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Number and mass of ultrafine, fine and coarse atmospheric particles during different seasons at Agra, India

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ABSTRACT:

In the urban atmosphere, adverse health effects of airborne particles cause great concern. Some toxicological studies show that ultrafine particles exert a much stronger physiological effect than the same mass of coarse particles due to an increase both in the number and relative surface area as compared to particles of larger size. The present study deals with the particle number, mass, volume and surface area - size distribution of atmospheric particulate matter to determine their relative proportions in the Ultrafine (< 500 nm), Fine (< 2.5µm) and Coarse (< 10 µm) particles at Agra. The particles collected were analyzed for their mass and number simultaneously during winter, summer and monsoon seasons using an optical particle counter Grimm monitor, 31-Channel Portable Aerosol Spectrometer model No: 1.109 in the range of 0.25-32 µm. The results indicated that the average number concentration was highest in summer (286399.1 cm^{-3}) followed by winter (109155.7 cm^{-3}) and monsoon season (67390.68 cm^{-3}). The concentrations were 2.63 and 4.24 times higher during winter and monsoon season than summer months. Higher concentration was attributed to local as well as long range transport of particles. The long range transport of aerosol particles is also supported by back trajectory analysis. The average number concentration of coarse particles was 2.41 times higher in summer season which indicate that dust storms during summer period have major proportion of coarse particles. The wash out effect during monsoon causes significant decrease of particles therefore the concentrations were found to be much lower.

Keywords:
Atmospheric aerosols, number concentration, mass concentration, EM/EDX.

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INTRODUCTION

Ambient particulate matter has attracted major public health and atmospheric science concern in the recent years. Especially, the size distribution of atmospheric aerosols, together with their composition, sources, and sinks, is a key element in understanding and managing aerosol effects on health (Harrison et al., 1999; Wichmann and Peters, 2000), visibility (Ramanathan et al., 2001) and climate (Seinfeld and Pandis, 1998). Because of the diverse sources, these particles have different morphologies and chemical compositions (Salma et al., 2001). There are number of properties of particles that are important for their role in atmospheric processes. These include in addition to their number concentration, mass, size, chemical composition and aerodynamic and optical properties. The determination of the origin of aerosols is often based on their morphological, mineralogical and chemical characteristics. For this reason, several multidisciplinary analytical techniques have been widely used for individual particle analysis (Esbert et al., 2001). One of the key tools selected for detailed investigation of the airborne particles is electron microscopy. Scanning electron microscopy with energy- dispersed analysis of X - rays (SEM/EDX) have already proven to provide information on the morphology, elemental composition and particle density of aerosols and to give us a better insight about the origin of particles that whether emitted from anthropogenic or the natural processes (Conner et al., 2001).

Asian aerosols have been shown to have a potential impact on atmospheric chemistry as well as global climate forcing (Clarke et al., 2004). Mineral aerosols produced from windblown soils are an important component of Earth's atmosphere system especially over arid and semi-arid regions of Asia. The Indo- Gangetic plains of South Asia are the largest alluvial tract in the world. The deposits include a thick layer of sand, clay, loam and silt contributing largely to soil related aerosols. The long range transfer of atmospheric particles through dust storms and thunderstorms from the Thar Desert lying towards the west of plains are frequently observed during the summer period which indicates the dominance of mineralogical particles.

The objective of the present work is to characterize aerosol number, mass, volume and surface area in various size ranges, to identify the chemical and structural behavior of individual atmospheric particles by scanning electron microscopy in conjugation with energy - dispersed X- ray analysis (SEM/EDX) and to compare the aerosol concentration during different seasons

Material and methods

Description of sampling site

The study was carried out at our campus site, Dayalbagh, which is surrounded by agricultural fields and is 10 km away from the industrial sector of the Agra city. Agra (27°10′N, 78°05′E, and 169 m.s.l.) is located in the north central part of India. Two thirds of its peripheral boundaries (SE, W and NW) are bounded by the Thar Desert of Rajasthan and therefore is a semiarid area characterized by loose, sandy, and calcareous soil containing an excess of salts. Agra's climate is tropical and strongly influenced by the aeolian dust blown from the Asian subcontinent and Thar Desert of Rajasthan. Meteorologically the year is divisible into three distinct seasons; summer (March–June), monsoon (July–September) and winter (October – February). Summer season is associated with strong hot dry westerly winds and high temperature ranging between 38 - 48 °C. Relative humidity in the summer ranges between 18 and 48%. The monsoon season is hot and humid, temperature ranges from 24 to 36 °C and the relative humidity ranges from 70 to 90%, while in winter season temperature drops below 2°C.

Ambient sampling

The particulate samples for TSP were collected

on the roof of Science Faculty building at a height of about 12 m above ground level using High Volume Sampler (Envirotech APM 460 BL) operated at a flow rate of 1.2 m^3/min for 24 hours on Quartz fiber filter paper. Before exposure, the quartz fiber filters were pre-heated in a muffle furnace at 800 °C for 3 hours to remove organic impurities and analyzed for SEM - EDX. The particulate matter in the ambient air was sampled simultaneously for particle number and mass concentrations. Particle number and mass concentrations were measured in the range of 0.25-32 µm using an optical particle counter Grimm monitor, 31-Channel Portable Aerosol Spectrometer model No: 1.109, a low-volume sampler that uses a light scattering technique to continuously measure particle number concentration and size distribution in an air stream at a flow rate of 1.2 L/ min ± 5% constant with controller for continuous measurement during the sampling period.

SEM- EDX Analysis

The aerosol samples were analyzed by SEM - EDX at National Institute of Oceanography, Goa. The SEM – EDX analysis was carried out with the help of computer controlled field emission scanning electron microscope SEM (JSM – 5800 LV) equipped with an energy dispersive X – ray system (Oxford 6841). The dry and loaded quartz filter papers were cut in 1 mm^2 from the centre of each sample. All the samples were mounted on plastic stubs for gold coating. A very thin film of gold (Au) was deposited on the surface of each sample using vacuum coating unit called Gold Sputter Coater (SPI – MODULE). The fine coating of gold can make the samples electrically conductive. The working conditions were set at an accelerating voltage of 20 kV, a beam current of 40 – 50 µA and the Si (Li) detector 10mm from the samples to be analyzed. X- Ray detection limit is ~0.1%.

RESULT AND DISCUSSION

Seasonal variation of Particle number and mass size distribution

Particle number and size influence the site deposition in the human respiratory tract and the consequent degree of toxicity. The statistics of particle number in different size ranges during different seasons are presented in **Table 1**. The particle in the diameter from 0.25 – 30 µm were divided into three ranges: 0.25 – 0.5 µm (ultrafine mode), > 2.5 µm (fine mode) and 2.5 - 30 µm (coarse mode).

The average number concentration was found to be highest in summer season (286399.1 cm^{-3}) followed by winter (109155.7 cm^{-3}) and monsoon season (67390.68 cm^{-3}). These concentrations were 2.63 and 4.24 times higher during winter and monsoon than summer months. During summer season the particle number concentration followed the pattern: coarse modes > fine > ultrafine. The coarse mode particles contributed the highest percentage (40.1%) of the measured total number of particles during dust event. Higher concentration was attributed to local as well as long range transport of particles during dust storms which occurred frequently during summer season and caused significant increase in atmospheric particles. Earlier

Table 1. Descriptive statistics of measured number concentration in different size ranges

Sampling season	Aerosol type	Mean (cm^{-3})	Median (cm^{-3})	Max. (cm^{-3})	Min. (cm^{-3})	SD
Summer	Ultrafine	221670.6	258837.6	288449.7	104153.3	76142.9
	Fine	301811.0	303719.9	3077 64.8	293350.2	5337.8
	Coarse	308855.7	308973.6	309006.7	308137.9	261.0
Winter	Ultrafine	69125.8	78826.3	92102.6	33983.9	23414.9
	Fine	111471.6	114162.3	124549.5	96484.5	10454.3
	Coarse	127681.9	128014.8	128083.8	125641.4	731.6
Monsoon	Ultrafine	41125.6	57205.4	68752.1	1618.3	29870.2
	Fine	74043.1	74617.8	75953.5	70742.5	1811.0
	Coarse	76246.6	76273.6	76279.3	76018.8	71.8

studies had also reported that coarse particles were more pronounced during the dust events (Pandithurai et al., 2008; Wang et al., 2006; Wu et al., 2008). The dust particles which were transported from Thar Desert of Rajasthan carried Aeolian dust which particularly comprised of large size mineral dust particles. These mineral dust particles mixed with the local soil dust and contributed significantly to coarse mode. Chun et al. (2001) and Lin (2001) had also shown that mineral dust was the major component of coarse mode particles while the fine particles contain primary particles from combustion sources and secondary materials such as sulphate, nitrate and ammonium formed by gas – to – particle conversion. The long range transport of the aerosol dust particles was also supported by back trajectory analysis (Fig 3 a). However, a significant decrease in particle number concentration during monsoon season could be attributed to wash out effect of particles during rainfall. To better understand the number and mass distribution among the various particle sizes, lognormal distribution function over a wide range of particle size of these parameters were plotted, which provided a good fit for a large range of data, that is, dN/ dlogDp and dM/dlogDp were plotted against Dp (Particle diameter) on a log scale where dN and dM were number and mass, respectively found in given size interval. **Fig 1a and 1b** show a plot of lognormal distribution function for number and mass during different seasons. The corresponding mass concentration of the particles varied from 0.9 - 1075 μgm^{-3} with an average of 241.8 ± 214 μgm^{-3} during summer while the particle mass varied from 4.5 - 171 μgm^{-3} with an average of 75.3 ± 52 μgm^{-3} during winter season. However, during monsoon the average mass was observed to be 13.9 ± 8.2 μgm^{-3} ranging from 0.04 – 26.5 μgm^{-3}. The average mass concentration of coarse particles was 3.72 times higher than winter while 22 times higher than monsoon. This significant increase could be attributed to the contribution of dust storms that had larger proportion of coarse particles. The results indicated that the particles with larger size constitued a major fraction to total mass as compared to ultrafine and fine particles. During winter season, the average ultrafine and fine particle mass concentration were found to be higher than summer and monsoon season, respectively. The average fine particle mass concentration was 34 μgm^{-3} which was 3 and 5 times greater than monsoon and summer season. The increase in fine particles during winter season indiacated the dominance of combustion activities (burning of biomass and biofuel) which were mainly contain fine and ultarfine particles. The total number and total mass concentrations were found to be poorly correlated (r^2 = 0.3). Tuch et al. (1997) at a site in Eastern Germany had also shown a poor correlation (r^2 = 0.5) between total number and total mass concentrations.

Variation of Particle surface area and volume distribution

The surface area and volume distributions were

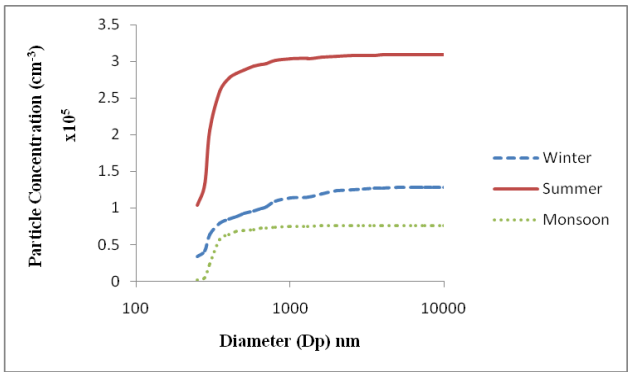

Fig 1 (a) Number size distribution of particles

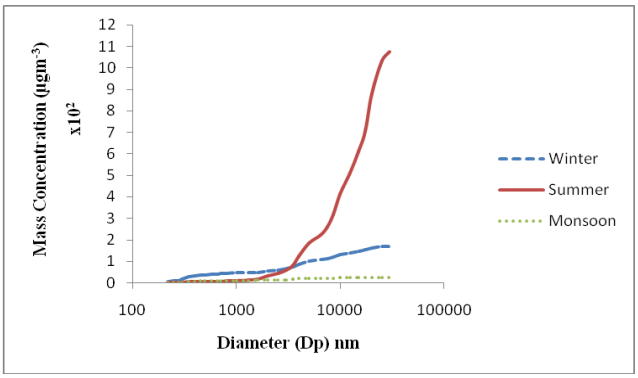

Fig 1 (b) Mass size distribution of particles

important when reactions of gases were considered at the surface of particles or reactions occurred within the particles themselves, like the oxidation of SO_2 to sulphates. Experimentally determined particle number concentrations of ambient aerosols were commonly described as the sum of three or more lognormal distributions. On the basis of literature data and assumption of particles to be spherical, surface area size and volume size distribution was derived from the particle number size distribution. The surface area and volume size distributions was calculated as:

$dS = dN^* \pi Dp^2$, $dV = dN^* \pi/6 Dp^3$ where, Dp= Particle diameter, dV = incremental volume, dS = incremental surface area, dN = incremental number concentration of particles.

The number of particles and particle surface area per unit mass increased with decrease in particle size during winter and monsoon period and indicated that the ultrafine particles contribute highest to number and surface area than fine and coarse particles. The surface area size distribution of the coarse particles ranged from 2.5 - 30 µm show that they contribute about 48% of the total fraction. However, the surface area of the particles varied from 1826 - 259428.6 $\mu m^2/cm^3$ while the volume of coarse particles within the size range of 2.5 - 30 µm show the largest fraction of 55% of the total volume of particles during summer season. Similar results had also been reported by Wu et al. (2008) that contribution of coarse mode particles increased significantly during summer season having dust storm events. The volume size distribution demonstrated that coarse mode particles make the largest part of airborne particles and also contributed a large amount to the surface area. **Fig 2a and 2b** show the average fraction of the particle number, mass, surface area and volume concentrations in the size range from 0.25 – 30 µm in the winter and summer season.

Back Trajectory Analysis

Air mass backward trajectories are one of the widely used methods for the identification of the origin and the pathway of air masses that arrive at any location. In the present study, the air mass backward trajectory analysis was carried out using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single – Particle Lagrangian Integrated Trajectory (HYSPLIT) model based on the GDAS global wind field developed by NOAA/ ARL (Draxler and Rolph, 2003). The back trajectory analysis indicated that during summer most of the air masses came from the west with a rapid moving speed carrying aeolian dust from the Thar Desert of Rajasthan **Fig 3a.** These strong air moving episodes during summer season resulted in a significant increase of aerosol particles. A five day air mass back trajectories were drawn for the 1500m height for monsoon season. The back trajectory results show the origin of southwest monsoon. **Fig 3b** show the moisture laden winds originated from Arabian Sea towards north central region of India. These moisture rich air masses when travelled

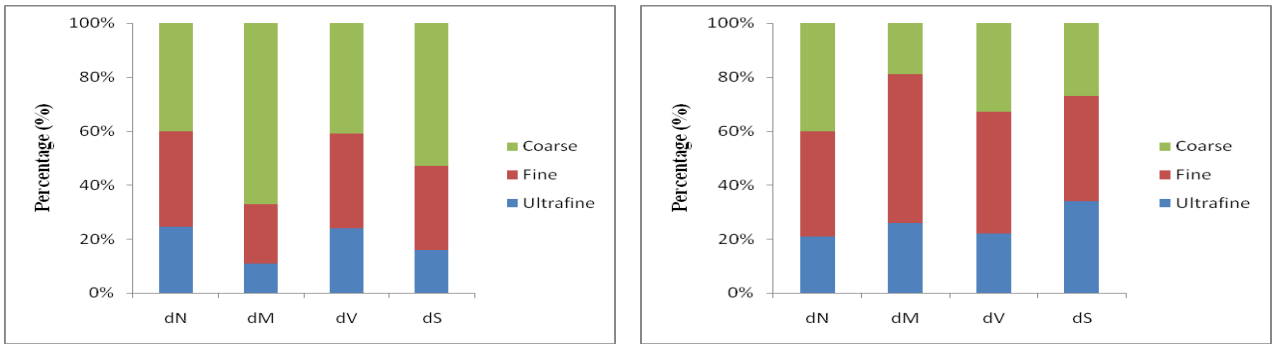

Fig 2. Percentage distribution of Number, Mass, Surface area and volume of airborne particles (a) During summer (b) During winter

from Indian Ocean to Indian Subcontinent were responsible for heavy rains during monsoon. However, the backward trajectory during winter season show that there is no long range transport of aerosols and the aerosol particles were anthropogenic in origin **(Fig 3c).**

SEM EDX Analysis

Scanning electron microscopy with energy dispersed analysis of X- rays (SEM/EDX) provided information on the morphology, elemental composition and particle density of aerosols. It also determined the origin of particles emitting from anthropogenic or natural processes. In the present study, diverse group of air borne particles were analyzed with the size ranging from 2µm to 70 µm.

The number concentration of aerosol particles increased during summer season **(Fig 4).** Individual particle analysis indicate that carbonaceous particles **(Fig 5)** were more predominant during the winter months and showed the presence of biomass burning which is also suggested by back trajectory analysis (Fig 3c). However, aluminosilicate particles which were mainly composed of Si and Al oxides with varying amount of Na, Mg, Ca, Fe and K, Fe/Ti oxides and silica particles were predominant during summer months which clearly indicated the dominance of crustal sources **(Fig 6).** The

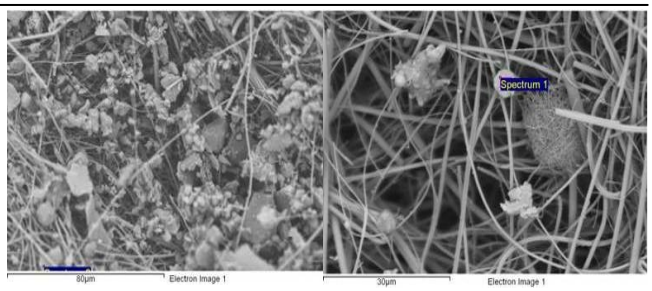

Fig 4. SEM images of bulk particles embedded in the filter paper (a) during summer (b) during winter

results show that the aluminosilicates/quartz contributed about 80% of the total particles analyzed indicating that the crustal materials were the primary contributor of aerosols.

The earlier studies on the elemental composition of aerosol particles at Agra reported by Parmar et al., (2001) further support the soil dominant nature of aerosol particles. However, during summer season, these group of particles showed a distinct increase which indicated the influence of local as well as long range transport of mineral dust particles this was further supported by back trajectory analysis which indicate that during summer most of the air masses came from the west carrying aeolian dust from the Thar Desert of Rajasthan (Fig. 3a).

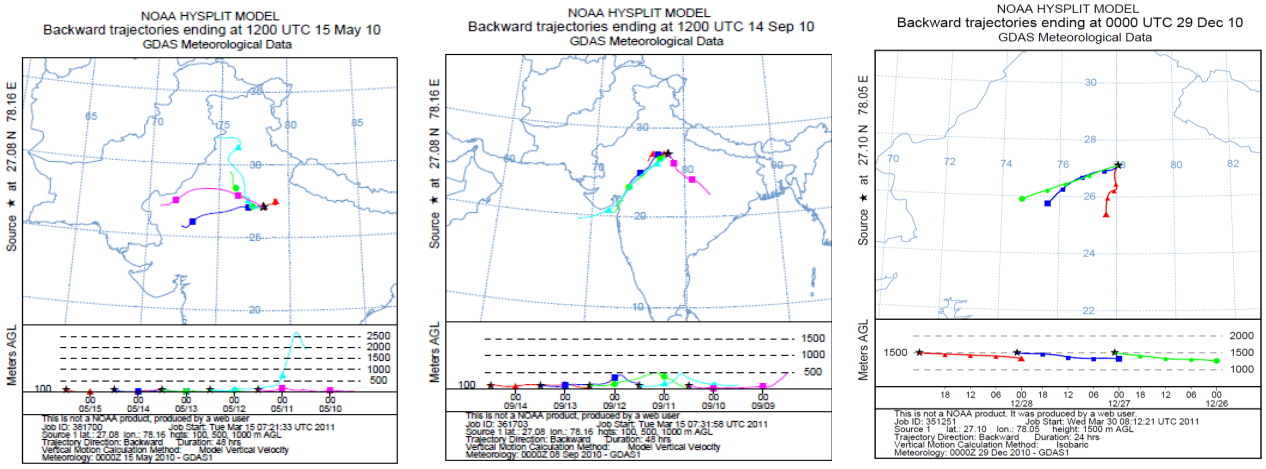

Fig. 3 The typical air mass trajectories arriving at the sampling site (a) Backward trajectory pointing to the dust source from northwest Asian desert region in summer (b)Backward trajectory air masses originated from Indian Ocean and travelling through the Indian sub continent (c) Backward trajectory during winter month indicating anthropogenic origin of aerosols.

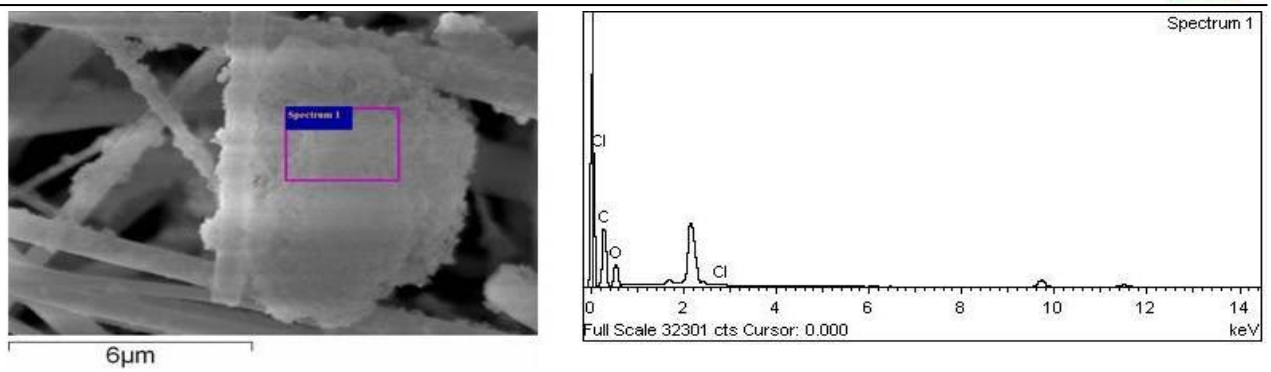

Fig 5. Scanning electron images of a single carbon particle with nearly spherical morphology dominated by C and O (> 90%)

CONCLUSION

The aerosol number concentration of coarse particles (2.5 - 30 µm) showed a significant increase during dust event in summer and indicated the long range transport of aerosol dust particles possibly originating from westerly winds carrying Aeolian dust which particularly comprised of large size mineral dust particles. The volume and surface area size distribution showed that coarse particles also contribute maximum to surface area and volume. Since total particle number and mass concentrations were poorly correlated (r^2 = 0.3) more insight into health related aspects of particulate air pollution would be obtained by correlating respiratory responses with mass and number concentration of ambient particles. The chemical composition and morphology of atmospheric particles were investigated using SEM – EDX system indicated that most of the particles were soil related indicating the dominance of crustal materials during summer season. However, a significant increase of carbonaceous particles during winter season showed the dominance of biomass burning which is also suggested by back trajectory analysis that the origins of these particles were mainly anthropogenic in nature. The stable and cold meteorological conditions dominating during winter also supported the accumulation of these anthropogenic ally originated particles within the atmosphere.

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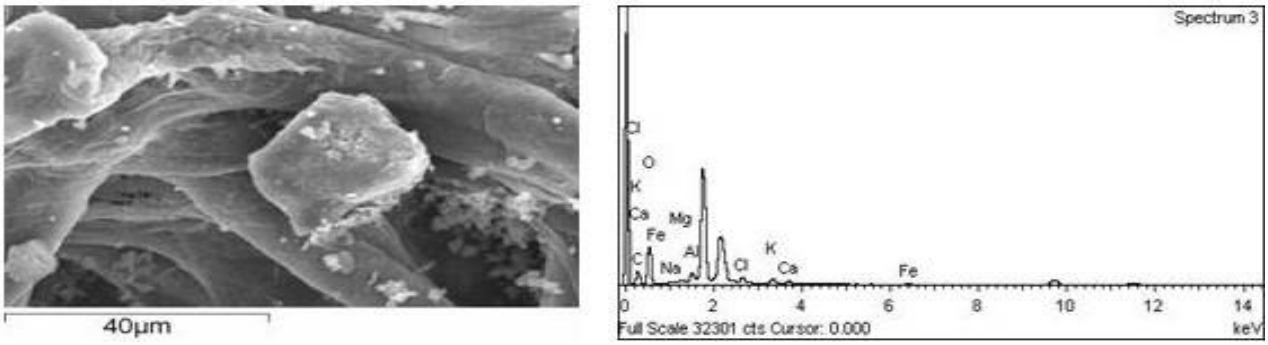

Fig 6. Scanning electron images of and energy – dispersive X- ray spectra of carbonaceous particles with varying amount of soil related components like Na, K, Mg, Ca and Al

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