Research article

Geochemical and electrical characterization of heavy metals in contaminated soils

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ABSTRACT

The present work is conducted in the industrial district south of greater Cairo (ElTabbin area). Some heavy metals like Mn, Co, Ni, Cu, Zn, Pb, Cr, Ba, Cd, Mo were determined in polluted soils during May, 2018. At the study area, results displayed that average heavy metals concentrations differ considerably. They are decreased from Mn to Cd (Mn > Ba > Zn > Cr > Ni > Co > Pb > Cu > Mo > Cd). The average ranges as follows: Mn (255.8–31448.2 ppm); Ba (145.2–17545.6 ppm); Zn (53.3–1589.9 ppm); Cr (26.7–311.3 ppm); Ni (29.7–114.1 ppm); Co (13.2–39.8 ppm); Pb (5.7–77.4 ppm); Cu (7.6–35.2 ppm); Mo (0.1–15.9 ppm) and Cd (0–1.5 ppm), respectively. ElTabbin area has heavy manufacturing activity at Egypt. ElTabbin area was chosen as a model for that contamination.

Electrical characteristics of some specimens from El- Tebbin area, Egypt, were taken and measured electrically at frequency range (10^{-3} to 100 kHz). Major and heavy elements were measured at specimens. Specimens were classified to three categories according to electrical properties. The changes were the consequence of change in minor and major mineral composition in the specimens. Electrical properties were able to recognize different specimens according to different levels of contaminants in the specimens. Texture, tortuosity and minor elements of heavy elements are the controlling factors that control electrical properties at specimens. High values concentration of Mn, Ni, and Zn heavy elements increases conductivity while the high concentration of Pb and Cu heavy elements decreases the conductivity. The mixing up of these different minor and major minerals and elements at specimen may lead to the change of conductivity values. The main controlling factors of the major elements are the Fe_2O_3 and SiO_2, while the main controlling factors of the minor elements are the Mn and Pb. Our main objective is to study mixing up of the different minor and major minerals and elements on electrical properties of rocks.

1. Introduction

Egypt alluvial soils are formed from disintegration of the metamorphic and eruptive rocks from Ethiopian Highlands. The mega cities at the world are facing serious environmental problems (Molina, 2004; Blokhuis et al., 1964).

The field mapping of the fluviatile and associated sediments of Nile Valley and the examination of a large number of boreholes, both deep and shallow, show that it is possible to conceive the Nile as having passed through five main episodes since the valley was cut down in the late Miocene time (Said, 1981). These are from the oldest to the youngest: Late Miocene, Late Pliocene, Proto-, Pre- and Neonile (Pleistocene). The local geology consists of quaternary deposits, floodplain sediments of Nile River (Said, 1993, 1990; Yousef, 1968).

Janos et al. (2010) mentioned that heavy metals are either leached into the ground or dissolved at the water and affecting growing food crops. Urbanization and industrialization increase rapidly, causing increased levels of environmental hazards (Robaa, 2003). Using bioremediation as a cleanup strategy for contaminated environments has increased due to its viability and cost-effectiveness (Alam et al., 2018).

ElTabbin area is located at South Greater Cairo. Melegy (2005), Melegy et al., (2002) reviewed the man-made impacts upon water quality of River Nile in Egypt. The previous work has shown that ElTabbin region is defined by relatively high level of anthropogenic soil contamination and stream sediments by potentially toxic elements (Cd, Se, As, Zn, Ba and Pb) (Melegy et al., 2010). The analysis of the soil extracted organic matter and sediments from ElTabbin reveal a marked contamination by anthropogenic products (Havelocova et al., 2014).

Increase of soil heavy metals increases the pollution effect and accordingly affects the human health (Melegy et al., 2010).

This paper aims to study refflationships between geochemical and electrical properties on heavy metals distribution at contaminated soils. ElTabbin (Egypt) area was chosen as a model for that contamination.

2. The investigated area

The investigated area ElTabbin is the heaviest industrial area in Egypt (Melegy, 2005). ElTabbin is located in the industrial district south of Greater Cairo on East side of River Nile with area about 30 km². It extends between longitudes (31° 16'- 31° 21' E) and latitudes (29° 45'– 29° 48' N) (Figure 1 A). The industrial activities include ferrous and non-ferrous metallurgical work, ceramics, firebricks, cement industry, coke factory, a power plant, chemical industries ...etc. Lack of effective planning has in recent years given rise to a number of social and environmental problems, including traffic congestion, sewerage, water supply and lack of appropriate landfill and land use planning (Gomaa et al., 2018; Havelcová et al., 2014; Melegy, 2005).

The emissions of the numerous and densely distributed industries are coupled with high temperature, lack of rain and predominant low wind speed. These factors favour the accumulation of pollutants on air over the district and supplement the formation of air pollution episodes. The situation temporarily worsens with dust laden winds especially during Spring and Autumn.

3. Materials and methods

3.1. Specimen collection

For determination of level of pollution 10 soil specimens were taken in spring period (May) in 2018 (Figure 1 B). Details of sampling sites are present at (Table 1).

The details of the collection of the specimens and preparation are present and discussed at (Melegy et al., 2010). At each site, six signs were marked. At each sign, 4 core specimens are collected at different depths (from 0 to 30 cm) randomly. This is used with a soil auger. Specimens are combined together to form one specimen. Specimens were kept in zip-locked plastic bags, air-dried at room temperature for 4 days, and broken into pieces to become fine powder. Then, coarse soil components were removed by sieving it with a 2 mm sieve. Sub-specimens of the sieved soils were then taken for heavy metals analysis. Volume of soil
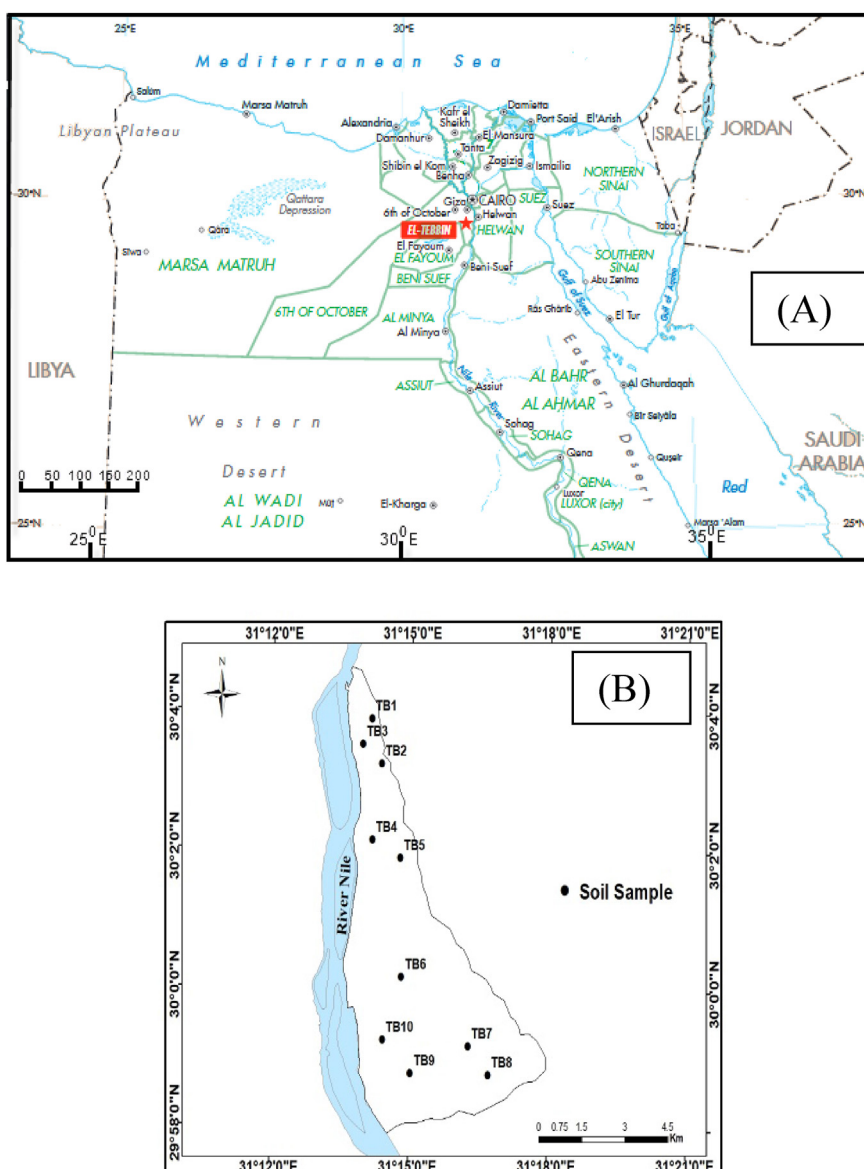

Figure 1. (A) The location map of the El-Tebbin Area, Egypt and (B) The sites of soil specimens in El- Tebbin Area, Egypt.

Table 1. The key of the studied specimens collected from ElTabbin area, Egypt (2018).

No. of specimens	Investigated sites	Major sources of pollution	ReZ (Ohm)
TB1	Arab Kafr Ei'llw	Cement factory	32400
TB2	Arab Kafr Ei'llw	Cement factory	28000
TB3	Arab Kafr Ei'llw	Cement factory	55900
TB4	Es El Tebbin	Asfor Co. of Metallorgy and Ceramics	24200
TB5	El Tebbin	Industrial Complexes	47900
TB6	El Shobak El-Sharqi	Industrial Complexes	17000
TB7	El Minya	Coke Factory	21000
TB8	El Minya, El Shurafa, El-Utaiyat	Coke Factory	14300
TB9	El-Utaijat	Industrial Complexes	17200
TB10	El Shobak	Industrial Complexes	17800

specimens, used for the electrical properties, were nearly 50 gm. These specimens were compressed to tablets for electric properties. The dimensions were one inch (2.67 Cm) in diameter and ~0.5 Cm in thickness. Major and trace elements were measured using X-ray fluorescence (XRF). Methodology used for calculating the amount of heavy metals in soil specimens are discussed in many papers (Shaltout et al., 2012).

LCR Hitester Impedance Analyzer (Hioki 3522-50) was used to measure the electrical properties. Electrical properties were measured at room temperature (~20 °C) and at relative humidity of (~65 %), at frequency range from .001 Hz up to 100 kHz, with 1 V (Gomaa and Alikaj, 2009; Gomaa and Kassab, 2016, 2017). A homogeneous mixture of specimens were used to be sure that the specimen is representative for the area of it. The variations of constituents change the values of the electrical properties, so the homogeneity of the specimens reduces these variations (Gomaa et al., 2019). Other details of electrical measurements details are present at (Gomaa, 2020).

4. Experimental results

There are a few analysis and discussions about the relation between the electrical properties and the major and trace elements of contaminations at rocks and minerals. Results of the chemical composition analysis (major and trace elements) of specimens of ElTabbin region (industrial district south of greater Cairo- ElTabbin area) are summarised in (Tables 2 and 3). Chemical analysis of major oxides (SiO_2, TiO_2, Al_2O_3, Fe_2O_3, MgO, CaO, Na_2O, K_2O, and P_2O_5) show that soils are dominantly has a siliceous type with small alumina enrichment.

Ranges of heavy minerals are Mn (255.8–31448.2 ppm); Ba (145.2–17545.6 ppm); Zn (53.3–1589.9 ppm); Cr (26.7–311.3 ppm); Ni

(29.7–114.1 ppm); Co (13.2–39.8 ppm); Pb (5.7–77.4 ppm); Cu (7.6–35.2 ppm); Mo (0.1–15.9 ppm) and Cd (0–1.5 ppm).

The available manganese concentration in soil specimens followed the order: TB8 > TB4 > TB2 > TB7 > TB1 > TB6 > TB9 > TB5 > TB10 > TB3. Maximum concentration of Mn was found to be 31448 ppm in TB8. The average concentration of Mn at the soil specimen was 3839.7 ppm. Although Mn is considered as an essential metal, but its high concentration may cause serious problems. Various reports are available which indicate high application of Mn metal from coke industry. The level of barium in soils is considered in the range from 145.2 ppm at TB3 specimen to 17545.6 ppm at TB8 specimen (~2096 ppm). Zinc also covers the class of essential heavy metal but may be supposed as toxic if present more than the maximum permissible limit. Concentration of zinc in soil specimens was found in the range of 53.3 ppm and 1589.9 at TB3 and TB8 specimens, respectively. Concentration of chromium varies from 26.7– 311.3 ppm (~111.04 ppm). The high concentration of Cr recorded at TB8 could be as a result of the steel producing industries in the area, chrome plating and pigment production is the main source of chromium contamination in that area. Nickel concentration ranges from 29.7-114.1 ppm (~63.26 ppm). The cobalt concentration at soil specimens varies from 13.2–39.8 ppm (~29.71 ppm). The highest concentration is 39.8 at TB9 specimen. The average concentration of lead at TB8 specimen is 77.4 ppm. Concentration of lead at the area ranges from 5.7-77.4 ppm. Main source of lead pollution in the area is due to electroplating of metals, excessive traffic and corrosion of lead pipes. Copper concentration varies from 7.6–35.2 ppm (~17.94 ppm). Copper accumulation at soil specimens may be due to steel manufacture industries. Similarly, concentration of Cd ranges from <0.001 ppm at TB1 specimen to 1.5 ppm at TB9 specimen with an average value of 0.6 ppm. Mean concentrations of Cd recorded at all sites were below the WHO/FAO (2001) permissible limit.

Table 2. Major elements in contamined soil in ElTabbin area.

TB10	TB9	TB8	TB7	TB6	TB5	TB4	TB3	TB2	TB1	Main Constituents (wt%)
34.8	36.7	10.5	44.5	45.5	27.1	40.9	42.9	40	42.	SiO_2
1.3	1.3	0.2	1.8	1.8	0.7	2.	1.2	2.	2	TiO_2
7.6	9.9	3.2	13.7	12.7	4.8	12.8	6.7	12.3	12.1	Al_2O_3
8.2	8.8	49.9	13.7	12.9	6.	13.	4.7	12.4	11.2	$Fe_2O_3^{tot.}$
2.2	3.4	1.2	2.6	2.4	2.	2.5	1.5	2.7	2.9	MgO
18.5	9.4	11.8	8.9	9.2	29.2	11.5	25.5	12.5	13.	CaO
2.3	4.6	0.9	0.8	0.8	1.1	0.8	0.7	0.8	1.1	Na_2O
1.8	1.8	0.4	1.5	1.5	1.1	1.1	1.1	1.1	1.2	K_2O
0.5	0.5	0.3	0.6	0.6	0.4	0.3	0.3	0.5	0.6	P_2O_5
14.5	12.4	16.7	10.8	11.3	22.3	13.6	13.3	14.6	12.5	LOI
4.8	6.5	1.5	0.3	0.5	3.	0.7	0.99	0.4	0.7	SO_3
2.9	4.4	0.8	0.2	0.3	1.7	0.1	0.5	0.1	0.3	Cl
99.4	99.6	97.4	99.4	99.3	99.3	99.3	99.5	99.5	99.5	Total

Table 3. Heavy metals in contamined soil in ElTabbin area.

Av	Max	Min	TB10	TB9	TB8	TB7	TB6	TB5	TB4	TB3	TB2	TB1	Trace elements (ppm)
3839.4	31448.2	255.8	471.6	548.9	31448.2	991.9	930.6	547.9	1158.3	255.8	1051.4	989.6	Mn
2096	17545.6	145.2	269.3	251.0	17545.6	378.6	387.4	414.3	737.6	145.2	411.4	420.1	Ba
269.7	1589.9	53.3	96.5	79.7	1589.9	136.7	136.1	163.6	188.0	53.3	130.8	121.9	Zn
111.	311.3	26.7	53.8	72.3	311.3	91.2	86.8	26.7	204.3	31.7	101.1	131.2	Cr
63.3	114.1	29.7	40.6	53.6	114.1	66.5	58.8	37.1	80.6	29.7	74.0	77.6	Ni
29.7	39.8	13.2	34.6	39.8	0	28.7	25.9	13.2	38.3	26.6	25.0	35.3	Co
28	77.4	5.7	14.4	9.6	77.4	21.3	19.2	48.7	54.0	5.7	17.3	12.2	Pb
17.9	35.2	7.6	12.7	12.8	27.9	19.2	16.6	12.7	35.2	7.6	18.5	16.2	Cu
2.4	15.9	0.1	0.6	1.6	15.9	0.1	0.3	1.1	0.6	0	1.1	0.1	Mo
0.6	1.5	0	0.2	1.5	1.2	0.6	0.0	0.4	0.002	0	0.3	<0.001	Cd

5. Discussions of electrical characteristics

There is a great change of the values of conductance in the specimens. These differences are argued to the great differences at mineral concentrations in the specimens (Gomaa, 2020a, b, c, d). Each specimen has a different value of minor or major element. The resulted electrical properties values are not limited to the total concentration of all these elements but, also, to the connections between these elements (this is what it is called texture between grains). Each specimen has a precipitation or sedimentation condition that is different from the other specimens. Also, the locations of specimens are different and accordingly the contaminants are different. TB1, TB2 and TB3 specimens are taken from (Arab Kafr Ei'llw) near a cement factory. TB4 specimen is taken from (Es ElTebbin) near a metallorgy and ceramics factory. TB5 and TB6 specimens are taken from (El Tebbin and El Shobak El-Sharqi) near an Industrial Complexes factory. TB7 and TB8 specimens are taken from (El Minya, El Shurafa, El-Utaiyat) near a Coke factory. TB9 and TB10 specimens are taken from (El-Utaijat and El Shobak) near an Industrial Complexes factory. Figure 2 shows the conductivity distribution with frequency of El Tebbin area. Conductivity curves values can be divided to three levels (high, moderate and low). TB10, TB9 and TB8 specimens have the highest conductivity values TB7, TB2 and TB3 specimens have the lowest conductivity values (Gomaa et al., 2009). TB6, TB5, TB4 and TB1 specimens have moderate conductivity values. The major elements TiO_2, Al_2O_3, Fe_2O_3 and MgO are supposed to be semiconducting elements, while SiO_2, CaO, Na_2O, K_2O, P_2O_5, SO_3 and Cl are supposed to be insulating elements. TB10, TB9 and TB8 specimens (Group A) have high conductivity values due to the presence of high TiO_2, Al_2O_3, Fe_2O_3 and MgO major elements, while the TB7, TB2 and TB3 specimens (Group C) have low conductivity values due to the presence of high SiO_2, CaO, Na_2O, K_2O, P_2O_5, SO_3 and Cl major elements. The other specimens (TB6, TB5, TB4 and TB1, Group B) have moderate values of conductivity due to the presence of a combination of the major elements. TB8 has very high total concentration of Fe_2O_3 major element (that increases the conductivity value), which is disseminated in the specimen, while it has the lowest value of the SiO_2 major element (that decreases the conductivity value). The combination of these major elements and the other elements in the specimen leads to the high values of conductivity at TB8 specimen
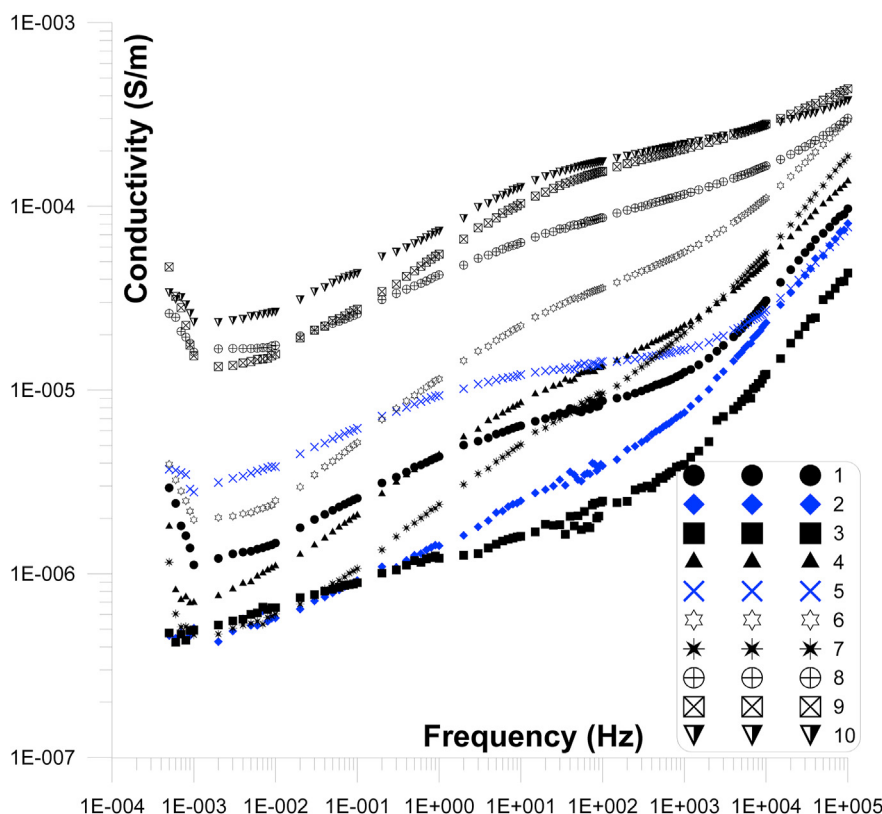

Figure 2. Variation of the conductivity of specimens as a function frequency of the El-Tebbin Area, Egypt.

and the other group (A) specimens (TB10 and TB9). On the other side, TB3 at Group (C), has the lowest conductivity value. TB3 specimen has very high concentration values of SiO_2 and CaO major elements (that decrease the conductivity value) in addition to relatively low concentration values of Al_2O_3, TiO_2 and the lowest conductivity value of Fe_2O_3 major element. Again, group B (TB6, TB5, TB4 and TB1) has moderate values of conductivity for the combination of all these major elements.

Really, the previous concentrations are not the only dominant factor in controlling the conductivity values. There are another dominant factor, which is called the texture and tortuosity of all these elements and the way that they are arranged together. Another factor is the minor elements (Table 3) of these heavy elements in the specimens. TB8 (Group A) has very high concentration of Mn, Ni, and Zn heavy elements (increases conductivity), while the other group specimens (TB9 and TB10) has relatively high values of these elements in addition to the relatively high values of the Pb and Cu heavy elements (decreases conductivity). Similarly, TB3 (Group C) has the lowest concentration values of Mn, Ni, and Zn heavy elements (increases conductivity), while the other group specimens (TB7 and TB2) has relatively high values of these elements in addition to the relatively high values of the Pb and Cu heavy elements (decrease conductivity). The mixing up of these different minor and major minerals and elements at specimen may lead to the change of conductivity values at these specimens. The main controlling factors of the major elements are the Fe_2O_3 and SiO_2, while the main controlling factors of the minor elements are the Mn and Pb.

Figure 2 shows three different regions for the variation of the conductivity with frequency. The first region is defined by a steep increase of conductivity with the increase of frequency at relatively low frequencies (slope ~0.25). The second region is defined by very low or nearly no increase of conductivity with the increase of frequency at relatively moderate frequencies. Finally, the third region is characterized, again, by a more or less increase of conductivity with the increase of frequency at relatively high frequencies (slope ~0.35). These three regions are very clear at specimen TB3 but they are changed from specimen to another

according to its combinations. Also, these three regions changes by decrease or increase (in frequency range) from one specimen to another.

Generally, as the frequency increases the conductivity increases as a direct result of the increase of charged particles energy. Semi-conductor clusters increase with the increase of their concentration and with the increase of frequency. Most of the specimens used here behave as semiconductors to relatively low conductors. Also, the loss of ignition (LOI) in the specimens adds humidity and accordingly conductivity values to the actual conductivity; this means that increase of LOI increases the conductivity of specimens. Generally, increase of frequency activates charged particles and elements to conquer energy barriers to become more conductive or semi-conductive and, finally, conductivity values increase.

Figure 3 shows the distribution of the dielectric constant (permittivity) with frequency of El Tebbin area. The values of the dielectric constant curves behave nearly as one group of levels. TB10, TB9 and TB8 specimens have the highest dielectric constant values. TB7, TB2 and TB3 specimens have the lowest dielectric constant values. TB6, TB5, TB4 and TB1 specimens have moderate dielectric constant values. The combinations of minor and major elements in the specimens lead to the present values of dielectric constant. The arrangement of the specimens according to conductivity values is nearly the same as their values according to dielectric constant (group A, B and C has the same arrangements). TB10 at Group (A) has the lowest dielectric constant value. TB3 at Group (C) has the lowest dielectric constant value.

Concentrations of minor or major elements and texture are not the only dominant factor in controlling the conductivity values. The mixing of these different minerals and elements at specimen may lead to the change of dielectric constant values. Again, the main controlling factors (on dielectric constant) of major elements are Fe_2O_3 and SiO_2 concentrations, while the main controlling factors of minor elements are the Mn and Pb.

Figure 3 shows three (or two at certain specimens) different regions for the variation of the conductivity with frequency. The first region is
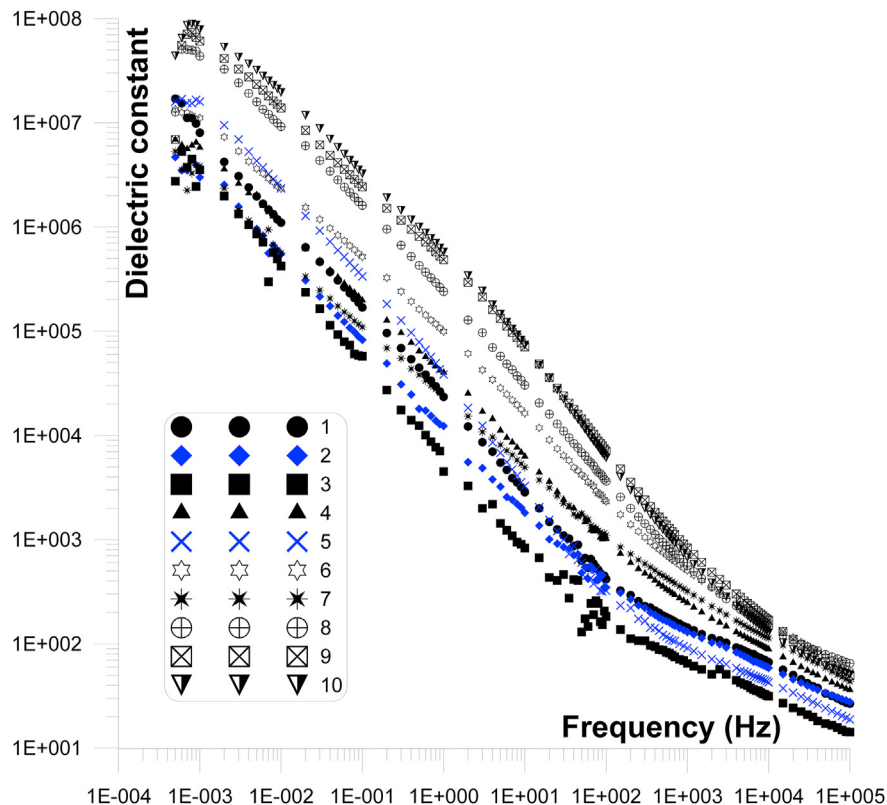

Figure 3. Variation of the dielectric constant of specimens as a function frequency of the El-Tebbin Area, Egypt.

defined by a steep decrease of dielectric constant with the increase of frequency at relatively low frequencies (slope ~0.75). The second region is defined by more increase of conductivity with the increase of frequency at relatively moderate frequencies. Finally, the third region is characterized, again, by a more or less flat decrease of dielectric constant with the increase of frequency at relatively high frequencies (slope ~0.33 to 0.65). These three regions are clear in specimens TB10 and TB9 but they are changed from specimen to another according to its combinations and concentrations of elements, while at the other specimens it is may be seen as two regions only. Also, the breadth of these regions changes by decrease or increase (with frequency range) from one specimen to the other.

Generally, as the frequency increases the dielectric constant decreases as a direct result of the decrease of distance between semi-conducting cluster, which decrease with the increase of the activity of charged particles with the increase of frequency. Frequency increase and accordingly, charged particles are activated to overcome energy barriers between clusters or grains to become more close to each other's and, finally, dielectric constant values increase (Gomaa et al., 2018).

The small insulating distances between semi-conducting clusters or grains motivate high values of dielectric constant.

Figures 4 and 5 show the distribution of the real and imaginary impedance of specimens with different concentrations of El Tebbinarea. The values of the real and imaginary impedance curves can be divided to three levels (high, moderate and low) complex impedance values. Group (A) specimens has the lowest real impedance values (~14.3–17.2 kOhm) at high relatively frequency (100 kHz). Group (C) specimen's have the highest real impedance values (~32.4–55.9 kOhm) at relatively high frequency (100 kHz). Group (B) specimens have moderate complex impedance values (~17.8–28.0 kOhm) at relatively high frequency values (100 kHz). Group (A) has low complex impedance values at relatively high frequency (100 kHz). Group (C) has high concentrations of SiO_2, CaO, Na_2O, K_2O, P_2O_5, SO_3 and Cl. TB3 specimen has the highest low complex impedance value and TB10 has the lowest low complex impedance value at relatively high frequency (100 kHz).

Concentrations of major and minor elements control the complex impedance values (Table 3). Mixing up of elements and minerals in specimens changes the texture that changes the complex impedance of these specimens (Gomaa and Eldiwany, 2020).

Figure 4 shows the distribution of all specimens. There are arcs (or lines) for all the specimens for the low frequency impedance. This arc (or line) is an indicator of the ratio of the real to the imaginary components in the specimens. When the angle (with the X-axis) decreases, then the specimen is more conductor (increase of total conductor conduction paths between electrodes) and vise verse. Figure 5 shows the distribution of real and imaginary impedance of each specimen. The detailed figure is a trial to show the details of each specimen. The curves can be distinguished to mainly two different regions. The low frequency region is represented by arcs (or lines). The arcs (or lines) are a representation of the interface grains of the specimens (Shaltout et al., 2012). The second region is the high frequency region, which is represented by a semicircle at the end of the arcs (or lines). The semicircles are a representation of the bulk material of the specimens. The semicircle is clear at some specimens (TB1, TB3, TB5, TB8 and TB 10) and at the other specimens it is hardly to be distinguished. The semicircles have different radii and different centers according to effect of the bulk material at the specimens (Gomaa and Abou El-Anwar, 2015).

Generally, semicircles mean that there is DC in addition to the AC conduction of the conductivity (Kassab et al., 2017). Arcs decrease in size to be a fraction of semicircle due to the increase of conductor minerals (Gomaa and Abou El-Anwar, 2019). The minor and major compositions (and texture) change and the electrical properties are changed and
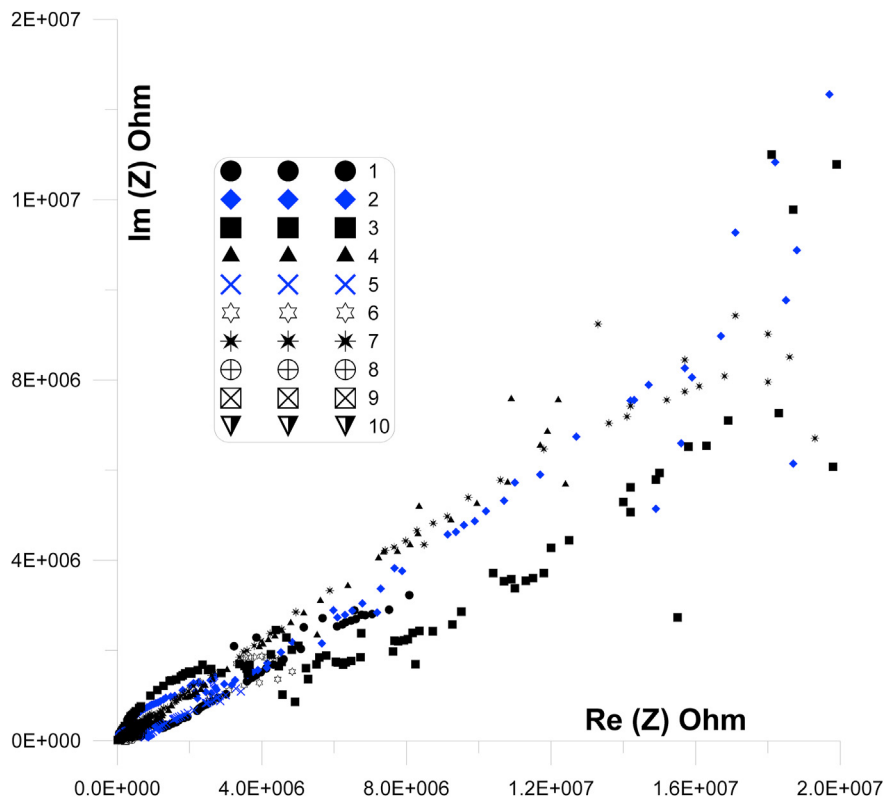

Figure 4. Variation of the real and imaginary impedance of specimens as a function frequency of the El-Tebbin Area, Egypt.
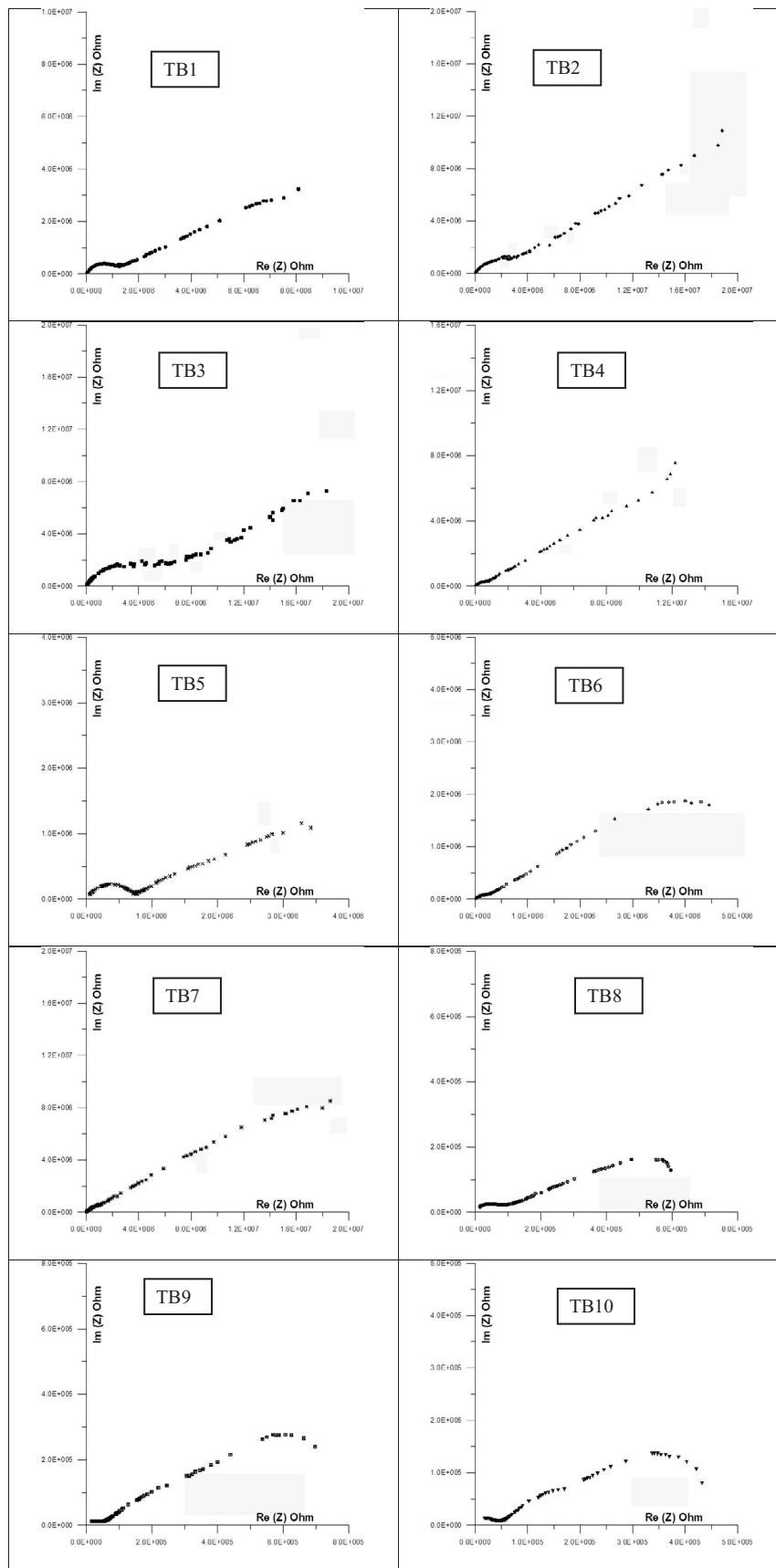
Figure 5. Detailed variations of the real and imaginary impedances of specimens as a function of frequency of the El-Tebbin Area, Egypt.

accordingly we can follow the contamination effect of minor and major compositions at the electrical properties.

6. Conculsion

At the study area, results displayed that average heavy metals concentrations differ considerably. They are decreased from Mn to Cd (Mn > Ba > Zn > Cr > Ni > Co > Pb > Cu > Mo > Cd). Maximum concentrations of Mn, Ba, Zn Cr, Ni, Pb and Mo were found to be 31448, 17546.6, 1589.9, 311.3, 114.1, 77.4 and 15.9 ppm, respectively, in TB8, which indicate high application of these metals from, coke the steel industries.

Electrical characteristics of some specimens from El- Tebbin area, Egypt, were taken and measured electrically at frequency range (10^{-3} to 100 kHz). Major minerals were measured in addition to the heavy metal elements in the specimens. Specimens were classified to three categories according to electrical properties. The changes were the consequence of change in minor and major mineral composition in the specimens. Electrical proeprties were able to ditiguish between different specimens according to different levels of contaminants in the specimens. Texture and tortuosity are the controlling factors that control the electrical proeprties in addition to the minor elements of the heavy elements in the specimens. High values concentration of Mn, Ni, and Zn heavy elements increases the conductivity while the high concentration values of the Pb and Cu heavy elements decreases the conductivity. The mixing up of these different minor and major elements and minerals in the specimen may lead to the change of conductivity values. The main controlling factors of the major elements are the Fe_2O_3 and SiO_2, while the main controlling factors of the minor elements are the Mn and Pb. Finally, we can conclude that the mixing up of the different minor and major elements at the specimens changes the apparent effect of the electrical properties. Many condensed measurements and discussions are needed to study qualititavely effect of these parameters.

Declarations

Author contribution statement

M. M. Gomaa, A. Melegy, H. Metwaly, S. Hassan: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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