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Variability of Carbonaceous Aerosols in Two Distinct Climatic Zones of the North-Western-Himalaya (India): An Expedition during Summer 2022

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The Northwestern Himalayas include two climatic zones: the temperate Kashmir Valley (KV) and the cold desert of Ladakh (LD), both hosting glaciers that sustain major Indo-Pak rivers. Carbonaceous aerosols (CA), key contributors to aerosol emissions, impact regional climate directly and indirectly. Aerosol were collected from four KV sites—Srinagar (SG), Qazigund (QZ), Pahalgam (PH), and Gulmarg (GM)—and two LD sites—Leh (LH) and Kargil (KG)—alongside real-time Black Carbon (BC) measurements during June-July 2022. Average BC concentrations were recorded as 6.36, 6.18, 6.10, 3.77, 1.10, and 0.94 µg/m' over GM, QZ, SG, PH, LH, and KG, respectively, with peaks observed at SG and PH in the morning and evening, while GM had an evening peak. KG and LH, the Cold Desert LD sites, have very weak BC concentration variation from morning to evening.

BC source apportionment shows fossil fuel contributions of 81-94% in KV and 82-87% in LD, with biomass burning contributing 6-19% in KV and 13-18% in LD. KV's bowl-shaped topography and higher anthropogenic activities increase fossil fuel BC, unlike LD's open topography and windy conditions. An inverse relationship was observed between wind speed (WS) and BC concentration, while rainfall showed a weaker correlation. BC was negatively correlated with relative humidity (RH) in SG and GM, whereas PH and KG showed weak RH correlations, and LH showed a positive RH-BC relationship.

OC/EC ratios indicate biomass burning as the primary CA source, with OC/EC increasing with altitude. Back-trajectory analysis identified regional and local winds blowing in KV and Indo-Gangetic Plain (IGP) as the primary wind source over KG and LH, with elevated BC levels linked to local winds over KV and when wind paths moved from KV in LD.

Keywords: Carbonaceous aerosols (CA), Organic carbon (OC), Elemental Carbon (EC), Black Carbon (BC), Kashmir Valley (KV), Ladakh (LD), Aethalometer, Back-trajectory analysis

1 Introduction

Aerosols exhibit diverse chemical compositions[1] that vary across spatial and temporal scales[2]. These variations significantly impact local and regional air quality[3], visibility, the global radiation budget, Earth's climate, and human health[4]. Among aerosols, Carbonaceous Aerosol (CA) is a key component, contributing up to 70% of the aerosol mass[5]. CA primarily consists of black carbon (BC) and organic carbon (OC)[6], with major sources including biomass burning (BB), fossil fuel combustion (FFC), and biogenic emissions[7]. Elemental Carbon (EC), quantified using offline thermal-optical methods, or BC, measured using real-time optical methods, constitutes a small fraction of CA by mass but plays a crucial role as the main absorber of visible light and a driver of global warming. In contrast, the impact of OC depends on its functional group composition and interactions with soot particles, leading it to either scatter light or contribute to warming[8]. Thus, quantifying and apportioning the sources of carbonaceous species that are OC and EC or BC in the pristine environments of the Himalayas, particularly the Kashmir Valley and Ladakh, is essential to understanding their implications for air quality, human health, and climate. It is highlighted that light-absorbing aerosols can alter regional atmospheric stability, influencing large-scale

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circulations along with hydrological cycle, which may explain recorded temperature and precipitation dynamics in India and China[9].

Organic carbon (OC) enters the atmosphere either directly through combustion processes such as cooking or indirectly through the mixed oxidation of volatile organic compounds (VOCs). It encompasses a diverse range of organic compounds forming from several sources and is classified into primary or secondary organic carbon based on its formation mechanism. Among its many species, compounds like polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) are recognized as potential mutagens or carcinogens. Additionally, water-soluble species of OC play a crucial role in cloud formation processes[10,11].

EC, also known as BC, is primarily released from partial combustion of fossil fuels and biomass[12]. BC contributes to positive radiative forcing by strongly absorbing solar radiation due to the high imaginary part (k) of its refractive index [13]. This absorption alters the global radiation budget through direct, semi direct, and indirect effects [14]. BC has a relatively short atmospheric longevity of 1 week to 10 days in the lack of precipitation [15]. Despite its brief duration, BC significantly deteriorates air quality[16] and exhibits the capacity to get transported to long distances, reaching remote regions devoid of local emission sources.

BC has gained significant attention from climate scientists for its exceptional light-absorbing properties, which drive various atmospheric feedback processes. These include modifications to the earth's radiation budget, forming cloud, broad-scale atmospheric circulation, air quality, visibility, and hydrological and carbon cycles[7,18]. The elevated concentrations of BC over the Indo-Gangetic Plain (IGP) make the Himalayan foothills particularly vulnerable, with adverse effects on Himalayan cryosphere[19]

BC is the largest contributor to global warming after carbon dioxide (CO_2), methane, along with volatile organic compounds (VOCs)[20-23]. Its radiative forcing, estimated as 0.4–1.2 Wm^{-2}, highlights its substantial impact on global warming[22] Projections indicate that average temperatures in the Indian subcontinent could increase by 3.5°C to 5.5°C by the end of the 21st century, while the Kashmir Himalayas may experience a higher rise of 6.43°C (±1.72)[24]. This temperature increase will exacerbate the melting of the Himalayan cryosphere[25,26] with BC and anthro-pogenic activities identified as major contributors to glacier retreat[27,28].

The Kashmir Valley, located in the Northwestern Himalayas having over 150 glaciers, is witnessing an alarming rate of glacier melt partly due to high BC concentrations. Elevated BC levels have significant implications for local and regional climate, environment, and human health[29]. Rapid urbanization in developing regions, particularly in China, South Asia, and Southeast Asia, has driven a sharp increase in BC emissions[30]. India and China together account for 25 to 35% of Earth's BC emissions, making them major hotspots[31]. Continuous BC monitoring in the Himalayas is crucial, given the region's role as a major reservoir of freshwater as snow and glaciers, which are rapidly melting due to climate change[32]. In pristine regions such as the Arctic and Antarctic, BC concentrations range from 10–140 ng m^{-3} and 0.1–30 ng m^{-3}, respectively [33,34]. In the Himalayas, BC levels have been reported at 0.09–2.7 μg m^{-3} on Mount Everest, 1.1–1.7 μg m^{-3} in Gilgit, 0.007–0.3 μg m^{-3} in Hanle, Ladakh, and 0.28–3.7 μg m^{-3} on the Machaoi Glacier in the Drass Valley [35-37].

Back trajectory analysis reveals that during non-summer months, dominant westerly air masses transport BC to Srinagar from Central Asian, West Asian, South Asian, African, and parts of European regions, along with local sources[38,39].

In recent years, the Kashmir Valley has emerged as the principal urban center in the Himalayan region, undergoing significant spatial expansion and rapid population growth[40,41]. Srinagar, the valley's urban hub, experiences a wide range of daily black carbon (BC) concentrations, varying from 0.56 μg/m³ to 40.16 μg/m³, with an average of 5.81 ± 5.31 μg/m³. These levels are higher than in most other Himalayan locations. Seasonal analysis reveals peak BC concentrations in November (13.89 ± 2.42 μg/m³) and the lowest in April (2.88 ± 0.57 μg/m³). Winter (8.60 ± 2.64 μg/m³) and autumn (8.31 ± 1.59 μg/m³) record elevated BC levels, whereas summer (3.23 ± 1.05 μg/m³) and spring (3.10 ± 0.66 μg/m³) show relatively lower concentrations. Increased temperature, precipit-ation, and wind speed significantly reduce BC levels. Biomass burning contributes to approximately 50% of BC emissions during autumn and winter but only 10% in summer. Elevated BC levels in the Kashmir Valley have critical implications for regional climate, human health, and the environment [29].

In contrast, Hanle in Ladakh records lower seasonal and annual BC levels compared to other

Himalayan stations, though still higher than those observed in the pristine Antarctic and South Pole environments[42]. In another study, it has been reported that aerosol radiative forcing over the western Trans-Himalaya exceeding the global average[43].

Carbonaceous aerosols (CAs) play a significant role in glacier melting by enhancing positive radiative forcing and reducing snow/ice albedo through deposition. These aerosols also impact the regional climate and glaciers of the Himalayas, often referred to as the Earth's "Third Pole," while posing risks to human health. This study investigates the qualitative and quantitative characteristics of CAs across six sites in the Kashmir Valley (Srinagar, Qazigund, Pahalgam, and Gulmarg) and the Ladakh cold desert (Kargil and Leh) during June and July 2022. The research aims to identify sources, analyze the influence of meteorological factors, and examine spatial and temporal variations of CAs. Analytical techniques included OC/EC analysis of TSPM using the thermal-optical method, BC measurements via an aethalometer, back-trajectory analysis of air masses using the HYSPLIT model, and meteorological data from nearby IMD stations was used.

This study is the first to comprehensively analyze CAs across six sites in the Kashmir Valley and Ladakh, employing thermal-optical and optical methods for OC, EC, and BC measurements and offering spatial comparisons. In recent years, numerous studies in India have focused on BC variability, its transport pathways, and its radiative impacts on high-altitude glaciers[44,45]. BC deposition on glaciers accelerates snow and ice melt, contributing to glacier retreat and impacting regional water cycles[46].

Despite this, limited observational studies have been conducted on BC, and even fewer on OC and EC, in the Kashmir Valley and Ladakh. While research on carbonaceous aerosols is available from other regions, the northwestern Himalayan region remains underexplored[47,48]. Given the low aerosol load in pristine Himalayan locations, TSPM (total suspended particulate matter, with diameters less than 40–50 μm) sampling was used to collect sufficient mass for chemical analysis. This field experiment provides the first insights into variations in BC, OC, and EC with altitude and their comparison across two distinct climatic zones: the temperate Kashmir Valley and the cold desert of Ladakh. The study integrates meteorological parameters to deepen our understanding of CAs in the northwestern Indian Himalayan region.

2 Experimental Section

2.1 Sites Description

According to the updated Köppen-Geiger climate classification, the J&K and Ladakh Himalayan region is categorized into 10 primary climate zones based on baseline climatic data. However, considering the specific climatic conditions of the region, the reported studies have simplified the classification into three distinct zones: the temperate zone encompassing the Kashmir Valley, the subtropical zone covering Jammu, and the cold desert zone, which includes Ladakh and the Gilgit-Baltistan regions[49]. Figure 1 illustrates the study area, while Fig. 2 provides background information on the sampling sites.

2.1.1 Kashmir Valley

Kashmir Valley a potent tourism region for domestic and international tourists, is situated in western Himalaya (known as Kashmir Himalaya) Srinagar is the main urban location of the valley situated almost at the center of the valley at 1592 meters from MSL, which can affect the valley's air quality on dispersal of winds. Kashmir Valley is a bowl-shaped topography surrounded by lofty snow-covered mountains of greater Himalaya from the north-east, Pir Panjal Mountain range from the south-west, and Shamshabari range in the north direction and is situated at an altitude of 1850m from MSL in north-western Himalaya. The valley is 135km long and 32 km wide, 15,520.3 km^2, with Jhelum a glacier and natural spring-fed major river. Kashmir is an administrative division of Jammu and Kashmir UT of India which includes 10 districts with Srinagar as a major urban location at the center which impacts the air quality of the valley. The Kashmir valley has a temperate type of climate four major seasons: cool spring (march to early May), pleasant summer (early May to late August with 15–18 degree Celsius temperature), cool autumn (mid-September to November), and cold winter with snowfall (December to February). The lofty mountain encircling the valley hinders the pollution dispersal from the valley. It has its highest precipitation in the form of rainfall in spring and in the form of snowfall in winter, whereas monsoon winds get weakened due to the Pir Panjal Mountain range in the south and southwest of the valley so usually do not have a major effect on the valley's weather. Cooking, lighting, heating, industrial and vehicular emissions are the common sources of pollution in the valley. From the beginning

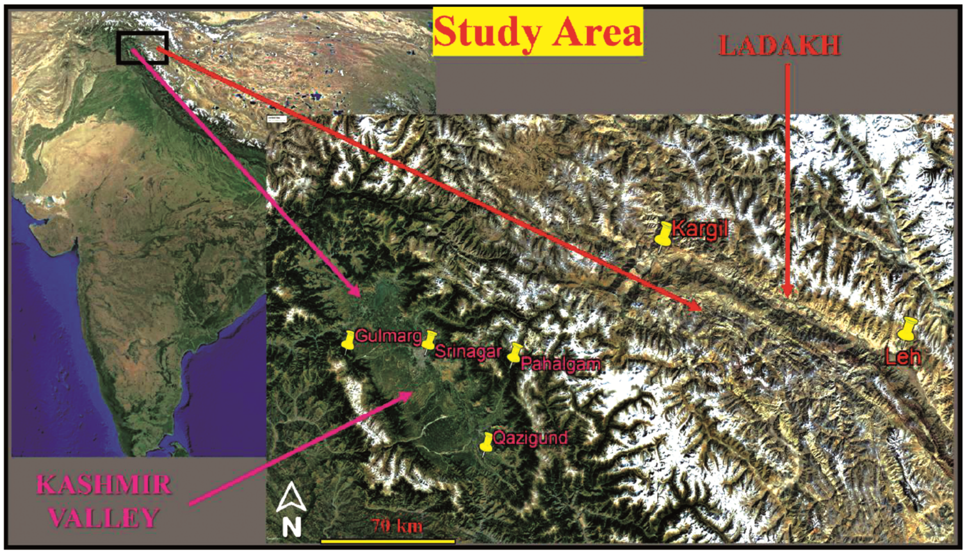

Fig. 1 — Study area: Map of India showing the study area Kashmir Valley and Ladakh regions in the northwestern Himalayas in a black-colored rectangle in left-hand side image and Zoomed-in map of Kashmir Valley (including sites, Srinagar, Qazigund, Pahalgam, and Gulmarg) and the Cold Desert Ladakh (including Kargil and Leh) on the right-hand side

Fig. 2 — Photographs of the sampling sites, Srinagar, Qazigund, Pahalgam, Gulmarg, Kargil, and Leh

of winter until March, biomass and biofuel are commonly used for household heating. Additionally, the combustion of dry twigs and leaves from deciduous vegetation and orchards from October to November is a significant practice in the Kashmir valley. In recent years, the Kashmir Valley has emerged as the dominant urban hub in the entire Himalayan region, experiencing substantial spatial expansion and grappling with rapid population growth [40,41]. The sampling over the KV has been done at three different i.e. south (Qazigund), east (Pahalgam), west (Gulmarg), and center i.e. Srinagar to have representative samples of the valley from every side. The elevations of sampling sites from sea level in meters are 1585, 1670, 2740, and 2650 of Srinagar, Qazigund, Pahalgam, and Gulmarg respectively.

2.1.2 Ladakh

Ladakh is the Union territory of India located in the northwestern Himalayas (west of the Tibetan plateau) with an average elevation of 6000 m from MSL lying in the rain shadow region possesses the arctic type climate having sparse vegetation and is considered as a “cold desert’’. It shares its boundaries with China and Pakistan and is important for the strategic and climatic point of view. It consists of rocky and sandy mountain peaks along with many high-altitude peaks covered by snow and glaciers throughout the year.

Ladakh is an arid mountain region experiencing harsh winter and cool summer with low humidity creating dry atmospheric conditions. Winds are generally high. It consists of two administrative districts Kargil and Leh having a total population of 2.74 lakh people depending upon glaciers and the Indus River for their water requirements. No major industrial activities are found in the Ladakh region, but the pristine climate and glaciers of the region are very sensitive to human activities affecting air quality which needs to be studied. The sampling was done at two different populated towns (locations), Kargil and Leh in early July. Until now only a few BC measurement data have been published from the Ladakh. No OC/EC (by thermal optical method) data has been published from the region till now.

2.2 Materials and Methods

Real-time BC measurement and TSPM sample collection have been carried out in summer 2022 at Srinagar from 13th to 17th June and again from 26th to 28th June, Pahalgam from 23rd to 25th June, Gulmarg from 29th June to 2nd July, Kargil from 7th to 11th July, and Leh from 13th to 15th July.

2.2.1 Black Carbon Measurement (By optical method)

2.2.1.1 Aethalometer and BC Measurement

The MicroAeth MA300 portable aethalometer, weighing 715 grams (25.22 ounces) and powered by a rechargeable battery, was utilized to measure black carbon (BC) at various locations across the Kashmir Valley and Ladakh. This instrument conducts real-time analysis across five wavelengths 880 nm, 625 nm, 528 nm, 470 nm, and 375 nm by detecting the rate of change of transmitted light caused by the constant deposition of particles onto a Polytetrafluoroethylene (PTFE) filter tape cartridge. Measurements were collected at 85 sampling locations.

The wavelength of 880 nm specifically measures the BC mass concentration, while 375 nm interprets Ultraviolet Particulate Matter (UVPM), which is suggestive of organic emissions such as smoke from wood, biomass burning, and tobacco smoke. The device simultaneously collects samples at two spots using different flow rates, and its automated tape advancement system ensures uninterrupted operation for extended periods without manual intervention. The MA300 employs the DualSpot® loading compensation method, which corrects for variations in particle optical properties in real-time based on differences in particle age and composition. During operation, the device maintained a flow rate of 150 ml/min, regulated by an internal mass flow meter with a closed-loop control system.

The optical attenuation (ATN) is calculated using Equation I, based on the transmitted light intensity from a blank filter spot (I_0) and a particle-deposited filter spot

$$ATN = -100 \ln (I/I_0) \quad \ldots (1)$$

The change of attenuated light (ΔATN) which is a function of time (Δt), filter spot size (S), flow rate (F), leakage factor (ζ), wavelength-dependent mass absorption cross-section (σair), multiple scattering parameters (C) and compensation parameter (k) is used to calculate the spectral BC mass concentration as in equation II:

$$BC_\lambda = S\,(\Delta ATN / 100)/F(1-\zeta)\sigma air C(1-kATN)\Delta t \quad \ldots (2)$$

Here, R(ATN) is the shadowing correction factor (constant).

The Spectral absorption coefficient for aerosols (βabs) at the wavelength (λ) of the Aethalometer is estimated using Equation III.

$$\beta_{abs}(\lambda) = BC_\lambda \times \sigma/C \times R(ATN_\lambda) \quad \ldots (3)$$

2.2.1.2 BC Source Apportionment

The identification of black carbon (BC) sources, such as fossil fuel combustion and biomass burning, is achieved by analyzing data from the UV (370 nm) and IR (880 nm) channels. Different aerosol types exhibit distinct absorption characteristics at these wavelengths. Organic carbon aerosols from biomass burning strongly absorb UV radiation, making the 370 nm wavelength highly sensitive to this source. In contrast, BC from fossil fuel combustion predominantly absorbs IR radiation at the 880 nm wavelength, which is the standard for measuring BC concentrations [51].

Thus, Light absorption measurements at UV and IR wavelengths indicate the relative contributions of black carbon (BC) from biomass burning and emissions from traffic in the atmosphere[52]. This approach effectively differentiates between the dominant sources of BC in a given environment.

2.2.2. OC EC (By thermal-optical method)

2.2.2.1 Sample Collection

Total suspended particulate matter (TSPM) was collected from five different sites in Kashmir Valley and

Ladakh during June and July 2022 on pre-weighed and pre-backed (for 4 hours at 450°C) 47 mm quartz filter paper. Oil-free suction pump with a 16 LPM flow rate was used for TSPM collection from morning to late evening along with a 47 mm filter paper holder attached with a silicon tube. A mass flow controller was used to maintain the flow rate. After collection, the filter papers were made moisture-free before weighing again by putting them in a silica gel-filled desiccator and then stored in air-tight poly-bags at 4°C temperature in a refrigerator to avoid loss of volatile components. TSPM concentrations have been calculated through the gravimetric method by using a sampler run-time, flow rate, and weight of the dust deposited on it. A blank filter paper was also conditioned in a similar way to know the background values of OC EC and TC.

2.2.2.2 OC EC Analysis

The analysis of organic carbon (OC) and elemental carbon (EC) in total suspended particulate matter (TSPM) samples was done using the OC-EC analyzer developed by Sunset Laboratory, Portland, USA. This analysis was performed at the Himalayan Aerosol Research Instrumentation (HARI) facility, a joint initiative of the Central University of Jammu (CU Jammu) and the Jammu & Kashmir Pollution Control Committee, located on the CU Jammu campus.

The instrument employs the thermal-optical transmittance (TOT) technique, adhering to the NIOSH-5040 method established by the National Institute of Occupational Safety and Health. It has a measurement range of 0.2 to 600 micrograms per square centimetre for both total carbon (TC) and organic carbon (OC), and a range of 0.2 to 30 micrograms per square centimetre for elemental carbon (EC). The device offers a detection limit of 0.10 micrograms per square centimetre.

The analysis involves heating the quartz filter samples in two distinct stages. In the first stage, the sample is heated in a non-oxidizing helium (He) atmosphere at a temperature of 870°C. During this step, carbon compounds are converted into carbon dioxide (CO_2), which is subsequently reduced to methane (CH_4) and measured using a Flame Ionization Detector (FID). The carbon content released during this phase is quantified as organic carbon (OC).

In the second stage, the sample is cooled to 550°C before being reheated to 870°C in an oxygen-rich atmosphere. This phase allows for the release of carbon compounds that are quantified as elemental carbon (EC). The total carbon (TC) concentration is then calculated by summing the values of OC and EC. This comprehensive process ensures precise identification and quantification of the carbonaceous components in the aerosol samples.

2.2.3 Back-trajectory analysis of winds and Meteorological Parameters

A portion of the carbonaceous aerosols (CA) observed at various sites can be attributed to non-local sources, transported over long distances. To identify the long-range transport pathways of these aerosols and their contribution to the observation sites, 72-hour backward wind trajectories were simulated for each day using the HYSPLIT model, available at NOAA's Air Resources Laboratory[53]. Meteorological parameters were incorporated into the model using Global Data Assimilation System (GDAS) 1-degree data. Wind trajectories were simulated at three different altitudes 100 m, 500 m, and 1000 m above ground level across all observation sites to pinpoint potential regions contributing to long-range aerosol transport.

The simulations revealed that, outside of the summer season, westerly winds play a dominant role in transporting black carbon (BC) to Srinagar from Central Asian, West Asian, South Asian, African, and parts of European regions. This transported BC adds to the pollution originating from local sources pollution [54].

To evaluate the impact of meteorological parameters on BC concentrations, daily variations in BC levels across all sites were analyzed alongside data on wind speed, rainfall, relative humidity, and temperature. Data of meteorological parameters was taken from the Indian Meteorological Department (IMD) observatories located either at or near the sampling sites: Srinagar, Qazigund, Gulmarg, and Leh observatories were co-located with the sampling locations, while Pahalgam and Kargil observatories were approximately 600 meters and 200 meters away from their respective sampling points.

3 Results and Discussions

3.1 BC Concentrations variation

3.1.1 Daytime Variation

Diurnal or daytime (morning to evening) variation of BC mass concentration is an important indicator of anthropogenic activities, sources, and impact of meteorology. Figure 3 shows the daytime variation of BC concentration from morning to evening over all the sites on all the days. A slight variation in the time of reading in the morning and evening is due to the reachability issue to the site during the field campaign. The data gap can be seen in the Figures because of the technical errors/glitches in the MicroAeth. Higher BC concentrations during morning and evening, whereas low concentrations during the day are observed at Srinagar city and Pahalgam. The reasons are low morning and evening temperatures which cause less convection and dispersion and low PBL which is inverse in the case of the daytime where the increase in temperature increases the PBL height, convection, and dispersion resulting in more dilution and dissipation of BC. Also, more traffic during morning and evening along with more combustion activities during cooking hours and running of diesel generators for electricity cause more BC in these hours. Gulmarg experiences more BC concentration in evening hours because of less dispersion due to changes in the height of PBL and use of diesel-run electricity generators along with cooking and heating activities. The major sources of BC in Srinagar are emissions from vehicles, household biomass burning, coal combustion, and BC coming from surrounding BC hotspots, whereas in Gulmarg the major sources of BC are vehicular exhaust and hardwood burning[55]. It is found the lowest concentration of BC at Srinagar and Gulmarg during summer. More BC concentration in the morning and evening times is also attributed to more RH[55].

The main local sources of BC over Kargil and Leh are traffic and household emissions. A constant BC concentration from morning to evening has been recorded at Kargil and Leh the two sites of Ladakh (Cold desert) which indicates fewer BC sources, constant regional transport of BC, and maximum dilution due to open desert topography and windy conditions of the sites. Constant BC concentration indicates the constant background source which can be the long-range transport of BC and is minimal as compared to the other sites. In another study also reported very weak diurnal variation of BC at Hanle, Ladakh[56]. No study has been published until now from these sites to our knowledge.

3.1.2 Daily Average with Meteorological Parameters

Figure 4 shows a daily variation of BC over all the sites and its variability with meteorology that is Wind Speed (WS), Relative Humidity (RH), Temperature (T), and Rainfall (RF). Table 1 shows mean values of BC, OC, EC concentration, OC/EC ratio, Wind Speed, Temperature, R.H, and Precipitation of different sampling days of respective sampling sites. One day BC measurement has been recorded over Qazigund as 6.19 $\mu g/m^3$ and respective OC, EC, OC/EC, WS, Temperature, R.H, and precipitation have not been mentioned in the table due to lack of proper observation on the site because of reachability issue during field campaign.

The BC mean concentration over different sites has been observed as 6.36±0.83 > 6.19±0.07 > 6.10±1.38 > 3.77±0.75 > 1.10±0.20 > 0.95±0.14 micrograms/m^3 over Gulmarg, Qazigund, Srinagar, Pahalgam, Leh, and Kargil respectively. Maximum mean concentration has been observed at Gulmarg a hill station (in Kashmir Valley) and minimum at Kargil (site of the cold desert Ladakh). More BC concentration is observed at all sites of Kashmir Valley (Srinagar, Qazigund, Gulmarg, Pahalgam) a temperate climate and more populated region compared to the sites of Ladakh (Kargil, Leh) a cold desert, arctic climate with less anthropogenic activities due to less population. Srinagar a major urban location, Qazigund a commercial activities site having more vehicular rush, particularly diesel-run trucks, and Gulmarg a major tourist activity site show a more BC concentration. Minimum BC concentration among sites of Kashmir Valley has been observed at the tourist place Pahalgam which is at the highest altitude among all. The observed average BC concentration over Srinagar is higher than the long-term annual and long-term summer observation by[57] whereas it is lower than the long-term observations of[58]. The higher BC concentration during observation time is also because of more vehicular rush in June and July because of more tourist rush and Srinagar as summer capital increasing anthropogenic activities. The BC concentration observed over Gulmarg is higher than the long-term annual and summer time reported in 2017[59]. No BC measurement has been reported from Qazigund and Pahalgam until now. Whereas other studies reported BC concentration between 0.114 $\mu g/m^3$ to 1.034 $\mu g/m^3$ over the Kalahoi Glacier at about 3700 meters AMSL about 16 km away from Pahalgam in Kashmir Valley[60].

Kargil and Leh sites of the cold desert Ladakh region have recorded the minimum BC concentration as compared with the sites of Kashmir valley is attributed to high altitude, less anthropogenic activities, minimum traffic emissions, windy conditions, minimum RH due to the desert climate and open topography. Leh town has observed more BC mass concentration compared to

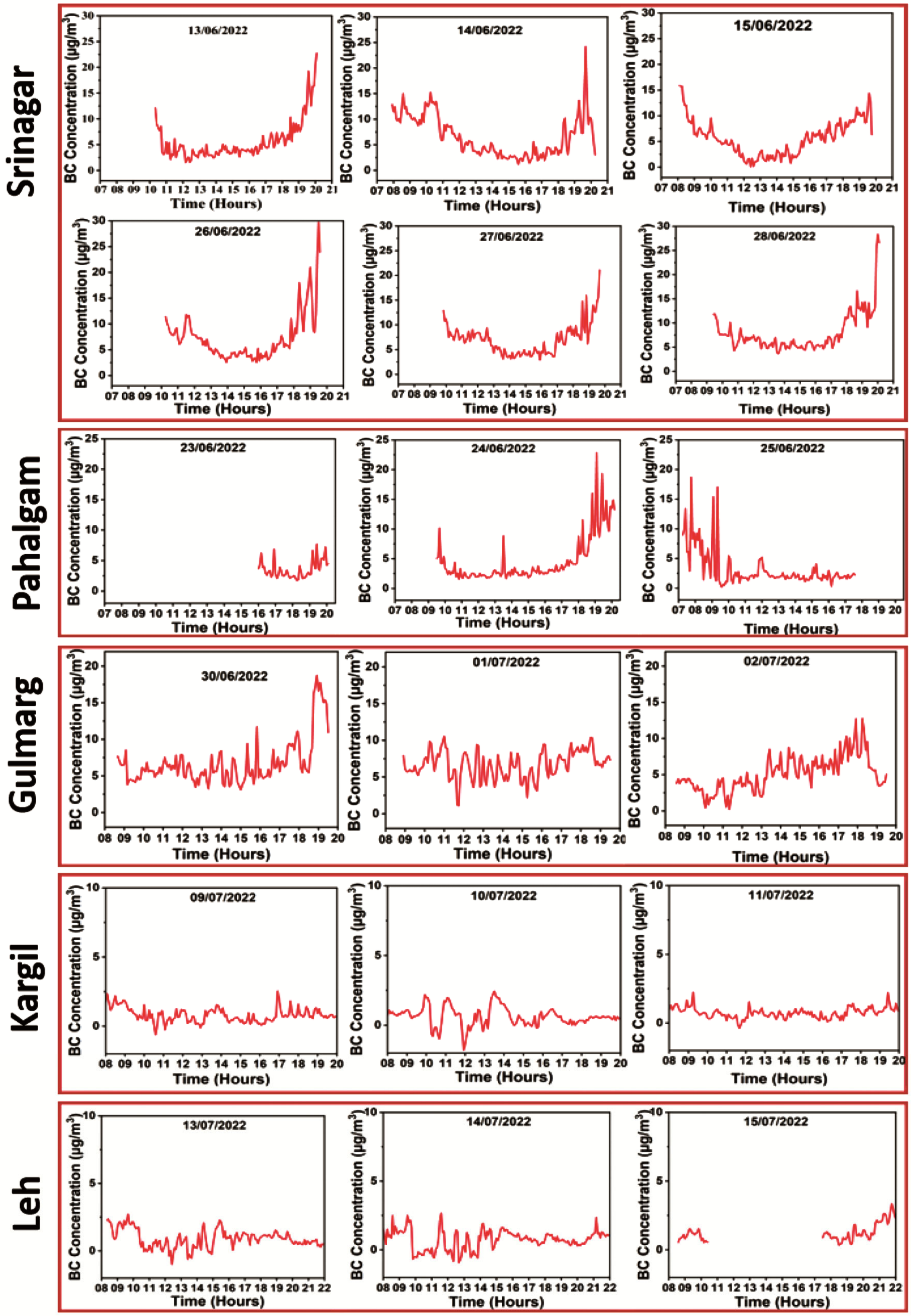

Fig. 3 — Daytime variation of BC concentration over Srinagar, Pahalgam, Gulmarg, Kargil, and Leh on different days

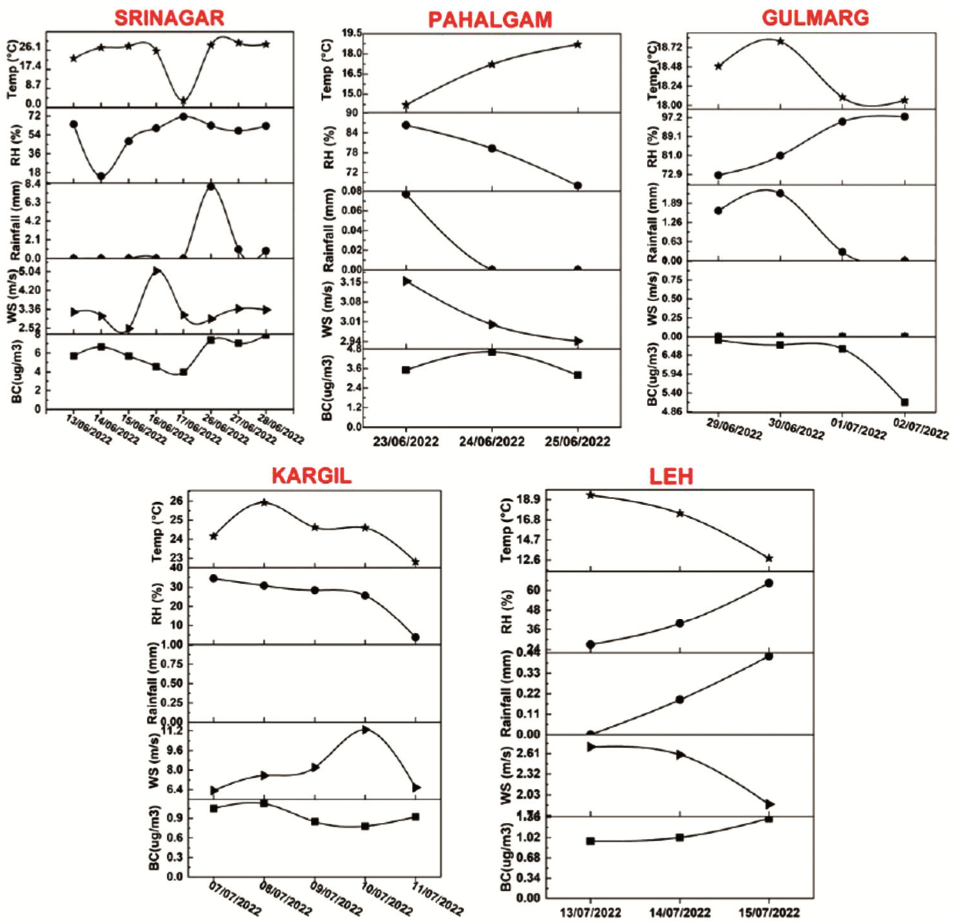

Fig. 4 — Daily variation of BC mass concentration along with meteorological parameters, WS, Rainfall, RH, and Temperature over the observation sites

Table 1 — Daily mean BC, OC, EC concentration, and OC/EC over the sampling sites.

Sites	Mean BC ($\mu g/m3$)	Mean OC ($\mu g/m3$)	Mean EC ($\mu g/m3$)	OC/EC	WS (m/s)	Temperature (°C)	RH (%)	Precipitation (mm)
SRINAGAR	6.10	36.61	5.34	6.86	3.33	26.81	55.17	1.24
PAHALGAM	3.77	41.89	3.01	13.92	3.03	16.71	77.85	0.03
GULMARG	6.36	44.85	7.59	5.91	Data not available	18.36	86.64	1.03
KARGIL	0.95	27.01	3.75	7.20	8.00	24.42	24.58	Data not available
LEH	1.10	27.13	0.22	123.32	2.40	16.54	43.66	0.20

Kargil because of comparatively more anthropogenic activities and vehicular emissions due to more population than Kargil. A few BC studies have been reported from the Hanle, Ladakh which is about 467 km away from Kargil and about 255 KM from Leh. A study reported less than 0.4 µg/m^3 BC mass concentration during a long-term measurement during 2009-2010 at Hanle which is 2-3 times less than the observed concentration at Kargil and Leh [61]. This difference is attributed to the large distance of Hanle from observation sites, populated observation sites, and more anthropogenic activities in 2022 compared to 2009-2010 in the Ladakh region. The BC mass concentration reported is higher than many high-altitude sites such as Hanle, Chirbasa near Gangotri glacier, Muztagh Ata, Qilian Mountain, Langtang Nepal, and Mukteshwar, except Kashmir Valley, Darjeeling, Shillong, Kullu, and Dehradun where BC mass concentration has been reported above than Kargil and Leh. No BC measurement has been reported from Kargil and Leh until now.

Daily variation of BC concentration on sampling days with WS, RF, RH, and T can be observed in Fig. 4. A general negative trend of BC mass concentration is observed with the WS over all the sites except Gulmarg where the WS data is not available. A weak correlation/trend of BC is observed with rainfall, and rare rainy days, and less/statistically insignificant data during sampling can be the reason for improper comparison. BC and RH show a negative trend with one another over Srinagar and Gulmarg explains the scavenging of BC from the atmosphere due to RH. A weak comparison/ relationship between BC and RH is seen over Kargil and Pahalgam. Whereas over Leh the BC increases with the increase of RH and it could be due to the availability of humidity over low humid regions causing BC to build up. BC and T are showing positive trends with one another at Srinagar, Gulmarg, and Kargil, opposite trends over Leh, and a weak correlation over Pahalgam.

3.1.3 BC Source Apportionment

Absorption at 370 nm (UV) and 880 nm (IR) wavelengths in aethalometers are widely used for BC source apportionment and are attributed to BC_{bb} (BC from biomass burning) and BC_{ff} (BC from fossil fuel). BC source identification over all the observation sites for all days shows the contribution of BC_{ff} several times greater than the BC_{bb}.

Figure 5 (a) shows the contribution of BC_{ff} and BC_{bb} in total BC by mass concentration. Srinagar has observed BC_{ff} concentrations as 3.38, 6.40, 3.90, 1.98, 2.92, 5.08, 5.33, and 5.63 µg/m^3 and BC_{bb} concentrations as 0.41, 0.56, 0.86, 0.47, 0.37, 0.55, 0.31 and 0.40 µg/m^3 on 13, 14, 15, 16, 17, 26, 27, and 28 June respectively. BC_{ff} and BC_{bb} concentrations over Qazigund have been observed as 4.17 and 3.70 µg/m^3 on 19 and 20 June respectively. At Pahalgam, the BC_{ff} and BC_{bb} concentrations have been recorded as 2.99, 2.79, and 3.08 micrograms/m^3 and 0.29, 0.52, and 0.44 microgram/m^3 on 23, 24, and 25 June respectively. Gulmarg has recorded BC_{ff} and BC_{bb} concentrations as 4.59, 4.34, and 2.46 microgram/m^3 and 0.46, 0.31, and 0.30 microgram/m^3 on 30 June, 1st July, and 2nd July respectively. Over Kargil, the BC_{ff} and BC_{bb} concentrations have been recorded as 1.09, 0.80, 0.59, 0.58, and 0.83 µg/m^3 and 0.21, 0.15, 0.09, 0.12, and 0.15 on 7, 8, 9, 10, and 11 July respectively. Whereas, the BC_{ff} and BC_{bb} concentrations over Leh are recorded as 0.59, 0.88, and 1.14 µg/m^3 and 0.13, 0.17, and 0.22 µg/m^3 on 13, 14, and 15 July respectively.

However**,** Fig. 5(b) shows the average share of BC_{ff}% as 89, 80, 88, 91, 85, and 83 and BC_{bb}% as 11, 20, 12, 09, 15, and 17 at Srinagar, Qazigund, Pahalgam, Gulmarg, Kargil, and Leh respectively. Approximately similar contributions from biomass burning and fossil fuel combustion have been reported over Srinagar [62,63] and over Gulmarg [63]. The BC_{bb}% at the Qazigund location is comparatively more and results in comparatively less BC_{ff}% is indicative of Qazigund as a rural environment where the biogenic contribution of BC is more than that of fossil fuel.

3.2 OC, EC, and their Comparison with BC over the Sites

OC and EC have widely been studied to know the source apportionment of the CA [64] and to examine the role of anthropogenic activities in air pollution globally in general and in developing nations such as India in specific. Anthropogenic biomass burning is

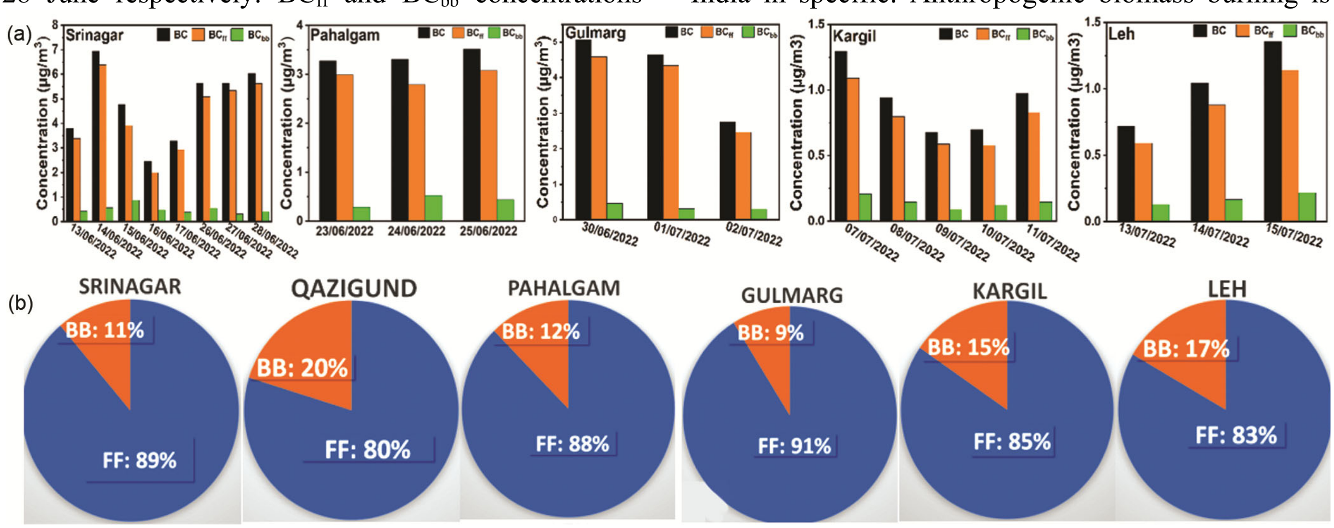

Fig. 5 — (a) Showing BC source apportionment, i.e. daily BC_{ff} and BC_{bb} mass concentration at all the observation sites, and (b) BC source apportionment as BC_{ff} and BC_{bb} in percentage over different sites

prevalent in Northern India[65] and the OC/EC ratios help in quantifying the fractional contribution of biomass-burning biogenic sources, and emissions from fossil fuels in carbonaceous aerosols [66]. Generally, the EC/TC ratios and EC concentration remain low over the different locations of pristine environments like the Himalayas. Whereas, higher EC concentrations have also been observed over many urban locations of the region due to increased anthropogenic activities and fossil fuel combustion. In this study, the OC/TC ratios over the observation sites have been studied along with their variation with the altitude and over different climatic zones i.e., Kashmir Valley and Ladakh. Figure 6 shows OC/EC ratios as 5.74, 9.25, 7.35, 6.33 on 14, 15, 26, and 28 June over Srinagar, 14.77, 13.66 on 24, and 25 June over Pahalgam, 7.98, 8.42, 3.71 on 30 June, 01 July, and 02 July, over Gulmarg, 4.96, 23.96, 18.50 on 08, 09, and 10 July over Kargil and 180.36, 99.49 on 13, and 14 July over Leh respectively. The average OC/EC values over different sites have been calculated as 7.17, 14.22, 6.70, 15.81, and 139.92 over Srinagar, Pahalgam, Gulmarg, Kargil, and Leh. The OC/EC ratios over Srinagar and Gulmarg are between 4 to 12 indicating biomass-burning sources[67] as major contributors of CA over the sites.

In a reported study diverse organic compounds in TSPM samples collected in Srinagar and observed a decrease in crustal input along with increased biomass combustion leading to higher organic content in the winter season [68].

Very high OC/EC ratios over Pahalgam, Kargil, and Leh show the minimum contribution of EC in CA over the sites. Figure 7 (a) shows the variation of EC/TC ratios calculated using the thermal method whereas BC was observed through the optical method with altitude. Values of both parameters show an identical trend with the increasing altitude. The concentrations can be seen decreasing with altitude except Gulmarg having the highest concentration because of more tourist activities over the sites and Kargil showing less concentration than the Pahalgam because of the Cold desert sites having open topography and windy conditions with minimum anthropogenic activities. Sites of the Ladakh region show less EC/TC and BC concentration values than the Kashmir Valley because of the cold desert climate and different topography, unlike the bowl-shaped topography of Kashmir Valley. OC/EC concentration, as shown in Fig. 7 (b), describes an increasing trend with altitude.

3.3 Back-Trajectory Analysis

Back trajectory analysis of winds coming over Srinagar, a central-most location of Kashmir Valley studied by different works shows most of the winds prevail from the west side along with a significant number of winds from the south, southwest, and southeast (Indo-Gangetic region).

During summer along with IGP a significant amount of BC_{bb} travels from the Mediterranean region where episodes of biomass burning can be observed during that time[67]. Another study showed that during

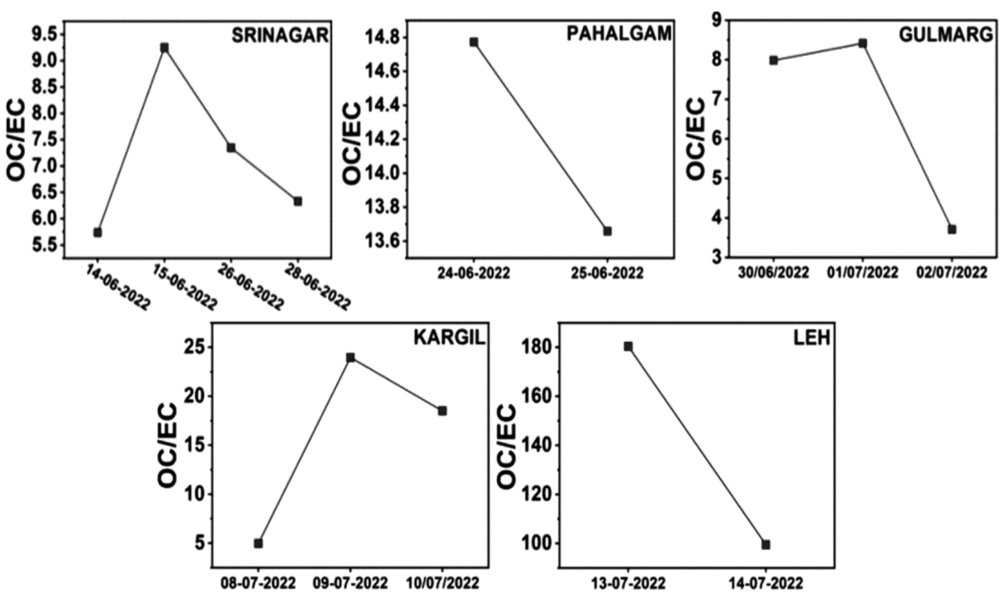

Fig. 6 — OC/EC over different observation sites

summer less than 30% of the winds coming to the valley are from westerlies, during that time low biomass burning occurs in the path from the middle-east to the valley, whereas the remaining air parcel comes from the southwest monsoon[69].

Figure 8 shows a Back-trajectory analysis of air masses coming over the observation sites analyzed using the HYSPLIT Model of Air Resource Laboratory shows the wind parcels come from the south, and southwest directions along with IGP and local (valley) regions during all days. Red, green, and blue colored lines show different heights of wind parcel movement as 1000, 500, and 100 meters above ground respectively. Over Srinagar on 26, 27, and 28

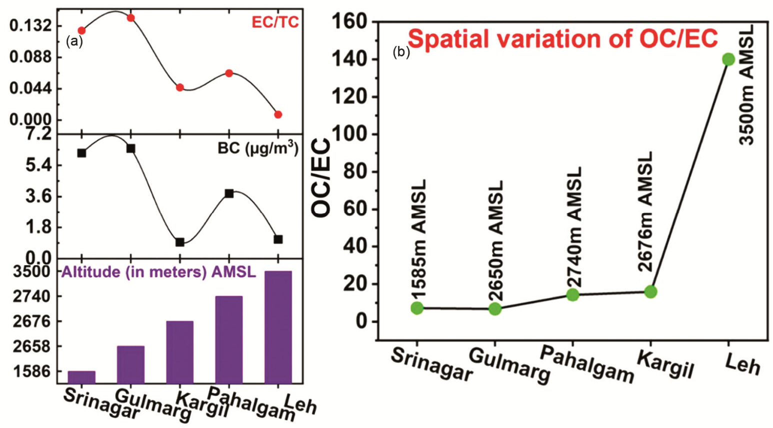

Fig. 7 — (a) BC mass concentration and EC/TC with altitude, and (b) Spatial OC/EC variation with altitude

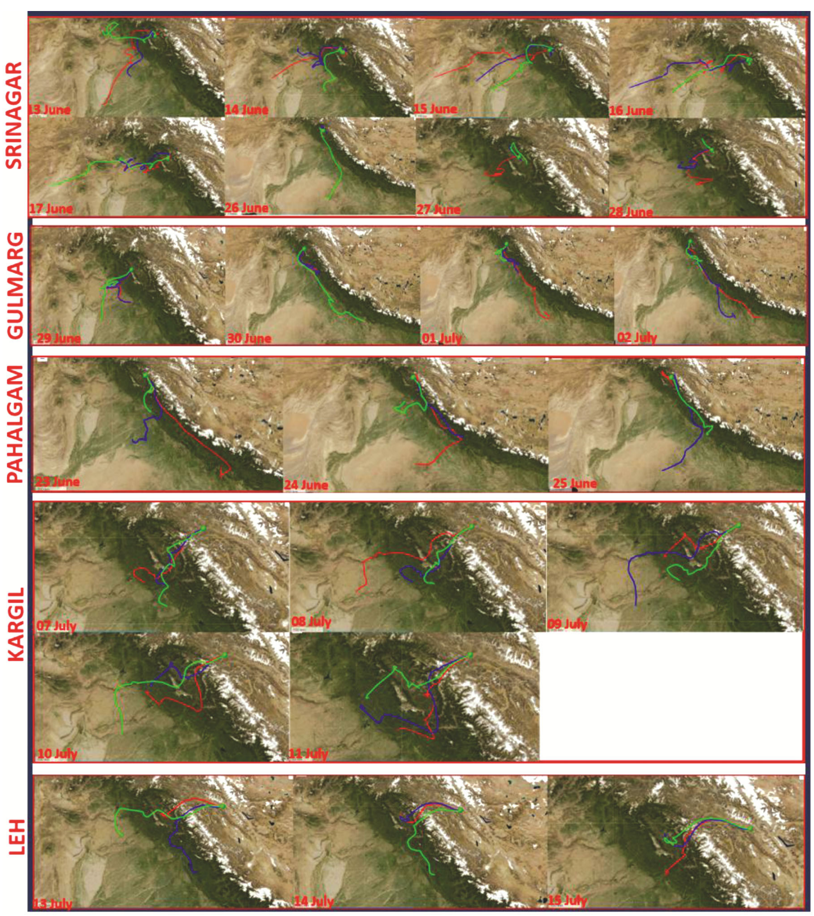

Fig. 8 — Back-trajectory analysis of air parcel movement over all the sampling sites on different days

June when the BC concentration was highest, the wind parcel movement was almost local, and on 13, 14, 15, 16, and 17 June when the BC concentration was observed as comparatively less than 26, 27, and 28 June, the wind movement was local along with a significant amount from the Mediterranean and south-west monsoon. At Gulmarg on 29 June the winds were local and the BC concentration was highest on that day, whereas on other days wind movements were monsoonal from IGP causing low BC over the observation site. Air parcel movement over the Pahalgam observation site on all days remains from IGP (southeast) regions. At Kargil, the wind parcel movement took the path of the Valley and is coming from the south and southeast of the valley which contributes to BC concentration over Kargil. Over Leh, on all three observation days, the wind parcel movement remained from IGP. The wind parcel movement along with the behavior of BC mass concentration over Kashmir valley describes the local (from valley) sources as prevalent over the sites. Whereas over Kargil the BC was more when wind parcel movement was from the valley side which means Kashmir valley is the source region of BC over Kargil and other regions of Ladakh Desert. IGP and KA are the sources of CA over Ladakh during the summer.

4 Conclusions

The purpose of the study is to monitor the carbonaceous aerosol (CA) (real-time BC and offline analysis of OC EC) in collected TSPM over the pristine environment of Kashmir Valley (KV) and Ladakh (LD). The study includes morning-to-evening variation and daily variation of BC concentration, the impact of met parameters on CA concentration, source apportionment of BC (i.e. BC_{ff} and BC_{bb}) including weightage quantification, and spatial percentage variation. The OC/EC, EC/TC vs EC, and back trajectory analysis of air masses coming over four different field sites of Kashmir valley (Srinagar, Qazigund, Gulmarg, Pahalgam) a temperate climate and two field sites (Kargil and Leh) of cold desert Ladakh along with their spatial variation along and altitudes have been studied.

BC variation from morning to evening at Srinagar and Pahalgam shows peaks in morning and evening, with low concentration in the daytime. At Gulmarg peaks in the BC concentration have been observed in the evening. Kargil and Ladakh the two sites of Ladakh show very weak BC variation from morning to evening. The high BC mass concentration has been observed over the Kashmir valley compared to the Ladakh region. Srinagar, Gulmarg, and Qazigund show comparatively higher BC mass concentrations. BC concentration shows a decreasing trend with the increase in altitude, except Gulmarg a hill station which shows the highest BC mass concentration. WS shows an inverse relationship with BC mass concentration, whereas rainfall shows a weak relationship probably because of a lack of precipitation during the sampling days. Over Srinagar and Gulmarg BC shows a negative trend with RH, whereas a weak relationship between RH and BC is observed at Pahalgam and Kargil. Leh shows a positive relationship between RH and BC mass concentration.

BC source apportionment reveals that fossil fuel is a major contributor to BC over the observation sites. Gulmarg has the highest contribution of BC_{ff} (91%) followed by Srinagar (89%). Over Leh, the BC_{ff} contribution has been observed as a minimum (83%) followed by Kargil (85%). BC_{bb} over Qazigund is observed as the highest (20%) due to its rural type of environment attributed to more biogenic sources of BC.

OC/EC over all the observation sites is observed as high and is attributed to biomass burning as the primary contributor of CA. Several times higher OC/TC ratio is observed over Leh indicating a minimum contribution of EC in CA. Both, BC measured through an aethalometer and EC through the thermal method have been compared which shows a similar trend over all the sites and a general decreasing trend with altitude.

Back-trajectory analysis shows local as well as regional winds prevalent over Kashmir Valley, and Kashmir Valley along with IGP as the source region of winds over Kargil and Leh. It has been observed that whenever local winds dominate the BC mass concentration increases. It has also been observed that whenever winds traveling to Kargil and Leh take the path of Kashmir valley the observed BC mass concentration increases, so KV is also contributing to BC over the Ladakh region.

The Kashmir Valley, with its bowl-shaped topography, high population density, and numerous urban and commercial centres, experiences significant anthropogenic activities, leading to increased levels of pollution, particularly carbonaceous aerosols (CA). In contrast, the Ladakh region, characterized by its open terrain, windy conditions, and sparse population, exhibits minimal anthropogenic influence resulting in minimal pollution. Consequently, the Kashmir Valley records higher CA concentrations and a greater contribution of fossil fuel combustion to black carbon (BC) mass concentration than Ladakh. Furthermore, the relatively humid conditions in the Kashmir Valley exacerbate aerosol accumulation by enhancing atmospheric stagnation, thereby increasing CA concentrations, and augmenting the light-absorbing properties of BC.

Fine particulate matter seriously risks human health, climate, ecosystems, visibility, and air quality. Its chemical composition primarily includes organic carbon (OC), elemental carbon (EC), and water-soluble inorganic ions (WSIC), which contribute to its adverse environmental and health impacts[70]. The Indo-Gangetic Plain (IGP) is known for elevated aerosol levels especially carbonaceous aerosols due to severe pollution, which typically peak in October and November. This increase in aerosol load is primarily due to seasonal factors such as agricultural activities, biomass burning, and industrial emissions. The onset of winter precipitation, influenced by western disturbances, contributes to a reduction in AOD through rainout processes, where aerosols are removed by precipitation. Additionally, stable meteorological conditions in the pre-winter period enhance the persistence of pollution, including carbonaceous aerosols, as weak winds and limited atmospheric mixing hinder the dispersion and removal of pollutants[71,72]. In the Himalayan region, particularly in Kashmir Valley and Ladakh, aerosol loading is highly dynamic, influenced by topography, meteorological conditions, and distinct anthropogenic activities. This contrasts with the Indo-Gangetic Plain (IGP), where aerosol levels are more consistent due to emissions from agriculture and industrial sources. During the pre-monsoon period in the IGP, dust storms contribute to elevated aerosol concentrations. These dust particles age and can develop coatings, including carbonaceous aerosols, altering their characteristics, and influencing air quality and radiative forcing [73].

This study will help the readers to know the mass concentrations, sources, and source region of CA over field sites of Kashmir Valley and Ladakh and their comparison along with a comparison between two different climatic zones (Kashmir Valley and Ladakh) of North-western Himalaya. It will also help for modeling studies and simulations of pollution over pristine environments like Kashmir and Ladakh and to know their impact on the environment and Himalayan Cryosphere. As this study is just a short-term field expedition, further long-term study of CA over Kashmir and Ladakh and their comparison is required.

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