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Characteristics and Variability of Carbonaceous Aerosols over a Semi Urban Location in Garhwal Himalayas

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Abstract

Extraction of organic carbon (OC) and elemental carbon (EC) were carried out over Srinagar, India, an ecologically sensitive semi-urban site in Garhwal Himalays. The PM_{2.5} sampling was carried out during January to December, 2017 over the site. The OC and EC were extracted from PM_{2.5} samples using a thermo optical OC/EC analyzer. Highest OC and EC concncentrations were found during postmonsoon $(17.67 \pm 1.1 \ \mu\text{g/m}^3 \text{ OC}$ and $6.34 \pm 0.75 \text{ EC}$) and Winter $(17.31 \pm 3.045 \ \mu\text{g/m}^3 \text{ OC}$ and $6.32 \pm 0.585 \ \mu\text{g/m}^3 \text{ EC}$) seasons are attributed to boundary layer dynamics and anthropogenic activities. The lower concentration of OC/EC was observed during monsoon season $(11.64 \pm 1.75 \ \mu\text{gm}^{-3} \text{ OC}$ and $3.46 \pm 0.19 \ \mu\text{gm}^{-3} \text{ EC}$) owing to wet scavenging of aerosols and minimum count of forest fire/biomass buring incidences. Both pre-monsoon and post-monsoon season concentrations are also influenced by biomass burning in the IGP (Indo-Gangetic Plain) region and forest fires in the adjecent areas. The OC/EC ratio sounds that vehicular exhaust and biomass burning are the major source of OC/EC over the site. Generation of PM2.5 is contributed by carbonaceous aerosols (OC and EC) over Srinagar. The pivotal role of meteorology in modulating OC/EC concentrations has been illustared in detail.

Keywords Himalayan valley \cdot Elemental carbon \cdot Organic carbon \cdot Seasonal variation \cdot Biomass burning \cdot Influence on meteorology

1 Introduction

Carbonaceous aerosols are potential climate forcing agents and majorly exist in size range below $2.5 \ \mu m$. Subset of these particles mainly include carbonaceous species like organic carbon

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(OC) and elemental carbon (EC). The chemical and optical properties of BC and EC are almost same and constitute the most solar absorptive particulates among the fine mode particulate matter (Chung and Seinfeld 2002; Watson et al. 2005). Carbonaceous aerosols account for almost 40% of total PM2.5 mass in urban areas (Seinfeld and Pandis 1998). Incomplete combustion of fossil fuels and biomass burning produce EC in the atmosphere. However, oxidation of volatile organic compound (VOC) generate secondary organic carbon in the atmosphere in addition to primarily emitted organic carbon (Turpin and Huntzicker 1995). Intense combustion processes release EC in to the atmosphere. Temperature and oxygen availability are the major factors determining the amount of EC formed during a combustion process. Low temperature and less availability of oxygen produces more EC and vice versa in a closed chamber combustion process (Panicker et al. 2018). Fresh EC aerosols are more or less hydrophobic in nature and barely probable to act as CCN in usual atmospheric conditions due to non-significant hygroscopic growth and cloud condensation nuclei (CCN) activity (Tritscher et al. 2011). The ageing processes changes the hygroscopicity, morphology and chemistry of these aerosols. Experimental evidences prove that even though EC is photo chemically inactive, it catalyses the photo chemical ageing of soot by enhancing oxidation of volatile organic compounds (Li et al. 2018). EC concentration often indicates the extent of anthropogenic pollution in the atmosphere and act as a potent global warming agent (Li et al. 2013). Furthermore the radiative forcing by carbonaceous aerosols increase atmospheric heating, which can in turn weaken the vertical temperature gradient, hence leading to stabilization of atmospheric column and suppression of rainfall (Dave et al. 2017; Satheesh and Ramanathan 2000). Pollutants which are located in Himalayan valleys get transported to high altitude by phenomena called valley wind which are induced by the thermal gradient between valley and mountain top (Bonasoni et al. 2012; Dhungel et al. 2018). These local convective activities are capable of transporting pollutants to elevated layers of atmosphere and even can dwell much above the boundary layer. These aerosols in Himalayan region could be transported to adjacent glaciers by large scale circulations. Latest research have proved that deposited carbonaceous aerosols over snow have the potential to melt glacier by intensifying the snow albedo feedback (Jacobi et al. 2015; Nair et al. 2013; Xu et al. 2015; Lau et al. 2010). Model and satellite observational studies show that pre-monsoon aerosol loading over the Himalayan region caused an excess heating of 2.5 °C in last 4 decades (Gautam et al. 2009). Forest fire episodes continue to increase in Himalayan foothills in recent years. The quantification and possible impacts of emitted carbonaceous aerosols is a topic of major concern. The influence of anthropogenic aerosols, especially of absorbing kind have potential implications in local as well as large scale weather patterns and also in the extreme weather scenarios happened in the foot hills of Himalaya (Hazra et al. 2017). Since the models are incapable of simulating accurate aerosol concentration due to complex terrain structure over Himalayas, the lack of aerosol observation especially in the central and southern part of Himalayan region is a major issue in understanding the influence of aerosols on different climatic processes (Nair et al. 2013). Cong et al. (2015) clearly illustrates the penetration of biomass burning to high altitude Himalayas through mechanism of mountain/valley wind system. In this paper we are discussing about the variability and characteristics of elemental and organic carbon over Srinagar in Garhwal Himalayas. Long term observations of carbonaceous aerosols are inadequate over Himalayan region. Since pollutants are transported to Glaciers at very high rate in recent years, objective of the study fixed as quantification and source apportionment of carbonaceous aerosol over a Himalayan valley region. The main aims of this particular study includes quantification, source identification, role of meteorology and variability of carbonaceous aerosols, which will further help to understand the impact of aerosols on weather and climatic patterns of Himalayan region through energy budget modulation.

2 Data and Methodology

2.1 Sampling Location

Observations were carried out over Srinagar, (30.2247° N, 78.7986° E), Uttrakhand, which is a valley region located in Garhwal Himalayan area and is the largest town in the Garhwal hills of Himalaya. Srinagar town lies in the bank of river Alaknanda (Fig. 1). The temperature of the location oscillates between a maximum of 38 °C in summer and a minimum of 2 °C in winter with an average annual temperature of 16 °C. Precipitation peaks with an average cumulative rainfall of 370 and 330 mm/month in the month of July and August respectively (Mishra 2017). Major local pollution sources include vehicular exhaust during tourist season and forest fire incidences during dry seasons of the year. Residential biomass burning doubles during winter time due to heating of premises in addition to cooking processes. November and December months witness minimum rainfall of the region. Humidity varied between 40 to 80% during the observation period. The peculiar geological features and proximity to central Himalayan Glaciers make this region unique in its weather patterns. Srinagar area is also under the influence of long range transported pollutants mainly from dust within and outside the country. Western disturbances (WD) have significant impact in this area and hence the humidity is comparatively high during winter season (Hunt et al. 2018; Madhura et al. 2014). Since the valley can trap pollutants, chances of higher mass concentration of pollutants may also influence the weather patterns of this area. Srinagar was also heavily impacted by extreme weather patterns like cloud burst and had undergone catastrophic damage during 2013 Uttarakhand flood (Das 2013; Rana et al. 2013).

2.2 Methodology

PM_{2.5} data sampling was conducted using fine particulate sampler (APM550, MFC). The flow rate of instrument was set at 16.7 l/min. Atmospheric air enters in to the system through an omni directional valve system, which allows only particles less than 10 µm of aerodynamic diameter. Air further goes in to a second impactor with an aerodynamic cut point of 2.5 μ m where the separation of PM_{2.5} and PM₁₀ take place. Fine particulate air then passed through a quartz filter paper (2500 QAO-UP, Palls Life Science Inc.,) of 47 mm diameter which collects the fine particulate matter. Quartz filter papers were sterilized and decontaminated by heating at a temperature of 900 °C in a muffle furnace prior to sampling. Desert Research Institute's (DRI) thermal/optical carbon analyser with IMPROVE A protocol was utilized to analyze exposed PM sample to extract OC and EC. A small circular punch with an area of 0.5 cm^2 from the particle adsorbed filter paper is heated in programmed temperature steps. Basic principle of

Fig. 1 Sampling location



the instrument includes preferential oxidation of carbonaceous particles like OC and EC under different temperature and environment. Organic compounds are exposed to non-oxidizing helium environment and heated up to 580 °C, while EC combustion takes place in an oxidizing environment with $2\% O_2$ up to a maximum of 850 °C. The remaining carbon particles are oxidized to carbon dioxide using a reaction with heated manganese dioxide. Methanation technique is used further to reduce CO₂ to CH₄. Flame ionization detector then quantifies the CH₄ equivalent of organic and elemental carbon (Holm 1999). OC1, OC2, OC3 and OC4 are extracted in programmed temperature steps of 140 °C, 280 °C, 480 °C and 580 °C respectively. Similarly EC1, EC2 and EC3 are extracted at temperatures of 580 °C, 740 °C and 840 °C respectively (Ali et al. 2016). The SOC (secondary organic carbon) in total organic carbon has been estimated using the equation as shown below (Ram et al. 2008; Satsangi et al. 2012).

Secondary organic carbon, SOC = OC_{tot} -POC Primary organic carbon, POC = $(OC/EC)_{min} \times EC_{tot}$

OC/EC ratio technique is a method to identify the source characteristics of carbonaceous aerosols (Ali et al. 2016). Lower values of OC/EC ratio indicate diesel or gasoline based vehicular emissions (Schauer et al. 1999). Higher values usually represent long range transported aerosols (Saarikoski et al. 2008).

3 Results and Discussions

3.1 Monthly and Seasonal Variation of OC/EC

The monthly and seasonal variation of EC, OC is shown in Figs. 2 and 3 respectively. EC showed an annual average of $5.21 \pm 1.41 \ \mu gm^{-3}$ over the region. It is observed that EC mass

was highest in November $(7.16 \pm 0.27 \ \mu g/m^3)$ and lowest in July $(3.38 \pm 0.37 \ \mu g/m^3)$. Seasonally the highest EC mass concentration was found in post-monsoon $(6.34 \pm 0.76 \mu \text{gm}^{-3})$ followed by winter $(6.32 \pm 0.59 \mu \text{gm}^{-3})$ seasons. The lowest EC concentration $(3.46 \pm 0.19 \mu \text{gm}^{-3})$ was observed in monsoon season. High and low OC concentration was respectively observed in November $(21.56 \pm 1.41 \mu \text{gm}^{-3})$ and August (8.81 $\pm 1.38 \ \mu gm^{-3}$). Seasonally OC mass showed same pattern as EC with highest concentration in post monsoon $(17.67 \pm$ $1.1 \mu \text{gm}^{-3}$) and lowest in monsoon ($11.643 \pm 1.75 \ \mu \text{gm}^{-3}$). The lowest concentration of EC and OC observed during the rainy season could be due to wash out processes and reduction in biomass burning. Fire maps clearly shows the reduction in the biomass burning during the season (Fig. 5a: f; b: g,h,i). The high OC/EC concentration during post-monsoon/winter is associated with both meteorological and anthropogenic reasons. The long range transport of carbon from Indo-Gangetic plane



Fig. 2 Monthly variation of EC and OC mass concentration with standard deviation at Srinagar in the year 2017 (Vertical lines on the bar indicates standard deviation)



Fig. 3 Seasonal variation of EC and OC with standard deviation at Srinagar during the year 2017 (Vertical lines on the bar indicates standard deviation)

and increase in vehicular emissions due to pilgrimage activities could be major reason for enhanced OC/EC in post-monsoon. In winter, biomass burning is high over the site to keep the surroundings warm. These emissions contribute to enhancement in carbonaceous aerosol concentrations. Temperature inversion induces a positive temperature gradient is a well known phenomena associated with trapping of pollutions in the boundary layer (Panicker et al. 2013; Baumbach and Vogt 2003; Largeron and Staquet 2016). Valleys are more susceptible to persistent inversion processes catalyzed by aerosol radiative cooling (Anquetin et al. 2002). The meteorological reasons are associated with the lowering of mixing layer heights due to temperature inversions during winter seasons which result in lower ventilation of pollutants over the site (Devara et al. 2002; Raj et al. 2002). However, the lower concentrations in monsoon months are due to aerosol scavenging processes. High amount of EC in seasons except monsoon is significantly contribute to bad air quality over the region. Smog is also a predominant condition at this valley region due to inversion in winter season which can negatively impact the vulnerable population of the region (Saikawa et al. 2019). Apart from the health effects, carbonaceous aerosols of absorbing kind influence the dynamics and microphysics of the cloud by modulation of energy fluxes (Vinoj et al. 2014; Khain et al. 2005).

Srivastava et al. (2011) reported a different BC pattern over Manora peak, another Himalayan site, where high BC was observed during pre-monsoon and lowest was in monsoon season. Several studies in different stations in high altitude Himalayan regions observed peaking of BC in pre-monsoon season while the opposite is happening during winter. Altitude of these stations play a crucial role in this, as the observation sites in Himalayas are already located in mid or upper boundary layer. In central Himalayan region like Srinagar the transportation of pollutants from IGP seems less prominent due to geographical aspects but more number of vehicles during pilgrimage season contribute high amount of carbonaceous pollutants in pre and post monsoon season. Nair et al. (2013) reported average BC concentration of 106 and 196 ng/ m³ at Hanle (western Himalaya, 4.5 km msl) and NCO-P (central Himalaya,) sites respectively. The same study also reported almost steady BC mass concentration at mid to higher altitudes in post-monsoon season. These values are much less when compared to BC values at Srinagar due to the large difference in altitude and nonexistence of local activities. It is observed that BC concentration decreases with increase in altitude in the lower Himalayan stations, means high concentration at Himalayan valley and lower concentrations at higher altitudes (Dumka et al. 2011; Nair et al. 2013). Our study also indicates comparatively high BC mass concentration over this valley region of Himalaya.

3.2 Source Characterization of OC/EC

Source characterization has been carried out using OC/EC ratio technique (Cao et al. 2004; Pio et al. 2011; Ali et al. 2016) and shown in Fig. 4. Typically, the OC/EC ratiol to 4.2 represent vehicular exhaust (Ali et al. 2016). A OC/EC ratio of 7.7 represents biomass burning (Schauer et al. 1999; Ali et al. 2016; Feng et al. 2009). A larger part of the OC/EC ratios in Srinagar lie below 4.2 suggests that vehicular exhaust is the major source of carbonaceous aerosols over the region. June month showed highest OC/EC ratio of 4.92 followed by the month of September (3.31). Highest OC/EC ratio was observed in monsoon season followed by post monsoon and winter seasons over Srinagar. However, another study showed high values of OC/EC ratio in winter in Eastern Himalayas indicating intrusion of long range transported aerosol over the region from IGP (Rajput et al. 2013). High OC/EC ratios in



Fig. 4 Box plot showing monthly variation of OC/EC ratio at Srinagar in the year 2017 (Asterisk and square signs indicates upper and lower quartile respectively. Line in the middle of the box symbolizes median)

monsoon are reported by sevaral other studies in urban areas like Pune and Jabalpur (Panicker et al. 2015; Ali et al. 2016). Long range transport of aerosols seems to be less prominent or masked by the dominance of local emissions since OC/EC values are much lesser than 12 in the observation region (Saarikoski et al. 2008).

The significant contributor in altering OC/EC ratio is the contribution of secondary organic carbon (SOC). Secondary organic carbon concentration $(3.71 \ \mu gm^{-3})$ is associated with high OC/EC ratio in June month as washout processes are less prominent due to very less precipitation and high emission rate. SOC contributed more in winter season $(3.13 \ \mu gm^{-3})$ followed by pre-monsoon season $(2.89 \ \mu gm^{-3})$ in total organic carbon mass concentration.

Fire maps (VIIRS satellite fire counts) were used to illustrate the intensity of biomass burning in neighbouring region throughout the year (Fig. 5a and b). The contributions of longrange transport were examined through NOAA hysplit back trajectory analysis (Fig. 6). It is seen that long range transport from adjacent countries are prominent during pre-monsoon and post-monsoon season. Winter transports of aerosol to Srinagar are mostly confined from aerosol loaded IGP regions. Back trajectory in winter shows that the long range transport by western disturbances also influence the aerosols in the region during winter (Times of India 2018; Dimri and Chevuturi 2016). South-westerly wind patterns are also clearly seen during the monsoon seasons predicting the probability of oceanic aerosols at the observation site as suggested by Shrestha et al. (2002). Forest fire incidences in Uttarakhand and adjacent areas in Himachal are also found to be significant during pre-monsoon and post-monsoon seasons. Lowest OC/ EC ratio of 2.37 observed in the month of April. Since all the OC/EC ratios are above 2, the formation of SOCs are indicated throughout the year (Satsangi et al. 2012). Lower values of OC/EC ratio indicate constant sources of contribution for OC and EC almost throughout the year. They also indicate the dominance of vehicular emissions on biomass burning and long range transport over the region. Lower OC/EC ratios are mainly attributed to higher primary carbonaceous aerosols as suggested by Castro et al. (1999). The other major factors

Fig. 5 a: Fire maps (10 day count) a) January b) February c) March d) April e) May f) June (Blue dot on the map indicates the observation location,Red dots on the map indicate the location of atleast one fire during the 10 day composite period). **b**: Fire maps (10 day count) g) July h) August i) September j) October k) November 1) December (Blue dot on the map indicates the observation location,Red dot on the map indicate the location of atleast one fire during the 10 day composite period)



Longitude





Fig. 5 (continued)

influencing the OC/EC ratio are spatio temporal variation in emission and transport. The geographical location of Srinagar plays significant role in trapping local emissions in the valley region which enhances the influence of local emissions in comparison with the long range transport.

3.3 Influence of Meteorology in OC and EC

Meteorology and boundary layer dynamics play an essential role in spatio-temporal distributions of particulate matter. Boundary layer height which is controlled by the turbulent kinetic energy (TKE) plays major role in surface mass concentration of pollutants. Meteorological parameters and boundary layer height is depicted in Fig. 7. Boundary layer height is obtained from MERRA-2 reanalysis (https:// giovanni.gsfc.nasa.gov/giovanni) data. Lower boundary layer (BL) height enhances the pollutant accumulation at lower level. However, higher BL height induces high ventilation of pollutants. In Srinagar BL height showed significant influence on the concentration of carbonaceous aerosol during winter and post monsoon seasons. The lower BL height found to induce more pollutant accumulation during the above two seasons. However, pre-monsoon showed high surface mass concentration of carbonaceous aerosol irrespective of deep boundary layer, majorly due to increased fire incidents in northern states of India. Correlation of meteorological parameters with elemental and organic carbon is shown in Fig. 8. OC showed statistically significant (at 95% level) correlation with RH. However, EC didn't show any statistically significant correlation with RH. This could be due to the fact that EC is a hydrophobic species, however OC has both water soluble and insoluble components (Ram et al. 2012). Relative humidity plays role in formation of SOCs and influence CCN activation by changing optical and chemical properties (Roberts et al. 2002). Since freshly emitted EC is highly hydrophobic, humidity has more or less no role in deposition, optical properties and chemistry of EC aerosol (Tritscher et al. 2011). Higher formation of SOC during pre-monsoon also could be



a reason for carbonaceous species in atmosphere. Increase in the production of SOC is mainly attributed to gas to particle conversion during pre-monsoon (Ali et al. 2016; Panicker et al. 2015). Moderately high values of correlation coefficients indicate that EC and OC are significantly influenced by minimum temperature and maximum temperature of the region. The variation of minimum temperature during the year mostly can be taken as the variation in morning boundary layer height. Since surface temperature is the key driving factor of mixed layer height, minimum and maximum temperature influence the concentration of surface pollutants by modulating boundary layer dynamics.

3.4 Carbon-PM Ratio and Contribution of EC-OC in Total Carbon

Table 1 shows contribution of total carbonaceous (TC) aerosols (OC + EC) in total $PM_{2.5}$. Carbon to PM ratio provides the extent of carbonaceous aerosol pollution in the measured particulate concentration. Higher amount of carbon, especially elemental and brown carbon (BrC) in PM often indicates high solar absorption and induce high atmospheric warming (Husain et al. 2008; Xie et al. 2017), which could be inappropriate in Himalayan regions like Srinagar considering the proximity to glaciers. It is observed that 24–32% of total

Fig. 7 Monthly variation of meteorological parameters Maximum temperature (T max(°C)), Minimum temperature, (T min(°C)),Boundary layer height (BL_HGT(m)),Relative humidity (RH(%))at Srinagar during the year 2017



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Fig. 8 Correlation of EC and OC with meteorological parameters **a**) Maximum temperature (Tmax) vs EC **b**) Minimum temperature (Tmin) vs EC **c**) Relative humidity (RH) vs EC **d**) Tmax vs OC **e**) Tmin vs OC **f**) RH vs OC



PM2.5 consists of carbonaceous aerosol particles in Srinagar. Pre-monsoon (28.55) followed by post monsoon (28.27) showed highest percentage of carbon aerosol in total $PM_{2.5}$. Forest fires in surrounding region, winds from IGP region and generation of SOCs together cause high surface mass concentration of total carbon aerosols. High solar radiation and less

humidity are conducive for gas to particle conversion and produce secondary organic carbon in pre-monsoon season (Komppula et al. 2009). OC contributed almost 73–76% to the total carbon particulate, while EC represented the rest 24–27%. One of the reasons for high carbonaceous aerosols is crop burning, and also most fire incidences were reported to

Table 1 Monthly percentage contribution of OC, EC in total carbon and total carbon(TC) in PM (Particulate matter) during 2017 at Srinagar, Garhwal

MONTH	PM ($\mu g/m^3$)	OC_mass (µg/m ³)	EC_mass (µg/m ³)	TC/PM (%)	OC/TC (%)	EC/TC (%)
JAN	101.47	18.45	6.83	24.91	72.99	27.01
FEB	93.38	16.17	5.82	23.54	73.53	26.46
MAR	72.98	17.10	6.34	32.12	72.94	27.05
APR	71.9	13.94	5.87	27.56	70.36	29.63
MAY	80.15	15.78	5.02	25.96	75.85	24.14
JUN	80.03	17.27	3.50	25.95	83.13	16.86
JUL	55.37	8.86	3.38	22.12	72.33	27.66
AUG	40.09	8.82	3.42	30.54	72.01	27.99
SEP	46.03	11.62	3.51	32.88	76.78	23.21
OCT	73.15	13.78	5.52	26.38	71.40	28.59
NOV	95.23	21.57	7.16	30.17	75.05	24.94
DEC	103.16	20.02	6.09	25.31	76.67	23.32

happen in pre-monsoon and post monsoon seasons (Sati and Juyal 2016). May and November months showed drastic increase in fire incidences in Haryana, Punjab and UP area. Crop burning found to be the reason for this high fire counts during post monsoon and early winter season. Another Himalayan observation site at Darjeeling showed good correlation between $PM_{2.5}$ and BC indicating similar source sink connection and significant contribution by BC in fine mode particulate matter (Sarkar et al. 2019).

4 Summary

- Continuous carbonaceous aerosol measurements were carried out at a central Himalayan semi urban location (30.2247° N, 78.7986° E) for 1 year (January – December, 2017).
- 2. 2. Elemental carbon and Organic carbon concentration found to be highest during post monsoon $(17.67 \pm 1.1 \ \mu g/m^3 \text{ OC}$ and $6.34 \pm 0.75 \text{ EC})$ followed by premonsoon $(17.31 \pm 3.045 \ \mu g/m^3 \text{ OC}$ and $6.32 \pm 0.585 \ \mu g/m^3 \text{ EC})$ seasons attributed to biomass burning and local anthropogenic activities.
- 3. Monsoon season showed lowest concentration of OC-EC $(11.643 \pm 1.75 \ \mu gm^{-3} \text{ OC} \text{ and } 3.46 \pm 0.19 \ \mu gm^{-3} \text{ EC})$ due to rapid wet scavenging processes and reduced biomass burning.
- Main sources of pollutants were identified as vehicular exhaust and biomass burning. OC/EC ratio indicates local production of pollutants dominates over long range transport over this region.
- Boundary layer dynamics plays significant role in trapping pollutants over the region. Inversion layer acts a lid over the boundary layer and stops the dispersion above BL. Low boundary layer height induced increased concentration of carbonaceous aerosols over the region.
- 6. In pre-monsoon season high concentration of pollutants are seen irrespective of deep boundary layer because of the increased pollution influx due to biomass burning from the northern Indian states.
- 7. OC contributed 73–76% of total carbon and EC contributed the rest 24–27% of total carbon.
- A significant amount (24–32%) of carbonaceous aerosol contributed to total PM_{2.5} over the region indicates bad air quality of this remote semi urban location in Himalayan region.

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