

ENVIRONMENTAL MAGNETIC AND GEOCHEMICAL CHARACTERIZATION OF ATMOSPHERIC PARTICULATE DUST IN VISAKHAPATNAM CITY, INDIA: A NEW TECHNIQUE OF TEXTILE MONITORING OF POLLUTION

Ravichandra Kammula^{1,2}, Srinivasa Rao Goddu¹, Prasada Rao P.V.V.³, Nathani Basavaiah⁴

¹Department of Physics, GITAM Institute of Technology, GITAM University, Visakhapatnam – 530 045, Andhra Pradesh, India

²Department of Geosciences, Dr. B.R. Ambedkar University, Etcherla-532 410, Srikakulam (District), Andhra Pradesh, India

³Department of Environmental Sciences, College of Sci. and Tech., Andhra University, Visakhapatnam – 530 003, Andhra Pradesh, India

⁴ Indian Institute of Geomagnetism, Kalamboli, New Panvel, Navi Mumbai – 410 218, India

Abstract—Magnetic analysis is done on 76 suspended particulate dust samples to detect hazardous levels in Visakhapatnam city (also known as Vizag) and distinguish between the areas affected by traffic and industrial pollution. A new method of sampling (Textile sampling method) is used to collect the suspended particles (SP). The correlation coefficient between weight of the SP obtained from textile method and classical High Volume Sampler (HVS) is found to be $R=0.99$. The variations of magnetic susceptibility (k) distinguish pollution source within the city and is mainly controlled by the local pollution effect. The percentage frequency dependence susceptibility (k_{fd} %) values range between 3 and 9% indicating high ultrafine percentage of magnetic grains in the Particulate Matter (PM) at various sampling sites. Higher k_{fd} % values are found at heavy traffic areas pointing out that vehicular emissions contain a higher percentage of ultrafine superparamagnetic (SP) grains than the SP in industrial zones. Magnetic mineralogy of the suspended dust is dominated by a magnetite-like phase. Hysteresis parameters measured for some samples are typical for pseudo-single domain magnetite. Inductively coupled plasma mass spectrometry (ICP-MS) analysis on few selected samples indicates very high concentrations of heavy metals Mn, Zn, Pb, Cr and Cu which are far beyond the permissible limit. The correlation of k_{fd} % with Cr and Ni is well expressed at a significance level of $P=0.05$ in our results. Also the correlation of magnetic susceptibility k with Cr, Mn and Ni is good indicating that the magnetic parameters respond to spatial distribution of heavy metals. The variation of k_{fd} % is interpreted as the relative degree of human health hazardous zones.

Index Terms—Cloth samplers, environmental magnetic properties, suspended particulate matter, air pollution, heavy metals.

I. INTRODUCTION

Air pollution is a serious problem in developing countries with fast growing industrialization and poor governmental regulation on environmental issues. These “Fine Suspended Particles” (FSP) vary in size, composition and origin. They originate mainly from combustion processes in vehicles, power plants and industries. FSP between $2.5 \mu\text{m}$ to $10 \mu\text{m}$ contain both natural dust materials and transient dust from roads and industries. FSP ranging from 100 nm to $2.5 \mu\text{m}$ mainly stem from gas to particle conversion, combustion and re-condensed organic and metal vapours [1]. FSP are considered as a threat for human health due to their chemical intolerance for the organism and their penetration into the human respiratory system. Many biological, geochemical, and toxicological studies have proved that FSP matter play a vital role for many cardiopulmonary diseases and lung cancer [2].

Most studies of monitoring air pollution are done in small scales; they are expensive and time consuming. With the existing instrumentation it is almost impossible to get a spatially comprehensive control over the situation especially in large cities of developing countries with poor environmental laws, and unprecedented vehicular and industrial emissions. It is therefore important to explore new methodologies to improve the spatial characterization of FSP matter. The application of fast and cost-efficient methods to detect and monitor hazardous suspended dust in fast growing cities of developing countries is applauded.

During the past two decades, magnetometry was explored as a tool to detect environmental pollution in high-resolution scales with relatively low costs and in less time [3-9]. One reason for progress may be the ease of access to materials including fly ashes, roads and road-side dusts, soils and other materials [3-9]. Magnetic properties are dependent on composition, grain size and source of the material, which enables a differentiation of the magnetic signal caused by natural (lithogenic, pedogenic) and anthropogenic (pollution) origins [6, 10-12]. Magnetometry turned out to be especially suitable for detecting heavy metal pollution caused by fly ash, it is based on the fact that during combustion processes both toxic heavy metals and strong magnetic particles (iron oxides or sulphides) are released into the environment [8-9, 13-14]. Measurement of the ferrimagnetic concentration and screening enhanced magnetic signals in polluted urban top-soils, sediments, and road dusts, provides a qualitative estimate of the degree of environmental pollution [8, 15-17].

The present study attempts to use magnetometry, notably magnetic susceptibility, to detect hazardous levels of particulate matter in Visakhapatnam city (also known as Vizag) and to distinguish between areas affected by road traffic and industrial pollution by studying ultrafine dust directly collected on cloth samplers. Previous attempts of studying the degree of environmental pollution using magnetic methods targeted tree leaves and pine needles as natural dust samplers [18-19]. Unavailability of tree leaves of the same species in the study region, and unpredicted rains washing out the dust, limit the possibility for such kind of bio-monitoring. To overcome this limitation [20], we propose an alternative

method using simple passive samplers in order to study environmental pollution in city areas (Textile monitoring method). So far unexplored and unexploited cloth receptors in India are being used to provide a nonmagnetic background for collecting atmospheric dust, and cloth samplers with a nap help to trap and retain ultrafine atmospheric particulate pollutants. Further consistency is achieved by using the same cloth-type with a nap of the same textile medium across the city areas.

II. STUDY AREAS

Visakhapatnam city covers approximately 110 sq. Km with 2.05 Million inhabitants and is one of the fastest developing cities in India. The growth of population is ~0.8 Million during the last decade. The city extends towards the west with the Bengal Sea in the east (Fig. 1a). The Simhachalam Hills and Dolphin Nose (Yarada Hills) flanks it in the north and south, respectively. The topographic shape of the city appears to be like a bowl. The geology of the region is represented by gneisses of Archean age belonging to the Eastern Ghats mobile belt. Quaternary deposits consist of red bed sediments, laterites, pediment fans, colluvium, alluvium and coastal sands. The region has a sub-humid to semi-arid monsoonal climate, with an average annual temperature of 32°C and an average annual rainfall of 950 mm (mostly coming through the northeast monsoon air masses). The wind direction is towards north in summer and turns south in winter. Major industries mainly deal with production of iron material, smelting, petroleum processing, ship building, port and transport activities, they are located in the south while residential and commercial areas are distributed in the north and north-east of the study area. Main roads run through these areas roughly parallel to the coastal road, and meet the highway at different locations (Fig. 1a).

III. SAMPLING PROCEDURE

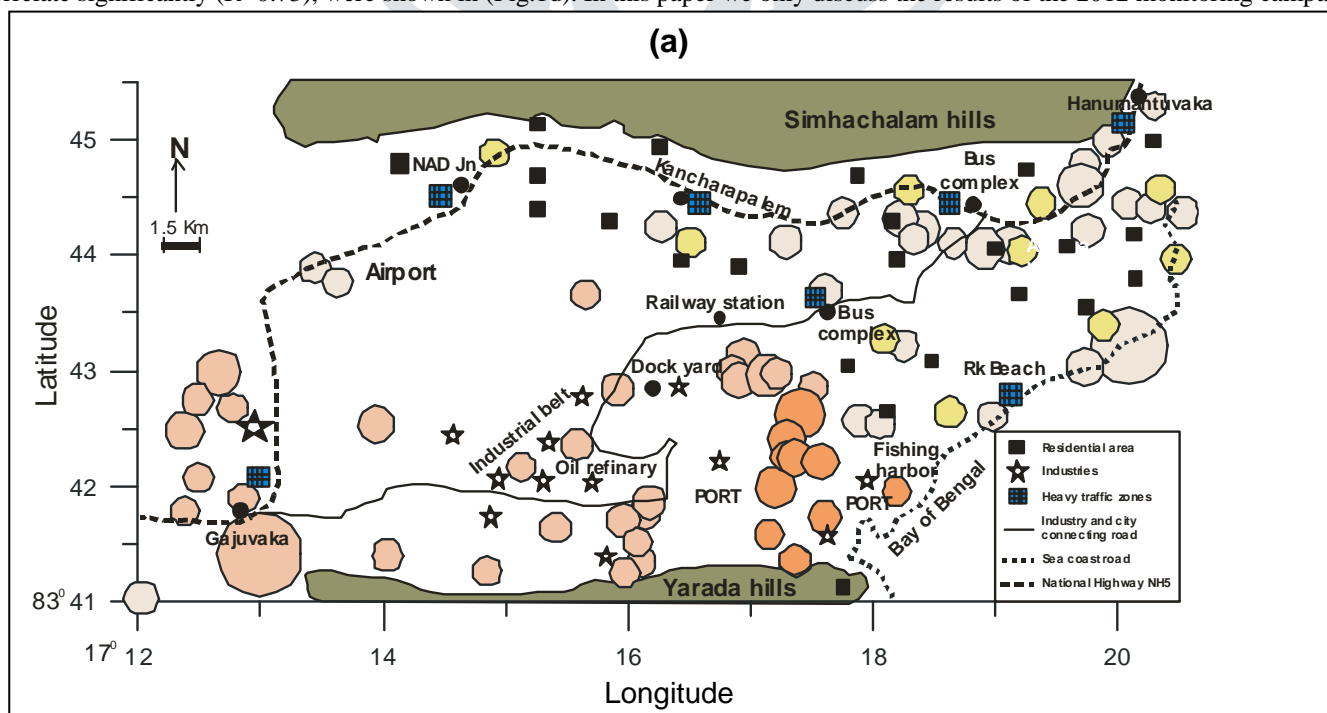
Nap textile medium raised (fuzzy) surface rug cloth was cut into pieces of equal dimension (10x10 cm). The corner of the cloth is pierced with the thread for tying it at the desired height. The cloths were weighed with a digital balance (accuracy 10 mg) in the laboratory and later hanged as dust collectors ca. 2.5 m above the ground level at 110 locations in the city. The hanged cloth flutters in the wind with both sides exposed and its thickness do not allow itself to fold. The coordinates and heights of the sample locations were determined by a Garmin GPS 78s. We choose sampling points avoiding direct exposure to roads, heavy traffic, known contaminated sites, and industries. To avoid direct influence from rain, we placed the cloths in sheltered positions.

IV. RESULTS

We left the cloths exposed to air for a period of two months i.e. from end of April to end of June 2012. Out of the 110 cloths installed, we could recover 76 from their respective locations. After recovery, we sealed the cloths in pre-weighted self-locked plastic bags and determined the total weight of the dust trapped by the cloths in the laboratory. The spatial distribution of the dust weight provides first information of the dust load in the study area (Fig.1a).

In order to check the suitability of the cloth samplers, we established traditional high volume samplers (HVS) in parallel at seven stations. The HVS sucked the suspended particles into a quartz filter with a rate of 1.2 litre/min for four hours during the day (between 10 am to 2 pm) during two subsequent days. The results of the two-days average dust weight of the HVS correlates very well with the dust weight collected by the cloth samplers (Fig.1b) with a correlation coefficient of $R=0.99$ ($N=6$), at one station the cloth could not be recovered or $R=0.85$ ($N=5$), excluding the result with the exceptional high value. The bivariate relationship between the sampling site elevation above sea level and the dust weight collected by the cloth samplers (Fig.1c) indicates a slight tendency of relatively higher dust weights at lower elevations of the city.

Further checking the reliability of the cloth sampler methodology, we performed a second monitoring campaign during April 2013 (four weeks), and increased the spatial resolution by installing cloth samplers at 160 sites (117 recovered). The spatial distributions of magnetic susceptibility for this campaign looks very similar than for the first one in 2012. Results from ten sites, for which data from both campaigns were obtained correlate significantly ($R=0.73$), were shown in (Fig.1d). In this paper we only discuss the results of the 2012 monitoring campaign.



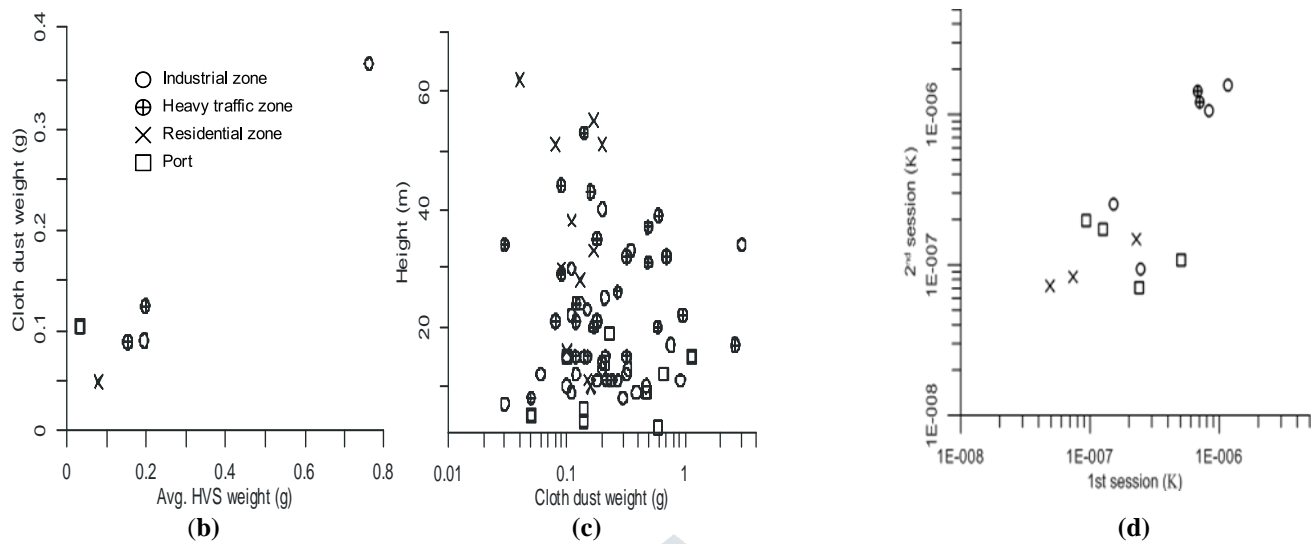


Figure 1.(a) The spatial distribution of dust weight in the area of proposed study,(b) Suspended dust weight correlation between High Volume Sampler HVS data and cloth samplers data,(c) Bivariate relation plot between sampling site elevation and collected dust, (d) Correlation between the spatial distributions of magnetic susceptibilities obtained in the first and second campaigns. Symbols of open circle, plus in circle, cross and open square represent samples of industrial, heavy traffic, residential and port areas.

MAGNETIC MEASUREMENTS

The samples were shaped as cylinders along with the plastic bags using a cellotape for performing magnetic measurements at the magnetic laboratory of Tübingen University, Germany. Susceptibility (k) and its frequency dependence were measured with an MFK1-FA instrument (AGICO) at frequencies of 976 and 15,616 Hz. The mass specific susceptibility (X) was found to have negative correlation with dust weight ($R = -0.34$), susceptibility (k) and dust weight shows positive correlation of $R = 0.68$, correlation between (X) and (k) is $R = 0.05$. The reason for this may be due to dust from different sources. In this paper we have not normalized the magnetic results to their dust weights. The k values of the 76 samples vary between wide ranges from 2.35×10^{-8} to 51.6×10^{-8} , except one sample having a very high value of 158×10^{-8} . Categorization into four groups of industrial, heavy traffic, residential and port demonstrates best the spatial distribution of k (Fig.2a). Pink, light pink, yellow and orange categories contain 28, 27, 10, and 11 samples, respectively. Their median values of k ($\times 10^{-8}$) are 15.45, 11.0, 5.79, and 14.8, respectively. Higher k values are predominant in the industrial and port zones, while medium to low values occur near main roads and in residential. The percentage frequency dependence ($k_{fd}\%$) was calculated with the formula $100 \cdot (k_{lf} - k_{hf}) / k_{lf}$. The $k_{fd}\%$ value indicates the relative contribution of ultrafine superparamagnetic (SP) particles of ferro(i)magnetic phases. For plotting the spatial distribution of $k_{fd}\%$ we again used four categories in Figure 2b. The median values for these categories are 5.7, 6.0, 7.2 and 5.6%. About 80% of the samples have $k_{fd}\%$ values $> 5\%$, and the highest values are observed in areas of heavy traffic zones, an oil refinery and smelting industries.

The intensity of isothermal remanent magnetization (IRM) was measured with a Molspin spinner magnetometer. An MMPM9 pulse magnetizer was used for imparting IRMs. The IRM at 2.5 T is regarded as saturation IRM (SIRM). After imparting an SIRM, a backfield of -300 mT was applied to determine the reverse IRM-300. From these experiments, we calculated a simplified S-ratio by $IRM-300/SIRM$. The S-ratio estimates the relative proportions of ferrimagnetic phases (magnetite, maghemite and greigite) and harder antiferromagnetic ones (hematite and goethite). The parameter "hard IRM" (HIRM), which is a measure for the absolute content of harder antiferromagnetic materials, was calculated by $0.5 \cdot (SIRM + IRM - 300mT)$. The spatial distribution of S-ratios, categorized into four groups (Fig.3c), shows median category values of 0.89, 0.86, 0.76 and 0.75. The correlation between magnetic concentration parameters (k , SIRM, HIRM) and the S-ratio (Figs. 3a-c) as well as $k_{fd}\%$ is moderate (Fig.3d). However, the spatial distribution map of S-ratios (Fig.3c) seems to indicate that the low and intermediate x values in the north eastern part (Fig.2a) are associated with a higher relative content of a harder magnetic fraction.

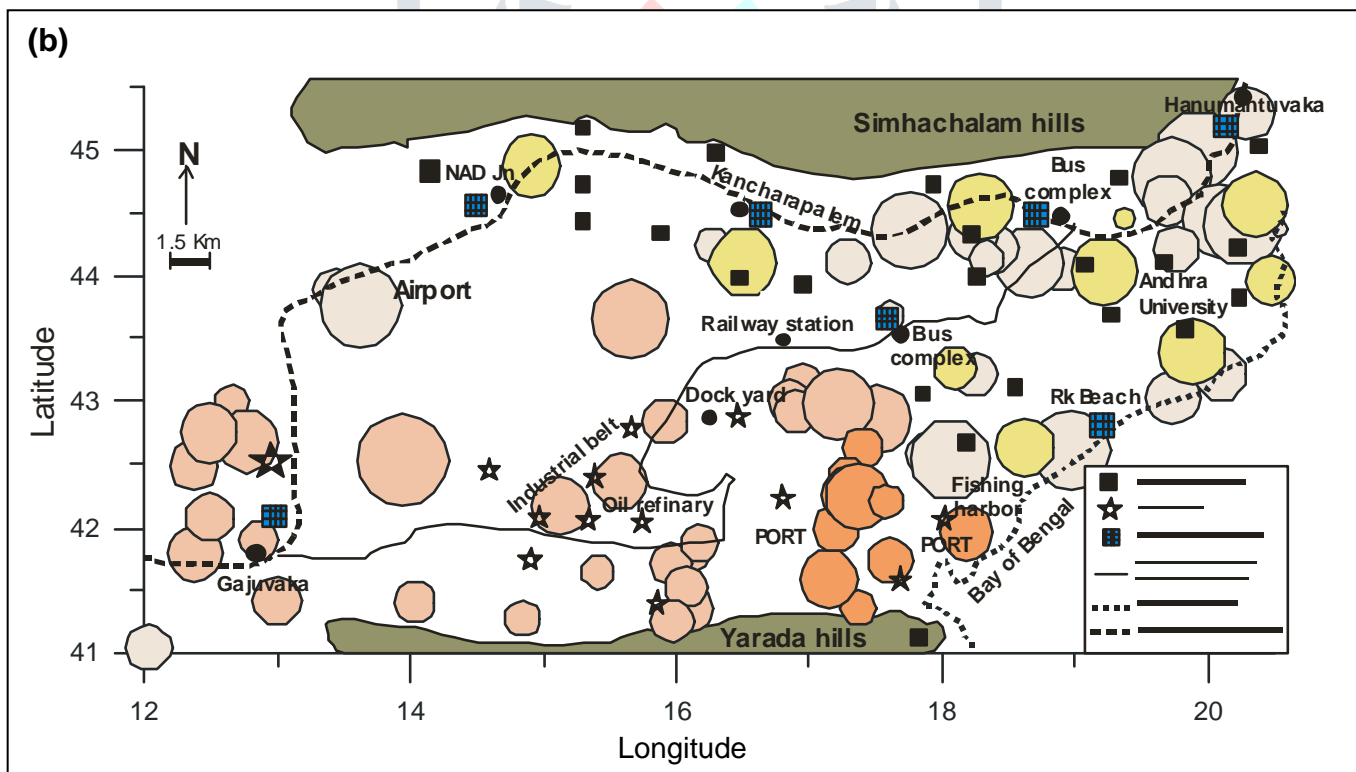
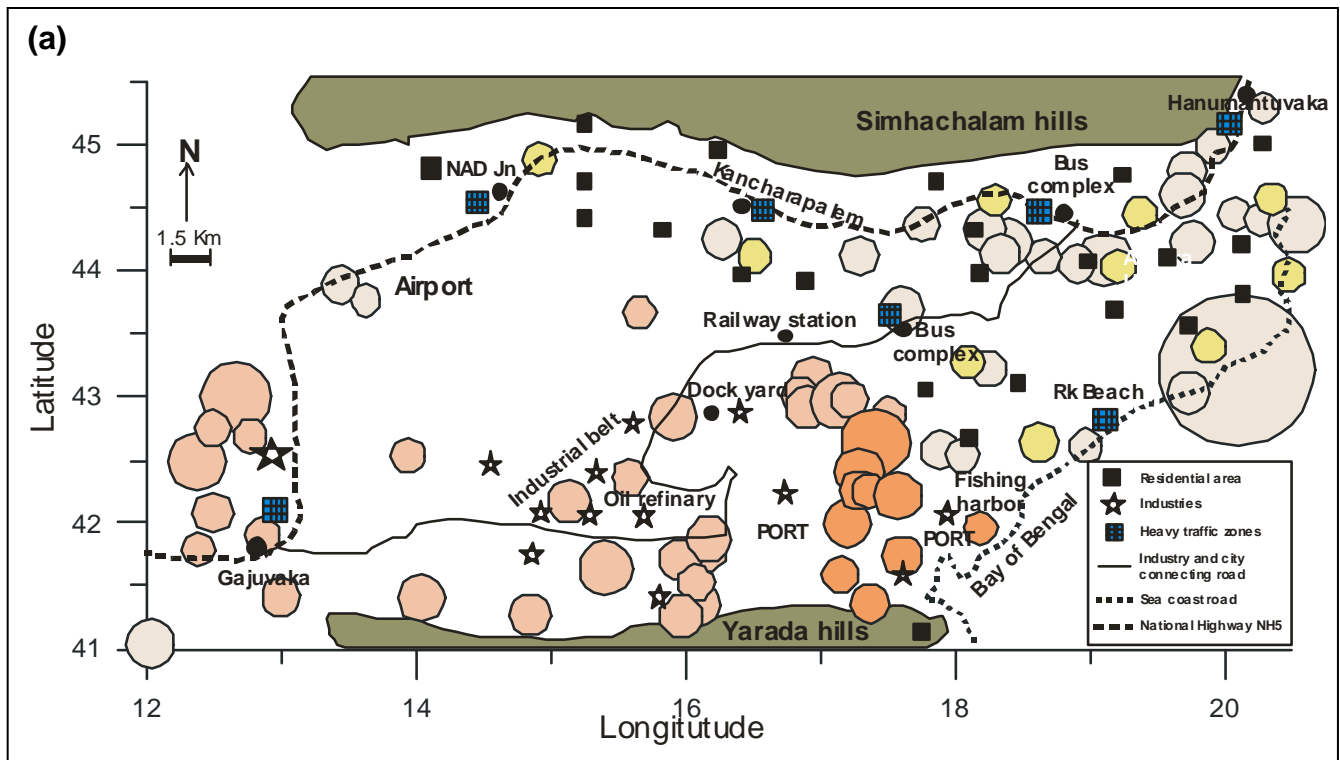


Figure 2. Spatial distribution of (a) magnetic susceptibility (k) and (b) percentage frequency dependence susceptibility $k_{fd}\%$ of dust loaded textiles. The dots represent different sampling points. The four categories of pink, light pink, yellow and orange contain 28, 27, 10, 11 samples, respectively with their median values of magnetic susceptibility ($k \times 10^{-8}$) of 15.45, 11.0, 5.79 and 14.8, respectively.

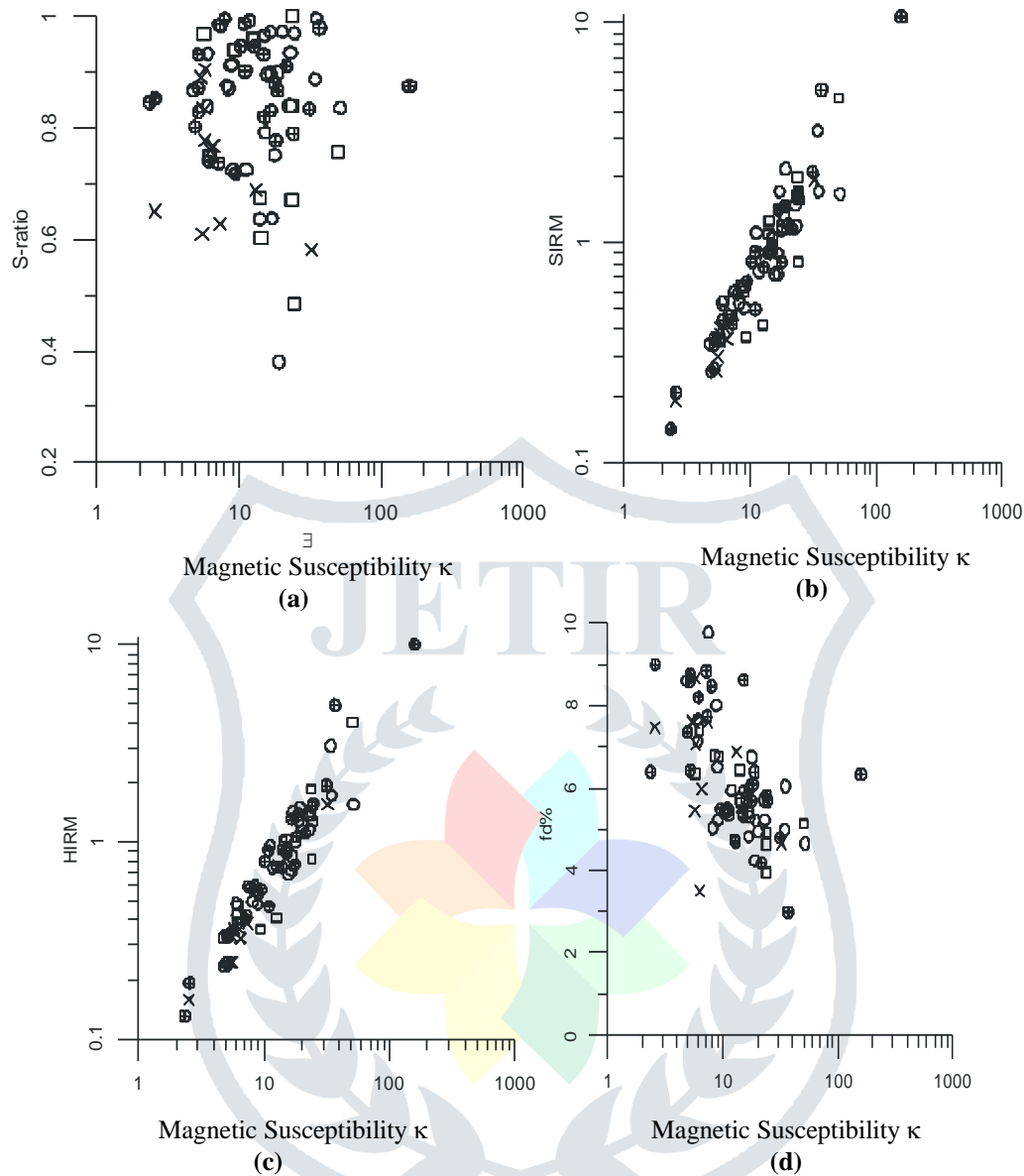


Figure 3. Cross plots of (a) S-ratio vs. magnetic susceptibility κ , (b) Saturation isothermal remanent magnetization $SIRM_{@2.5T}$ vs. magnetic susceptibility κ , (c) Hard remanent magnetization HIRM vs. magnetic susceptibility κ and frequency dependent susceptibility $kfd\%$ vs. magnetic susceptibility κ . Symbols of open circle, plus in circle, cross and open square represent samples of industrial, heavy traffic, residential and port

Based on the high and low κ values we selected a few samples for identifying the magnetic mineralogy by thermomagnetic runs of κ . By gently patting the cloths, we obtained few mg of dust for measuring κ -T curves (Fig.4). For running the measurements from -196°C to 700°C we used a KLY-3 Kappabridge and a CS-3 temperature unit. The Verwey transition and a Curie temperature $\sim 580^{\circ}\text{C}$ are observed in almost all samples that confirms the presence of magnetite. An increase of κ in the heating curves starting at $\sim 500^{\circ}\text{C}$, is caused by secondary magnetite formation at elevated temperature. The gradual increase of κ with temperature in the low-temperature curves may indicate the presence of pseudo-single domain (PSD) and single domain particles (SD) of magnetite. However, also larger multi-domain (MD) particles of magnetite are indicated by the peak at around -140°C that is caused by the isotropic point of magneto crystalline anisotropy [22].

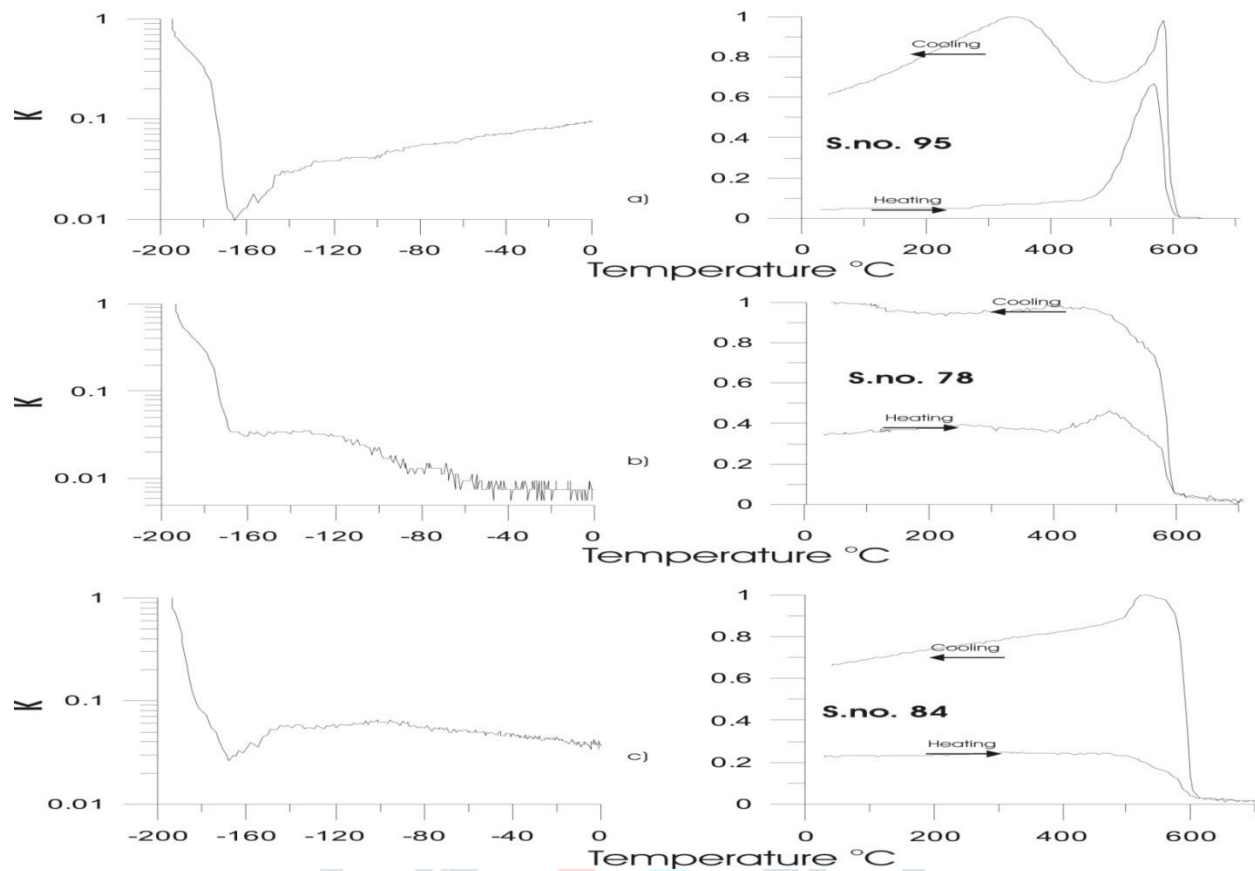


Figure 4. Thermo-magnetic analysis of low and high temperature variations of magnetic susceptibility. The direction of heating and cooling is indicated by arrows. The collected cloth sample numbers (S.no.): 84- Hindusthan Shipyard Entrance, 78- Andhra Petro Chemicals Limited (APCL) and 95- Port Area.

Moreover, we performed hysteresis measurements using an alternating gradient force magnetometer Micromag 2900 (Princeton Measurements). From slope corrected curves, we determined the remanent saturation magnetization (M_{rs}), saturation magnetization (M_s), coercivity (H_c), and the coercivity of remanence (H_{cr}) from additional backfield measurements. The two hysteresis ratios M_{rs}/M_s vs. H_{cr}/H_c plotted in the Day-diagram (Fig.5) [23, 24], plot over a close range of the PSD field. In combination with the thermomagnetic curves and the variable kfd% values, we assume that this represents a mixture of a large grain size spectrum extending from MD to SD range and even the SP range. Also, the plot of weight of suspended dust (g) vs. susceptibility (k) over four different regions is shown in Figure 6.

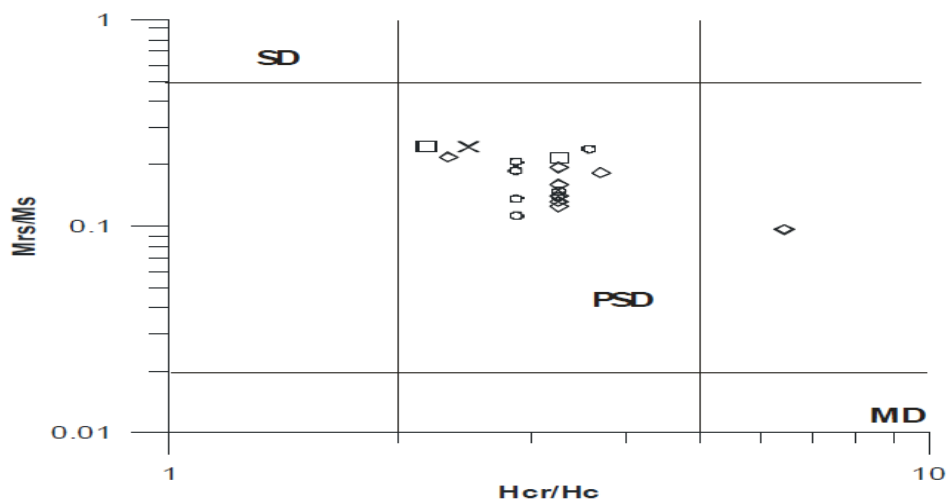


Figure 5. Day-diagram of the ratios M_{rs}/M_s and H_{cr}/H_c for textile samplers. Grain size boundaries for Single Domain (SD)–Pseudo Single Domain (PSD)–Multidomain (MD) are according to Dunlop [24]. Symbols of open circle, plus in circle, cross and open square represent samples of industrial, heavy traffic, residential and port areas respectively.

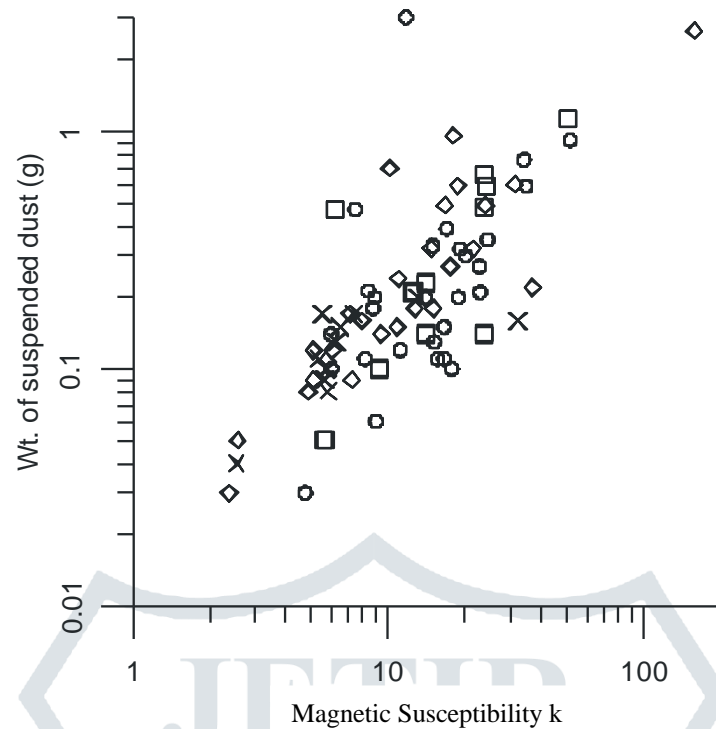


Figure 6. Magnetic susceptibility (k) as a function of the weight of suspended dust (g) (1st campaign data). Symbols of open circle, plus in circle, cross and open square represent samples of industrial, heavy traffic, residential and port areas respectively.

GEOCHEMICAL ANALYSIS

In order to test the correlation between the magnetic signal and heavy metals concentrations, we selected nine samples for geochemical analysis. Criteria for sample selection were the representation of sites with (1) high and low magnetic concentration values, and (2) different locations in the study area. The samples were digested in 1.5 ml HCl and 0.5 ml HNO₃ in Savillex beakers on a hot plate (80°C) for 24 hours. The dissolved samples were dried, taken up in 1 ml HNO₃ and dried again. This step was twice repeated to ensure full conversion to nitride form. Finally, the dry sample was taken up in 2% HNO₃, centrifuged, and the supernatant was diluted for measurement. Metal concentrations of Cr, Mn, Co, Ni, Cu, Zn, Cd and Pb were determined using a ThermoFisher Scientific iCAP Qc quadrupole ICP-MS instrument at Tuebingen University, Germany. A mixture of single element standards in three different concentrations was used for calibration and tested against USGS reference material SCo-1. All samples contained relatively large amounts of fine textile fibres and the measured values given in (Table 1) have ca. 7% uncertainty, which results from analytical and weighing errors, and from dilution. A correlation coefficient matrix of magnetic parameters and heavy metal contents is given in Table 2.

Table 1. Results of ICP-MS done on suspended dust (in ppm) from selected samples. Symbols # and * indicate 2-3 mg and 0.42 mg dust that was used for analysis due to paucity.

Sample ID	Cr	Mn	Co	Ni	Cu	Zn	Cd	Pb
I 123*	138.4	1,155.0	9.9	31.8	97.2	332.1	8.8	266.3
I 78#	75.4	705.4	8.8	38.3	65.7	317.2	6.0	392.1
I 95	66.3	1,980.8	12.1	37.2	62.4	512.8	0.7	419.7
P 02	57.4	981.8	25.7	36.0	580.7	576.3	1.3	70.0
P 60	74.6	1,536.4	17.1	50.8	183.3	415.0	1.3	250.1
R 49#	240.0	994.6	16.5	120.7	74.7	156.7	0.5	108.7
T 17	74.4	893.2	11.9	46.0	115.0	680.1	5.3	73.7
T 67	63.6	962.5	9.7	32.8	89.8	294.6	0.8	80.5
T 84#	91.3	958.9	8.6	139.0	83.5	238.3	1.6	76.0

V. DISCUSSION

Anthropogenic air pollution in Visakhapatnam city has several origins but the results from a specific sampling station could reflect local pollution sources in particular. The higher and lower absolute weights of the suspended dust obtained at different areas may indicate tentative hotspots of pollution in the study area. In general, suspended particles that evolved from natural processes such as wind erosion are coarser than those that originated from combustion processes like vehicular and industrial emissions [25, 26]. The highest amounts of collected dust mass were found at the port, in the industrial area and in heavy traffic zones as expected. The results of susceptibility (k) ($R=0.78$) and its frequency dependence ($R=-0.66$), and the collected dust weight (Fig.1b) between the HVS and our cloth samplers correlate well. These observations show

that the applied simple sampling technique is reliable. Besides providing the opportunity for a time-saving and spatially high-resolution monitoring, the cloth samplers have also the advantage of collecting only finer particles because coarser ones will easily drop down due the gravity effect.

Table 2. Correlation matrix between magnetic and geochemical parameters. Correlations of the standardized data set. Underlined correlations are significant at $p=0.05$. Number of samples $N=10$.

	Cr	Mn	Co	Ni	Cu	Zn	Cd	Pb	Dust Wt. (g)	LF Sus	% FD Sus	SIRM 2.5T	IRM 0.3T	S-Ratio	HIRM	Height (m)
Cr	1.00	-0.16	0.06	0.56	-0.26	-0.51	0.05	-0.13	-0.39	-0.56	<u>0.71</u>	-0.42	-0.38	-0.06	-0.25	0.09
Mn	-0.16	1.00	0.06	-0.20	-0.12	0.16	-0.35	0.50	<u>0.76</u>	0.53	-0.04	0.50	0.44	-0.16	0.02	-0.14
Co	0.06	0.06	1.00	-0.08	<u>0.88</u>	0.42	-0.31	-0.26	-0.13	-0.07	0.00	-0.12	-0.17	-0.30	0.01	-0.40
Ni	0.56	-0.20	-0.08	1.00	-0.23	-0.52	-0.34	-0.40	0.33	-0.48	0.42	-0.29	-0.22	0.31	-0.13	-0.15
Cu	-0.26	-0.12	<u>0.88</u>	-0.23	1.00	0.49	-0.14	-0.33	-0.21	0.07	-0.31	-0.02	-0.06	-0.15	0.21	-0.46
Zn	-0.51	0.16	0.42	-0.52	0.49	1.00	0.20	0.04	0.46	0.18	-0.18	-0.03	-0.08	0.00	-0.25	0.05
Cd	0.05	-0.35	-0.31	-0.34	-0.14	0.20	1.00	0.29	-0.19	-0.20	0.07	-0.31	-0.29	0.12	-0.16	0.18
Pb	-0.13	0.50	-0.26	-0.40	0.33	0.04	0.29	1.00	0.56	0.39	-0.14	0.26	0.22	-0.01	-0.11	-0.20
Dust Wt. (g)	-0.39	<u>0.76</u>	-0.13	-0.33	-0.21	0.46	-0.19	0.56	1.00	0.59	-0.16	0.44	0.40	0.16	-0.17	0.02
K	-0.56	0.53	-0.07	-0.48	0.07	0.18	-0.20	0.39	0.59	1.00	<u>-0.81</u>	<u>0.95</u>	<u>0.93</u>	0.37	<u>0.67</u>	-0.44
% FD Sus	<u>0.71</u>	-0.04	0.00	0.42	-0.31	-0.18	0.07	-0.14	-0.16	<u>-0.81</u>	1.00	<u>-0.78</u>	<u>-0.78</u>	-0.44	<u>-0.80</u>	0.56
SIRM 2.5T	-0.42	0.50	-0.12	-0.29	-0.02	-0.03	-0.31	0.26	0.44	<u>0.95</u>	<u>-0.78</u>	1.00	<u>0.99</u>	0.40	<u>0.80</u>	-0.47
IRM 0.3T	-0.38	0.44	-0.17	-0.22	-0.06	-0.08	-0.29	0.22	0.40	<u>0.93</u>	<u>-0.78</u>	<u>0.99</u>	1.00	0.49	<u>0.82</u>	-0.47
S-Ratio	-0.06	-0.16	-0.30	0.31	-0.15	0.00	0.12	-0.01	0.16	0.37	-0.44	0.40	0.49	1.00	0.40	-0.44
HIRM	-0.25	0.02	0.01	-0.13	0.21	-0.25	-0.16	-0.11	-0.17	<u>0.67</u>	<u>-0.80</u>	<u>0.80</u>	<u>0.82</u>	0.40	1.00	-0.57
Height (m)	0.09	-0.14	-0.40	-0.15	-0.46	0.05	0.18	-0.20	0.02	-0.44	0.56	-0.47	-0.47	-0.44	-0.57	1.00

The strong positive correlation ($R=0.94$) shown in Fig.3b between SIRM and magnetic susceptibility (k) suggest that the magnetic concentration signal is dominated by the ferrimagnetic fraction. The k values in our study area are higher closer to the industrial and port zones, whereas medium to low values occur at traffic zones and residential areas. This could indicate that the susceptibility values are controlled by different local sources of pollution or that there is a distance effect to larger sources. The correlation coefficients between the susceptibility (k) and weight of the suspended dust are 0.21, 0.91, 0.78, 0.83 at industrial area, heavy traffic, residential and port zones respectively.

Lower S-ratio values are predominant at heavy traffic zones and also in the industrial zone Fig.3a. HIRM is an indicator of the total concentration of weakly magnetic high coercivity imperfect antiferromagnetic minerals. The positive correlation of HIRM with k ($R=0.85$) shown in Fig.3c and this indicates that when susceptibility is higher also the absolute contribution of the hard magnetic fraction is increased. R value of HIRM with k is relatively low at industrial zone ($R=0.77$) and higher at heavy traffic zones ($R=-0.96$), residential areas ($R=0.97$) and port ($R=0.94$). There is no correlation between k and the S-ratio for the overall data Fig.3a. However, the R values are quite different when separating the samples from the industrial area ($R=0.02$), heavy traffic zones ($R=-0.01$), residential areas ($R=-0.13$) and port ($R=-0.22$). Thus this may indicate that the magnetic signal is influenced by the different sources of pollution with a distance effect in the study area. Traffic related dust seems to contain a larger fraction of hard coercive FSP than in industrial areas, while the dust in the residential areas is containing relatively more magnetite. The latter seems to be related the influence of the industrial emissions that influence the atmospheric FSP over longer distances than traffic emissions. In high traffic zones, both contributions are important, leading to the observed increase of both absolute k values and HIRM as well as a complex relationship of the S-ratio with k . The decreasing trend of $kfd\%$ with higher k values (Fig.3d) indicates that the traffic emissions contain a larger fraction of ultrafine SP particles.

Studies have shown that the heavy metals Cr, Cu, As, Co, Cd, Pb, Zn and Ni are mainly associated with the industrial and automobile sources [5, 27, 28]. In our study the average values of Cr, Mn, Co, Ni, Cu, Zn, Cd and Pb are found to be 100.8, 1079.0, 12.9, 60.2, 146.1, 388.5, 3.1, 185.9 ppm, respectively. These observed values are well above the permissible limits. Gajghate et al. [29] have studied the trace elements of PM10 dust at three locations in Visakhapatnam city and they concluded that Al, Mn and Fe are predominantly of natural origin (i.e., lithogenic), whereas Cd, Cr, Zn and Pb have anthropogenic sources. In our study the presence of high concentrations of Mn, Zn, Pb, Cr, and Cu, indicates that the samples are highly influenced by anthropogenic sources while the contribution of dust with natural origin is quite low. Our results show very high concentrations of Mn (705.4 to 1536.4 ppm). The highest Mn values stem from samples closer to port area. The reason for these high values could be due to fine Mn ore that is exported via the port. The correlation between magnetic parameters and Mn is moderate in our results which may result from superimposed sources. The correlation of $kfd\%$ with Cr ($R=0.71$) is high, and also Ni is fairly correlated, however partly positive (Ni) or negative (Cu, Zn). Also the correlation with k with Cr, Mn and Ni is good while there is no correlation with the other measured elements. It again supports a complex mixture of different sources. Strzyszczyk [30] showed that the concentrations of ferrimagnetic materials have decreased to 0.24–0.89% at distances of 500 m away from a contaminated source (cement plants) and reached 0.02–0.16% at a distance of >3 km. The correlation between magnetic parameters and toxic metals in our study may not be due to structural incorporation [31] or adsorption [32] of heavy

metals in iron oxides, but more likely due to common sources and co-precipitation [33] contributing magnetic particles and heavy metals with different distance effects.

VI. CONCLUSIONS

- (1) The textile method is successful in detecting environmental pollution, and is suitable as a cost-efficient and fast method for monitoring suspended particle matter.
- (2) The content of heavy metals in Visakhapatnam city is beyond the permissible limits similar to big metropolitan cities in the world.
- (3) The correlation between the susceptibility and absolute weight of the suspended dust indicates that the susceptibility can be used as a proxy for spatial distribution of pollution levels in the city.
- (4) The magnetic concentration signal of the suspended dust is largely controlled by the pollutant from local source than the far distance. Thus the results enable us to know the extent of pollution zones.
- (5) Finer suspended particles (FSP) that are more hazardous to human health are released from vehicular traffic as indicated by higher percentage frequency dependence susceptibility (kfd%) values.

VII. ACKNOWLEDGMENTS

R.K. thanks CSIR, Govt of India for financial support, vide CSIR Award No.: 09/1067(0002)/2012-EMR-1 and S.G.to DST, Govt of India for support through research project. We are thankful to Visakhapatnam GITAM University for providing infrastructure and laboratory facility during progress of the work.

REFERENCES

- [1] WHO Report, Bonn, Germany, "Health aspects of air pollution with particulate matter, ozone and nitrogen dioxide," 2003.
- [2] A. Valavanidis, K. Fiotakis, T. Vlachogianni, "Airborne particulate matter and human health: toxicological assessment and importance of size and composition of particles for oxidative damage and carcinogenic mechanisms," *J. Environ. Sci. Health C Environ. Carcinog. Ecotoxicol. Rev.*, 26(4), pp. 339–362, 2008.
- [3] P.J. Flanders, "Collection, measurements and analysis of airborne magnetic particulates from pollution in the environment," *J. Appl. Phys.*, 75, pp. 5931–5936, 1994.
- [4] A. Kapicka, N. Jordanova, E. Petrovsky, S. Ustjak, "Magnetic stability of power-plant fly ash in different soil solutions," *Phys. Chem. Earth*, 25, pp. 431–436, 2000.
- [5] S.R. Goddu, E. Appel, D. Jordanova, F. Wehland, "Magnetic properties of road dust from Visakhapatnam (India) -relationship to industrial pollution and road traffic," *Phys. Chem. Earth*, 29, pp. 985–995, 2004.
- [6] N. Jordanova, D. Jordanova, T. Tsacheva, "Application of magnetometry for delineation of anthropogenic pollution in areas covered by various soil types," *Geoderma*, 144(3–4), pp. 557–571, 2008.
- [7] N.K. Meena, S. Maiti, A. Shrivastava, "Discrimination between anthropogenic (pollution) and lithogenic magnetic fraction in urban soils (Delhi, India) using environmental magnetism," *J. Applied Geophysics*, 73, pp. 121–129, 2011.
- [8] N. Basavaiah, U. Blaha, P.K. Das, K. Deenadayalan, H. Schulz, "Evaluation of environmental magnetic pollution screening in soils of basaltic origin: Results from Nashik thermal power station, Maharashtra, India," *Environmental Science Pollution Research*, 19, pp. 3028–3038, 2012.
- [9] N. Basavaiah, R.D. Mohite, P.U. Singare, A.V.R. Reddy, R.K. Singhal, U. Blaha, "Vertical distribution, composition profiles, sources and toxicity assessment of PAH residues in the reclaimed mudflat sediments from the adjacent Thane Creek of Mumbai," *Marine Pollution Bulletin*, 118, pp. 112–124, 2017.
- [10] M. Hanesch, G. Rantitsch, S. Hemetsberger, R. Scholger, "Lithological and pedological influences on the magnetic susceptibility of soil: Their consideration in magnetic pollution mapping," *Sci. Total Environ.*, 382, pp. 351–363, 2007.
- [11] A. Kapicka, E. Petrovsky, H. Fialova, V. Podrazsky, I. Dvorak, "High resolution mapping of anthropogenic pollution in the Giant Mountains National Park using soil magnetometry," *Stud. Geophys. Geod.*, 52, pp. 271–284, 2008.
- [12] U. Blaha, N. Basavaiah, K. Deenadayalan, D.V. Borole, R.D. Mohite, "Onset of industrial pollution recorded in Mumbai mudflat sediments, using integrated magnetic, chemical, 210Pb dating, and microscopic methods," *Environ. Sci. & Tech.*, 45, 686–692, 2011.
- [13] C. Spiteri, V. Kalinski, W. Rösler, V. Hoffmann, E. Appel, MAGPROX-Team, "Magnetic screening of a pollution hotspot in the Lausitz area, eastern Germany: correlation analysis between magnetic proxies and heavy metal pollution," *Environ. Geol.*, 85, pp. 109–117, 2005.
- [14] U. Blaha, B. Sapkota, E. Appel, H. Stanjek, "Micro-scale grain-size analysis and magnetic properties of coal-fired power plant fly ash and its relevance for environmental magnetic pollution studies," *Atmospheric Environ.*, 42, pp. 8359–8370, 2008.
- [15] R. Mitchell, B.A. Maher, "Evaluation and application of biomagnetic monitoring of traffic derived particulate pollution," *Atmospheric Environ.*, 43, pp. 2095–2103, 2009.
- [16] U. Blaha, E. Appel, H. Stanjek, "Determination of anthropogenic boundary depth in soil profiles and semi-quantification of heavy metal loads using magnetic susceptibility," *Environ. Pollut.*, 156, pp. 278–289, 2008.
- [17] D. Jordanova, S.R. Goddu, K. Kotsev, K. Jordanova, "Industrial contamination of alluvial soils near Fe–Pb mining site revealed by magnetic and geochemical studies," *Geoderma*, 192, pp. 237–248, 2013.
- [18] J. Hofman, I. Stokkaer, L. Snauwaert, R. Samson, "Spatial distribution assessment of particulate matter in an urban street canyon using biomagnetic leaf monitoring of tree crown deposited particles," *Environ. Pollut.*, 183, pp. 123–132, 2013.
- [19] P. Gautam, U. Blaha, E. Appel, "Magnetic susceptibility of dust loaded leaves as a proxy of traffic related heavy metal pollution in Kathmandu city, Nepal," *Atmospheric Environ.*, 39, pp. 2201–2211, 2005.
- [20] L.W. Cao, E. Appel, S.Y. Hu, M.M. Ma, "A method of monitoring discriminating pathways of traffic-derived pollutants," *Environ. Pollut.*, 205, pp. 97–102, 2015.

- [21] J.W. King, J.T. Channell, "Sedimentary magnetism, environmental magnetism and magnetostratigraphy," *Rev. Geophysics*, pp. 358–370, 1991.
- [22] Y. Syono, "Magnetocrystalline anisotropy and magnetostriction of Fe₃₀₄-Fe₂Ti₀₄ series with special application to rock magnetism," *Jpn. J. Geophys.*, 4, pp. 71–143, 1965.
- [23] R. Day, M. Fuller, V. Schmit, "Hysteresis properties of titanomagnetites: grain-size and compositional dependence," *Phys. Earth Planet. Interiors*, 13, pp. 260–267, 1977.
- [24] D.J. Dunlop, "Theory and application of the Day plot (Mrs/Ms versus Hcr/Hc) 1. Theoretical curves and tests using titanomagnetite data," *J. Geophys. Res.*, 107, pp. 2056–2060, 2002.
- [25] S. Mbengue, Y.L. Alleman, P. Flament, "Size-distributed metallic elements in submicronic and ultrafine atmospheric particles from urban and industrial areas in northern France," *Atmospheric Research*, 135, pp. 35–47, 2014.
- [26] R.W. Kozłowska, G. Majewski, P.O. Czechowski, "The size distribution and origin of elements bound to ambient particles: a case study of a Polish urban area," *Environ. Monit. Assess.*, 187, pp. 240–248, 2015.
- [27] P.K. Hopke, R.E. Lamb, D.F.S Natusch, "Multi-elemental characterization of urban roadway dust," *Environ. Sci. & Tech.*, 14, pp. 164–172, 1980.
- [28] A.Waheed, M. Morgan, S. Kashif, A. Ali, A. Ishfaq, A. Muhammad, A. Ishaq, "Roadside dust contamination with toxic metals along industrial area in Islamabad, Pakistan," *Nuclear Science and Techniques*, 25, pp. 030201-1–030201-6, 2014.
- [29] D.G. Gajghate, A.D. Bhanarkar, P.P. Pipalatkari, "Atmospheric Concentration and Fluxes of Trace Elements in Visakhapatnam city of India," *Int. J. Environ. Eng.*, 5(2), pp. 111–128, 2013.
- [30] Z. Strzyszczyk, "Gehalt an Ferromagnetika in den von der Immission der Zementindustrie in der Wojewodschaft Opole beeinflussten Böden (Contents of ferromagnetics in soils of Opole region contaminated by cement-industry immissions)," *Mitt der Deutschen Bodenkundl. Gesellschaft*, 76, pp. 1477–148, 1995.
- [31] V.S. Coker, A.G. Gault, C.I. Pearce, G. van der Laan, N.D. Telling, J.M. Charnock, D.A. Polya, J.R. Lloyd, "XAS and XMCD evidence for species-dependent partitioning of arsenic during microbial reduction of ferrihydrite to magnetite," *Environ. Sci. & Tech.*, 40, pp. 7745–7750, 2006.
- [32] G. Zhao, X. Wu, X. Tan, X. Wang, "Sorption of heavy metal ions from aqueous solutions: a review," *Open Colloid Sci. J.*, 4, pp. 19–31, 2011.
- [33] Y. Wang, G. Morin, G. Ona-Nguema, F. Juillot, G. Calas, J.G.E. Brown, "Distinctive arsenic (V) trapping modes by magnetite nanoparticles induced by different sorption processes," *Environ. Sci. & Tech.*, 45, pp. 7258–7266, 2011.

