Research Article

# Assessment of carbonaceous fractions in ambient aerosols at high altitude and lowland urban locations of Kumaon Province, Indian Himalaya

Amit Kumar[1,2] · Sachchidanand Singh[2] · Niraj Kumar[3] · Narendra Singh[4] · Krishan Kumar[1] · Sapna Chourasiya[5]

## Abstract

The present study investigates the characteristics of carbonaceous species like organic carbon (OC) and elemental carbon (EC) in ambient total suspended particulates (TSP) at Bhimtal (high altitude urban, 1413 m asl) and Pantnagar (lowland urban, 224 m asl) sites of Kumaon province in Uttarakhand, Indian Himalayan region during winter and summer 2017–2018. Ambient TSP samples were taken on quartz filters with high volume sampler followed by OC and EC quantification using IMPROVE_thermal optical reflectance protocol. Results showed that distinct seasonal differences in carbonaceous species levels were observed at both sites, while day- and night-time concentrations did not show notable variations. Further, total carbonaceous aerosols (TCA) at Pantnagar were approximately 3.0 and 1.3 times higher than Bhimtal for winter and summer, respectively, where estimated TCA accounted for ~ 30% to total TSP at both sites. Among quantified eight carbon fractions, OC2 and OC3 at Bhimtal while EC1 and EC2 at Pantnagar were the most abundant components. The char-EC and soot-EC concentrations showed a similar seasonal pattern where char contributed significantly as 89–90% to total EC at both sites. The observed OC/EC ratios suggested the formation of secondary organic carbon and char-EC/soot-EC ratios implied biomass burning as major sources for carbonaceous aerosols. Pearson correlation analysis indicated that char-EC showed significant higher correlations with OC and EC than soot-EC which infers different formation mechanisms of char and soot. Most of the carbonaceous parameters exhibited contrasting positive and negative correlations with the boundary layer height, temperature, and solar radiation at Bhimtal and Pantnagar, respectively.

Keywords  Carbonaceous aerosols · Himalaya · Carbon fractions · Char/soot · Meteorological parameters

## 1 Introduction

In the last few decades, numerous studies have been reported concerning carbonaceous species of ambient aerosols and their impact on local/global climate, atmospheric chemistry, air quality, biodiversity and human health [1–6]. Carbonaceous aerosols are ubiquitous in nature mainly consisted of organic carbon (OC) and elemental carbon (EC) which have significant portions in atmospheric aerosols [7, 8]. OC comprises a diversity of organic compounds, and their atmospheric sources could be both primary and secondary in origin. Primary OC may release from fossil fuel combustion/biomass burning as fine particulates and mechanical processes of the

ecosystem like plant spores and pollens, soil organic matter and decomposed vegetative debris as coarse particulates [9]. Atmospheric chemical reactions between volatile organics and gas-phase precursors through low vapour pressure, optimum temperature and solar radiation are known for secondary OC emissions [3, 10]. However, EC (similar to graphitic carbon) is exclusively primary in origin and occurred as residues mainly derived from incomplete combustions of biomass and fossil fuels [4, 11]. These carbonaceous species influence visibility, atmospheric radiative forcing, cloud physics and climatic systems [12, 13]. It is reported that OC has both warming and cooling effects on the atmosphere, while EC has warming characteristics and acts as the second most contributing agent after carbon dioxide in enhanced global warming. OC and EC both consisted of diverse ranges of carcinogenic and mutagenic compounds which cause morbidity and mortality of cardiovascular and respiratory disorders [14, 15].

In recent times, carbonaceous fractions are determined in the aerosol as bulk content of OC and EC along with eight carbon sub-fractions like OC1 to OC4, OP (organic pyrolysed carbon) and EC1 to EC3. The quantification of individual substance in OC fraction at the molecular level is still very challenging due to the complicated molecular structures of carbonaceous aerosols [16]. The variability of spatial/temporal distribution of carbon fractions significantly influences the physico-chemical properties of carbonaceous aerosols [17, 18]. EC is further classified into two groups, namely char-EC (formed at relatively low combustion temperatures and limited access of air) and soot-EC (formed at higher temperatures via gas phases processes by incomplete combustion) [19]. Nowadays, these eight fractions, char-EC and soot-EC are being used as indicators for source profiling of the carbonaceous aerosols [20–22]. Char-EC and soot-EC possess differences in physical, chemical and optical characteristics; therefore, they influence climate and environment in different ways [8]. Char-EC has the property of strong absorption in the UV region, whereas soot strongly absorbs solar light with little spectral dependence [23].

Significant roles of carbonaceous aerosols on the regional and global atmosphere have received increasing and special attention among the scientific community [9, 24–26]. The unprecedented urbanization along with rapid commercial and industrial development leads to deterioration of air quality in urban areas worldwide. Several research and reports have been documented which pertains to the determination of carbonaceous aerosols in the urban set-up where commercial, industrial activities and huge transportation networks are occurred [13, 27]. However, limited measurements concerning carbonaceous aerosols have been conducted over urban city/town/remote location in high altitude regions across the

world. Very few studies dedicated to carbonaceous aerosols have been reported from the Indian Himalayan region. The contribution of black carbon to total aerosol optical depth and direct radiative forcing was estimated in detail by Srivastava et al. (2011) over Manora Peak in the Indian Himalayan foothills [28]. Kumar and Attri [29] investigated the role of biomass combustion in carbonaceous aerosols in the western Himalayas and Rajput et al. [30] reported an abundance of carbonaceous aerosols in the foothills of northeast Himalaya [29, 30]. Recently, Sandeep et al. [31] measured OC and EC concentrations for one year over Srinagar, India, an ecologically sensitive semi-urban site in Garhwal Himalayas, and suggested that boundary layer dynamics and anthropogenic activities are responsible for seasonal variations [31]. However, no field campaigns were previously performed regarding comparative study of carbonaceous aerosols in lowland and high altitude regions of Indian Himalayas.

It is thus imperative to examine the sources and load of carbonaceous aerosols in the urbanized location of the mountainous region during different seasons and compare it with the lowland (Tarai) region. Therefore, we investigated the levels of carbonaceous species in total suspended particulates (TSP) at high altitude urban site (Bhimtal) and Tarai area (Pantnagar) in the Kumaon province of the Indian Himalayan region. The primary objectives of the present study include (a) determination of seasonal and day–night characteristics of carbonaceous components in TSP; (b) quantification of eight carbon fractions along with char and soot; (c) source attribution using char and soot ratios; (d) contributions of carbonaceous components to TSP and its comparison with earlier studies and (e) finally correlation analysis among carbonaceous components and meteorological parameters.

## 2 Materials and methods

### 2.1 Study area

Total suspended particulates (TSP) sampling campaign was conducted at two locations at Bhimtal (high altitude urban) and Pantnagar (lowland urban) in the Kumaon province of Uttarakhand, western Himalaya. The geographical locations of the sampling sites are shown in Fig. 1. Bhimtal is the hill station/small town of Kumaon province in the lower Himalayas which is geographically situated at 29.35°N and 79.53°E with a population of 7722 [32]. It is located approximately 22 km away from the Nainital city at an altitude of 1413 m asl and has recently witnessed dramatic urbanization and economic growth in the last two decades. It is bound by a dense forest of oak, pine, and shrubs where the climate is characterized
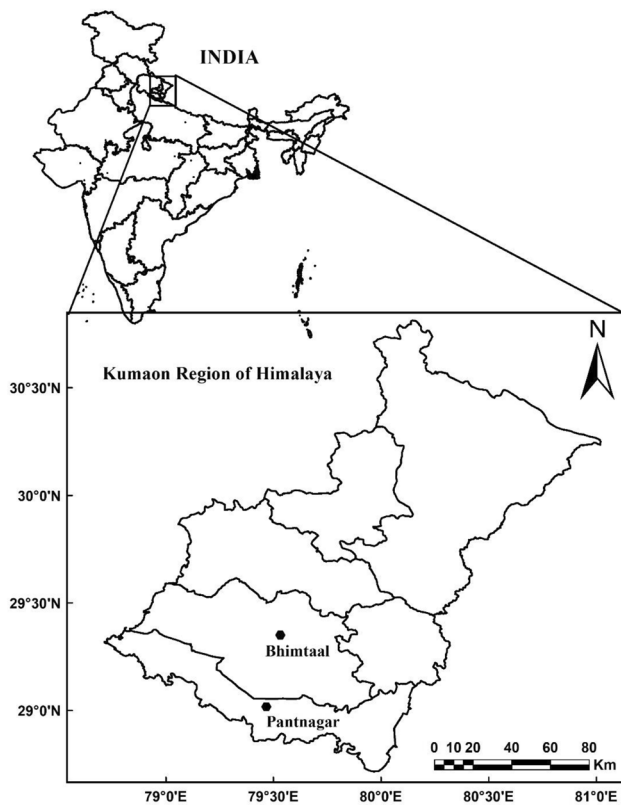
**Fig. 1** Map showing the geographical locations of sampling sites at Bhimtal and Pantnagar in Kumaon region, Western Himalaya

by pleasant summers (15–29 °C) and winters (4–18 °C). Due to the belief of holy land in mountainous regions, pilgrimage activities and aesthetic natural beauty, thousands of tourists visit here and go to higher altitudes via this region every year. This has increased anthropogenic activities and energy consumption through fossil fuel burning led to the deterioration of air quality. There were no major industrial activities near or surroundings of the sampling location. The elevation, location, energy consumption, religious activities and prevailing meteorological condition make Bhimtal a unique urban site as compared to other urban regions.

Another sampling site was selected at Pantnagar in Udham Singh Nagar district of Uttarakhand, India. It is geographically located at 29.01°N and 79.47°E at an elevation of 224 m asl in the "Tarai" region (lowland urban area) in Himalayan foothills. Pantnagar town with a population of 35,820, is a mixture of the university campus (G. B. Pant University of Agriculture and Technology) and industrial estate (431 industries spread over 1310 ha of land consisted of food production, electroplating, automobile industries, etc.) [33]. The climate of Pantnagar is characterized by humid sub-tropical conditions and accounted for varying climatic extremes throughout the year. In general,

summer and winter temperatures in this region are in the range of 26–42 °C and 8–15 °C, respectively, with an annual rainfall of 1300–1400 mm. Pantnagar has a flat topography and is also known as a centre of attractions for tourists near its vicinity like Jim Corbett National Park, Nainital, and Mukteshwar.

## 2.2 Sampling

High volume air sampler (Envirotech APM 450, India) was used to collect ambient TSP at both sampling sites with a flow rate of $1.1 \pm 0.1$ m$^3$/minute. TSP samples were collected for five consecutive days during the winter and summer seasons at both sites for daytime (07:00 to 19:00 h) and night-time (19:00 to 07:00 h) periods. At both study sites, sampling was performed during November (2017) and May (2018) for winter and summer seasons, respectively. Twelve hours TSP samples were collected on quartz filters (PALLFLEX™; 2500 QAT-UP, size: 20.0 cm, 25.4 cm). Before sampling, quartz filters were pre-combusted at a temperature of 900 °C in a muffle furnace to remove any residual carbon or impurities [34]. After sampling, all exposed filter samples were kept in zip-lock plastic bags and stored in a refrigerator until OC and EC analysis to avoid evaporation of deposited VOCs. TSP mass loadings (µg) were deduced from gravimetric measurements done before and after sampling using a weighing machine (AD—50 B, ADAIR DUTT Instruments Pvt. Ltd), and corresponding concentrations (µg/m$^3$) were evaluated by dividing mass loadings to sampled air volume. Flow rate and duration of the sampling period were used to estimate the volume of air sampled. Quartz filters were equilibrated in a desiccator for 24 h at constant temperature (23–25 °C) and relative humidity (40–45%) for 4–5 h [35]. Field blank filters were also taken into account by exposing filter paper in the sampler without running the pump for same sampling duration. In order to remove positive sampling artefact, the estimated sample results were corrected by the subtraction of mean values of observed blank concentration [34, 36]. The precision of the weighing process was < 10 µg for the blank filters and < 20 µg for the filter samples. All necessary precautions were strictly considered during the handling of quartz filters to avoid any possibility of contamination.

## 2.3 Chemical analysis

TSP quartz filters were analysed for OC and EC quantifications using the DRI model 2001 Thermal/Optical Carbon analyser (Atmoslytic Inc., Calabasas, CA). A small punch aliquot of 0.5 cm$^2$ from each filter was positioned at a quartz tube inside the thermal desorption chamber of the analyser. The analytical procedure consisted of two

stages of stepwise heating (thermal differentiation) following IMPROVE-A (Interagency Monitoring of Protected Visual Environments) protocol produce four organic carbon fractions and three elemental carbon fractions [18, 37]. OC fractions as OC1, OC2, OC3 and OC4 were volatilized at respective temperatures of 140 °C, 280 °C, 480 °C and 580 °C in a non-oxidizing helium atmosphere. Further, a pyrolysed carbon fraction (OP) was determined when reflected laser light attained its original intensity after oxygen was added to the combustion atmosphere. Moreover, EC fractions as EC1, EC2 and EC3 were produced at 580 °C, 740 °C and 840 °C, respectively, in the presence of 2% oxygen/98% helium. After analysis, a datasheet was generated which reports data of eight carbon fractions separately. According to IMPROVE-A protocol, OC is represented as OC1 + OC2 + OC3 + OC4 + OP, while EC as EC1 + EC2 + EC3 − OP [37]. Further, char-EC is determined by EC1 minus OP, while soot-EC designates the sum of EC2 and EC3 [21]. The OC/EC analyser is auto-calibrated every day using known concentrations of $CH_4$ before and after sample analysis. Replicate analysis was also carried out for every ten samples run where the difference estimated was smaller than 5% for the total carbon (TC, OC + EC) and 10% each for OC and EC. Detection limits for OC and EC were calculated as 0.25 μg/m$^3$ and 0.04 μg/m$^3$, respectively. OC and EC concentrations were also estimated in blank filters and average blank concentrations have been used to get a correction in the sample results. It is important to note that fraction of carbonate carbon (CC) in the aerosol samples have not been chemically removed for the present study.

## 3 Results and discussion

### 3.1 Seasonal and day-/night-time variations of OC, EC, char-EC and soot-EC

Twelve hours seasonally averaged and day–night-time atmospheric concentrations of TSP along with different carbonaceous components during the observation period for two studied sites are documented in Table 1. Firstly, it can be seen that the levels of TSP and OC/EC concentrations at Pantnagar (urban, lowland Tarai) site were higher than Bhimtal (urban, high altitude) site. Higher concentration values of TSP and carbonaceous species at Pantnagar could be due to industrial processes and seasonal agricultural practices. Banerjee et al. [38] reported higher pollution at the same site because of the development of industrial estate [38]. The seasonal analysis suggested that TSP and OC/EC concentrations were higher during summer in contrast to winter at Bhimtal while contrasting results at Pantnagar with higher and lower concentrations of these parameters during winter and summer, respectively. At Bhimtal, mean concentrations of TSP/(OC)/(EC) were found to be 89.7/(12.9)/(3.6) μg/m$^3$ and 109/(18.7)/(5.4) μg/m$^3$ for winter and summer, respectively. On the other hand, respective TSP/(OC)/(EC) concentrations at Pantnagar were observed as 224/(35.3)/(15.6) μg/m$^3$ and 180/(23.1)/(8.8) μg/m$^3$ for winter and summer.

The day-to-night-time (D/N) ratios at Bhimtal for winter and summer were 1.5 and 1.2 for TSP, 1.2 and 1.1 for OC and 1.3 and 1.0 for EC, respectively. Higher daytime concentrations of OC and EC at Bhimtal could be attributed to more tourist activities during the daytime. However, irrespective of high night-time TSP concentrations at Pantnagar, the D/N ratios for winter and summer were 1.2 and 1.2 for OC and 1.2 and 1.1 for EC, respectively. Higher TSP concentrations during night-time than daytime at

**Table 1** Statistical summary of seasonal and day-/night-time concentrations of different TSP components (μg/m$^3$) at Bhimtal and Pantnagar

| | Bhimtal | | | | Pantnagar | | | |
|---|---|---|---|---|---|---|---|---|
| | Winter | | Summer | | Winter | | Summer | |
| | Avg ± SD | D/N | Avg ± SD | D/N | Avg ± SD | D/N | Avg ± SD | D/N |
| TSP | 89.7 ± 24.6 | 103/70.1 | 109 ± 18.1 | 120/98.6 | 224 ± 49.8 | 200/247 | 180 ± 36.9 | 173/188 |
| OC | 12.9 ± 4.4 | 14/11.8 | 18.6 ± 3.4 | 19.5/17.7 | 35.3 ± 4.0 | 37.9/32.7 | 23.1 ± 4.5 | 25.2/21.1 |
| EC | 3.6 ± 1.0 | 4.1/3.1 | 5.4 ± 1.3 | 5.5/5.3 | 15.6 ± 2.2 | 17.1/14 | 8.8 ± 3.0 | 9.4/8.3 |
| Char-EC | 3.2 ± 1.0 | 3.8/2.7 | 4.9 ± 1.2 | 5.0/4.8 | 13.7 ± 2.3 | 15.2/12.2 | 7.9 ± 3.0 | 8.4/7.5 |
| Soot-EC | 0.4 ± 0.05 | 0.4/0.4 | 0.5 ± 0.1 | 0.5/0.4 | 1.8 ± 0.4 | 1.9/1.8 | 0.9 ± 0.2 | 0.9/0.8 |
| OM | 20.6 ± 7.0 | 22.3/18.9 | 29.8 ± 5.5 | 31.3/28.3 | 56.5 ± 6.4 | 60.7/52.4 | 37.0 ± 7.2 | 40.3/33.7 |
| EM | 4.0 ± 1.1 | 4.6/3.4 | 5.9 ± 1.4 | 6/5.8 | 17.1 ± 2.4 | 18.8/15.4 | 9.7 ± 3.3 | 10.3/9.1 |
| TCA | 24.2 ± 7.7 | 26.4/22 | 35.2 ± 6.6 | 36.7/33.6 | 72.1 ± 8.4 | 77.6/66.4 | 45.8 ± 9.7 | 49.7/42 |

*SD* Standard deviation, *D* day, *N* night, *OM* organic matter, *EM* elemental matter, *TCA* total carbonaceous aerosols

the lowland urban region of Pantnagar suggested mixing height plays an important role in the diurnal variation. TSP was observed to be low during daytime when mixing height was higher (ranged from 417 to 1081 m). In contrast, higher concentrations occurred during night-time when mixing height was lower in the range of 180–458 m. Higher night-time concentrations could be associated with trapping of TSP due to the lowering of mixing height [39]. Similar higher concentrations are generally noticed during the night-time (than daytime) in urban areas [27, 40]. The temporal variability of TSP along with carbonaceous species for two studied sites is shown in Fig. 2.

Along with OC and EC, other carbonaceous components like Total Carbonaceous Aerosols (TCA), Organic Matter (OM) and Elemental Matter (EM) in ambient TSP for two studied sites are also reported (Table 1). The quantity of OM was estimated by the multiplication of observed OC concentrations with a factor of 1.6 [41], while EM amounts have been estimated by multiplying the EC concentrations with 1.1 [42]. Further, TCA in the atmosphere is evaluated by the summation of OM and EC (TCA = OC × 1.6 + EC) [43]. The conversion factors for OC to OM have a wide range of values (1.2–2.1) reported in previous literature [44]. The lower values of the conversion factors (1.2–1.4) are used for the organic aerosol having lower SOA fraction; however, higher values were considered for aerosols with

higher SOA fractions. Similar to OC/EC concentrations, Pantnagar exhibited higher mean values of OM, EM and TCA as compared to the values of Bhimtal. The concentration ratios of Pantnagar to Bhimtal showed for winter and summer were 2.7 and 1.2 for OM, 4.3 and 1.6 for EM and 3.0 and 1.3 for TCA, respectively. At Bhimtal, the mean values of OM, EM and TCA were noted to be higher as 1.7, 1.5 and 1.5 times during summer in contrast to winter. However, Pantnagar exhibited contrasting respective results as 1.5, 1.8 and 1.6 times higher during winter as compared to summer.

The observed variations of TSP and carbonaceous species at two selected sites could be explained by the combined phenomenon of emission sources (biomass burning, coal combustion and vehicular exhaust), their emission rates and prevailing meteorological conditions. In the context of high altitude Bhimtal site, seasonal and day/night variations of TSP and OC/EC concentrations can be understood by the phenomenon of vertical movement of planetary boundary layer and distant transport [45]. Summer time exhibited higher values of pollutants because of an increase in convective mixing and extent of convective boundary layer due to more surface heating, which resulted in a conducive environment for the upliftment of aerosols from the valley region [46]. Besides this, higher summer concentrations could also be due to an increase in

**Fig. 2** Temporal variations of TSP along with OC, EC and TCA concentrations (μg/m³) in Bhimtal and Pantnagar
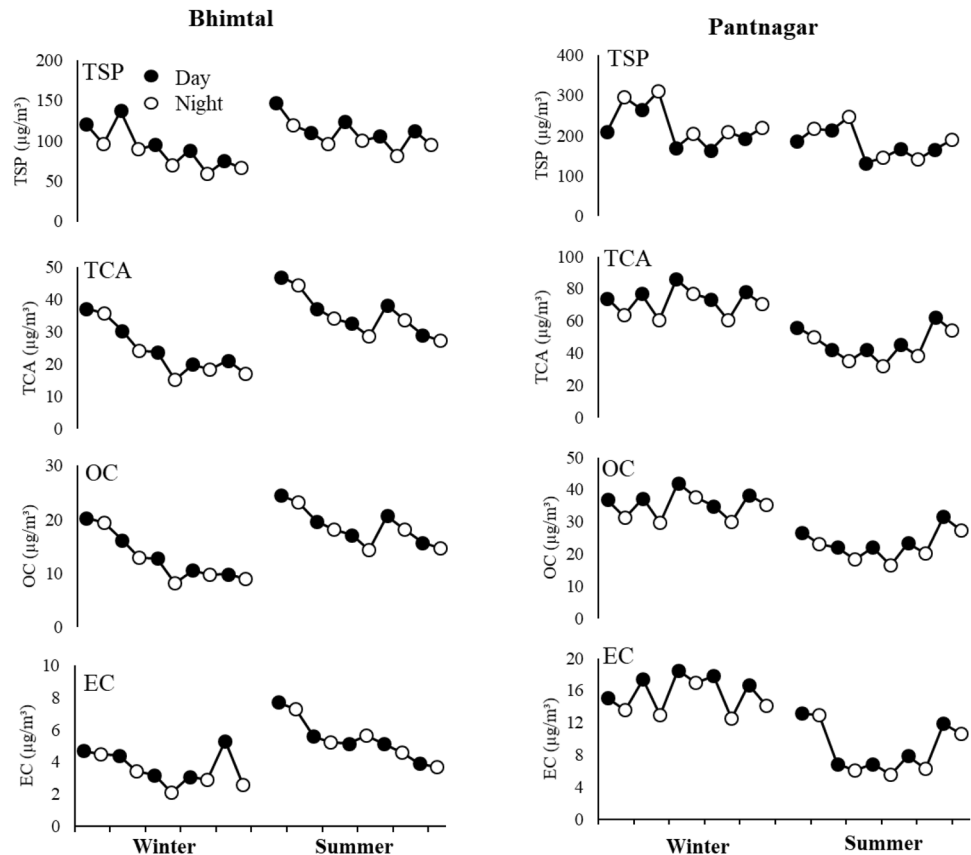
vehicular emissions due to tourism and pilgrimage activities. In the case of Pantnagar, TSP and OC/EC exhibited higher and lower concentrations during winter and summer, respectively, similar to the observed results of plain urban areas [14, 27]. Besides enhanced fuel consumption, formation of the inversion layer and stable conditions during the winter period resulted into the trapping of carbonaceous aerosols could also be the main reasons for higher concentrations during winter [23]. Further, vertical mixing and increased planetary boundary layer could be associated with lower carbonaceous pollutants during summer.

The backward air trajectory analysis over Bhimtal and Pantnagar could also explain the wind patterns and long-range transport of pollutants. 120 h air mass back-trajectory plots at three different heights (500 m, 1000 m and 1500 m above ground level) have been plotted using the HYSPLIT model, developed by NOAA (http:// ready. arl.noaa.gov/HYSPLIT.php). The representative air mass back-trajectories for each season over Bhimtal and Pantnagar are shown in Fig. 3a, b. It can be seen that winters are dominated by south and south-westerly air masses, which passes through the central part of India prior to entering in high altitude Bhimtal region. In contrast, transported air masses during summer largely came from the northeast region pass over the Indo-Gangetic plain before reaching Bhimtal. Higher summer-time concentrations of carbonaceous aerosols at Bhimtal are attributed to coming winds from the Indo-Gangetic plain (highly polluted region). At Pantnagar, air masses were observed to be mostly arriving from north/north-west directions originated from Pakistan and Afghanistan during winter and pass through agricultural states of Himachal Pradesh, Punjab and Haryana before entering to the receptor site. Agricultural residues burning practices in the neighbouring states could be associated with higher carbonaceous aerosols during the winter season. Pantnagar experienced shorter trajectories
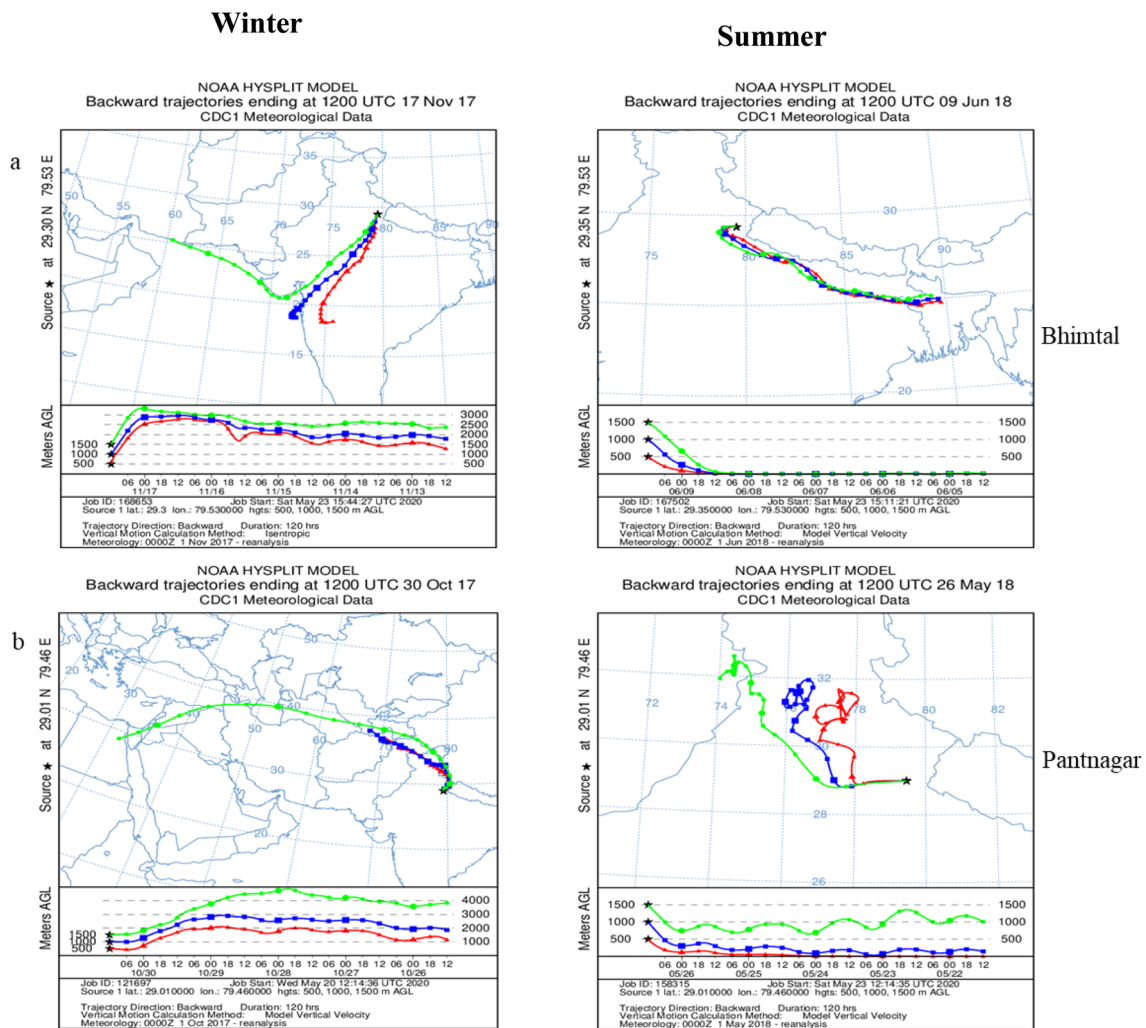


**Fig. 3** Representative air mass back-trajectories over sampling sites **a** Bhimtal and **b** Pantnagar during winter and summer seasons

during summer infers dominance of local pollution along with distant transport.

## 3.2 Characterization of eight carbon fractions at studied sites

The present section deals with the occurrence of different carbon fractions at two studied sites. The analytical procedure of IMPROVE TOR protocol consisted of different evaporation or oxidation temperatures for carbon fractions (OC1–OC4, OP and EC1–EC3) which have unique characteristics and emission sources [37]. Source profiling of carbonaceous aerosols is done based on the abundance of carbon in each of these eight carbon fractions [1, 19]. Based on the previous literature, OC1 and EC1 infer biomass burning, OC2 and OC3 might be emitted from SOC and biomass burning sources, OC4, EC2 and EC3 designate coal combustions and vehicular emissions and OP could be an indication of increased water-soluble organic carbon [19]. According to Zhang et al. (2012), higher contents of OC3 and OC4 reflect road dust as pollution sources [4].

The percentage seasonal contribution of OC and EC fractions in TSP for two studied sites is shown in Fig. 4. These carbon fractions at Bhimtal accounted in the order of OC2 (28.9%) > OC3 (21.1%) > EC1 (20.2%) > OC4 (11.8%) > OP (9.0%) > OC1 (6.9%) > EC2 (1.7%) > EC3 (0.6%). On the other hand, the average contributions of these fractions at Pantnagar were ranked in the following order: EC1 (25.5%) > OC2 (24.7%) > OC3 (21.5%) > OP (11.6%) > OC4 (8.1%) > OC1 (5.7%) > EC2 (2.4%) > EC3 (0.8%). OC2 and OC3 were the main carbon fractions in Bhimtal, while EC1 and OC2 were the prevalent carbonaceous species in Pantnagar. Therefore, fractions contribution at the two sites suggested the occurrence of major sources like vegetative burning, coal combustion and vehicular emissions.

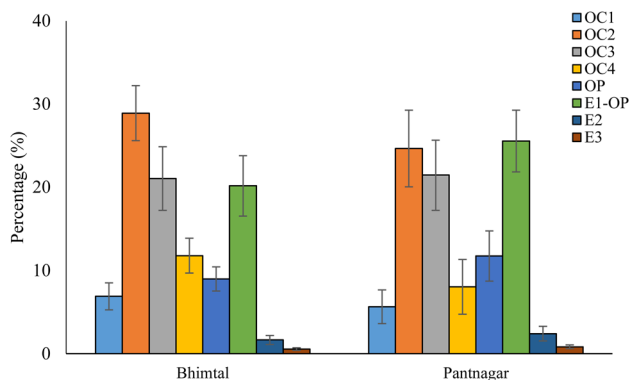In the present study, observed contributions of four organic carbon fractions (OC1, OC2, OC3 and OC4) to TC



**Fig. 4** Percentage contributions of eight carbon fractions to total carbon at studied sites

were noted to be higher than urban area of Costa Rica, Central America which ranged from 0.9% to 19.8% in $PM_{10}$ and 1.5% to 16.6% in $PM_{2.5}$, respectively [10]. Similarly, the average abundance of OC1 to OC4 was also observed to be lower (4.3–19.9%) in the Pearl River Delta Region, China [1]. The study carried out in urban areas of Hanoi, Vietnam has suggested OC3 and EC1 were predominant in $PM_{10}$ and $PM_{2.5}$, while OC3 and OC2 were the prevailing carbonaceous fractions of $PM_{0.1}$ [11]. The EC2 and EC3 contributions of the present study were comparable with the contributions reported in Tianjin, China, and lower in the Pearl River Delta Region, China [1, 3].

## 3.3 Char- and soot-EC variations and source identification using char-EC/soot-EC ratios

Based on the analytical methods, EC is also categorized into two groups commonly known as char-EC and soot-EC which have different physico-chemical and optical properties. In general, char-EC is mainly derived as residues from incomplete combustion or heating of carbonaceous matter in limited access of air, while soot-EC is formed from gas–particle re-condensation at higher temperature burning process [19, 21]. The larger particle size of char-EC makes higher rate of deposition which indicated local combustion emissions and the smaller size of soot-EC favours long-range transport resulted into a longer lifetime [47].

The average concentrations of char-EC and soot-EC at Bhimtal were $3.23 \pm 1.02$ μg/m³ and $0.38 \pm 0.05$ μg/m³ (winter) and $4.92 \pm 1.97$ μg/m³ and $0.47 \pm 0.11$ μg/m³ (summer), respectively. However, respective concentrations of analogue parameters at Pantnagar were $13.7 \pm 2.3$ μg/m³ and $1.82 \pm 0.45$ μg/m³ (winter) and $7.9 \pm 3.0$ μg/m³ and $0.88 \pm 0.18$ μg/m³ (summer). Overall, char-EC and soot-EC at Pantnagar were approximately 2.7 and 9.6 times higher than the Bhimtal. The observed lower soot-EC levels at both sites indicated less influence of vehicular and industrial emissions. Seasonal variations of char-EC and soot-EC may be attributed to differences in fuel combustions and meteorological factors (mixing height, temperature, etc.) [21]. Higher mixing height during summer favours the dispersion phenomenon resulting in the lowering of pollutant concentrations.

In the previous, a quantitative assessment of OC/EC mass ratios was used to identify the carbonaceous sources [1, 48]. OC/EC ratio method is not widely accepted now due to some of the limitations as it is influenced by photochemical reactivity and secondary organic aerosol (SOA) formation. In contrast, char-EC/soot-EC is considered as an effective tool than OC/EC ratios to infer primary carbonaceous source because they avoid the great bias of dependency on SOA [23]. Cao et al. [20] reported char-EC/

soot-EC ratios of 11.6 for biomass burning and 1.9 for coal combustion [20]. In the present study, the mean values of OC/EC showed almost similar values during winter/(summer) as 3.6/(3.5) and 2.3/(2.8) at Bhimtal and Pantnagar, respectively, which infers the SOC formation. The estimates of char-EC/soot-EC showed distinct seasonal variations as 14.6/(10.6) and 12.0/(9.1) during winter/(summer) at Bhimtal and Pantnagar, respectively. For source characterization, the observed ratios of the present study suggested carbonaceous aerosols are mainly derived from biomass combustion where higher winter period values could be due to enhanced biomass burning activities. Strong biomass burning activities have been observed during winter as compared to summer in north and northwest regions of India which is nearby our sampling sites (Fig. S1). While comparing with char-EC/soot-EC ratios of vehicular exhaust which is suggested to be lower than 1, summer-time ratios values showed still higher values. It could be due to the dominance of biomass burning and residential emissions even in the summer period. These observed results were consistent with the previous studies across the world [19, 24, 29]. Further, char-EC/soot-EC in the current study is much lower than the ratios observed in Nanjing [14].

## 3.4 Contributions of carbonaceous components to TSP and comparison of observed data set with earlier studies

The relative proportions of different carbonaceous components to total TSP are shown in Table 2. The proportion of carbon to TSP mass can be useful to understand the magnitude of carbonaceous aerosol pollution in observed particulate estimations. The occurrence of a large proportion of carbon (especially EC) in the particulates may lead to the enhancement of solar absorptions followed by an increase in atmospheric warming. Further, it could largely affect Himalayan provinces like our studied locations Bhimtal and Pantnagar are close to glaciers [31, 49]. As shown in Table 2, percentage contributions of OC, EC and TCA to TSP at Bhimtal were 14%, 4% and 27% for winter

and 17%, 5% and 32% for summer, respectively, while at Pantnagar analogue parameters accounted for 17%, 7% and 34% for winter and 13%, 5% and 26% for summer, respectively. Overall, OC accounted for almost 78% and 71% of the total carbon particulates for Bhimtal and Pantnagar, respectively, while EC represented the rest respective 22% and 29%. At the Bhimtal, char-EC accounted for approximately 19.6% and 20.5% to total carbon during winter and summer, respectively, while 27.0% and 24.7% contributions at Pantnagar. It is suggested that the proportions of carbonaceous species to atmospheric TSP at studied sites were different where higher contributions at Bhimtal and Pantnagar were during the summer and winter periods, respectively. Besides local combustion sources, higher concentrations of carbonaceous species in aerosols at two sites could be associated with crop burning practices during pre-monsoon (May/June) and winter (November) in nearby regions of Haryana, Punjab and Uttar Pradesh.

An investigation of one-way analysis of variance (ANOVA) test was implemented to determine the seasonal and day-/night-time differences for OC, EC, char, soot, OC/EC and char/soot using SPSS statistical software (Table 3). It can be seen that TSP along with most of the carbonaceous parameters showed marked seasonal differences ($p < 0.05$) at both sites which could be associated with seasonal variations of sources, their emission rates and meteorological conditions. Conversely, insignificant differences ($p > 0.05$) of day and night-time concentrations for most of the carbonaceous parameters were noted at both sites.

Table 4 documents the comparative account of OC, EC, char-EC and soot-EC concentrations observed in TSP from different regions across the world. It can be observed that OC and EC levels at Bhimtal were comparable with high altitude urban site of other regions as respective values of 13.5 μg/m³ and 5.1 μg/m³ at Kangra, India [29] and 15.3 μg/m³ and 5.2 μg/m³ at Srinagar, India [31]. However, OC and EC concentrations were observed to be much higher than Langtang, Nepal as 2.0 μg/m³ and 0.38 μg/m³ [50] and Tennessee, USA, as 3.3 μg/m³ and 0.55 μg/m³, respectively [51]. Pantnagar, a lowland urban region, exhibited higher carbonaceous aerosols than Bhimtal where OC, EC values were comparable with results of urban regions as 37.7, 7.8 μg/m³ in Delhi, India [27] and 38.6, 8.4 μg/m³ in Xian, China [24] while experienced much higher values than Saitama, Japan as 5.5 μg/m³, 2.9 μg/m³ [23], Shanghai, China, as 10.1 μg/m³, 2.5 μg/m³ [52] and Huangshi, China, as 22 μg/m³, 6.2 μg/m³ [8]. The observed char-EC and soot-EC of the present study were noted to be higher than those observed in Dharamshala, India [53], Jiangxi, China [54], Saitama, Japan [23] and Shanghai, China [52].

**Table 2** Statistical summary of seasonal percentage contributions of carbonaceous components to TSP at studied sites

|  | %OC/TSP | %EC/TSP | %TCA/TSP | %OM/TSP | %EM/TSP |
|---|---|---|---|---|---|
| *Bhimtal* | | | | | |
| Winter | 14.3 ± 2.8 | 4.1 ± 1.2 | 27.0 ± 4.9 | 22.9 ± 4.4 | 4.6 ± 1.3 |
| Summer | 17.2 ± 2.9 | 4.9 ± 0.9 | 32.4 ± 5.2 | 27.5 ± 4.6 | 5.4 ± 0.9 |
| *Pantnagar* | | | | | |
| Winter | 16.7 ± 4.7 | 7.4 ± 2.3 | 34 ± 9.8 | 26.7 ± 7.6 | 8.1 ± 2.6 |
| Summer | 13.3 ± 3.4 | 5.0 ± 1.6 | 26.2 ± 6.6 | 21.2 ± 5.4 | 5.5 ± 1.7 |

**Table 3** One-way ANOVA of seasonal and day-/night-time variability in TSP and carbonaceous components at studied sites

| Sampling sites | Comparison criteria | TSP | OC | EC | Char-EC | Soot-EC | OC/EC | Char/soot |
|---|---|---|---|---|---|---|---|---|
| Bhimtal | Between seasons | 4.1[a](0.06[b]) | 10.6 (0.00) | 11.5 (0.00) | 11.4 (0.00) | 4.9 (0.03) | 0.08 (0.77) | 8.9 (0.00) |
| | Between day and night | 7.2 (0.01) | 0.9 (0.37) | 0.9 (0.35) | 0.9 (0.34) | 0.2 (0.63) | 0.03 (0.86) | 5.8 (0.02) |
| Pantnagar | Between seasons | 4.9 (0.04) | 41.1 (0.00) | 32.8 (0.00) | 23.8 (0.00) | 37.4 (0.00) | 8.3 (0.00) | 4.6 (0.04) |
| | Between day and night | 2.2 (0.15) | 2.0 (0.17) | 1.2 (0.29) | 1.8 (0.3) | 0.2 (0.68) | 0.002 (0.96) | 0.001 (0.96) |
| Averaged of two sites | Between sites | 21.6 (0.00) | 17.7 (0.00) | 16.2 (0.00) | 51.4 (0.00) | 106 (0.00) | 39.7 (0.00) | 3.7 (0.06) |

[a]$F$ value;

[b]$p$ value

**Table 4** Comparison of OC, EC, char-EC, soot-EC, OC/EC and char/soot measurements ($\mu g/m^3$) in present study with cities across the world

| Reference | Location | Size fractions | OC | EC | Char-EC | Soot-EC | OC/EC | Char/soot |
|---|---|---|---|---|---|---|---|---|
| Present study | Bhimtal (high altitude urban) | TSP | 15.6 | 4.5 | 4.1 | 0.4 | 3.5 | 10.3 |
| | Pantnagar (lowland semi-urban) | TSP | 29.2 | 12.2 | 10.8 | 1.4 | 2.4 | 7.7 |
| Kaushal et al. [53] | Dharamshala, India (high altitude) | $PM_{10}$ | 5.0 | 2.5 | 2.1 | 0.4 | 2 | 5.3 |
| Sandeep et al. [31] | Srinagar, India (high altitude) | $PM_{2.5}$ | 15.3 | 5.2 | – | – | 2.9 | – |
| Kumar and Attri [29] | Kangra, India (high altitude) | $PM_{10}$ | 13.5 | 5.1 | 5.0 | 0.19 | 2.6 | 26.3 |
| Carrico et al. [50] | Langtang, Nepal (high altitude) | $PM_{2.5}$ | 2.0 | 0.38 | – | – | 5.3 | – |
| Li et al. [54] | Jiangxi, China (high altitude) | $PM_{2.5}$ | 3.8 | 1.3 | 0.91 | 0.36 | 2.9 | 2.5 |
| Tanner et al. [51] | Tennessee valley, USA | $PM_{2.5}$ | 3.3 | 0.55 | – | – | 6 | – |
| Bisht et al. [27] | Delhi, India (urban) | $PM_{2.5}$ | 37.7 | 7.8 | – | – | – | – |
| Han et al. [24] | Xian, China (urban) | $PM_{2.5}$ | 38.6 | 8.4 | 6.9 | 1.6 | 4.6 | 4.3 |
| Kim et al. [23] | Saimata, Japan (urban roadside) | $PM_{2.5}$ | 5.5 | 2.9 | 2.7 | 0.3 | 1.9 | 9 |
| Cao et al. [52] | Shanghai, China (urban) | TSP | 10.1 | 2.5 | 1.7 | 0.7 | 4.0 | 2.4 |
| Zhan et al. [8] | Huangshi, China (urban industrial) | $PM_{10}$ | 22 | 6.2 | 5.3 | 0.9 | 3.5 | 5.9 |
| Zhu et al. [22] | Xuenshan, Taiwan (urban tunnel) | $PM_{2.5}$ | 23.9 | 18.9 | 18.1 | 0.8 | 1.3 | 22.6 |

## 3.5 Correlations among carbonaceous components and meteorological parameters

Correlation analysis was performed among TSP mass, carbonaceous components and meteorological parameters for two studied sites during the observation period and is presented in Table 5. A significant positive correlation between OC and EC was observed at Bhimtal ($r = 0.82$) and Pantnagar ($r = 0.94$) suggested the combination of common combustion sources for carbonaceous species. Higher correlations were similar to those found in reported results of previous studies [21, 41]. As compared to soot-EC, char-EC was highly correlated with TC, OC and EC at both sampling sites which indicated different formation mechanisms for char and soot. It is also concluded that emissions sources of soot-EC could be different from OC and char-EC. Among the carbonaceous components, char-EC showed the strongest correlation with EC at both sites which were consistent with dominated char-EC of 90% and 88.5% contributions to total EC at Bhimtal and Pantnagar, respectively. It is suggested that the occurrence of common burning practices like biomass burning and residential heating. The moderate correlation between soot-EC and EC implied that variations in EC concentrations were also considerably influenced by soot-EC. The correlation between char-EC and soot-EC at both sites were found to be weaker than those between OC and EC. The occurrence of no correlations between OC/EC and char-EC/soot-EC at Bhimtal suggested source variations for carbonaceous aerosols emissions, while Pantnagar exhibited anti-correlation between them.

The association between EC and char-EC, EC and soot-EC and soot-EC and char-EC using regression plots at both sites is illustrated in Fig. 5. It can be seen that char-EC showed the strongest correlation with EC at both sites which were consistent with observation results conducted in previous studies [23, 24]. However, soot-EC exhibited moderate correlations with EC at Bhimtal ($r^2 = 0.65$) and weak correlations at Pantnagar ($r^2 = 0.36$). Char-EC vs soot-EC showed weaker correlations as $r^2 = 0.60$ (Bhimtal) and $r^2 = 0.32$ (Pantnagar). Poor correlations at Pantnagar suggested mixed sources of biomass burning, coal combustion and vehicular emissions.

**Table 5** Pearson correlation analysis among observed carbonaceous components in TSP and meteorological parameters

|  | TC | OC | EC | OC/EC | Char-EC | Soot-EC | Char/soot | PBL | SR | Temp | WS | RH |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| *Bhimtal* | | | | | | | | | | | | |
| TC | 1.00 | | | | | | | | | | | |
| OC | 0.99** | 1.00 | | | | | | | | | | |
| EC | 0.89** | 0.82** | 1.00 | | | | | | | | | |
| OC/EC | 0.10 | 0.23 | − 0.36 | 1.00 | | | | | | | | |
| Char-EC | 0.89** | 0.82** | 0.98** | − 0.36 | 1.00 | | | | | | | |
| Soot-EC | 0.68** | 0.62** | 0.80** | − 0.36 | 0.77** | 1.00 | | | | | | |
| Char/soot | − 0.02 | − 0.01 | − 0.04 | 0.05 | 0.01 | − 0.60 | 1.00 | | | | | |
| PBL | 0.41 | 0.41 | 0.37 | 0.01 | 0.35 | 0.35 | − 0.10 | 1.00 | | | | |
| SR | 0.52* | 0.51* | 0.48* | − 0.01 | 0.45* | 0.55* | − 0.29 | 0.89 | 1.00 | | | |
| Temp | 0.61** | 0.60** | 0.57** | 0.01 | 0.54* | 0.70** | − 0.43 | 0.81** | 0.92** | 1.00 | | |
| WS | − 0.22 | − 0.20 | − 0.24 | 0.05 | − 0.26 | − 0.07 | − 0.21 | 0.55* | 0.32 | 0.28 | 1.00 | |
| RH | 0.06 | 0.03 | 0.14 | − 0.19 | 0.13 | 0.45* | − 0.62** | − 0.54* | − 0.26 | − 0.10 | − 0.38 | 1.00 |
| *Pantnagar* | | | | | | | | | | | | |
| TC | 1.00 | | | | | | | | | | | |
| OC | 0.99** | 1.00 | | | | | | | | | | |
| EC | 0.97** | 0.94** | 1.00 | | | | | | | | | |
| OC/EC | − 0.72** | − 0.63** | − 0.85** | 1.00 | | | | | | | | |
| Char-EC | 0.97** | 0.93** | 0.99** | − 0.85** | 1.00 | | | | | | | |
| Soot-EC | 0.60** | 0.59** | 0.60** | − 0.44 | 0.56** | 1.00 | | | | | | |
| Char/soot | 0.70** | 0.66** | 0.72** | − 0.70** | 0.76** | − 0.08 | 1.00 | | | | | |
| PBL | − 0.33 | − 0.30 | − 0.36 | 0.40 | − 0.36 | − 0.18 | − 0.25 | 1.00 | | | | |
| SR | − 0.57** | − 0.56* | − 0.57** | 0.47* | − 0.57** | − 0.37 | − 0.35 | 0.87 | 1.00 | | | |
| Temp | − 0.62** | − 0.61** | − 0.62** | 0.49* | − 0.61** | − 0.39 | − 0.39 | 0.85** | 0.98** | 1.00 | | |
| WS | − 0.53* | − 0.54* | − 0.49* | 0.28 | − 0.48* | − 0.49* | − 0.14 | 0.56** | 0.62** | 0.66** | 1.00 | |
| RH | 0.48* | 0.46* | 0.50* | -0.48* | 0.50* | 0.35 | 0.30 | − 0.79** | − 0.94** | − 0.91** | 0.59 | 1.00 |

*PBL* Planetary boundary layer, *SR* solar radiation, *Temp* temperature, *WS* wind speed, *RH* relative humidity

*$p < 0.05$, **$p < 0.01$ (correlation coefficient and significance test).

Besides the emission sources, the occurrence of carbonaceous components in any region is also dependant on prevailing meteorological conditions. Meteorological data set like planetary boundary layer (PBL), temperature (T), solar radiation (SR), wind speed (WS) and relative humidity (RH) during the observed period have been obtained from ERA–5 reanalysis data set, developed by the ECMWF (https://cds.climate.copernicus.eu/cdsapp#!/home). At Bhimtal, carbonaceous components showed weak to moderate positive correlations ($p < 0.01$) with meteorological variables T > SR > PBL. The positive correlations of carbonaceous components with PBL suggested higher concentrations during daytime with an increase in PBL. The atmospheric boundary layer stays far below the observation site during the night-time. The strong convective motions formed in the valley regions results into vertical transport of pollutants through upslope winds favours the higher concentrations during daytime. In contrast, night-time concentrations are observed to be lower could be due to deepening in PBL. Further, higher temperature and

solar radiation along with low wind speed during daytime favour the increased carbonaceous concentrations. Our results are consistent with the observed meteorological association at the high altitude site of Manora Peak [55]. In contrast, Pantnagar exhibited weak to moderate negative correlations ($p < 0.01$) for carbonaceous components with meteorological parameters T > SR > PBL. Pantnagar is a lowland region having flat topography in the vicinity of the Himalayan foothills which does not show phenomena like mountain-valley convective motion and vertical movement of atmospheric pollutants. The good negative correlations of carbonaceous components with PBL, T and SR suggested lower concentrations during the daytime could be attributed to increased dispersion and dilution phenomenon due to elevated PBL. Similar results of inverse relationships of carbonaceous components at Pantnagar are in agreement with the other low-altitude urban sites [27, 56]. WS is known for the characteristics of cleanup function which influence the occurrence of pollutant concentrations [56]. Weak negative correlations
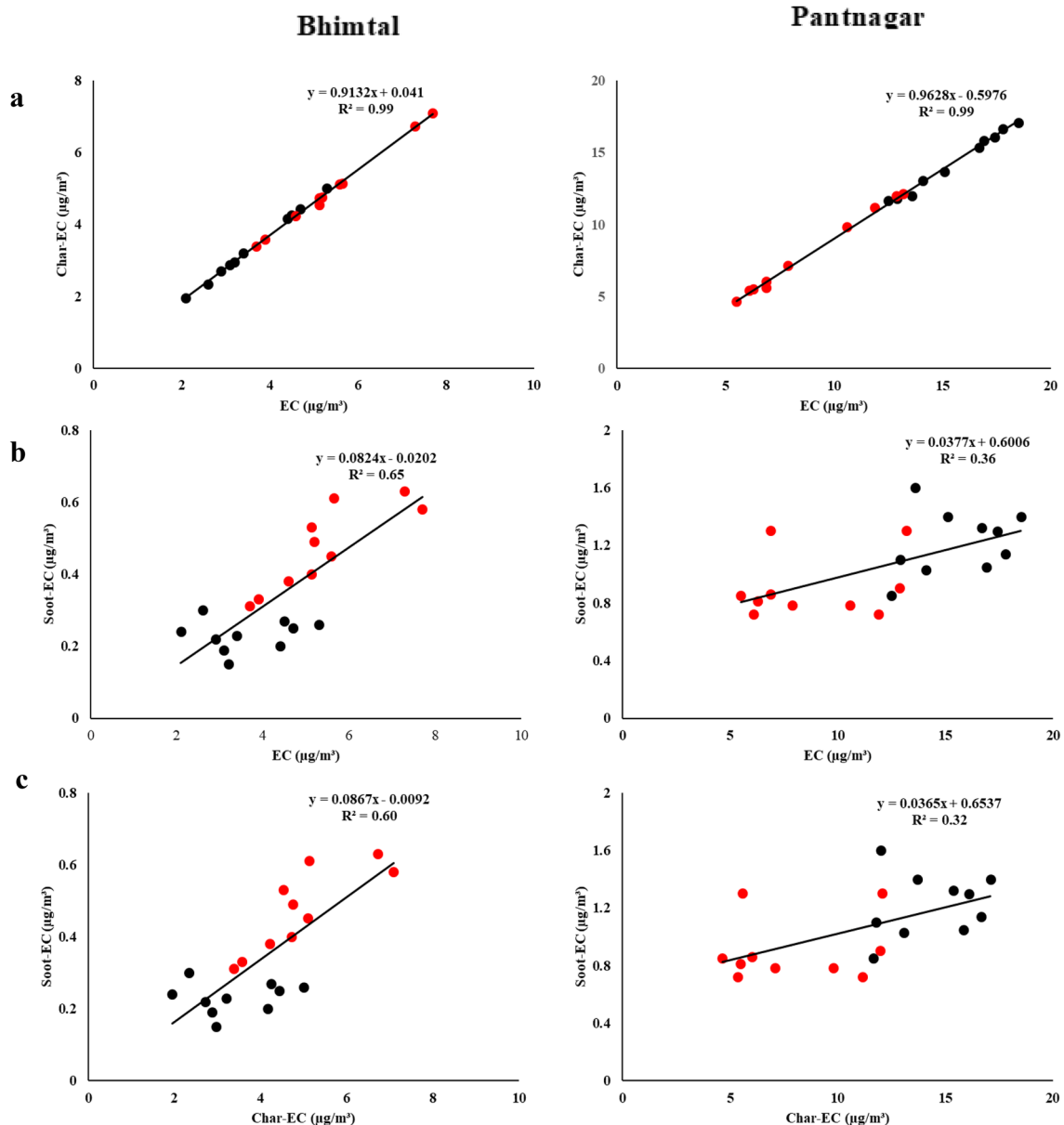
**Fig. 5** Regression plots between **a** EC and char-EC, **b** EC and soot-EC and **c** char-EC and soot-EC at Bhimtal and Pantnagar

were found for OC and EC with WS at Bhimtal. However, good negative correlations were observed for OC and EC with WS suggested the cleanup function of wind at Pantnagar. No significant correlation was observed for most of the carbonaceous components with RH at Bhimtal, while moderate positive correlations were found at Pantnagar.

## 4 Conclusions

Carbonaceous components of particulates are considered as one of the important agents for influencing atmospheric chemistry and local/regional air quality. This work

reports the first measurements in Bhimtal (high altitude urban) and Pantnagar (lowland urban) regions which cover the comparative assessment of different eight carbon fractions in carbonaceous aerosols, correlations among carbonaceous species and meteorological factors along with source attribution using char/soot ratio. Distinct seasonal differences of TSP, OC, EC and TCA concentrations were observed with respective average values of 99.4 $\mu g/m^3$, 15.8 $\mu g/m^3$, 4.5 $\mu g/m^3$ and 29.7 at Bhimtal, while 202 $\mu g/m^3$, 29.2 $\mu g/m^3$, 12.2 $\mu g/m^3$ and 58.9 $\mu g/m^3$ at Pantnagar. Higher summer values at Bhimtal could be attributed to the vertical movement of pollutants due to convective motions formed in the valley region. In contrast, Pantnagar

exhibited higher values in winter that could be associated with the low planetary boundary layer, low temperature and stable atmospheric conditions. Percentage contributions of OC, EC and TCA to total TSP were for almost 15.5%, 4.5% and 29.5 at Bhimtal and 15%, 6% and 30% at Pantnagar, respectively. Among the eight carbon fractions, OC2 and OC3 accounted for 29% and 21% of TC at Bhimtal, respectively, while EC1 and OC2 contributed for 26% and 25% of TC at Pantnagar, respectively. Char-EC and soot-EC showed almost similar seasonal patterns as that of OC and EC where char contributed significantly as ~ 90% and 88.5% to total EC at Bhimtal and Pantnagar, respectively. The char/soot ratios at both sites indicated that carbonaceous aerosols were mainly derived from biomass and vegetative burning. Correlation analysis among carbonaceous components and meteorological parameters has been conducted, and results demonstrated that these parameters were weak to moderate positively correlated with PBL, T and SR at Bhimtal and, however, showed anti-correlation at Pantnagar with these parameters.

The present study is preliminary work in this region and could provide a basic understanding of carbonaceous aerosols, formation mechanisms and source attributions. It is noted that smaller sampling periods at two sampling sites are the limitation of the current study; therefore, future studies need a more extensive sampling campaign and long-term quantitative approach along with modelling tools for different size fractions of aerosols.

## Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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