



Wintertime Variation in Carbonaceous Components of PM₁₀ in the High Altitudes of Himalayas [†]

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Abstract: Carbonaceous aerosols play a significant role in the Earth's atmospheric system by affecting visibility, the hydrological cycle, the climate, radiative forcing, and human health. The present study analyses PM₁₀ samples that were collected at three distinct urban locations (Mohal-Kullu, Nainital, and Darjeeling) over the Himalayan region of India during winter 2019. The mass concentrations of PM₁₀ were recorded as $51 \pm 16 \mu\text{g m}^{-3}$, $38 \pm 9 \mu\text{g m}^{-3}$, and $52 \pm 18 \mu\text{g m}^{-3}$ for Mohal-Kullu, Nainital, and Darjeeling, respectively. Organic carbon (OC) dominated over elemental carbon (EC) and was found to be 50.2%, 42.8%, and 47% of total carbon (TC) at Mohal-Kullu, Nainital, and Darjeeling, respectively. The respective mass concentrations of carbonaceous species [OC, EC, water-soluble organic carbon (WSOC), and total carbonaceous aerosol (TCA)] were higher at Mohal-Kullu (OC: 11.1 ± 5.3 , EC: 4.2 ± 1.9 , WSOC: $5.3 \pm 1.3 \mu\text{g m}^{-3}$, and TCA: $22.1 \pm 10.4 \mu\text{g m}^{-3}$) followed by Darjeeling (OC: 5.4 ± 2.0 , EC: 2.7 ± 1.0 , WSOC: $3.9 \pm 1.3 \mu\text{g m}^{-3}$, and TCA: $22.1 \pm 10.4 \mu\text{g m}^{-3}$) and Nainital (OC: 2.9 ± 1.0 , EC: 1.3 ± 0.6 , WSOC: $2.1 \pm 0.6 \mu\text{g m}^{-3}$, and TCA: $6.7 \pm 2.4 \mu\text{g m}^{-3}$). The OC/EC and WSOC/OC ratio at Mohal-Kullu (2.6 ± 0.3 , 0.6 ± 0.2), Nainital (2.0 ± 0.4 , 0.7 ± 0.2), and Darjeeling (2.3 ± 0.5 , 0.7 ± 0.2), respectively, indicates the dominance of fossil fuel combustion (coal and vehicular exhaust), with signified additional contribution from secondary organic carbon (SOC).

Keywords: carbonaceous aerosols; water-soluble organic carbon; secondary organic carbon; trajectories

1. Introduction

Atmospheric aerosols are well known as a major pollutant worldwide due to their complex composition, their effects on visibility, and their effects on the heat balance of the Earth [1,2]. High loading of aerosol poses severe implications for human health, global climate change, and the Earth's radiation budget [3,4]. Carbonaceous aerosols (CAs) are the key component of particulate matter (PM), constituting 20–70% of coarse particulate matter [4]. Carbonaceous components of PM are classified as organic carbon (OC) and elemental carbon (EC), in terms of their optical and physical properties [4,5]. OC is emitted from combustion or indirectly from the heterogeneous oxidation of volatile

organic compounds (VOCs). OC has a wide variety of organic compounds originating from various sources and it is further classified as primary organic carbon (POC) and secondary organic carbon (SOC), in terms of its formation. EC is emitted primarily from the incomplete combustion of biomass (BB) and fossil fuel combustion (FFC) [6–8]. As the CAs play a crucial role in atmospheric chemistry, Earth’s radiation budget, human health, and the air quality of the region, it is very important to measure the concentrations of the carbonaceous particles to understand their transport, their sources, and their deposition.

The Himalayan region is regarded as pristine and vulnerable to environmental change due to regional and global change [6]. There has been little research on carbonaceous particles, their sources, their movement, and their climatic effects [7–13] but are not adequate for studying the detailed effects of CAs on the environment. The present study on CAs at different altitudes of the Himalayas fills this gap to some extent

The present study estimates the concentration of CAs at different locations (Mohal-Kullu, Nainital, and Darjeeling) in the Himalayan region of India during the winter season (January–February 2019). We chose the winter season for the study because of the increased BB and CC activity for heating purposes, as well as the steady and stagnant air conditions that can lead to large concentrations of CAs. Using ratios, we identify the origins and their contribution to CAs across the different study locations.

2. Methodology

2.1. Observation Sites

We conducted the study over different sites in the Himalayan region of India i.e., Mohal-Kullu, Nainital, and Darjeeling.

Mohal-Kullu: G. B. Pant’s National Research Institute of Himalayan Environment and Sustainable Development, Mohal-Kullu (31.9° N, 77.11° E, and 1154 m a.s.l.) is located in the western Himalayan region. The study site has a sub-temperate environment and receives heavy rain during the winter period.

Nainital: Aryabhatta Research Institute of Observational Sciences (ARIES), Nainital is located on Manora hill (29.39° N, 79.45° E, 1959 m a.s.l.) in the central Himalayan region.

Darjeeling: Bose Institute, Darjeeling (27.01° N, 88.15° E, 2200 m a.s.l.) is located in the eastern Himalayas. The study site is geographically adjacent to the Indo-Gangetic plain (IGP) region (a global hotspot for heavy aerosol loadings) [10]. A detailed description of the sampling location has been discussed in previous studies [7–10,14–16].

2.2. Sample Collection and Analysis

The PM₁₀ samples were collected during the winter season (January–February 2019) twice every week for 24h using a respirable dust sampler with a flow rate of 1.2 m³ min^{−1} on pre-baked (550 °C for 5 h) Pall flex Tissuequartz filters (20 cm × 25 cm at three urban locations of Mohal-Kullu, Nainital, and Darjeeling.

The measurements of carbonaceous species (OC, EC) in the PM₁₀ samples collected over the study locations were carried out using an OC/EC analyzer. This instrument works on the thermal–optical approach. WSOC in the PM₁₀ samples was quantified using a TOC analyzer (Model: TOC L_{CPH/CPN}, Shimadzu), which works on the principle of catalytic-oxidation combustion at high temperatures of 680 °C. A WD-XRF (wavelength dispersive X-ray fluorescence) spectrometer was used for the quantification of the elements in PM₁₀, ranging from Barium to Uranium. Details of the OC/EC analyzer, TOC analyzer, and WD-XRF spectrometer are available in references therein [9–12].

3. Result and Discussions

3.1. Mass Concentration of PM₁₀ and Carbonaceous Components

In the present study, the average PM₁₀ concentrations at Mohal-Kullu, Nainital, and Darjeeling were 51 ± 16 µg m^{−3}, 38 ± 9 µg m^{−3}, and 52 ± 18 µg m^{−3}, respectively.

The findings that were acquired for the study sites are comparable to the previously reported PM₁₀ aerosol mass and carbonaceous components at different sites in India (Table 1).

In the present study, high concentrations of OC (Darjeeling: $5.36 \pm 1.74 \mu\text{g m}^{-3}$; Nainital: $2.85 \pm 0.75 \mu\text{g m}^{-3}$; Mohal-Kullu: $10.49 \pm 4.56 \mu\text{g m}^{-3}$), EC (Darjeeling: $2.67 \pm 0.87 \mu\text{g m}^{-3}$; Nainital: $1.30 \pm 0.46 \mu\text{g m}^{-3}$; Mohal-Kullu: $4.06 \pm 1.99 \mu\text{g m}^{-3}$), and WSOC (Darjeeling: $3.93 \pm 1.17 \mu\text{g m}^{-3}$; Nainital: $2.04 \pm 0.54 \mu\text{g m}^{-3}$; Mohal-Kullu: $5.23 \pm 1.35 \mu\text{g m}^{-3}$) were observed (Figure 1a). The high concentrations of carbonaceous components might be due to the increased contribution from wood burning for household heating, open biomass burning, and the stable atmospheric conditions leading to pollution deposition within the lower atmosphere [4,6]. The observed OC and EC concentrations are in good accordance with other reported studies (Table 1). The SOC and POC were quantified in this study using the EC tracer method [5].

Table 1. Comparison of carbonaceous components and their mass ratios over high altitude locations in India.

Sampling Site	Altitude (m asl)	Time Period	PM ₁₀ ($\mu\text{g m}^{-3}$)	OC ($\mu\text{g m}^{-3}$)	EC ($\mu\text{g m}^{-3}$)	WSOC ($\mu\text{g m}^{-3}$)	OC/EC	WSOC/OC	References
Mohal-Kullu (31.9° N, 77.11° E)	1154	January–February 2019	51 ± 16	10.4 ± 4.5	4.0 ± 1.9	5.2 ± 1.3	2.6 ± 0.3	0.55 ± 0.23	Present study
Nainital (29.39° N, 79.45° E)	1959	January–February 2019	38 ± 9	2.8 ± 0.7	1.3 ± 0.4	2.0 ± 0.5	2.3 ± 0.5	0.74 ± 0.15	Present study
Darjeeling (27.01° N, 88.15° E)	2200	January–February 2019	52 ± 18	5.3 ± 1.7	2.6 ± 0.8	3.9 ± 1.1	2.0 ± 0.3	0.74 ± 0.22	Present study
Pohara (32.2° N, 76.2° E)	750	January–April 2015	52 ± 19	6.8 ± 2.3	4.8 ± 2.0	-	1.5 ± 0.4	-	Kaushal et al., 2018
Dharamshala (32.2° N, 76.3° E)	1350	February–April 2015	39 ± 23	5.0 ± 3.0	2.5 ± 0.6	-	2.0 ± 1.0	-	Kaushal et al., 2018
Manora Peak (29.39° N, 79.45° E)	1950	February–March 2005	138 ± 78	11.6 ± 5.9	1.8 ± 0.8	-	6.6 ± 0.3	-	Ram et al., 2008
Palampur (32.1° N, 76.5° E)	1300	March 2013	47 ± 7	6.7 ± 2.2	1.6 ± 0.9	-	4.3	-	Sharma et al., 2014
Kullu (32.2° N, 76.3° E)	1154	March 2013	34 ± 1	4.8 ± 1.6	1.9 ± 0.7	-	2.9	-	Sharma et al., 2014
Manora Peak (29.39° N, 79.45° E)	1950	2014–2017	32.1 ± 2.7	8.1 ± 6.0	2.4 ± 1.5	-	-	-	Srivastava and Naja, 2021

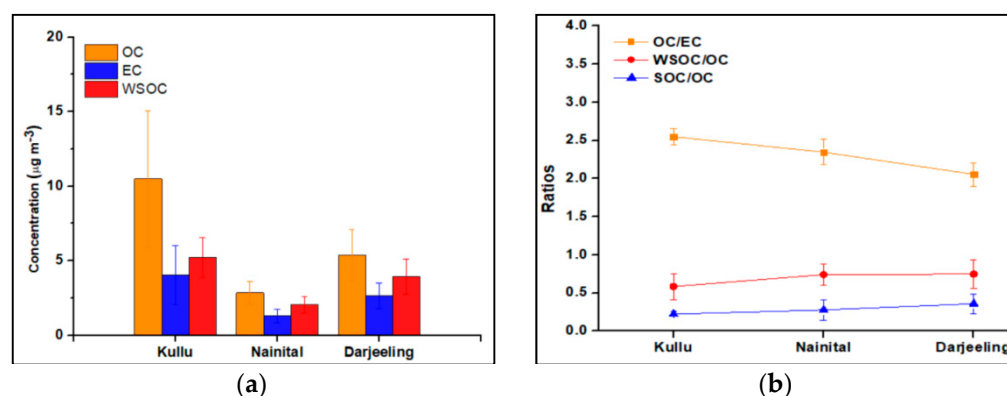


Figure 1. (a) Mass concentration of carbonaceous components of PM₁₀ (b) diagnostic ratios of carbonaceous components at different study sites of the Indian Himalayan Region IHR.

At Mohal-Kullu, Nainital, and Darjeeling, an average POC concentration was observed as $8.24 \pm 3.73 \mu\text{g m}^{-3}$, $2.13 \pm 0.93 \mu\text{g m}^{-3}$, and $3.41 \pm 1.27 \mu\text{g m}^{-3}$, respectively, during the study period (Table 1). The SOC concentration was observed as $2.90 \pm 1.97 \mu\text{g m}^{-3}$, $0.75 \pm 0.36 \mu\text{g m}^{-3}$, and $1.99 \pm 1.10 \mu\text{g m}^{-3}$ at Mohal-Kullu, Nainital, and Darjeeling, respectively.

3.2. Diagnostic Ratios and Scatter Plots

The diagnostic ratios of OC/EC and WSOC/OC are important for the identification of CA emission sources and secondary organic aerosol (SOA) formation and their removal

rates and the aging of the atmospheric aerosols [17–20]. Figure 1b shows the OC with EC and the WSOC with OC ratios at Mohal-Kullu (2.6 ± 0.3 , 0.6 ± 0.2), Nainital (2.3 ± 0.4 , 0.7 ± 0.1), and Darjeeling (2.06 ± 0.37 , 0.7 ± 0.2), respectively, which indicates the frequent practices of biomass burning during the season and the formation of SOC due to the transported carbonaceous species (Figure 2) from the nearby IGP region [9–22]. The acquired ratios of OC with EC and WSOC with OC are in good agreement with earlier reported studies [11–22].

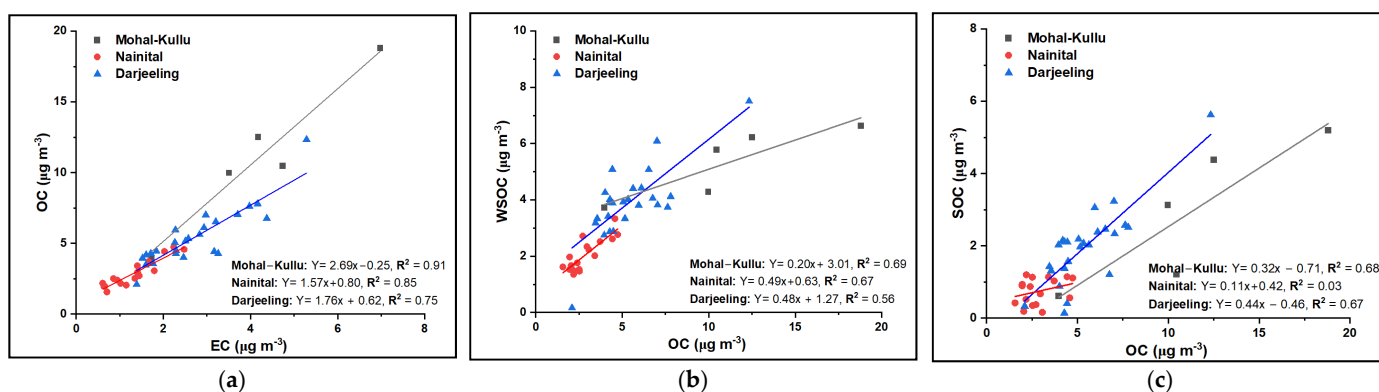


Figure 2. Scatter plots of carbonaceous components of PM_{10} (a) OC with EC (b) WSOC with OC (c) SOC with OC at different sites of the IHR.

The OC and EC exhibited a significant correlation throughout the study period (Darjeeling: $R^2 = 0.765$; Nainital: $R^2 = 0.854$; Mohal-Kullu: $R^2 = 0.912$) (Figure 2a), indicating similar sources of CAs (fossil-fuel combustion and BB) and the emission potential of the sources [4,20,22]. Throughout the study, a significant correlation of WSOC with OC (Figure 2b) was observed over the study sites (Mohal-Kullu: $R^2 = 0.693$; Darjeeling: $R^2 = 0.563$; Nainital: $R^2 = 0.671$), which might be due to water-soluble SOA formation [6,11,20,22]. Over the study regions, the influx of pollutants from the IGP region and the local sources could be the major contributing sources of the carbonaceous species (Figure 3).

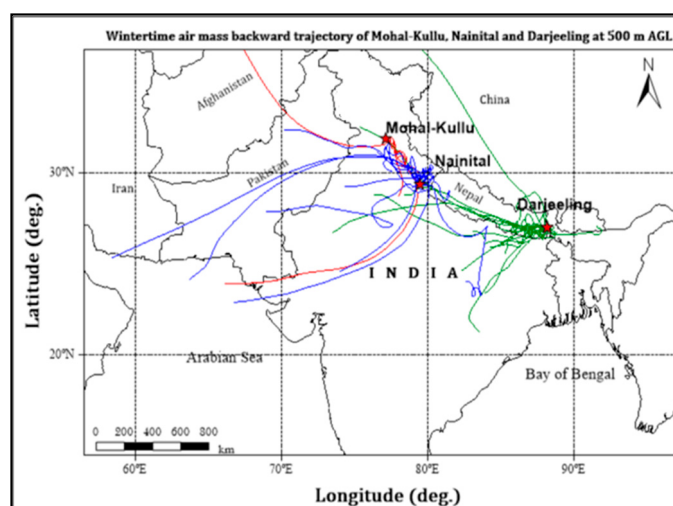


Figure 3. Wintertime air-mass backward trajectories of the study sites at 500 m AGL.

4. Conclusions

The CAs that are associated with PM_{10} have been studied during the winter season at different sites of the IHR, i.e., Mohal-Kullu, Nainital, and Darjeeling. The mean concentration of PM_{10} was $51 \pm 16 \mu\text{g m}^{-3}$, $38 \pm 9 \mu\text{g m}^{-3}$, and $52 \pm 18 \mu\text{g m}^{-3}$ at Mohal-Kullu,

Nainital, and Darjeeling, respectively. The major results of the study are summarized below as follows:

- In the present study, the winter concentrations of OC (Darjeeling: $5.36 \pm 1.74 \mu\text{g m}^{-3}$; Nainital: $2.85 \pm 0.75 \mu\text{g m}^{-3}$; Mohal-Kullu: $10.49 \pm 4.56 \mu\text{g m}^{-3}$), EC (Darjeeling: $2.67 \pm 0.87 \mu\text{g m}^{-3}$; Nainital: $1.30 \pm 0.46 \mu\text{g m}^{-3}$; Mohal-Kullu: $4.06 \pm 1.99 \mu\text{g m}^{-3}$), and WSOC (Darjeeling: $3.93 \pm 1.17 \mu\text{g m}^{-3}$; Nainital: $2.04 \pm 0.54 \mu\text{g m}^{-3}$; Mohal-Kullu: $5.23 \pm 1.35 \mu\text{g m}^{-3}$) were observed due to the enhanced contribution from wood burning for household purposes, open biomass burning, and stable atmospheric conditions.
- Overall, the diagnostic ratios of OC with EC, WSOC with OC, and SOC with OC showed their positive association with carbonaceous components and a major influence of BB as a source of carbonaceous species over the IHR. A linear regression analysis was performed among the carbon components OC, EC, WSOC, and SOC for more information on the sources of CAs. The OC and EC exhibited significant correlation throughout the study period, which can be attributed to the common nature of their sources;
- The long-range transported aerosols from the IGP region and the surrounding areas contribute to the carbonaceous species, along with local emissions.

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