ORIGINAL PAPER

Heavy metals and their sources, potential pollution situations and health risks for residents in Adıyaman province agricultural lands, Türkiye

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Received: 2 July 2022 / Accepted: 24 October 2022 / Published online: 13 November 2022 © The Author(s), under exclusive licence to Springer Nature B.V. 2022, corrected publication 2022

Abstract In this study, the contents of heavy metals (HMs) such as Al, Cd, Co, Cr, Cu Fe, Mn, Ni, Pb and Zn in soil samples collected from 403 sampling locations of the agricultural lands of Adıyaman Province (Türkiye) were determined by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). The mean concentrations of Al, Cd, Co Cr, Cu Fe, Mn, Ni, Pb and Zn HMs were detected 28,986, 3.60, 15, 127, 52.67, 45,830, 817, 62.40, 10.75 and 66.25 mg kg^{-1} , respectively. These results showed that the average concentrations of Cd, Cr, Cu, Fe, Mn and Ni exceeded the Upper continental crust average. To determine and to evaluate the contamination status and distribution of HMs in agricultural soils, metal pollution parameters such as enrichment factor (EF), geoaccumulation index (I_{geo}) , contamination factor (Cf), pollution load index, potential ecological risk factor (Er), and potential ecological risk index (RI) were used. Factor analyses (FA) and principal component analyses (PCA) indicated that Cd, Cr and Ni levels were infuenced by anthropogenic sources, Fe by both lithological and anthropogenic sources, and other HMs by lithogenic origins. For both children and adults, the hazard index (HI) and total hazard index (THI) values of HMs were < 1 , suggesting that non-carcinogenic health risks to residents through

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ingestion, inhalation pathways, and dermal contact were currently absent. In addition, the cumulative carcinogenic risk (CCR) results were within the acceptable risk range $(10^{-4} \text{ to } 10^{-6})$. The results showed that children were more sensitive to the non-carcinogenic and carcinogenic efects of HMs.

Keywords Adıyaman agricultural soils · HMs · PCA and FA · Pollution sources · Risks

Introduction

The main source of agricultural production is soils. Agricultural soils, which took thousands of years to form, is the only resource that cannot be produced and is impossible to renew. Soil is a natural resource that provides living things with basic needs such as food, medicine, and clean water (Soil Survey Staf, [2014\)](#page-16-0). The rapid urbanization and industrialization demands of human beings pose signifcant threats to soils such as pollution, salinization, decrease in biodiversity organic matter and erosion in recent years (European Commission (EC), [2006;](#page-14-0) Aytop & Şenol, [2022](#page-14-1)). Soils are considered to be the most exposed part of the biosphere to the accumulation of HMs (Marchand et al., [2011](#page-16-1)). HMs, regardless of their sources, are often associated with soil pollution (Zhang et al., [2017\)](#page-17-0). The ability of soils to accumulate HMs depends on their physical and chemical properties, as well as the type of soil and the nature of

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heavy metals (Kabata-Pendias, [2011\)](#page-15-0). HMs, which is quite stable in soil, is not washed and does not decompose (Lionetto et al., [2012;](#page-15-1) Mazurek et al., [2017](#page-16-2); Mmolawa et al., [2011](#page-16-3)). The critical sources from which humans take harmful HMs into their bodies are agricultural products (Harmanescu et al., [2011\)](#page-15-2). Soils contaminated with HMs can be seen as a potential and real environmental worry (Islam et al., [2016](#page-15-3); Jia et al., [2018;](#page-15-4) Motuzova et al., [2014\)](#page-16-4). The level of pollution in agricultural soils, which is also important for human and animal health, should be carefully monitored (Wong et al., [2002\)](#page-17-1). HMs resources can be from anthropogenic efects or natural processes (lithogenic and pedogenic) (Akbay et al., [2022;](#page-14-2) Huang et al., [2018;](#page-15-5) Li et al., [2009](#page-15-6), [2018](#page-15-7); Mazurek et al., [2017](#page-16-2); Rivera et al., [2015](#page-16-5); Wang et al., [2012](#page-17-2)). The highest concentrations of HMs are usually found in the topsoils, because surface layers, especially organic horizons, are highly skilled at binding HMs (Acosta et al., [2015](#page-13-0)). The efect of anthropogenic inputs on the accumulation of HMs in soils is greater than that of natural resources (Dong et al., [2018](#page-14-3); Ni et al., [2018\)](#page-16-6). The main anthropogenic sources for HMs in soils are exhaust emissions, domestic wastes, industrial works and agricultural activities such as fertilizer and pesticide applications (Muhammad et al., [2011](#page-16-7); Chen et al., [2015;](#page-14-4) Antoniadis et al., [2017](#page-14-5); Dong et al., [2018;](#page-14-3) Huang et al., [2018](#page-15-5); Ni et al., [2018;](#page-16-6) Kumar et al., [2019](#page-15-8)). The natural spread of metals to the environment is usually caused by forest fres and events such as release from plants, abrasion of rocks, erosion and volcanic eruptions (Can et al., [2021;](#page-14-6) Kapahi & Sachdeva, [2019;](#page-15-9) Muradoglu et al., [2015](#page-16-8); Ozturk et al., [2017](#page-16-9)).

The best method of combating HMs pollution in agricultural areas is to take the necessary measures without allowing them to accumulate in the soils, because cleaning HMs from contaminated soils is a very difficult, time-consuming and costly task (Hu et al., [2020](#page-15-10); Varol et al., [2021\)](#page-17-3). In order to prevent agricultural soil pollution, it is extremely important to determine the pollution status, environmental and ecological risks of HMs and to reveal their sources. In addition, estimating the human health risks posed by HMs is important for making decisions on the management of soil pollution (Deng et al., [2020](#page-14-7); Fei et al., [2019](#page-15-11); Varol et al., [2020](#page-17-4), [2021\)](#page-17-3). While soil contamination indices such as EF, *I*geo, Cf, PLI, Er and RI are used to determine the pollution status of soils (Baltas et al., [2020](#page-14-8); Fei et al., [2019;](#page-15-11) Ma et al., [2017](#page-15-12); Mazurek et al., [2019](#page-16-10); Shaheen et al., [2020;](#page-16-11) Varol et al., [2020,](#page-17-4) [2021\)](#page-17-3), health risk assessment indices such as HI and CR are applied to reveal risks arising from exposure to TMs (Baltas et al., [2020](#page-14-8); Deng et al., [2020](#page-14-7); Jia et al., [2018;](#page-15-4) Rinklebe et al., [2019](#page-16-12); Varol et al., [2020,](#page-17-4) [2021\)](#page-17-3). FA, PCA and correlation analyses are generally used to identify possible input sources of HMs in soils and to determine the relationships among HMs (Kumar et al., [2019](#page-15-8); Ma et al., [2017;](#page-15-12) Ni et al., [2018](#page-16-6); Varol et al., [2020](#page-17-4), [2021](#page-17-3)). It is suggested that these risk indices should be evaluated together in order to be used efectively in soils of a particular region (Fei et al., [2019](#page-15-11); Jia et al., [2018](#page-15-4); Rinklebe et al., [2019](#page-16-12); Varol et al., [2020](#page-17-4), [2021\)](#page-17-3). For this reason, many researchers focus on studies on the environmental and ecological risks of HMs pollution in soil and its efects on human health (Gujre et al., [2021](#page-15-13); Mahurpawar, [2015](#page-15-14); Mishra et al., [2019;](#page-16-13) Varol et al., [2020](#page-17-4), [2021;](#page-17-3) Yaylalı-Abanuz, [2011](#page-17-5); Zeng et al., [2019](#page-17-6)). However, it is seen that these studies in the literature are studies on the determination and monitoring of the concentrations of HMs: pollution index studies using heavy metal concentrations and pollution levels in agricultural soils in Türkiye, and health risk assessment index studies showing how much of these can afect human health are extremely rare (Malkoç et al., [2010;](#page-16-14) Arslan & Çelik, [2015;](#page-14-9) Sungur, [2016;](#page-17-7) Baltaş et al. [2020;](#page-14-8) Varol et al., [2020;](#page-17-4) Varol et al., [2021](#page-17-3)). In addition, there are no detailed data on the heavy metals in their soil and their health risks for people living in Adıyaman Province located in the southeast of Türkiye. Therefore, generating data on the current state of HM pollution in the agricultural lands of Adıyaman are very important for human health and will also shed light on future research. In this context, the aim of the present study is to determine total concentrations of 10 HMs (Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn) in 403 surface soils collected from Adıyaman Province agricultural areas, to defne the possible sources of HMs by applying Pearson correlation, PCA and FA, and to estimate the ecological and environmental risks of HMs by using EF, I_{geo} , Cf, PLI, Er and RI. It is also to assess both non-carcinogenic and carcinogenic health risks for adults and children (residents) exposed to HMs.

Materials and methods

Study area

Adıyaman Province is located in the Middle Euphrates section in the Southeastern Anatolia Region of Türkiye. Adıyaman Province is between 37° 25′ and 38° 11′ north latitudes and 37° 25′ and 39° 15′ east longitudes. Its area is 7614 km^2 , with lakes 7871 km^2 , and its altitude is 669 m. The climate of the mountainous region to the north of the Anti-Taurus Mountains that divide Adıyaman from east to west and the climate of the region to the south are diferent from each other. In the south, summers are hot and dry, and winters are rainy and mild; in the north, summers are cool and dry, and winters are cold and rainy. The climate of the province, which acts as a bridge between the Eastern Anatolia and the Mediterranean Regions, is diferent from the other provinces in the region due to this feature. After the formation of the Atatürk

Dam Lake area, there has been a softening in the climate of the province and an increase in the humidity rate. The prevailing winds in the province are in the north, northwest and northeast directions (Anonymous, [2022a](#page-14-10)). The industry of Adıyaman Province generally consisted of small businesses (Anonymous, [2022b\)](#page-14-11). Its population is 632148, and the number of registered farmers is 28967 people. The agricultural area is 2244544 km^2 (TOB, 2022). There are many mineral deposits and businesses in the province (MTA, [2022\)](#page-16-15). In addition, 26.02% of Türkiye's oil is produced in Adıyaman (4,300,000 barrels) (TPAO, [2022\)](#page-17-9).

Soil sampling and analyses

In this study, sampling points were chosen to represent all agricultural areas of Adıyaman Province (Fig. [1](#page-2-0)). Thus, a total of 403 surface $(0-20 \text{ cm})$ soil samples were collected between April 2016 and May

Fig. 1 Adıyaman Province and sampling stations

2018. Samples were taken from agricultural lands every 2.5 km according to the grid sampling method. A composite soil sample was obtained at each sampling point by mixing four random subsamples. Collected soil samples were placed in nylon bags and taken to the laboratory.

All samples were naturally air-dried. Then, they were passed through a 2 mm sieve and stone, gravel and plant parts were removed. The sieved samples were pulverized with a morter and pestle, passed through a 0.5 mm sieve and stored in clean polyethylene bottles. In this study, 10 HMs, namely Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn, were analyzed in soil samples. These were chosen because they cause a lot of soil pollution and health risks (Rinklebe et al., [2019](#page-16-12)). The content of the ten HMs was determined in the Soil, Plant and Water Analysis Laboratory of Kahramanmaraş East Mediterranean Transitional Zone Agricultural Research Institute authorized by the Ministry of Agriculture and Forestry of Türkiye. Soil samples were digested in Tefon vessels containing a mixture of HCl and $HNO₃$ at 1:3 ratio (Aqua regia wet digestion method) using a wet digestion system in a CEM MARS 6 (USA) microwave oven. The solutions were then diluted with ultrapure water to a volume of 50 mL. Concentrations of 10 HMs were measured by an Agilent 5100 (USA) brand inductively coupled plasma‒optical emission spectrometry (ICP‒OES). Assurance and control of results were performed using method blanks, certifed reference material (CRM) (LGC6187, river sediment) and replicates. Merck's (Darmstadt, Germany) standard solutions were used for the calibration curves. In this study, one CRM was digested and analyzed in every 21 soil samples. Recovery of HMs in CRM ranged from 90.9 to 108.3% (Table [1\)](#page-3-0). An analysis for a soil was done two times (in two repetitions) and the arithmetic mean of the results of these two analyses was used in the data analysis. It was done this way in all soils.

Environmental risk assessment

Enrichment factor (EF)

EF is used to aveluate soil pollution levels and the possible impact of human activities on HMs concentrations in soils (Loska et al., [2004;](#page-15-15) Taşpınar et al., [2021;](#page-17-10) Varol et al., [2020;](#page-17-4) Wu et al., [2018](#page-17-11)). EF is calculated with the following formula:

Table 1 Parameters values used to the health risk heavy metals i children and a

$$
EF = \left[\frac{C_i}{C_{\text{ref}}}\right] \text{sample} / \left[\frac{C_i}{C_{\text{ref}}}\right] \text{background}
$$
 (1)

In the formula, C_i is the concentration of HM of the soil sample. C_{ref} is the content of reference HM (Bern et al., [2019\)](#page-14-12). In this study, Fe was used as a reference HM due to its high content (Aytop, [2022;](#page-14-13) Taşpınar et al., [2021](#page-17-10)). UCC values reported by Rudnick and Gao [\(2004](#page-16-16)) were used as background concentrations of HMs. The EF classes are presented in Table [2.](#page-4-0) The EF values were also classified by Sutherland (2000) (2000) : low enrichment (0.5–2), moderate enrichment (2–5), signifcant enrichment (5–20), very high enrichment $(20-40)$ and extremely high enrichment (>40) .

Geoaccumulation index (Igeo)

*I*geo was found by Müller [\(1969](#page-16-17)) to determine and classify the pollution level of HMs in soil. I_{geo} is calculated with the following formula:

$$
I_{\rm geo} = \log_2 \left[\frac{C_i}{1.5 \times B_i} \right] \tag{2}
$$

In the formula, B_i is the geochemical background value of HM (Rudnick & Gao, [2004](#page-16-16)) and Ci is the

concentration of HM. A coefficient of 1.5 is used to minimize the efect of possible variations in the background values of natural processes in the soil (Al-Haidarey et al., 2010 ; Baltas, 2020). I_{geo} classes are given in Table [2.](#page-4-0) I_{geo} index was also classified in 6 diferent groups by Muller (Buccolieri et al., [2006](#page-14-15)). These were unpolluted (< 0) , unpolluted to moderately polluted $(0-1)$, moderately polluted $(1-2)$, moderately to highly polluted (2–3), strongly polluted (3–4), strongly to extremely polluted (4–5) and extremely polluted (>5) , respectively.

Contamination factor (Cf)

Cf is used to determine the level of HMs contamination in soils (Hakanson, [1980;](#page-15-16) Varol et al., [2020](#page-17-4)). Cf is calculated with the following formula:

$$
C_f^i = \frac{C^i}{C_n^i} \tag{3}
$$

in this formula, C_i is the content of HM and C_f^i is the background (or pre-industrial) concentration of HM (Rudnick & Gao, [2004](#page-16-16); Varol et al., [2020](#page-17-4), [2021](#page-17-3)). The Cf classes are given in Table [3.](#page-5-0) Qingjie et al. [\(2008](#page-16-18)) also classifed Cf values as low contamination

Table 2 Relative bioavailability factor, dermal absorption fraction, oral reference dose, oral slope factor, gastrointestinal absorption, inhalation reference concentration, particulate emission factor and inhalation unit risk values for each heavy metal

Heavy metal	Relative bio- availability factor (RBA) (unitless)	Dermal absorption fraction (ABSd) (unitless)	Oral refer- ence dose $(RfDo)$ (mg/ $kg-day)$	Oral slope factor (CSF ₀) (mg/ kg -day $)^{-1}$	Gastro- intestinal absorption (GIABS) (unitless)	Inhalation reference concentra- tion (RFC) $(mg\ m^{-3})$	Particulate emission factor (PEF) $(m^3 \text{ kg}^{-1})$	Inhalation unit $risk$ (IUR) (μ g $\rm m^{-3}$) ⁻¹
Al	1	0.001	$\mathbf{1}$		1	0.005000	1.36×10^{9}	
Cd	1	0.001	0.01		0.025	0.00001	1.36×10^{9}	0.0018
Co	1	0.001	0.0003		1	0.000006	1.36×10^{9}	0.0090
Cr^*	1	0.001	0.003	0.5	0.025	0.000100	1.36×10^{9}	0.0840
Cu		0.001	0.04		1		1.36×10^{9}	$\overline{}$
Fe	1	0.001	0.7		1		1.36×10^{9}	
Mn		0.001	0.024		0.04	0.000050	1.36×10^{9}	
Ni		0.001	0.02		0.04	0.000090	1.36×10^{9}	0.0003
Pb		0.001	0.0014		1		1.36×10^{9}	\equiv
Zn		0.001	0.3		1		1.36×10^{9}	
References	USEPA (2019d)	USEPA (2004)	USEPA (2019e) Jia et al., 2018)	USEPA (2019e)	USEPA (2019e)	USEPA (2019e)	USEPA (2019e)	USEPA (2019e)

factor $(< 1$), moderate contamination factor $(1-3)$, considerable contamination factor (3–6) and very high contamination factor (>6) .

Pollution load index (PLI)

PLI shows the overall pollution status of the studied area and combines the Cf values of all HMs (Kowalska et al., [2018](#page-15-17); Rinklebe et al., [2019](#page-16-12)). PLI is calculated with the following formula (Baltas et al., [2020](#page-14-8); Madrid et al., [2002](#page-15-18)):

$$
PLI = \sqrt[n]{C_{f1}xC_{f2}xC_{f3}x, ..., xC_{fn}}
$$
 (4)

In the formula, *n* is the number of HMs analyzed. If the $PLI < 1$, the investigated area is not contaminated by metals, but if the PLI>1, the investigated area is contaminated by metals (Chakravarty & Patgiri, [2009\)](#page-14-19).

Ecological risk assessment

Potential ecological risk factor (Er)

Er is used to evaluate the potential ecological risk of a single HM in the soil examined (Hakanson, [1980](#page-15-16)). The formula for Er is as follows:

$$
E_r^i = T_r^i \times C_f^i \tag{5}
$$

In the formula, T_r^i is the toxic response factor of HM, they are 30, 2, 5, 5, 5 and 1 for Cd, Cr, Cu, Ni, Pb and Zn, respectively (Hakanson, 1980). C_f^i is the contamination factor of HM. The Er classes are given in Table [3](#page-5-0). Hakanson also classified Er values as low (<40), moderate (40–80), considerable (80–160), high $(160-320)$ and very high ecological risk (>320).

Potential ecological risk index (RI)

RI is calculated to determine the level of ecological risk caused by multi-HMs in the soil (Hakanson, [1980;](#page-15-16) Varol et al., [2020](#page-17-4), [2021](#page-17-3)). The formula for RI is as follows:

$$
\sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \times C_f^i
$$
 (6)

In the formula, *n* is the number of HMs $(n=6$ in this study) and E_r^i is the potential ecological risk factor of HMs. RI classes are given in Table [2](#page-4-0). Qingjie vd (2008) (2008) also classified RI values as; RI < 150, low ecological risk; 150≤RI<300, moderate ecological risk; $300 \leq RI < 600$, considerable ecological risk; and RI>600, very high ecological risk.

Human health risk assessment

In this study, we tried to determine the health risks of HMs in Adıyaman Province soils for children and adults residing here. We evaluated both non-carcinogenic and carcinogenic health risks for children and adults exposed to HMs in soil through accidental ingestion, dermal contact and inhalation (Li et al., [2017;](#page-15-19) USEPA, [2019a;](#page-17-22) Varol et al., [2020,](#page-17-4) Varol et al., [2021\)](#page-17-3). HMs in soil were calculated using hazard quotients (HQs) (USEPA, [2007](#page-17-23); Jia et al., [2018](#page-15-4); Wu et al., [2018\)](#page-17-11). Carcinogenic health risks were estimated only for Cr due to the lack of carcinogenic slope factors (ingestion and dermal) of other HMs. The non-carcinogenic risks (HQs) and carcinogenic risks (CRs) of HMs for resident by ingestion, dermal contagion and inhalation routes were calculated using the below formulas (USEPA, [2019b](#page-17-24)). All terms in these equations appear in Tables [1](#page-3-0) and [2](#page-4-0).

Non-carcinogenic risks:

$$
HQ_{\text{ingestion}} = \frac{Cs \times IRS \times RBA \times EF \times ED}{BW \times AT \times RfDo \times 10^6} \quad (7)
$$

$$
HQ_{\text{dermal}} = \frac{Cs \times SA \times AF \times ABSd \times EF \times ED}{BW \times AT \times RfDo \times GIABS \times 10^6}
$$
\n(8)

$$
HQ_{\text{inhalation}} = \frac{Cs \times EF \times ED}{AT \times RFC \times DEF}
$$
 (9)

Carcinogenic risks:

$$
CR_{\text{ingestion}} = \frac{Cs \times IFS \times RBA \times CSFO}{AT \times 10^6} \tag{10}
$$

In the formula, $IFS = \frac{EF \times EDC \times IRSa}{BWc} + \frac{EF \times EDA \times IRSa}{BWa}$

$$
CR_{\text{dermal}} = \frac{Cs \times DFS \times ABSd \times CSP}{AT \times GIABS \times 10^6} \tag{11}
$$

$$
\begin{array}{c} \text{In} \qquad \qquad \text{the} \qquad \qquad \text{formula,} \\ \text{DFS} = \text{DFS} = \frac{\text{EF} \times \text{EDc} \times \text{SAc} \times \text{AFc}}{\text{BWc}} + \frac{\text{EF} \times \text{EDa} \times \text{SAa} \times \text{AFa}}{\text{BWa}} \end{array}
$$

$$
CR_{\text{inhalation}} = \frac{Cs \times EF \times ED \times IUR \times 1000}{AT \times PEF} \quad (12)
$$

In this study, the hazard index (HI) and total carcinogenic risk (TCR) values were determined using the formulas numbered 13 and 14:

$$
HI = HQingestion + HQdermal + HQinhalation \t(13)
$$

$$
TCR = CR_{\text{ingestion}} + CR_{\text{dermal}} + CR_{\text{inhalation}}
$$
 (14)

Also, in this study, the RSL calculator developed by the USEPA [\(2019c\)](#page-17-25) was used to validate all the estimated outcomes associated with health risks.

According to the USEPA (2001) (2001) report, if HI < 1 it is unlikely to have a negative efect on the health of the individual exposed to HMs. However, noncarcinogenic health effects can be seen if $HI > 1$ (Eziz et al., [2018\)](#page-14-20). The acceptable range of TCR is 1×10^{-4} to 1×10^{-6} (USEPA, [1991a\)](#page-17-12). It is accepted that there is no signifcant health risk for humans for TCR values below 1×10^{-6} (Fryer et al., [2006;](#page-15-20) Hu et al., [2012\)](#page-15-21).

Statiscal analyses

Pearson correlation analysis $(p < 0.05)$ was performed to determine the relationships between HMs in soils. After all the data to be analyzed were standardized with z-scale transformation, principal component (PCA) and factor analyses (FA) were performed to determine potential sources of HM in the soil. In addition, Kaiser–Meyer–Olkin (KMO) tests and Bartlett's sphericity were used to test the suitability of all data for PCA and FA. All statistical analyses were done in SPSS 25.0 statistical program.

Results and discussion

The concentrations of HMs in soils

Some descriptive statistics of the ten HMs in the agricultural lands of Adıyaman are given in Table [3](#page-5-0). pH values of only 23 of the total 403 soil samples collected were < 7 , in 380 soils they were $= 7$ and > 7 . The average of pH values of all soils was 7.53. Fe was the HM with the highest amount. Al and Mn followed this. Cd, Pb and Co were lesser amounts than the

other HMs. HMs were ranked from highest to lowest as Fe>Al>Mn>Cr>Zn>Ni>Cu>Co>Pb> Cd according to their determined averages (Table [3](#page-5-0)). When the values of HMs we determined in the study area were compared with the HMs values of UCC (Rudnick & Gao, [2004\)](#page-16-16), it was understood that Zn concentrations were very close to each other. Al, Co and Pb were lower than their UCC values. Cd, Cr, Cu, Fe, Mn and Ni values were approximately 40, 1.4, 1.9, 1.2, 1.1 and 1.3 times higher than the respective UCC values (Table [3](#page-5-0)). This shows that as a result of anthropogenic activities, Cd, Cr, Cu, Fe, Mn and Ni are enriched in the soils.

In general, the average values of all HMs except Cd and Cr were determined above the limit values set by the Turkish Soil Pollution Control Regulation (SPCR, [2005](#page-16-19)). The maximum amounts of Cd and Cr were approximately 1.2 and 1.3 times higher than the corresponding limit values of SPCR ([2005\)](#page-16-19), respectively. Cd in 146 samples (36.2%), Co in 18 samples (2.5%), Cr in 239 samples (24.7%) and Ni in 345 samples (85.6%) exceeded the limit values of SPRC [\(2005](#page-16-19)). Cu, Pb and Zn did not exceed the limit values in any instance (Table 3). When the concentrations of HMs we found were compared with those of worldwide soil HMs (Kabata-Pendias, [2011](#page-15-0)), the Pb and Zn concentrations were approximately 2.5 and 1.1 times lower than the worldwide average values, respectively. However, Cd, Co, Cr, Cu, Mn and Ni were approximately 8.8, 1.3, 2.1, 1.4, 1.7 and 2.2 times higher than their worldwide average values, respectively (Table [3\)](#page-5-0). When we compare the average HM values of European soils reported by Kabata-Pendias ([2011\)](#page-15-0), it is understood that only Pb and Zn are below the averages, while other HMs exceed the averages (Table [3\)](#page-5-0). However, according to the maximum allowable concentrations (MAC) of HMs in the soil (Kabata-Pendias, [2011\)](#page-15-0), it is observed that there is an excess of 2.40 mg kg^{-1} only in Ni concentration (Table [3](#page-5-0)). In this study, the average concentrations of HMs were also compared with the concentrations in agricultural soils of Greece, Iran and China (Table [3](#page-5-0)). While the concentrations of Co, Cr, Fe, Mn, Ni, Pb and Zn of the agricultural lands of Adıyaman Province were lower than the soils of the Mouriki-Thiva region of Greece (Antibachi et al., [2012\)](#page-14-16), the concentration of Cu was 1.7 times higher. While the Co concentrations of the study area soils were found close to the Co contents of the lands of Isfahan, Iran

(Esmaeili et al., [2014](#page-14-17)), the values of Cd, Cr, Cu, Fe and Mn were found to be lower and Al, Ni, Pb and Zn contents also were found higher. Again, the concentrations of Cd, Cr, Cu and Ni of the soils we studied were higher than those of the agricultural soils of the city of Daye in China (Du et al., [2015\)](#page-14-18), while the concentrations of Cu, Pb and Zn were lower.

The average concentrations of HMs in the current study were also compared with HMs in agricultural soils of diferent regions in Türkiye (Table [3\)](#page-5-0). The Cr, Ni and Pb contents in the study were below than the concentrations in Sinop province (Baltas et al., [2020](#page-14-8)), while the Cu, Fe and Mn contents were above. Zn amounts were very close to each other. Cr, Cu, Fe and Mn concentrations in the soils of the Harran Plain (Varol et al., [2020\)](#page-17-4) were below than those in this study, while Al, Ni and Zn contents were higher. Co and Pb amounts were found to be very close to each other. Cr, Ni and Pb contents were lower than those in Çanakkale province (Sungur & İşler, [2021](#page-17-21)), while Cd and Cu contents were higher. Compared to the Alpu Plain soils (Taşpınar et al., [2021\)](#page-17-10), it was seen that the Cd, Cu, Mn and Zn contents of Adıyaman soils were higher and the Co, Cr, Ni and Pb contents were lower (Table [3\)](#page-5-0). Ni and Pb concentrations were lower than those in Malatya province (Varol et al., [2021](#page-17-3)). Aluminum, Cd, Co, Cr, Cu, Fe and manganese were high. The Zn amounts were determined very close to each other (Table [3](#page-5-0)).

These diferent the concentrations of HMs in various parts of the world may be because of spatial heterogeneity in human activities (anthropogenic activities) and in soil properties (natural mineral degradation) (Aytop, [2022;](#page-14-13) Varol et al., [2020,](#page-17-4) [2021\)](#page-17-3).

Environmental risk assessment of HMs

Descriptive statistics of EF, I_{geo} , Cf, Er, PLI and RI results used in the assessment of environmental and ecological risks for Adıyaman soils are presented in Tables [3](#page-5-0) and [4](#page-9-0). When the soils of the study area were examined in terms of average EF values, it was determined that the soils were very highly enriched in terms of cadmium (35.00) $(20 < EFCd < 40)$ and moderately enriched in terms of nickel (3.28) $(2 \times EF_{Ni} \leq 5)$. In addition, the soils were minimally enriched in terms of Al (0.51), Co (0.57), Cr (1.29), Cu (1.57), Fe (1.00), Mn (1.08), Pb (0.66) and Zn (1.10) (EF_{Al, Co, Cr, Cu, Fe, Mn, Pb and Zn} < 2). Enrichment factors of HMs in Adıyaman soils were listed as $\mathrm{EF}_{\mathrm{Cd}}>\mathrm{EF}_{\mathrm{Ni}}>\mathrm{EF}_{\mathrm{Cu}}>\mathrm{EF}_{\mathrm{Cr}}>\mathrm{EF}_{\mathrm{Zn}}>\mathrm{EF}_{\mathrm{Mn}}>\mathrm{EF}_{\mathrm{Fe}}>\mathrm{E}$ $F_{\rm Pb}$ > $EF_{\rm Co}$ > $EF_{\rm Al}$.

Cd (4.31) and Ni (0.86) had positive mean I_{geo} index values, while other HMs had negative mean I_{geo} index values. From the results, it was understood that Adıyaman soils were strongly to extremely polluted by Cd, unpolluted to moderately polluted by Ni and unpolluted by other heavy metals (Table [4](#page-9-0)).

Cf values showed consistent results with EF values. Four (Al, Co, Ni and Fe) of the 10 HMs of which Cf average values were examined remained below the Cf contamination index value and they indicated low contamination $(< 1$). Zn (1.03) , Mn (1.05) , Cr (1.27) and Cu (1.51) showed moderate contamination (1–3). Ni (3.22) showed considerable contamination (3–6). However, very high contamination (>6) was detected in cadmium (35.07) (Table [4\)](#page-9-0).

The mean PLI value of the soils of research area was 1.40, indicating that the soils were polluted by HMs. According to the pollution indices, the pollution of Adıyaman agricultural soils was caused by HMs such as Cd, Ni, Cu, Cr, Mn and Zn. The reason for these HMs may be the pesticides applied by the farmers to their own farmland, chemical fertilizers and the contaminated irrigation waters they used. Kabata-Pendias ([2011\)](#page-15-0), Rutigliano et al. [\(2019](#page-16-20)), Baltas et al. (2020) (2020) , Varol et al. (2020) and Varol et al. [\(2021](#page-17-3)) reported that chemical fertilizers and pesticides applications increased concentrations of HMs in soils. In addition, the use of agricultural waters contaminated with HMs in feld, vineyard and garden irrigation is an indication that these waters are a source of soil pollutants (Ahmad et al., [2016;](#page-13-1) Varol et al., [2021](#page-17-3)).

Ecological risk assessment of HMs

The descriptive statistics of Er and RI are given in Table [3](#page-5-0) and [4](#page-9-0). In the study area, Zn and Cr HMs had the lowest Er values, while Cd and Ni had the highest Er values. However, mean Er values of Cr, Cu, Ni, Pb and Zn were less than 40, indicating that had low ecological risk. But Cd (1052) showed very high potential ecological risk. In this study, Er values ranged from 1.03 to 1052. The fact that the average RI value was 1083 meant that the ecological risk of Adıyaman agricultural soils was very high. Similarly, very high values for RI were also reported for agricultural soils

	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
EF										
Mean	0.51	35.00	0.57	1.29	1.57	1.00	1.08	3.28	0.66	1.10
Minimum	0.23	17.21	0.21	0.45	0.75	1.00	0.39	0.75	0.20	0.60
Maximum	0.83	83.86	1.53	3.48	4.15	1.00	2.86	13.94	1.59	3.38
Standart deviation	0.09	16.08	0.21	0.39	0.43	0.00	0.36	1.80	0.20	0.33
Standart error	$0.00\,$	0.80	0.01	0.02	0.02	0.00	0.02	0.09	0.01	0.02
Median	0.51	26.90	0.52	1.23	1.49	1.00	0.99	2.88	0.64	1.00
I_{geo}										
Mean	-1.68	4.31	-1.59	-0.38	-0.08	-0.69	-0.64	0.86	-1.34	-0.61
Minimum	-3.62	1.92	-4.17	-2.55	-1.89	-2.83	-3.47	-1.58	-3.50	-2.07
Maximum	-0.60	6.09	0.40	1.27	1.11	0.32	0.76	3.08	0.17	0.97
Standart deviation	0.53	0.82	0.72	0.64	0.55	0.52	0.63	0.84	0.57	0.45
Standart error	0.03	0.04	0.04	0.03	0.03	0.03	0.03	0.04	0.03	0.02
Median	-1.64	4.16	-1.57	-0.33	-0.04	-0.66	-0.61	0.87	-1.29	-0.61
Cf										
Mean	0.50	35.07	0.56	1.27	1.51	0.99	1.05	3.22	0.64	1.03
Minimum	0.12	5.67	0.08	0.26	0.40	0.21	0.14	0.50	0.13	0.36
Maximum	0.99	102	1.97	3.61	3.23	1.87	2.55	12.70	1.68	2.95
Standart deviation	0.17	20.95	0.29	0.56	0.54	0.32	0.41	2.05	0.24	0.34
Standart error	0.01	1.04	0.01	0.03	0.03	0.02	0.02	0.10	0.01	0.02
Median	0.48	26.78	0.50	1.19	1.46	0.95	0.98	2.74	0.61	0.98
	Er						PLI	R1		
	Cd	Cr	Cu	Ni	Pb	Zn				
Mean	1052	2.53	7.57	16.11	3.19	1.03	1.40	1083		
Minimum	170	0.51	2.02	2.51	0.66	0.36	0.36	178		
Maximum	3060	7.23	16.16	63.48	8.42	2.95	2.68	3092		
Standart deviation	629	1.11	2.72	10.27	1.22	0.34	0.46	635		
Standart error	31.31	0.06	0.14	0.51	0.06	0.02	0.02	31.63		
Median	803	2.38	7.31	13.68	3.07	0.98	1.35	833		

Table 4 Some descriptive statistics of enrichment factor (EF), pollution factor (Cf), geoaccumulation index (I_{geo}), ecological risk factor (ER), pollution load index (PLI) and ecological risk index (RI) values of HMs in soils

of India (Kumar et al., [2019\)](#page-15-8) and China (Wu et al., [2019\)](#page-17-27). However, high RI values for soils are rarely reported in the literature.

Multivariate analysis of soil HMs

Pearson correlation matrix was calculated to examine the relationships between HMs (Table [5\)](#page-10-0). Very significant positive correlations $(P<0.01)$ were found among all HMs. Positive and highly correlated HMs may have a common source, interdependence and the same behavior (Baltas et al., [2020;](#page-14-8) Dong et al., [2018;](#page-14-3) Pan et al., [2016;](#page-16-21) Varol et al., [2020](#page-17-4)). The results showed that there were high positive correlations $(r > 0.40^{**}; P < 0.01)$ between Al, Co, Cr, Cu, Fe, Mn, Pb and Zn at the 1% level. It shows that these HMs in Adıyaman agricultural soils were originated from similar sources and anthropogenic activities. No heavy metals were found to show a negative relationship. PCA and FA were used with standardized data for a more efective evaluation of HM values in Adıyaman agricultural soils. KMO score (0.76) and Bartlett's test of sphericity $(p < 0.001)$ showed that the data set was appropriate for PCA and FA.

In this study, the program identifed two components (PC1 and PC2) that explained 74.8% of the

Table 5 Relationships between heavy metals themselves

HMs	Al	C _d	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Al										
C _d	$0.335**$									
Co	$0.422**$	$0.370**$								
Cr	$0.499**$	$0.474**$	$0.740**$							
Cu	$0.602**$	$0.644**$	$0.497**$	$0.518**$						
Fe	$0.844**$	$0.639**$	$0.660**$	$0.730**$	$0.751**$					
Mn	$0.597**$	$0.400**$	$0.579**$	$0.446**$	$0.607**$	$0.643**$	1			
Ni	$0.218**$	$0.309**$	$0.744**$	$0.870**$	$0.334**$	$0.498**$	$0.286**$	1		
Pb	$0.688**$	$0.616**$	$0.339**$	$0.435**$	$0.592**$	$0.679**$	$0.595**$	$0.195**$		
Zn	$0.593**$	$0.570**$	$0.321**$	$0.464**$	$0.710**$	$0.651**$	$0.461**$	$0.236**$	$0.778**$	

**Correlation is significant at the level 1% ($P < 0.01$)

Table 6 Varimax rotated component matrix for HMs

Parameter	PC1	PC ₂		
Al	0.789	0.215		
C _d	0.676	0.252		
Co	0.306	0.846		
Cr	0.376	0.863		
Cu	0.794	0.301		
Fe	0.786	0.507		
Mn	0.667	0.329		
Ni		0.952		
Pb	0.889			
Zn	0.849	0.112		
Eigenvalues	5.90	1.58		
% of variance	59.00	15.76		
Cumulative %	59.00	74.76		

Bold values represent strong and moderate loadings Extraction Method: Principal Component Analysis Rotation Method: Varimax with Kaiser Normalization

total variance with an eigenvalue >1 . The first variable component (PC1) was loaded by Al, Cd, Cu, Fe, Mn, Pb and Zn, while the second variable component $(PC2)$ was loaded with Co, Cr and Ni (Table [6](#page-10-1)). All of them had strong positive charges (>0.6) . PC1 represented 59% of the total variance, while PC2 represented 15.8%. Chandrasekaran et al. [\(2015](#page-14-21)) and Baltas et al ([2020\)](#page-14-8) suggested that HMs in PC1 were caused by the degradation of the parent material (lithogenic activities).

In particular, due to the fact that the Al and Pb averages of the Adıyaman lands remained below the

UCC averages, the Zn average was similar to the UCC average, and in addition, the EF, I_{geo} and Cf values of these three HMs were low, pointig that they were loaded entirely as a result of natural activities. Cu and Mn averages were above the UCC averages. However, the low EF, I_{geo} and Cf values of these HMs indicated that they were loaded as a result of natural activities. But for Cd, the situation was diferent. Because both the Cd average in the soils was higher than the UCC average and the EF (35.00), I_{geo} (4.31) and Cf (35.07) values were very high, it shows that the Cd was loaded in a result of anthropogenic activities. In general, basaltic igneous rocks are rich in HMs such as Cd and Cu, while sedimentary rocks containing silt and clay contain large amounts of Cd, Cu, Mn, Pb and Zn (Mishra et al., [2019;](#page-16-13) Muradoglu et al., [2015](#page-16-8)).

In Adıyaman, there is Mount Nemrut, which is an extinct volcano. There are also clay, Cu, Pb, Zn, Mn (MTA, [2022](#page-16-15)) and petroleum reserves (TPAO, [2022\)](#page-17-9) from underground resources. The above-mentioned rocks and underground riches explain the lithogenic sources. Al, Fe and Mn are among the most abundant elements in the earth's crust. Fe was distributed between the two components (PC1 and PC2), this status was indicating also a lithogenic origin although anthropogenic activities were greater in the area studied. Fe was found as a mixed source (lithogenic and anthropogenic source) in Adıyaman agricultural soils. Similar cases have been reported in other studies (Baltas et al., [2020;](#page-14-8) Kelepertzis, [2014\)](#page-15-22). There are apatite (raw phosphate rock) Fe ore deposits in Adıyaman Province (MTA, [2022](#page-16-15)). In addition, ironcontaining microelement fertilizers are frequently

used in agricultural soils. In general, phosphate fertilizers contain all heavy metals found as components in phosphate rock (Dissanayake & Chandrajith, [2009](#page-14-22); Mortvedt, [1996\)](#page-16-22). Co, Cr and Ni in PC2 also showed strong (> 0.7) positive loading (Table [6\)](#page-10-1). The Co average in the soil is lower than the UCC average. Since EF (0.57), I_{geo} (−1.59) and Cf (0.56) values were also low, it is understood that loading of Co is the parent material and pedogenic processes. The average of Cr is higher than the average of UCC. According to the Cf value (1.27), it was understood that there was moderate loading in Cr as a result of anthropogenic activities. The mean value of Ni was higher than the mean value of UCC. Since EF (3.28) , I_{geo} (0.86) and Cf (3.22) values were found to be moderate and signifcant, human-induced loading was observed in Ni also. Agricultural products grown in Adıyaman are generally wheat, corn, barley, cotton and chickpeas (TOB, [2022](#page-17-8)). According to the report prepared by the Provincial Directorate of Environment for Adıyaman, nitrogen, phosphorus and potassium fertilizer consumption in 2019 is 32806, 16,295 and 2986 tons, respectively. In the same report, it was reported that pesticide consumption in agricultural areas was 306 tons (ÇŞB, [2020](#page-14-23)). The loading of these metals is therefore likely related to anthropogenic activities such as irrigation water contaminated with industrial waste, fertilization and pesticides. This topic was supported by PCA and FA analysis results, enrichment factor and correlation results.

Potential child and adult health risk assessment

In this study, non-carcinogenic HQ_{ingestion}, HQ_{inhalation}, HQ_{dermal}, HI, total HI (THI), CHQ, carcinogenic $CR_{insection}$, $CR_{inhalation}$, CR_{dermal} and TCR values were calculated for both children and adults (Table [7](#page-12-0)). In this study, HQingestion values for children were listed as Fe>Co>Cr>Mn>Al>Cd>Ni>Cu>Zn>P b, HQingestion values for adults, $Fe > Co > Cr > Mn$ $> A l > Pb > C d > Ni > Cu > Zn$ for adults. HQ_{inhalation} values for both children and adults were listed as $Mn > A l > C_0 > Cr > Ni > Cd$, while HQ_{dermal} values were listed as Cr>Mn>Cd>Ni>Fe>Co>Al>P $b > Cu > Zn$ for both children and adults (Table [7](#page-12-0)). The cumulative HQ (CHQ) values of the three intake pathways for children followed the $CHQ_{\text{ingestion}}$
(0.7320) > CHQ_{dermal} (0.0884) > $CHQ_{\text{inhalation}}$ (0.0884) > CHQ_{inhalation} (0.0190) sequence, while the CHQ values for adults were $CHQ_{\text{ingestion}}$ $(0.0648) > CHQ_{\text{inhalation}}$ $(0.0190) > CHQ_{\text{dermal}} (0.0169)$ (Table [7\)](#page-12-0). These values were below the risk threshold and were unlikely to have a negative non-carcinogenic efect on health for children and adults exposed to HMs through ingestion, inhalation routes and dermal contact pathways in Adıyaman soils.

HQ, HI and total HI (THI) values of HMs levels of Adıyaman soils for both adults and children were < 1 . This was also suggests that the HMs we studied, which were transmitted to humans through ingestion, inhalation pathways and dermal contact, carry insignifcant non-carcinogenic risks. Similar results were reported by Praveena et al. ([2018\)](#page-16-23), who studied the surface soils of the Klang region in Malaysia. Of the 10 HMs examined in the study, their HI for children was higher than for adults. Likewise, the THI value for children was 8.33 times higher than for adults, indicating that children were more sensitive to non-carcinogenic health risks than adults. Similar results have been reported in previous studies (Baltas et al., [2020](#page-14-8); Deng et al., [2020;](#page-14-7) Rinklebe et al., [2019;](#page-16-12) Shaheen et al., [2020](#page-16-11); Sun et al., [2021](#page-17-28); Varol et al., [2020,](#page-17-4) [2021](#page-17-3)). HI values for children decreased in Fe $>Cr>Co>Mn>Al>Cd>Ni>Cu>Zn>Pb$, while HI values for adults were $Mn > Cr > Fe > Co > Pb >$ $Cd > Ni > Cu > Zn$ (Table [7](#page-12-0)). The oral CHQ values of all HMs accounted for 87.25% and 64.28% of THI for children and adults, respectively. The fndings showed that the negative impact of the oral route on the health of children and adults was greater than the inhalation and dermal contact routes. Lian et al. [\(2019](#page-15-23)), Xiao et al. ([2020\)](#page-17-29) and Deng et al. [\(2020](#page-14-7)) also reported similar results. Cr's carcinogenic risk (CR) values through oral, inhalation pathways and dermal contact and total carcinogenic risk (TCR) values remained within USEPA's acceptable 10^{-4} and 10^{-6} risk limits (Table [7\)](#page-12-0). It has been determined that Cr HM does not currently have carcinogenic risks for 3 receptors in Adıyaman soils. The CRs of Cd, Co, Cr and Ni for the inhalation pathway were also within or below the acceptable risk limits (Table [7](#page-12-0)). These results showed that there were no carcinogenic health risks from exposure to Cd, Co, Cr and Ni for residents in the territory of the study area. The fndings were also in agreement with previous studies (Deng et al., [2020;](#page-14-7) Sun et al., [2021;](#page-17-28) Varol et al., [2020,](#page-17-4) [2021\)](#page-17-3). TCR (total CR) values decreased according to Cr>Co>Ni>Cd order. CCR (cumulative CR)

Table 7 Carcinogenic (CR, CCR, TCR and CTCR) and non-carcinogenic (HQ, CHQ, HI and THI) risks from soil HMs for child and adult residential receptors in Adıyaman

Table 7 Carcinogenic (CR, CCR, TCR and CTCR) and non-carcinogenic (HQ, CHQ, HI and THI) risks from soil HMs for child and adult residential receptors in Adıyaman

values for the three intake pathways followed the order of $CCR_{inhalation} > CCR_{ingestion} > CCR_{dermal} > CCR$ inhalation value was 8.87 and 18.32 times higher than $CCR_{ingestion}$ and CCR_{dermal} values, respectively. The CCRinhalation value accounted for 86.58% of the CTCR (cumulative TCR) value. Cr was the largest contributor to CTCR through oral and dermal contact and was the highest. It contributed 9.78% and 4.74% to TCR, respectively.

Conclusions

HMs such as Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn were measured using inductively coupled plasma-optical emission spectrometry (ICP–OES) in soil samples collected from 403 sampling points of agricultural soils in Adıyaman Province, Türkiye. The average concentrations of Cd, Cr, Cu, Fe, Mn and Ni HMs were higher than the average UCC values, while the average concentrations of Al, Co and Pb HMs were lower than the average UCC values. Ortalama Zn değeri ise UCC'nin ortalama Zn değerine çok yakın bulundu. The pollution index values of Cd such as EF, I_{geo} , Cf and Er were determined more than the pollution index values of other investigated HMs. Therefore, Cd had high environmental and ecological risk. Other HMs had low to moderate environmental and ecological risk. Since the RI $(1083) > 600$, it indicated that there was a "very high degree of ecological risk" in the soils of the study area. PLI was determined as>1 in Adıyaman agricultural soils. Therefore, contamination caused by HMs was detected. Pearson correlation analysis was used to determine the relationships among these HMs and PCA and FA methods were applied to defne the pollution sources. The PCA and FA results used showed that Cd, Cr and Ni from anthropogenic sources, Fe from both lithogenic origins and anthropogenic sources (fertilizers and pesticides) and other HMs came from lithogenic sources. These results showed that concentrations of Cd, Cr, Ni and Fe HMs were high in agricultural soils. Therefore, it is recommended to control the excessive use of chemical fertilizers and pesticides and the use of contaminated irrigation water to prevent soil contamination. In this study, intake from soil was the most important pathways for human exposure to HMs. Three intake pathways for both children and adults CHQ values were lower than the risk threshold, which showed that there were no health risks on the territory of the province of Adıyaman. In addition, the CR, TCR, CCR and CTCR values of Cd, Co, Cr and Ni HMs were below the acceptable risk limit of USEPA of 10^{-4} . These suggest that the carcinogenic health risks from the intake of HMs do not occur for residents. Since this study is designed to represent the agricultural areas of the entire province, it can be used as a model in future studies. In addition, this study results show that the intensive use of chemicals and contaminated irrigation waters in agricultural areas can be used as a model for organizing routine follow-up programs in those areas and monitor soil pollution risks and health risks of residents.

Acknowledgements It was utilized data from the project titled "Determination of plant nutrients and potential toxic element contents of agricultural soils in Kahramanmaraş, Malatya and Adıyaman provinces, creation of the database and mapping" supported by the General Directorate of Agricultural Research and Policies of the Ministry of Agriculture and Forestry of the Republic of Türkiye in this study.

Author contributions CHY wrote and checked the main manuscript. Again all the tables and fgures were prepared and checked by CHY.

Funding The author has not received any funding for publishing this manuscript in a journal.

Declarations

Confict of interest The author declare that there is no fnancial/commercial confict of interest in this paper.

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Environmental Monitoring and Assessment, 189(9), 1–14. <https://doi.org/10.1007/s10661-017-6140-y>

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