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Research Article
Spatiotemporal Variation of Heavy Metals Pollution in the Inegol Plain, Turkey

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Abstract

Background and Objective: The impact of pollution in the vicinity of overcrowded cities and from industrial effluents has reached a disturbing magnitude and is arousing public awareness. Therefore, the objective of this research was to determine the concentrations and characterize the spatial distribution of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) heavy metals in soil of Inegol Plain by using geostatistical approaches combined with GIS based on 17 soil samples in both winter and summer periods. **Methodology:** Ordinary kriging interpolation technique was used to estimate a variable at an unmeasured location from observed values at nearer locations. Moran's I and Getis-Ord Gi were used to investigate spatial autocorrelation of soil pollutants. **Results:** Very high heavy metal concentration levels were found throughout the study area. The local spatial autocorrelation analysis made clear that all heavy metals had important hotspots and coldspots and these areas should be targeted by best management practices to reduce heavy metal loads. Attention must be given to industrial activities performed within these high-risk areas. **Conclusion:** Results from this study can be used by ecologists, planners and managers to protect soil supplies.

Key words: Heavy metals, GIS, kriking, soil pollution, spatial autocorrelation

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Environmental quality and pollution is one of the most important problems in today's world. Industrialization and urbanization are the two main factors that decrease environmental quality. Thus, the relationship between the urban environment and human activities is revealed through the increase of pollutants in the natural environment[1].

Environment pollution affects the quality of pedoshphere, hydrosphere, atmosphere, litosphere and biosphere[2]. One of the main problems of pollution in overpopulated areas is soil contamination[3,4] via heavy metal contamination[5]. Soils could be contaminated by emissions from industrial areas and by high metal wastes[6,7]. Most commonly found heavy metals are arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn) and mercury (Hg)[4,7]. Heavy metals can be stored easily in the environment because they do not biodegrade[8]. The accumulation of heavy metals in the soil is from natural or anthropogenic sources. Natural accumulation of heavy metals in the soil occurs as a result of separation of rock piles and other pedagogic processes. In this situation, accumulation of heavy metals in the soil shows relatively lower concentration[9]. Industrial waste, mining activities, melting processes, sewage, atmospheric pollution, vehicles and burning of fossil fuels make up the anthropogenic sources of heavy metals in urban areas[10]. In this situation, more pollutants accumulate and spread in the area.

Protection of soil is vital for sustainable development and protection of ecosystems and biodiversity[10] because soil is the critical environment for interactions between bedrock, water, air and the biological environment. Contamination of the soil by heavy metals caused by disposal of urban wastes creates serious health problems[11-14]. Heavy metals directly harm human health through skin absorption and breathing dust[15] adversely affecting the central nervous system[11].

While analyzing the effect of heavy metals for environmental management and monitoring, the spatial distribution and concentration levels of pollutants must be determined[11-13]. Geographic Information Systems (GIS) and geo-statistics have opened up new ways of analyzing spatial distributions and spatial patterns of soil pollutants and to study spatial uncertainty and hazard assessment. The GIS can be a successful decision support system for environmental management and planning using different spatial models. The GIS technology and GIS-based geo-statistical techniques are widely used to analyze spatial distribution of heavy metals in the soil[1,12,13]. Several studies used GIS tool to show the spatial distribution of heavy metal pollution in urban areas[2,13].

The working group initiated by the European Soil Bureau suggested a program to provide a database for heavy metals in soil[16]. Databases for heavy metals in soils commenced in July, 2000 for all 15 Member States of the European Union. New and more detailed soil information for all of Europe is required to address heavy metal contamination. However, in Turkey, relatively little data are available on the extent of soil pollution.

As many cities in Turkey, Inegol is experiencing rapid urbanization and industrialization. Analyzing the effects of rapid urbanization and industrialization, which cannot be noticed in a short period is important for environment management and monitoring. It is hypothesized that heavy metals accumulated in urban soils would present a potential risk to the ecosystem. Therefore, the proposed objective of the study is to assess the spatial distribution and levels of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) heavy metal concentration in the soil of Inegol Plain by using geostatistical approaches combined with GIS. Specific objectives are to determine levels of heavy metal concentration in soils, to determine the spatial distribution of heavy metal concentrations and to specify the area above a specified threshold.

MATERIALS AND METHODS

Data and analysis

Site description: Study area is the Inegol Plain on the Bursa preference in the Marmara Region in the Western Turkey (Fig. 1). This area is located approximately between 40°09' Northern latitude and 29°49' East longitude. The study area shows basin characteristics in terms of morphology, hydrology and geology[17,18]. The whole study area covers a total land area of 142 km^2. The mean annual temperature is 13.8°C and the average annual rainfall is 427 mm. Formation of the plain is quaternary alluvium formed or deposited in a stream or river and this quaternary soil cover and alluvium, forming along the valley overlie the older units.

The human population of Inegol has been increasing steadily over the years: 76,908 in 1965, 215,375 in 2009 and 242,232 in 2014. The most important economic activities in the area are agriculture, wood working, furniture-making, wood products, textiles, tourism, food and machinery industry[19]. Although, Inegol has very fertile soil for agricultural functions, industry is the most important economical activity in the plain.

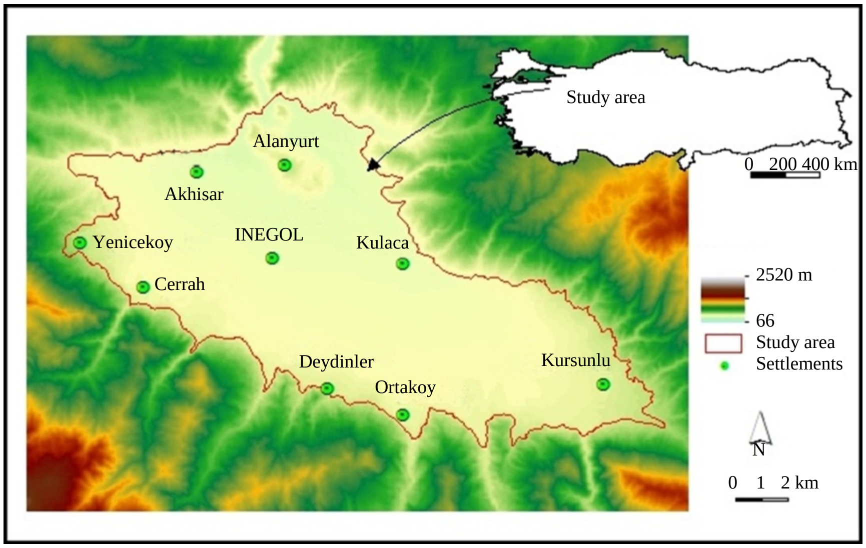

Fig. 1: Location map of the study area

Data and evaluation of soil samplers: For characterization of soil contamination, samples were obtained using a systematic grid-system. The study area was divided into a 3.0×3.0 km grid which included settlements, forested areas, agricultural areas and industrial areas. Samples were collected using the "Systematic random" method from soil pits at 0-20 cm depths, because anthropologic sourced pollutants accumulate on upper layers of soil[20-22]. Soil samples were taken from 17 different locations in July, 2014 and February, 2014. One of the samples was collected from randomly and four were collected from around the main sampling point. Five profiles were dug with a shovel at each grid. Equal amounts of these five samples were mixed and homogenized. Samples were collected with a plastic spoon and kept in 2 L PVC packages for transport and storage. On the border areas when sampling positions on the grid were close to paths, rivers or buildings, an alternative sampling position nearby was selected. The GPS was used to determine sampling locations.

After soil samples were air dried, they were sieved with a 2 mm sieve to remove stones and coarse materials, etc. Later, 20 g soil samples were taken for analysis. Total content of heavy metals were analyzed by "Inductively Coupled Plasma Mass Spectrometry (ICP-MS)" in the Bulent Ecevit University Science and Technology Application and Research Center. Next, pollutant concentrations were interpolated over the whole study area using geostatistical tools developed in GIS.

Geo-statistics and autocorrelation: Kriking techniques were used to analyze the spatial distribution of heavy metals in the study area and to predict the variable values of unsampled locations with help from the values of sampled locations. Ordinary kriking and indictor kriking are widely used methods in geostatistical analysis of soil contamination[23-25], because kriking methods have greater accurate in mapping and display of heavy metals in soil compared to other interpolation methods[26]. Ordinary kriking was used to create regional distribution maps and indictor kriking method was used to characterize the hazard posed by heavy metal concentrations at a specific threshold value in the study area. The indicator kriking was carried out by considering the attribute of the As>20 ppm, Cd>1 ppm, Cr>100 ppm, Cu>50 ppm, Ni>30 ppm, Pb>50 ppm and Zn>150 ppm as point measurements with values 0 and 1. Limit values in Turkey Soil Quality Control Regulation-TKKY[27] were used.

Local statistics are important because the magnitude of spatial autocorrelation of pollutants were not necessarily uniform over the area[28]. In this study, it is preferred to use global Moran's I[29] and Getis-Ord General G statistics to indicate general spatial autocorrelation. In order to capture the spatial heterogeneity of spatial autocorrelation and to identify the location of clusters, we used the Getis-Ord Gi* statistic developed by Getis and Ord[30] to identify spatial clusters of statistically significant high or low attribute values by indicating whether high values or low values (but not both) tend to cluster in a study area. A high value of Local Gi indicates hot-spots, while a low value indicates coldspots.

RESULTS AND DISCUSSION

Spatiotemporal distribution of as in soils in inegol plain: A small amount of As concentration is essential in animals[31] and beneficial in plants[32]. According to Asiam[33], As has generally been used as an indicator element to identify mineral deposits. However, high concentrations of As in soil may endanger the health of plants, foods and water for human beings[34,35]. As a result, As received new and global attention in the mid 1990s. Brandstetter *et al.*[36] indicated that As polluted environments are abundant in many regions of the world.

Concentrations of As in the soil in the plain are higher in winter than summer (Fig. 2a, b). The highest As concentrations (Fig. 2a) in winter are in the Southeastern part of the plain around the Kursunlu where there are mineral water facilities. Oylat hot spring is close to Kursunlu and also to kınık and Fresa mineral water facilities. Smedley *et al.*[37] reported that human beings can be exposed to As by soil with high levels of As, especially from mine wastes. Most affected areas are close to metal processing activities[38,39]. Mean As concentration in winter is over 20 ppm, which is the limit of Turkey Soil Quality Control Regulation. Mean As concentration in summer is over only in the northwestern side of the plain.

We conclude that there is a significant positive spatial autocorrelation in As concentration among the plain (Table 1). Positive z score indicates clustering of high values in the plain. There is a less than 1% likelihood that this high-clustered

Table 1: Results of global spatial autocorrelation

	As	Cd	Cr	Cu	Ni	Pb	Zn
Summer							
Moran's index	0.95	0.86	0.72	0.95	0.77	0.88	0.89
z-score	16.24	14.73	12.43	13.37	10.93	15.13	15.31
p-value	0.00	0.00	0.00	0.00	0.00	0.00	0.00
General G	0.06	0.06	0.05	0.06	0.07	0.06	0.06
z-score	3.92	2.34	-0.03	5.28	4.80	3.25	0.80
p-value	0.00	0.02	0.98	0.00	0.00	0.00	0.42
Winter							
Moran's index	0.88	0.86	0.73	0.82	0.73	0.83	0.75
z-score	15.20	14.76	12.67	14.07	12.78	14.28	12.90
p-value	0.00	0.00	0.00	0.00	0.00	0.00	0.00
General G	0.06	0.06	0.05	0.06	0.06	0.06	0.06
z-score	2.00	4.18	-1.21	3.43	0.56	3.98	2.73
p-value	0.05	0.00	0.23	0.00	0.57	0.00	0.01

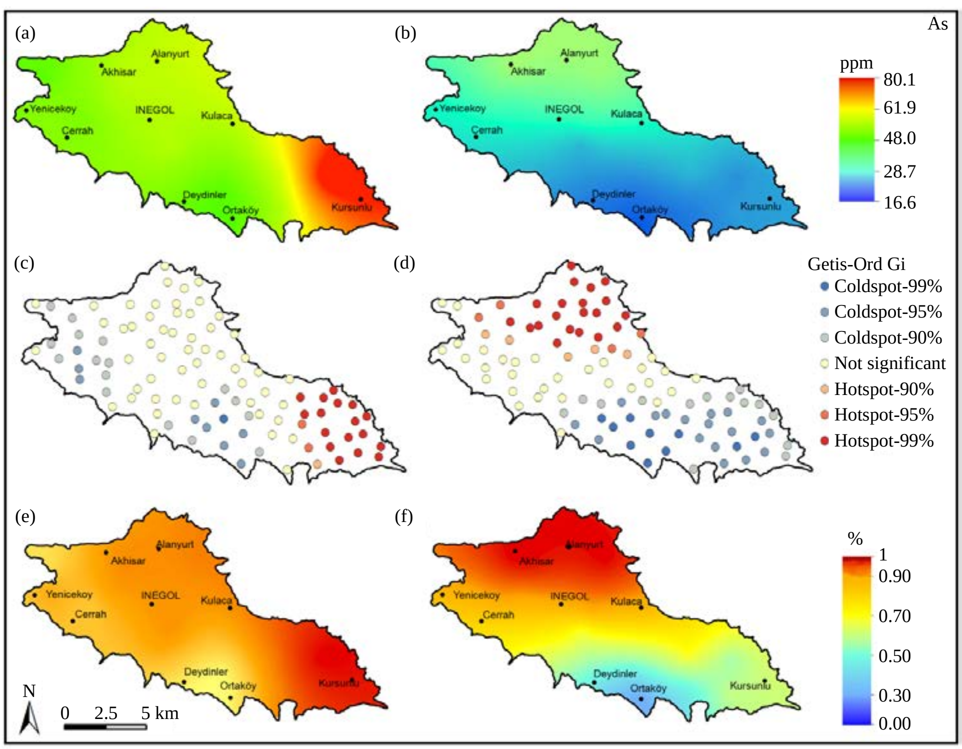

Fig. 2(a-f): As concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, (b) Spatial distribution in summer, (c) Local G-statistics in winter, (d) Local G-statistics in summer, (e) Probability of As values exceeding 20 ppm, in winter and (f) probability of As values exceeding 20 ppm in summer

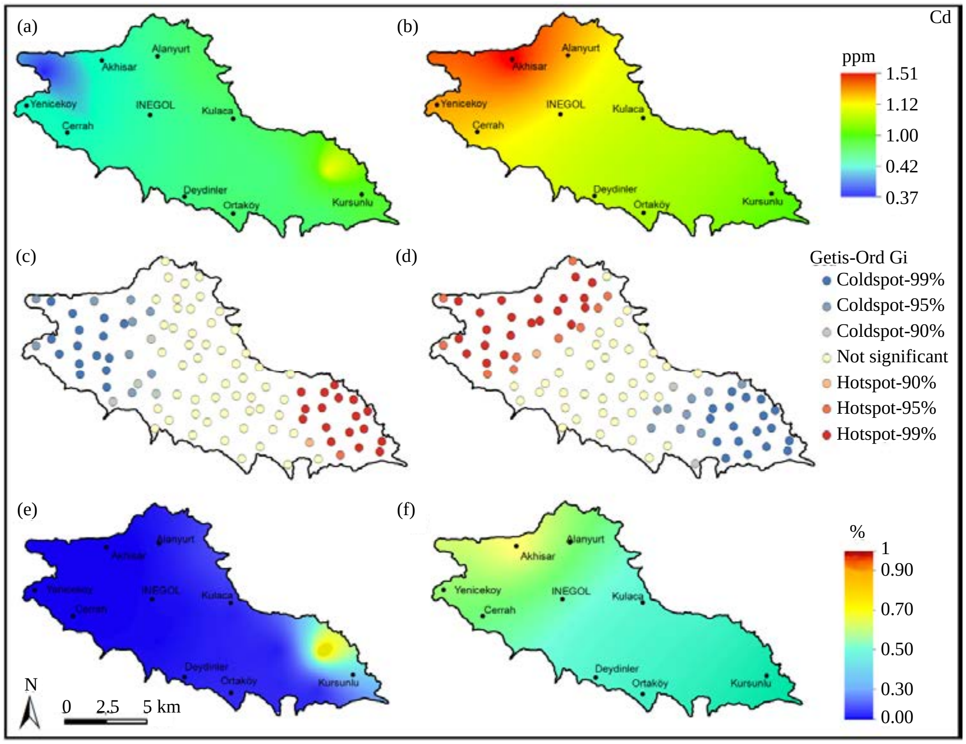

Fig. 3(a-f): Cd concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, (b) Spatial distribution in summer, (c) Local G-statistics in winter, (d) Local G-statistics in summer, (e) Probability of Cd values exceeding 1 ppm, in winter and (f) probability Cd values exceeding 1 ppm in summer

pattern could be the result of random chance. Two apparent "Hotspots" in the Fig. 2c and d are remarkable: In around Kursunlu in winter and in Northeast of the Inegol, around the Organized Industrial Zone in summer.

The indicator kriging was carried out by considering the attribute of the 20 ppm threshold level. A map showing the probability of exceeding the threshold is shown in Fig. 2. The indicator kriking map (Fig. 2e, f) indicates that As content of soils is very high all over the plain. It is determined that only a small part of the plain in the Southwest in summer is under the 50% probability. This indicates that the plain is contaminated with As and thus requires treatment of the soil. The plain is mainly farming communities and this high possibility could be causing low crop yields.

Spatiotemporal distribution of Cd in soils in inegol plain: The Cd is known as a non-essential metal and it presents in the soil naturally. Soil Cd is increased by atmospheric deposition, human activities and agricultural management like applications of fertilizers and sludge[40]. Because Cd is a highly toxic and carcinogenic metal, it has a strong effect on crop quality[41] and it threatens human health[42]. Because it could be dangerous for humans, risk assessments of Cd on human health in Europe have been done[43].

Opposite of As, concentrations of Cd in the soil in the plain is higher in summer than winter (Fig. 3a, b) In winter, the highest Cd concentrations are in the southeastern part of the plain, around the Kursunlu where there are mineral water facilities. It is hypothesized that Cd concentration increase in the edge of the plain, because of geothermal and mineral waters in winter. However, in summer high concentrations of Cd are in the North part of the study area, around the Inegol. In winter, the South part of the Inegol plain has low concentrations of Cd. Previous studies suggest similar results. For example, Olatunji *et al*.[44] reported Cd concentrations related with industry. They emphasized that concentration levels increase according to intense population, traffic and industrial activities[45]. Akpoveta *et al*.[45] found that the farther away you go from urban areas, the lesser concentration of Cd you observe. In this study area, around the Inegol, which has an organized industrial zone, the highest Cd concentration are seen.

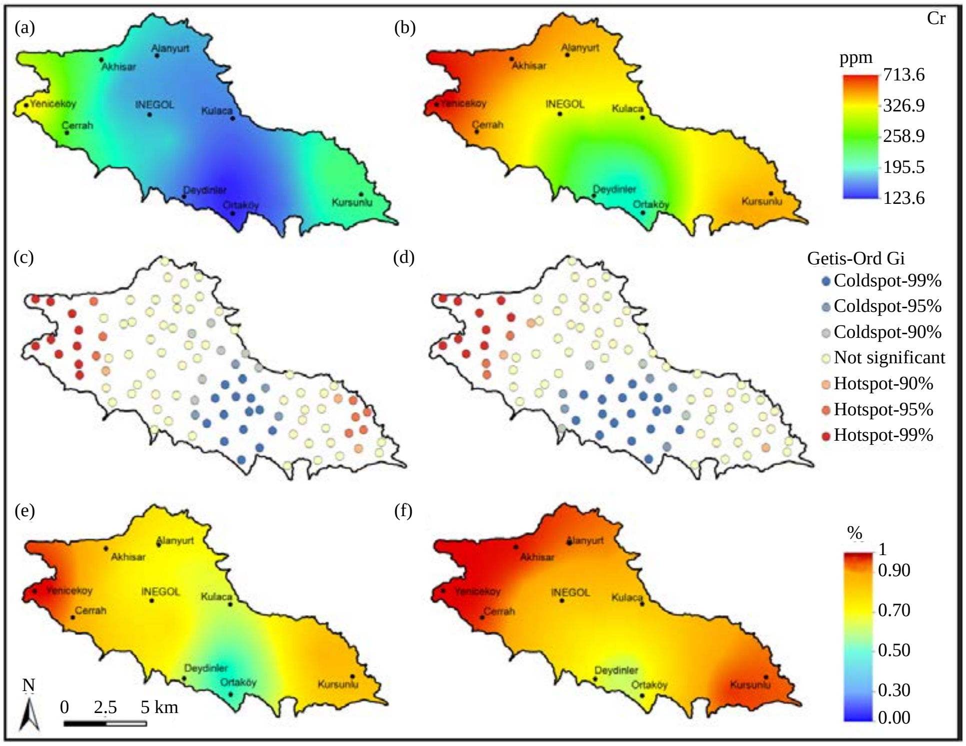

Fig. 4(a-f): Cr concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, (b) Spatial distribution in summer, (c) Local G-statistics in winter, (d) Local G-statistics in summer, (e) Probability of Cr values exceeding 100 ppm, in winter and (f) Probability Cr values exceeding 100 ppm in summer

According to the results of Moran index, Cd has a significant positive spatial autocorrelation and there is less than a 1% likelihood that this clustered pattern could be the result of random chance (Table 1). As with As, two hotspots are remarkable, as seen in Fig. 3c and d. These hotspots are the same as the hotspots of As: Around the Kursunlu in winter and around the Organized Industrial Zone in summer. Most of the industrial factories, particularly metallurgical activities are located in and around the Inegol Organized Industrial Zone. This individual hotspot in the Southeast quadrant is close to geothermal water source that is an agricultural field known for production of fruits.

The seasonal maps showing the probability of exceeding the 1 ppm threshold are reported in Fig. 3e and f. The distribution of the probability Cd shows higher values at the Southeast in winter and Northwest in summer. These areas are very high for Cd contamination. Maps show that the area with the higher probability of exceeding the 1 ppm threshold is wider during the summer, but not higher that 57%.

Spatiotemporal distribution of Cr in soils in inegol plain: The Cr concentration increases due to industrial growth, especially metal, chemical and tanning industries. Chandra *et al.*[46] reported that water resources and indirectly soil can also be contaminated instead by Cr from the tanning industry. According to Nriagu[47], the causes of Cr contamination in the soil are metallurgical industries, textile industrial wastes of dye and tannery and manures. The soil in this study area is rich with these types of industries.

In our study area, it is found that concentrations of Cr in summer are higher than in winter (Fig. 4a, b). A number of studies found the same result[48,49] with important differences between hot and wet periods: concentrations increase in hot periods. According to Yildiz *et al.*[50], soil temperature, organic matter amount and existence of other metals change the concentration levels and affect the metal accumulation in the soil. Also, Liu *et al.*[12] reported that irrigation with the river water is another important reason for higher Cr in soil. Irrigation could be the cause of high Cr concentrations during summer, on the plain. In both winter and summer periods, mean Cr concentrations are over 100 ppm, which is the limit value of Turkey Soil Pollution Control Regulation (TKKY). The level of Cr pollution in winter has been observed to be high especially in the Northwest where dense industrial activities are present being close to Inegol Organized Industrial Zone (OIZ). The level of Cr pollution in summer has been observed to be high in the Southwestern side of the plain, being close to hot and cold geothermal water sources. Oylat hot springs are not in the study area but close to the study border. However, kınık and Fresa mineral water facilities are in the field. It is possible that the increase in spatial change of Cr in Kursunlu, in the southeastern part of the study area, is related to these variables. Ozdemir and Ucar[51] reported that heavy metal concentrations in soil are higher close to geothermal facilities. According to Camgoz *et al.*[52] natural hot and cold waters carry more chemicals than drinking water. Moreover, because those mineral waters are hotter and saltier, it makes it easy for elements to go into solution, therefore increasing metal levels[53].

According to the Moran index, there are significant positive autocorrelations of Cr: Higher or lower values correlate more closely in the plain. The hotspots and coldspots, of Cr were identified. One hotspot is remarkable as seen in Fig. 4c and d in and around the Yenicekoy. This individual hotspot in the northwest quadrant was close to an industrial area.

The seasonal maps showing the probability of exceeding the threshold 100 ppm was reported in Fig. 4e and f. Probability is higher than 66% in summer and 58% in winter, showing that the plain is under risk of Cr contamination. The highest at-risk areas for contamination areas are in the Northwest and Southeast borders of the plain due to the same reason mentioned before.

Spatiotemporal distribution of Cu in soils in inegol plain: The Cu is a widely used metal in many fields: Transportation, manufacturing, currency, transportation of electricity, construction and agriculture. The Cu is released to water as a result of natural weathering of soil. However, it is difficult to determine natural concentrations of Cu in soil because of additional anthropogenic inputs[54].

In the study area, summer Cu concentrations are higher than winter concentrations. As with As and Cd, winter time high concentrations of Cu were determined around Kursunlu in the Southeast part of the plain (Fig. 5a, b). The Cu concentrations increased during the summer in the northeast part of the plain. As mentioned before, this side of the plain is not only populated heavily but also industrialized heavily.

A number of studies found that increased Cu concentration is related to the intensity of industrial activities[55-57]. The EU[58] reported that rail transport and road

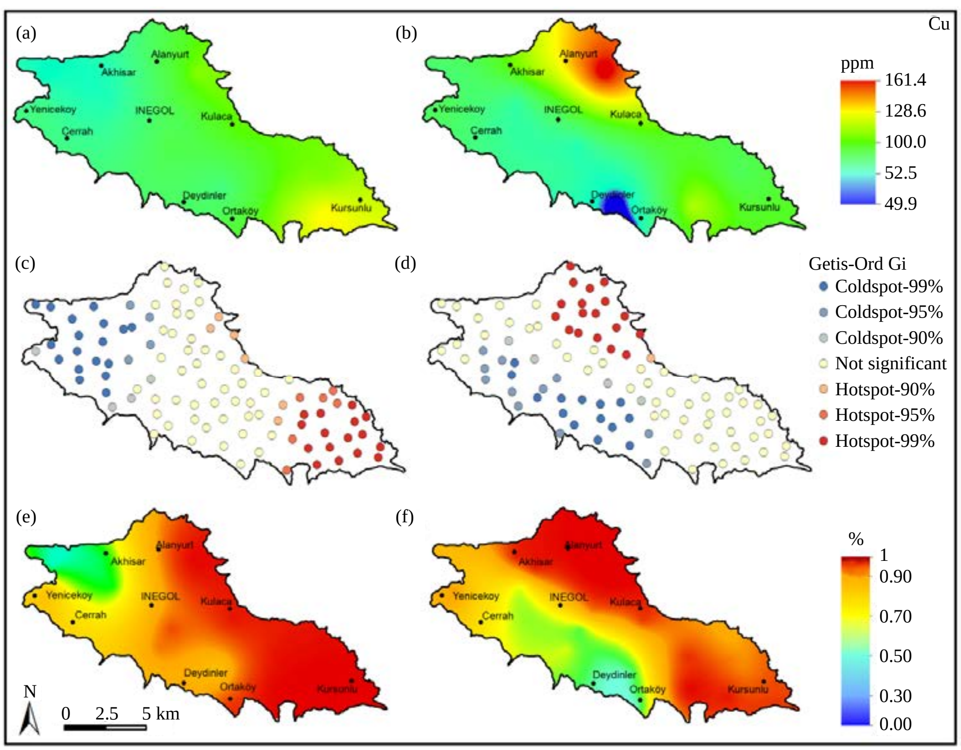

Fig. 5(a-f): Cu concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, (b) Spatial distribution in summer, (c) Local G-statistics in winter, (d) Local G-statistics in summer, (e) Probability of Cu values exceeding 50 ppm, in winter and (f) probability Cu values exceeding 50 ppm in summer

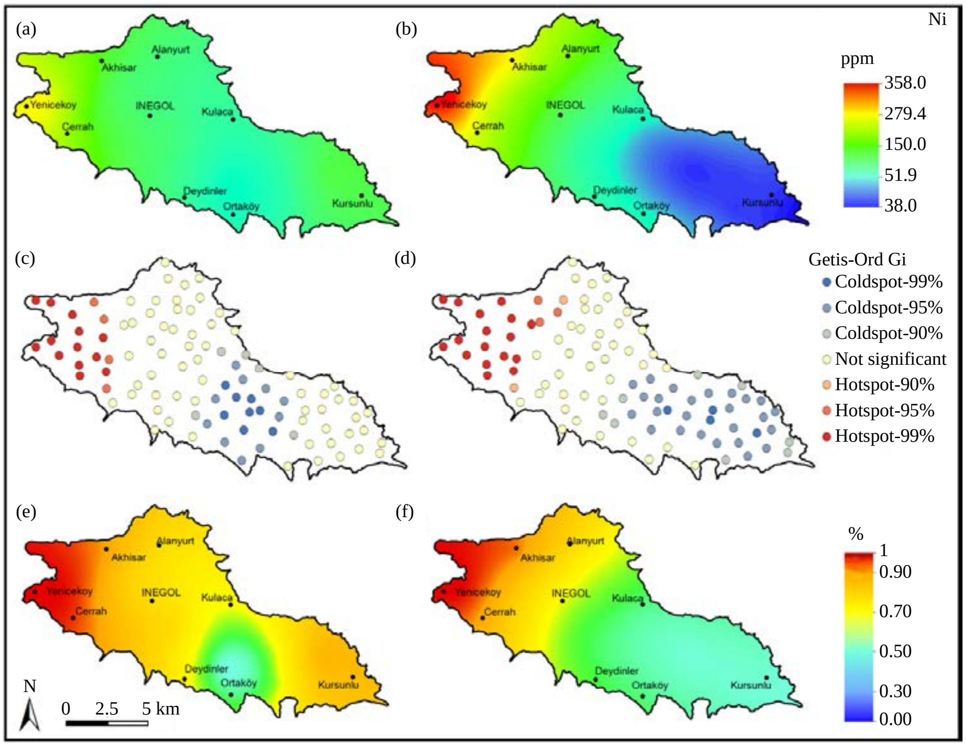

Fig. 6(a-f): Ni concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, (b) Spatial distribution in summer, (c) Local G-statistics in winter (d) Local G-statistics in summer, (e) Probability of Ni values exceeding 30 ppm, in winter and (f) Probability Ni values exceeding 30 ppm in summer

transport could cause local input in Cu concentration in soil. In addition, in many studies[59-61] it is indicated that Cu concentration increases near roads. The wood preservation products industry is very common in the study area. The Cu used in wood preservation products could cause increased Cu concentrations in soil[62,63]. Other important sources of high Cu concentrations are mining and smelting activities, wood treatment facilities and application of high doses of Cu pesticides[62,64].

As with other heavy metals, Cu shows positive spatial autocorrelation on the plain (Table 1) and there is a less than 1% likelihood that this clustered pattern could be the result of random chance. Two hotspots are detected, around the Kursunlu in winter and around the northeast of the Inegol in summer, which is a densely industrialized and settled region (Fig. 5c, d).

A map showing the probability of exceeding the threshold value of 50 ppm is reported in Fig. 5e and f. It is determined that almost all of the plain is over the 50% probability, especially in the East-southeast. All of the study area is at risk for copper contamination and thus, treatment of soils is required.

Spatiotemporal distribution of Ni in soils in inegol plain: The Ni is an essential metal which has many industrial and commercial applications. However, increases in industrialization uses could cause increased Ni concentration in the soil: Ni contamination as a result of anthropogenic activities could cause serious problems[65]. Both in summer and winter high Ni concentrations were determined to be in the same area in the Northwestern part of the plain, around the Yenicekoy, where there are intensive industrial activities and intensive settlements (Fig. 6a, b).

The ATSDR[66] noted that localized Ni contamination occurs near a smelter and plating works, as with Yenicekoy. Addo *et al.*[67] reported that vehicles along with cement facilities were responsible for heavy metal contamination. On the other hand, around the Ortakoy shows the lowest concentrations in winter. This area is the ones far from industrial activities. In the summer, concentrations are changing from one place to another place in the plain.

The Ni shows a significant high-clustered pattern and there is less than 1% likelihood that this clustered pattern could be the result of random chance (Table 1). As with Cr, there is one hot spot in the northwestern side of the plain where there are intensive industries (Fig. 6c, d).

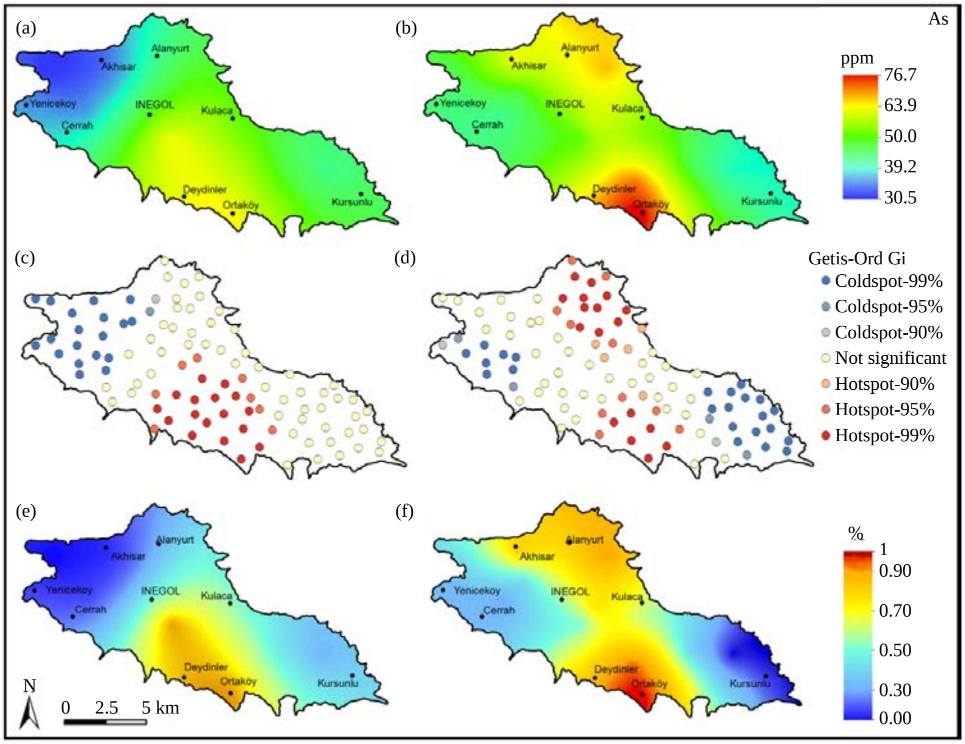

Fig. 7(a-f): Pb concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, b) Spatial distribution in summer, (c) Local G-statistics in winter, (d) Local G-statistics in summer, (e) Probability of Pb values exceeding 50 ppm, in winter and (f) Probability Pb values exceeding 50 ppm in summer

The distribution of the probability of Ni concentration shows higher values at the Northwestern and Southeastern border of the plain. It is determined that Northwestern side of the plain has more than 90% probability (Fig. 6e, f), evidence of high risk.

Spatiotemporal distribution of Pb in soils in inegol plain: The Pb is present naturally in soil and has different purposes[68]. However as suggested by the U.S. Department of Health and Human Services[69], Pb may contaminate humans in various ways, including consumption of water or food and ingestion of soil contaminated with Pd. A national geometric mean of 16 ppm has been determined as the naturally occurring Pb level in soil by The U.S. Geological Survey[70]. This amount is not very high. The Pb contamination is an ecological problem due to the high toxicity on human health in soils[71].

Concentration levels for Pb in summer is higher than winter (Fig. 7a, b). It is seen that Pb level is higher in the central part of the plain where there are intense planted agricultural activities. Sonmez *et al.*[72] reported that heavy metals like Cr, Pb, Cd, Cu, Ni and Zn get to the soil from the fertilizers and pesticides from agricultural activities. Zhang *et al.*[71] reported high levels of Pb from mining and smelting activities. Indeed, urban development, mining and smelting activities and irrigation by wastewater largely contributed to Pb accumulation in agricultural soils[71].

According to Moran I and General G (Table 1), there are high-clustered pattern in Pb concentrations. This could not be the result of random chance. In the central part of the plain there are the intensive agricultural activities, making it a significant hotspot for Pb (Fig. 7c, d).

In both winter and summer, mean Pb concentrations are over 50 ppm, which is the limit value of Turkey Soil Pollution Control Regulation (Fig. 7e, f). The distribution of the probability shows higher values at the central part of the plain (>70%). Thus, the central part of study area is under risk for Pb contamination.

Spatiotemporal distribution of Zn in soils in inegol plain: Although, Zn is naturally present and ubiquitous and is also an essential plant nutrient, when it is present in excess, Zn become toxic for living organisms[73]. Human activity contributes significantly to the increase of Zn concentrations in soil. For example, the application of inorganic fertilizers in agriculture could be the reason for local increases in Zn concentration[74]. Hotz and Brown[75] reported that it affects on average one-third of the world's population in different countries.

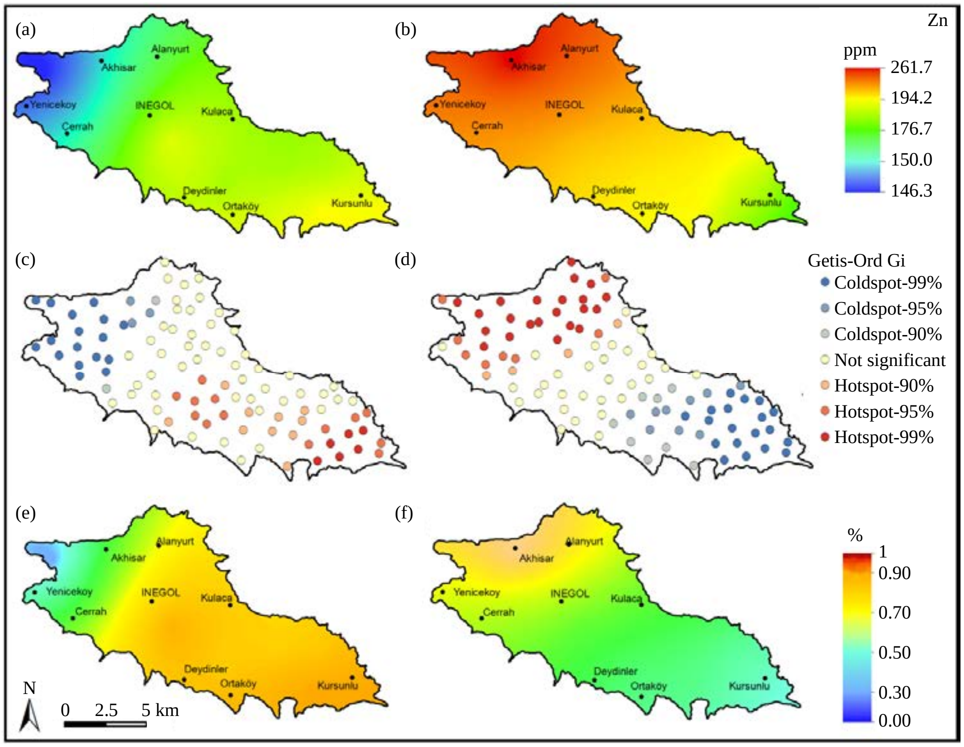

Fig. 8(a-f): Zn concentration of the surface soils in the Inegol Plain, (a) Spatial distribution in winter, (b) Spatial distribution in summer, (c) Local G-statistics in winter, (d) Local G-statistics in summer, (e) Probability of Zn values exceeding 150 ppm, in winter and (f) Probability Zn values exceeding 150 ppm in summer

In the study area, summer concentrations of Zn are higher than in winter (Fig. 8a, b). Mostly, in the winter time the Southeast side has higher levels, whereas in summer, the Northeast side has higher levels. As described earlier, these two regions are characteristic with high heavy metal concentrations. In previous studies, it has been found that in areas with high population, traffic density and industry[9,76,77] there is a potential risk of Zn pollution. Fekiacova *et al*.[73] reported that major contamination sources of Zn are industrial activities, like mining and refinery, fossil fuel combustion and chemical industries, urban activities and agriculture.

According to Table 1, there is a significant clustered pattern of Zn concentration which is significantly high in winter but not in summer. In summer, the pattern does not appear to be significantly different than random. However, local Getis Ord Gi shows one hotspot in summer and two in winter (Fig. 8c, d), potentially at-risk for Zn contamination.

The probability of exceeding the threshold 150 ppm is reported in Fig. 8e and f, between 46 and 86% in winter and between 56 and 75% in summer. This shows that there is a moderate risk for Zn. Figure 8e and f shows that the area with the higher probability of exceeding the threshold is wider during the winter period.

CONCLUSION

The present study examines the spatial distribution and levels of heavy metal concentration in the soil of Inegol Plain by using geostatistical approaches combined with GIS. Monitoring of soil quality for decision-making and environmental policies evaluation requires a proper level of accuracy and information. The finding show that Ordinary and Indicator Kriking seems to be accurate and significant in the identification of heavy metal vulnerable zones. We suggest that maps could be used to con rm most locations of suspected pollution sources. In the study area, the maps clearly demonstrated that:

- Variations of As, Cd, Cr, Cu, Ni, Pb and Zn were strongly related to the location of industrial plants, agricultural activities and hot and cold springs in the study area
- Concentration levels of most of the heavy metals, except As, were higher in summer than in winter
- All heavy metals had important hotspots and coldspots, indicating strong increasing processes for all heavy metals in soil in the plain

This study explored spatial pattern of heavy metals in the entire Inegol Plain based in 2014 using Moran's I and Getis-Ord General G analysis. The global spatial autocorrelation statistics proved to be a useful measure of overall clustering, while spatial autocorrelation was an important tool for detecting local spatial patterns for possible polluting areas. Because of the significant spatial pattern of these heavy metals in the plain, their remediation in the soil would be more difficult.

The spatial probability maps could be useful for determining the natural or anthropogenic factors on heavy metal concentration in soil in the Inegol Plain. For this reason, the results obtained from this study, relatively cheap and easy to update, can be used by local landuse planners, ecologists and managers to protect soil supplies and monitoring programs in vulnerable areas in the plain. Kursunlu in the Southwest area of the plain, around the Inegol Organized Industrial Zone and around Yenicekoy should be targeted by best management practices to reduce heavy metal loads. It is suggested that attention should also be given to industrial activities performed within the high risk areas.

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REFERENCES

1. Rodriguez-Salazar, M.T., O. Morton-Bermea, E. Hernandez-Alvarez, R. Lozano and V. Tapia-Cruz, 2011. The study of metal contamination in urban topsoils of Mexico city using GIS. Environ. Earth Sci., 62: 899-905.
2. Parveen, N., A. Ghaffar, S.A. Shirazi and M.N. Bhalli, 2012. GIS based assessment of heavy metals contamination in surface soil of urban parks: A case study of Faisalabad city-Pakistan. J. Geogr. Nat. Disast., Vol. 2. 10.4172/2167-0587.1000105
3. Hronec, O., J. Vilcek, J. Toma, P. Adamiin and E. Huttmanova, 2010. Environmental components quality problem areas in Slovakia. Mendelova Univerzita Brno, pp: 225.
4. Salah, E., A. Turki and S. Noori, 2013. Heavy metals concentration in urban soils of Fallujah city, Iraq. J. Environ. Earth Sci., 3: 100-113.
5. Al Obaidy, A.H.M.J. and A.A.M. Al Mashhadi, 2013. Heavy metal contaminations in urban soil within Baghdad city, Iraq. J. Environ. Protect., 4: 72-82.
6. Khan, S., Q. Cao, Y.M. Zheng, Y.Z. Huang and Y.G. Zhu, 2008. Health risks of heavy metals in contaminated soils and food crops irrigated with wastewater in Beijing, China. Environ. Pollut., 152: 686-692.
7. Caglarirmak, N. and A.Z. Hepcimen, 2010. Effect of heavy metal soil pollution on food chain and human health. Akademik Gida, 8: 31-35.
8. Sumer, A., S. Adiloglu, O. Cetinkaya, A. Adiloglu, A. Sungur and C. Akbulak, 2013. An investigation of some heavy metals (Cr, Ni, Pb) pollution of karamenderes basin soils in Canakkale. Tekirdag Ziraat Fakultesi Dergisi, 10: 83-89.
9. El-Gammal, M.I., R.R. Ali and R.M.A. Samra, 2014. Assessing heavy metal pollution in soils of Damietta Governorate, Egypt. Proceedings of the International Conference on Advances in Agricultural, Biological and Environmental Sciences, October 15-16, 2014, Dubai, UAE., pp: 116-124.
10. Valladares, G.S., O.A. de Camargo, J.R.P. de Carvalho and A.M.C. Silva, 2009. Assessment of heavy metals in soils of a vineyard region with the use of principal component analysis. Scientia Agricola, 66: 361-367.
11. Li, X., S.L. Lee, S.C. Wong, W. Shi and I. Thornton, 2004. The study of metal contamination in urban soils of Hong Kong using a GIS-based approach. Environ. Pollut., 129: 113-124.
12. Liu, X.M., J.J. Wu and J.M. Xu, 2006. Characterizing the risk assessment of heavy metals and sampling uncertainty analysis in paddy field by geostatistics and GIS. Environ. Pollut., 141: 257-264.
13. Lu, Y., W. Yin., F. Zhu and G. Zhang, 2010. The spatial distribution and sources of metals in urban soils of Guangzhou, China. Proceedings of the 19th World Congress of Soil Science, Soil Solutions for a Changing World, August 1-6, 2010, Brisbane, Australia, pp: 77-80.
14. Sofianska, E., K. Michailidis, V. Mladenova and A. Filippidis, 2013. Multivariate statistical and GIS-based approach to identify heavy metal sources in soils of the Drama plain, Northern Greece. Proceedings of the Bulgarian Geological Society National Conference with International Participation Geosciences, December 12-13, 2013, Sofia, Bulgaria, pp: 131 -132.
15. Su, C., L.Q. Jiang and W.J. Zhan, 2014. A review on heavy metal contamination in the soil worldwide: Situation, impact and remediation techniques. Environ. Skeptics Critics, 3: 24-38.
16. Langenkamp, H., O. Duwel and J. Utermann, 2001. Trace element and organic matter contents of European soils. Progress Report First Results of the Second Phase of the Short Term Action Joint Research Centre (JRC), Ispra.
17. Hosgoren, M.Y., 1975. Geomorphology of the inegol Basin. Istanbul University Institute of Geography, Istanbul.
18. Yucesahin, M.M., 2003. In view of urbanization process urban settlements in Inegol district. Cografi Bilimler Dergisi, 1: 75-95.
19. Yakut, Y., 2006. XIX. Yuzyil sonlarinda inegol (C-233 Numarali H.1313/M.1892 yilina ait ser'iye sicili kayitlarina gore). Master's Thesis, Suleyman Demirel Universitesi Sosyal Bilimler Enstitusu, Isparta.

20. Wei, B. and L. Yang, 2010. A review of heavy metal contaminations in urban soils, urban road dusts and agricultural soils from China. Microchem. J., 94: 99-107.
21. Parth, V., N.N. Murthy and P.R. Saxena, 2011. Assessment of heavy metal contamination in soil around hazardous waste disposal sites in Hyderabad city (India): Natural and anthropogenic implications. E3 J. Environ. Res. Manage., 2: 27-34.
22. Yao, H., J. Lu, X. Yuan, J. Wu, J. Zhao, X. Yu and Y. Zhou, 2014. Concentrations, bioavailability and spatial distribution of soil heavy metals in a long-term wastewater irrigation area in North China. Clean Soil Air Water, 42: 331-338.
23. Odeh, I.O.A. and A. Onus, 2008. Spatial analysis of soil salinity and soil structural stability in a semiarid region of New South Wales, Australia. Environ. Manage., 42: 265-278.
24. Hani, A. and M.T. Karimineja, 2010. Toxic metal distribution in soils of Kaveh Industrial city Iran. World Applied Sci. J., 8: 1333-1342.
25. Hani, A., N. Sinaei and A. Gholami, 2014. Spatial variability of heavy metals in the soils of ahwaz using geostatistical methods. Int. J. Environ. Sci. Dev., 5: 294-298.
26. Mahmoudabadi, E., F. Sarmadian, G.H. Savaghebi and Z. Alijani, 2012. Accuracy assessment of geostatistical methods for zoning of heavy metals in soils of urban-industrial areas. Int. Res. J. Applied Basic Sci., 3: 991-999.
27. TKKY., 2005. Soil pollution control regulation. Official Gazette No. 25831, TKKY., Turkey.
28. Moran, P.A.P., 1950. Notes on continuous stochastic phenomena. Biometrika, 37: 17-23.
29. Anselin, L., L. Lozano and J. Koschinsky, 2006. Rate transformations and smoothing. Spatial Analysis Laboratory, Department of Geography, University of Illinois, Urbana-Champaign, pp: 1-85.
30. Getis, A. and J.K. Ord, 1992. The analysis of spatial association by use of distance statistics. Geog. Anal., 24: 189-206.
31. Uthus, E.O., 1992. Evidence for arsenic essentiality. Environ. Geochem. Health, 14: 55-58.
32. Evans, G., J. Evans, A. Redman, N. Johnson and R.D. Foust, 2005. Unexpected beneficial effects of arsenic on corn roots grown in culture. Environ. Chem., 2: 167-170.
33. Asiam, E.K., 1996. Environmental assessment of gold beneficiation: Arsenic audit and impact on the Obuasi environs. Ghana Min. J., 2: 17-20.
34. Lokuge, K.M., W. Smith, B. Caldwell, K. Dear and A.H. Milton, 2004. The effect of arsenic mitigation interventions on disease burden in Bangladesh. Environ. Health Perspect., 112: 1172-1177.
35. Thornton, I., 1983. Applied Environmental Geochemistry. Academic Press, London, ISBN: 9780126906400, Pages: 501.
36. Brandstetter, A., E. Lombi and W. Wenzel, 2000. Arsenic-Contaminated Soils: I. Risk Assessment. In: Remediation of Hazardous Waste Contaminated Soils, Wise, D., H. Trantolo, H. Inyang and E. Cichon (Eds.). Marcel Dekker Inc., New York, pp: 715-737.
37. Smedley, P.L. and D.G. Kinniburgh, 2002. A review of the source, behaviour and distribution of arsenic in natural waters. Applied Geochem., 17: 517-568.
38. Jonnalagadda, S.B. and G. Nenzou, 1997. Studies on arsenic rich mine dumps. II. The heavy element uptake by vegetation. J. Environ. Sci. Health Part A: Environ. Sci. Eng. Toxicol., 32: 455-464.
39. Baroni, F., A. Boscagli, L.A. di Lella, G. Protano and F. Riccobono, 2004. Arsenic in soil and vegetation of contaminated areas in southern Tuscany (Italy). J. Geochem. Explorat., 81: 1-14.
40. Tiller, K.G., M.J. Mclaughlin and A.H.C. Roberts, 1999. Environmental Impacts of Heavy Metal in Agroecosystems and Amelioration Strategies in Oceania. In: Soils and Goundwater Pollution and Remediation, Huang, P.M. and I.K. Iskander (Eds.). CRC Press, Boca Raton, USA., ISBN: 9781566704526, pp: 1-41.
41. Vairavamurthy, M.A., W.S. Goldenberg, S. Ouyang and S. Khalid, 2000. The interaction of hydrophilic thiols with cadmium: Investigation with a simple model, 3-mercaptopropionic acid. Mar. Chem., 70: 181-189.
42. Yong, R.N., 2001. Geoenvironmental Engineering: Contaminated Soils, Pollutant Fate and Mitigation. CRC Press, Boca Raton, USA., ISBN: 9781420036886, Pages: 320.
43. EU., 2007. Cadmium metal. Part I environment (Vol. 72). European Union Risk Assessment Report, Of ce for Of cial Publications of the European Communities, Luxembourg.
44. Olatunji, A.S., A.F. Abimbola and O.O. Afolabi, 2009. Geochemical assessment of industrial activities on the quality of sediments and soils within the LSDPC industrial Estate, Odogunyan, Lagos, Nigeria. Global J. Environ. Res., 3: 252-257.
45. Akpoveta, O.V., S.A. Osakwe, B.E. Okoh and B.O. Otuya, 2010. Physicochemical characteristics and levels of some heavy metals in soils around metal scrap dumps in some parts of Delta State, Nigeria. J. Applied Sci. Environ. Manage., 14: 57-60.
46. Chandra, P., S. Sinha and U.N. Rai, 1997. Bioremediation of Cr From Water and Soil by Vascular Aquatic Plants. In: Phytoremediation of Soil and Water Contaminants, Kruger, E.L., T.A. Anderson and J.R. Coats (Eds.). American Chemical Society, Washington, DC., pp: 274-282.
47. Nriagu, J.O., 1989. A global assessment of natural sources of atmospheric trace metals. Nature, 338: 47-49.
48. Yahaya, M.I., S. Mohammad and B.K. Abdullahi, 2009. Seasonal variations of heavy metals concentration in abattoir dumping site soil in Nigeria. J. Applied Sci. Environ. Manage., 13: 9-13.

49. El-Serehy, H.A., H. Aboulela, F. Al-Misned, M. Kaiser, K. Al-Rasheid and H.E. El-Din, 2012. Heavy metals contamination of a mediterranean coastal ecosystem, Eastern Nile Delta, Egypt. Turk. J. Fisher. Aquat. Sci., 12: 751-760.
50. Yildiz, M., H. Terzi and B. Urusak, 2011. [Chromium toxicity and cellular responses in plants]. Erciyes Universitesi Fen Bilimleri Enstitusu Dergisi, 27: 163-176.
51. Ozdemir, M.A. and H. Ucar, 2006. Water pollution in urban ecosystem of Afyonkarahisar. Afyon Kocatepe Universitesi Sosyal Bilimler Dergisi, 8: 123-158.
52. Camgoz, B., M.M. Sac, M. Bolca, F. Ozen, O.G. Oruc and N. Demirel, 2010. [Investigation of radioactive and chemical contents of thermal waters; Izmir Seferihisar region representative]. Ekoloji, 19: 78-87, (In Turkish).
53. Kilinc, R. and I. Yokas, 1987. Toprak, bitki ve su kaynaklarinda bulunan radyasyon miktarlari ve cevresel onemi. E.U.Z.F. Dergisi.
54. Elberling, B., H. Breuning-Madsen, H. Hinge and G. Asmund, 2010. Heavy metals in 3300-year-old agricultural soils used to assess present soil contamination. Eur. J. Soil Sci., 61: 74-83.
55. Cubukcu, A. and N. Tuysuz, 2005. The environmental impacts of Karadeniz Copper Industries (KBI) smelting plant, fertilizer industry (TUGSAS) and the Industrial Park Facilities (IPF) in Tekkekoy, Samsun. Jeoloji Muhendisligi Dergisi, 29: 44-72.
56. Tahir, N.M., P.S. Chee and M. Jaafar, 2007. Determination of heavy metals content in soils and indoor dusts from nurseries in Dungun, Terengganu. Malay. J. Anal. Sci., 11: 280-286.
57. Ozkul, C., 2008. Effect of industrialization on soil heavy metal concentrations in Izmit (Kocaeli) region. Uygulamalı Yerbilimleri, 2: 1-9.
58. ECA., 2008. Voluntary risk assessment reports-copper and copper compounds. European Chemicals Agency (ECA). http://echa.europa.eu/web/guest/copper-voluntary-risk-assessment-reports
59. Garcia, R., I. Maiz and E. Millan, 1996. Heavy metal contamination analysis of roadsoils and grasses from Gipuzkoa (Spain). Environ. Technol., 17: 763-770.
60. Dierkes, C. and W.F. Geiger, 1999. Pollution retention capabilities of roadside soils. Water Sci. Technol., 39: 201-208.
61. Akkus, S., T. Bal, N.M. Karaaslan, E. Yaman, E. Kilinc and M. Yaman, 2013. Fractionation of Ni, Cr and Cu from soil by sequential extraction procedure and determination by inductively coupled plasma optical emission spectrometry. CLEAN-Soil Air Water, 41: 1229-1234.
62. Hasan, A.R., L. Hu, H.M. Solo-Gabriele, L. Fieber, Y. Cai and T.G. Townsend, 2010. Field-scale leaching of arsenic, chromium and copper from weathered treated wood. Environ. Pollut., 158: 1479-1486.
63. Mench, M. and C. Bes, 2009. Assessment of ecotoxicity of topsoils from a wood treatment site. Pedosphere, 19: 143-155.
64. Komarek, M., E. Cadkova, V. Chrastny, F. Bordas and J.C. Bollinger, 2010. Contamination of vineyard soils with fungicides: A review of environmental and toxicological aspects. Environ. Int., 36: 138-151.
65. Scott-Fordsmand, J.J., 1997. Toxicity of Nickel to Soil Organisms in Denmark. In: Reviews of Environmental Contamination and Toxicology, Ware, G.W., H.N. Nigg and A. Bevenue (Eds.). Vol. 148, Springer, New York, ISBN: 9781461222644, pp: 1-34.
66. ATSDR., 2005. Toxicological profile for nickel. Agency for Toxic Substances and Disease Registry (ATSDR), U.S. Department of Health and Human Services Public Health Service, Atlanta, Georgia, pp: 397.
67. Addo, M.A., E.O. Darko, C. Gordon, B.J.B. Nyarko and J.K. Gbadago *et al*., 2012. Evaluation of heavy metals contamination of soil and vegetation in the vicinity of a cement factory in the Volta region, Ghana. Int. J. Sci. Technol., 2: 40-50.
68. Nriagu, J.O., 1978. The Biogeochemistry of Lead in the Environment. Elsevier/North-Holland Biomedical Press, USA., ISBN: 9780444800503, pp: 18-88.
69. CDC., 1991. Preventing lead poisoning in young children: A statement by the Centers for Disease Control. U.S. Department of Health and Human Services, Public Health Service, Centers for Disease Control, Atlanta, GA., USA., October 19991.
70. Shacklette, H.T. and J.G. Boerngen, 1984. Element concentrations in soils and other surficial materials of the conterminous United States. U.S. Geological Survey Professional Paper 1270, U.S. Government Printing Office, Washington, DC.
71. Zhang, X., D. Chen, T. Zhong, X. Zhang, M. Cheng and X. Li, 2015. Evaluation of lead in arable soils, China. CLEAN-Soil Air Water, 43: 1232-1240.
72. Sonmez, I., M. Kaplan and S. Sonmez, 2008. Effect of chemical fertilizers on environmental pollution and its prevention methods. Derim Dergisi, 25: 24-34.
73. Fekiacova, Z., S. Cornu and S. Pichat, 2015. Tracing contamination sources in soils with Cu and Zn isotopic ratios. Sci. Total Environ., 517: 96-105.
74. Cakmak, I., A. Yilmaz, M. Kalayci, H. Ekiz, B. Torun, B. Erenoglu and H.J. Braun, 1996. Zinc deficiency as a critical problem in wheat production in Central Anatolia. Plant Soil, 180: 165-172.
75. Hotz, C. and K.H. Brown, 2004. Assessment of the risk of zinc deficiency in populations and options for its control. Food Nutr. Bull., 25: 191-204.
76. Vural, A. and E. Sahin, 2012. Preliminary findings of trace elements accumulation in soil and acacia for vicinity of state main road passing through Gumushane city center. Gumushane Univ. J. Sci. Technol. Inst., 2: 21-35.
77. Lin, Y.P., B.Y. Cheng, H.J. Chu, T.K. Chang and H.L. Yu, 2011. Assessing how heavy metal pollution and human activity are related by using logistic regression and kriging methods. Geoderma, 163: 275-282.