

Estimation of Carbonaceous Emission Impact on Urban Soil-Dust in Delhi

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Abstract: It is well established that mineral dust is responsible for high levels of particulates in the air over North India. Mixing of dust with anthropogenic air pollutants influences the optical properties of the atmosphere affecting the visibility, biogeochemistry, local climate and air quality. Strong interaction of carbonaceous aerosols with mineral dust further facilitates such phenomenon. Carbonaceous components such as organic carbon (OC) and elemental carbon (EC) play significant role in regional and global climate by altering the radiation budget of the surface, whereas carbonate carbon (CC) helps in buffering the atmospheric acidity. In order to investigate such interactions, this paper presents the concentrations of different carbonaceous components in atmospheric dust and soil at three sites in the National Capital Region of Delhi. Results showed that high levels of carbonaceous components and their spatial distribution had close correspondence with local anthropogenic activities such as vehicular emission, biomass burning, crustal components from exposed land surfaces, construction activities, etc. High OC and EC enrichment in dust as compared to soil have suggested their abundance in urban dust due to the significant interaction of atmospheric dust with urban anthropogenic emissions of EC and OC. The estimation of carbonaceous fractions in soil and dust filter samples had high accuracy due to matrix match standard was developed to quantify the content. Recovery of 98% was achieved for a synthetic mixture of charcoal and carbonate representing the matrix of real samples. The study demonstrated that abundance of carbonaceous components in atmospheric dust is an effective indicator of pollution from increased human activities in urban areas.

Keywords: Urban dust, Synthetic standard, OC, EC, Carbonate carbon.

Introduction

Increasing global concern of the atmospheric carbon contents has resulted in intensive research on carbonaceous aerosols i.e. Organic carbon (OC), Elemental carbon (EC) and Carbonate carbon (CC). They all are becoming one of the ubiquitous issues of environment. Carbonaceous aerosols, especially OC and EC, are one of the most important factors affecting earth's radiation budget (IPCC, 2014). This is evidently confirmed by its growing significance in the atmospheric processes of biogeochemical cycle, hydrological cycle,

local as well as global climate change and air quality (Jacobson, 2001, 2002; Menon et al., 2002). However, an abundance of mineral dust over the tropical region has remarkably modified its role in the atmospheric processes. Continuous entrainment of mineral dust in the atmosphere has led to an increasing interaction of urban pollutants with the atmospheric components, especially over the South Asian region (Rodriguez et al., 2012). These interactions have been commonly observed to occur in a two-stage process. During the first stage, when the concerned pollutant and the dust come into physical contact, external mixing is initiated

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which, over a period of time, leads to the second stage. The latter is marked by a transformation in the chemical composition of dust itself through internal mixing (Raes et al., 200; Baker and Croot, 2010).

Carbonaceous aerosols play a very important role in global and regional climate change (Haywood and Shine, 1995; Penner et al., 1998; Jacobson, 2001; Chung and Seinfeld, 2002; Roeckner et al., 2006). OC along with sulfate and nitrate aerosols are primarily responsible for the scattering of solar radiation in the atmosphere and thus causes negative radiative forcing as opposed to the positive radiative forcing of greenhouse gases, whereas BC or EC is mainly responsible for the absorption of solar radiation which heats the upper atmosphere and concomitantly cools the surface. Due to very high absorption efficiency of EC in the atmosphere, it is considered as the second most important factor responsible for global warming after CO₂ (Ramanathan and Carmichael, 2008).

Regions with high solar radiation and elevated EC concentrations are responsible for the so called Brown Clouds covering large areas of Asia. Surface dimming led by Brown Clouds over Asian regions resulted in the alteration of hydrological cycle and thus affecting monsoon (Ramanathan and Carmichael, 2008). IPCC (2007) pointed that EC deposition on snow substantially alter the total radiative forcing by the absorption of solar radiation on snow surface, which ultimately results in the melting of glaciers (Forster et al., 2007; Yttri et al., 2009). Many sources emit EC as soot particles which contain up to 50% organic matter and on mixing with sulfate particles by agglomeration; they form internally mixed soot-sulfate particles. Organic matter significantly enhances the absorption capacity of EC by a factor of two to four (Fuller et al., 1999; Jacobson, 2001; Roeckner et al., 2006). OC also acts as cloud condensation nuclei (CCN) due to its hygroscopic growth in the atmosphere and thus play an important role in cloud droplet formation (Gelenscer, 2004).

Increasing emissions of these carbonaceous aerosols into the atmosphere from various anthropogenic activities such as burning of fossil fuels and biofuels for industrial, vehicular and domestic purposes have aggravated the local climatic pattern in the form of erratic rainfall patterns, drought, deterioration of air quality and consequent health impacts (IPCC, 2007). Resuspension of the soil-derived dust and bioaerosols are considered as an important natural source of OC in the atmosphere, whereas black carbon or EC is only emitted from combustion processes (Gelenscer, 2004). Inorganic carbon or CC mostly comes from mineral

dust, road dusts and construction activities. Thus, CC is becoming an important atmospheric component over the tropics owing to the abundance of mineral dust from dry lands (Tegen and Fung, 1995).

In Indian region, acidity caused by SO₂ and NO₂ over the semi arid tracts of the Indo Gangetic regions is significantly buffered by the alkaline nature of atmospheric dust (Kulshrestha et al., 2003; Ho et al., 2003). Increasing urban sprawl and their growing energy consumption in a Delhi megacity, on the other hand, has remarkably increased the ambient concentration of carbonaceous components in the atmosphere (Singh et al., 2005; Tiwari et al., 2013). Such increased emissions of these carbonaceous components in the atmosphere have ultimately led to its higher deposition into the soils and sediments, which on resuspensions with soil dust and road dust cause the degradation of local air quality. As a consequence, the quality of urban air has been adversely affected resulting in severe health implications.

The present work is a modest effort made to make a meaningful contribution to the existing knowledge on this facet. This work mainly focuses on estimating the concentration of carbonaceous components (OC, EC and CC) in coarse dust at three different sites and their respective local soil, which is quite an untouched aspect in Indian context. The site selection has been made keeping in mind the varying land use patterns and anthropogenic activities over the study area. Enrichment of carbonaceous components in dust with respect to soil was also analyzed for the purpose of assessing and ascertaining the level of pollution.

Methodology

Sampling Sites

National Capital Region (NCR) of Delhi is located at 28° 36' N, 77° 13' E, ~ 240 m above mean sea level and considered as one of the most polluted cities in the world (Figure 1). Delhi ranks second in population, having population density 11,297 per km² with land area 1,484 km². Considering the large diversity in land use patterns and human activities three sites were selected (Table 1).

Collection of Dust and Soil Samples

For the collection of free fall dust, 47 mm Pallflex Tissuequartz (2500QAT-UP) microfibre filters were used, which was mounted on 60 mm petridish and placed about 15 feet above the ground at all the three sites (Saxena et al., 1992). To prevent the blowing of

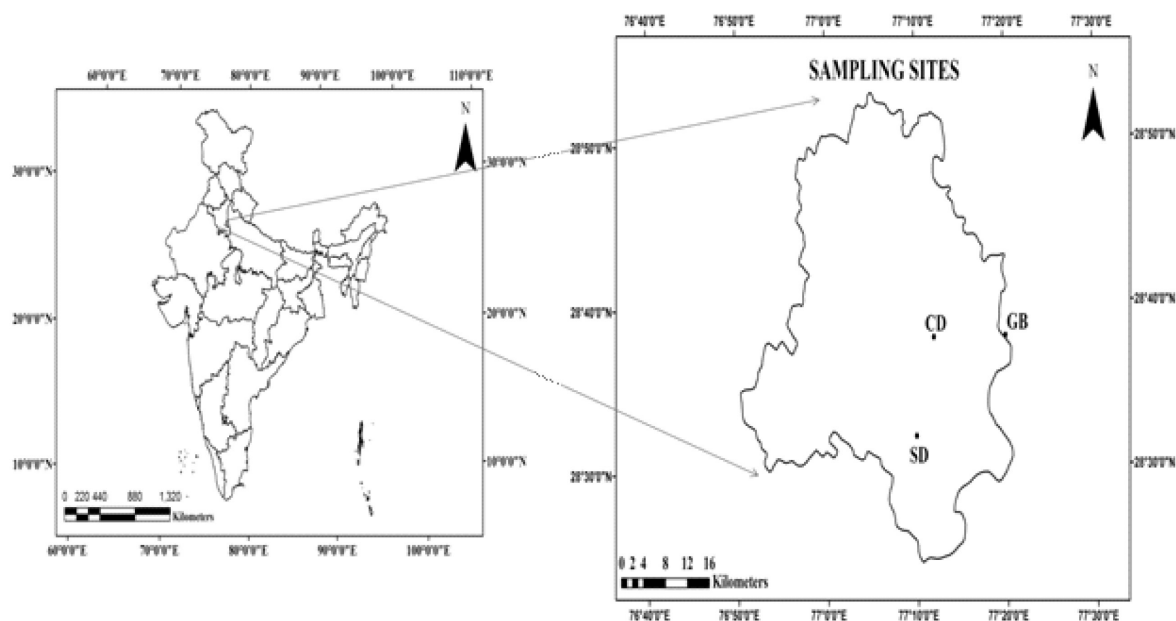


Figure 1: Sampling sites in Delhi, NCR.

Table 1: Description of three sampling sites in Delhi, NCR

No.	Sites	Landuse type	Population density	Abbreviations
1	Jawaharlal Nehru University (South Delhi) (28°32' N, 77°10' E)	Residential/institutional	11060 per km ²	SD
2	Connaught Place (Central Delhi) (28°38' N, 77°12' E)	Commercial	27730 per km ²	CD
3	Vaishali (Ghaziabad) (28°38' N, 77°20' E)	Residential	2400 per km ²	GB

sample filters, a rubber washer was implanted onto the filters and the petridishes were kept onto the wooden holder. For dust collection, petridishes were kept in a little shady platform of buildings to avoid the damage of quartz filters from external factors such as rain, birds, etc. Samples were left exposed for the time interval of 15 days. A total of 28 samples (11, 9 and 8 samples from SD, CD and GB, respectively) were collected in the duration of six months between June and November 2014 excluding discarded and damaged samples (two from CP and three from GB). The collected samples were stored at room temperature until analysis. Filters were weighed with 0.00001 g sensitivity microbalance (Mettler Toledo AD265-S/FACT) before and after sampling to get dust deposition on the filter. One field blank and one filter blank were also analyzed in order to avoid the over or underestimation during analysis. The dustfall flux was calculated with the help of difference in the initial and final weights of each sample.

Soil samples were also collected for the measurement of various carbonaceous components from non-perturbed area. After removal of 5 cm upper layer, soil was collected in polythene bags. Visible impurities were carefully handpicked from the samples. Dried soil samples were sieved with 200 mesh to get smooth and homogenous soil. Soil samples were collected from three different locations at each of the sites from least disturbed crustal surfaces.

Soil Sample Preparation

To analyze the carbonaceous components (CC, OC and EC) in soil through thermal/optical method, it is necessary to disperse the soil on the quartz filter which can be placed in the sample port of the analyzer. For the homogenous dispersion of soil samples onto the quartz filters, a closed glass chamber was used. The procedure of soil dispersion is illustrated in Figure 2. The procedure was followed with keeping in mind that

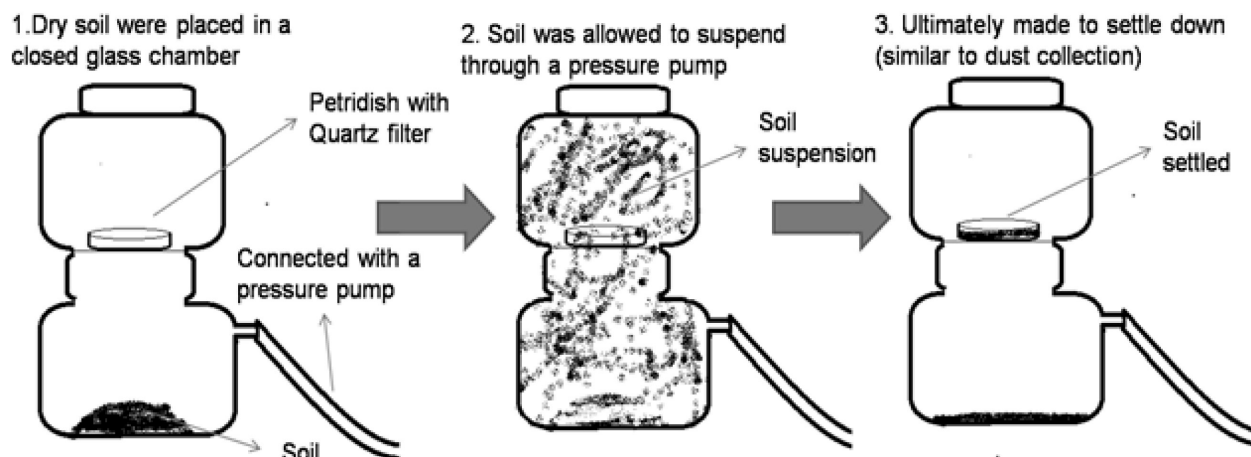


Figure 2: Preparation of soil samples on quartz filters.

it should be more or less representing the dust settling on a surface. Soil sample filters were weighed before and after the procedure to get the accurate soil loading (in grams).

Carbon Analysis

The OC, EC and CC fractions were determined in the dust samples by using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Analyzer. Thermal/Optical Reflectance (TOR) method was selected for the determination of carbonaceous fractions using acid pre-treatment followed by IMPROVE-A (Interagency Monitoring of PROtected Visual Environments) protocol (Han et al., 2007, 2009a; Chow et al., 2007) which was completed through the following two major steps.

Step 1: Determination of CC

For the analysis of CC, 0.4N HCl was used as a neutralizing acid of carbonate in samples. A filter punch of 0.5 cm² area was cut from the sample which was placed in the analyzer. After passing He gas at 40 cm³/min for 90 seconds, 25 µl of HCl was injected onto the filter piece with a micro syringe. Due to reaction with HCl, CO₂ was released from the carbonate, which was measured as CH₄ same as in IMPROVE-A carbon analysis (Chow et al., 2008). The field blank and filter blank were also analyzed to remove artifacts.

Step 2: Determination of OC and EC

After the analysis of CC, the sample was left for 900 seconds within the analyzer which allowed the evaporation of all the moisture from the sample piece by passing pure He gas. Four fractions of OC (OC1, OC2, OC3 and OC4 at 120°C, 240°C, 480°C and 580°C, respectively in pure He atmosphere) and three

EC fractions (EC1, EC2 and EC3 at 580°C, 740°C and 840°C respectively in 98% He and 2% O₂ mixed gas) were produced. Pyrolyzed Organic Carbon (OP) fraction was obtained when reflected laser light reaches its initial value after injection of oxygen to the analysis environment (Chow et al., 2008). Thus, following formula was used for the determination of OC, EC and TC.

$$OC = OC1 + OC2 + OC3 + OC4 + OP$$

$$EC = EC1 + EC2 + EC3 - OP$$

$$\text{Total Carbon (TC)} = OC + EC + CC$$

In this regard, Quality Assurance/Quality Control procedure has been described by Cao et al. (2003). One field blank and one filter blank were analyzed to avoid overestimation in the analysis. Replicate samples were also analyzed to find the precision of dust and soil samples through the analyzer.

Preparation of Artificial Known Samples

In order to validate the method of soil sample preparation, matrix matched three artificial known samples were prepared to analyze different carbonaceous fractions. Small amount of charcoal was suspended in a closed glass chamber through a pressure pump to produce its dust, which was allowed to settle on a pre-weight quartz filter. The amount of such dust deposited on the filter was determined by calculating the difference in the weight of the filter. This was treated as artificial known samples. Similarly, the known samples of pure CaCO₃ and a mixture of charcoal + CaCO₃ in 4:1 ratio were prepared. Analysis results of these three samples are given in Table 2.

Carbon Analysis of Replicate Samples

Two sets of blank filters and dust samples were analyzed in replicates by the Thermal Optical analyzer using the IMPROVE_A TOR method. CC showed a precision of 94% with an artifact of <6%. After acid pre-treatment, i.e. after the removal CC from the sample punch, OC and EC were analyzed. This showed a precision of the order of 8 to 10% for blank filters and 8 to 11% for dust samples. Such difference between replicates could be attributed to the very high absorption capacity of the quartz filter for gases and volatile compounds (Weingartner et al., 2003). Besides this, handling process and small uneven distribution of particles on the filter surface could also be implicated for the differences occurring between the replicates of the dust samples. Chow et al. (1993, 2005) have reported the precision for analyzer for atmospheric samples of the order of 5-10%. Han et al. (2007) have also reported the difference of replicate samples of OC and EC between 3 and 13%.

Results and Discussion

Concentration of Carbonaceous Components in Dust and Soil Samples

Concentrations of different carbonaceous fractions (OC, EC and CC) along with total carbon concentration (TC) at three sites in NCR are summarized in Figure 3.

Dust Samples

Concentrations of different carbonaceous fractions in urban dust samples are summarized in Table 3. Mean concentration of OC, EC and CC in dust at SD site were estimated as 64.8 mg/g, 5.7 mg/g and 5.5 mg/g, respectively. At CD site, their concentrations were recorded to be 66.5 mg/g, 6.8 mg/g and 6.8 mg/g, respectively, however, at GB site as 38.5 mg/g, 3.3 mg/g and 4.6 mg/g, respectively. Being a major commercial hub of NCR, CD site showed the highest fractions of OC and EC owing to the increased fossil fuel combustions from its heavy traffic density. Interferences

Table 2: Analysis of three artificial known samples

	<i>Taken (mg)</i>	<i>Found (mg)</i>	<i>% Recovery</i>
Charcoal	4.83	4.27	88%
CaCO ₃	0.09	0.08	89%
Charcoal + CaCO ₃ (4:1)	0.56	0.55	98%

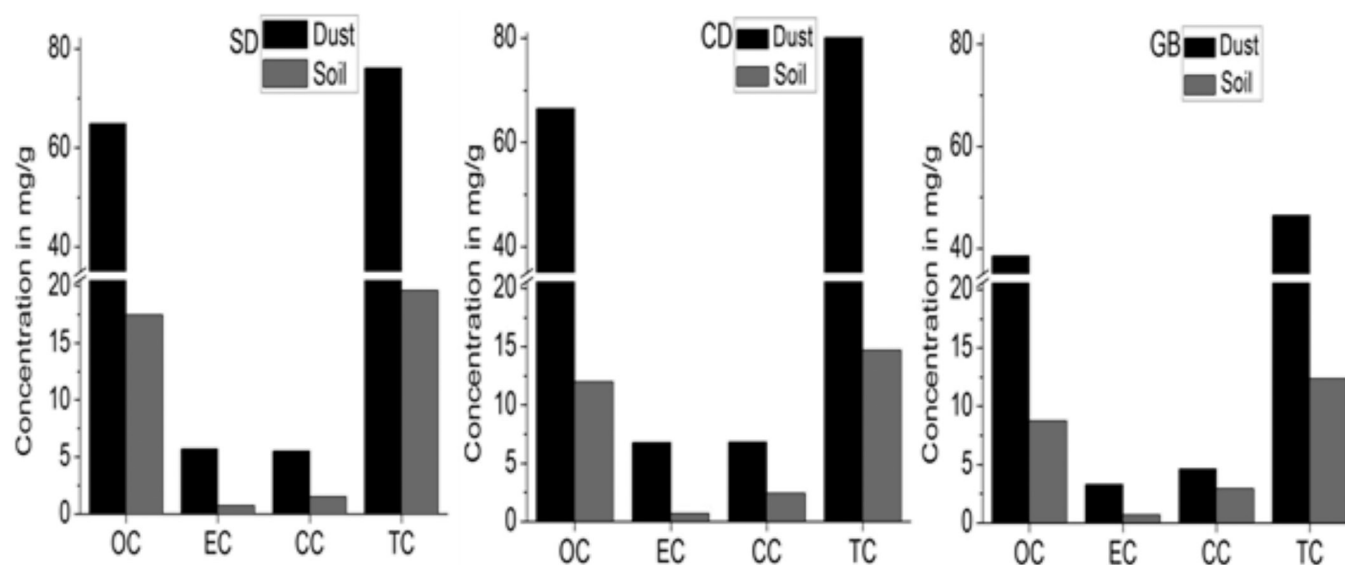


Figure 3: Concentrations of carbonaceous fractions (OC, EC, CC and TC) in atmospheric dusts at three sites and their respective soils.

Table 3: Concentrations of carbonaceous fractions (mg/g) in dust samples at three sites in NCR

Carbon fractions	SD (n = 11)		CD (n = 10)		GB (n = 10)	
	Mean	Range	Mean	Range	Mean	Range
OC	64.8±12.5	26.0-180.8	66.5±11.1	39.2-147.4	38.5±4.3	19.6-54.2
EC	5.7±0.9	0.7-9.1	6.8±1.1	2.3-13.0	3.3±0.7	0.7-6.4
CC	5.5±0.7	2.6-10.6	6.8±0.8	3.8-10.7	4.6±0.4	2.0-5.7
TC	76.0±12.8	36.1- 192.6	80.1±11.8	46.1-162.0	46.5±4.8	24.7-65.0

from the crustal sources, on the other hand, resulted in a higher fraction of CC over EC at GB site which has been characterized by its extensive deforestation and construction activities. Such results of OC and EC were also found to be comparable to other studies (Han et al., 2007, 2009b).

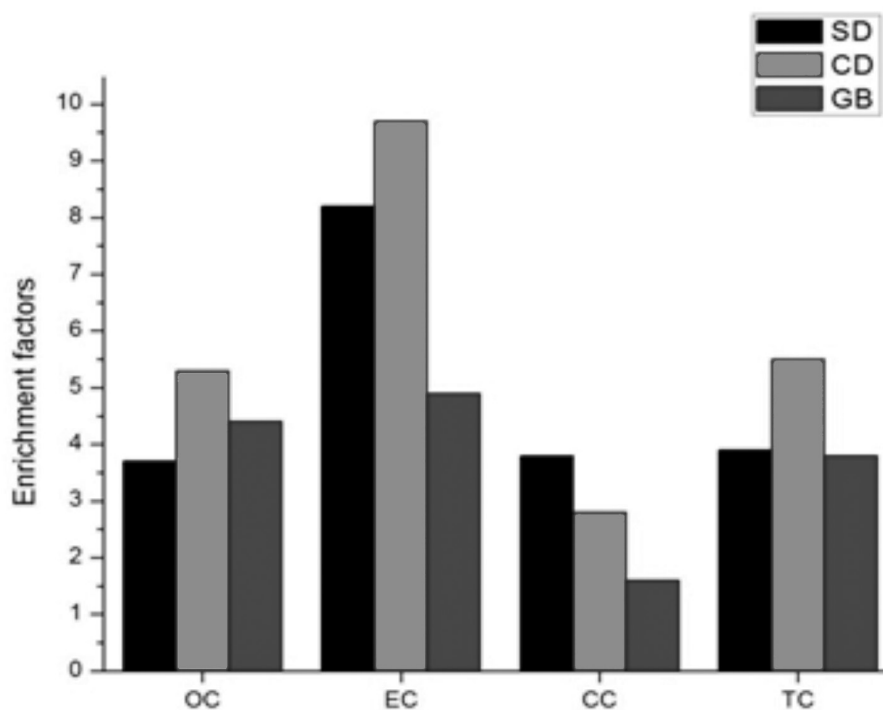
Soil Samples

Average concentration of OC, EC and CC of soil samples at SD site were 17.4 mg/g, 0.7 mg/g and 1.5 mg/g, respectively (Figure 3). Their mean concentrations at CD site were observed to be 12.0 mg/g, 0.7 mg/g and 2.4 mg/g, respectively, whereas their concentrations at GB site were 8.8 mg/g, 0.7 mg/g and 2.9 mg/g, respectively. Such results of carbonaceous fractions in soil samples of three sites were found to be in close correspondence with their respective dust samples. Highest fraction of OC at SD site could be attributed to its high vegetation cover resulting in a high level of

organic matter to its local soil. EC, on the other hand, showed the lowest contribution to the carbon fraction at all the sites. However, it was found to be comparable to Xi'an soil where it ranged from 0.3 to 2.11 mg/g and country park soils of urban (4.8 mg/g) and non-urban (0.3 mg/g) areas (Ho et al., 2003; Han et al., 2009b). Higher concentration of CC in soil samples at all the sites suggested towards the alkaline nature of Indian soil (Khemani et al., 1985; Kulshrestha et al., 2003). Overall, the concentration of TC was observed to be 19.6 mg/g, 15.1 mg/g and 12.4 mg/g at SD, CD and GB sites, respectively.

Enrichment Factor

In order to assess the anthropogenic influence of atmospheric dust, Enrichment Factor (EF) was calculated for each site as shown in Figure 4. It can be defined as the concentration of carbonaceous fractions in

**Figure 4: Enrichment factor of OC, EC, CC and TC of atmospheric dusts of three sites in NCR.**

atmospheric dust divided by their concentration in soil representing its background levels (Han et al., 2009b). Highest EF values were reported for EC fraction and lowest for CC fraction. Interaction of dust with traffic emissions has resulted in the very high EF value of EC fraction (9.7) at CD site whereas it was lowest at GB site (4.9). Han et al. (2009b) have reported the enrichment of road dust in Xi'an of an order of 12.4 suggesting great influence of non-natural sources. EF values of OC fraction, on the other hand, were reported to be the highest at CD (5.5), again showing the high influence of traffic emissions. Lowest EF value for OC was found at SD site (3.7). This could be attributed to the higher content of organic matter leading to high OC fraction in dust contributed from the soil resuspension at SD site. Therefore, high enrichment of OC and EC in dusts suggested towards the greater human influence from various anthropogenic activities such as burning of fossil fuel and biofuels.

Owing to the alkaline nature of Indian soils, EF values of CC were observed to be lowest amongst the carbon fractions (Khemani et al., 1985; Kulshrestha et al., 2003). EF values for CC were found to be 3.7, 2.8 and 1.6 at SD, CD and GB sites, respectively. Lowest enrichment of CC at GB sites primarily due to windblown soil dust due to increasing deforestation and construction activities in the area. Higher enrichment of CC at other sites can also be attributed to long range transport of mineral dusts from North-western desert (Thar) of India.

Relative Contributions of Carbonaceous Fractions in TC in Dusts and Soil

The relative contribution of different carbon fractions (OC, EC and CC) in total carbon (TC) in dust and soil of three sites is shown in Figure 5. It is clear from the figures that OC contributes more than three quarters of TC in both dust as well as the soil across all the sites except for GB soil, where OC contributes 71% of TC. At each site EC percentage in dust samples were higher than soil samples, indicating EC contribution from urban emissions. However, percentage CC was found to be higher in soil than urban dust at each site, suggesting the higher contribution of CC in the total carbon content of soil. At GB site, CC fraction was found to be about one quarter of TC, representing high loading of crustal components in GB soil which is also a primary source of CC in the atmosphere caused by soil resuspension. Land use and land cover changes highly affect the carbonaceous fraction loading in urban dust along with other anthropogenic activities (Kumar et al., 2014).

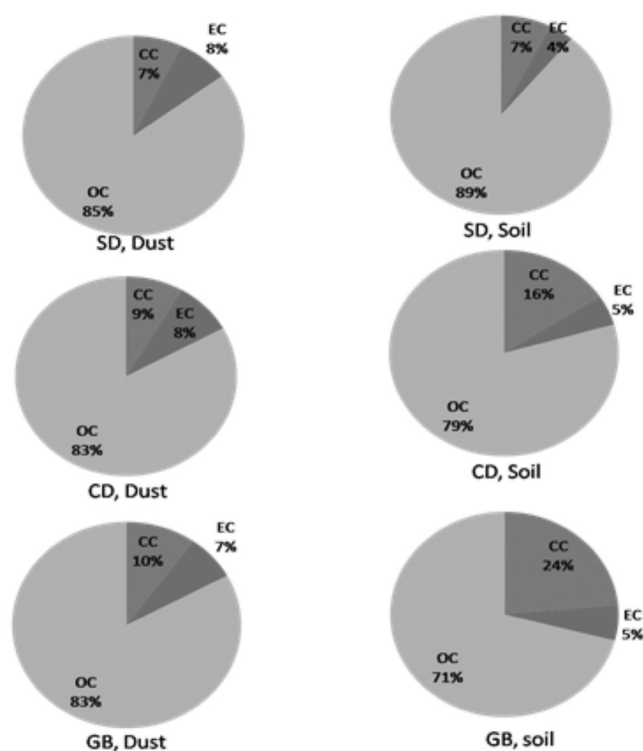


Figure 5: Relative contribution of OC, EC and CC in urban dusts and soil of three sites.

Conclusions

The method developed for the estimation of EC, OC and CC in soil and dustfall is highly reliable showing recovery of 98% for a sample using synthetic mixture of known quantities of charcoal and carbonate. The method provided results with high accuracy due to matching matrix of the samples and synthetic standards. The inference drawn from the findings of the paper indicates that the enrichment of carbonaceous components in the atmospheric dust is significant. So far as the abundance of the different constituents of carbon (OC, EC and CC) is concerned, it is underscored that the EC enrichment in urban dust is found to be disproportionately high as compared to other constituents viz. OC and CC across all the sites. Since all the sites are situated within an urban perimeter, the high levels of EC are noteworthy which has consequently severe health implications. High levels of EC emission also have significant impact on regional climate, radiation budget, rainfall patterns etc. Greater vehicular and industrial emissions are the hallmarks of urban centres, which is shown by the multifold enrichment of dust with EC and OC as compared to soil. Very high concentrations of EC and OC at CD site is suggestive of the significant influence

of traffic emissions of urban dust. CC enrichment with anthropogenic emissions was found to be lowest among all constituents suggesting their contribution mostly from natural sources. The findings of the study are suggestive of the high influence of urban anthropogenic activities to the atmospheric dust. The study is very useful in measuring the regional and global carbon budget.

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