Mustafa Kemal Üniversitesi Tarım Bilimleri Dergisi, 30 (3), 866-885
Mustafa Kemal University Journal of Agricultural Sciences, 30 (3), 866-885
https://dergipark.org.tr/tr/pub/mkutbd
e-ISSN: 2667-7733

ARAŞTIRMA MAKALESİ **RESEARCH ARTICLE**

Micrometal sources and potential pollution status in intensive and monoculture agriculture

Yoğun ve monokültür tarımda mikrometal kaynaklar ve potansiyel kirlilik durumu

Tuğba ŞİMŞEK[1], Nilgün KALKANCI[1], Cafer Hakan YILMAZ[2], Gökhan BÜYÜK[3]

[1]*Ministry of Agriculture and Forestry Pistachio Research Institute Directorate, Gaziantep, Türkiye.*
[2]*East Mediterranean Transitional Zone Agricultural Research of Institute, Kahramanmaraş, Türkiye.*
[3]*Adıyaman University, Agriculture Faculty, Adıyaman, Türkiye.*

ARTICLE INFO

Article history:
Recieved / Geliş: 19.06.2025
Accepted / Kabul: 16.09.2025

Keywords:
Osmaniye
Agricultural soils
Health risks
Pollution sources
Mikrometaloit

Anahtar Kelimeler:
Osmaniye
Agricultural soils
Sağlık riskleri
Kirlilik kaynakları
Mikrometaloit

Corresponding author/Sorumlu yazar:
Gökhan BÜYÜK
gbuyuk@adiyaman.edu.tr

ABSTRACT

In this study, the concentrations of micrometalloids (MMs) such as Fe, Mn, Cu, Zn, Ni, Cd, Co, Pb and Cr were determined in 165 soil samples collected from 0-20 cm depth by determining their locations from agricultural lands in Osmaniye province.The average concentrations of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn were detected 3.30, 0.16, 103, 18.52, 0.16, 17093, 126.3, 232.61, 2.71 and 52.19 mg kg^{-1}, respectively. The results of this study showed that Ni, Co and Cr are due to geological erosion of the parent material, Fe, Mn and Cd are due to both lithogenic and different anthropogenic sources and Pb is due to atmospheric factors. The average concentrations of Ni, Cd and Cr among MMs in the investigated agricultural soils exceeded the UCC concentrations used for the calculation of environmental and ecological risk indices. Although non-carcinogenic and carcinogenic risks do not pose a threat to the inhabitants of the region at the moment, the accumulation of MMs and the level of contamination of soils indicate that they may pose a threat in the near future. Furthermore, HQ and HI values for ingestion and dermal were found to be higher in children than in adults, indicating that children are more affected by MMs in agricultural soils. the high concentration of a MM in soils where pollution is investigated may directly depend on the parent material of the soils studied, as in this study. Therefore, in such studies, sampling should be done from the main material.

ÖZET

Bu çalışmada, Osmaniye ilindeki tarım arazilerinden konumları belirlenerek 0-20 cm derinlikten toplanan 165 toprak örneğinde Fe, Mn, Cu, Zn, Ni, Cd, Co, Pb ve Cr gibi mikrometaloidlerin (MMs) konsantrasyonları belirlenmiştir. Ortalama Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb ve Zn konsantrasyonları sırasıyla 3.30, 0.16, 103, 18.52, 0.16, 17093, 126.3, 232.61, 2.71 ve 52.19 mg kg^{-1} olarak tespit edilmiştir. Bu çalışmanın sonuçları Ni, Co ve Cr'nin ana materyalin jeolojik erozyonundan, Fe, Mn ve Cd'un hem litojenik hem de farklı antropojenik kaynaklardan, Pb'nin ise atmosferik faktörlerden kaynaklandığını göstermiştir. İncelenen tarım topraklarındaki MM'ler arasında Ni, Cd ve Cr'nin ortalama konsantrasyonları, çevresel ve ekolojik risk endekslerinin hesaplanmasında kullanılan UCC konsantrasyonlarını aşmıştır. Kanserojen olmayan ve kanserojen riskler şu anda bölge sakinleri için bir tehdit oluşturmasa da, MM'lerin birikimi ve toprakların kirlenme seviyesi yakın gelecekte bir tehdit oluşturabileceklerini göstermektedir. Ayrıca, yutma ve dermal için HQ ve HI değerleri çocuklarda yetişkinlere göre daha yüksek bulunmuştur, bu da çocukların tarım topraklarındaki MM'lerden daha fazla etkilendiğini göstermektedir. Kirliliğin araştırıldığı topraklarda bir MM'nin yüksek konsantrasyonu, bu çalışmada olduğu gibi, doğrudan çalışılan toprakların ana materyaline bağlı olabilir. Bu nedenle bu tür çalışmalarda ana materyalden örnekleme yapılmalıdır.

Cite/Atıf Şimşek, T., Kalkancı, N., Yılmaz, C.H. & Büyük, G. (2025). Micrometal sources and potential pollution status in ıntensive and monoculture agriculture. *Mustafa Kemal Üniversitesi Tarım Bilimleri Dergisi, 30* (3), 866-885. https://doi.org/10.37908/mkutbd.1722760

INTRODUCTION

Agriculture is a fundamental activity for human life and plays a critical role in food production and nutrition. However, the use of fertilizers, pesticides, and industrial waste in modern agricultural practices can cause heavy metal accumulation in soil and water resources. Heavy metals such as lead, cadmium, arsenic, mercury and nickel have the potential to accumulate in soil and plants entering the food chain. This poses serious risks to human health and threatens the balance of the ecosystem.

Ingestion of heavy metals through food can lead to chronic health problems such as cancer, nervous system damage, kidney and liver dysfunction. The effects on human health, especially at low doses under prolonged exposure, can occur over many years. Therefore, heavy metal pollution in agricultural production is of great importance for both public health and sustainable agricultural policies. Soil health is the capacity of soil to continue to function as a vital living ecosystem that nourishes plants, animals and people, and links agriculture and soil science to policy, stakeholder needs and sustainable supply chain management (Lehmann et al., 2020). "Heavy metals" refers to 'heavy metals' (eg. Cd, Co, Hg, Pb) and metalloids (eg. Se, Sb, As) with atomic number greater than 20 (Sanaei et al., 2020) and density greater than 5 g cm^{-3} (Alloway, 2013). Health risk assessments that consider the uncertainty of exposure and toxic factors reveal that soil heavy metal contamination in China is severe, with Pb, Cd, Zn, Ni and As being the dominant contaminants (Peng et al., 2022).

The objectives of this research were to identify potential sources and numbers of MMs using multivariate statistical methods, to assess the ecological-environmental risks of MMs using individual and multiple indices, and to assess both non-carcinogenic and carcinogenic human health risks for local residents exposed to MMs in soil.

MATERIALS and METHODS

Study area

Located in the eastern part of the Mediterranean Region and the Çukurova Plain, Osmaniye lies between 35° 52'-36° 42' East Meridians (longitudes) and 36° 57'-37° 45' North Parallels (latitudes). It is bordered by Gaziantep to the east, Hatay to the south, Adana to the west, and Kahramanmaraş to the north. Its surface area is 3,279.9 km^2, 121 meters above sea level and 20 kilometers from the Mediterranean Sea.

Although it varies in mountainous and lowland areas, it shows typical Mediterranean climate characteristics TSMS, 2024). Although the climate of Osmaniye province varies in mountainous and lowland areas, it has the characteristics of Mediterranean climate. In general, summers are hot and dry, winters are mild and rainy. Precipitation is higher in winter and fall than in other months. The lowest temperatures are in December and January and the hottest in July and August. Precipitation is not evenly distributed throughout the year. According to 1986-2017 data, the average annual precipitation is 828.0 mm.

Soil sampling and determination of MMs concentrations

The sampling was carried out by creating 2.5 x 2.5 km grids by taking into account the areas where irrigated agriculture, dry agriculture, peanut, wheat, barley, cotton, vineyard, olive, citrus, etc. are cultivated within the borders of Osmaniye province. A total of 165 soil samples were taken from the points falling on the agricultural areas and from 0-20 cm depth. Coordinates were determined with GPS (Figure 1).

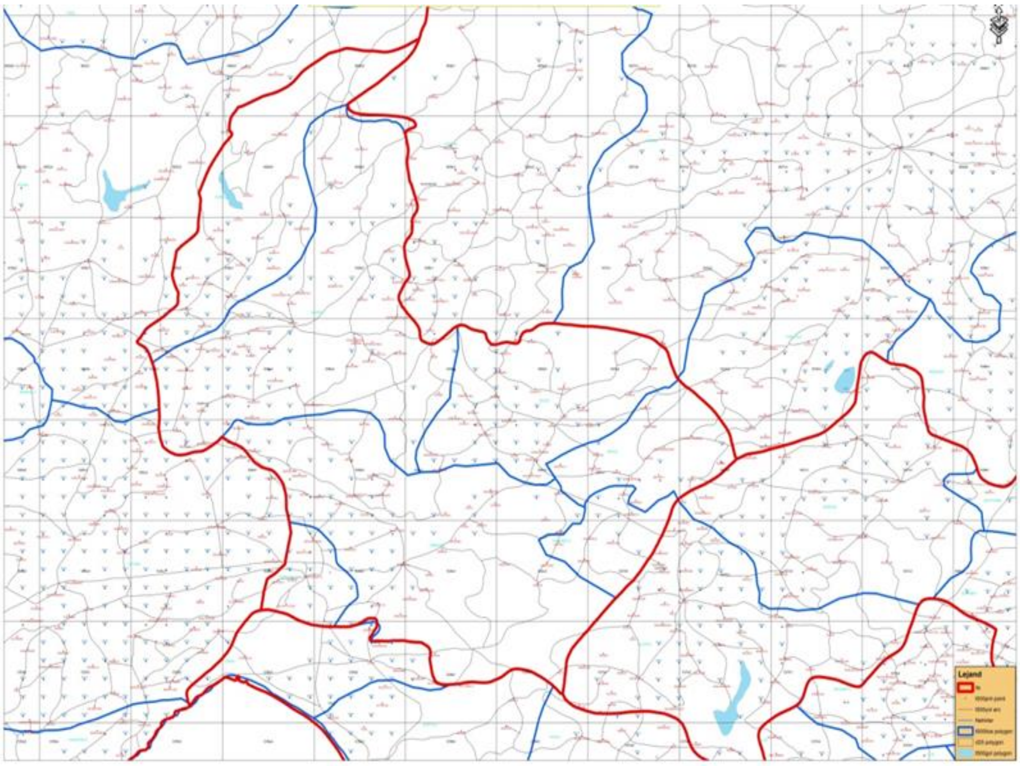

Figure 1. Points of soil samples taken from the study area
Şekil 1. Çalışma alanından alınan toprak örneklerinin noktaları

The soil samples prepared for analysis were subjected to analysis with 'king water' in Cem brand Mars 6 model microwave oven according to the wet digestion method as described by Jackson (1958). The extract obtained was read in Agilent brand 5100 model ICP-OES and total micrometabolites (Fe, Mn, Cu, Zn, Ni, Cd, Co, Pb and Cr) concentrations (mg kg^{-1}) were determined. Each soil sample was analyzed twice (in duplicate).

Environmental and ecological risk indices of MMs

The environmental risks of MMs in agricultural soils were determined by calculating five different pollution indices such as geoaccumulation index (Igeo), enrichment factor (EF), contamination factor (Cf), pollution load index (PLI) and Nemerow pollution index (NPI). In addition, ecological risk index (RI), Nemerow risk index (NRI) and ecological risk factor (ERF) were calculated to determine the ecological risk levels of MMs in soils. Upper continental crust (UCC) contents were used in the calculation of individual indices (Igeo, EF, Cf and Er) of MMs (Yılmaz, 2023). The impact and pollution level of MMs on soil ecosystems are estimated by individual indices such as Igeo, EF, Cf and Er (Hakanson, 1980; Müller, 1981). The cumulative risks of synergistic effects of MMs for the soil ecosystem and the overall pollution level are estimated by NPI, PLI, NRI and RI (Men et al., 2020). The environmental and ecological risk assessment indices used in the study are given in Table 1.

Estimation of MM sources in agricultural soils by multivariate analysis

To determine whether MMs in agricultural soils come from common sources or different sources, the relationships between them were examined. Pearson correlation test was used for this purpose. PCA-FA was used to determine the potential pollution sources and numbers of MMs. Before the tests, z-scale transformation was applied to the data obtained from the research area and the values were normalized. KaiserMeyerOlkin (KMO) score and Bartlett sphericity test were used to check the suitability of the data for PCA-FA.

Health risks of MMs in agricultural soils

In the study, the health hazards from exposure to MMs in agricultural soils through ingestion, inhalation and dermal contact were analyzed separately for children and adults living in the region with the values determined by USEPA (2023a). This is because the most common routes of human exposure to MMs from soil are through ingestion, inhalation and dermal contact (Deliboran et al., 2024). MM hazard quotients (HQ) and carcinogenic risks (CR) were estimated for non-carcinogenic and carcinogenic health risks, respectively, using the following formulas reported by USEPA (2023b). The reference values used in the formulas are given in Tables 1 and 2.

In the study, CHQ: cumulative hazard coefficient, HI: hazard index, ΣHI: total HI, CCR: cumulative carcinogenic risk, ΣCR: total carcinogenic risk and CΣCR: cumulative ΣCR were determined as can be understood from the above formulas.

The non-carcinogenic risks:

$$\text{HQingestion} = \frac{\text{Cs x IRS x RBA x EF x ED}}{\text{BW x AT x RfDo x } 10^6}$$

$$\text{HQinhalation} = \frac{\text{Cs x EF x ED}}{\text{AT x RfC x PEF}}$$

$$\text{HQdermal} = \frac{\text{Cs x SAx AF x ABSd x EF x ED}}{\text{BW x AT x RfDo x GIABS x } 10^6}$$

The carcinogenic risks:

$$\text{HQinhalation} = \frac{\text{Cs x EF x ED}}{\text{AT x RfC x PEF}}$$

$$\text{CRingestion} = \frac{\text{Cs x IFS x RBA x CSFo}}{\text{AT x } 10^6}$$

$$\text{IFS} = \frac{\text{EF x EDc x IRSa}}{\text{BWc}} + \frac{\text{EF x EDa x IRSa}}{\text{BWa}}$$

$$\text{CRdermal} = \frac{\text{Cs x DFS x ABSd x CSFo}}{\text{AT x GIABS x } 10^6}$$

$$\text{DFS} = \frac{\text{EF x EDc x SAc x AFc}}{\text{BWc}} + \frac{\text{EF x EDa x SAa x AFa}}{\text{BWa}}$$

$$\text{CRinhalation} = \frac{\text{Cs x EF x ED x IUR x 1000}}{\text{AT x PEF}}$$

$$\text{CHQ} = \sum HQs \text{ of MMs}$$

$$\text{HI} = \text{HQing} + \text{HQinh} + \text{HQder}$$

$$\Sigma\text{HI} = \sum HIs \text{ of MMs}$$

$$\text{CCR} = \sum CRs \text{ of carcinogenicMMs}$$

$$\Sigma\text{CR} = \text{CRing} + \text{CRinh} + \text{CRder}$$

$$\text{C}\Sigma\text{CR} = \sum \Sigma\text{CRs of carcinogenicMMs}$$

RESULTS and DISCUSSIONS

MM concentrations in agricultural soils

Some descriptive statistics of micrometalloids (MM) contents of the soils where different crops were grown in the study area are shown in Table 2. The mean concentrations of Fe, Mn, Cu, Zn, Ni, Cd, Co, Pb and Cr were 17093, 126.30, 18.52, 52.19, 232.61, 3.30, 0.16, 2.71 and 103.00 mg kg-1, respectively. In this study, Zn (28.32%) had a lower CV value, indicating that these MMs are affected by lithogenic sources, but Co (126.64%), Cr (120.44%), Pb (115.30%), Ni (114.65%), Mn (112.70%), Cd (48.21%), Fe (45.71%) and Cu (63.60%) had higher coefficients of variation (CV) (Table 2), indicating that the distribution of MMs in soil is not homogeneous and anthropogenic sources may be effective in the study area. (Deliboran et al., 2024; Fei et al., 2019; Zhang et al., 2021). Mn, Cu and Co do not show a normal distribution as their kurtosis and skewness values are >2. This indicates that these MMs may be affected by human activities (Xie et al., 2023). However, according to the kurtosis and skewness values of other MMs, there may be a normal distribution.

Table 1. Environmental and ecological risk evaluation indexes used in the research
Çizelge 1. Araştırmada kullanılan çevresel ve ekolojik risk değerlendirme indeksleri

Indexes	Riskor contamination degree	Equations of indexes	Explanations
Pollution indices			
Geoaccumulation index (Igeo)	Igeo ≤ 0 → Unpolluted 0 < Igeo < 1 → Unpolluted to moderately polluted 1 < Igeo < 2 → Moderately polluted 2 < Igeo < 3 → Moderately to heavily polluted 3 < Igeo < 4 → Heavily polluted 4 < Igeo < 5 → Heavily to extremely polluted Igeo ≥ 5 → Extremely polluted	$\mathrm{Igeo}=\log_2[\frac{\mathrm{Ci}}{\mathrm{Bi\ x\ 1.5}}]$	Bi is background value of element (i); Ci is the content of element (i) (Muller, 1969)
Enrichment factor (EF)	EF < 2 → Minimal enrichment 2 ≤ EF < 5 → Moderate enrichment 5 ≤ EF < 20 → Significant enrichment 20 ≤ EF < 40 → Very high enrichment EF ≥ 40 → Extremely high enrichment	$\mathrm{EF}=\frac{[\frac{\mathrm{Ci}}{\mathrm{Cref}}]\mathrm{sample}}{[\frac{\mathrm{Ci}}{\mathrm{Cref}}]\mathrm{background}}$	Ci represents the MM content in soil, and Cref is the reference MM (Fe) for geochemical normalization (Mazurek et al., 2017)
Contamination factor (Cf)	CF < 1 → Low contamination 1 < CF < 3 → Moderate contamination 3 < CF < 6 → Considerable contamination CF ≥ 6 → Very high contamination	$\mathrm{Cf}=\frac{\mathrm{Ci}}{\mathrm{Cb}}$	Ci represents the amount of the MM, and Cb is the previous industrial reference level of the MM (Hakanson, 1980)
Pollution load index (PLI)	PLI = 0 → No contamination 0 < PLI < 1 → Baseline contamination PLI > 1 → High contamination	PLI= $\sqrt[n]{\mathrm{Cf1\ x\ Cf2\ x\ Cf3\ x.....x\ Cfn}}$	Chakravarty and Patgiri (2009).

Table 1 (continued). Environmental and ecological risk evaluation indexes used in the research
Çizelge 1 (devamı). Araştırmada kullanılan çevresel ve ekolojik risk değerlendirme indeksleri

Nemerow pollution index (NPI)	NPI ≤ 0.7 → Unpolluted 0.7 < NPI ≤ 1 → Warning line of pollution 1 < NPI ≤ 2 → Low polluted 2 < NPI ≤ 3 → Moderately polluted NPI > 3 → Strongly polluted	$\mathrm{NPI} = \sqrt{\frac{[\mathrm{Cfaverage}]^2 + [\mathrm{Cfmaximum}]^2}{2}}$	Cfaverage and Cfmax are the average and maximum values of values of all investigated MMs (Men et al., 2021)
Ecological risk indices			
Ecological risk factor (Er)	Er < 40 → Low potential ecological risk 40 ≤ Er < 80 → Moderate potential ecological risk 80 ≤ Er < 160 → Considerable potential ecological risk 160 ≤ Er < 320 → High potential ecological risk Er ≥ 320 → Very high potential ecological risk	$E_r^i = T_r^i \times C_f^i$	T_r^i is the toxical reaction factor of MM and with respect to Hakanson it is 5, 1, 5, 30, 2 and 5 for Cu, Zn, Ni, Cd, Cr and Pb,respectively. C_f^i is the contagion factor of MM (Hakanson, 1980)
Nemerow risk index (NRI)	NRI ≤ 40 → Low risk 40 < NRI ≤ 80 → Moderate risk 80 < NRI ≤ 160 → Considerable risk 160 < NRI ≤ 320 → High risk NRI > 320 → Very high risk	$\mathrm{NPI} = \sqrt{\frac{[\mathrm{Eraverage}]^2 + [\mathrm{Ermaximum}]^2}{2}}$	Eraverage and Ermaximum are the average and maximum values of E_r^i(Men et al., (2020)
Ecological risk index (RI)	RI < 150 → Low ecological risk 150 ≤ RI < 300 → Moderate ecological risk 300 ≤ RI < 600 → Considerable ecological risk RI ≥ 600 → Very high ecological risk	$\sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} (T_r^i \times \frac{C_i}{C_b}) \longrightarrow (\frac{C_i}{C_b} = Cf)$	E_r^i is the single ecological risk that MMs and with respect to Hakanson it is 5, 1, 5, 30, 2 and 5 for Cu, Zn, Ni, Cd, Cr and Pb, respectively.Cf is the contagion factor of MM (Hakanson, 1980)

Determined in the Turkish Soil Pollution Control Regulation (TSPCR); Upper Continental Crust (UCC) proposed by Rudnick and Gao (Rudnick et al., 2003); the Canadian Soil Quality Guidelines (CSQG) (CCME, 2007); the average MM contents of European (ES) and World (WS) soils and the maximum allowable concentrations (MAC) of MMs in soils as recommended by Kabata-Pendias (Kabata-Pendias, 2011) were compared with the average concentrations of MEs determined in this study (Table 2). The results showed that the mean concentrations of Ni (232.61 mg kg^{-1}), Cd

(3.30) and Cr (103.00) of the study area soils exceeded the Ni, Cd and Cr concentration values of TSPCR (75, 3 and 100 mg kg^{-1}, respectively), CSQG (50, 1.4 and 64 mg kg^{-1}, respectively), UCC (47, 0.09 and 92 mg kg-1, respectively), ES (37, 0.28 and 94.8 mg kg^{-1}, respectively) and WS (29, 0.41 and 70 mg kg^{-1}, respectively). It also exceeded the amount of Ni (60 mg kg^{-1}) of MAC. The averages of the other MMs analyzed were lower than the MM concentrations of the comparison soils, except for Cu (17.30 mg kg^{-1}) content of ES (Table 2). In this study, the average MM concentrations in agricultural soils where different crops were grown were also compared with the MM values of the soils of Mouriki-Thiva region of neighboring Greece (Antibachi et al., 2012).

Daye City region of China (Du et al., 2015) and Harran Plain (Varol et al., 2020) where intensive agriculture is practiced in our country (Table 2). Accordingly, MM concentrations of Osmaniye agricultural soils were found to be above the Cr (24 mg kg^{-1}) contents of Mouriki-Thiva soils, Ni (25.80 mg kg^{-1}), Cd (1.41 mg kg^{-1}) and Cr (60.7 mg kg^{-1}) concentrations of Daye City soils, and Ni (89 mg kg^{-1}) and Cr (85 mg kg^{-1}) contents of Harran Plain soils. These data show that the spatial heterogeneity of agricultural soils in different parts of the world and the different activities of humans have an impact on the concentrations of MMs in soil (Yılmaz, 2023).

Environmental and ecological risk indices determining the pollution status of MMs in agricultural soils

Igeo, EF and Cf indices were calculated for the level of pollution and enrichment of a single element or a specific MM in the soil, while PLI and NPI were calculated to see the overall pollution levels of MMs as a whole. The Igeo, EF, Cf, PLI and NLI indices determining the environmental pollution status of MMs in soils are presented in Figure 2 and Table 4. Table 1 was used for the risk classification of the indices.

In this study, the mean Igeo values of Ni and Cd were found to be 0.84 and 4.44, respectively. It is understood that soils are 'unpolluted to moderately polluted' by Ni and 'heavily to extremely polluted' by Cd. However, the mean Igeo values of other MMs are negative, indicating that soils are 'unpolluted' by these MMs. Within the scope of this study, according to Müller's (1969) Igeo classification:

Cd soil is heavily to extremely contaminated, and the sources of this metal are likely related to strong anthropogenic (human-induced) activities (e.g., agricultural pesticides, industrial waste). Low-level contamination may be present for Ni. Since the average Igeo values for other metals are negative, the environmental levels of these elements are considered to be within natural limits and are not thought to cause contamination.

The average EF values of MMs in the investigated agricultural soils were found to be Co (0.02)< Mn (0.41)< Pb (0.42)< Fe (1.00)< Cu (1.72)< Zn (2.06)< Cr (2.10)< Ni (9.38)< Cd (83.93) (Figure 2 and Table 4). According to these average EF results, Co, Mn, Pb, Fe and Cu showed 'minimal enrichment' in agricultural soils, while Zn and Cr showed 'moderate enrichment', Ni showed 'significant enrichment' and Cd showed 'extremely high enrichment'. When Table 4 is analyzed for the Cf values of the study; Cd (36.705) and Ni (4.949) averages are >4 and it is understood that agricultural soils are significantly polluted by these MMs. From the results, it is seen that agricultural soils have 'very high contamination' by Cd and 'considerable contamination' by Ni. Since the CfCr average of the research area is 1.120, it is 'moderate contamination' in terms of Cr, and since the Cf average values of the other MMs are <1, they have received 'low contamination' levels. Similarly, in the literature, Cd is reported to be anthropogenic in origin (particularly from phosphate fertilizer use, industrial activities, and wastewater) and to have high Cf values in agricultural areas (Alloway, 2013; Kabata-Pendias, 2011). For example, Zhang et al. (2018) stated in a study conducted in China that the Cf value of Cd in agricultural soils is often above 6 and that this situation is critical in terms of environmental risk.

The “significant level of contamination” detected for Ni may also be related to the use of metal-containing pesticides and fertilizers. On the other hand, the average CfCr value of 1.120 for Cr indicates that this metal causes “moderate contamination” in the study area. This suggests that natural geological sources may be influential. Indeed, Cr is typically rock-derived, and Cf values in agricultural soils are generally reported in the range of 1–3 (Li et al., 2014). In contrast, the Cf values of other heavy metals (e.g., Zn, Cu, Pb) being below 1 indicate that these metals are at a “low contamination” level in the study area and are largely present at natural levels. These findings are consistent with studies conducted in similar agricultural areas (Rahman et al., 2012; Wei & Yang, 2010).

The mean value of PLI of MMs in agricultural soils was 0.458 and was categorized as 'baseline contamination', while the mean value of NPI of the studied soils was 26.20 and was categorized as 'strongly polluted'. Figure 2 and Table 4 show that the results of individual pollution indices such as Igeo, EF and Cf are consistent with each other. These results show that most of the agricultural soils are highly polluted by Cd, highly polluted by Ni, and moderately polluted by Zn and Cr. In contrast, Fe, Mn, Cu, Co and Pb showed low contamination. The research area was formed on different parent materials (basalt, basaltic tuff, marl, alluvial, colluvial, limestone, ophiolite, serpentine) in Osmaniye province (Lavkor, 2006) and is intensively farmed.

The high levels of MMs such as Cr and Ni may be due to the presence of serpentine in the bedrock and the mineralization of chromium. Cr and Ni can be released into agricultural ecosystems and natural water systems, especially during mineral weathering (Oze et al., 2008). With rapid industrialization and the widespread application of agricultural fertilizers, pesticides, etc. in agricultural activities in recent years, agricultural lands are faced with micrometaloids pollution (Zu et al., 2008). Industrial activities such as mining can also increase soil Cd levels. In the study area, intensive mining and intensive chemical fertilization (phosphate) to increase agricultural production may cause Cd accumulation (Şimşek et al., 2021). In addition, even one of the PLI and NPI multiple pollution indices was high, indicating that the combined pollution effects of MMs may cause high pollution in most of the agricultural soils.

Table 2. Some descriptive statistics of MM results of investigated soils and data from other studies around the world (mg kg^{-1})

Çizelge 2. İncelenen toprakların MM sonuçlarının bazı tanımlayıcı istatistikleri ve dünyadaki diğer çalışmalardan elde edilen veriler (mg kg^{-1})

Osmaniye	Fe	Mn	Cu	Zn	Ni	Cd	Co	Pb	Cr	Reference
Average	17093	126.30	18.52	52.19	232.61	3.30	0.16	2.71	103.00	
Minimum	3096	7.43	5.80	19.40	15.92	0.68	0.00	0.02	6.46	
Maximum	45596	924.48	72.74	105.18	988.63	8.63	1.09	13.02	529.90	
Median	7812.7	142.34	8.07	14.78	266.70	1.59	0.20	3.13	124.05	
Standart deviation	610.07	11.11	0.63	1.15	20.83	0.12	0.02	0.24	9.69	In this research
Standart error	15385	82.40	17.35	52.15	95.89	2.93	0.08	1.59	40.22	
Coefficient of variation (CV, %)	45.71	112.70	43.60	28.32	114.65	48.21	126.64	115.30	120.44	
Kurtosis	-0.005	11.454	13.901	1.193	0.915	-0.115	5.764	1.449	1.882	
Skewness	0.654	3.010	2.736	0.673	1.466	0.717	2.255	1.402	1.665	
Studies in the world										
TSPCR[a]	-	-	140	300	75	3	20	300	100	(TSPCR, 2005)
CSQG[b]	-	-	63	200	50	1.4	40	70	64	(CCME, 2007)
UCC[c]	39200	774	28	67	47	0.09	17.30	17	92	(Rudnick and Gao, 2003)
ES[d]	-	524	17.30	68.1	37	0.28	10.40	32	94.8	(Kabata-Pendias, 2011)
WS[e]	-	488	38.9	70	29	0.41	11.30	27	70	(Kabata-Pendias, 2011)
MAC[f]	-	-	150	300	60	5	50	300	200	Kabata-Pendias, 2011)
Mouriki-Thiva, Greece	46600	1010	32	67	1591	-	54	277	24	(Antibachi et al., 2012)
Daye city, China	-	-	105	159	25.80	1.41	-	43.7	60.7	(Du et al., 2015)
Harran Plain, Türkiye	37505	679	27	68	89	-	16	10.6	85	(Varol et al., 2020)

a: Turkish Soil Pollution Control Regulation; b: Canadian soil quality guidelines; c: Upper continental crust; d: European soils; e: Worldwide soils; f: Maximum allowable concentrations

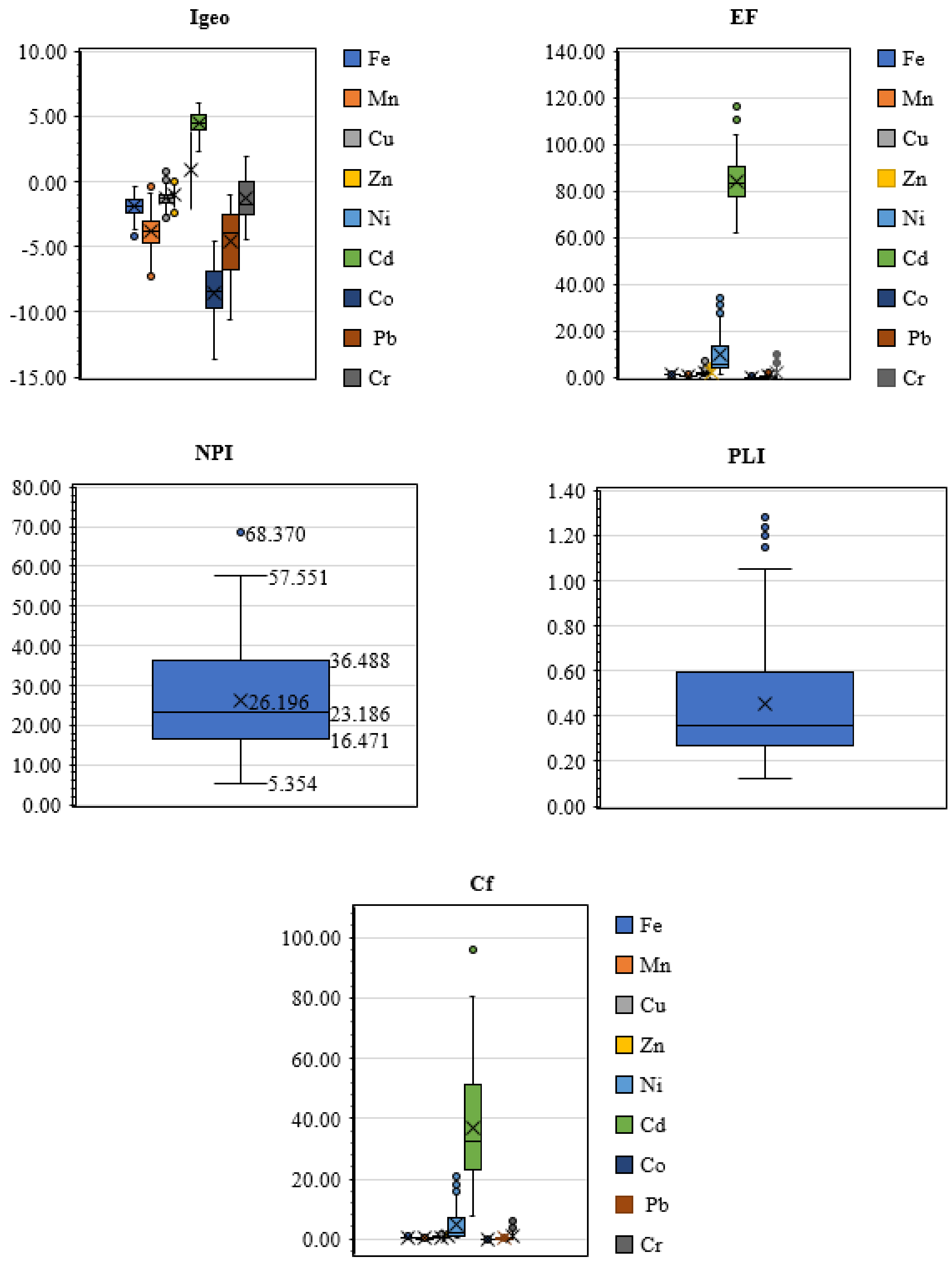

Figure 2. Graphs of the averages of environmental risk indices (EF, Igeo, Cf and NPI)

Şekil 2. Çevresel risk endekslerinin (EF, Igeo, Cf ve NPI) ortalamalarının grafikleri

The Er index was calculated for the individual ecological risks of MMs in the soil, and the NRI and RI indices were calculated to see the ecological risks of MMs as a whole. The averages of the Er, NRI and RI indices, which determine the ecological pollution status of MMs in soils, are shown in Figure 3 and Table 4. Table 1 was also used for pollution classifications.

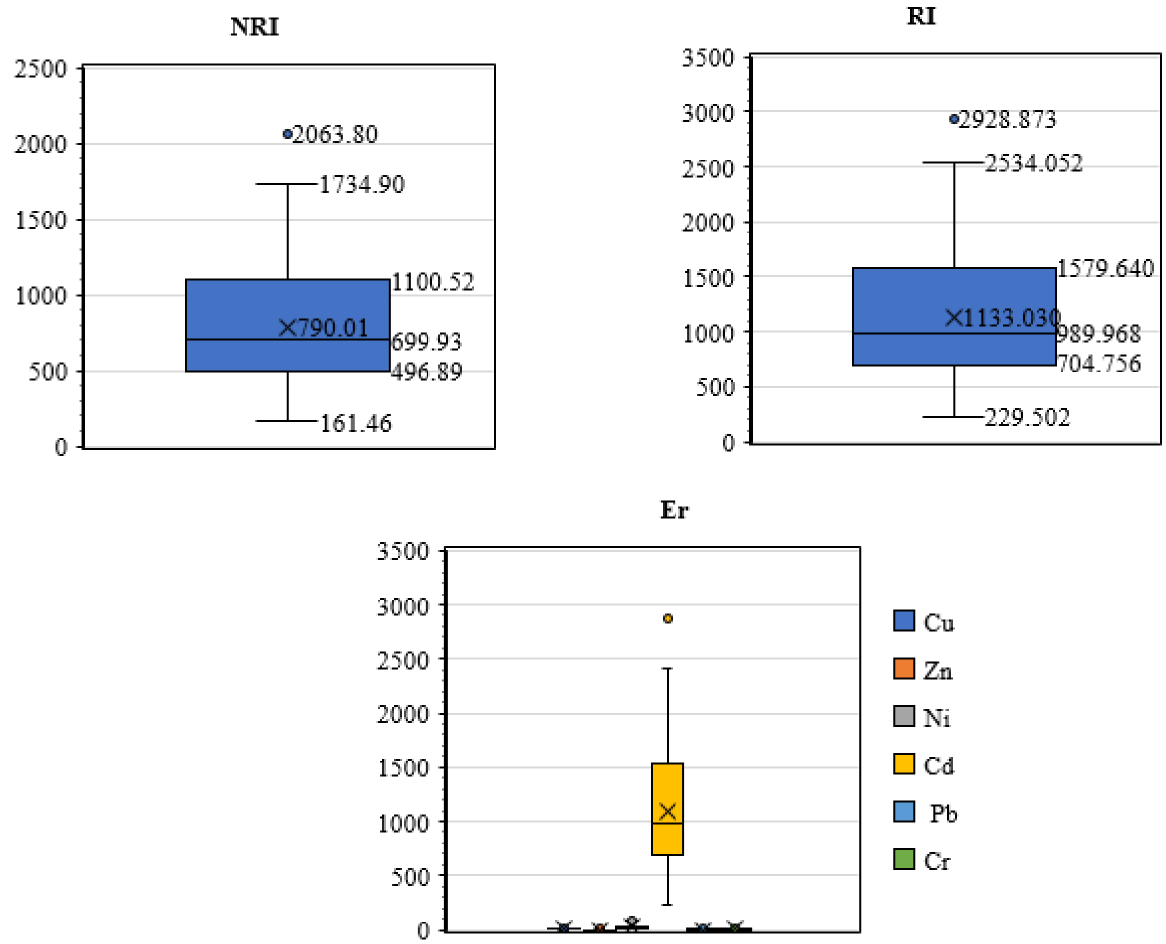

Figure 3. Graphs of the averages of ecological risk indices (Er, NRI and RI)

Şekil 3. Ekolojik risk endekslerinin (Er, NRI ve RI) ortalamalarının grafikleri

The mean values of MMs in agricultural soils were found to be 3.31 for Cu, 0.78 for Zn, 24.75 for Ni, 1101.16 for Cd, 0.80 for Pb and 2.24 for Cr. According to Table 1, since the mean values of Cu, Zn, Ni, Pb and Cr are < 40, these MMs pose 'low potential ecological risk' in agricultural soils. Cd has a mean value of ER > 320, indicating 'very high potential ecological risk' (Table 3). The mean value of NRI of MMs in the study was (790.01)> 320 (Figure 3 and Table 4), indicating 'very high risk'. The mean RI of MMs was 1133.03 and >600 (Figure 3 and Table 4). RI values similar to our results were also reported by Kumar et al. (2019) and Wu et al. (2018). In addition, high RI values are very rare in the literature. In the study area, intensive mining and intensive use of phosphate fertilizers and pesticides to increase agricultural production caused Cd enrichment in agricultural soils and this toxic MM contributed the most to NRI and RI. When Table 4 is analyzed, it can be predicted that the maximum concentrations of Ni MM in individual indices are higher than the high limit values compared to the limit values in Table 1, and this element may pose a threat for the future in agricultural soils. However, it should not be ignored that the use of UCC data determined by Rudnick and Gao (2003) in the calculation of individual indices may be misleading as to whether soils are contaminated by MMs (Aytop et al., 2023). This is because high concentrations of an MM in the soils under investigation may be due to different anthropogenic activities, as in this study, or may be directly related to the parent material of the soils under study. Therefore, sampling of the parent material should also be carried out in such studies.

Sources of contamination of MMs in agricultural soils

The relationships between MMs in the study soils determined using Pearson correlation matrix are given in Table 3. The table shows that there are significant and very high relationships at P≤0.01 level between Fe and Ni, Cd and Cr (r: 0.700**, r: 0.975** and r: 0.703**, respectively), between Ni and Cd and Cr (r: 0.739** and r: 0.846**, respectively) and between Cd and Cr (r: 0.714**). The least significant relationships were found between Mn and Zn and Co at P≤0.05 (r: 0.198* and r: 0.154*, respectively). Low to moderate correlations were found among other MCCs, significant at the P≤0.01 level.Some researchers have reported that regular and highly related MCCs may have a common source, interdependence or similar action (Pan et al., 2016). The significant and high correlations identified in the present study indicate that the MMs in the investigated agricultural soils originate from common sources and especially from lithologic activations.

Table 3. Relationships among MMs

Çizelge 3. MM'ler arasındaki ilişkiler

MMs	Fe	Mn	Cu	Zn	Ni	Cd	Co	Pb	Cr
Fe	1								
Mn	0.490**	1							
Cu	0.377**		1						
Zn	0.520**	0.198*	0.554**	1					
Ni	**0.700****	0.318**			1				
Cd	**0.975****	0.464**	0.384**	0.528**	**0.739****	1			
Co	0.231**	0.154*			0.424**	0.281**	1		
Pb		0.235**		0.223**				1	
Cr	**0.703****	0.425**			**0.846****	**0.714****	0.424**		1

** Correlation is significant at the P≤0.01 level

* Correlation is significant at the P≤0.05 level

In order to determine the pollution sources and numbers of MMs in agricultural soils, the data obtained from the research area were normalized by z-scale transformation before PCA and FA tests. In addition, Bartlett's test and KMO score were checked to determine whether the data were suitable for PCA and FA. The KMO score (0.766, P> 0.50) and Bartlett's test of sphericity (0.0000, P< 0.05) showed that the data set was suitable for FA and PCA (Fei et al., 2019; Varol et al., 2021). PCA/FA results are given in Figure 4 and Table 4. Accordingly, three principal components (PCs) with eigenvalues > 1 and explaining 76.42% of the total variance were obtained. The first PC showed a strong '+' loading for Fe (0.736), Mn (0.455), Ni (0.911), Cd (0.766), Co (0.621) and Cr (0.909). Therefore, the most important component in the study was found to bePC1, which explained 45.14% of the total variance. Therefore, the most important component in the study was PC1, which explained 45.14% of the total variance. These data suggest that lithogenic activities due to geochemical erosion of the parent material were active in the first PC (Chandrasekaran et al. 2015).

Some of the soils of the research area consist of serpentine parent material. Serpentine soils rich in heavy metals such as Co, Cr and Ni also contain high levels of Fe (Avcı, 2005). In addition, Mamat et al. (2014) reported that metalloids such as Mn, Ni and Cr in soils are mainly influenced by geochemical sources and are generally derived from geological origin. The second PC was dominated by Fe (0.530), Cu (0.863), Zn (0.844) and Cd (0.534) and explained 19.17% of the total variance. In PC2, Fe, Cu, Zn and Cd accumulated in the study soils as a result of anthropo activities such as mining and agricultural activities. The General Directorate of Mineral Research and Exploration of Turkey (MTA, 2024) reported that Fe is mined in the study area. Although there is mining in the region, the main sources of MMs dominated by PC2 are thought to be chemical fertilizers (containing phosphorus and microelements) and pesticides applied intensively to increase crop yields. Fertilizers of animal origin are also used in the region.

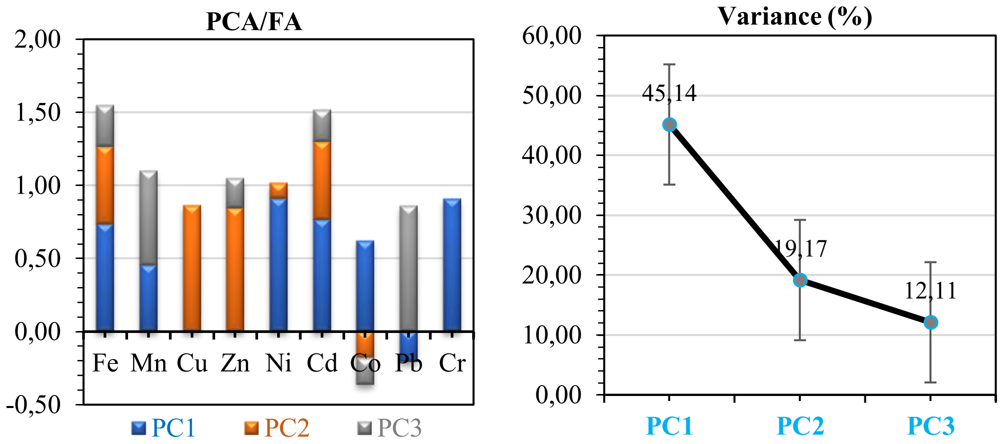

Figure 4. PCA/FA plots of MMs in the research area soils

Şekil 4 . Araştırma alanı topraklarındaki MM'lerin PCA/FA grafikleri

Cu and Zn may have accumulated in the soil due to retention by organic matter and clay. The results of the environmental and ecological indices indicate that of these MMs, only Cd may be toxic. The third PC was highly loaded with Mn (0.645) and Pb (0.858). PC3 contributed only 12.11% of the total variance. This component is attributed to mixed non-natural activities such as industrial, mining and agricultural activities and atmospheric factors. It was concluded that Mn may have accumulated in the soils of the research area as a result of the use of industrial and mining wastewater and agricultural waters containing fertilizer and pesticide residues. The use efficiency of fertilizers and pesticides is generally low. Therefore, they are mostly lost to soil, water and air (Tudi et al., 2021). In the study, Pb was considered as an atmospheric factor. Airborne Pb may have originated from fumes from vehicle exhausts, which occur frequently throughout the study area (Chandrasekaran et al., 2015). In the study area, charcoal or firewood is produced from unsuitable forest trees and the trees used in this production are burned for 15-20 days with some techniques. Emissions from these and fossil fuels from transportation could be the source of Pb in surface soils. In a study conducted to determine Cd and Pb pollution and their regional distribution in an agricultural area, it was reported that Pb pollution in surface soil was caused by exhaust gases and Cd pollution was caused by phosphorus fertilizers (Merry & Tiller, 1991). Cd has been controlled by both anthropogenic and lithogenic sources (Aytop, 2023).

Health risks to humans from MMs in agricultural soils

The health risk consequences of MMs for children and adults within the study area region are presented in Figure 5 and Table 5.

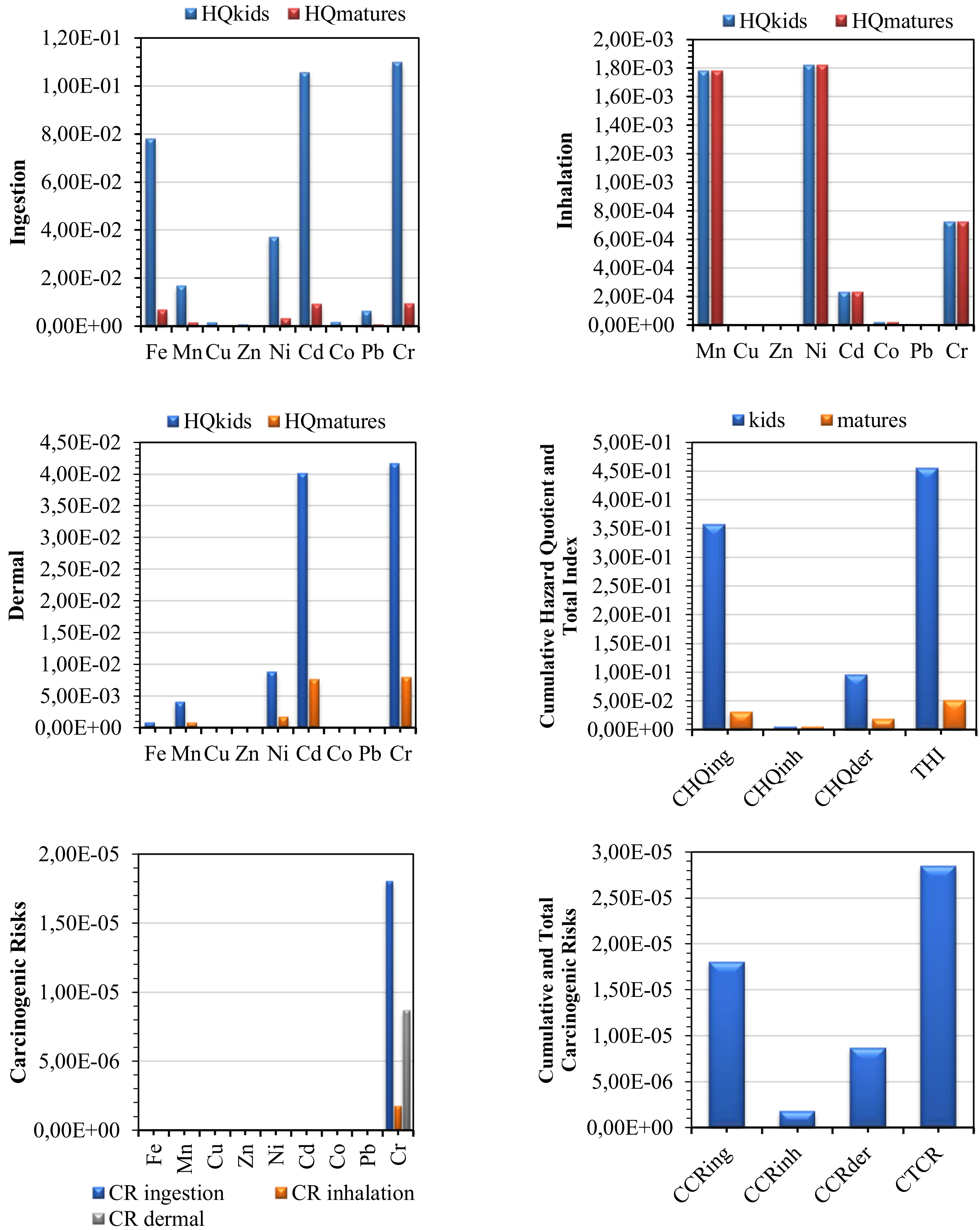

Figure 5. Health risks of averages of MMs in agricultural soils on kids and adults (HQing-HQinh-HQder, CHQing-CHQinh-CHQder, THI, CCRing-CCRinh-CCRder and CTCR)

Şekil 5. Tarımsal topraklardaki MM ortalamalarının çocuklar ve yetişkinler üzerindeki sağlık riskleri (HQing-HQinh-HQder, CHQing-CHQinh-CHQder, THI, CCRing-CCRinh-CCRder ve CTCR)

At present, these values should be interpreted as not posing a risk to either children or adults. But he HQ, CHQ, HI and ΣHI values of MMs calculated for both children and adults are <1.0, indicating that non-carcinogenic risks are not currently seen for children and adults through exposure routes to MMs in soils through skin contact, inhalation and ingestion. Similar findings were also reported by Praveena et al. (2018). However, children's HQing Cd and Cr MMs were higher and closer to 1.0 than adults. This indicated that these two MMs may pose a future risk for children. The CHQ values for the three exposure pathways followed the order CHQing (3.57E-01 and 3.06E-02, respectively) > CHQder (9.54E-02 and 1.82E-02, respectively) > CHQinh (4.58E-0.3 and 4.58E-03, respectively) in both children and adults (Table 5). In addition, the ΣHI value for children was 8.94 times higher than that of adults, indicating that children are more susceptible to the adverse health effects of MMs in soil. Some researchers have reported similar results (Shaheen et al., 2020; Varol et al., 2020; Sun et al., 2021; Wang et al., 2023).

The carcinogenic risk values of MMs determined from the research soils are given in Figure 5 and Table 5. In carcinogenic risk assessment; CR< 10^{-6} (very low), 10^{-6}< CR<10^{-5} (low), 10^{-5}< CR<10^{-4} (moderate), 10^{-4}< CR<10^{-3} (high) and CR > 10^{-3} (very high), these five classifications are used (Tepanosyan et al., 2017). According to this classification; Ni (1.22E-08), Cd (1.20E-09) and Co (2.84E-10) values through CRinh were found to be <10^{-6} (very low), Cr (1.74E-06 and 8.67E-06) values through CRinh and CRder (1.74E-06 and 8.67E-06, respectively) were found to be 10-6< CR <10^{-5} (low) and Cr (1.80E-05) value through CRing was found to be 10^{-5}< CR <10^{-4} (medium). The results indicate that carcinogenic risks from exposure to Ni, Cd, Co and Cr in agricultural soils are not currently present for residents in the region. ΣCR's Ni, Cd and Co values and class ranges were the same as CRinh. The Cr class of ΣCR was similar to the Cr class of CRing. The ingestion, inhalation and dermal values of CCR were similar to the Cr values and risk classes of the CR exposure routes. The value and carcinogenic risk class of CΣCR was similar to the Cr value and risk class of ΣCR. All these results are between and below the USEPA's (2023a) acceptable risk range of 1×10-4 to 1×10-6, indicating that carcinogenic risks from exposure do not pose a danger to residents for the time being. However, in the future, Cr may pose a carcinogenic risk to residents through ingestion. The CCRing value was higher than the CCRinh and CCRder values, indicating that the receptors were first exposed to MMs in soil through ingestion. Similar to previous studies (Zeng et al., 2019; Tudi et al., 2021), Cr was the largest contributor to ΣCR and CΣCR in this study. Although Cd was found to be the most important pollutant in agricultural soils in this study, the potential exposure level of Cr was found to be higher than that of Cd, which is attributed to the higher amount of Cr in surface soils than Cd.

In conclusion, this study was carried out to determine the potential environmental and ecological pollution risks of MMs in agricultural soils in Osmaniye province, their potential sources, and to investigate the health risks they pose for the residents of the region. The research results showed that Ni, Co and Cr in the soils resulted from geological weathering of the parent material, Fe, Mn and Cd from both lithogenic and different anthropogenic sources, and Pb from atmospheric factors.

One of the important conclusions of this research is that the use of UCC data in calculating individual indices may be misleading as to whether soils are contaminated by MMs. Because the high concentration of a MM in soils where pollution is investigated may directly depend on the parent material of the soils studied, as in this study. Therefore, in such studies, sampling should be done from the main material.

One of the important results of this research is that the use of UCC data in the calculation of individual indices may be misleading as to whether soils are contaminated by MMs. The averages of Ni, Cd and Cr among the MMs in the agricultural soils investigated exceeded the UCC concentrations used in the calculation of environmental and ecological risk indices. Again, in the study, the average Ni and Cr concentrations were found to be above the Ni and Cr amounts of the MAC and Harran Plain agricultural soils in Urfa, Türkiye. According to Igeo, EF and Cf results, it was determined that agricultural soils were 'heavily to extremely polluted', 'extremely high enrichment' and 'very high contamination' by Cd, respectively. is. Because the high concentration of a MM in soils where pollution is investigated may directly depend on the parent material of the soils studied, as in this study. Therefore, in such studies, sampling should be done from the main material.

Again, according to Er results, it was determined that the soils had a 'very high potential ecological risk' in terms of Cd. Again, Cd made the biggest contribution to the NPI, NRI and RI results. For this reason, soils have 'strongly polluted', 'very high risk' and 'very high ecological risk', respectively, in terms of Cd. All non-carcinogenic risk values (HQ, CHQ, HI and ΣHI) were below 1, indicating no non-carcinogenic health risk for children and adults

Except for HQinh, other HQ and HI values of MMs were higher in children than in adults, indicating that children were more affected by MMs in agricultural soils. CR, CCR, ΣCR, and CΣCR were within and below the USEPA acceptable risk range. This explained that there were no carcinogenic risks for the residents of the region for now.

STATEMENT OF CONFLICT OF INTEREST

The authors declare no competing interests.

AUTHOR'S CONTRIBUTIONS

The authors declare that they have contributed equally to the study.

STATEMENT OF ETHICS CONSENT

This article does not require ethical approval as there are no studies with human or animal subjects.

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