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Research Article

Variability in TSPM BOUND EC/OC and BC with respect to ambient meteorological conditions in Srinagar J&K India

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ABSTRACT

More than a yearlong investigation (2008-2009) was carried in Srinagar, in the North-Western folds of Himalayas, involving the estimation of Total Suspended Particulate Matter (TSPM) and different carbonaceous fractions associated with it namely, Total carbon (TC), Elemental carbon (EC), Black Carbon (BC) Organic carbon (OC), no such direct estimates were available at that time for the valley and neither have they been updated till date. The samples collected on quartz filter using high volume sampler in a time series sequence and analyzed reflected marked seasonal dependence. The variability observed in carbonaceous contents of ambient aerosols was found to be similar as generally observed from other urban areas, however the relief along with prevalent meteorology was found to have a profound impact on the concentration of TSPM as well its constituent carbonaceous fractions. The higher values of TSPM were observed during summer months and the contribution of OC to TC as well. The contribution of EC to Tc was higher during colder months, EC arising from combustion sources attributed largely to petrogenic sources (fossil fuel) could be distinctly seen to be augmented by the biomass burning during autumn and winter; also increase in the OC content mainly starting in spring and continuing through to summer can be to attributed to active biogenic sources.

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INTRODUCTION

The carbonaceous aerosols (CA) as a part of ambient aerosols' chemical composition, represents the carbon in different forms. Determination of the Total carbon (TC), Black carbon (BC), Elemental carbon (EC), and Organic carbon (OC) present in the CA fraction of aerosols is done by using different methods (spectral, light scattering, thermal and chemical analysis). The reliable estimates of different carbon forms in ambient aerosols provide valuable information needed to appraise CAs impact on the surrounding environment regime to reflect upon the climate change concerns [1–3]. The contributions to CA having different forms (BC, EC, OC) can arise from primary or secondary sources and these sources may be natural or

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anthropogenic in character and activity. Emission of BC and EC mostly arise from an incomplete combustion of fuels (diesel, gasoline, CNG, biomass), while other carbon forms (OC) may also originate from biological sources (bacteria, pollen, fungi, plant waxes). Volatile Organic Carbon compounds (VOCs) initially emitted into the atmosphere in gaseous form is subsequently transformed into aerosol particles through their photo-oxidation in atmosphere, also becoming part of CAs, and they are designated as secondary Organic aerosols (SOA) [4–6].

To have a meaningful understanding about the temporal and spatial variability associated with the chemical composition of aerosols, it is imperative to keep in perspective the climate regime under which these samples are collected and analyzed [7–9]; as the prevalent climate variables would significantly have an impact on CAs concentration and compositional profile. To characterize the CAs present in the ambient environment, it is essential to have their reliable estimation (TC, BC/EC and OC) from different climate zones, where the investigation takes into account the region's meteorology. Quantitative estimation of TC, BC, OC, and EC contents associated with the aerosol load at different locations, in view of their ability to impact the climate, is an important scientific endeavor whereby the calculations can be done to determine the proportion of warming caused by their presence [10].Present understanding about carbonaceous aerosols (CA), based on the work done in different regions helps establish their role in affecting several atmospheric processes, *i.e.* influencing the radiative forcing; atmospheric chemistry; cloud condensation and associated precipitation[11–15]. Number of studies investigating CA has focused mainly on the presence of OC fraction to create an inventory of aerosol bound organic compounds [3, 16–20]. Few have looked at the presence of BC contents in aerosols from different regions [21, 22]. The established impact of carbon forms in aerosols also initiated research to understand the role of different carbon emitting sources (EC, OC or BC) [23–26]. The influence of the seasonal changes on the overall profile of carbon compounds present as a part of CA fraction was also investigated [27–32]. From Indian region few investigations have characterized aerosol associated carbon forms and their time dependent variability, but these investigations are limited mainly to Metropolises [29, 33]. Consequently, it necessitated to initiate a long-term investigation from this particular climatic zone of Himalaya, where no reliable information about CA was available till date.

The geophysical attributes of the Himalayas hold importance for the regional climate regime prevalent in the Northern part of the Indian sub-continent [34], and also is a vital source of fresh water to the ten largest rivers of Asia. The anticipated climate change related concern, and the role of particulate abundance to adversely affect the climate change regime in Himalayas has necessitated the need to have a reliable direct estimate about their (particulate) abundance, composition (BC, EC, TC, OC). So far little information about the characteristics of particulate matter from this Himalayan region exists [9]. The political uncertainty in this part has constrained the initiation of a systematic ground based long term investigation in this part.

MATERIAL AND METHOD

Sampling

Weekly Ambient aerosol samples collected from August 2008 till Dec 2009 on a Quartz fiber filter paper were analyzed to estimate aerosol load, and specific samples from the batch collected in 2009 (one from every month) were further analyzed for associated carbonaceous fractions (TC, EC, OC and BC). The sampling was done in Srinagar city (1524m ASL; 74°56′, 75°79′ E and 33°18′, 34°45′ N) located in Kashmir (15,520.3 km^2) valley surrounded by the Himalayas Figure 1. The region is endowed with temperate climate. The annual rainfall in this region is about 651 mm, temperature varies between 39° C to − 5° C [35]. The samples were collected using a High-Volume Sampler (HVS) kept on the roof top of a building located within a residential area. The sampling site was around 5km away from the main commercial center (Lal Chowk) in the middle of the city. The HVS was mounted on a flat surface ~ 9 meters above the ground. Prior to the start of the sampling, the filters were appropriately conditioned as per the established standard practice [36, 37]. The filter sheets were coded, weighed and stored in air tight containers before being transported to the sampling site. Post sampling the filters were stored in A4 size butter paper envelopes and covered with aluminum foil. All samples were stored under refrigerated condition till their analysis. Appropriate field blanks were simultaneously collected.

Aerosol Sample Analysis

Collected TSPM load on the filter matrix was weighed on a calibrated Sartorius CP224S microbalance having a precision to measure 0.1 mg. The estimation of TC, OC, and EC fractions present in the samples was done using a carbon analyzer Model 2001AThermal/Optical Carbon Analyzer (TOR/TOT) from Dessert Research Institute (DRI), Reno, NV USA. For the analysis a 0.5 cm^2sample punch was removed from the quartz filter sheets having TSPM sample and placed in a Quartz analyzer boat and loaded onto the instrument. The analysis followed Improve A protocol of EPA [38–40].

The measurement of BC content present in the samples was done by using a UV-Visible spectrometer equipped with an integrated sphere accessory capable of measuring surface reflectance and absorbance values [41–43]. A standard curve was generated from BC values reliably estimated from Optical Transmissometer OT-21– Magee Scientific Company Berkeley, California [44]. The procedure followed, in brief, involved loading of the stamped out 25mm discs from the filter sheet into the integrating sphere

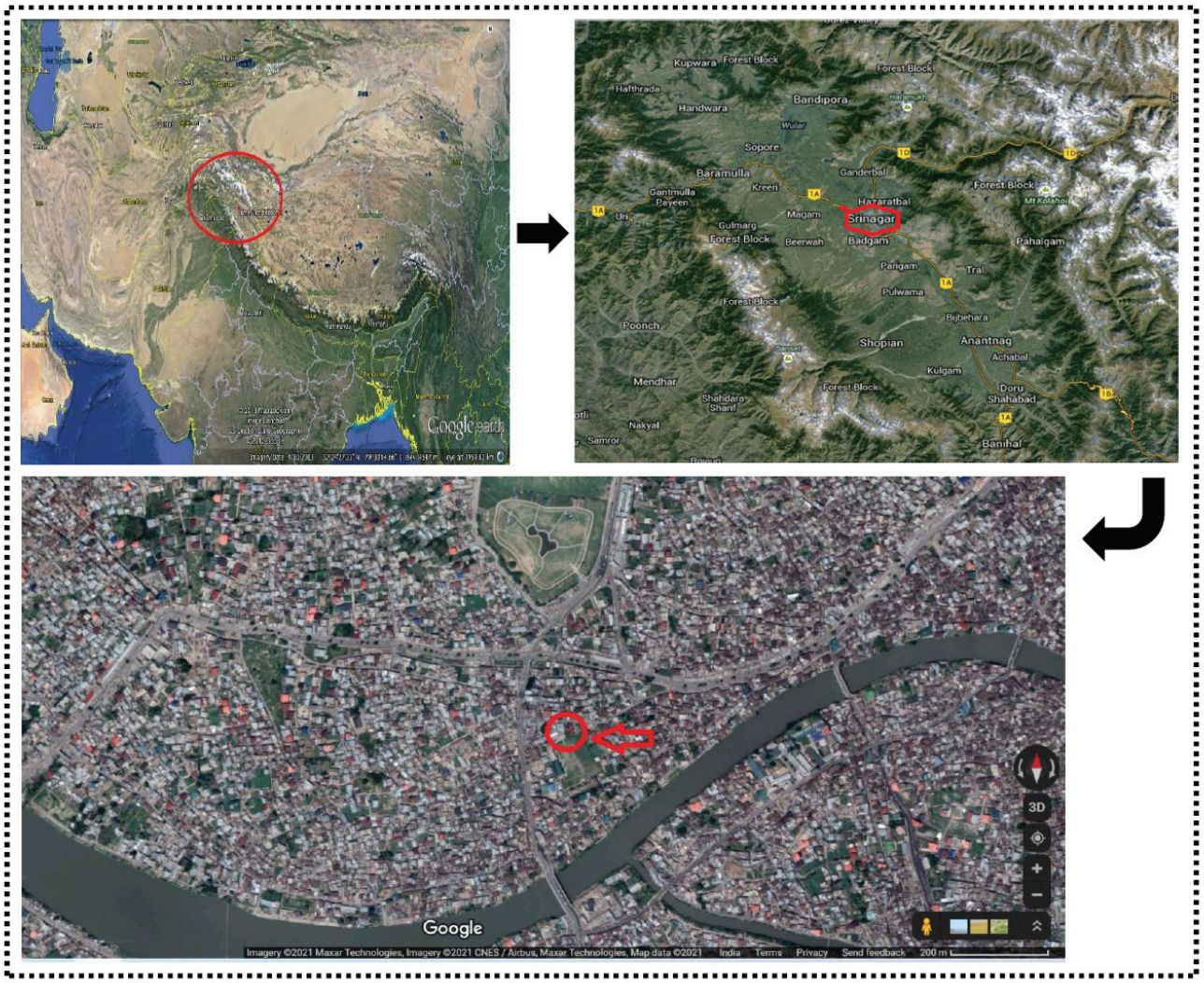

Figure 1. Study area and sampling location.

chamber of UV Spectrophotometer (UV-2450 Shimadzu) and recording the transmittance at 550 nm. The measured transmittance was converted to BC surface density μg/m². All measurements done on the collected samples were corrected with respect to the corresponding field blanks. The relevant data of meteorological parameters for the sampling duration was sourced from Air Resources Laboratory, National Oceanic and Atmospheric Administration <http://www.ready.noaa.gov/>. Additionally, the determination of the long-term trends in temperature of the region surrounding the sampling location was also done by subjecting the long-term time series of the ambient environment's temperature taken over 65 five years (1948-2012) to a time series analysis. The data was taken from NCEP/NCAR Reanalysis. The extraction of the trends present in the proxy temperature data was done by using Ensemble Empirical Mode Decomposition (EEMD) approach, a well-established mathematical tool developed initially by Huang et al. [45].

Meteorology of the Sampling Region

Relevant meteorology data for this investigation was procured from Air Resources Laboratory (http://ready.arl.noaa.gov/READYcmet.php), from the GDAS archive overview with resolution of 1°. Plots of average monthly variation of selected meteorological parameters [PBL (m), T (°C), Dp (°C), RH (%), WS (ms^{-1}) and PPT (mm)] were constructed (Supplementary Figure 1).

RESULTS

The onset of the increase in the warming can be seen to have started subsequent to the year 1965, since then the extent of warming in this region has been around 0.7°C Figure 2. Though at first look extent of warming does not

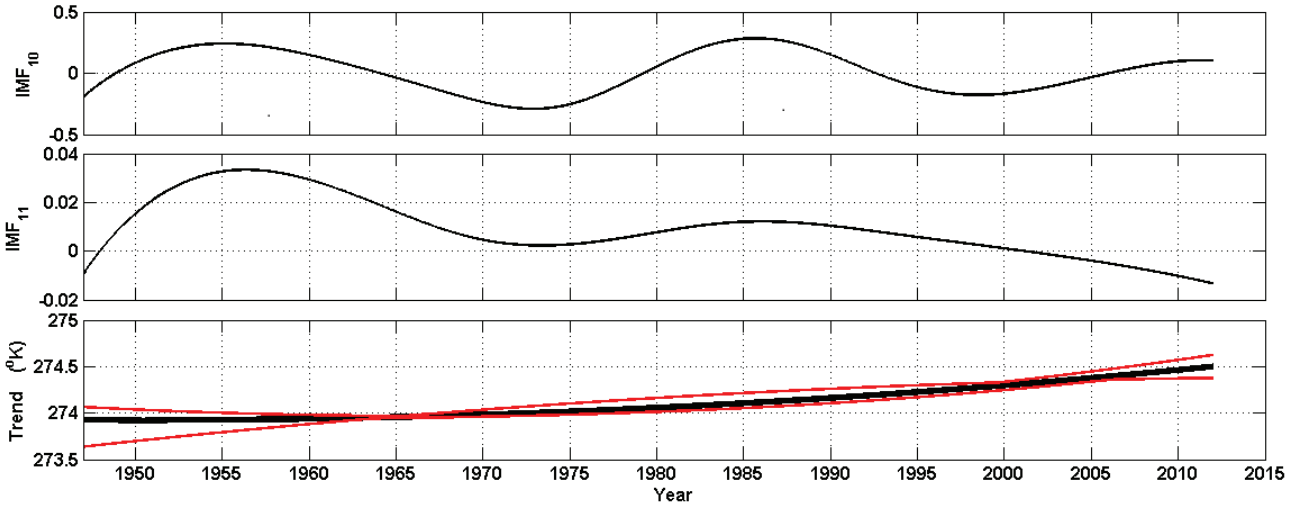

Figure 2. Extracted ensemble average trend in temperature over 65years (thick black line), two red lines surrounding the trend line represent the sensitivity region at 65% confidence.

seem alarming, however, taking into account the sensitivity of the region, especially the presence of alpine glaciers that are a source of water to some of the major rivers of the entire subcontinent, climate change induced warming can be very detrimental. Therefore, identifying its potential contributors becomes a really important endeavor.

The analysis of the observed variability in the aerosol load was looked at with respect to the changes in the ambient meteorological factors predominantly PBL to understand source modulations Figure 3; corresponding wind roses (Supplementary Figure 2) as well as wind back trajectories were also constructed for the sampling duration to identify their major contributing sources. Specific TSPM samples from year 2009 (mid-monthly) were analyzed for different associated carbon forms (TC, EC, BC, OC), OC and EC and corresponding results are given in Table 1, whereas the plotted OC values as a function of EC Figure 4 help elucidate the source variability between the two. Figure 5 presents the comparison of the thermal based measurements of EC with the Optical based BC estimates from the quartz samples collected during 2009.

DISCUSSION

Meteorology Impact on the Temporal Variability of the Ambient TSPM Profile

The TSPM load exhibits a distinct temporal variability augmented by the ambient meteorological conditions The monthly average TSPM concentrations ranged from the 169.6±109.4 μg/m^3 for August 2008 (minimum) to 633.6±101.1 μg/m^3 for November 2008, whereas the highest concentration for 2009 was observed in late spring 423±160.8 (Figure 3). In an attempt to explain TSPM variability in terms of the contributing sources, the loads were scaled with respect to the variability in the ventilation

Table 1. Variations displayed by TSPM, TC, OC, EC and their ratios for the select samples from year 2009

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
	Q1	Q2	Q3	Q4	Q5		Q6	Q7	Q8	Q9	Q10	Q11
TSPM (μgm^{-3})	116.2	286.2	296.5	603.2	317.2	-	311	422	213	296.5	237	211.5
BC (μgm^{-3})	3.64	3.15	2.34	4.88	2.62		2.70	2.57	2.64	2.89	2.73	2.54
TC (μgm^{-3})	31.1	44	35	50	33.4	-	37.6	33.1	39	55	50	43
OC[R] (μgm^{-3})	21.6	31.4	26	43	28.4	-	27.3	25	22.4	34.7	38	31
EC[R] (μgm^{-3})	9.5	12.6	9	7.3	5.0	-	10.3	8.3	16.4	20.4	11.8	12
OC/TC	0.7	0.7	0.7	0.9	0.9	-	0.7	0.8	0.6	0.6	0.8	0.7
EC/TC	0.3	0.3	0.3	0.2	0.2	-	0.3	0.3	0.4	0.4	0.2	0.3
OC/EC	2.3	2.5	2.9	5.8	5.7	-	2.7	3.0	1.4	1.7	3.2	2.6

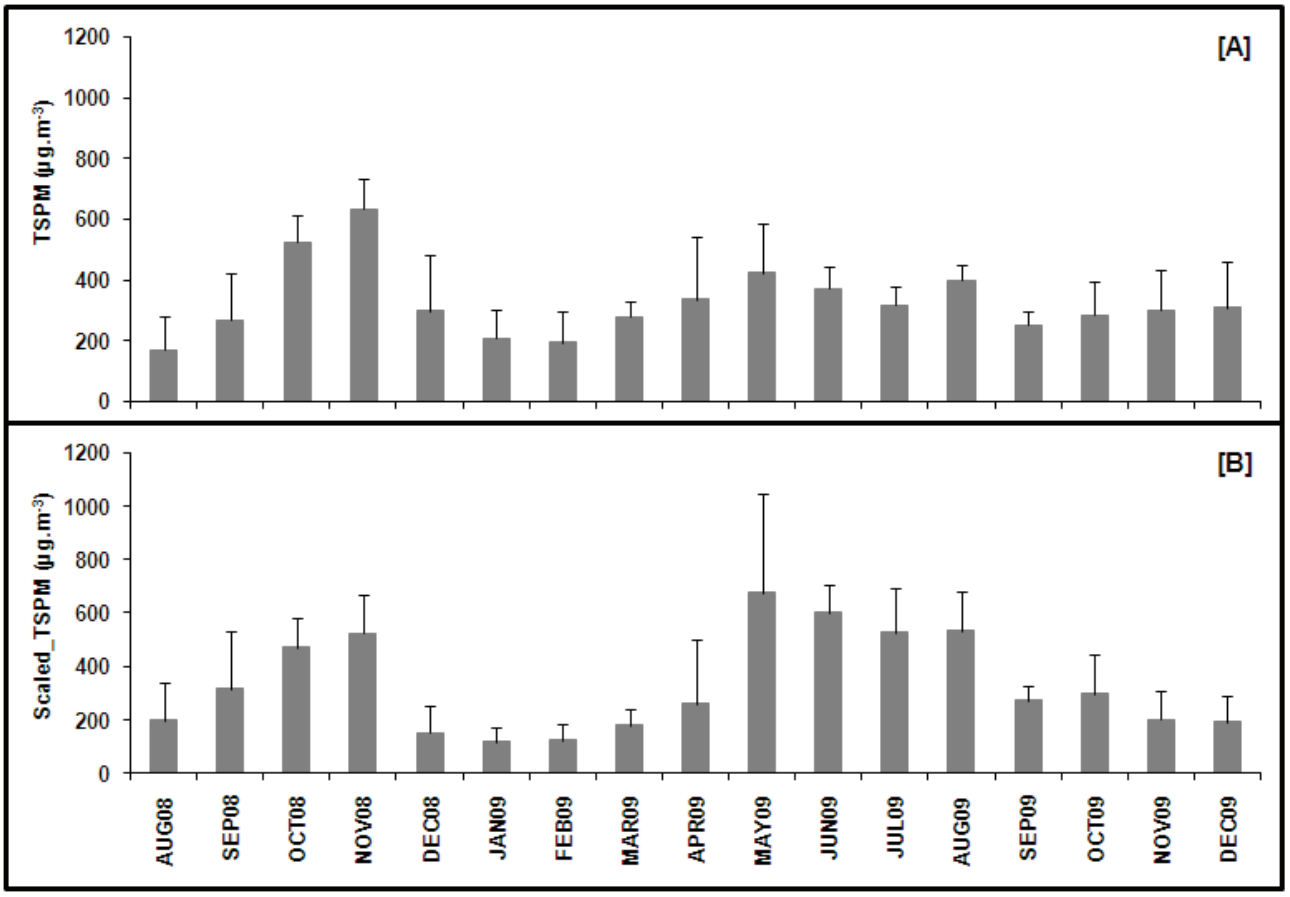

Figure 3. Histogram plots of monthly average values with standard deviation are given for TSPM load in panel [A], the Scaled TSPM values normalized to VCF ratio is shown in the panel [B] of the figure.

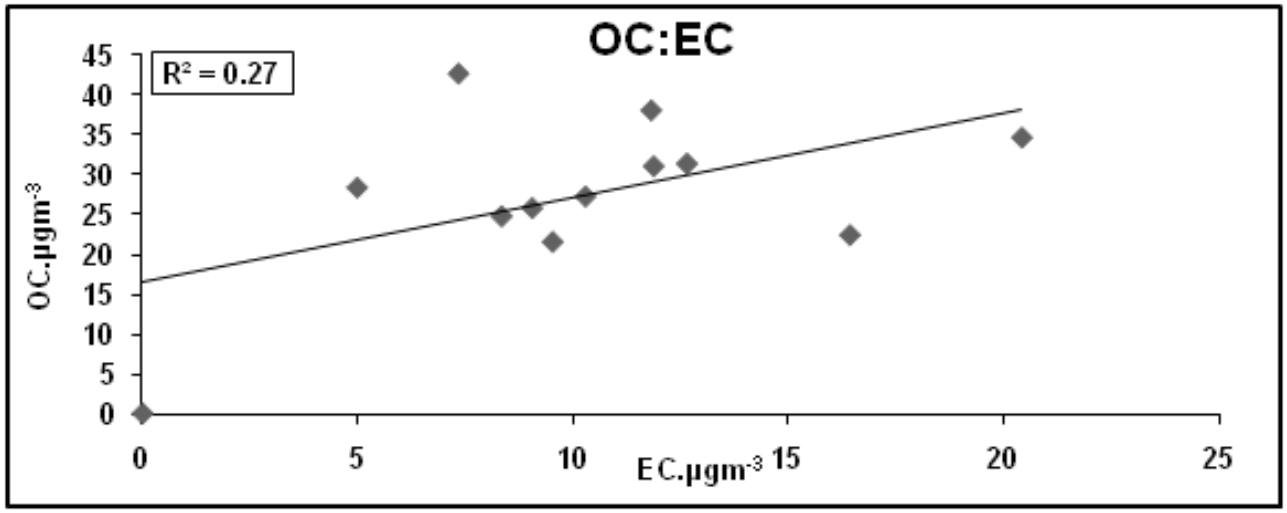

Figure 4. Variation in the OC/EC ratios observed for the samples (2009) collected on Quartz filter matrix values are reflectance (R) corrected.

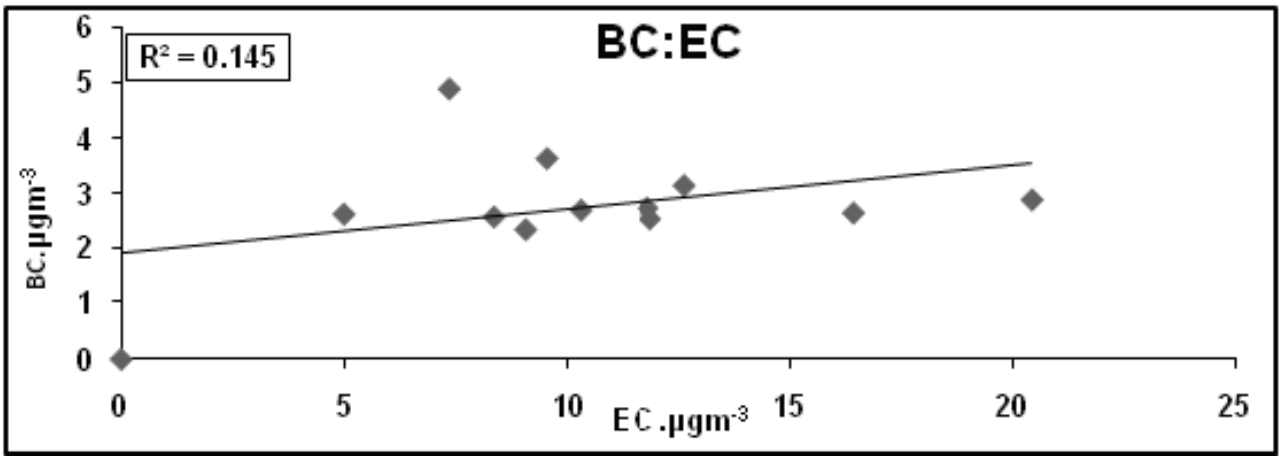

Figure 5. Comparison between Thermally estimated EC using DRI analyzer Employing improve A Protocol with the estimates of BC done by Using Optical transmissometer.

coefficient (VCF), which provides the modulations in mixing volume, for complete discussion refer [9]. Lower TSPM load– in August and December 2008, and January to April in 2009, can be attributed to the rain-wash out effect [46] as highest precipitation occurred during these months. The observed higher values of TSPM (scaled as well as normal), during summer months can be explained on account of the heightened biogenic activity increased re-suspension of the ground deposits and even transported particulates from the regions far removed from the sampling region [9, 47–49].

Temporal Profile of TC, OC, EC Fractions Associated with Ambient Aerosol Samples

In case of heavily loaded samples, (Table1) as is the case in present study, the transmission corrected values are susceptible to a bias, resulting in an uncertainty in the measured OC: EC split, consequently only the reflectance corrected values were considered [50, 51]. The value for OC was maximum in April 2009 (45.2 µg/m^3); minimum value was registered in January, 2009 (23.4 µg/m^3). The maximum EC concentrations, were obtained in October, 2009 (20.6 µg/m^3); and a minimum occurred in April 2009 (7.7 µg/m^3). The OC/EC ratios for the year 2009 are plotted in Figure 4; the ratio was maximum in April 2009 (5.8 µg/m^3) and minimum in September 2009 (1.43 µg/m^3) respectively.

Though the OC concentrations present in the samples, in the year 2009 was found to be minimum in January sample, but its contribution to the TSPM load was maximum (18.5 %), suggesting a selective washout of the coarser and hydrophilic components composing the ambient aerosol's matrix load [48]; this observation coincided with the high precipitation in the region. The washout would magnify the concentrations of the hydrophobic fraction of the aerosols; this aspect is also corroborated by the corresponding increase observed in the percentage proportion of elemental carbon present in the sample. The OC concentration in February were higher on account of the increased aerosol load, however its contribution to TSPM load was low, even though the overall OC source profile is expected to remain invariant. On the whole OC, EC and TC contents were higher in winter and autumn months. The highest OC concentrations corresponded with the high TSPM load, and this was recorded in the spring season (April) and can be explained on the basis of a spurt in the vegetation blooming and pollination after prolonged winter dormancy [52] this observation was also supported by higher OC: EC ratios. However, its (OC) contribution (%) to the ambient aerosol loads was minimum this is expected for the coarser aerosol size fractions [52, 53]; since both OC as well as TC contributions to the total aerosol load were low, it can be stated that the overwhelming proportion of the ambient aerosol load was predominantly of non-carbonaceous nature [52]. During summer the OC content was, on an average, lower than that found in the winter samples, but higher than that in the samples collected in spring. This suggests an increase of inputs from the active vegetative sources: correspondingly, the elemental carbon content also registered an increase in summer when compared with the values obtained in spring. These results again indicate that while the organic concentrations during summer season (July) tend to rise, their contribution to the total aerosol load is somewhat offset due to the higher elemental carbon inputs arising from the increased fossil fuel usage as well as higher contribution from coarse crustal input [54]. The OC and EC concentrations, both, declined during August, i.e., before they increased in September onwards to reach a higher value in autumn and early winter. It was interesting to note that during this period there was a pronounced increase in the EC concentrations relative to the OC concentrations as suggested by the lowering of OC/EC ratios, which alludes to an increased activity of the combustion-based source emissions (both petrogenic and biomass burning being prevalent in the valley), and their contributions to OC as well as

to EC [52, 55, 56]. Above stated explanation is supported by the plotted OC values as a function of EC (Figure 4); the extent of correlation (~0.5) between the two suggests that the contributions to EC and OC show a significant source deviation.

However, it is important to state that during autumn and early winter there was a higher correlation (0.8) between the two; which suggests that during this period they were mostly contributed by a common source. The average OC/EC ratio for the samples collected during 2009 was 3.1±1.4, a typical value for an urban area, suggesting an overall high fossil fuel combustion-based inputs [54].

Variability in Thermally Derived Elemental Carbon and Optically Determined Black Carbon

The average concentration of BC observed in this investigation, compared to what has been reported from other major urban locations of the country was much lower [57, 58] but they are way above what has been reported from other urban areas worldwide [59]. However, the observed concentrations were higher than that reported from the coastal and high-altitude mountainous region [60, 61].

Comparison of the thermal based measurements of EC with the Optical based BC estimates from the quartz samples collected during 2009 showed that the EC estimates were consistently higher than the estimated BC (Figure 5). BC, as a function of EC, shows a poor correlation (0.38); it is apparent that they are not in agreement and it is further suggested that deriving a BC value by fixing the attenuation coefficient (OT-21) might be significantly underestimating the BC. This becomes more crucial whenever there is a higher input to the aerosol matrix from the wood /biomass burning and the collected particulate matter is aged or highly mixed [52, 62]. Interestingly, the extent of deviation in EC: BC was seen to be more pronounced in the samples collected during the period (Sep-Oct) in which most of the post-harvest, pre-winter biomass burning takes place, in addition this is also the period during which most of the social celebratory events (weddings, etc.) take place in the valley; these events consume copious amount of wood for different activities (cooking, heating, etc.). This part of the year is also characterized by large scale biomass burning for charcoal production in preparation of long winters ahead.

CONCLUSIONS

Temporal profiling of carbonaceous fraction present within the ambient TSPM collected from Kashmir valley, which has unique climate regime and geophysical setting, was done. The bulk characterization of the Total Carbonaceous content (OC, EC, and BC) components presents in the collected ambient aerosol load revealed predominance of anthropogenic inputs, although significant but lesser proportion can be attributed to biogenic inputs. The average BC values present were indicative of the semi-urban to urban character of the study area. Both EC and OC contents of associated with the samples peaked during winter and autumn, which indicated that the in addition to high petrogenic input, biomass burning in this period could also a significant contributing source. The observed OC: EC ratios again reinforced the importance of the combustion-based emissions, which contributes to the ambient aerosol load in this region. The observed decline in carbonaceous content (both OC and EC) in spring indicated higher inputs of coarser fraction (crustal sources) into the ambient aerosols, whereas finer carbonaceous fraction (combustion sources) contributed more to the load during winter months. Slight increase in the OC fraction present in the summer month's samples suggested a sustained input from biogenic-sources. The marked deviation in EC and BC values reinforces the common observation that BC estimations are better source indicators when reported in real time as pose to sampled values.

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AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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