Geochemical investigations regarding the variability of metal pollution in the Amvrakikos Bay, Greece

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Abstract

The Amvrakikos Bay is one of the most important ecosystems in the Mediterranean with great environmental, ecological and biological value. However, over the years, water quality has suffered severe degradation and is now one of the most polluted environmental areas in Greece. In the present study, using the analytical data of concentrations of precipitated metal deposits at the bottom of Amvrakikos Gulf, at its three largest lagoons: Logaros, Tsoukalio and Rodia, and by applying the indicators PLI, Igeo and EF, an assessment of the level of pollution of the area was made. The use of these indicators may show the degree of pollution of the area with heavy metals, and reveal the source of pollution i.e. whether the heavy metals are of anthropogenic or of natural processes, assessing simultaneously the degree of anthropogenic effect. Based on the PLI indicator, the marine zone in Menidi and Paliovarka as well as the Rodia and Tsoukalio lagoons are considered to be polluted. According to the Igeo Index, the sampling areas are unchanged to slightly burden by heavy metals. The largest charge is caused by Ni, followed by Mn and Pb. Based on EF, the concentrations of Cr, Mn, Co, Pb and Ni are significantly influenced by the various anthropogenic activities.

Keywords: Amvrakikos, bay, metal pollution indices, marine sediment metal pollution, management of marine environment.

1. Introduction

The Amvrakikos Bay is located on the northwest coast of Greece. The area occupies about 400 km[2] and is one of the largest semi-bays with particular morphology. It is characterized by a limited water renewal, as communication with the open sea is achieved only in the area of Preveza, through a narrow channel approximately 600 m wide. The bottom of the sea is covered with sludge and sand. The waters are blurred and show a strong eutrophication phenomenon (Bearzi *et al.,* 2008). It presents high concentrations of nutrients, making it a highly productive habitat. The complex system of Amvrakikos lagoon includes at least 15 lagoons, with a total area of more than 70 km[2]. Their main characteristic is their small size, shallowness and their influence from a similar tidal effect on their opening (Christia and Papastergiadou, 2006).

Due to the limited water renewal, only coming from the surface waters of the Ionian Sea, the issue of Amvrakikos waters quality arises, especially in the deepest layers of the bay. Combined with the run-off to the northern part of the rivers, carrying sediments with organic material and increased concentrations of metals and nutrients, the existing situation is even worse. The major sources of pollution from human activities that burden the Amvrakikos Bay are, industrial waste water, sewage, surface run-off (mainly from agriculture and livestock farming).

Environmental geochemical studies can help in assessing the quality of the environment as well as the determination of the sources of pollutants, their behavior and other characteristics. This knowledge is necessary in any application of remediation methodologies and waste management for the prevention of pollutants in getting into the food chain and in determining safe criteria regarding the quality of soils and drinking water (Varnavas *et al.,* 2012). Based on geochemical studies of marine sediments and waters, environmental conditions have been assessed in many Greek seas (Voutsinou *et al.,* 1997) and Greek bays such as the Saronic Gulf (Sklivagou *et al.,* 2008; Sklivagou *et al.,* 2001), Thermaikos Gulf (Voutsinou and Varnavas, 1995), Patraikos Bay (Varnavas, 1991), Euboicos Bay (Voutsinou and Varnavas, 1985), but also in other small or large bays (Achilleopoulos and Varnavas, 1996; Varnavas *et al.,* 2012; Varnavas, 1989; Varnavas *et al.,* 1992; Voutsinou and Varnavas, 1992; Varnavas *et al.,* 1987a; Panagos *et al.,* 1989; Varnavas *et al.,* 1987b; Varnavas *et al.,* 1984) and lagoons (Varnavas, 2005), giving interesting results.

The aim of this paper is to assess the environmental status of the Amvrakikos Gulf water system, using the metal concentration data in sediment samples at the Amvrakikos Gulf, more specifically at its three largest lagoons: Logaros, Tsoukalio and Rodia, and by applying the indicators PLI, Igeo and EF, an assessment of the level of pollution of the

area was made. The use of these indicators may show the degree of pollution of the area with heavy metals and reveal the source of pollution i.e. whether the heavy metals are of anthropogenic or of natural processes, assessing simultaneously the degree of anthropogenic effect.

2. Materials and methods

According to the study by Ferentinos *et al.* (2010), the bottom of the bay is covered by a uniform black layer of mud, 3-10 cm thick (Figures 1 and 2). The intense black color is the proof of the anoxic conditions prevailing at the bottom. As can be seen from the bottom profile, its clay content is estimated between 28 and 33% while sludge ranges from 65 to 68%. Sand fractions are less than 3% of the sediment. Underneath the black layer, another layer, greenish green and over 50 cm thick, is present. It contains 38-41% clay and 50-60% sludge. The black layer contains 5-8.6% total carbon (TOC), while the second layer contain only 5% TOC, (Ferentinos *et al.,* 2010).

In coastal water systems, the presence of metals, commonly referred to as heavy metals, is frequent. Some of these, such as aluminum and iron, have a natural presence and are found at high relative concentrations. Some others, such as mercury, cadmium, selenium etc., are rarely present and their concentrations are usually low. Human activities can produce industrial and municipal waste, agricultural residues etc., increasing the concentrations of heavy metals. As a consequence, they accumulate in fine sediments and living organisms, creating toxicity, causing dysfunction in organ functions and changes in population distribution and abundance.
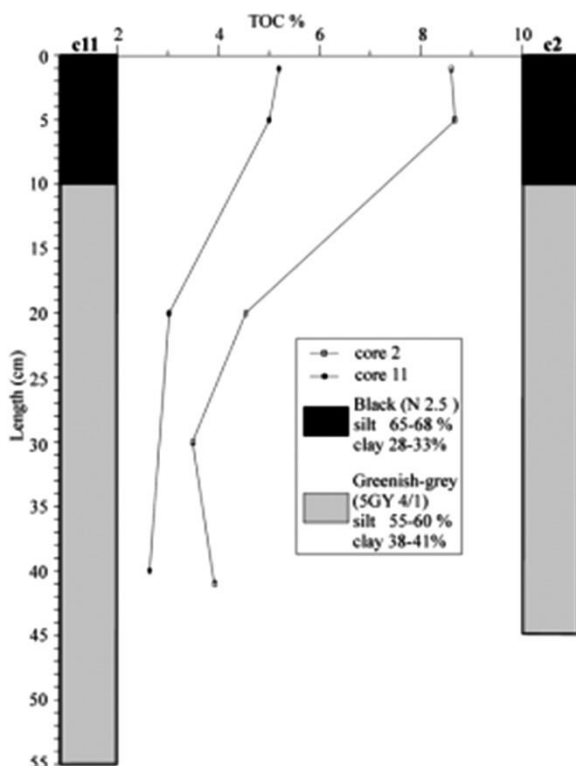

Figure 1. Bottom description and TOC profile, at two different locations in Amvrakikos (Ferentinos *et al.,* 2010)

In Table 1, the data from the determination of heavy metals in sediments of the Amvrakikos Marine Region (A2, A10, A12, A14) and lagoons (L1, L2, L3) in December 2009 (Amvrakikos Management Consortium, 2009), are presented. The data were used in the research project "Development of Management Tools and Drafting of a Master Plan of the hydrological basins of the rivers Louros and Arachthos and Amvrakikos Bay". Sampling sites were chosen so as to reflect in extended range fitness Amvrakikos Gulf.

- A2. Pogonitsa (38 ° 57'37.05 "N, 20 ° 48'34.50" E). A2 station, 8.5m deep, is close to two large aquaculture units. It is also representative of the northern part of the Gulf.

- A10. Menidi (39 ° 1'39.70 "N, 21 ° 7'7.88" E). The station A10, 14.6 m deep, is located outside the Menidi City Center. It is also close to large fish farms.

- A12. Paliovarka (38 ° 54'6.39 "N, 21 ° 3'36.45" E). The A12 station, 20 m deep, is representative of the conditions prevailing in the southern part of the Amvrakikos Gulf between the two urban centers of Vonitsa and Amfilochia.

- A14. Vonitsa (38 ° 56'10.47 "N, 20 ° 53'43.21" E) The A14 station, 14 m deep, is located in the port of Vonitsa.

- L1. Rodia (39 ° 5'19.57 "N, 20 ° 48'41.20" E), L2. Tsoukalio (39 ° 3'38.07 "N, 20 ° 49'42.60" E) and L3. Logarou (39 ° 2'33.55 "N, 20 ° 54'33.11" E) L1, depths of 3.03 m, L2, depth 1.6 m and L3 1.8 m deep, are representative of biotic and abiotic conditions prevailing in the three largest and most important lagoons of the North Amvrakikos Gulf.

The high content of heavy metals, in the lagoon sediment, is due to the geological composition of their soils (Karageorgis, 2007). In the lagoons of Rodia and Tsoukalio, we can observe, higher values of lead concentration than in the rest of the bay, associated with the anoxic conditions of the bottom of the two lagoons (Calvert *et al.*, 2001).

The data of Table 1 are expressed in dry μg/g weight of sediment. Concentrations of metals were measured by atomic absorption spectrometry.

3. Heavy metal pollution indices

Estimation of pollution levels in the bottom sediment of a coastal area is based on a series of indicators, such as:

3.1. Pollution load index, PLI

The Pollution Load Index (PLI) is the concentration of each heavy metal relative to the metal concentration value in an average non-polluted precipitate. The index is calculated as follow (Tomlinson *et al.,* 1980):

a) Concentration Factor (CF) for each metal and at each sampling location:

$$CF = C \text{ metal}/C \text{ background value} \tag{1}$$

C metal: metal concentration in polluted sediments

C background value: background value of that metal.

The background concentrations of the respective metals Enrichment Factor are shown in Table 2.

b) Calculation for each sampling point of the total charge of all heavy metals:

PLI sampling position = (CF1×CF2×CF3×....×CFk)$^{1/k}$ (2)

CFk: the pollution factor of the k-bone metal at the specific sampling site

k: number of metals

c) Sampling positions displaying a similar PLI-location are grouped into bands and the pollutant load index (PLI) for each zone is calculated as follow:

PLI zone = (PLI1×PLI2×…×PLIm)$^{1/m}$ (3)

PLIm: the pollutant of the m-th sampling position

m: the number of sampling sites constituting that zone

d) Calculation of the general pollution load index for the studied sea bay, or lake according to the individual indicator of the zones based on the ratio:

PLI environment = (PLI×PLI2×…×PLIn)$^{1/n}$ (4)

PLIn: the pollutant of the n-th zone

n: the number of zones constituting the particular environment.

3.2. Geoaccummulation index, Igeo

The Geoaccumulation Index (Igeo) compares the current concentrations of heavy metals in a sample to the corresponding pre-industrial levels. The index is expressed as follow (Muller, 1969):

(5)

Igeo = log2 Cn/(1.5 ×Bn)

Cn: measured concentration of element n in the sediments

Bn: geochemical background for the element n

1.5: is used to minimize lithological effects.

3.3. Enrichment factor, EF

The Enrichment Factor (EF) is expressed as follow (Huu, 2010):

EF = (Tracer/Normalizer) sample/ (Tracer/Normalizer) background   (6)

Where (tracer/normalizer) Sample and (tracer/normalizer) background respectively are the concentrations of metals (in µg/g of dry sample) in the sample and in the non-contaminated area.

Typically, as a normalizer we use a reference metal whose concentration is not due to anthropogenic effect. Such metals are iron and aluminum (Mediolla *et al.,* 2008).

Table 1. Concentrations of heavy metals in sediments of the Amvrakikos sea area and lagoons (Amvrakikos Management Consortium, 2009)

Metals	Cu µg/g	Cr µg/g	Ni µg/g	Co µg/g	Mn µg/g	Pb µg/g	Zn µg/g
EF - A2	27.9	86.8	310	32.5	1487	31.2	28.1
EF - A10	36.1	71.3	203	27.1	1172	21.1	43.5
EF - A12	47.3	76.9	223	31.2	484	28.1	50.1
EF - A14	19.7	27.5	68.6	22.9	896	33.6	17.3
EF - L1	34.9	68.5	161	28.0	941	43.0	39.6
EF - L2	34.5	107.8	154	28.8	957	39.7	40.9
EF - L3	29.8	57.0	146	27.5	1058	30.0	29.1

Table 2. Heavy metal background table (Turekian and Wedepohl, 1961)

Metals	Cu µg/g	Cr µg/g	Ni µg/g	Pb µg/g	Zn µg/g	Co µg/g	Mn µg/g	Fe µg/g
Background concentrations	45	83	68	20	95	19	600	47200

Table 3. CF values calculated for each element

Metals	Cu	Cr	Ni	Pb	Zn	Co	Mn	Fe
CF$_{metal}$ - A2	0.62	1.05	4.56	1.56	0.02	1.71	2.48	0.513
CF$_{metal}$ - A10	0.8	0.86	2.99	1.06	0.46	1.43	1.95	0.409
CF$_{metal}$ - A12	1.05	0.93	3.28	1.41	0.53	1.64	0.81	0.428
CF$_{metal}$ -A14	0.44	0.33	1.01	1.68	0.18	1.21	1.49	0.176
CF$_{metal}$ - L1	0.78	0.83	2.37	2.15	0.42	1.47	1.57	0.386
CF$_{metal}$ - L2	0.77	1.3	2.26	1.99	0.43	1.52	1.6	0.449
CF$_{metal}$ - L3	0.66	0.69	2.15	1.5	0.31	1.45	1.76	0.407

4. Results and discussion

The calculation of the PLI, EF and Igeo indices was done according to equations (1) to (6). The concentrations of metals for each sampling area are reported in Table 1. The background concentrations of the respective metals Enrichment Factor are shown in Table 2.

4.1. Pollution load index, PLI

In Table 3, the CF calculations for each element, at all sampling positions, carried out according to equation (1), are presented. In Table 4, the results of the PLI for each sampling position, according to equation (2), are presented.

Table 4. PLI calculated for all elements in each sampling point

PLI - A2	0.83
PLI - A10	1.02
PLI - A12	1.04
PLI - A14	0.59
PLI - L1	1.02
PLI - L2	1.1
PLI - L3	0.91

4.2. Geoaccummulation index, Igeo

The total PLI environment according to equation (4) for all the above sampling positions is PLItotal = 0.91.

Table 5 shows the Igeo calculations for each element at all sampling locations, according to equation (5).

4.3. Enrichment FACTOR, EF

In Table 6, EF calculations for each element, at all sampling positions according to equation (6), are presented. Iron was used as the reference metal, the presence of which is natural and the concentration is not due to human influence.

5. Evaluation of results

5.1. Pollution load index

The Pollution Load Index can show us whether an area or part of an area is contaminated by heavy metals (Harikumar *et al.,* 2009).

- For PLI>1 values, the area is considered contaminated.

- For PLI values <1, the area is considered to be unpolluted.

According to the above represented calculations, at 4 sampling points (A10, A12, L1, L2), the PLI was higher than the unit. Thus, the marine zone in Menidi and Paliovarka and the lakes of Rodia and Tsoukalio is considered to be polluted. At the other 3 sampling points (A2, A14, L3), the PLI was less than one. Thus the marine zone in the Pogonitsa and Vonitsa and Logaros lagoons is considered to be unpolluted. Figure 2 shows the map with the sampling positions and the indication of the pollution that characterizes them.

The total PLI environment for all the above sampling positions is PLItotal = 0.91. This value, although less than a unit, is very close to it, so the result is not reassuring. We cannot, however, consider it a representative value for the entire Amvrakikos bay because the sampling points used were few in relation to the volume of the bay.

Figure 2. Area contaminated by heavy metals according to the PLI Index, in Amvrakikos Bay

High PLI values at L1 - Rodia and L2 - Tsoukalio lagoons are determined. Similar studies (Karagiorgos *et al.,* 2006), explain that higher contents of heavy metals in this area due to the fact that the lagoon system is supplied by terrigenous material originating in the Rivers Arachthos and Louros and also directly by the weathering of rock formations. Due to the anticlockwise water circulation prevailing in the Amvrakikos Gulf, particle-rich riverine waters from the Louros River entrain the Tsoukalio and Rodia lagoons. The fact that the positions A10 and A12 are close to urban centers is the most likely reason for the high value of PLI index.

Table 5. Igeo calculated by equation (5)

Metals	Cu	Cr	Ni	Pb	Zn	Co	Mn	Fe
Igeo - A2	-1.275	-0.52	1.604	0.057	-6.084	0.189	0.724	-1.549
Igeo - A10	-0.903	-0.804	0.993	-0.51	-1.712	-0.07	0.381	-1.875
Igeo - A12	-0.513	-0.695	1.128	-0.09	-1.508	0.131	-0.89	-1.809
Igeo - A14	-1.777	-2.179	-0.57	0.163	-3.042	-0.32	-0.01	-3.094
Igeo - L1	-0.952	-0.862	0.658	0.519	-1.847	-0.03	0.064	-1.96
Igeo - L2	-0.968	-0.208	0.594	0.404	-1.801	0.015	0.089	-1.74
Igeo - L3	-1.18	-1.127	0.517	0	-2.292	-0.05	0.233	-1.883

5.2. Geoaccumulation index

The Igeo Index is used to assess the degree of metal pollution in terrestrial, aquatic and marine environments. The degree of pollution from metals is distinguished in

6 classes of contamination. Table 7 shows the degree of sediment contamination, according to the Igeo Index.

As can be seen from Table 8, the Amvrakikos bay is more burdensome Ni metal, followed by the Mn and Pb metals.

All metals belong to classes 0.1 and 2. This means that the regions according to the Igeo Index are from unchanged to slightly damage. A possible explanation of these results is the charge by human activities such as industrial waste, urban waste water (lacking sewage treatment facilities) and surface runoff (mainly agriculture and livestock).

Table 6. EF calculated by equation (6)

Metals	Cu	Cr	Ni	Pb	Zn	Co	Mn
EF - A2	1.21	2.04	8.89	3.04	0.04	3.34	4.83
EF - A10	1.96	2.1	7.3	2.58	1.12	3.49	4.78
EF - A12	2.46	2.16	7.66	3.28	1.23	3.84	1.88
EF - A14	2.49	1.89	5.74	9.57	1.04	6.86	8.5
EF - L1	2.01	3.95	6.14	5.58	1.08	3.82	4.07
EF - L2	1.71	5.33	5.04	4.42	0.96	3.37	3.55
EF - L3	1.63	1.69	5.28	3.69	0.75	3.56	4.33

Table 7. Igeoclassification (Muller, 1969)

Igeo	Igeo - Class	Description of sediment quality
< 0	0	Uncontaminated
0-1	1	Uncontaminated to moderately contaminated
1-2	2	Moderately contaminated
2-3	3	Moderately to strongly contaminated
3-4	4	Strongly contaminated
4-5	5	Strongly to extremely strongly contaminated
5	6	Extremely contaminated

Table 8. Gradient load of Amvrakikos sediments based on the maximum Igeo class for each metal

Igeo	Igeo - Class	Metals
< 0	0	Cu, Cr, Zn, Fe
0-1	1	Pb, Co, Mn
1-2	2	Ni
2-3	3	
3-4	4	
4-5	5	
5	6	

According to Karageorgis *et al.* (2001), high element (Pb, Cu, and Zn) contents and Igeo (geoaccumulation index) values were found for the clay fractions in the northern part of Pagassitikos Gulf. This enrichment is attributed to the discharge of raw domestic and industrial effluents of Volos city and port before the WWTP was installed. The dispersal of pollutants is essentially controlled by diffusion from point sources (city, port and WWTP) and is limited to Volos Bay. Relatively high Mn levels are ascribed to digenetic formation of manganese carbonates (antigenic phase), whereas Cr and Ni are elevated due to weathering of ultrabasic formations on land.
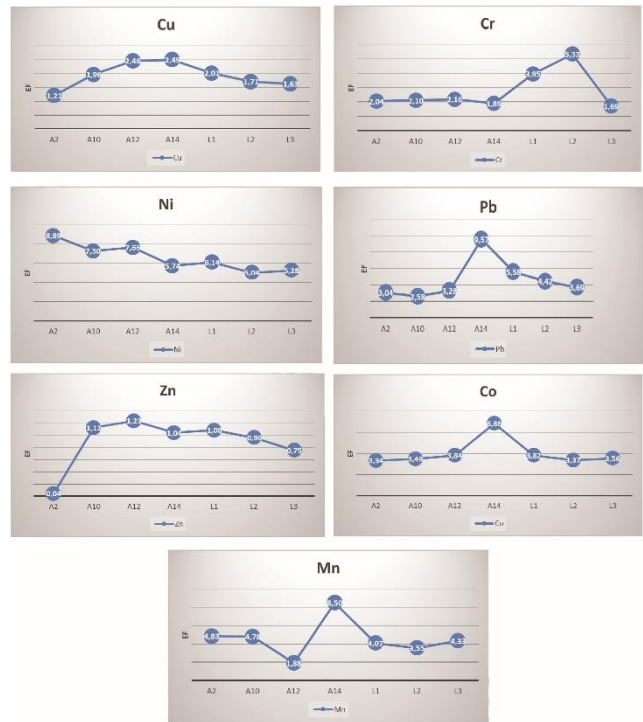

Figure 3. Diagrams Enrichment Factor of Cu, Cr, Ni, Pb, Zn, Co, Mn

5.3. Enrichment factor

The Enrichment Factor is used to distinguish between metals originating from anthropogenic activities and from natural processes, assessing the degree of anthropogenic effect. In Table 9, the categories of infection according to the Enrichment Factor are presented.

Table 9. Five contaminant categories on the basis of the EF (Sutherland, 2000)

EF	Contamination Degree
< 2	Deficiency to minimal enrichment
2-5	Moderate enrichment
5-20	Significant enrichment
20-40	Very high enrichment
40	Extremely high enrichment

Figure 3 shows the graphs of the Metal Enrichment Factor, based on the calculations of Table 9.

Zn is the metal with the lowest degree of enrichment, with values ranging from 0.04 to 1.23, while Cu and has a minimum of enrichment with values from 1.21 to 2.49. Mn and Cr belong to the categories of minimum and significant enrichment, with maximum values of 8.5 and 5.33 and minimum values of 1.88 and 1.89 respectively. Co and Pb show moderate to significant enrichment with minimum values of 3.34 and 2.58 and maximum values of 6.86 and 9.57 respectively. It is worth noting that Pb showed the maximum enrichment value of all the metals studied. Finally, Ni belongs to the category of significant enrichment, since all of its values exceed 5, with a minimum value of 5.04 and a maximum of 8.89. Based on the above, it is conclude that the concentrations of Cr, Mn,

Co, Pb and Ni are significantly affected by various anthropogenic activities.

Similar study from Argostoli Bay (Panagos *et al.,* 1990) of the metals examined Mo, Pb, Cr, Ni, Co and Zn were found to be enriched relative to normal shallow water sediments, their degree of enrichment being in the following order Mo › Pb › Cr › Ni › Co › Zn. The highest values of Pb in Argostoli Bay, would suggest that they should be related to the dissemination of oil in sea from the oil tanks occurring at the eastern coast.

Highest values of EF of Cu, Pb, Co and Mn in Vonitsa area observed. Due to the fact that this position is very close to the Vonitsa city and there present are significantly influenced by the various anthropogenic activities. Also at Panagos *et al.* (1990), study highest values of Cu and Zn are found at the outfalls of domestic sewage of Argostoli and Lixouri area.

Survey was performed at the beginning of the operation of Athens' sewage primary treatment unit, in the he Saronikos Gulf, indicated a chronic oil-pollution in thesediments. The industrially impacted Elefsis Bay sediments suffered from heavy petroleum pollution, while the sediments near the sewage outfall had a moderate degree of pollution (Sklivagou *et al.,* 2008). The concentration of heavy metals, in Thermaikos Gulf, increased with decreasing mean grain size, suggesting their association with the fine fraction of the sediments, the clays and the organic matter, (Voutsinouu and Varnavas, 1995). In Aetoliko, Greece lagoon, large quantities of pollutants are float on the top of an intermediate cold layers, in the form of particulate matter, (Varnavas, 2005). This studies shows that it is necessary to continuously monitor the natural, chemical and biological characteristics, as well as to take appropriate protective measures immediately in order to preserve the entire ecosystem.

6. Conclusions

Climate conditions, area morphology, intense stratification, poor water circulation and renewal, nutrient enrichment, low oxygen concentrations, and high concentrations of metals in sediment are the main causes of degradation of the Amvrakikos ecosystem.

Based on the PLI indicator, for the determination of the metal pollution in the area, the marine zone in Menidi and Paliovarka as well as the Rodia and Tsoukalio lagoons are considered polluted, while the sea zone in the area of Pogonitsa, Vonitsa and the Logaros lagoon are considered unpolluted.

According to the Igeo Index, the sampling areas are unchanged to slightly burden by heavy metals. The largest charge is caused by Ni, followed by Mn and Pb.

Based on EF, the concentrations of Cr, Mn, Co, Pb and Ni are significantly influenced by the various anthropogenic activities.

For the above reasons, it is necessary to continuously monitor the natural, chemical and biological characteristics, as well as to take appropriate protective measures immediately in order to preserve the entire

ecosystem. The present approach and their results can be used to monitor the long-term effects in the area.

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