the fungal spores was documented the major contributor to total OC with 3.1 % (0.03 %–19.8 %) over the East China Sea (Kang et al., 2018). All above strengthened the importance of plant-debris and fungal spores to the aerosol burden in the atmosphere.

3.7.3 Biogenic SOC and anthropogenic toluene SOC

Biogenic secondary organic carbon (B-SOC) and anthropogenic aromatic SOC (A-SOC) from the oxidation of isoprene, monoterpenes, sesquiterpene and toluene were assessed using the tracer-based method proposed by Kleindienst et al. (2007). This method has been applied successfully in numerous aerosol studies (Shen et al., 2015;Ding et al., 2016a;Kang et al., 2018). The mass fraction of tracer compounds in SOC (FSOC) for an individual precursor was calculate based on the smog chamber simulations. The calculation formula as following:

$$FSOC = \frac{\sum_i [\mathrm{tri}]}{[SOC]}$$

where [tri] is the concentration of tracer i and [SOC] is the concentration of SOC. The conversion factors of FSOC were 0.155±0.039, 0.231±0.111, 0.0230±0.0046 and 0.0079±0.0026 μg μgC^{-1} for isoprene, monoterpenes, sesquiterpene and toluene, respectively (Kleindienst et al., 2007).

The total calculated concentrations of B-SOC vaired from 0.41 to 2.77 μg m^{-3} with an annual mean concentration of 1.36±0.49 μg m^{-3}, a higher concentration of 1.43±0.48 μg m^{-3} in monsoon and lower concentration of 0.86±0.20 μg m^{-3} in winter (Fig 10g). The B-SOC/OC showed a higher average percentage of 10.1%±3.34% in the monsoon season (Fig 11c), indicating B-SOC was an important OC sources in Bode during this period. During post-monsoon, B-SOC/OC declined to 5.36% (Fig 11d). The B-SOC/OC showed the lowest value of 1.52%±0.70% in winter (Fig 11e), indicating that B-SOC had minor contributions to elevated OC in winter. The annual average concentration of A-SOC was 2.45± 1.45 μg m^{-3}, which is higher than the B-SOC. The highest A-SOC concentration was obtained in winter (3.27±1.25 μg m^{-3}) (Fig.10h). A-SOC was the second most important contributor to OC after BB-OC. It is not only derived from increased fossil fuel combustion and the subsequent oxidation, but also from BB emissions.

In total, SOC (including B-SOC and A-SOC) reconstructed using the formula above in this section was 3.81±1.63 μg m^{-3}, accounting for 15.0%±8.99% of OC.

3.7.4 Possible sources of the unidentified OC

On the whole, BB contributed one-fourth (24.9%±10.4%) of the OC in Bode, followed by A-SOC (8.82%±5.55%), B-SOC (6.19%±4.49%), fungal-spores (3.15%±2.86%) and plant-debris (1.42%± 1.03%) (Fig. 11a). Nevertheless, there is still part of OC (55.5%) that we were not able to be attributed to any specific sources based on the tracers analyzed in current study. There are partly uncertainties caused by the organic tracer analyses (estimation of measurement uncertainty was shown in Table S2). Furthermore, fossil fuel combustion and soil dust could be also notable fractions of OC in Bode aerosols. Additionally, low molecular weight (LMW) dicarboxylic acids from both primary and secondary sources is also a remarkable contributor to atmospheric organic aerosols (Kawamura and Bikkina, 2016). Humic-like substances and amines can constitute another fraction of OC, but not well studied (Wu et al., 2018;Laskin et al., 2015). Therefore, the possible contributions of the unidentified OC (55.5%) from various sectors need further investigation, which is better to comprehensively understand the sources of South Asian aerosols and will be very useful for the targeted pollution control measures in this region.

4. Summary and conclusions

Field measurements of atmospheric aerosols were conducted in a semi-urban site (Bode) of the KV, Nepal, from April 2013 to April 2014. The organic tracers from primary and secondary organic aerosols were determined. A distinctive seasonality was observed for various aerosol species. Higher concentrations of OC, EC, anhydrosugars, phenolic compounds and resin acid were observed in winter and pre-monsoon seasons, while their concentrations were lower during the wet (monsoon) period. Levoglucosan was the dominant species of the total identified tracers with an average concentration of 788 ng m^{-3}. We observed high abundances of monosaccharides in the pre-monsoon season and of sugar alcohols in the wet period, and lower levels in winter because of the reduced plant activities. I-SOA tracers represented a majority among B-SOA tracers with a maximum in the monsoon. The seasonal variation of M-SOA tracers was controlled by monoterpenes emission and BB. DHOPA exhibited higher concentrations during the winter and pre-monsoon seasons.

The likely OC sources were further evaluated for their contributions to observed total OC using tracer-based methods. BB-OC contributed a major fraction (24.9%) to OC in Bode, followed by A-SOC (8.8%), B-SOC (6.2%), fungal spores (3.2%) and plant debris (1.4%). The highest contribution of BB-OC, 36.3%, occurred during post-monsoon season. A-SOC, B-SOC, fungal spores and plant debris all made larger contributions during the monsoon. The higher BB-OC and the A-SOC contributions imply that some BB and anthropogenic components are widespread in the KV and thus represent the main contributors affecting the regional air quality in the KV region.

The present study clearly shows that the chemical constituents and sources of OC strongly vary with seasons, as a result of diverse air pollution sources in the valley across four seasons. The heavy BB and the subsequent oxidant emissions are anticipated to cause larger contributions of B-SOC to OC. Understanding OC's climate impacts is a frontier area of research, because a large uncertainty still exists in the estimation of OC radiative forcing. Our study implies that since BB is a major source of ambient OC, the fraction of OC that absorbs light (referred to as brown carbon) and also acts as cloud condensation nuclei, needs to be further studied in order to better understand radiative effects of OC on regional climate change. The current source contribution estimates from the tracer-based methods do not accurately evaluate the large temporal variations from all kinds of sources. Contributions from other sectors (ca 55.5%), including low molecular weight dicarboxylic acids (Kawamura and Sakaguchi, 1999;Kawamura and Bikkina, 2016), need further investigation to better understand the atmospheric aerosols from both urban and rural sources such as the KV and other sites in the Himalayan foothills and the Indo-Gangetic Plain regions. These observations of the severe air pollution, particularly the particular matter pollution, provide valuable support for air pollution control measures, especially in determining which sources and sectors to first focus on the KV and the surrounding region, in order to reduce the air pollution from being severe to become much cleaner in the near future. In addition, the current study based on the molecular level-source apportionment of OC in heavy polluted region from South Asia provides a much more specific quantification of source estimation for OC, which is different from previous studies based on the bulk carbonaceous aerosol using radiocarbon (^{14}C) measurements, PMF and CBM.

There are additional improvements for future studies to be addressed in the supporting information file. The key recommendations are as follows: (i) much more tracers need to be identified to explain the other sources of organic aerosols in the KV; (ii) the conversion factors of tracers to organic carbon from local emissions are critical for more precise source apportionments and therefore, a number of studies on the emission characteristics will be valuable; (iii) comprehensive methods (e.g., carbon isotope and modeling) need to be integrated together for the source apportionment of organic aerosols in the KV; (iv) the influences of BB on the formation of secondary organic aerosols could be further studied, especially during the heavily polluted dry season, with additional simultaneous measurements of precursors (e.g., NOx and O_3), $PM_{2.5}$ and so on at the same time; (v) to better understand the atmospheric processes of various chemical species, investigations of size-segregated aerosols are especially needed in the heavy polluted KV.

Data availability. Raw data are archived at the Institute of Tibetan Plateau Research, Chinese Academy of Sciences, and are available on request by contacting the corresponding author.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1 Concentrations of TSP, major ions, OC, EC (μg m^{-3}) and molecular tracers in the aerosols from Bode, Kathmandu Valley (ng m^{-3}).

	Annual			Pre-monsoon			Monsoon			Post-monsoon			Winter		
Compounds	Mean	Median	SD	Mean	Median	SD	Mean	Median	SD	Mean	Median	SD	Mean	Median	SD
TSP	256	213	166	381	366	171	120	107	52.4	225	254	71.6	353	348	68.5
SO_4^{2-}	10.8	6.15	9.83	17.2	16	7.49	4.1	2.6	4.04	4.81	4.24	2.38	27.3	24.1	6.79
NO_3^-	5.21	3.8	4.35	8.82	8.41	4.41	2.34	1.85	1.65	3.52	3.65	0.92	9.47	8.11	4.43
NH_4^+	5.92	3.46	6.16	8.57	6.71	5.65	1.99	1.31	2.58	3.72	3.65	1.48	17.5	15.6	3.06
Ca^{2+}	7.96	5.82	6.85	11.6	8.98	8.97	4.47	3.59	3.66	6.21	6.01	1.51	13.8	15.6	5.02
K^+	2.43	1.5	2.82	3.87	2.42	4.27	1.15	0.94	0.96	1.97	2.03	0.46	4.03	3.39	1.45
Cl^-	2.15	1.18	2.25	2.72	2.27	1.83	0.73	0.67	0.32	1.68	1.78	0.4	6.94	7.87	2.23
Na^+	3.28	2.93	1.58	3.15	1.68	2.38	3.3	3.61	1.18	2.8	2.84	0.12	4.21	4.28	0.49
Mg^{2+}	0.61	0.39	0.54	0.95	0.67	0.7	0.32	0.24	0.25	0.41	0.4	0.07	1.07	1.09	0.39
OC	38.7	24.2	32.7	59.4	46.9	37.9	14.6	14.7	3.76	31.8	32.9	12.7	62.8	53.6	20.6
EC	9.92	9.34	5.33	14.4	13.85	5.24	5.61	4.76	1.8	9.37	9.95	2.19	11	9.89	3.48
OC/EC	3.78	3.09	2.37	4.44	3.29	3.23	2.71	2.54	0.69	3.31	3.01	0.93	5.86	5.3	1.75
Anhydrosugars															
Levoglucosan	788	631	685	1214	900	705	204	188	60.1	863	836	252	1391	1120	535
Galactosan	44.4	33.8	40.3	68.5	51.4	42.7	13.2	13.1	5.55	34.8	33.8	10.9	85.6	72.4	32.8
Mannosan	50.6	34.5	45.1	71.8	61.8	42.3	16	16.7	5.5	39.5	39.6	12.5	116	106	45
Subtotal	883	688	765	1354	974	781	233	218	68.2	937	922	273	1592	1268	611
Monosaccharides															
Glucose	124	114	60	137	118	65.1	129	122	47	143	149	48.7	39.9	38.7	12.5
Fructose	58.2	53.2	28.3	68.4	66.9	29.1	57.1	48.2	26.8	58.2	54.7	21	27	22.5	10.2
Trehalose	40.8	35.6	22	48	53.1	22.4	40.2	35.5	22.8	38.9	40.1	12.5	20.1	16.8	9.17
Sucrose	48.3	40.3	27.4	64.7	55.5	31.6	38.7	38.7	11.9	56.6	45	27.1	18.4	16.6	6.37
Xylose	26.5	20.5	18.1	37.8	30.9	20	13.2	13.4	5.68	24.9	26.5	9.33	38.6	37.5	14.1
Subtotal	298	285	127	356	333	141	278	249	93.5	322	318	104	144	135	35.8
Sugar alcohols															
Mannitol	86.9	77	55.3	84.6	78.8	38.7	114	102	61.4	63.4	53.6	34.6	18.1	19.3	6.02
Arabitol	68.4	60.6	39.8	68.9	65.3	30.3	86.6	68.9	44.5	42.6	33.5	22.5	26.1	24.7	9.13
Sorbitol	14.2	12.7	8.02	13.1	12.6	5.96	17.9	15.9	9.31	13.1	13.2	4.65	5.82	4.97	2.72
Erythtitol	43.1	36.5	28.8	35.8	34	15.4	56.9	47.5	33.1	48.6	39.1	31.8	12.4	10.2	7.6
Subtotal	213	192	126	202	198	84.8	275	245	143	168	151	80.1	62.5	61.8	19.7
Total sugars	1394	1206	813	1913	1570	919	787	727	249	1427	1367	327	1798	1462	651
Phenolic compounds and resin acid															
Vanillic acid	15.3	11.3	11.3	20.8	15.8	12.9	7.1	6.84	1.94	14.3	12.3	4.95	26.9	30.5	9.37
Syringic acid	17.1	11.6	13.7	23.7	17.1	15.6	7.82	7.52	2.67	13.7	12.5	4.54	32	37.8	12.3
p-Hydroxybenzoic acid	19.8	15.5	12.3	26.1	19.3	16.7	14.5	13	4.57	14.2	14.6	3.76	23.6	23.1	9.15
Dehydroabietic acid	13.8	12.6	6.19	16.3	15.4	6.52	10.3	9.7	2.32	10.9	9.3	3.1	21.4	20.1	6.76
Subtotal	66.1	49.7	41.4	86.9	67	49.2	39.7	39.6	9.22	53	47.5	15.6	104	112.8	36.2

	Annual			Pre-monsoon			Monsoon			Post-monsoon			Winter		
Compounds	Mean	Median	SD	Mean	Median	SD	Mean	Median	SD	Mean	Median	SD	Mean	Median	SD
Phthalic acid esters															
Diethyl (DEP)	16.6	15.6	8.41	19.4	17.5	11.6	15.4	15.5	4.42	14.9	13	6.25	12.9	9.3	6.49
Di-n-butyl (DnBP)	56.2	48.5	25.6	63.6	59.8	30.7	52.2	48.4	20.9	55.4	45.3	22.8	46.4	41.3	21.8
Bis-(2-ethylhexyl) (DEHP)	438	378	200	495	466	239	407	377	162	431	353	177	361	322	170
Subtotal	510	444	230	578	545	276	474	445	184	501	408	204	420	372	196
Isoprene tracers															
2-Methylglyceric acid	34.2	30.2	14.8	45.9	48.8	15.2	25.1	24.5	6.91	36.2	34.9	9.86	25	24.7	8.56
2-Methylthreitol	30.4	27.3	19.5	22	22.8	10	45	44.9	20.6	27.4	26.3	7.39	8.4	7.9	3.83
2-Methylerythritol	64.1	58.3	39.6	45	44.5	20.4	97.3	98.9	38	53	52	9.78	18.5	18.7	4.63
2-Methylterols[a]	94.4	84.3	58.9	67	68.1	30.4	142	142	58.3	80.5	78.3	17	27	26.6	8.39
C5-Alkene triols[b]	45	39.2	29.4	30.4	28.8	13.4	68.8	69.1	30	35.1	30.6	18	17.5	15.3	4.88
Subtotal	174	160	86.2	144	150	53.4	236	220	87.2	152	145	39.3	69.5	72.4	19.3
Monoterpene tracers															
cis-Pinonic acid	26	24.5	11.6	32.3	30	13.8	21	19.1	8.04	28	29.1	5.92	20.6	20	8.01
Pinic acid	11.9	10.8	4.48	11.4	10.4	3.78	12.7	12.3	5.07	13.7	15.6	5.25	8.83	8.67	0.92
3-Hydroxyglutaric acid	10.6	9.16	6.85	13.9	12.1	7.75	5.75	4.88	3.01	16	15.4	4.18	11.5	10	4.72
3-MBTCA[c]	10.8	9.53	7.36	16.9	15.1	7.32	5.14	4.3	3.12	11.9	9.61	3.63	9.3	8.89	3.75
Subtotal	59.3	55.6	24.6	74.6	72.1	28.3	44.6	42.6	14.4	69.5	65.7	12.6	50.2	49.6	10.8
Sesquiterpene tracer															
β-Caryophyllenic acid	6.43	5.35	3.93	8.61	7.15	4.29	3.66	2.85	2	8.35	7.2	3.02	7.16	6.6	2.76
Total B-SOA tracers[d]	136	234	50.4	133	229	43.9	156	275	53.9	131	217	26.2	75.6	129	14.2
Toluene tracer															
DHOPA[e]	19.4	16.5	11.5	22.8	19	15.5	15	13.4	5.69	17.3	18.2	5.94	25.8	23	9.84
Total SOA tracers[f]	259	249	94.7	250	242	84.9	299	286	101	247	243	48.2	153	162	29.4

[a] Sum of 2-methylthreitol and 2-methylerythritol.

[b] C5-Alkene triols: 3-Methy-2,3,4-trihydroxy-1-butene, cis-2-Methyl-1,3,4-trihydroxy-1-butene and trans-2-Methyl-1,3,4-trihydroxy-1-butene.

[c] 3-MBTCA: 3-methyl-1,2,3-butanetricarboxylic acid.

[d] Sum of 2-methylglyceric acid, 2-methylterols, C5-Alkene triols, cis-pinonic acid, pinic acid, 3-hydroxyglutaric acid and 3-MBTCA.

[e] DHOPA:2,3-dihydroxy-4-oxopentanoic acid.

[f] Sum of 2-methylglyceric acid, 2-methylterols, C5-Alkene triols, cis-pinonic acid, pinic acid, 3-hydroxyglutaric acid and 3-MBTC and 2,3-dihydroxy-4-oxopentanoic acid.

Table 2 Linear correlation coefficients (R^2) among major ions and OC, EC in aerosols in Bode, Kathmandu Valley

	SO_4^{2-}	NO_3^-	NH_4^+	Ca^{2+}	nss-Ca^{2+}	Mg^{2+}	nss-Mg^{2+}	K^+	nss-K^+	Cl^-	Na^+	OC	EC
SO_4^{2-}	1.00												
NO_3^-	0.78**	1.00											
NH_4^+	0.87**	0.69**	1.00										
Ca^{2+}	0.48**	0.58**	0.62**	1.00									
nss-Ca^{2+}	0.48**	0.58**	0.62**	1.00**	1.00								
Mg^{2+}	0.61**	0.71**	0.69**	0.91**	0.91**	1.00							
nss-Mg^{2+}	0.65**	0.76**	0.70**	0.88**	0.88**	1.00**	1.00						
K^+	0.40**	0.59**	0.55**	0.78**	0.78**	0.85**	0.83**	1.00					
nss-K^+	0.40**	0.60**	0.55**	0.77**	0.77**	0.85**	0.83**	1.00**	1.00				
Cl^-	0.67**	0.58**	0.76**	0.41**	0.40**	0.48**	0.50**	0.37**	0.37**	1.00			
Na^+	0.05*	0.08*	0.23**	0.48**	0.48**	0.37**	0.28**	0.40**	0.39**	0.09*	1.00		
OC	0.59**	0.61**	0.57**	0.32**	0.32**	0.48**	0.53**	0.36**	0.36**	0.48**	0.01	1.00	
EC	0.35**	0.43**	0.24**	0.12**	0.12**	0.23**	0.27**	0.16**	0.16**	0.24**	0.02	0.36**	1.00

*: $P<0.1$.

**: $P<0.001$.

Table 3 Linear correlation coefficients (R^2) among monosaccharides and sugar alcohols in aerosols from Bode, Kathmandu Valley

	Glucose	Fructose	Sucrose	Trehalose	Xylose	Mannitol	Arabitol	Sorbitol	Erythtitol
Glucose	1								
Fructose	0.77***	1							
Sucrose	0.55***	0.55***	1						
Trehalose	0.30***	0.23***	0.28***	1					
Xylose	0.001	0.06*	0.19***	0.05*	1				
Mannitol	0.51***	0.51***	0.23***	0.14***	0.002[a]	1			
Arabitol	0.44***	0.50***	0.22***	0.16***	0.0005	0.77***	1		
Sorbitol	0.51***	0.53***	0.20***	0.13**	0.0002	0.83***	0.68***	1	
Erythtitol	0.46***	0.40***	0.16***	0.13**	0.007	0.77***	0.62***	0.77***	1

a Negative values indicate negative correlations.
*p<0.1
**p<0.01.
***p<0.001.

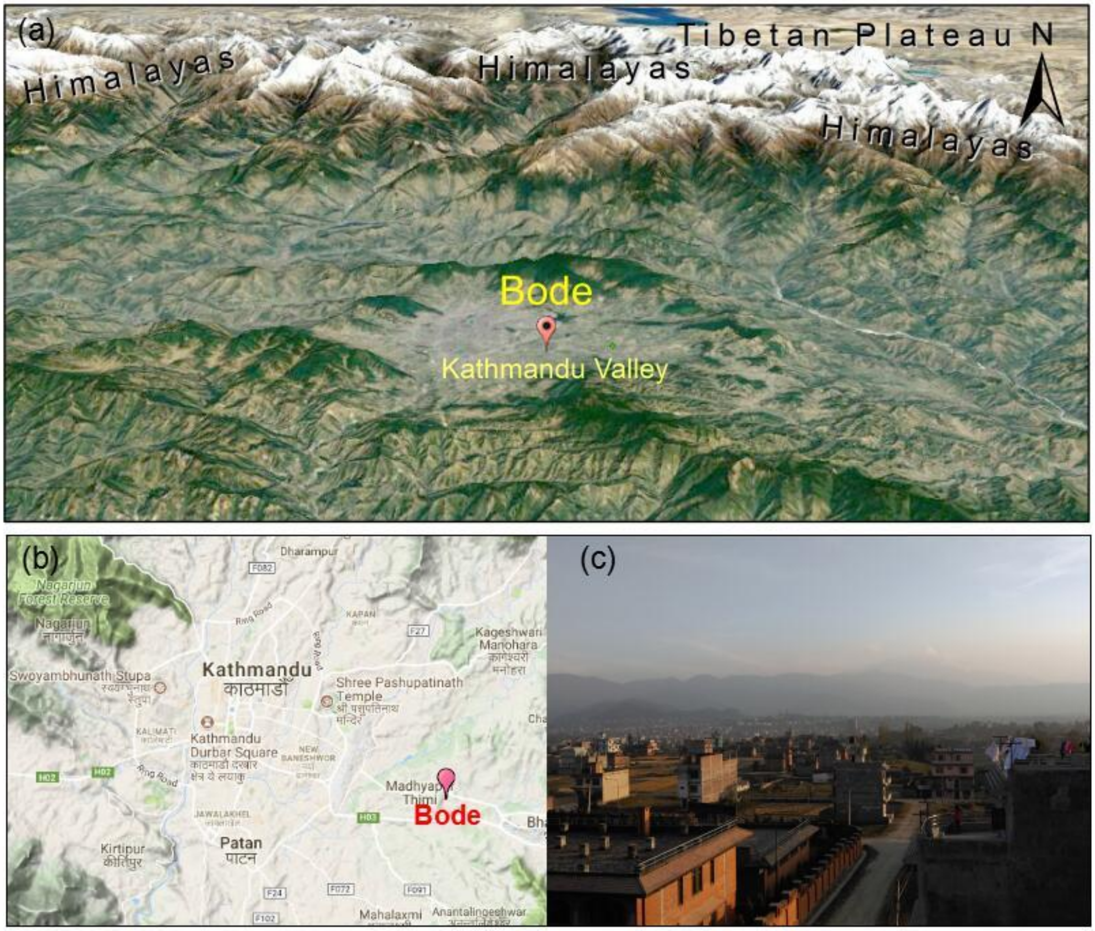


Fig. 1. Location of measurement site: (a) Kathmandu Valley, (b) urban measurement site at Bode in Kathmandu Valley, (c) air pollution observed from the Bode site in the afternoon.

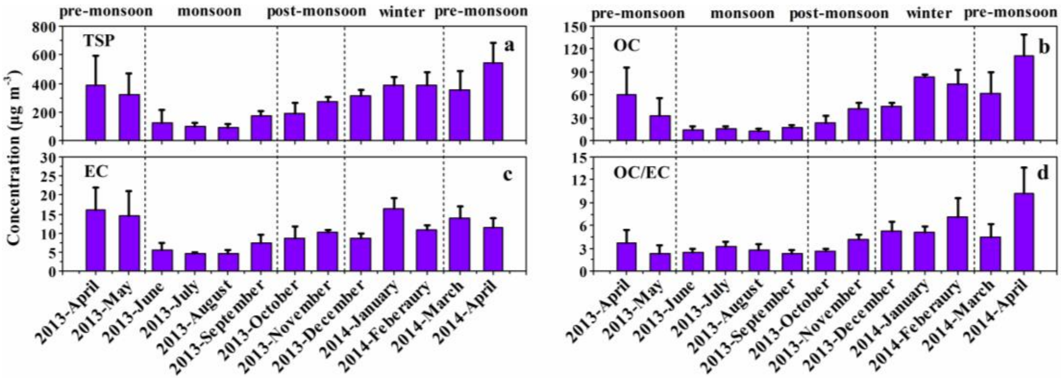


Fig. 2. Monthly variations of TSP, OC, EC, OC/EC ratios at Bode site, Kathmandu Valley during April 2013-April 2014.

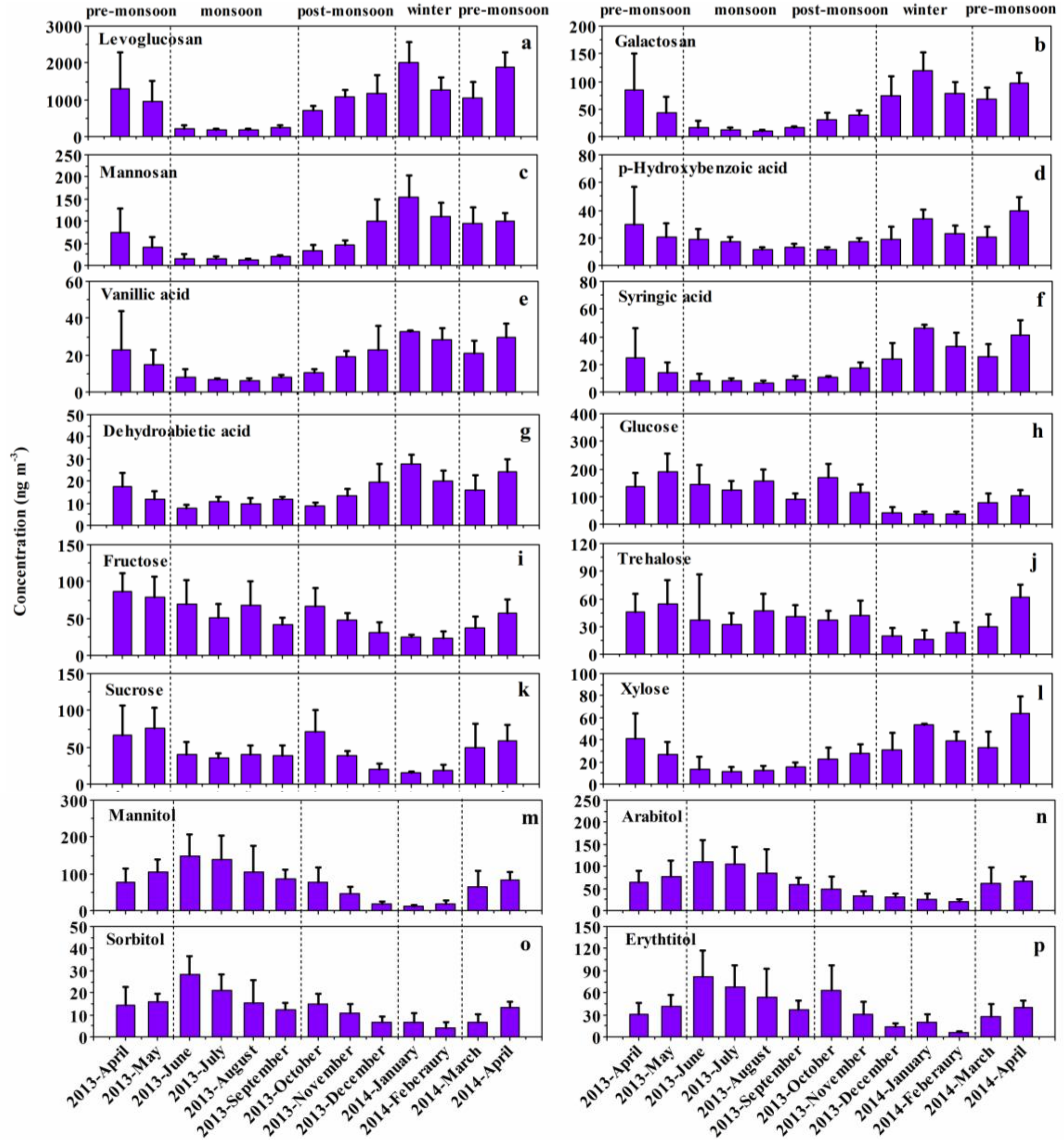
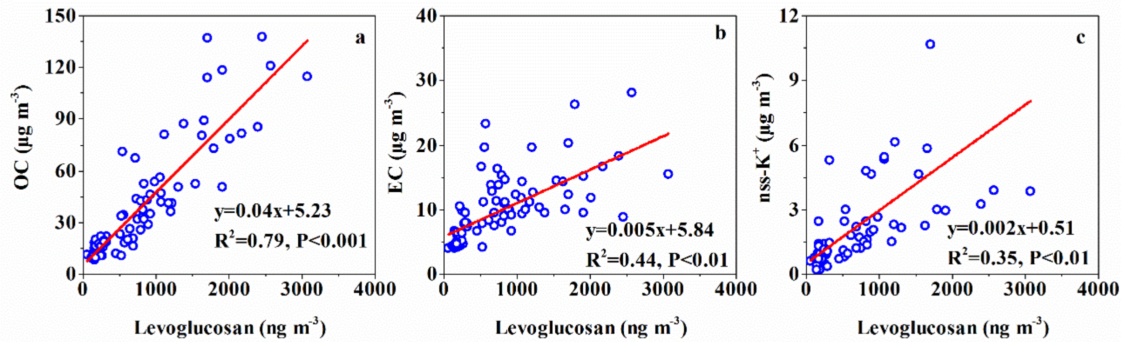


Fig. 3. Monthly variations of biomass burning tracers, monosaccharides and sugar alcohols at Bode site, Kathmandu Valley during April 2013-April 2014.

986 

987 **Fig. 4.** Correlations between (a) levoglucosan and OC, (b) levoglucosan and EC, (c) levoglucosan and 988 nss-K^+ in Bode aerosols during the sampling period (April 2013 to April 2014).

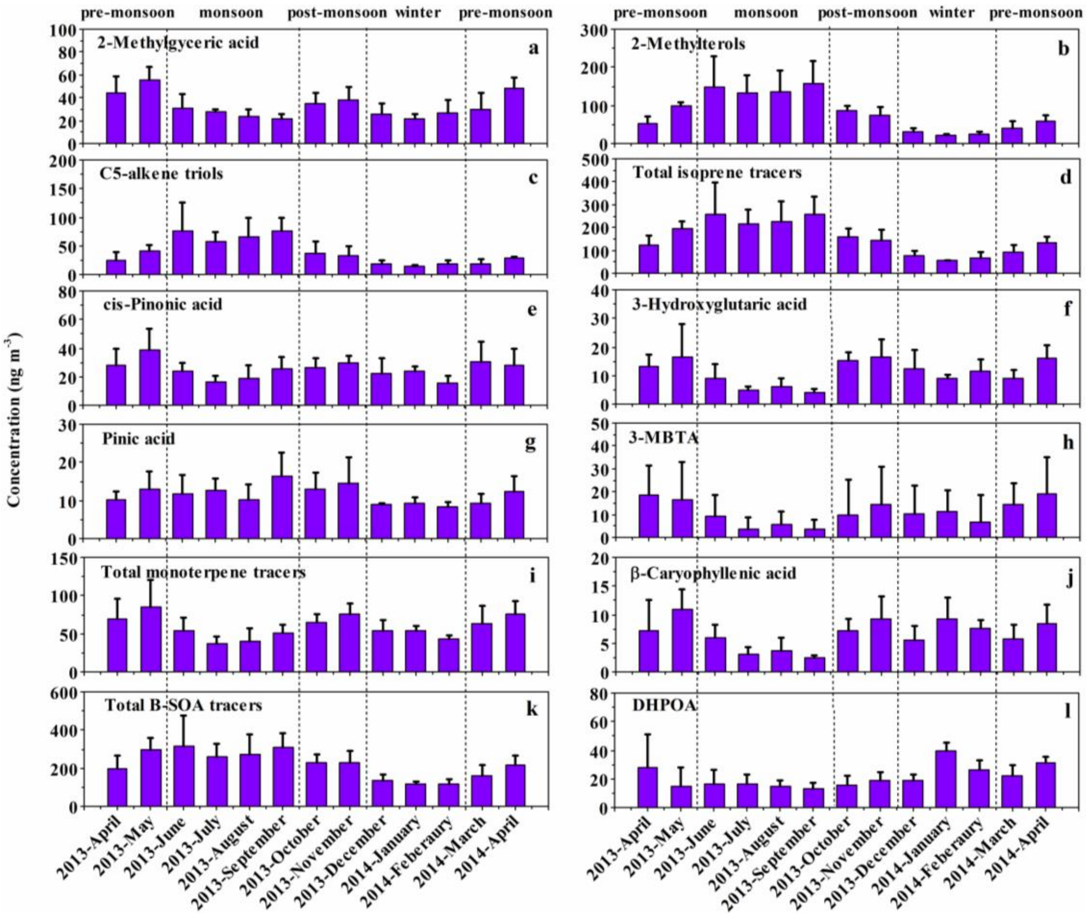


Fig. 5. Monthly variations of B-SOA tracers, total isoprene tracers, total monoterpene tracers, *β*-caryophyllenic acid, total B-SOA tracers and DHPOA at Bode site, Kathmandu Valley during April 2013-April 2014.

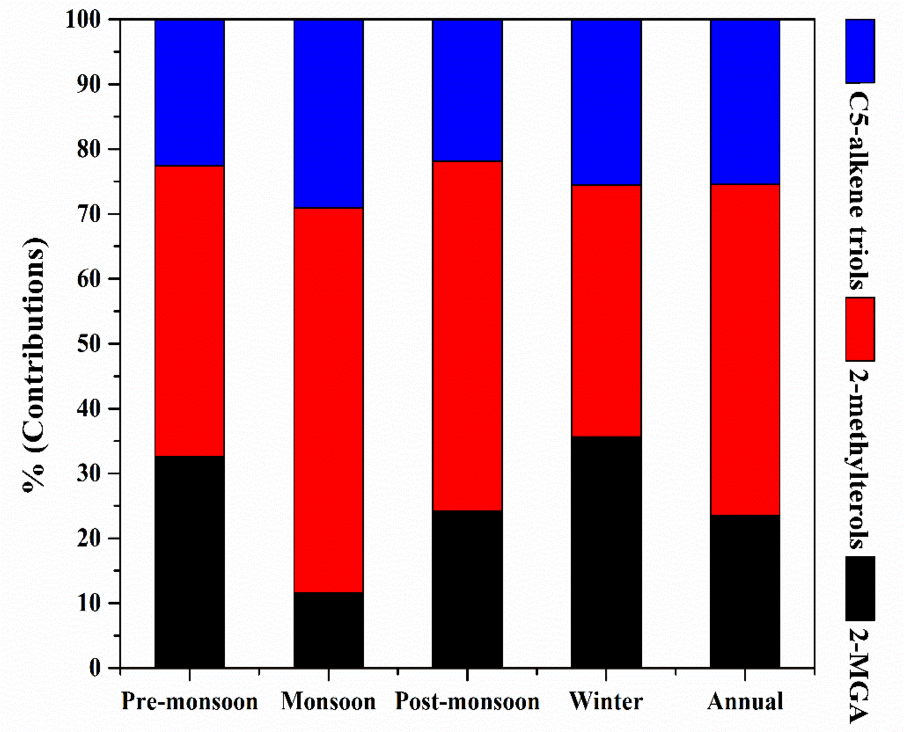


Fig. 6. The percentage contributions of the isoprene SOA tracers to the total during different seasons in the atmospheric aerosols from Kathmandu.

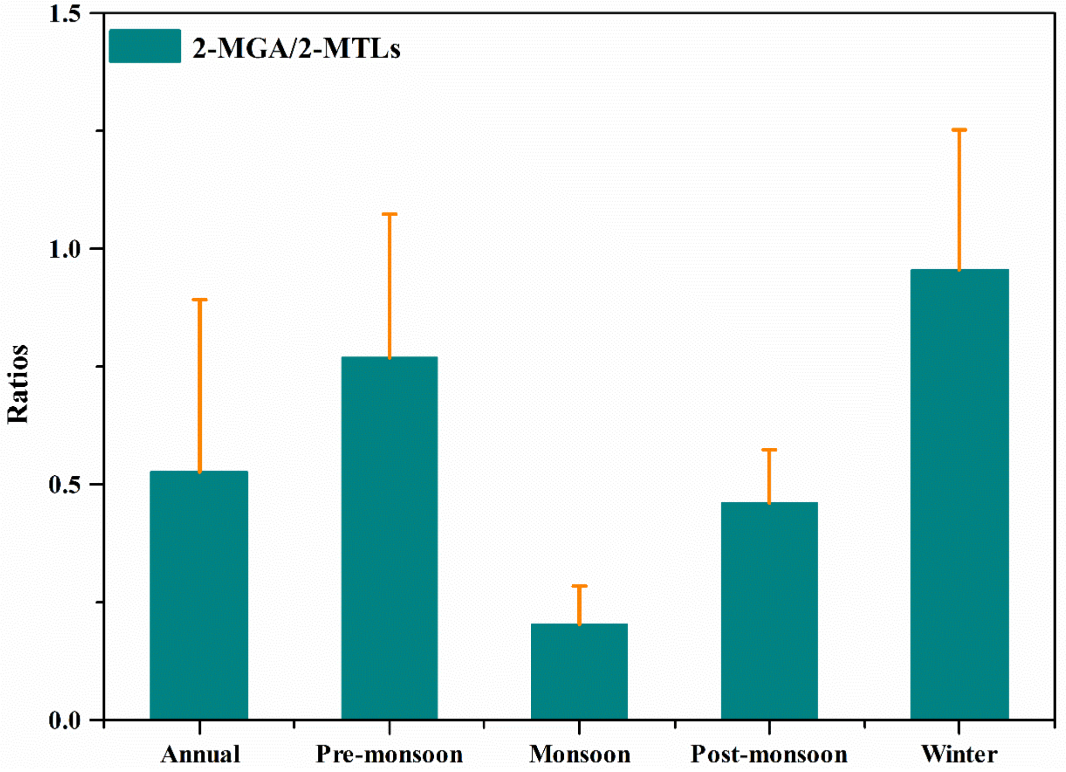


Fig. 7. Ratios of 2-MGA/2-MTLs during different seasons in Bode, Kathmandu.

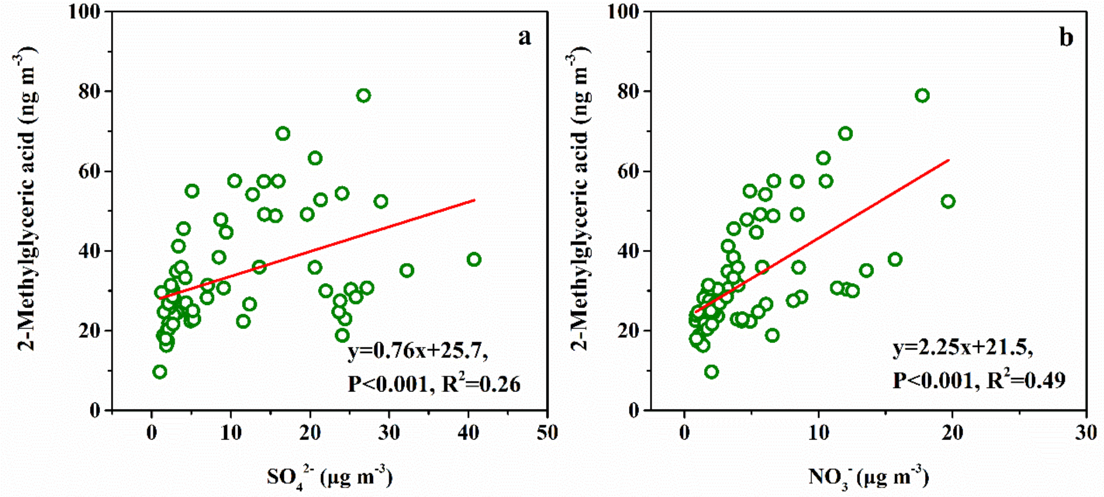


Fig. 8. Concentration correlation between (a) 2-methylglyceric acid (2-MGA) and SO_4^{2-}, (b) 2-methylglyceric acid and NO_3^- in the aerosols from Bode, Kathmandu.

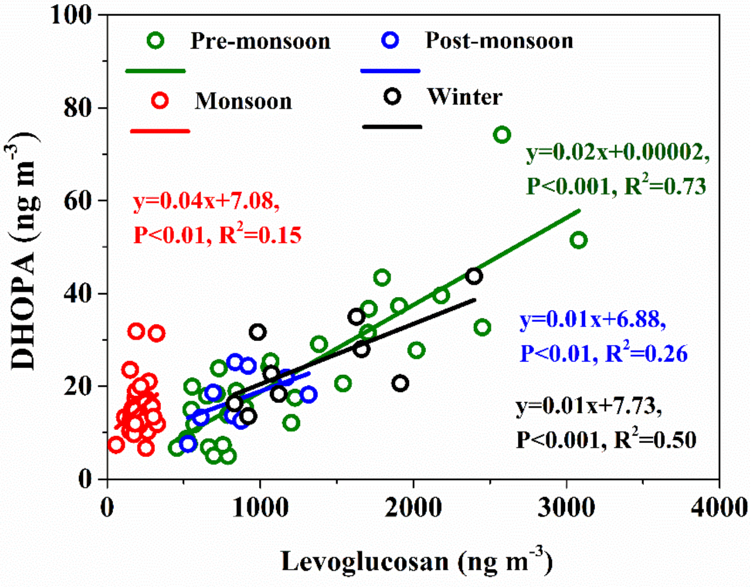


Fig. 9. Correlation between 2,3-dihydroxy-4-oxopentanoic acid (DHOPA) and levoglucosan in Bode aerosols during the sampling period (April 2013 to April 2014).

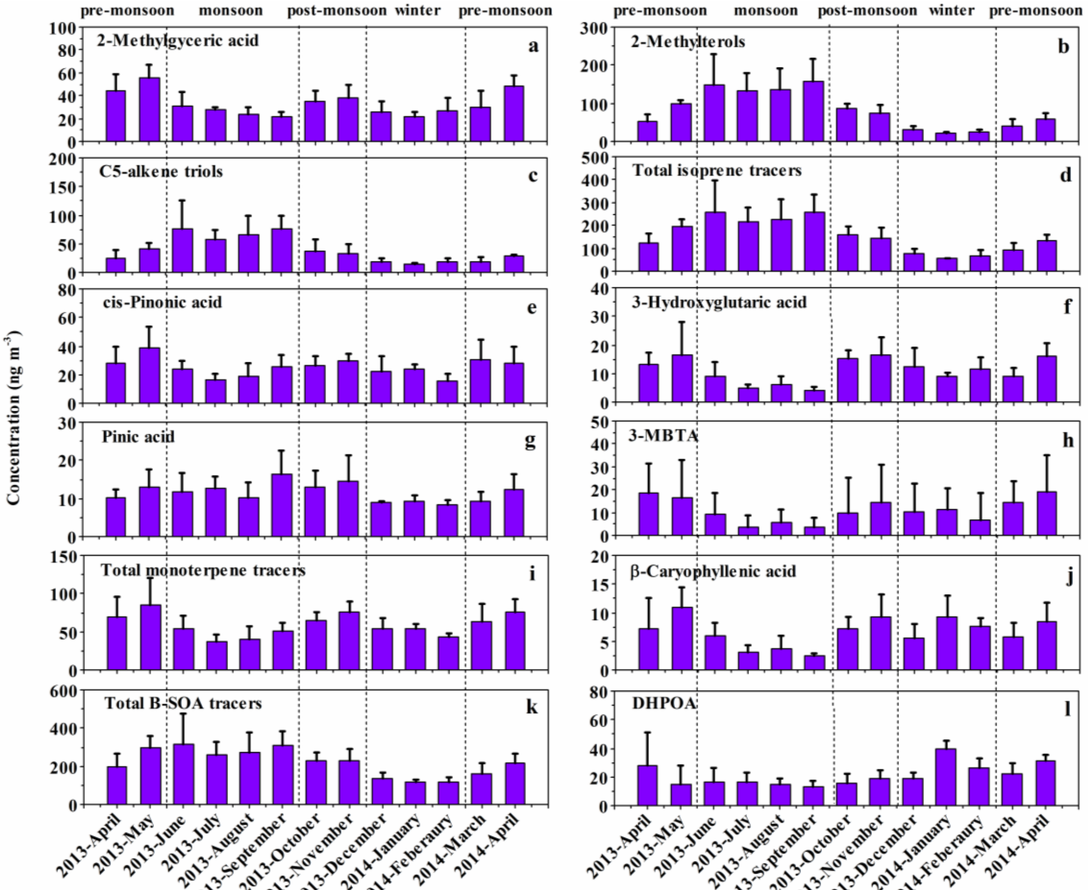


Fig. 10. Monthly concentration variations of (a) BB-OC, (b) plant-debris-OC, (c) fungal-spore-OC, (d) SOC-isoprene (I-SOC), (e) SOC-monoterpenes (M-SOC), (f) SOC-sequiterpene (SOC-C), (g) SOC-isoprene+monoterpenes+sequiterpene (B-SOC), (h) SOC-toluene (A-SOC), and (i) total SOC that were estimated using a tracer-based method at Bode site, Kathmandu Valley during April 2013-April 2014.

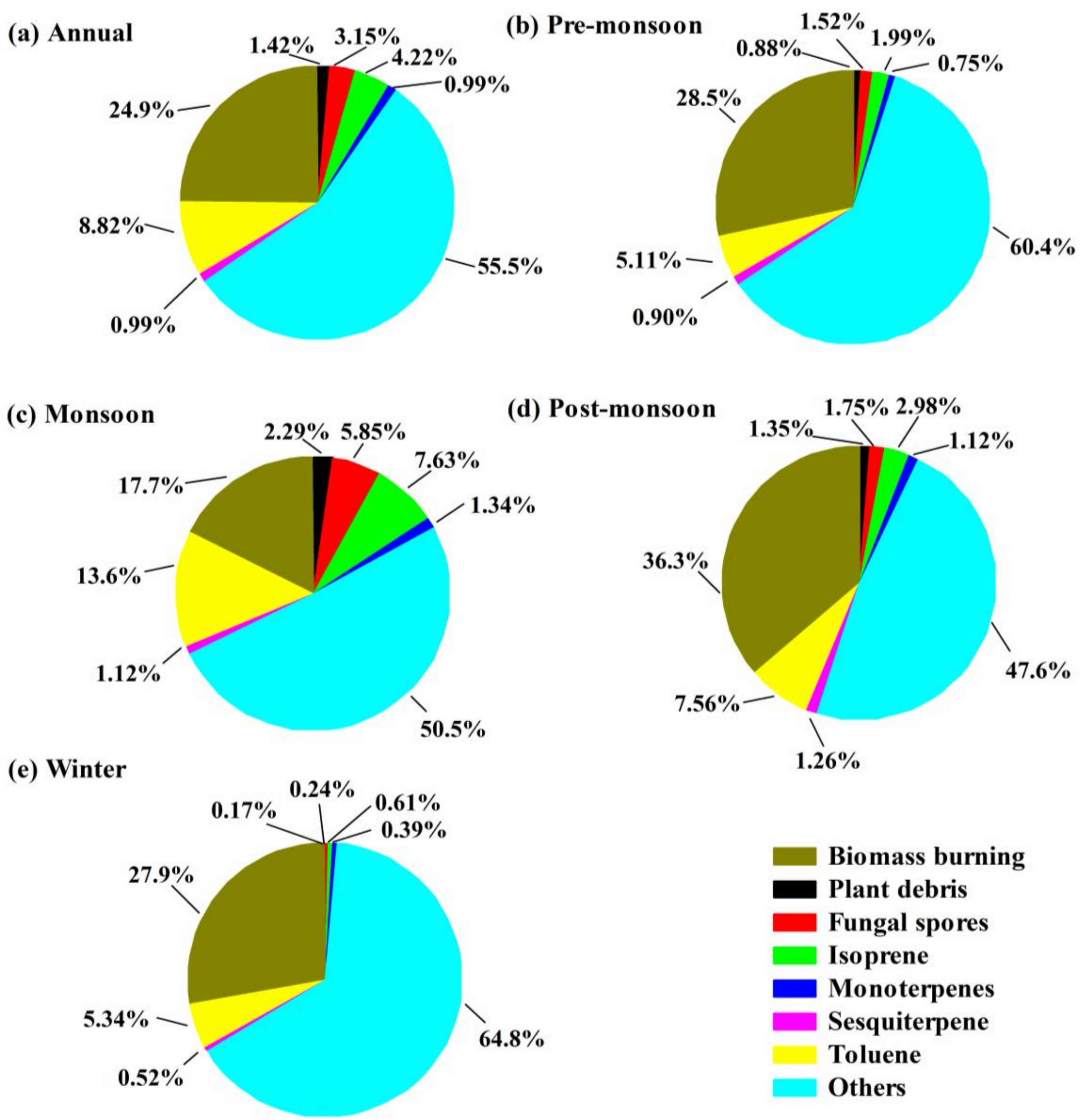


Fig. 11. Pie-charts showing contributions from different sources to organic carbon based on the estimation of tracer-method in Bode, Kathmandu Valley: (a) annual, (b) pre-monsoon, (c) monsoon, (d) post-monsoon and (e) winter.