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Spatial distribution of magnetic properties in urban soils and correlation with heavy metals pollution: A case study from the Thai Nguyen iron and steel industrial zones, Thai Nguyen city, Vietnam

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Abstract

Both natural and anthropogenic factors influence urban soils. Anthropogenic activities have caused the accumulation of heavy metals in the soil environment. Therefore, soil pollution significantly reduces environmental quality and affects human health. In this study, magnetic susceptibility measurements have been used for pollution monitoring. The objectives of this research are to determine the spatial variability of magnetic properties, and to selected heavy metals, and to evaluate effects of land use on their variability in the surface soils of the Thai Nguyen iron and steel industrial zones, Thai Nguyen city. The concentration of selected heavy metals including Cu, Zn, Pb, Fe, Cd, and the magnetic parameter, magnetic susceptibility at low frequency (χ_{lf}), in the urban and industrial land top - soils (0 - 5 cm) samples are significantly higher than that in the agricultural land soils in the study area. The spatial distribution of the selected heavy metals and χ_{lf} in the study area has suggested that activities at the urban and industrial land sites caused higher pollution as compared to that at the study sites of other land uses. Magnetic susceptibility using the MKF1 - A meter links to a computer operated using Safyr 7.0 software, the measurements have been done at low (0.47 kHz) frequency susceptibilities - χ_{lf}. The parameters for low-frequency mass magnetic susceptibility ranges between 14,1 × 10^{-8} m^3 kg^{-1} and 67,54 × 10^{-4} m^3 kg^{-1} with an average value of 15,34 × 10^{-4} m^3 kg^{-1} and standard deviation of 22,28 × 10^{-4} m^3 kg^{-1}. The concentrations of Cu and Zn are likely to have been affected by anthropogenic sources, whereas natural sources in the study area are mainly controlled Cd. Moreover, the attention of soil Pb and Fe in the study area could be affected by both lithologic and anthropogenic sources. The magnetic parameters used to be a proxy measure for the degree of heavy metal contamination and become a potential method for the detection and mapping of contaminated soils.

Key Words: *Magnetic susceptibility; Heavy metals; Urban soils; Spatial analysis; Thai Nguyen city; GIS*

1. Introduction

The use of the magnetic properties of soils is mainly due to the specific behaviour of metals compounds which almost exclusively control the magnetic order in the soil. Several authors have demonstrated the concentration of iron oxides, by weathering and pedogenesis in soils (Lamouroux and Ségalen, (1969); Paquet, (1970); Sadiki, (1991)). Together with the concentration of heavy metals from the parent rock to the top-soils, there is always an increase in the magnetic susceptibility of soil particles. The vertical

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evolution of the nature and content of magnetic minerals allows us to know about the state of the soil. Thus the constituents of stable, undisturbed soil show the base of the profile towards the surface, a regular increasing evolution of the magnetic susceptibility.

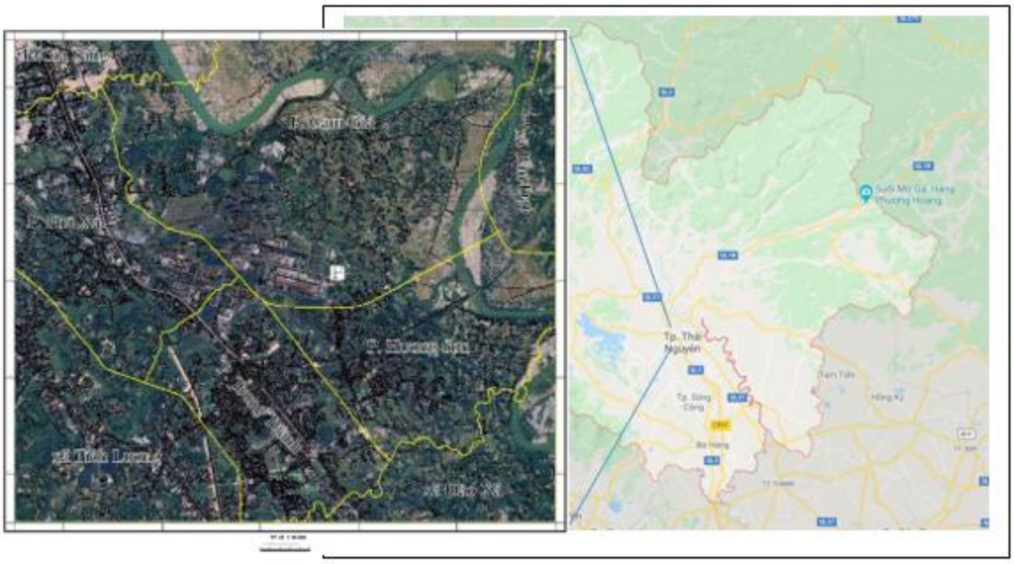

Figure 1. Geographical map study area, Thai Nguyen city

In contrast, a soil which has undergone a disturbance shows more disordered and generally is the low of magnetic susceptibilities. It allows for measurement the concentration of magnetic crystals, grain size, shape and type of the magnetic minerals presented in a sample (Mullins, (1977), Dearing et al., (1985); Beget et al., (1990); Dearing, (1999); Dearing et al., (2001); Meglish et al., (2008); Blundell et al., (2009); Kanu et al., (2013a) and (2013b)). Pollutants from human activities into the atmosphere will eventually settle and accumulate in the soil (Ayoubi and Adman, V, (2019); Ayoubi and Karami, M, (2019)). Accumulation of these anthropogenic particles derived from human activities such as one taking place at the steel ironworks (i.e., welding, painting, vehicular discharges and dust, poor disposal of spare part, etc.), resulted in significant enhancement of soil magnetic susceptibility (Kapicka et al., (2002), Caggiano et al., (2005); El - Hassan et al., (2009); Mahamed et al., (2011); Mohammad et al., (2012); Murdock et al., (2012); Kuceret al., (2012); Kanu et al., (2013a) and (2013b); Oluyide et al., (2019); Li XD., Poon CS., Liu PS., (2001)). In the particles state, these particles usually contain heavy metals and toxic elements.

There has been a growing interest in using magnetic techniques for monitoring environmental pollution, and many other publications studied the relationship between heavy metals accumulation in soil and magnetic susceptibility parameter. Kanu et al., (2013b), has published the results of their research of top - soil samples from parts of Jalingo, Taraba State, N - E Nigeria to assess soil contamination levels and to determine pollution hotspots using the magnetic parameter. Similarly, Xue Song Wang and Yong Qin, (2005); Yang T, Liu Q, Chan L, and Cao G, (2007) studied the correlation between magnetic susceptibility and heavy metals distribution in the city of Xuzhou, China. They opined that magnetic susceptibility could be used as a proxy for soil heavy metals contaminations.

The urban ecosystem is characterized as a complex composite of both the natural and anthropogenic factors. Heavy metals may come from various sources in urbanized areas, including vehicle emissions, industrial discharges, and activities (Thornton, (1991); Li et al., (2001)). The urbanization in the form of increased population density, intense industrialization and excessive exploitation of natural resources and has its consequential deteriorating impacts on the overall structures and functions of the eco - system (Antrop, (2000); Qureshi et al., (2010)). Because of the effectiveness of the integration of chemical composition and magnetic properties in the study of urban soil pollution degree, we have applied a similar technique to characterize and quantify the degree of pollution at Thai Nguyen iron and steel industrial zone, Thai Nguyen city, Viet Nam.

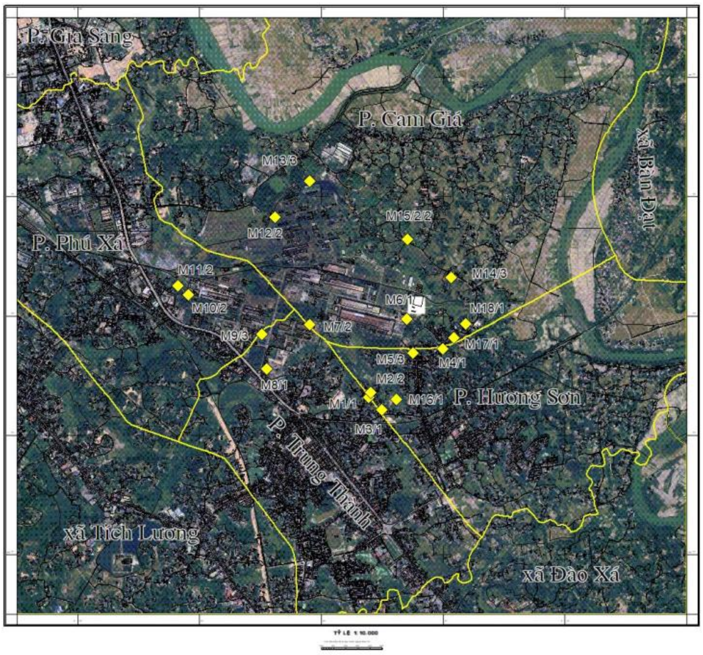

Figure 2. Sample location map of Thai Nguyen iron and steel industrial zones

2. Materials and methods

2.1. Study area

Thai Nguyen city is located in the northwestern part of Ha Noi capital, one of the provinces of Viet Nam. The main wind direction is from the northwest, although in general, wind velocities are low. This enhances the deposition of particulates within the city. It has very convenient transportation facilities, and its

current urban population exceeds 420,000 inhabitants. Nowadays, steel production and processing still play an essential role in the economic life of the city, together with the chemical industry and other activities. Therefore, urban top - soil may have accumulated heavy metals and magnetic materials over a long period, and a mixture of several sources has to be expected (Figure 2 - Table 1).

2.2. The sample magnetic susceptibility

The soil samples were collected in a suitable plastic bags test container of about 200 grams, by using a plastic spoon. These samples were sent to the laboratory where they were screened to remove macroscopic traces of stones, glass, plastic, rubber, animal, and plant matter to ensure that the analyzed materials were free from such contaminants. The samples were air - dried at room temperature in the laboratory in a few days to reduce the mass contribution of soil moisture and to avoid any chemical reaction and to avoid contact with metal objects (Kanu et al., (2013a), (2013b)). The samples were grinded and sieved through a 1 mm sieve mesh and stored in well - labelled plastic containers for magnetic susceptibility measurements and chemical analysis.

Table 1. Sample points and coordinates

ID	Sample	UTM-X	UTM-Y
1	M1/1	590385,2061	2383276,421
2	M2/2	590401,7802	2383320,582
3	M3/1	590493,1442	2383177,481
4	M4/1	590996,2314	2383677,219
5	M5/3	590747,5944	2383644,836
6	M6/1	590698,9522	2383920,31
7	M7/2	589900,5483	2383875,639
8	M8/1	589546,1337	2383510,303
9	M9/3	589510,4044	2383793,906
10	M10/2	588908,4001	2384120,049
11	M11/2	588821,6868	2384195,194
12	M12/2	589615,1184	2384756,72
13	M13/3	589901,1997	2385054,409
14	M14/3	591062,5774	2384263,75
15	M15/2	590703,6688	2384573,227
16	M16/1	590610,3094	2383261,584
17	M17/1	591083,357	2383767,041
18	M18/1	591179,2067	2383883,937

Magnetic susceptibility measurement was performed on each of the collected samples using the MKF1 - A meter linked to a computer operated with Safyr 7.0 software. For all the measures, the sensitivity was set at 1.0. Measurements were made three times; Firstly, air reading and sample reading; Secondly, air reading before and after each cycle for drift correction (Kanu et al., (2013b)). The MKF1 - A sensor was a handy laboratory sensor that made use of 10 cm^3 samples in plastic containers. It could take measurements at two different frequencies, i.e., at 470 Hz (low frequency) and 4700 Hz (high frequency). When the 10 cm^3 cylindrical plastic bottles were in use, the accuracy of the MKF1 - A meter was 1% (Dearing, (1999)). In this study, the susceptibility measurements were done at low - frequencies (470 Hz), which were further used to compute the frequency - dependent susceptibility.

Table 2. Statistic Summary of magnetic susceptibility of soil samples in Thai Nguyen iron and steel industrial zones (all values in $m^3\ kg^{-1}$)

ID	Sample	KRe [SI]	KIm [SI]	KVol [SI]	KMass [m^3/kg]
1	M1/1	0,0004699	0,00002113	0,0004699	0,0021411828
2	M2/2	0,0008194	0,00003027	0,0008194	0,0031976117
3	M3/1	0,0009313	0,00003060	0,0009313	0,0000006217
4	M4/1	0,0002324	0,00999400	0,0002324	0,0000002879
5	M5/3	0,0002024	0,00969900	0,0002024	0,0000001687
6	M6/1	0,0001567	0,00622000	0,0001567	0,0000001410
7	M7/2	2,0690000	0,00006725	2,0690000	0,0016030000
8	M8/1	8,3750000	0,00024850	8,3750000	0,0067540000
9	M9/3	6,0410000	0,00017480	6,0410000	0,0036750000
10	M10/2	7,2200000	0,00027010	7,2200000	0,0065340000
11	M11/2	0,0169800	0,00051330	0,0169800	0,0000147800
12	M12/2	0,0009640	0,00003811	0,0009640	0,0000007484
13	M13/3	2,9510000	0,00009282	2,9510000	0,0024970000
14	M14/3	0,0249500	0,00074520	0,0249500	0,0000212400
15	M15/2/2	1,3400000	0,00005247	1,3400000	0,0011590000
16	M16/1	0,0034760	0,00043700	0,0002340	0,0000003910
17	M17/1	0,0021340	0,00048700	0,0003457	0,0000016870
18	M18/1	0,0000456	0,00001276	0,0003426	0,0000187800
Min		0,0000456	0,0000128	0,0001567	0,000000141
Max		8,3750000	0,0099940	8,3750000	0,006754
Mean		1,5581868	0,0016186	1,5579238	0,001534
SD		2,76368648	0,00331587	2,76384327	0,002228
CV		177,365547	204,863898	177,405549	145,20586

Bartington Instruments Ltd., linked to the MKF1 - A dual - frequency sensor (470 Hz and 4700 Hz).

2.3. The sample chemical composition

Samples (approx: 0.2 g) were dissolved in a hot HF - HNO_3 - HCl acid mixture (approx. 15 ml), and refluxed with the acid mixture if the sample was only partly dissolved. Fe, Cd, Cu, Pb, and Zn concentrations were measured by inductively coupled plasma - Mass spectrometry (ICP - MS).

Table 3. Chemical analysis results of heavy metals for selected top - soil within the study area in mg kg⁻¹

ID	Sample	Cu (mg/kg)	Zn (mg/kg)	Cd (mg/kg)	Pb (mg/kg)	Fe (mg/kg)
1	M1/1	35	185	2	245	4108
2	M2/2	20	203	2	107	6213
3	M3/1	25	172	3	146	3070
4	M4/1	23	165	4	142	2984
5	M5/3	28	175	3	137	3118
6	M6/1	22	185	2	156	3247
7	M7/2	26	251	3	162	4092
8	M8/1	35	512	4	205	7436
9	M9/3	30	845	5	235	7254
10	M10/2	45	120	3	155	5852
11	M11/2	16	71	0,9	22	1642
12	M12/2	22	170	5	143	3051
13	M13/3	27	297	6	166	4672
14	M14/3	18	387	7	237	3356
15	M15/2/2	25	229	3	154	3804
16	M16/1	23	165	3	140	2992
17	M17/1	24	170	2	85	3052
18	M18/1	26	272	1	97	4363
	Min	16	71	0,9	22	1642
	Max	45	845	7	245	7436
	Mean	26,11	254,11	3,272	151,89	4128,11
	SD	6,86	178,69	1,64	55,79	1592,27
	CV	26,27	70,32	50,31	36,73	38,57

2.4. Morphology and SEM

SEM observed morphology of particles on magnetically enhanced samples, obtained by extraction from suspended material using a hand - magnet. Statistical and graphic analyses Statistical calculations were performed with the SPSS V.23 software. Contour plots were created with Arc - Map 10.6.

3. Results and discussion

3.1. Distribution of magnetic susceptibility

The mass - specific magnetic susceptibility values for the top - soil samples collected randomly within the study area are given in table 2. The values for low - frequency mass magnetic susceptibility range between 0,01 × 10^{-5} m^3 kg^{-1} and 675,40 × 10^{-5} m^3 kg^{-1} with an average value of 153,44 × 10^{-5} m^3 kg^{-1} and standard deviation of 222,81 × 10^{-5} m^3 kg^{-1}. These values of the magnetic susceptibility measurements at the steel ironworks are much higher than the observed values outside the steel ironworks. Values presented by Yang et al., (2007) from China, Mücella, (2010) from Turkey, Mohamed et al., (2011) and Ayoubi and Karami, (2019) from Iran reveals high variability of the magnetic susceptibility values but are all in the range of 10^{-5} (SI) as well.

As mean concentrations of the said of magnetic susceptibility in the urban soils of Thai Nguyen iron and steel industrial zones were found to be significantly different from each other, the degree of variability within the concentrations of magnetic susceptibility in the soil can be shown and compared with other metals more comprehensively by using the coefficients of variation: (CV = (SD × 100)/mean), rather than by Standard deviation (SD) itself. A modified version of Aweto ranking (1982) (CV ≤ 20%, little variation; 20 < CV ≤ 50%, moderate variation; CV ≥ 50%, hight variation) was suggested by Phil - Eze., (2010) as follows: if CV ≤ 20%, it shows low variability; 21% < CV ≤ 50% it is regarded as moderate variability; 51% < CV ≤ 100% it is observed as high variability; while 100% of the above CV is considered as exceptionally high variability. The same ranking has been used in the present study.

The GIS - based maps were selected according to their capability to visualize spatial relationships. The distribution of magnetic susceptibility is shown in figure 3. Higher values are concentrated in the southwest part where a lot of industrial plants (e.g., steel ironworks) are located, followed by a commercial zone where the magnetic minerals may be derived from the traffic emissions. The values of magnetic susceptibility are relatively smaller in public greens compared to the industrial zone.

3.2. Results of the geochemical analysis

Results of the heavy metal concentrations for the top - soil samples collected randomly within the steel ironworks are given in table 3. The concentration of the heavy metals of the selected top - soil decreases from Fe to Cd: Fe > Zn > Pb > Cu > Cd. The concentrations of Fe, Zn, and Pb in the steel ironworks are much higher than that of other selected heavy metals. The reason is a gradual accumulation from various pollution sources over time, including factories processing pig steel and iron, and other related activities in the steel ironworks. Too higher concentrations of Fe compared to other metals in the steel ironworks are likely to have come from metallurgical sources such as steel and iron. We did a parallel analysis for the top - soil samples collected randomly outside of the study area. It shows that the mean concentration of the selected heavy metals is much lower than their concentrations at the station, respective.

This also reveals that the higher values observed at the station could be due to the anthropogenic inputs. The value of the coefficient of variation (CV) also reveals the variability in the distribution of the heavy metals in the study area. CV ≤ 20% indicates little variability, 20% < CV ≤ 50% implies moderate variability, while 50% < CV ≤ 100% indicates high variability and CV value greater than 100% is regarded as exceptionally high variability. From the results, the low - frequency magnetic susceptibility (X_{lf}) and all the heavy metals show low variability in the soil.

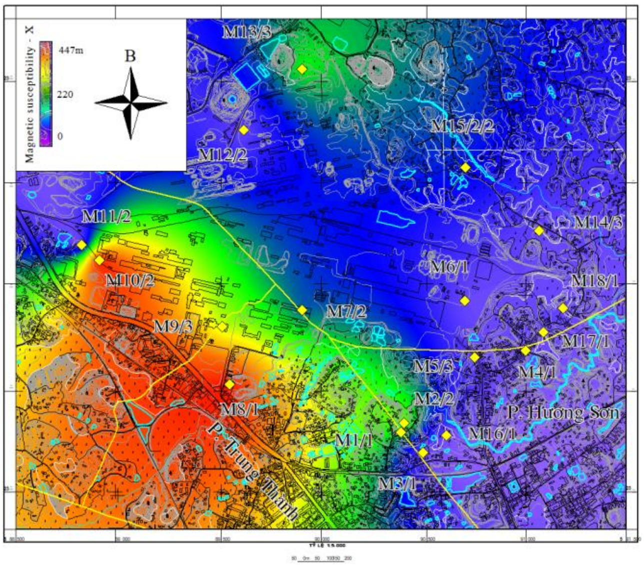

Figure 3. Map of distribution of magnetic susceptibility in the city of Thai Nguyen iron and steel industrial zones urban top - soil

3.3. Pearson correlation analysis

The Pearson correlation coefficients (R) between the heavy metals concentrations and magnetic susceptibility values and between different elements are presented in table 4.

Table 4. Pearson correlation matrix between magnetic susceptibility and heavy metals

		Kmass [m^3/kg]	Fe (mg/kg)	Pb (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Cd (mg/kg)
Kmass [m^3/kg]	Pearson Correlation						
	Sig. (2-tailed)						
Fe (mg/kg)	Pearson Correlation	**,872****					
	Sig. (2-tailed)	,000					
Pb (mg/kg)	Pearson Correlation	,375	***,477****				
	Sig. (2-tailed)	,125	,045				
Zn (mg/kg)	Pearson Correlation	,413	**,696****	**,605****			
	Sig. (2-tailed)	,088	,001	,008			
Cu (mg/kg)	Pearson Correlation	**,761****	**,593****	,431	,187		
	Sig. (2-tailed)	,000	,009	,074	,457		
Cd (mg/kg)	Pearson Correlation	,140	,212	**,622****	***,493****	,002	
	Sig. (2-tailed)	,578	,398	,006	,038	,993	

**. *Correlation is significant at the 0.01 level (2-tailed).*

*. *Correlation is significant at the 0.05 level (2-tailed).*

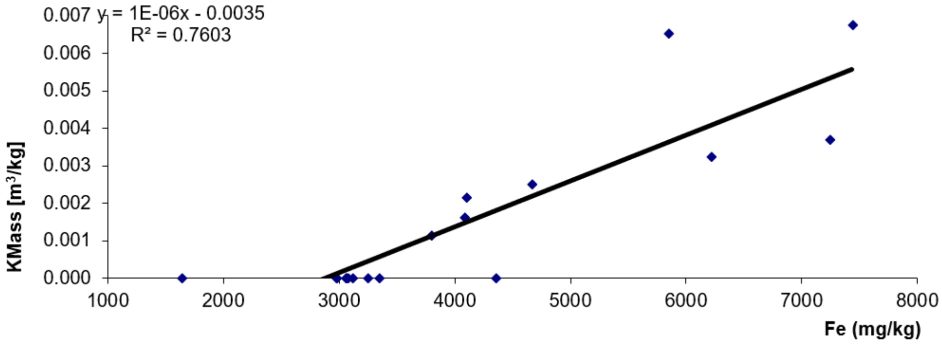

Figure 4. The correlation between Fe and magnetic susceptibility

The correlation analysis between the magnetic susceptibility and the heavy metals concentrations was conducted further to investigate the relationship between the sources of the heavy metals. The results were classified according to the correlation coefficient R as follows: 0,8 < |R| < 1, suggests a strong correlation; 0,5 < |R| < 0,8, suggests a significant correlation; 0,3 < |R| < 0,5 suggests a weak correlation; and |R| < 0,3 suggests an insignificant correlation.

The resulting demonstrated that concentrations of Fe (R = 0,872), Cu (R = 0,761), were strongly correlated with the magnetic susceptibility (Figure 4). Likewise, Cu (R = 0,593), Zn (R = 0,696) represented correlation with Fe; and Zn (0,605), Cd (0,622) represented correlation with Pb were also strongly correlated. Cd - Zn, and Fe - Pb showed an insignificant correlation.

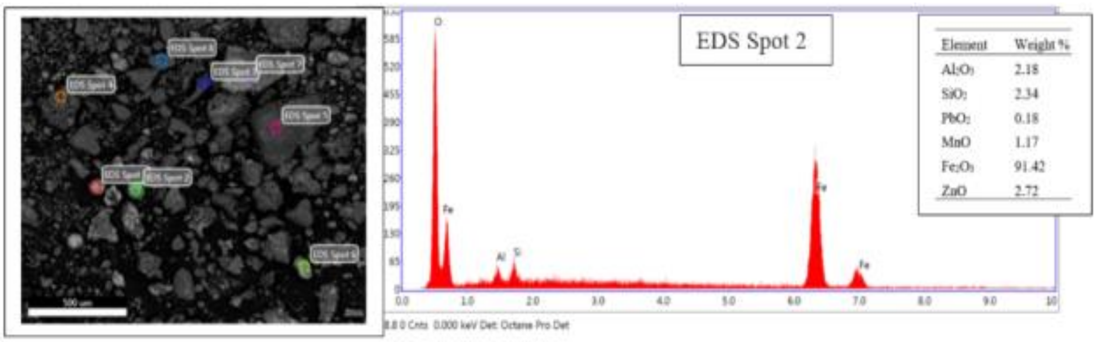

Figure 5. Image EDS spectra of metals oxide on the sample

The correlation between magnetic susceptibility measurements and heavy metals content shows a good relationship between ferrimagnetic oxides and heavy metals in the study area. This is a result of the fact that heavy metals are adsorbed onto the surface of pre - present ferrimagnetic in the environments or are subsumed into the lattice structure of the ferrimagnetic. A strong and positive correlation between the selected heavy metal and also with magnetic susceptibility was observed in table 4, indicating a regular pollution source. Therefore, the high correlation coefficient between the magnetic sensitivity measurements and the heavy metal content can be used as an indicator of the degree of contamination.

3.4. Morphology and SEM - EDS

The SEM and EDS are capable of providing imaging information about the morphology and surface texture as well as the elemental composition of the sample. SEM observation indicates that metal particle particles are characterized by the large heterogeneity of particle size and shape (Figure 6). Three morphologies of the sample can be identified: spherule, irregular-shaped, and aggregate particles. The representative SEM images for the spherule - shaped sample are shown in figure 6, indicating the sample present in spherical particles of various sizes. The morphology of these samples is consistent with the magnetosphere from the previous studies.

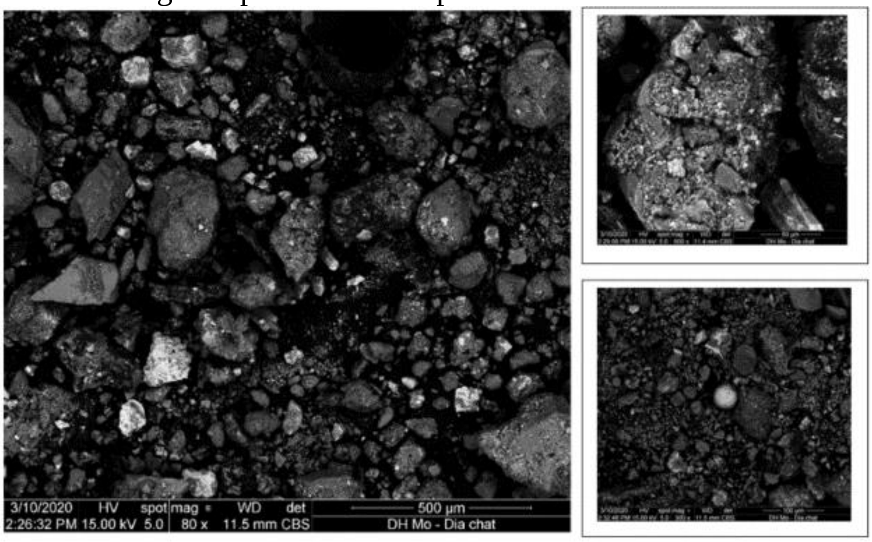

Figure 6. SEM images showed the spherule - shaped in different sizes

The spherule - shaped samples can be observed practically in all urban soils of our study. The size of spherical samples ranges from tens to hundreds of micrometers. EDS analysis reveals that the concentrations of Fe in these spherical samples are 91,42%. These spherules are typical products of coal - burning, such as power plants, industries, and domestic heating systems. Therefore, the most likely source of these spherules is the fuel high - temperature combustion, and the most likely source of the irregular slag is the iron - smelting and steel production.

4. Conclusion

Magnetic susceptibility measurement was carried out on 18 top - soil samples randomly collected from the study area, using the Bartington MKF1 - A meter linked to a computer operated using Safyr 7 software. The measurements were done at low (0.47 kHz) frequency susceptibilities. The results reveal high magnetic susceptibility values at the steel ironworks compared with the values observed outside steel ironworks. Magnetic particles present in urban soils may be formed by various technogenic processes, including coal-burning, industrially derived dust, slag, vehicle emissions, etc. This significant magnetic enhancement indicates a high concentration of ferrimagnetic minerals in the soil and thus evidence of pollution due to the activities in the study area. Otherwise, the results confirm the presence of a considerable amount of technogenic magnetic particles in urban top - soils. Furthermore, it also points out that the magnetic enhancement is of anthropogenic source than pedogenic and lithogenic. Chemical analysis of heavy metals also reveals higher values in the area industry.

The samples of different origins show very characteristic morphological and mineralogical forms. Three representative morphologies of samples can be observed:

spherical, irregular-shaped, and aggregate particles. The ferrimagnetic minerals of samples mainly occur in the form of magnetite and hematite with different stoichiometric and morphological forms. The distinctive morphological and magnetic mineralogical features of samples could serve as an indicator of source identification for soil pollution in urban soils.

The results also indicate that the strong correlation observed between the heavy metals and magnetic susceptibility. It also indicates a strong affinity of heavy metals to magnetic materials. Hence, since the magnetic susceptibility parameter is cost-effective, fast and can cover a very large area in a short time, it becomes essential and should be used as a preliminary method/tool to identify polluted spots before applying the geochemical method that is time - consuming and expensive.

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