Spatial and Temporal variations of Organic Chemical compounds in Ambient Air from Ahmedabad City, Gujarat, India

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Abstract
Organic contaminants linked to atmospheric particulate matter have been a major source of worry across the world since the last decade. The ambient air samples were collected from three sites in Ahmedabad city for spatial and temporal variation using a High-Volume Air Sampler (HVAS) to examine the morphology and elemental composition of fine and ultrafine particles by using a Scanning electron microscope (SEM) and an energy dispersive X-ray system (EDAX) from $PM_{2.5}$. The Si, C, O, Cl, Na, Al, K, Ca and Mg were found to be quite abundant in the EDAX spectra. Transition elements such as Ti, Zn, Cu, S and Fe were registered, indicating the influence of industry in liberating the abundance of fine and ultrafine particles.

In addition, the organic chemical compounds were screened by using GC–MS. A total of 230 organic chemicals were detected, including 187 aliphatic and 43 aromatic chemical compounds at three different locations. Results show that aliphatic and aromatic hydrocarbon compounds were found maximum in the urban area followed by industrial and rural areas.

Keywords: HVAS, Spatial and Temporal Variation, Elemental Composition, Organic Compounds, Atmospheric Particulate Matter.

Introduction

Air pollution is one of the world's most pressing issues nowadays which is caused due to presence of substances in the atmosphere that are not only harmful to the health of humans and other living beings but also cause damage to the climate or materials[10]. It has a significant role in numerous illnesses, teratogenicity, mutagenicity and carcinogenicity and causes an estimated 7 million premature deaths per year[22]. Approximately four million people died in 2019 because of fine particle ambient air pollution in Asia. Eastern Europe has the greatest mortality rate[1]. Global issues such as global warming and climate change are mainly caused by air pollution[28,32]. Air pollution is a result of increased urbanization and industrialization across the world, particularly in developing nations such as India. According to World Bank Annual Report[2], organic and inorganic pollutants with particulate matter aerodynamic size 2.5 µg are also characterized as effective air pollutants.

Interactions between atmospheric organic pollutants and their derivatives have awakened some interest over the years. Chemical reactions amongst organics are possible, even without catalysts. A significant number of organic pollutants are released from point and non-point sources. These are the substances that are not commonly monitored but potentially enter the environment[15]. They are also known as contaminants of emerging concern (CECs) for which no regulations are currently established[35]. These pollutants may be mobile and persistent in the air, water and atmosphere at low concentrations[13].

The presence and distribution of chemical compounds associated with the particulate matter around the planet are influenced by the atmosphere. Due to their aerodynamic diameter, the emissions of these particulate matter compounds, notably $PM_{2.5}$, in the urban, rural and industrial environments are known to represent a primary hazard to human health. This $PM_{2.5}$ can enter the body through breathing, causing carcinogenic, mutagenic and teratogenic effects[4,19,24]. Natural and anthropogenic sources (fossil fuel combustion, biomass combustion, plastic combustion, secondary organic transformations and marine sources) produce organic compounds, which make up a large portion of PM[11].

Furthermore, alkanes, alkenes, alkanoic acids, aromatic hydrocarbon compounds, aromatic acids and plasticizers are found in the organic matter of aerosol particles and are employed as the source, transit and receptor tracers for volatile and organic chemical substances [26]. It has been proposed that metallurgical enterprises may release a good number of elements into the atmosphere such as Si, C, O, Cl, Na, Al, K, Ca, Ti, Zn, Cu and S may correlate to vehicle emissions [33] as well as an increased understanding of natural and anthropogenic sources of trace elements. The presence of Bisphenol A (BPA) in surface waters across the world was determined and its effects on human estrogen levels are widely affected. It is the need of the hour to determine the occurrence and distribution of inorganic and organic compounds in ambient air - $PM_{2.5}$ in urban area, industrial and rural area of Ahmedabad city, Gujarat, India.

Material and Methods

Study Area: Gujarat's largest city Ahmedabad (23.0225° N, 72.5714° E) also known as Manchester of the East, is situated in the central region on the banks of river Sabarmati which is known for leading industrial sectors such as chemicals, petrochemicals, cement, ceramic, textiles and engineering. Sampling was carried out in three specific sectors (Fig. 1): (1) Industrial area (S1) - 2.9785° N, 72.6314° E (2) Urban area (S2) - 23.0748° N, 72.5356° E (3) Rural area (S3) - 23.1424° N, 72.5146° E.

Sample collection: The particulate matter ($PM_{2.5}$) samples from the atmosphere were taken using a High-Volume Air Sampler – HVAS (Model: NON-PNS-Combo sampler) in the metropolitan city of Gujarat during Post-monsoon, Pre-monsoon and Monsoon seasons. The particles samples were collected using micro–glass fibre filter (GFF) with a diameter of 47 mm. The particulate matter samples were run for 24 hours at a flow rate of 17 L/min. The concentration of particulate matter is calculated using the gravimetric method. To remove organic contaminants, filters were pre-baked at 100°C for 1 hour before exposure[14]. Before and after sampling, each filter paper was weighed and the net mass concentration was measured. $PM_{2.5}$ filter was individually wrapped in aluminum foil and stored in a polypropylene (PP) container.

To prevent evaporation of the volatile components, all samples were kept in the refrigerator at 4°C until further examination[27]. Measurement of meteorological parameters like temperature, wind speed, light intensity and relative humidity was measured with the help of 4 in 1 anemometer, humidity meter, light meter and thermometer/model: LM-8100.

SEM-EDAX analysis: The filter paper size was utilized to quantify the morphological and elemental composition present for SEM-EDAX analysis. at Sophisticated Instrumentation Centre for Applied Research and Testing – SICART, Vallabh Vidyanagar, Anand. The weight percentage of each element present in the spectrum was determined using an EDAX instrument with a silicon drift detector that quantifies the element and performs elemental mapping with a maximum energy resolution at 129Ev for three sites.

GC-MS analysis: $PM_{2.5}$ filters were allowed to cool to room temperature before being transferred to 15 mL PP tubes for further analysis. The extraction solvent mixture of methanol and dichloromethane (DCM) 1:1 was utilized with certain changes[16]. For the extraction of the target compounds, 2 mL of solvent was introduced to the PP tube followed by ultrasonication (Sonicator, USA) at 50% frequency for 15 minutes. The sample tubes were well mixed using Vortexer for 1 minute before centrifugation for 5 minutes to obtain the supernatant, which was then transferred to new PP tubes and the operation was repeated twice to obtain a total of 6 mL of extract. The extract was evaporated using a hot water bath for up to 1 mL and kept in the refrigerator until the analysis[17].

To determine the presence of organic contaminants, the analysis of the extract was done by using a GC 2010 plus coupled with a Shimadzu Mass Spectrometer at the Central Salt and Marine Chemicals Research Institute in Bhavnagar with a flow rate of 1 mL/min, a cone voltage of 0.3 kV and an ion source temperature of 300°C and helium gas as the carrier.

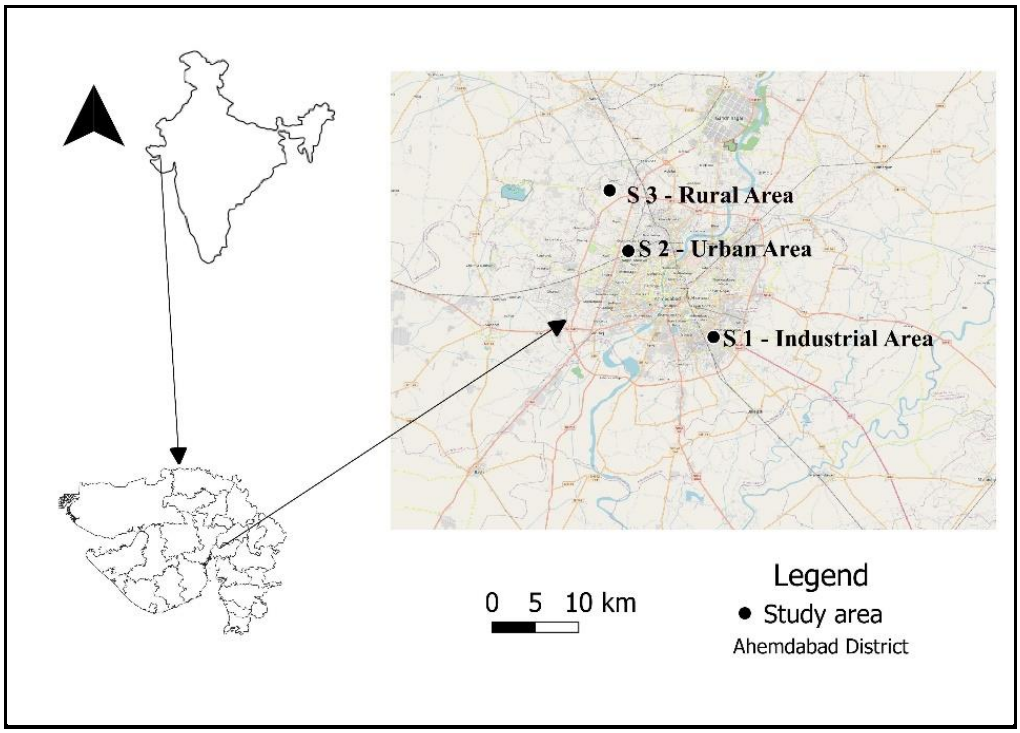

Fig. 1: Three selected sites of Study area in Ahmedabad, Gujarat, India

Table 1
Sampling Information and Mass Concentration

S.N.	Area Type	Sampling seas on	Latitude ° N Longitude ° E	Concentration (µg/m^3)	Light Intensity (Lux)	Temp. (°C)	R. Humidity (%)	Wind Speed (km/h)
1	Industrial	Post monsoon	22.9785, 72.6314	139.96	11890	25.6	47.08	7.4 km/h
2	Urban		23.0748, 72.5356	143.96	15676	38.9	41.20	5.14 km/h
3	Rural		23.1424N, 72.5146	92.12	14207	31.6	35	3.7 km/h
4	Industrial	Pre monsoon	22.9785, 72.6314	131.98	12260	34.5	23	9.7 km/h
5	Urban		23.0748, 72.5356	133.88	19653	44.3	21.10	3.86 km/h
6	Rural		23.1424, 72.5146	83.56	15134	35.1	26.70	9.81 km/h
7	Industrial	Monsoon	22.9785, 72.6314	130.78	11175	30.7	62.20	11.6 km/h
8	Urban		23.0748, 72.5356	132.45	14845	37.3 °C	46.20	7.32 km/h
9	Rural		23.1424, 72.5146	82.52	13942	34.58°C	38.73	10.9 km/h

All the identified aliphatic and aromatic chemical values were calculated using the chromatogram's peak height percent. For each chemical, chromatograms from Library: NIST17.lib m/z and PubChem were used to identify it. The interference was removed by performing a blank during the organic chemistry analysis.

Results and Discussion

Mass concentration is the most important criterion to determine air quality. Table 1 shows the maximum $PM_{2.5}$ concentrations was 143.96 (µg/m^3), 133.88 **(**µg/m^3**)** and 132.45 (µg/m^3) at urban area during Post-monsoon, Pre-monsoon and Monsoon season respectively. According to National Ambient Air Quality Standards, the concentration of all the samples was within the range. The combined impact of climate processes and anthropogenic outrush from numerous regional sources such as industrial emissions, automotive exhaust, waste incineration and fossil fuel combustion could explain the elevated amounts of particulate matter at these sampling locations.[29]

Elemental Concentration: To understand the sources and transport of pollutants and to have a better idea of vehicular pollution, EDX-spectra are used. In the industrial area (S1), the concentrations of elements (%) were: O (29.77) > Si (22.59) > C (19.35) > Na (6.92) > Zn (5.67) > K (3.97) > S (3.28) > Mg (0.49) > Ti (2.04) > Ca (1.98) > Al (2.66) > Fe (0.79) > Cl (0.51) during post-monsoon season. In the season of pre-monsoon, concentrations were: O (31.21) > Si (21.58) > C (19.24) > Na (6.81) > Zn (4.13) > K (3.27) > S (1.86) > Mg (0.21) > Ti (1.8) > Ca (1.87) > Al (2.59) > Fe (0.76) > Cl (4.47) and during monsoon season, the concentrations were: O (44.37) > Si (23.47) > C (11.3) > Na (7.93) > Zn (3.81) > K (2.27) > S (0.42) > Mg (0.14) > Ti (1.74) > Ca (1.45) (Fig. 2).

In the Urban area (S2), the concentrations of elements were: (%), O (33.5) > Si (23.51) > C (16.77) > Na (7.61) > Zn (4.74) > K (3.06) > Cl (2.78) > Al (2.74) > Ti (1.98) > Ca (1.79) > S (0.96) > Mg (0.27) > Fe (0.28) during post-monsoon season. In the season of pre-monsoon, concentrations were: O (37.11) > Si (26.16) > C (12.46) > Na (8.67) > Zn (4.48) > K (3.1) > Al (3.02) > Ti (2.07) > Ca (1.7) > S (0.51) > Mg (0.12) > Cl (0.3) > Fe (0.29) > and during monsoon, season the concentrations were: O (42.13) > Si (27.21) > C (7.09) > Na (8.15) > Zn (4.6) > Br (3.66) > K (2.94) > Ti (2.33) > Ca (1.54) > S (0.17) > Mg (0.17). (Fig. 3).

In the rural area (S3), the concentrations of elements (%) were: O (27.7) > Si (22.6) > C (14.6) > Zn (6.8) > Al (5.26) > Na (4.42) > K (3.53) > Cl (3.53) > S (2.78) > Ti (2.43) > Ca (2.36) > Fe (1.64) > Cu (1.44) > Mg (0.51) during post-monsoon season. In the season of pre-monsoon, concentrations were: O (31.28) > Si (26.38) > C (13.49) > Zn (5) > Al (5.09) > Na (6.1) > K (3.96) > Ca (2.4) > Ti (2.31) > Cl (1.97) > S (0.83) > Fe (0.79) > Mg (0.4) and during monsoon season, the concentrations were O (41.76) > Si (26.5) > C (8.03) > Na (7.45) > Zn (5.15) > Br (3.66) > K (2.89) > Ti (2.35) > Ca (1.96) > S (0.16) > Mg (0.1). (Fig. 4). In all sites, Si, C and O elements were dominant during all three seasons. The biological particles such as microorganisms and fragments of all varieties of living matter (i.e. viruses, bacteria, fungal spores, pollen, plant debris and animal matter) were present in the atmosphere for C and O. Besides this, the highest concentration of C is

possibly due the vehicle pollution.

Pure Si has natural and anthropogenic origins[21]. Cement, glass, bricks, clay, pottery and other construction materials all contributed to silica. Consequently, it is equally possible that these particles are derived from building construction and destruction. However, just trace amounts of other crucial tracers (Cu, Fe, Al, Cl, Mg, Ti, Ca, S, K, Zn and Na) are present[5-9,18,23,27,30,37]. Scanning electron micrograph shows the different types of elemental shapes like Subangular morphology, platy-shaped structure, irregular morphology, globular clustered structure and spherical shaped structure[34] (Figs. 5, 6, 7).

Qualitative analysis of Organic chemical compounds: A total of 230 organic chemical compounds were detected in $PM_{2.5}$ size collected from Ahmedabad. Among these, 187 were aliphatic compounds and 43 were aromatic compounds. The peak height % of the detected compound was considered and the detection of these substances was completely variable and independent of any specific particle size.

Total aliphatic hydrocarbon (TAHs): Road crustal emissions are responsible due to incomplete combustion of fossil fuels, petroleum residues and wood combustion. Ceramic dust might be an important source for n- alkanes in the cold period[34]. Out of 187 aliphatic compounds 143 n-alkane compounds including short-chain and long-chain (C6 to C54), 42 n-alkanoic acid compounds and 2 alkenes were present. Total short-chain alkanes were 129 ($C_n \leq C_{26}$) which are mainly derived from anthropogenic emissions as well as fossil fuel input contributing to the production of short-chain n-alkanes. while a total of 14 long-chain n-alkanes ($C_n > C_{26}$) were identified which were typical of the biogenic source[36].

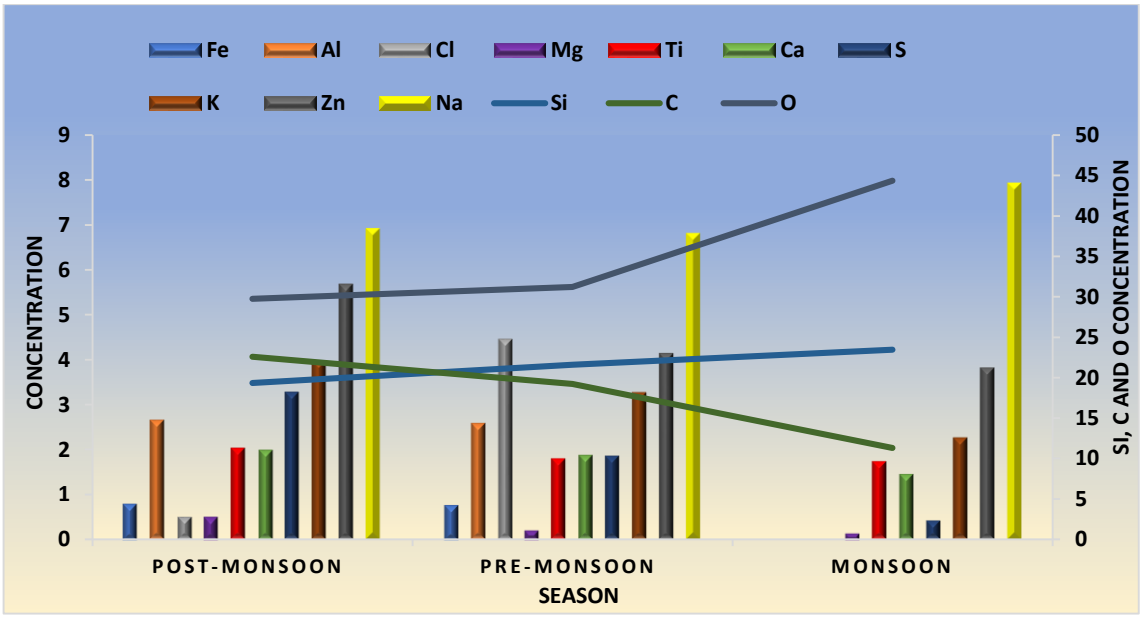

Fig. 2: Temporal variation in the concentrations of Elements at Ahmedabad Site-1 Industrial area

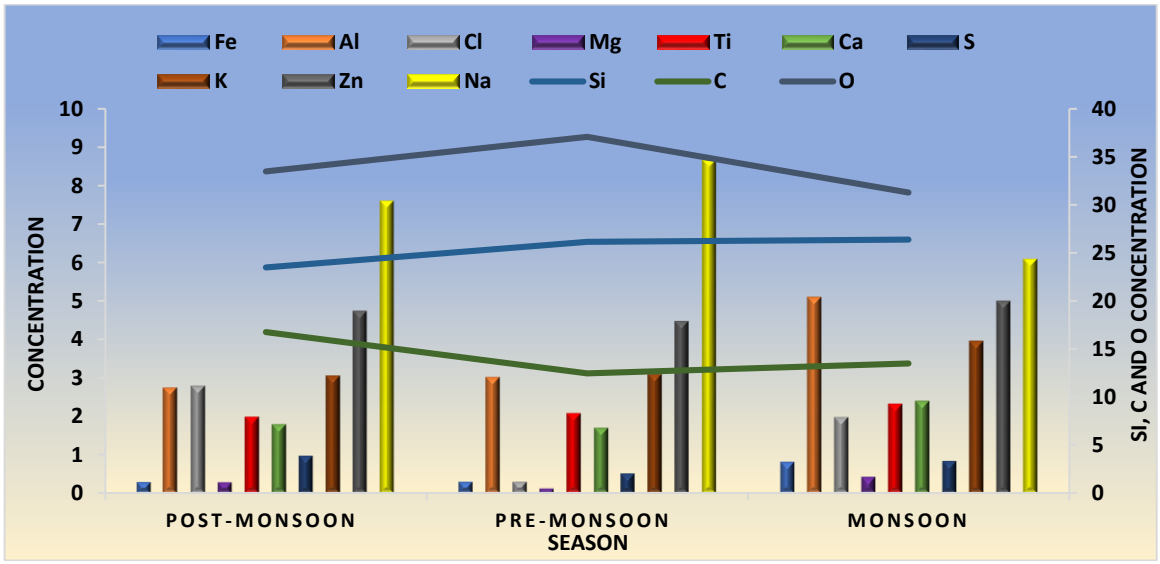

Fig. 3: Temporal variation in the concentrations of Elements at Ahmedabad Site-2 Urban area

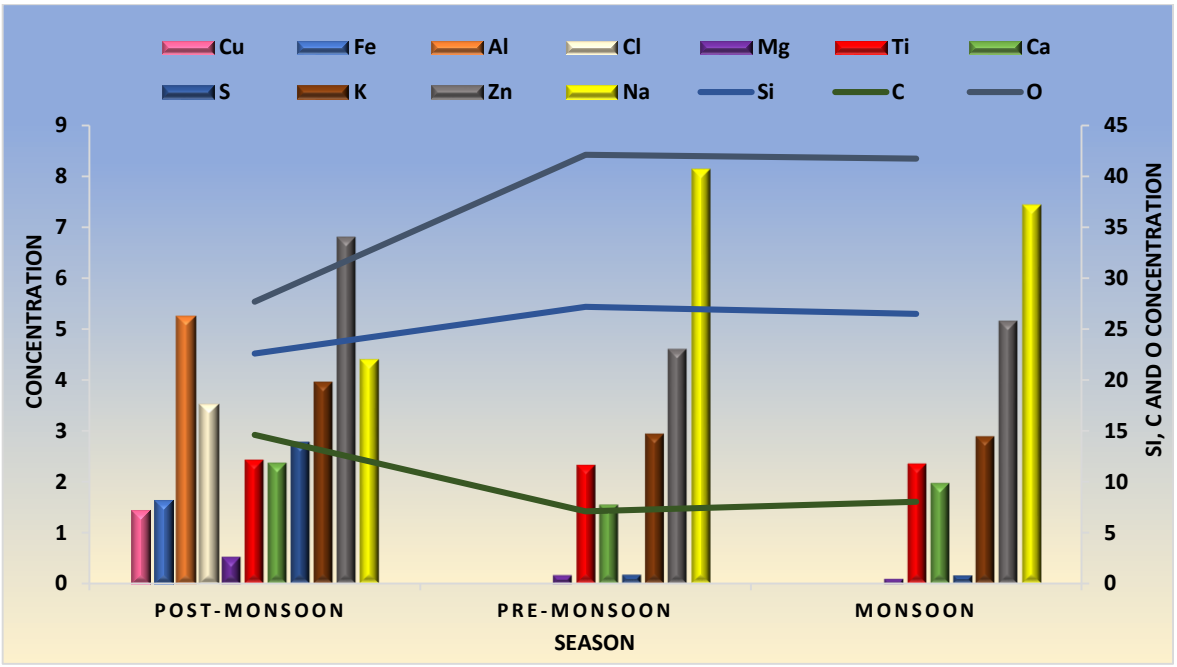

Fig. 4: Temporal variation in the concentrations of Elements at Ahmedabad Site-3 Rural area

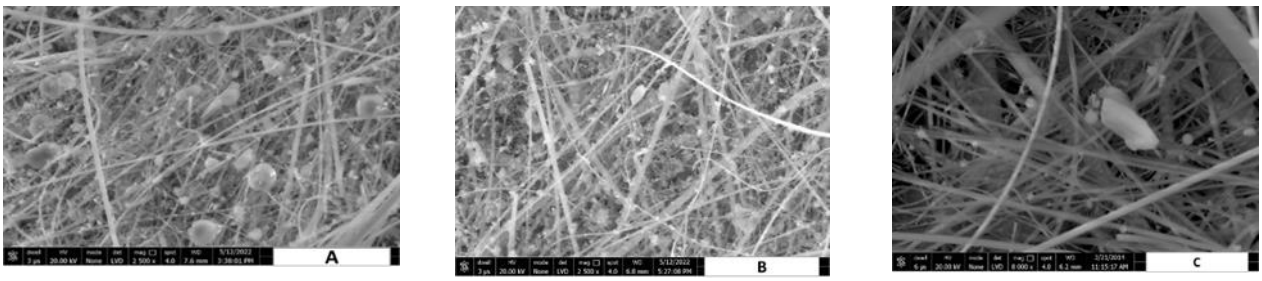

Fig. 5: A, B and C Scanning electron micrographs and EDAX spectrum of aerosols at Site-1 Industrial area Post-Monsoon, Pre-Monsoon and Monsoon season respectively.

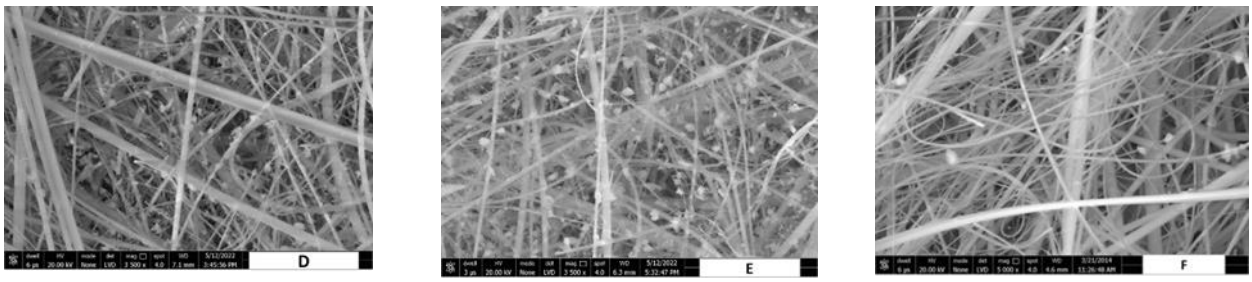

Fig. 6: D, E and F Scanning electron micrographs and EDAX spectrum of aerosols at Site-2 Urban area, Post-Monsoon, Pre-Monsoon and Monsoon season respectively.

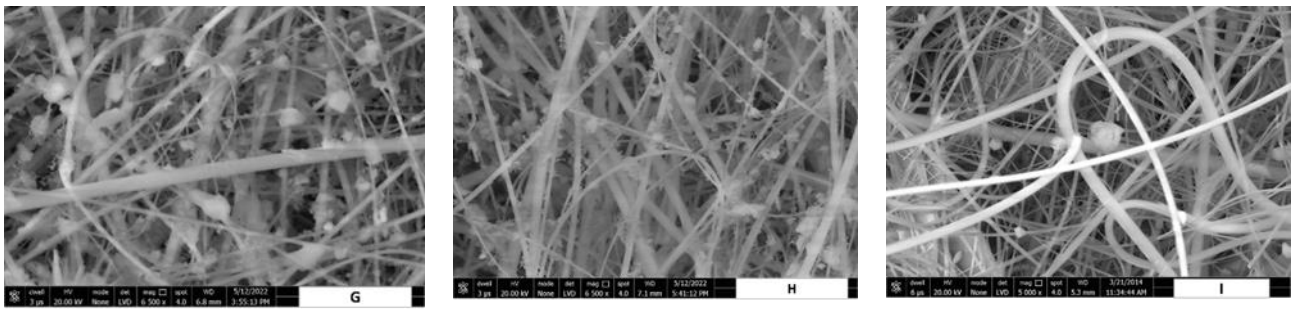

Fig. 7: G, H and I Scanning electron micrographs and EDAX spectrum of aerosols at Site-3 Rural area, Post-Monsoon, Pre-Monsoon and Monsoon season respectively.

Table 2
The $PM_{2.5}$ fine particulate matter associated with Organic chemical compounds.

Season	Site	Peak height (%)	No. of Compounds	Dominant compounds from 2.0
Post-Monsoon	Industrial area	0.0-0.5	21	Octadecane, 3-ethyl-5-(2-ethylbutyl)-
		0.51-1.0	13	Hexacosane, 1-iodo-
		1.01-2.0	9	1-Monopalmitin, 2TMS derivative
		≥2.0	9	Eicosane
	Urban Area	0.0-0.5	31	Nonane, 4,5-dimethyl-
		0.51-1.0	22	6-Methoxy-2-hexanol, TMS derivative
		1.01-2.0	8	3-(2-Carboxy-1-methyl-ethoxy)-5-phenyl-pent
		≥2.0	12	Butylated Hydroxytoluene
	Rural area	0.0-0.5	30	Dodecane, 2-methyl-
		0.51-1.0	9	1-Monopalmitin, 2TMS derivative
		1.01-2.0	8	Bis(2-ethylhexyl) phthalate
		≥2.0	6	Eicosane
Pre-Monsoon	Industrial area	0.0-0.5	31	Benzaldehyde, 4-(1-methylethyl)-
		0.51-1.0	8	Triacontane, 1-iodo-
		1.01-2.0	9	Dodecane, 2,6,11-trimethyl-
		≥2.0	9	Phenol, 2,4-bis(1,1-dimethylethyl)-, phosphite
	Urban Area	0.0-0.5	34	Decane, 1-iodo-
		0.51-1.0	11	Octacosane
		1.01-2.0	9	1-Monopalmitin, 2TMS derivative
		≥2.0	10	Phenol, 2,4-bis(1,1-dimethylethyl)-, phosphite
	Rural area	0.0-0.5	19	Decane, 3,7-dimethyl-
		0.51-1.0	16	Heptadecane
		1.01-2.0	7	1,3,5-Trisilacyclohexane
		≥2.0	10	Phenol, 2,4-bis(1,1-dimethylethyl)-, phosphite
Monsoon	Industrial area	0.0-0.5	27	Phthalic acid, nonyl pentadecyl ester
		0.51-1.0	11	Octadecane, 5-methyl-
		1.01-2.0	9	Bis(tridecyl) phthalate
		≥2.0	8	Eicosane
	Urban Area	0.0-0.5	39	Undecane, 5,7-dimethyl-
		0.51-1.0	14	Octadecane, 1-iodo-
		1.01-2.0	10	Heptacosanoic acid, methyl ester
		≥2.0	7	Phenol, 2,4-bis(1,1-dimethylethyl)-, phosphite
	Rural area	0.0-0.5	40	Sulfurous acid, 2-propyl tetradecyl ester
		0.51-1.0	11	Phenol, 3,5-bis(1,1-dimethylethyl)-
		1.01-2.0	6	Hexadecanoic acid, 2-hydroxy-1-(hydroxymet
		≥2.0	6	Eicosane

Based on GC-MS results, organic compounds are classified into 4 different groups based on their peak (%) height. Among alkane compounds, eicosane, hexadecane, heptadecane, 2,6,10,15-tetramethyl, heneicosane and methyl stearate were dominant in the atmosphere (Table 2). Hexadecanoic acid methyl ester, Tridecanoic acid 4,8,12-trimethyl methyl ester, heptacosanoic acid methyl, decanedioic acid, bis(2-ethylhexyl) ester, eicosanoic acid were predominant in the atmosphere. In the industrial area, long-chain alkane was registered higher during the post-monsoon season, there was not much variation encountered in the number of short chain alkane, however, alkanoic acid was found greater during the monsoon season (Fig. 8).

The number of n-alkane was more in the urban and rural area during all three seasons. There was not much more variation registered for alkene and the maximum number of n-alkanoic acid was documented during pre-monsoon (Figs. 9 and 10).

Total aromatic hydrocarbons: A total 43 aromatic compounds were identified including PAHs, aromatic acids and plasticizers. They are a class of contaminants that are frequently found in the atmosphere. They have high melting and boiling points, low vapor pressure and very low water solubility. Phenol, 2,4-bis(1,1-dimethylethyl), phosphite, 7,9-Di-tert-butyl-1-oxaspiro (4,5) deca-6,9-diene-2,8-dione, cyclohexasiloxane, dodecamethyl- were found in the atmosphere with maximum peak height. Aromatic acids like 1,2-Benzenedicarboxylic acid, bis(2-methylpropyl) ester, Benzenepropanoic acid, 3,5-bis(1,1-dimethylethyl)-4-hydroxy-, methyl ester were detected in the extracted sample.

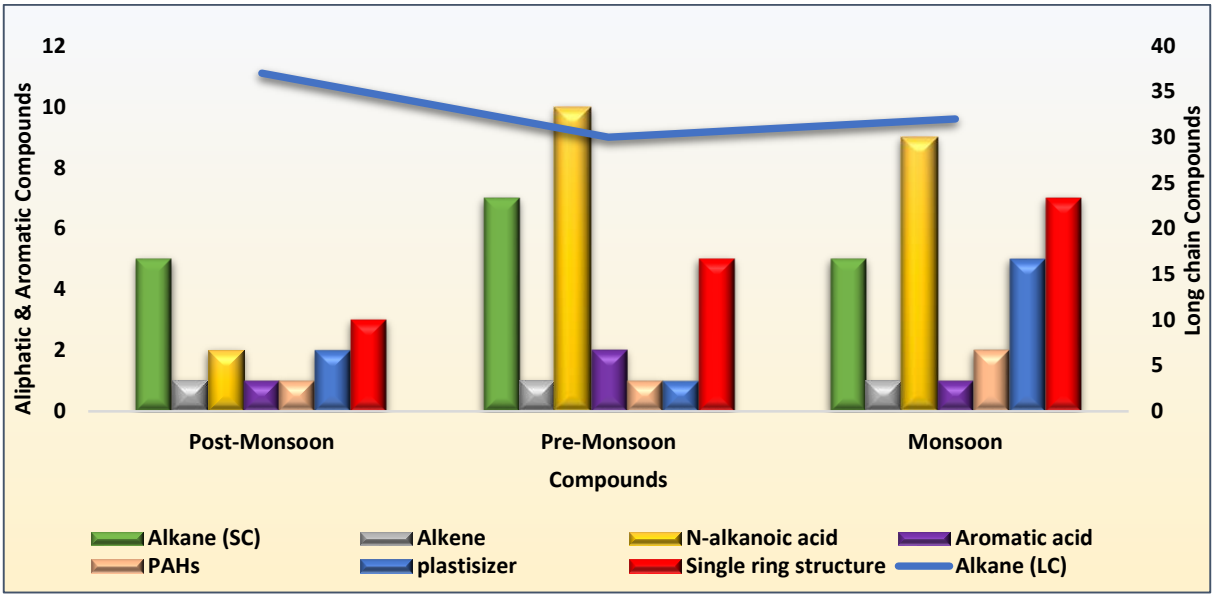

Fig. 8: Organic pollutants at Site-1 Industrial area, Ahmedabad

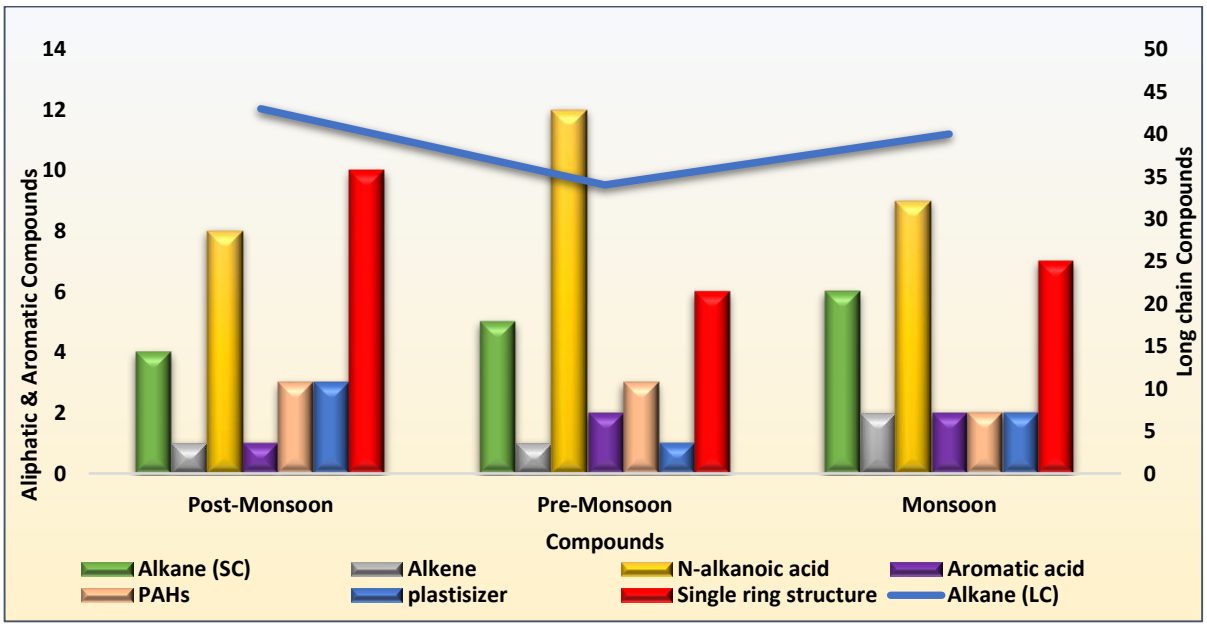

Fig. 9: Organic pollutants at Site-2 Urban area, Ahmedabad

Table 3
Total aromatic hydrocarbons

S.N.	Name	Functionality
1	1,2-Benzenedicarboxylic acid, bis(2-methylpropyl) ester	Aromatic acid
2	1,3-Benzenedicarboxylic acid, bis(2-ethylhexyl)ester	
3	Benzenepropanoic acid, 3,5-bis(1,1-dimethylethyl)-4-hydroxy-, methyl ester	
4	1H-Imidazole, 4,5-dihydro-2-(phenylmethyl)-	PAHs
5	2-Isopropyl-5-methyl-1-heptanol	
6	2-tert-Butyl-4,6-bis(3,5-di-tert-butyl-4-hydroxybenzyl)phenol	
7	4-[2-(4-Fluorophenyl)ethyl]piperidine	
8	5,5'-Di(ethoxycarbonyl)-3,3'-dimethyl-4,4'-dipropyl-2,2'-dipyrrylmethane	
9	7,9-Di-tert-butyl-1-oxaspiro(4,5)deca-6,9-diene-2,8-dione	
10	cyclohexasiloxane, dodecamethyl-	
11	Tris(2,4-di-tert-butylphenyl) phosphate	
12	Urs-12-en-28-al	
13	Phenol, 2,4-bis(1,1-dimethylethyl)-, phosphite	
14	Bis(2-ethylhexyl) phthalate	Plasticizer
15	Dibutyl phthalate	
16	Phthalic acid, 7-bromoheptyl butyl ester	
17	Phthalic acid, butyl undecyl ester	
18	Bis(tridecyl) phthalate	
19	Di-n-octyl phthalate	
20	Phthalic acid, bis(7-methyloctyl) ester	
21	Phthalic acid, nonyl pentadecyl ester	
22	1,1,3,6-tetramethyl-2-(3,6,10,13,14-pentamethyl-3-ethyl-pentadecyl)cyclohexane	Single ring structure
23	1,2-Bis(trimethylsilyl)benzene	
24	1,3,5-Trisilacyclohexane	
25	1H-imidazole-2-methanol, 1-decyl-	
26	2,4-Di-tert-butylphenol	
27	2,6-Lutidine 3,5-dichloro-4-dodecylthio-	
28	2-t-Butyl-3-methyl-1-(2,4,6-triisopropyl-benzenesulfonyl)-imidazolidin-4-one	
29	3-(2-Carboxy-1-methyl-ethoxy)-5-phenyl-pent	
30	3-Ethyl-3-methylheptane	
31	Benzaldehyde, 4-(1-methylethyl)-	
32	Benzene, (2,3-dimethyldecyl)-	
33	Butylated Hydroxytoluene	
34	Cyclohexanol, 2,4-dimethyl-	
35	L-(-)-Fucose, tetrakis(trifluoroacetate), benzyloxime	
36	Phenol, 3,5-bis(1,1-dimethylethyl)-	
37	p-Trimethylsilyloxyphenyl-bis(trimethylsilylox	
38	p-Trimethylsilyloxyphenyl-bis(trimethylsilyloxy)ethane	
39	Silane, cyclohexyldimethoxymethyl-	
40	Thymol, TBDMS derivative	
41	2,2,3,5,6,6,7-Heptamethyl[1,4,2,3,5,6,7] dioxapentasilepane	
42	Cyclotrisiloxane, hexamethyl-	
43	Ethyl homovanillate, TMS derivative	

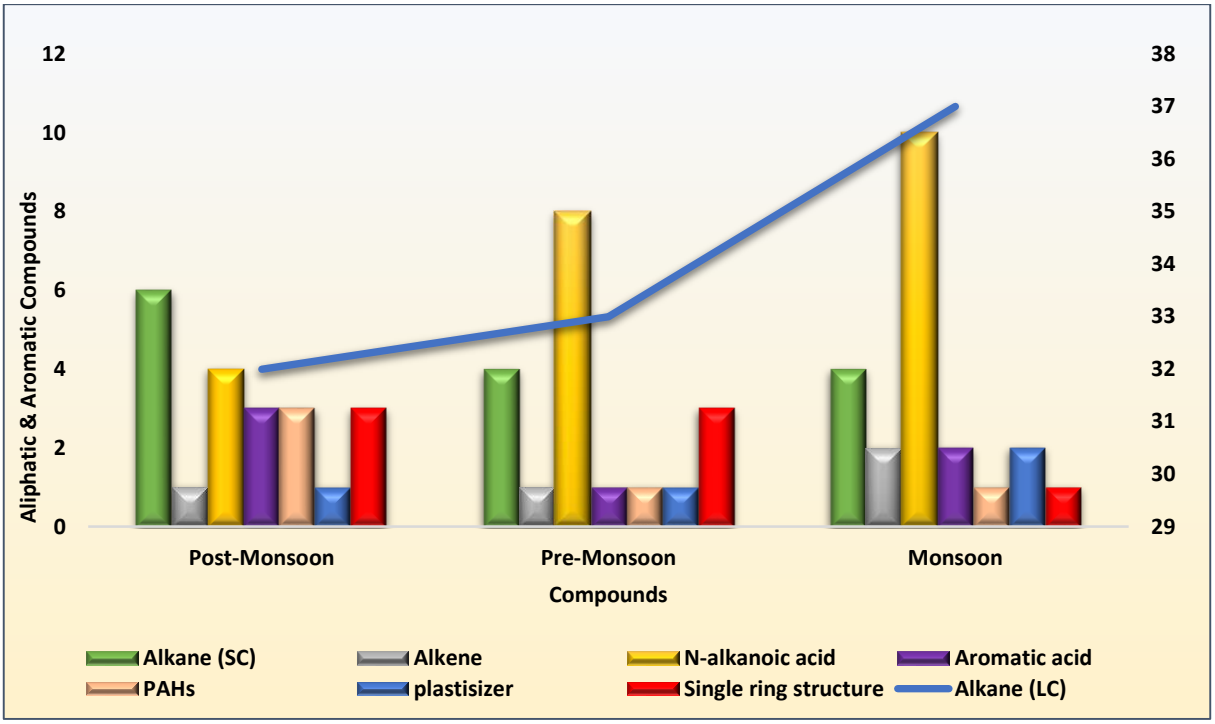

Fig. 10: Organic pollutants at Site-3 Rural area, Ahmedabad

The most abundant and frequently detected plasticizers were phthalic acid, bis(7-methyloctyl) ester, di-n-octyl phthalate, Bis(tridecyl) phthalate. The industrial area contains a greater number of plasticizers (Fig. 8) whereas the number of PAHs and single ring aromatic compounds were maximum in the urban area (Fig. 9) while the aromatic acids were found higher in the rural area as compared to industrial and urban area (Fig. 10) due to the incomplete combustion of organic materials which release polycyclic aromatic hydrocarbons from home heating, coal gasification, liquefying plants, coal-tar pitch and asphalt manufacture.[34]

Conclusion

The cold weather during the winter season encourages ambient particles in the atmosphere to live longer, resulting in a higher concentration of particulate matter ($PM_{2.5}$). The SEM-EDAX device was used to analyse the elemental and morphological content of air particles. The findings demonstrate that these airborne particles were both natural and anthropogenic in origin, accounting for most of the particles. The organic chemical analysis was done using a GC-MS instrument and was grouped by indices scale based on the peak height of the fine and ultrafine particulates and a total of 230 compounds were detected with different distribution patterns.

The study provides vital information about the occurrence and distribution of the ambient air organic pollutants in three study areas of Ahmedabad where compound like eicosane was found in all the study area during all three-season confirming that such compounds bioaccumulate in the food chain and atmosphere which demand further detail study on spatial, temporal variations and its impact on human health issues.

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