



Consideration of geo-statistical analysis in soil pollution assessment caused by leachate breakout in the municipality of Thermi, Greece

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ABSTRACT

Soil heavy metal pollution is the driving force to various health problems as well as reprehensible soil quality. Landfill leachates are one of the focal sources of soil and underground water pollution. A total number of 120 soil samples were taken from three different soil horizons and tested for heavy metal content, and their physical and chemical properties were measured. Approximately, 1,500 m³ of leachates were released into the main local stream and pour to an area of 800 ha used for agriculture practices and olive cultivation. The designated area is located in the municipality of Thermi, Prefecture of Thessaloniki, Greece. Total extractable concentrations of seven different heavy metals were detected using atomic absorption spectroscopy (AS, Cd, Pb, Zn, Cu, Mn, and Ni)—and were elucidated to be slightly above the average of the universal allowable concentrations. Inverse distance weighting interpolator was implemented, and weighted linear combination was used to assess the overall pollution risk. Different multivariate analyses were implemented to point out the relationship between the experimented heavy metals. The risk of having soil heavy metal pollution at Tagarades is incontestable; nevertheless, the underground water of Tagarades in the meantime is not under jeopardy but the underground water pollution threat constantly exists.

Keywords: Groundwater quality; Geo-statistical analysis; Heavy metals pollution; Soil quality; Remediation techniques

1. Introduction

Soils secure vital functions to humans and livestock such as crop production, store and cycle water to sustain human demands [1]. Soils in principle are heterogeneous and contain a multiplicity of soil features during the process of soil formation [2]. Mineral fractions,

organic content, air cavities, and living organisms are the main constituents of many soil types [2].

Heavy metals occur naturally in soils as a result of the continuous geological process, specifically, underground materials erosion and alteration [3]. Several heavy metals are essential to different biological processes in terms of enzymatic activity upon which many life forms depend on [4]. However, chronic

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exposure even at very low concentration levels to any heavy metals was categorized as a human and/or environment health risk [5].

In 1987, United State Environmental Protection Agency (USEPA) lists 13 heavy metals as priority pollutants. This list contains arsenic, beryllium, cadmium, chromium, copper, lead, manganese, mercury, nickel, selenium, silver, thallium, and zinc. In 2002 at the sixth Community Environment Action Programme, soils had been subjected to specific protection policies at EU community level based on Soil Framework Directive (1999/45/EC). A common framework has become justified in order to articulate the efforts of the member state to improve the protection of soils and their sustainable use and also to control trans-boundary soil degradation effects [5,6]. The communal apprehension in many developed urban communities is the term pollution [7,8]. Various health problems related to pollution is well documented and well recognized by the general community [9]. Eventually, many types of pollutions are usually reached and preserved in soils due to the continuous action between the anthropogenic activities and different elements of the environment [10].

Agricultural practices, industrialization in addition to other anthropogenic activities, are principally the sources of different kinds of soil pollutions. Soil pollution induced by heavy metals threatens human and livestock due to its toxicity [11]. Extended scholar works have pointed out the different sources of soil pollutions and their impacts on the health of both human and environment, e.g., vehicle emissions [12,13], quarrying activities [14,15], agricultural practices [13,16], and manufacturing and municipal wastes [12,17].

Landfill leachates contain toxic agricultural and industrial wastes which disposed into municipal systems with no sequence of proper treatments comprise a major threat to the surrounding groundwater resources [18]. Contamination of the underlying groundwater resources by landfill leachates was reported in several literatures [19,20]. Constructional design of landfill plays an important role in determining the foreseen risk of groundwater pollution by the leachates. Recent landfill sites constructed with liners effectively limit the leakage of the leachates unless the liners break [21,22]. Leachate carries several organic and inorganic compounds and enters through contaminated soil to the food chain and, as a result, of bioaccumulation causes serious health problems to humans [23].

In 1981, a landfill was established in the designated study area. In the first years of operation, between 1981 and 1987, the disposal of municipal wastes in the landfill was uncontrolled. In order to improve the disposal standards, in 1987, several

engineering interventions took place aiming to transform the area into a sanitary landfill. Lately, the landfill covers approximately an area of 60 ha and serves more than one million people from the broader area of Thessaloniki city.

The aim of the current research was to investigate the extent of heavy metal pollution through different soil horizons within the designated study area due to the leakage of landfill leachates.

2. Materials and methods

2.1. Study area

The study area is located in Thermi municipality in the vicinity of the villages Tagarades, Trilofos, and Agia Paraskevi, prefecture of Thessaloniki, in North Greece. The study area is shown by the orthorectified image in Fig. 1. The waste load of the landfill is 1.368 in/d. Landfill leachates are congregated in the adjacent lagoon. Lately, a firebreak took place in the landfill and the walls of the lagoon bent. An excess of 1,500 m³ of leachates then was released into a local stream network and contaminated the surrounding area of 800 ha, and land owners noticed that leachates remained for roughly 10 months. The contaminated areas are principally used for agricultural activities, irrigated and non-irrigated annual crops. The agriculture activities comprise mainly olives cultivation as well as annual crops. Recently, more than 6 million tons of urban wastes have been disposed in the area.

2.2. Soil analyses

A total number of 120 soil samples were collected from the study area where the leakage took place.

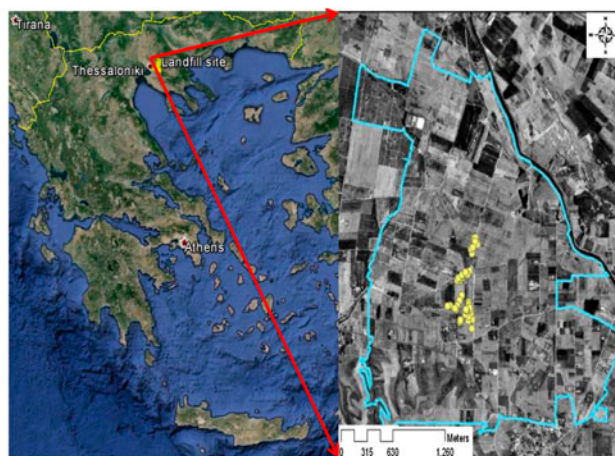


Fig. 1. Location of the study area.

Forty soil samples were collected from each different soil horizon (A: <30, B: 30–70, and C: <100 cm) located adjacently to the main water stream in the area. Soil samples were collected from upstream toward downstream in accordance with farmers' and land owners' permission; this was a limitation factor of better distribution of soil sample. The sample collection started after a major rain incident in succession to the fire-break. Physical (clay, silt, and sand) and chemical analyses (pH, calcium carbonate, electric conductivity (EC), and organic content) were performed including all samples.

2.3. Heavy metals detection

For heavy metal extraction, 100 mg of soils was weighed into a 50-mL volumetric plastic tube. To each tube, 10 mL of 4% acetic acid or 5 mL of 0.1 M HCl was added. For the acetic acid extractions, the tubes were allowed to sit overnight in darkness (up to 24 h). The tubes containing the HCl extractant were heated in a water bath at 37°C in darkness for one hour with agitation. This was followed by one hour in the water bath in darkness without agitation. These extractions were then filtered through 0.45- μ m membranes prior to analysis. The liquid sample is aspirated and mixed as an aerosol with combustible gasses (acetylene and air). The mixture is ignited in a flame of temperature ranging from 2,100 to 2,800°C. Heavy metals detection was performed following USEPA [24], using atomic absorption spectrometer (Perkin Elmer 2100).

2.4. Statistical analysis

2.4.1. Principal component analysis

Principal component analysis is an important data transformation technique used in several environmental complexed problems [25]. PCA is used basically to discriminate multidimensional variables into distinguishable elements. Adopted transformation of Rumel [26] is a reduction technique that leads to describe variables which are non-correlated with the first component containing most variance and the succeeding component.

2.4.2. Geo-statistical analysis

Geo-statistical analysis under GIS environment was used to determine the spatial variability of the heavy metal content of the soils. Samples for laboratory tests were taken from the study area in Tagarades in a random system from 0 to 30 cm depth. Spatial

distribution equation was carried out according to Weibel [27], and the interpolation equation was carried out according to Stoer and Bulirsch [28].

3. Results and discussion

3.1. Soil physical analysis

The bulk of Tagarades soil is sand as the percentage of sand in the tested soil is calculated to be about 52%, while both of silt and clay are calculated to be 22 and 26%, respectively. The majority of Tagarades soils are falling into two groups according to soil textural classes (Fig. 2). The first group is composed of sandy loam and sandy clay loam soil while the other group is composed of clay loam and minor loam soil according to soil textural classes adopted by FAO [29].

3.2. Soil chemical analysis

Different types of chemical analyses were applied to the designated 40 soil samples to elaborate the chemical properties that affect heavy metal behavior in Tagarades soil. pH, EC, CaCO₃ content, and cation exchange capacity are the most common used analyses variables, as shown in Table 1. The pH level can affect the solubility and the mobility of heavy metals; soils of Tagarades are shifted toward slightly alkaline soils. Alkaline soils tend to have high sodium, potassium, magnesium, and calcium contents. The latter two elements tend to form calcareous deposits on buried structure with protective properties against heavy metal corrosion. CaCO₃ content in the experimented

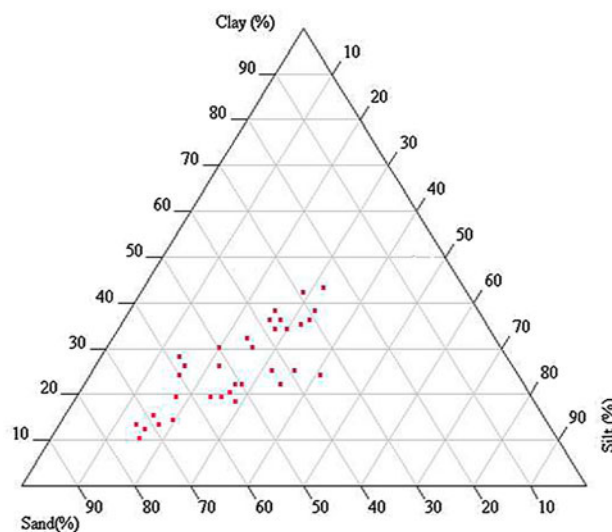


Fig. 2. Soil textural classes Tagarades area.

Table 1
Soil chemical analyses in Tagarades

Sample #	pH	EC (mS/cm)	CaCO ₃ (%)	K ⁺ (mg/l)	Na ⁺ (mg/l)	Ca ²⁺ (mg/l)	Mg ²⁺ (mg/l)	OC (%)
1	8	0.79	1.9	44	86	175	12	1
2	7.8	0.46	3.1	7.8	48	47	4.2	0.9
3	7.8	0.41	1	17	19	58	4.5	1.1
4	7.5	1.03	0.4	45	130	66	7.1	1.4
5	8	2.11	2	71	370	106	17.5	1.6
6	8.1	0.74	1.8	56	190	46	0.6	1.7
7	7.9	0.69	2.9	3.2	170	25	3.2	1.9
8	8	0.62	1.8	41	77	26	0.4	0.9
9	7.6	0.35	0.4	11	29	19	5.1	1.6
10	7.9	0.85	0.7	41	150	12	6.7	1.7
11	8.1	1.28	1.3	57	220	52	9.3	1.9
12	7.8	1.63	1.1	42	230	98	10.7	1.6
13	7.1	0.34	1.5	16	30	26	3.6	1.4
14	7.5	0.37	0.4	27	6	50	7.1	1.4
15	6.6	1.88	0.9	52	210	132	21	1.8
16	7.4	4.45	0.7	200	500	383	49.1	1.8
17	7.2	6.32	0.9	110	710	642	79.2	1.8
18	7.7	1.23	0.9	48	180	94	29.8	1.3
19	6.9	0.26	0.7	6.9	33	15	39	0.3
20	6.8	0.29	0.9	1.7	27	18	51.7	0.1
21	7.7	0.41	0.6	3.8	46	6	72.7	0.3
22	7	0.48	0.1	22	22	16	7.4	0.2
23	7	1.1	0.2	38	160	30	8.2	0.8
24	6.9	1.35	0.1	110	98	51	12.5	0.4
25	6.7	1.29	0.2	0.2	150	92	17	0.2
26	6.5	1.68	0.1	2	3	108	23	0.2
27	6.3	0.7	0.2	18	58	71	16.4	0.4
28	7.9	0.78	1.1	42	80	125	27	0.3
29	5.2	0.51	0.1	18	26	85	16.2	0.1
30	6.9	1.39	0.4	16	180	155	30.4	0.2
31	7.6	1.62	2.6	73	160	128	17.1	0.4
32	7.9	0.4	2.2	17	32	69	7.5	0.2
33	7.9	0.78	1.8	90	33	92	7.9	0.7
34	7.7	0.58	1.5	17	32	86	2	0.5
35	7.7	0.51	1.4	58	190	190	24.9	0.4
36	6.6	1.37	0.1	45	170	106	17.5	0.6
37	6.9	0.7	0.2	20	19	96	12.8	0.6
38	7.9	0.83	1.3	35	86	90	10.5	0.6
39	6.8	1.46	0.2	64	160	129	20.9	1.5
40	8	0.63	0.8	37	17	78	10.1	1.5

soils is generally at low values. In soils with low CaCO₃ content, the risk of deterioration of the metals can be predicted from the soil type. Soil pH of Tagarades is classified to be slightly alkaline as it is shown in Fig. 3(a). The grand mean of the 40 tested soil samples is 7.37 with an extremely high pH value of 8.1. On the other hand, the lowest pH value is 5.2 which are classified to be slightly acidic. EC of tested soil intended to have an average value of 1.11 mS/cm. The maximum value of EC was measured in midstream at

soil sample number 17 and had the value of 6.23 (mS/cm) while the minimum measured value was in midstream at soil sample number 19 (0.26 mS/cm) as shown in Fig. 3(b). Total CaCO₃ content (Fig. 3(c)) has an average value of 1.08%, which is safe for growing crops with an extremely high value located upstream at soil sample number 2 (3.1%).

Soluble salts are tested in Tagarades soil to inspect the deicing effect of the salts on the heavy metal mobility. The tested soluble salts are shown in the

following Fig. 3(d)–(g) for K^+ , Na^+ , Ca^{2+} , and Mg^{2+} (mg/l). Figures show that the extreme values are located only in midstream at soil sample numbers 16 and 17 for all the tested soluble salts (200, 500, 383.4, and 49.1 mg/l, respectively).

Organic carbon percentages are clearly distinguished at soil sample number 20 (midstream), where almost the first half of the soil samples is above the average (0.92%), meanwhile, the second half is below the average. The maximum percentage is 1.9% at

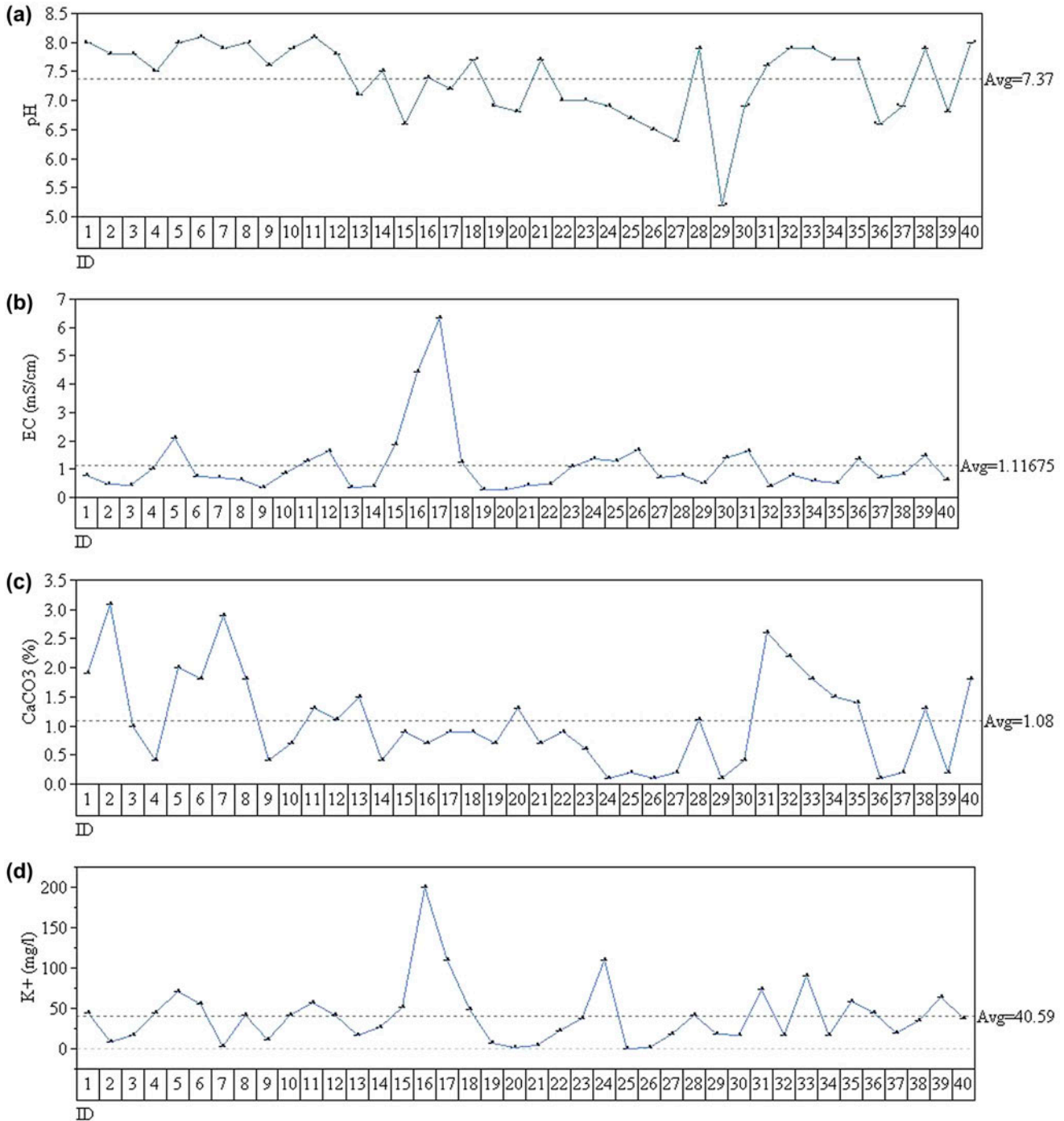


Fig. 3. Tagarades soil chemical analysis: (a) pH, (b) EC (μ S/cm), (c) total $CaCO_3$ (%), (d) soluble K^+ (mg/l), (e) soluble Na^+ (mg/l), (f) soluble Ca^{2+} (mg/l), (g) soluble Mg^{2+} (mg/l), and (h) organic carbon (%).

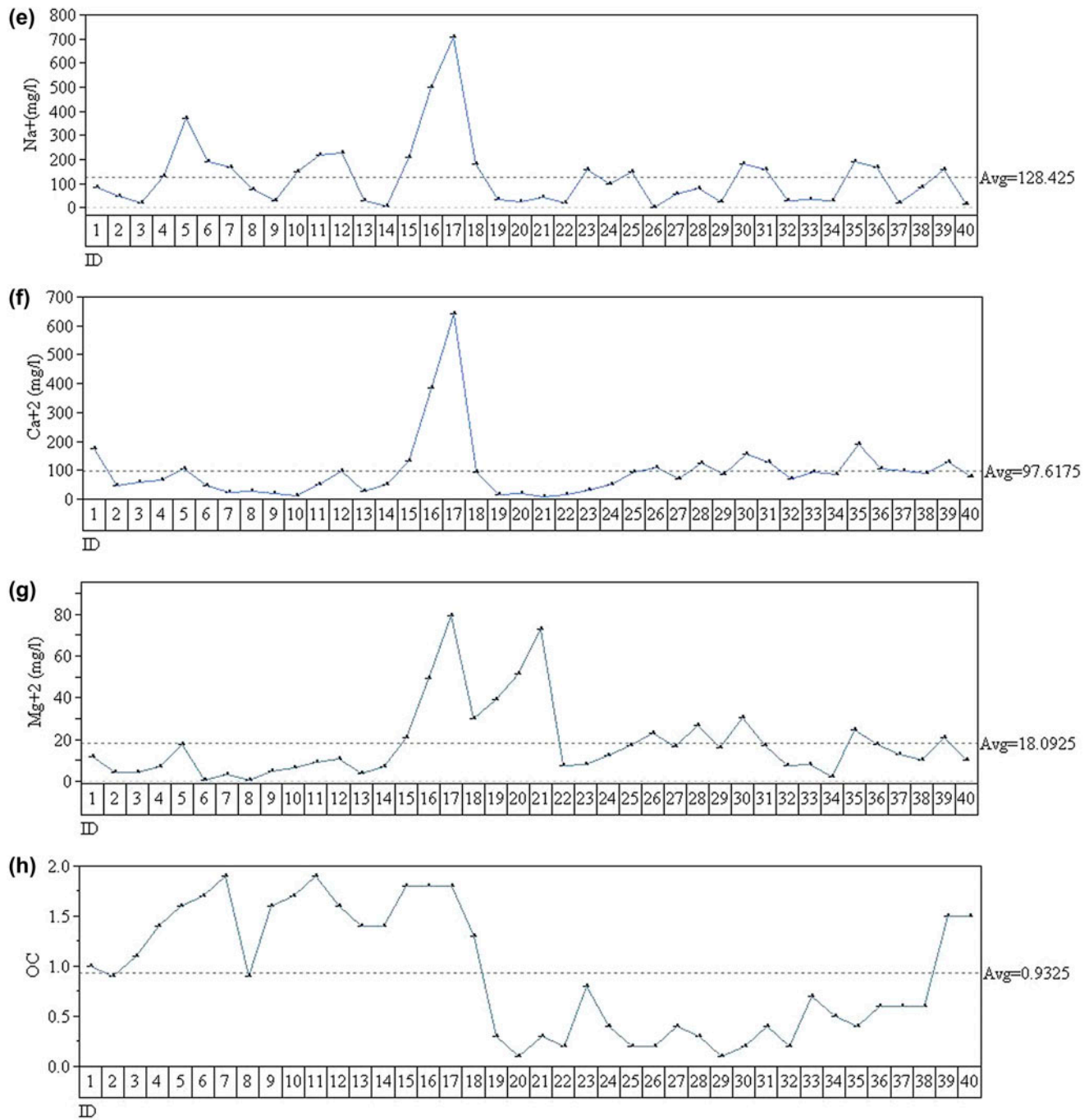


Fig. 3. (Continued).

upstream soil sample number 7, but on the other hand, the minimum percentages are 0.1% at downstream soil sample number 20 and 29 as it is shown in Fig. 3(h).

The organic substance in soil is the most vital constituent concerning metal retaining [30]. Organic

substance modifications may enhance metals solubility by producing intermediate compounds that chelate the metals to avoid their sorption and to endorse percolating through materialization of soluble metal complexes [31].

3.3. Heavy metal detection

Atomic absorption spectrophotometer detects seven different elements described to be heavy metals in the 40 selected soil samples of the Tagarades area. Detected metals (As, Pb, Cd, Zn, Cu, Mn, and Ni) were ordered accordingly to their toxicity effects [32]. The detected total extractable concentrations of the tested heavy metals are verified to be slightly higher than the maximum allowable values of heavy metals in soil used in different countries [33] as shown in Table 2.

Detected heavy metals fell into two categories based on the heavy metals mobility in the tested soil. The first category includes As, Pb, Cd, Zn, and Cu. This heavy metal category tends to accumulate in Tagarades soil at a depth of 100 cm. More than 50% of these heavy metal total extractable concentrations are found at a depth of 100 cm as is calculated to be as follows: 52.6, 67.8, 67.9, 69.2, and 81.8% for Cd, Cu, Pb, Zn, and As, respectively. On the other hand, the second category includes only Mn and Ni, where the measured total extractable concentrations were considerably low—28.6 and 38.9% for Ni and Mn, respectively.

Previous research of Norrström and Jack [34] reported that the larger part of As, Pb, Zn as well as Cd and Cu is presented in a chemical fraction which is available to leach when exposed to a high salt concentration, reduction condition, or decreased pH. This doubtlessly can give one rationalization to high total extractable concentration through the soil profile. Heavy metal transportation, especially a downward movement through Tagarades soil profile, may not reach the groundwater in the near future. It is a risk from the long-term point of view, and the risk would potentially be higher, especially in high-precipitation seasons. Generally, Cd and Cu total extractable concentrations in the designated study area are much lower than As, Pb, and Zn due to its relatively low

corrosion rate, attributable to the affinity of these heavy metals to bind with clay materials [35].

Principle component correlation and analysis suggest that the examined heavy metal total extractable concentrations also fall into two groups, where As, Pb, Cu, and Zn form the first component, while Cd, Mn, and Ni form the second one. As and Zn total extractable concentrations are distinguished from the remaining elements of the first component. Furthermore, Cd total extractable concentration is also distinguished from the remaining elements of the second component (Fig. 4).

The scatter plot matrix and the cluster correlation were performed to visualize the former multivariate analyses in a way to support the principle component analyses of having two components. From the scatter plot matrix, Mn and Ni were obviously strongly correlated. Additionally, As and Zn is not correlated to any other heavy metal total extractable concentration, and both of them are distinguishable from the remaining weathered heavy metals. The remaining heavy metals are more or less correlated (Fig. 5).

The group of clustered elements includes Pb, Zn, and Cu as heavily influenced by anthropogenic means that are caused primarily by landfill site leachate [36]. However, the first cluster of heavy metals group including Cd, Mg, and Ni shows the influence of the anthropogenic input from agrochemicals practices and fertilizers causing soil deterioration as well as soil erosion [37,38]. Nevertheless, considering the landfill leachates as a source of pollution of the second and the third clustered group containing the remaining investigated heavy metals, it cannot be confirmed due to the absence of soil sample inside or nearby the landfill site.

3.4. Geo-statistical analysis

Consistent cross-validation method was implemented to evaluate different interpolator accuracies.

Table 2

Maximum allowable limits (L.A.M.) for heavy metals in soil (ppm) used in different countries following Kabata-Pendias [33]

Heavy metals	Austria	Poland	Great Britain	Germany	Greece	Detected values
As	3	5	5	3	5	4
Pb	100	100	100	500	300	380
Cd	5	3	3	2	3	8
Zn	300	300	300	300	300	485
Cu	100	100	100	50	140	145
Mn	200	200	150	200	300	6,410
Ni	100	100	50	100	75	90

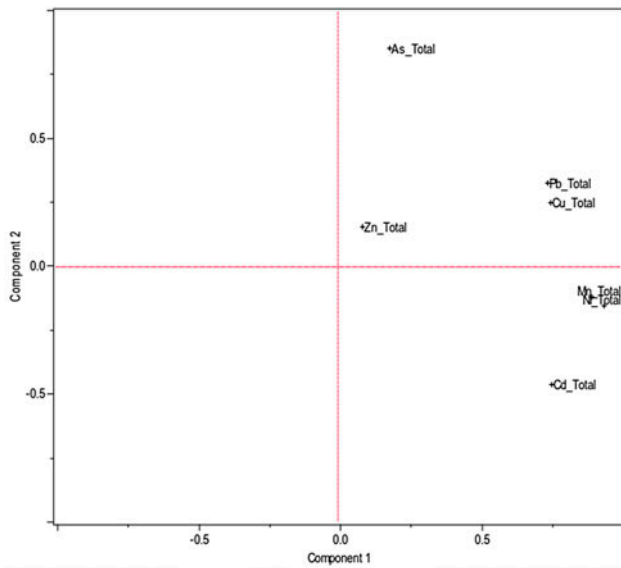


Fig. 4. Principal components analysis.

Inverse distance weighting interpolator is the adopted method for statistical reasons, as it has the lowest root-mean-square error (RMS). RMS is calculated to be 0.0146, 0.0151, 0.0153, and 0.0154 for inverse distance weighting, global polynomial, local polynomial, and Kriging, respectively. The buffer zone of the predicted values meant to be 250 m surrounding the tested sites, and the extent of the buffer zone is based on the maximal heavy metal mobility in most favorite soil following Warrick et al. [39] and Grathwohl et al. [22].

Natural breaks classification method was used to reclassify the prediction risk maps into four classes according to their toxicity levels into low, moderate, high, and severe [40]. The role of the classifier is based on squared error minimization from a class's means by creating internally homogenous groups but maintaining heterogeneity between classes.

Different interpolation techniques were used to produce a precise overall risk map of heavy metal pollutions based on the root-mean-square error (RMSE)

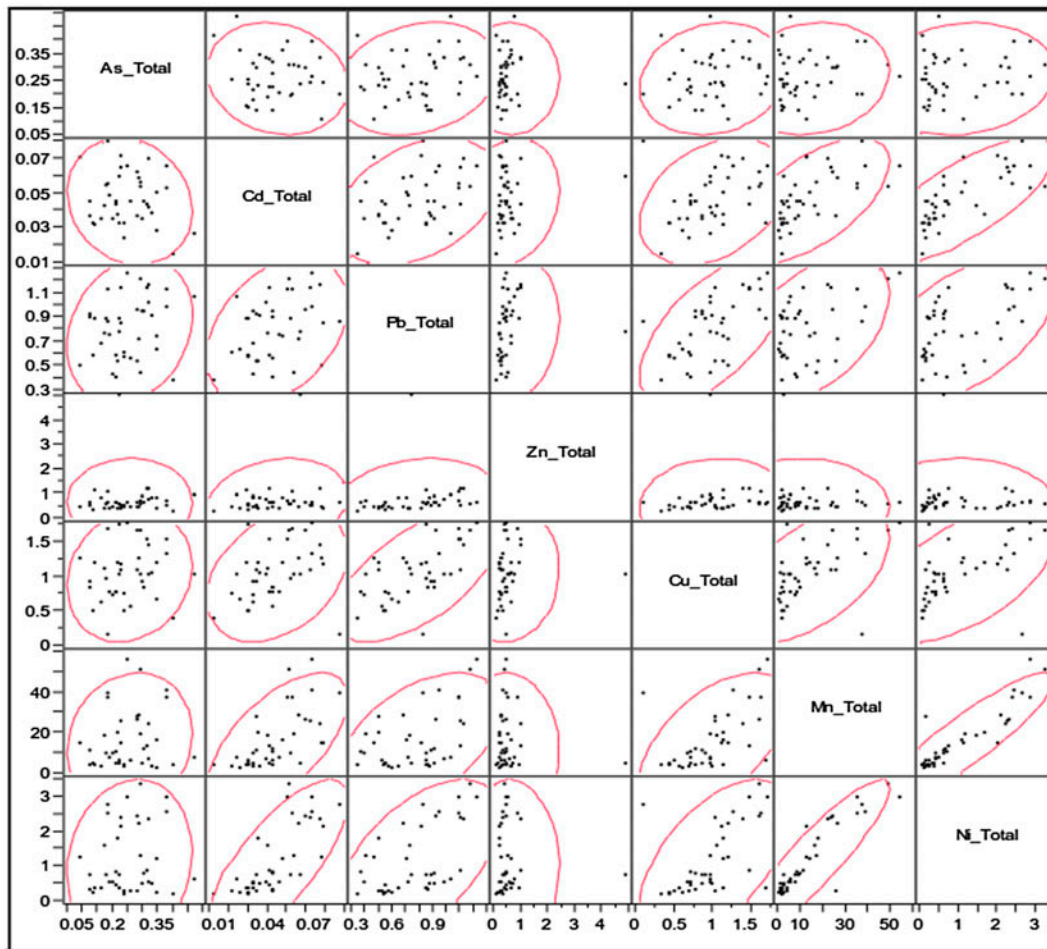


Fig. 5. Scatter plot matrix.

Table 3
Overall risk prediction map eigenvector of weights

Heavy metal	Weight	Rank
As	0.4266	7
Pb	0.2545	6
Cd	0.1434	5
Zn	0.0833	4
Cu	0.0464	3
Mn	0.0272	2
Ni	0.0186	1

evaluation. Principally, radial basis functions (RBF) showed better results [41]. This could be explained due to the fact that the RBF is considered as an exact interpolation technique, which means that the predicted values are identical to the measured values [42]. The interpolation is based on a mathematical function that smooths the generated surfaces by minimizing the surfaces curvature.

To predict the overall risk pollution map, the study requires an approach to identify parametric values in modeling the risk pollution according to multi-criteria analysis (MCA). In the absence of an original method with some guidelines for establishing priorities or aggregation rule, the need for MCA becomes essential and requires information on the relative toxicity of each heavy metal risk [39]. Under spatial decision support system environment, weight linear combination approach was implemented to assign different weights for each heavy metal total concentration to finalize the overall risk map according to heavy metal lethal

toxicity rank. Table 3 describes the different weight sets with a consistency ratio equal to 0.09. The relative toxicity of each heavy metal risk (Rank) is following Robert [32] according to the tested heavy metal lethal dose (Fig. 6).

Several remediation techniques are potentially considered in the current designated area. Isolation and containment, physical barriers made of steel, cement, bentonite, and grout walls can be used for capping, vertical and horizontal containment [43]. Solidification/stabilization technologies are usually applied by mixing contaminated soils or treatment residuals with a physical binding agent to form a crystalline or polymeric framework surrounding waste particles. In addition to micro-encapsulation, some chemical fixation mechanisms may improve waste leach resistance [24]. Soil washing is an ex-situ remediation technology that uses a combination of physical separation and aqueous-based separation unit operations to reduce contaminant concentrations to site-specific remedial goals [44]. Soil flushing is also an *in situ* extraction of contaminants from the soil via an appropriate washing solution. Water or an aqueous solution is injected into or sprayed onto the area of contamination, and the contaminated elutriate is collected and pumped to the surface for removal, recirculation, or onsite treatment and reinjection [24]. Phytoremediation uses plants to remove pollutants from the environment. The use of metal-accumulating plants to clean soil and water contaminated with toxic metals is the most rapidly developing component of this environmentally friendly and cost-effective technology [45].

4. Conclusions

The fate and the mobility of heavy metals fractions are determined by contact duration of heavy metals in soils and their interactions with soil features. Tagarades soils are with low clay content, slightly alkaline, and with the considerable content of organic matter, which plays an essential role in heavy metals temporary bindings with a surface area of soil particles. During rainy seasons with high precipitation, the potential risk of groundwater contamination by heavy metal corrosion products being transported through soil horizons is realizable. Clay content controls the horizontal and the vertical movements of heavy metal due to its ability to retain vertical permeability over horizontal one. The potential of having non-source groundwater pollution is very susceptible due to the larger time window of heavy metal corrosion products interactions with groundwater resources during winter time. The heavy metal examined values are slightly higher than the range of average metal concentration

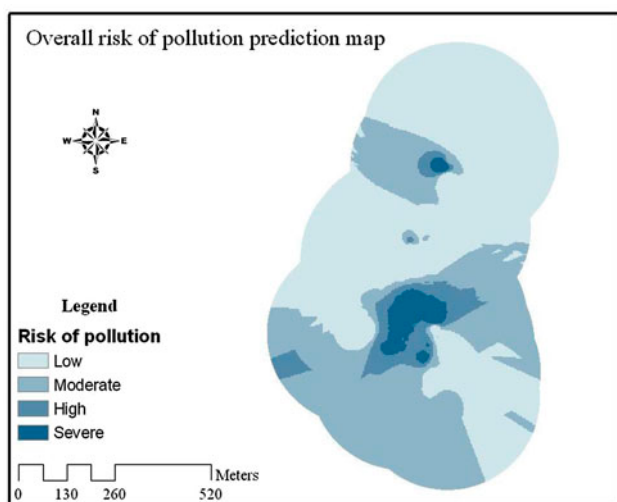


Fig. 6. Overall risk of pollution prediction map following IDW method.

toxicity level in Tagarades soils; therefore, proper remediation technique must be adopted. Finally, groundwater resources are plainly subjected to heavy metal contamination.

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