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Analysis of trace elements in atmospheric particulate in various regions of Tamil Nadu, India.

R. Senthil Kumar[1], P. Rajkumar[2]

[1]Department of Physics, T.J.S. Engineering College, Peruvoyal- 601 206, Tamilnadu, India.
[2]Department of Physics, Sethupathy Govt. Arts College, Achunthan Vayal - 632 502, Ramanathapuram District, Tamilnadu. India.

Abstract: The aim of the study to assess the element in the air concentration and the potential ecological risk of trace elements in Tamilnadu, India. The samples were subjected to various methods to find out the different elements like Na, Pb, Mn, K, Fe, Al, Ca, Mg and Si of the air dust particles which collected from various places in Tamilnadu. The results of concentration of trace elements in air were discussed. Nemerow pollution indices indicated heavy pollution with metals. The enrichment factor also calculated for the minerals. FESEM-EDX studies also included for better understanding of elemental irregular structures.
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Keywords: Air pollution, Air suspended particles, Trace elements, Enrichment Factor, Field Emission Scanning Electron Microscope - Energy Dispersive X-Ray.

Introduction

The Man breathes nearly 22000 times a day and inhales approximately 15 kg of air per day. But then, the air is abundantly available over the surface of the earth, which it contains a lot of impurities. In India, the problem of air pollution has reached alarming proportions due to concentration of industries and vehicles. Air quality management in India is being studied with more rigorous. Many Asian cities are facing environmental crisis due to severe air pollution[1]. Various epidemiological studies have conducted to provide an evidence for an association between acute Particulate Matter (PM) exposures and increases in mortality and morbidity among people who are suffering with respiratory and cardiovascular diseases[2, 3]. Elements are released to the atmosphere from both anthropogenic and natural sources. The epidemiological studies have found an association between the fine particle concentrations and increased human health effects[4, 5].

Atmospheric particulate matter is from many sources and differed in terms of physical and chemical characteristics, effects on human health and the ecosystem, persistence in the atmosphere and the ability to react with each other[6]. Particulate matter (PM) emissions are caused by traffic exhaust and combustion processes which are predominant in the fine size range (particle diameter<1 μm). Whereas mechanically produced particles and road dust particles are mainly found in the coarse size fraction (particle diameter>1 μm). Most emissions like traffic and industry are related to emission or atmospheric dilution processes which are varied within a few hours[7] Moreover, these small particles are preferentially deposited in the human respiratory system [8, 9].

According to various studies, heavy metals pollution in the environment is mainly derived from anthropogenic sources[10, 11]. In Asian cities the high level of human activities in and around transport corridors is occurring in the common place, the incidence of people is having high health problem[12]. The Windsor, Ontario exposure assessment study[13] is launched in 2005, its aim is to provide accurate and representative human exposure data collected across the city for indoor and outdoor residential locations, and personal environments. Urban air pollution is responsible for 865000 premature deaths every year, about 60%of which occur in Asia[14]. In Indian, most of the mega cities are polluted one in the world. Air concentration of a number of air pollutants is much higher than levels recommended by the World Health Organization[15]. The effect of air pollutants on the macro-morphological features of plants are focused mainly on the leaf injury symptoms such as leaf tip burns, intervene necrosis in Mangiferaindica[16]; discoloration (chlorosis) associated with cell collapse and necrosis and a general overall waxy appearance of leaves of some perennial weeds[17].

Materials and Methods

A large variety of field techniques for assessing settled dust have been described in the literature[18, 19]. Keeping the above problem in view, the suspended particulate matter (SPM) has been undertaken in Tamilnadu. The air suspended particles which are deposited on tissue papers at the height of 20 feet in roadside and land area were collected[20, 21]. These tissue papers were washed in distilled water and the settled dust particles at the bottom of the container are then dried at 110ºC in the oven and are used for analysis. The samples were collected from 38 towns and cities of Tamilnadu, which are labeled as 1 to 111. The samples were mainly collected from the Vehicular area (say Bus stand), Industrial area and Residential area which covers almost all major districts of Tamilnadu. The selected sampling areas (40 out of 111) are listed out with their latitude and longitude in the table 1.

Different techniques are used to find the trace minerals. The concentration of elements is measured in the Chemical Institute of Research and Training (Chemical analysis and consultancy services) 239/131, s.e.e.d colony, alagapuram, Salem – 636 016, Tamilnadu. All reagents used in this study were of analytical reagent grade and distilled water was used for preparation of solutions.The samples were dropped into a solution. Initially 2 grams of sodium hydroxide were taken in a nickel crucible. The electric Bunsen furnace at 90 divisions was set and waits for half an hour. Samples of 50 grams were added after the complete melting of sodium hydroxide and covered the crucible. The sample kept in dull red hot condition for 5 minutes. The crucible were taken out and kept it on porcelain tile and cool it at room temperature. Crucible has filled with a ¾ th of distilled water and kept it covered overnight and added 10 ml of HCL (conc). This solution is taken in a 250 ml standard flask after cooling it in a room temperature; it is transferred into a plastic bottle. Silica and Alumina have estimated by the Ammonium molybate method and Alizarin red s method. The trace elements of Fe, Ca, Mn, Na, Mg, K, has estimated by 1,10 Phenonthoroline Method, Titration method involving EDTA, Periodate Method, Flame Photometric Method, Titration method involving, Flame Photometric Method[22-25].

The Scanning Electron Microscope (SEM) has played an important role for many years in research and development. The samples have analyzed with Field Emission Scanning Electron Microscope with Energy Dispersive X-Ray (FESEM- Hitachi-japan – model no: su6600 & EDX - Horiba- japan, model no: 8121-H) in National center for Nano Science and Nano Technology, University of Madras, Tamil Nadu, India. It utilizes advanced Variable Pressure Technology and an improved Schottky field emission electron source that provides exceptional imaging and high probe current with great stability. The Variable Pressure mode allows the operator to change vacuum conditions in the sample chamber from high vacuum (≤10-4Pa) to low vacuum (10-300Pa). It is a new and versatile Field Emission SEM for a diversified range of applications including observations and analyses of advanced materials which have become increasingly important in modern science and engineering.

Table No.1: Latitude and Longitude of sampling sites

				Sample Numbers		
City/ Town	**Latitude**	**Longitude**	**District**	**Vehicular Area**	**Residential Area**	**Industrial Area**
Kanchipuram	N 12° 50' 3"	E 79° 42' 13"	Kanchipuram	1	2	3
Cuddalore	N 11° 45' 0"	E 79° 45' 0"	Cuddalore	10	11	13
Hosur	N 12° 43' 58"	E 77° 49' 48"	Krishnagiri	23	25	26
Erode	N 11° 20' 32"	E 77° 43' 38"	Erode	36	37	38
Tiruppur	N 11° 6' 43"	E 77° 21' 15"	Tiruppur	39	40	42
Karur	N 10° 57' 26"	E 78° 4' 51"	Karur	49	50	51
Tiruchirappalli	N 10° 47' 25"	E 78° 42' 16"	Tiruchirappalli (Trichy)	52	53	
Madurai	N 9° 55' 30"	E 78° 7' 11"	Madurai	70	72	73
Sivakasi	N 9° 26' 54"	E 77° 47' 53"	Virudhunagar	79	80	103
Tuticorin	N 8° 45' 50"	E 78° 8' 5"	Tuticorin	82	83	85
Tirunelveli	N 8° 43' 58"	E 77° 42' 0"	Tirunelveli	86	87	89
Tiruvarur	N 10° 46' 16"	E 79° 38' 13"	Thiruvarur	95		96
Mayiladuthurai	N 11° 6' 6"	E 79° 39' 7"	Nagapattinam	97		98
Sriperumbudur	N 12° 58' 2"	E 79° 56' 30"	Kanchipuram		105	
Ayanavaram	N 13° 6' 1"	E 80° 13' 45"	Chennai		110	109
Koyambedu	N 13° 4' 8"	E 80° 11' 28"	Chennai	111		

Results and Discussion

Concentration of elements in airborne Samples

The concentrations of elements in various samples are summarized in fig 1 (a-i). This study is exposed that the concentration of elements in the samples of various places in Taminadu, where the effluents were directly discharged from the various sources. The decrease of elements contents in various cities of airborne particles. The ranking order of mean element contents in the residential area of various cities was Si>Mg>Ca>Fe>Al>k>Na>Mn>Pb respectively. The trends of element content in our studied samples were consistent with the results reported by the Hong Kong Environmental Protection Department[26], where Si was identified as the most abundant in the air particulate sample followed by Mg and Ca, and the least was Pb. Our results were compared with the National Institute for Environmental Studies and certified air particulate SRM 1633a and Detection limit (L.O.D. µg m^{-3})[27]. The ranking order of mean element contents in the Vehicular area of various cities was Si>Ca>Mg> Fe>Al>k>Na>Mn>Pb respectively. In vehicular area, Si was identified as the most abundant element and least was Pb. The mean element contents in industrial areas were Si>Ca>Mg> Fe>Al>Na>K>Mn>Pb respectively. In an industrial area Si was identified as the most abundant element and least element was Pb. This study revealed that all the examined metals concentrations in residential areas were around compared values. However, the levels of trace element content in the air dust samples were verified with the reference values which were suggested by WHO (1993)[28]. The Pb concentrations in all the studied samples were found to be lower than all the prescribed limits, which might be due to the fallout of various sources emissions and discharge effluents. The median concentration of all trace elements in samples has observed to decrease in the following order: Vehicular area> Industrial Area> Residential area However, standard permissible limits given in the table 2. Our measured values in percentage (%) have shown in the figure 1 (a-i).

Table No:2: Standard permissible limits for trace elements

Elements	certified air particulate SRM 1633a (%)	Detection limit (L.O.D. µg m-3)	The National Institute for Environmental Studies
Na	0.17	0.053	0.192±0.008
Mg	0.455	0.063	0.101±0.005
Al	14	0.047	0.33±0.02
Si	22.8	0.035	-
K	1.88	0.030	0.115±0.008
Ca	1.11	0.025	0.53±0.02
Mn	-	0.002	-
Fe	9.4	0.018	-
Pb	-	0.014	219± 9

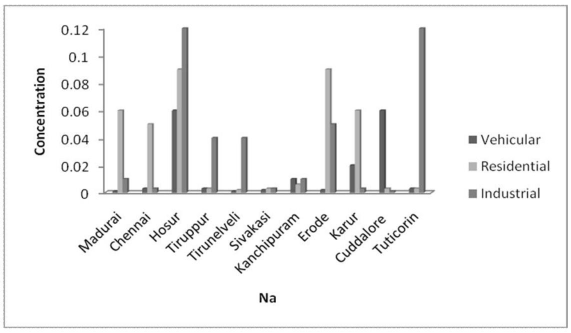


Figure: 1a

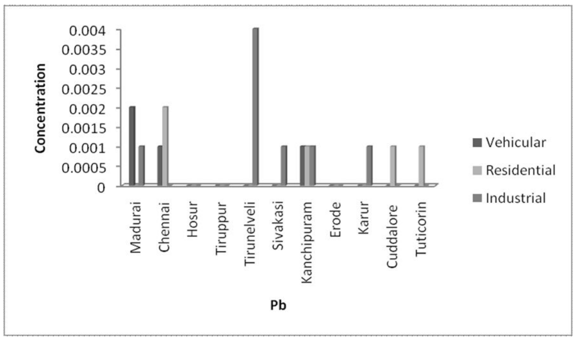


Figure: 1b

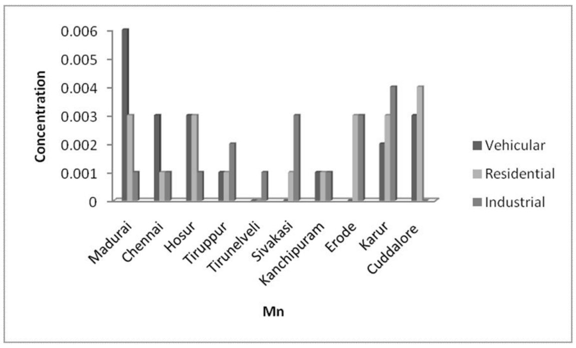
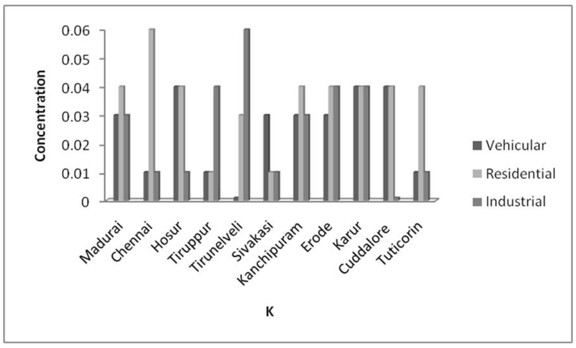


Figure: 1c



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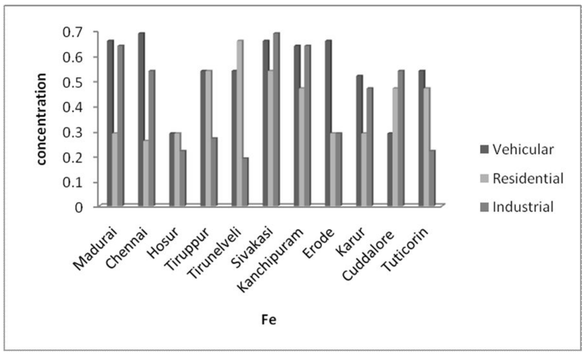

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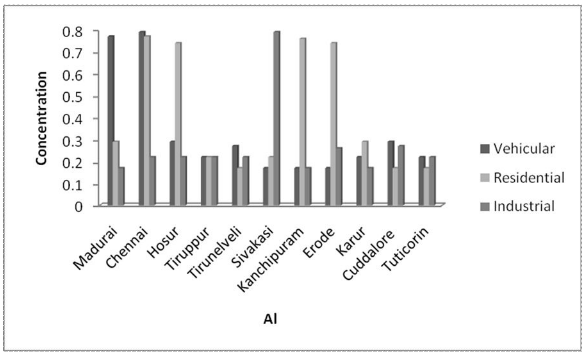

Figure: 1f

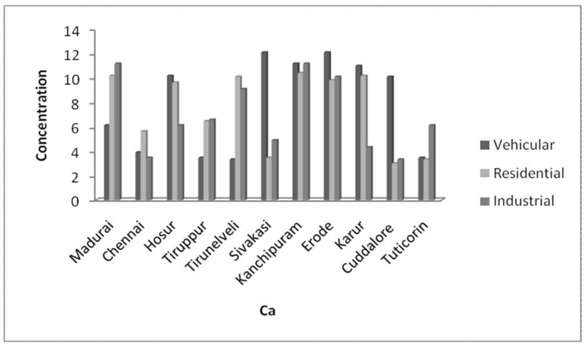

Figure: 1g

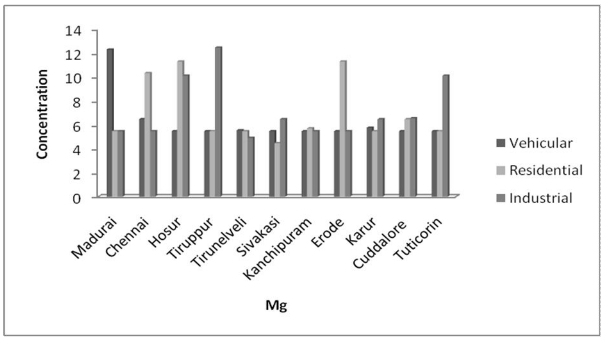

Figure: 1h

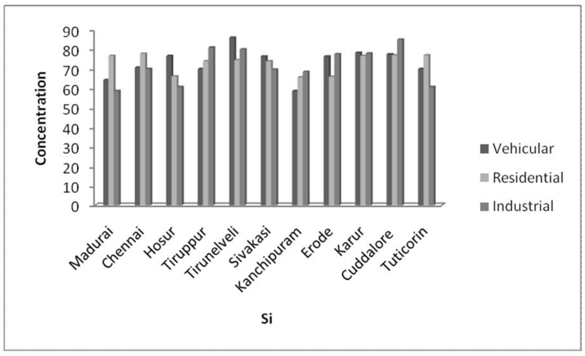

Figure: 1i

Figure: 1 (a,b,c,d,e,f,g,h,i) shows the various mass concentration (%) of Na, Pb, Mn, K, Fe, Al, Ca, Mg, Si respectively for various places in Tamilnadu.

Enrichment Factor (EF)

The contribution level of crustal and anthropogenic sources of trace elements is associated with PM and it was estimated here by calculating the enrichment factor (EF) for each metal particulate fraction based on the Earth's crustal mean abundance of the metals given by Sharma, 1994[29]. Each EF was calculated by using the following equation, in which Al is set as a reference element due to its distribution in coarse particles:

$$EF = \frac{\left(\frac{E}{Al}\right)_{Air}}{\left(\frac{E}{Al}\right)_{Crust}}$$

Where (*E/Al*) Air and (*E/Al*) Crust are the concentration ratios of element *E* to *Al* in airborne particles and the upper continental crust, respectively. Generally, if EF is close to unity for any element *E*, this element may have a crustal source. Whereas, if the EF value is over 10, the element will have a significant contribution from non-crustal sources[30, 31]. The element Pb has the lowest enrich value and Mg and Si have the high value enrichment in vehicular area. The lead element has lowest and calcium has highest enrichment value in residential areas. Similarly, lead has the lowest and Magnesium element has highest enrichment value in industrial area. The enrichment values of selected samples are shown in the figure 2 (a-c). Generally, trace element in air may cause serious health implications including high blood pressure, digestive problems, nerve and kidney disorders, memory and concentration problems, muscle and joint pain and also it affects the immune systems and sometimes it may lead to cancer.

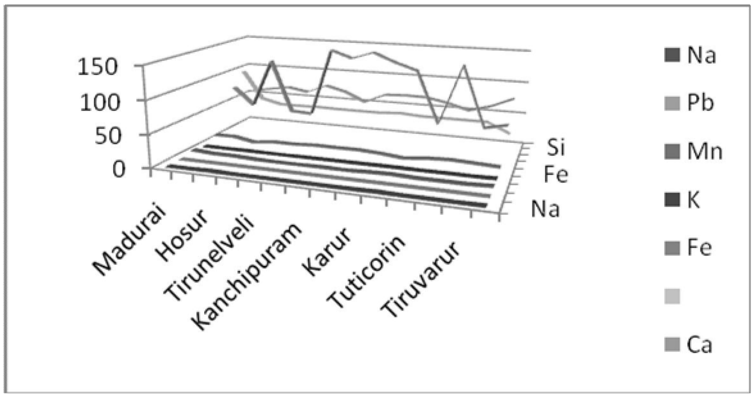

Figure: 2a Vehicular area

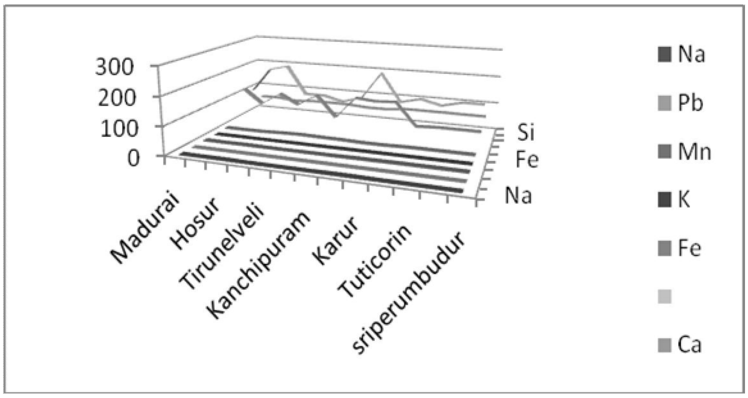

Figure: 2b Residential Area

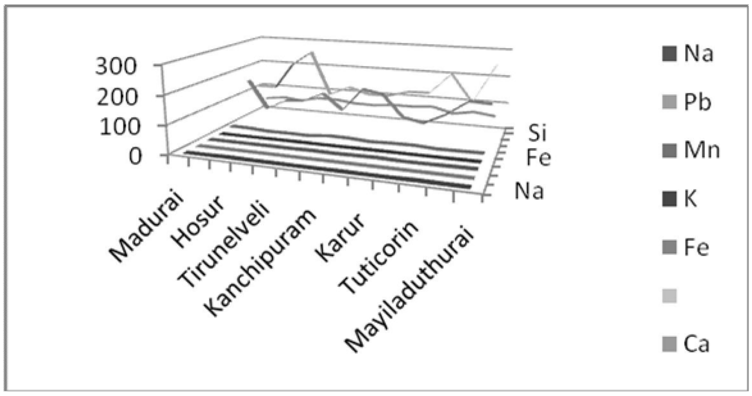

Figure: 2c Industrial Area

Figure 2: (a-c) shows the enrichment values of selected samples in different areas.

Pollution Evaluation method

The Composite index method (Nemerow Index) according to Liang et al. (2011)[32] was adopted to evaluate the quality of the air of the study area. The composite Index method was used in the single pollution index, which was more directly reflects the pollution of environmental indicators.

Single pollution index (P_i) = Ci/Cref

Where P_i is single pollution index, Ci represents to the mean concentration of trace element from at least five sampling and Cref indicates the evaluation of criteria value. The value of evaluation criteria has taken from a Major element composition of the Upper Continental Crust as given in wt. % from either surface exposures or glacial clays[33]. These values were first given by Condie 1993.The Nemerow Composite Index method was taken into account all the individual evaluation factors of the most contaminated elements.

Nemerow Index (P_s)= $\sqrt{(P_{ave}^2 + P_{max}^2)/2}$

Where P_{ave} is the average of the single pollution Index of all the metals and P_{max} is the maximum value of the single pollution index of all the metals. The quality of the environment is classified into 5 grades from Nemerow pollution index: (P_s<0.7, safety domain; 0.7<P_s<1.0, precaution domain; 1.0< P_s<2.0, slightly pollution domain; 2.0<P_s<3.0, moderately polluted domain; and P_s<3.0 seriously polluted domain[34]. The values of Nemerow Composite Pollution Index (P_s) were calculated for vehicular residential and industrial areas are presented in table 3.

The distribution of elements in the air in the industrial area had a higher P_s value than vehicular and residential area. In all areas Ca, Mg and Si were high potential ecological risks, whereas the other elements Na, Pb, Mn, K, Fe and Al were with low ecological risk. By considering, the potential ecological risk assessment of the study area, the common elements of the vehicular, residential and industrial areas had normal potential ecological risk and Si was found to be the main factor which causing the serious risk of the air. Wei et al. (2010)[35] reported significantly higher potential ecological risk of heavy metals of road dust in N W China. It's also important to know that the air of the study area, which was the translation of the high Nemerow Composite Index recorded in the study area due to the various operation/activities at the areas. This report is consistent with the finding of several authors. Liang et al. (2011), Wu et al. (2010), and Qiu (2010) reported that high potential ecological risks were recorded in their studies [32, 36, 37].

Table No. 3: The calculated values of Nemerow Composite Pollution Index (P_s) for vehicular residential and industrial areas

	Na	Pb	Mn	K	Fe	Al	Ca	Mg	Si	P_s
Vehicular Area P_i (n=14)	0.0050	2.52E-05	0.02551	0.00911	0.15612	0.01931	1.78352	0.89578	1.21384	1.30179
Residential Area P_i (n=13)	0.00933	3.17E-05	0.03517	0.01242	0.1211	0.02391	1.90851	3.00502	1.2251	2.18248
Industrial Area P_i (n= 13)	0.0129	3.62E-05	0.02747	0.01034	0.1189	0.02023	2.06857	3.22676	1.19136	2.34119

6. Physical and chemical characterization of particles

Field emission scanning electron photomicrographs with EDX shows deposited particles/elements observed on the selected samples general appearance have shown in figure 3 (a, b, c, d, e, f, g, h) Particles were present in a wide range of diameters up to 100 µm. Particles from anthropogenic sources, mostly emitted from high temperature in the combustion of processes are characterized by their irregular surfaces. This type of particles occurs not only an individual particles, but also in aggregate form as agglomerates of similar-sized particles and individual large particles carrying several smaller attached particles[38]. Both, fine and coarse particles were reported to be responsible for increased the human respiratory deceases. The chemical composition of the particles suggested that the most abundant particles were Si, Al, Fe, Mg, N, S, Ca, K, Cl particles rich in Al, Si, Ca, Fe and K. In general, air particles containing C, Al, Si, K, Ca; and particles liberated by the local industrial processes and vehicle concentration. Among the particles which are contained in the trace metals, the most abundant particles were in aggregate form, where Si is the major element associated with lower concentrations of V, Na, Cl, and Pb. According to the particle morphology and chemical composition indicated by the FESEM-EDX procedure, it may be suggested that the particles have deposited in the different areas, were mostly originated from the traffic or from the re suspended particles and possibly by the local sources.

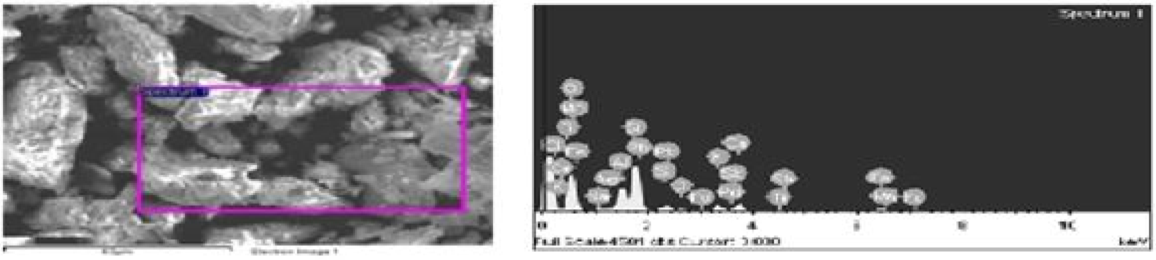

Figure: 3(a,b) shows the FESEM and EDX image of sample no 23

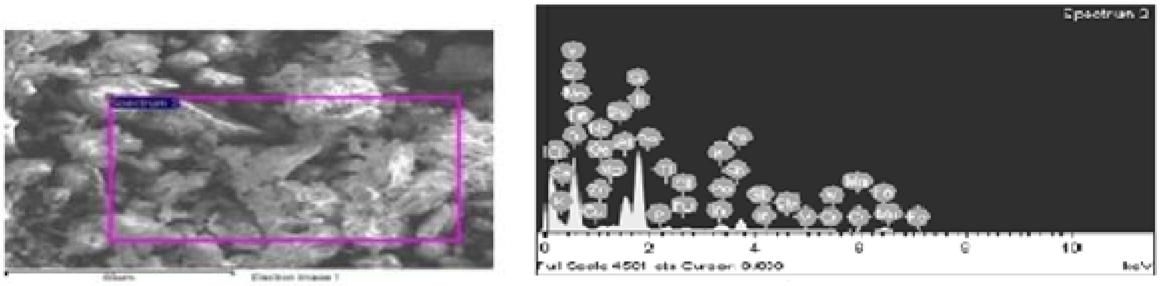

Figure: 3(c,d) shows the FESEM and EDX image of sample no 25

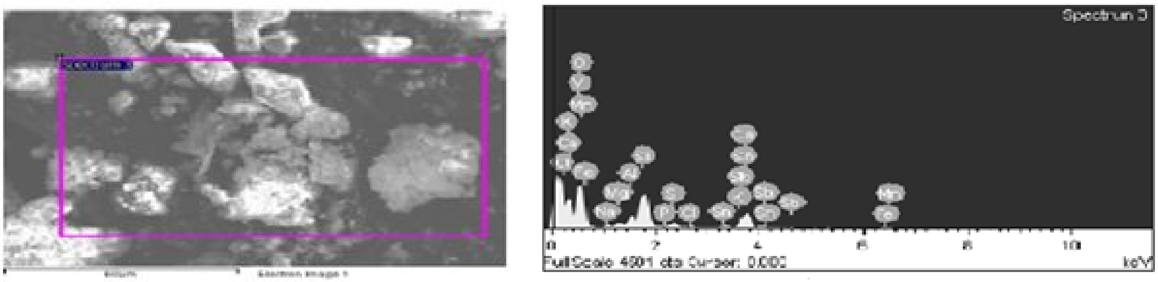

Figure: 3 (e,f) shows the FESEM and EDX image of sample no 26

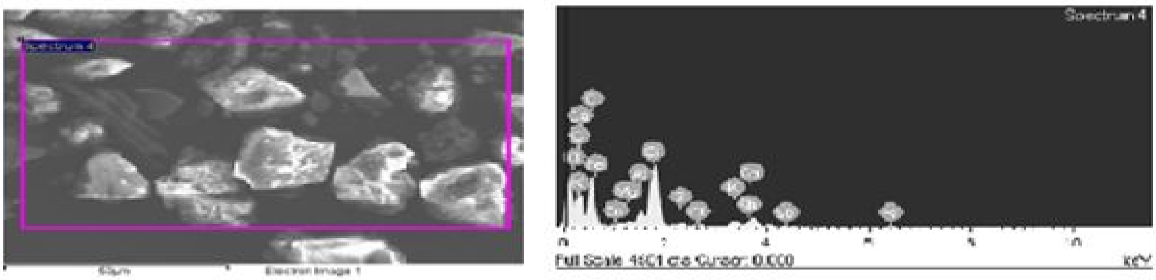

Figure: 3 (g,h) shows the FESEM and EDX image of sample no 70

Conclusion

The concentrations of elements in the samples of various places in Taminadu have discussed. Generally, Si, Mg and Ca have higher concentrations than other elements in residential, Industrial and vehicular areas. Vehicular area has a higher concentration than other areas. Contribution levels of anthropogenic sources for different elements have calculated by the enrichment factor. The element Pb has lowest enrichment and Si has highest enrichment in all the studied areas. Potential Ecological risk assessments have calculated with Nemerow Composite Index and single pollution index methods. The element Si has found the main factor in causing the serious ecological risk of the air. From FESEM-EDX the different elements have identified and samples have more irregular aggregates which composed mostly of the various elements. This study concludes that the concentration of Mg, Ca and Si in the residential, vehicular and industrial area in Tamilnadu are the result of air pollution; the vehicular activities are engaged with transport and anthropogenic activities. The trace elements, especially Si, Mg and ca have great potential in the area of ecological function. The High mobility of Si, Mg and Ca are coupled with its heavy load in the air pose a serious environmental concern.

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