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Heavy metal concentrations in the soil near illegal landfills in the vicinity of agricultural areas—artificial neural network approach

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Abstract

Purpose To anticipate the impact of illegal landfills, development of new models should become a part of environmental risk management strategies. One of such approaches includes applications of the artificial neural network (ANN). The main objective of this study was to elucidate the impact of illegal landfilling on the surrounding soil environment and human health, as well as to establish an artificial neural network (ANN) models for predicting the hazards of illegal landfilling as an effective tool in decision-making and environmental risk management.

Methods The identification of heavy metals source in soil was performed by principal component analysis (PCA). To assess the sensitivity of the soil ecosystem to heavy metal concentrations, Soil Quality standards and quantitative indices were used. The possible health effects were valued using the average daily doses (ADDs), hazard quotient (HQ), hazard index (HI), and carcinogenic risk (CR). ANN modeling was used for the prediction of heavy metal concentrations in the soil based on landfill size, municipality size, the number of residents, plant species, soil, and landform types.

Results The average values of the pollution indexes for Cd were in the moderately contaminated and very high contamination categories. The *HQ* values were lower than the safe level. Cr and Pb posed a significant *CR* for adults and children, and Ni for children. The ANN models have exhibited good generalization power and accurately predicted the output parameters with a high value of the coefficient of determination.

Conclusion Concerning heavy metal concentrations, illegal landfills near agricultural soil have a significant impact on the soil ecosystem and people's health. The developed ANN models can be applied generally to anticipate the heavy metal concentrations in soil, according to the before mentioned input parameters, with high accuracy.

Keywords Soil quality · Heavy metals · Illegal landfills · Ecological risks · Health risks

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1 Introduction

In the final life cycle of discarded products, landfilling is one of the most widespread methods (Hamid et al. 2018). Landfills are important for the proper disposal of solid waste; however, they have significant social and environmental impacts. The most demanding environmental worry regarding landfills is the release of methane gas, carbon dioxide, water vapor, nitrogen, hydrogen, and non-methane volatile organic compounds (Vaverková et al. 2019). The landfills destroy natural habitats for wildlife. Further, landfills release numerous pollutants to the environment via landfill leachate (El-Salam and Abu-Zuid 2014; Han et al. 2016a), causing the contamination and/or degradation of the soil/water (Dąbrowska et al. 2018; Koda et al. 2017; Vaverková et al. 2018). The high toxicity can spread throughout the whole surface of the Earth, polluting it. In that way, they become a threat to the safe environment for the future generation (Pawłowski 2011). Especially, illegal landfills have been identified as a serious environmental problem, which can have a major impact on soil quality and health (Wright et al. 2018; Vaverková et al. 2019).

Waste-related problems are linked to several United Nations (UN) Sustainable Development Goals (SDGs): 3rd, ensure good health and well-being; 6th, ensure clean water and sanitation; 8th, to promote decent work and economic growth; 9th, build a sustainable industry, resilient infrastructure, and foster innovation; 11th, make sustainable cities and communities; 12th, ensure responsible consumption and production; 14th, conserve life below water; 15th, to protect, to restore, and to promote life on the land. To realize SDGs, soil quality has to be of high importance. Land Degradation Neutrality sets the ambitious goal of increasing the soil quality resources needed to support ecosystem services and improve food security across the world (Solomun et al. 2018).

Much like education or healthcare, waste management requires a country to have a proper system in place. In Serbia and many similar countries where the solid waste management system is not simply poor, but virtually nonexistent, a large number of sites have become increasingly polluted (Krčmar et al. 2018). Waste is disposed of in an inadequate manner because the implementation of national and European legislation (Directive 2008/98/EC; Council Directive 1999/31/EC) is neither systematic nor sufficient (Krčmar et al. 2018). To achieve Land Degradation Neutrality's ambitious goal, the waste management system in Serbia at the moment must be built from scratch. The first step in controlling illegal landfills as waste management practice involves primarily public education of residents and businesses about the consequences that improperly disposed of materials can have on the environment. An entire waste management system requires decision-making on multiple plains: building sanitary landfills, a waste sorting system in place, establishing a market price of waste management services, etc. Further, the development of new models should become a part of environmental risk management strategies, especially from the aspect to anticipate the impact of illegal landfills. One of such approaches includes applications of the artificial neural network (ANN).

Analysis using neural networks is very useful for large series of data. The obtained data on the concentrations of heavy metals in the soil are linked with other information for the investigated locations. Neural networks clean and reprocess the data, remove extreme values, and approximate missing data. Neural networks first learn based on measured values, validate them, and test different models to which these sets of values correspond. It is important to choose the right network architecture. During training, the network will iteratively adjust its internal parameters (weights and biases) to minimize the difference between its predictions and the actual metal concentrations. After training, the performance of the neural network on the test set is evaluated. Metrics such as coefficient of determination (r^2), reduced chi-square (χ^2), mean bias error (*MBE*), root mean square error (*RMSE*), mean percentage error (*MPE*), a sum of squared errors (*SSE*), and average absolute relative deviation (AARD) can be used to assess the model's accuracy in predicting the concentrations of toxic metals. Neural networks are often considered "black-box" models because they lack explicit interpretability.

According to the literature, several investigations were connected to the applications of the ANN approach in environmental risk assessment and prediction. In the study by Spitz and Lek (1999), environmental impact management involves dealing with complex ecological systems and analyzing nonlinear relationships. Conventional techniques often fall short in addressing these complexities, making it akin to predicting the output of a black box. However, ANNs have emerged as a promising solution for modeling such situations. ANNs have demonstrated their ability to learn intricate relationships between environmental variables and impact assessment, providing valuable predictions that can guide managers in efficiently distributing their actions between prevention and protection measures. In the investigation of Shi and Li (2007), an ANN model was proposed for the eco-environment background value validation and prediction. A three-layer back propagation ANN model was designed to automatically learn the internal relationship using a training set of known eco-environmental attributes across the study area. The paper by Li et al. (2008) aimed to offer a practical evaluation methodology for product environmental impact by exploring existing environmental impact assessment methodologies and proposes an ANN approach to estimate missing data, providing a potential remedy for current tools. In the article by Jahani et al. (2014), the ANN modeling tool was used to predict forest degradation based on ecological and associated factors. The results showed that the multi-layer feed-forward network, specifically the optimized forest degradation model, outperforms other degradation models in this regard. In the study by Xiong et al. (2019), an entropy weight method was employed to create a vulnerability assessment system and a Levenberg–Marquardt back-propagation neural network model for the susceptibility assessment. Based on these assessments, a risk assessment was conducted, revealing that approximately 70% of the slopes were in high-susceptibility areas with a considerable landslide risk.

Since, illegal landfill areas occur most frequently on the peripheries of inhabited areas, on agricultural and forest margins, in ditches, as well as at other places, the current study was focused on determining the impact of illegal landfills on the heavy metal concentrations in soil in the vicinity of agricultural areas. The main objective of this study was to elucidate the impact of illegal landfilling on the surrounding soil environment and human health, as well as to establish ANN models for predicting the hazards of illegal landfilling as an effective tool in decision-making and environmental risk management. In order to rich the objective, the following tasks were carried out: (i) determination of the heavy metal concentrations; (ii) identification of the sources of heavy metals; (iii) assessment of the sensitivity of the soil ecosystem to heavy metal concentrations; (iv) assessment of the impact of illegal landfills on human health; and (v) assessment of the impact of landfill size, municipality size, the number of residents, plant species, soil, and landforms types on the heavy metal concentrations using ANN models.

2 Materials and methods

2.1 Study area, sampling, and analysis

2.1.1 Study area

In this study, Vojvodina was selected for the case study. Vojvodina is the Autonomous Province (AP) in the Republic of Serbia located in the southeastern part of the Pannonian (Carpathian) Basin. It is the northernmost part of Serbia, bordered by Hungary to the north, Croatia to the west, and Romania to the east (Fig. 1).

2.1.2 Sampling

A total of 1640 soil samples were collected from 164 sites of illegal landfills near agricultural margins on the territory of the AP Vojvodina. Sampling was performed on five measuring points at each landfill so that one measuring point was in the center of the landfill, i.e., the place where the waste has been for a long time, and four more measuring points were located on the edges of the landfill. Soil samples were collected at two depths (0–30 cm and 30–60 cm). Approximately 1.5 kg of surface soil was stored in polyethylene bags in a field cooler.

2.1.3 Sample analysis

Each soil sample (0.4 g) was digested in the microwave digester (ETHOS 1, Advanced Microwave Digestion System, MILESTONE, Italy) using 7 ml of concentrated HNO_3 and 2 ml of H_2O_2. The digestion solution was diluted with deionized water and heavy metal concentrations were determined

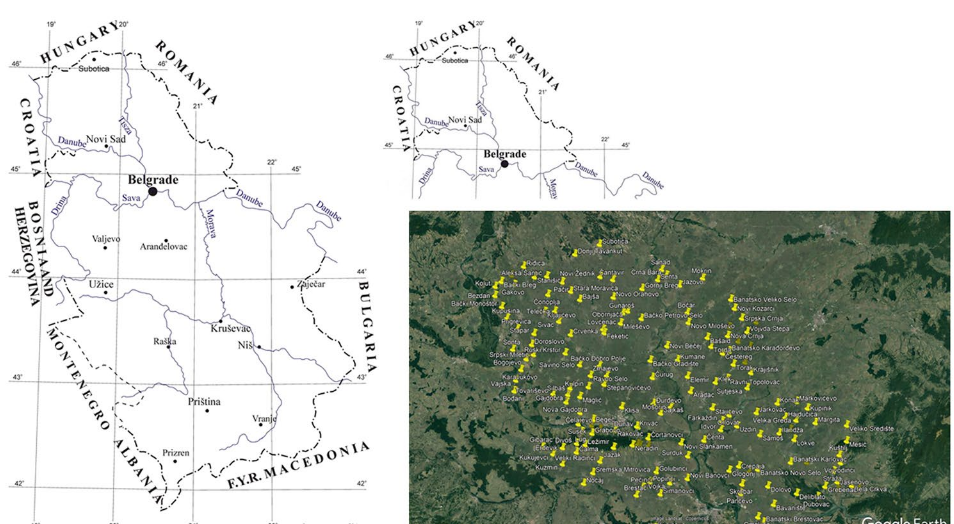


Fig. 1 Study area

using inductively coupled plasma with optical emission spectrometry (ICP-OES system Thermo iCAP 6500 Duo). Total Hg concentration was analyzed using Direct Mercury Analyzer DMA 80 Milestone. The quality assurance and quality control (QA/QC) procedures were conducted using the BCR-141R and BCR-142R reference materials. The procedures used to detect soil heavy metals were relatively accurate, the relative standard deviation was less than 5%, and the recovery percentages were within ± 10%.

3 Results and discussion

3.1 Determination of heavy metal concentrations in the soil

The basic descriptive statistics of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), and mercury (Hg) concentrations (mg kg^{-1}) in the soil near illegal landfills in the vicinity of agricultural areas are presented in Table 1. The mean ± SD (range) concentrations (mg kg^{-1}) for As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg in soil samples were 6.12 ± 7.33 (0.09–140), 1.41 ± 1.00 (0.03–19.9), 32.9 ± 19.7 (0.50–327), 28.1 ± 34.3 (4.22–902), 27.1 ± 17.3 (0.97–329), 16.8 ± 31.1 (1.06–734), 96.4 ± 162 (2.01–5418), and 0.05 ± 0.12 (0.001–1.54), respectively (Table 1).

3.2 Identification of the sources of heavy metals

Principal component analysis (PCA) technique was applied to identify the source of heavy metals (natural or anthropogenic) in the soil near illegal landfills in the vicinity of agricultural areas. The PCA results of heavy metal concentrations calculated by Minitab 17.0 (AppOnFly, Inc., San Francisco, CA, USA) are shown in Fig. 2. The PCA results based on the Varimax rotation (Eq. 1) show that three principal components (PCs) were extracted for the eight heavy metals in soil samples with an accumulated contribution of 61.91%.

The coordinate rotation technique in PCA analysis is commonly used to discover an $m \times m$ orthogonal rotation matrix U = [u_{ij}] applied to the factor matrix. In this transformation, the rows retain the representation of the initial factors, while the columns embody the new factors. Among the various rotation methods, Varimax stands out as the most widely used. It strives to enhance the disparities between the loading factors, all while upholding orthogonality between axes. The central objective of the Varimax method is to maximize the value of V. To achieve this, an iterative algorithm is employed, comprising the subsequent steps. Each of these steps is elucidated using the loading factors in matrix form:

$$V = \frac{1}{k} \cdot \sum_{j=1}^{m} \left[\sum_{i=1}^{k} \left(\frac{b_{ij}^2}{\varphi_i} \right)^2 - \left(\frac{1}{k} \cdot \sum_{i=1}^{k} \frac{b_{ij}^2}{\varphi_i} \right)^2 \right] \tag{1}$$

The rotation of a two-column matrix involves the multiplication by a matrix following this structure (Eq. 2):

$$A = \begin{bmatrix} \cos\theta & -\sin\theta \\ \sin\theta & \cos\theta \end{bmatrix} \tag{2}$$

where θ signifies the rotation angle.

A short explanation of this iterative procedure describes that subsequent to each rotation of this nature, a fresh loading factors matrix is generated, which subsequently becomes the focus of the ensuing substep. Each rotation generates a fresh loading factors matrix, which is then employed in the subsequent substep. Each element is computed as the difference between the squares of the elements within the corresponding row of the two columns under rotation. This iterative process persists until either minimal alterations are observed in the resulting matrix or a predetermined number of iterations have been completed.

The first three PCs have eigenvalues greater than 1, and the scree plot shows that the eigenvalues start to form a straight line after the third PC. The PC1 has large positive associations with As, Cd, Cr, and Ni (Fig. 2). The As concentrations in the soil depend on the origin of the rocks, but their level increases to pollution by human activities. The value of coefficients of variation (CV) for As in soil samples was 119.61%. Cr and Ni are known to be geochemically associated in nature and originate from similar lithogenic parental materials (Guan et al. 2019; Wang et al. 2020). The CV values for Cr and Ni were 59.83% and 63.96%, respectively. Although Cd is mainly derived from anthropogenic sources, the CV value of Cd in this study was 71.37%. The PC2 has large positive associations with Cu, Pb, and Zn (Fig. 2). The CV values for Cu, Pb, and Zn were 122.41%, 185.31%, and 168.90%, respectively. The CV values higher than 100% indicate that the distribution of

Table 1 Descriptive statistics of heavy metal concentrations (mg kg^{-1}) in soil near illegal landfills in the vicinity of agricultural areas

	As	Cd	Cr	Cu	Ni	Pb	Zn	Hg
Mean	6.12	1.41	32.9	28.1	27.1	16.8	96.4	0.05
Median	5.06	1.10	27.6	20.8	23.8	11.7	61.3	0.02
Min.	0.09	0.03	0.50	4.22	0.97	1.06	2.01	0.001
Max.	140	19.9	327	902	329	734	5418	1.54
SD	7.33	1.00	19.7	34.3	17.3	31.1	162	0.12

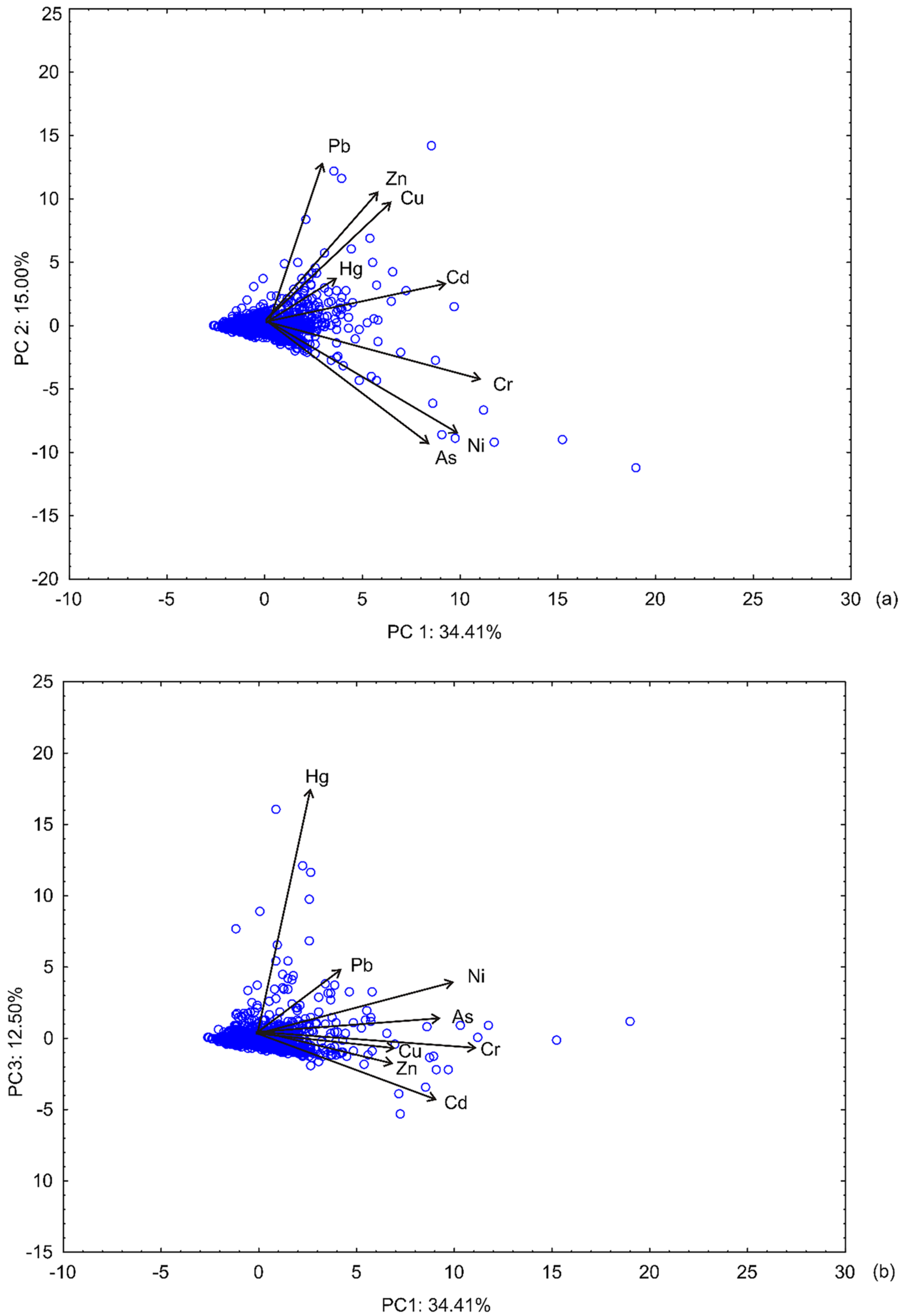


Fig. 2 The PCA scree plot of heavy metals in soil near illegal landfills in the vicinity of agricultural areas

heavy metals is strongly disrupted by human activities (Wang et al. 2020). Human activities can produce a high variation in the concentrations of heavy metals (Liu et al. 2021). Cu, Pb, and Zn are typical "urban" metals. Due to anthropogenic activities, they are emitted in large quantities that are concentrated in urban areas (De Miguel et al. 1998; Liu et al. 2015; Wang et al. 2020). The PC3 has large positive associations with Hg (Fig. 2). Using PCA or other measures, Hg is often classified as an isolated group in source identification (Lv 2019; Wang et al. 2020). Atmospheric deposition has a large contribution to the Hg contents in soil (Guan et al. 2019).

3.3 Assessment of the sensitivity of the soil ecosystem on heavy metal concentrations

To evaluate the sensitivity of the soil ecosystem on heavy metal concentrations near illegal landfills in the vicinity of agricultural areas, Soil Quality standards and quantitative indices were used. The obtained results were compared with "target" values of heavy metals in soil regulated by the legal norms of Serbia ("Off. Gazette of RS" No. 30/2018 and 64/2019), the European Union (EU) (Waseem et al. 2014), and the Food and Agriculture Organization/World Health Organization (FAO/WHO) (Ogundele et 2015; Onyedikachi et al. 2018).

To classify the degree of pollution, different quality indices were applied: geo-accumulation index (I_{geo}) (Müller 1979), pollution index (PI), Nemerow integrated pollution index ($NIPI$) (Nemerow 1974), and potential ecological risk index (RI) (Håkanson 1980). These several methods have different reference contents, initial design purposes, and analysis focuses, and each method has its advantages in one aspect, and some deficiencies in other aspects (Dung et al. 2013).

The I_{geo}, proposed by Müller (1979), was used for assessing the contamination of sediments. Since then, it has been widely applied for assessing the contamination of soils, compared with geochemical background values. The value of I_{geo} can be calculated by Eq. (3) (Müller 1979):

$$I_{geo} = log_2 \frac{C_i}{1.5B_i} \tag{3}$$

where C_i is the measured concentration of i metal in the soil; B_i is the geochemical background concentration of i metal in the continental crust (Taylor 1964). The constant 1.5 allows to analyze natural fluctuations in the concentration of a given metal in the environment and to detect very small anthropogenic influences. There are seven classes based on I_{geo} value: < 0, 0–1, 1–2, 2–3, 3–4, 4–5, ≥ 5, representing uncontaminated, uncontaminated to moderately contaminated, moderately contaminated, moderately to heavily contaminated, heavily contaminated, heavily to extremely contaminated, and extremely contaminated (Yaqin et al. 2008).

To evaluate the contamination level for every single metal and for all studied metals in each soil samples PI and $NIPI$ (Nemerow 1974) were used. The values of PI and $NIPI$ can be calculated by Eqs. (4) and (5) (Han et al. 2016b):

$$PI = \frac{C_i}{S_i} \tag{4}$$

$$NIPI = \sqrt{\frac{PI_{max}^2 + PI_{ave}^2}{2}} \tag{5}$$

where C_i is the concentration of i metal in the soil; S_i is soil quality critical value or standard of i metal. Serbian Soil Quality critical value for As was 29, Cd 0.8, Cr 100, Cu 36, Ni 35, Pb 85, Zn 140, and for Hg was 0.3 ("Off. Gazette of RS" No. 30/2018 and 64/2019). PI_{max} is maximum PI value of all studied metals; PI_{ave} is the average PI value of all studied metals. The PI classifies four grades: < 1, 1–2, 2–3, ≥ 3 representing uncontaminated, uncontaminated to moderately contaminated, moderately to strongly contaminated, and strongly contaminated, and $NIPI$ values are classified as < 0.7, 0.7–1, 1–2, 2–3, ≥ 3, representing no pollution, the warning threshold, low pollution, moderate pollution, and severe pollution (Liu et al. 2021).

Håkanson (1980) introduced RI to assess the degree of ecological risk of metals in soil or sediments. The value of RI can be calculated by Eqs. (6)–(8) (Håkanson 1980):

$$C_f^i = \frac{C_D^i}{C_n^i} \tag{6}$$

$$E_r^i = T_r^i C_f^i \tag{7}$$

$$RI = \sum_{i=1}^{n} E_r^i \tag{8}$$

where C_f^i is the contamination factor; C_D^i is the present concentration of metals; C_n^i is the background concentrations of metal in the continental crust (Taylor 1964); E_r^i is the potential risk of individual metal; T_r^i is the toxic-response factor for a given metal. The toxic-response factor for As was 10, Cd 30, Cr 2, whereas Cu, Ni, Pb was 5, Zn 1, and Hg was 40 (Håkanson 1980). C_f^i classes suggested by Håkanson (1980) are < 1, 1–3, 3–6, > 6 representing low contamination, moderate contamination, considerable contamination, and very high contamination. Hakanson (1980) defined five categories of E_r^i and four categories of RI. Ecological risk levels of E_r^i are $E_r^i < 40$ low risk, $40 \le E_r^i < 80$ moderate risk, $80 \le E_r^i < 160$ considerable risk, $160 \le E_r^i < 320$ high risk, and $320 \le E_r^i$ serious risk. General levels of RI are $RI < 150$ low grade risk, $150 \le RI < 300$ moderate risk, $300 \le RI < 600$ severe risk, and $600 \le RI$ serious risk (Wu et al. 2010).

The mean concentration of As in soil samples (Table 1) did not exceed the limit values according to Serbian Soil Quality (29 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019) and the EU standards (20 mg kg^{-1}) (Waseem et al. 2014). Agricultural practices may be an important source of As, as its contents may be raised in fertilizer, pesticides, manure, and sludge (Kabata-Pendias and Mukherjee 2007). The previous study has shown a linear relationship between the concentration of As in soil and its transfer into the plants (Waseem et al. 2014). The mean value of Cd in soil samples was above the recommended limit set by Serbian Soil Quality standard (0.8 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019), but below the recommended limit set by EU standard (1–3 mg kg^{-1}) (Waseem et al. 2014). The major sources of Cd pollution in soil are atmospheric deposition and P-fertilizers. Up to 90% of Cd pollution remains in the top, 15 cm depth layer of soil (Kabata-Pendias and

Mukherjee 2007). The high concentration of Cd has toxic effects on the beneficial microbes, disturbing their metabolic process, and inhibiting their growth (Ahmad et al. 2021). The mean concentration of Cr in soil samples was found under the limit set by Serbian Soil Quality (100 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019) and the EU standard (100–150 mg kg^{-1}) (Waseem et al. 2014). Sewage treatment plants from industrial and residential sources discharge substantial amounts of Cr (Kabata-Pendias and Mukherjee 2007). Cr contamination considerably affects the biological activities of soil, especially chernozem's biota is highly affected by the toxic effects of Cr (Ahmad et al. 2021). An excess amount of Cu in the soil becomes toxic to some beneficial microorganisms and plants, for the reason that it can inhibit the mineralization of P and N (Ahmad et al. 2021). Various important sources such as sewage sludge, fertilizers, agrochemicals, manures, industrial by-product wastes, and the quality of irrigation waters have contributed to increased Cu levels in agricultural soils (Kabata-Pendias and Mukherjee 2007). In soil samples around the landfill sites, Cu was found in higher concentrations. High Cu concentration could be an indication of the migration of leachate into the surrounding soils (Ahmad et al. 2021). In this study, Cu concentration was found in concentrations lower than the permissible limit defined by Serbian Soil Quality (36 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019), EU (50–140 mg kg^{-1}) (Waseem et al. 2014), and FAO/WHO standards (36–75 mg kg^{-1}) (Onyedikachi et al. 2018). The recommended limit for Ni by Serbian Soil Quality ("Off. Gazette of RS" No. 30/2018 and 64/2019) and WHO standards is 35 mg kg^{-1} (Ogundele et al. 2015). Ni is a serious pollutant that is released from metal processing operations and from the increased combustion of coal and oil. Sewage sludges and P-fertilizers are also important sources of Ni in agricultural soils (Kabata-Pendias and Mukherjee 2007). The mean concentration of Pb in soil samples was under the standard limit set by Serbian Soil Quality (35 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019) and EU standards (50–300 mg kg^{-1}) (Waseem et al. 2014). Between numerous natural and anthropogenic sources of Pb contamination, the impact of industrial emissions and previously used leaded petrol are considered to be of the greatest environmental risk (Kabata-Pendias and Mukherjee 2007). Higher concentrations of Pb present in the soil significantly affect the microbial mass, worsening soil fertility by reducing the rate of nutrient cycling (Ahmad et al. 2021). The mean value of Zn in soil samples was below the recommended limit set by Serbian Soil Quality standard (140 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019), but above the recommended limit set by WHO standard (50 mg kg^{-1}) (Ogundele et al. 2015). It is a very difficult task to estimate Zn pollution in soils. Anthropogenic Zn in forest soils is mostly of industrial origin, whereas in agricultural soils it may originate from numerous other sources (e.g., fertilizers, atmospheric deposition, pesticides, leaching from galvanized materials, sewage sludge, manure, slag, waste, and ashes). The fate of the Zn from several sources differs depending upon its chemical species and their affinity to soil and soil parameters (Kabata-Pendias and Mukherjee 2007). The mean concentration of Hg in soil samples was found under the standard limits set by Serbian Soil Quality (0.3 mg kg^{-1}) ("Off. Gazette of RS" No. 30/2018 and 64/2019) and WHO (0.08 mg kg^{-1}) (Onyedikachi et al. 2018). Hg enters soils from various sources: atmospheric fall out and rainfall, sludge application, disposal of industrial and domestic solid waste products, sewage, Hg-based pesticides, and municipal incinerator ash (Kabata-Pendias and Mukherjee 2007).

Figure 3 shows the I_{geo}, C_f^i, E_r^i, and *PI* values for heavy metals in soil samples near illegal landfills in the vicinity of agricultural areas. The I_{geo} values for As, Cd, Pb, and Zn in soil samples were in the uncontaminated to extremely contaminated category; for Cu and Hg in the uncontaminated to heavily contaminated; and for Cr and Ni in the uncontaminated to a moderately contaminated category according to the defined I_{geo} classes (Yaqin et al. 2008). The average I_{geo} values for Cr, Cu, Ni, Pb, Zn, and Hg were in the uncontaminated category; for As in the uncontaminated to moderately contaminated; and for Cd in the moderately contaminated category.

Values of C_f^i for Cr and Ni in soil samples were in the low contamination to considerable contamination category; and for As, Cd, Cu, Pb, Zn, and Hg in the low contamination to very high contamination category according to the defined C_f^i classes (Håkanson 1980). The average C_f^i values for Cr, Cu, Ni, and Hg were in the uncontaminated category; for Pb and Zn in the moderate contamination; for As in considerable contamination; and for Cd in the very high contamination category. The average E_r^i values for As, Cr, Cu, and Hg were less than 40, which indicated that these heavy metals posed a low potential ecological risk. In contrast, the mean E_r^i(Zn) and E_r^i(Pb) values suggest a considerable, and E_r^i(Ni) and E_r^i(Cd) very high potential ecological risk level according to the defined E_r^i classes (Håkanson 1980). The average *PI* values for As, Cr, Pb, and Hg metal showed no pollution; for Cu, Ni, and Zn the warning threshold; and for Cd high pollution status (Liu et al. 2021).

To quantify the sensitivity of the study soil ecosystem to the heavy metal concentrations, *RI* and *NIPI* were also calculated. The average *RI* value is 564, which showed considerable potential ecological risk according to the defined *RI* classes (Håkanson 1980). The average *NIPI* value was 10.6, indicating severe pollution soil status, ranging from 2.33 to 27.4. It should be noted that Cd, Cr, and Zn contributed 74% of the total *NIPI* value.

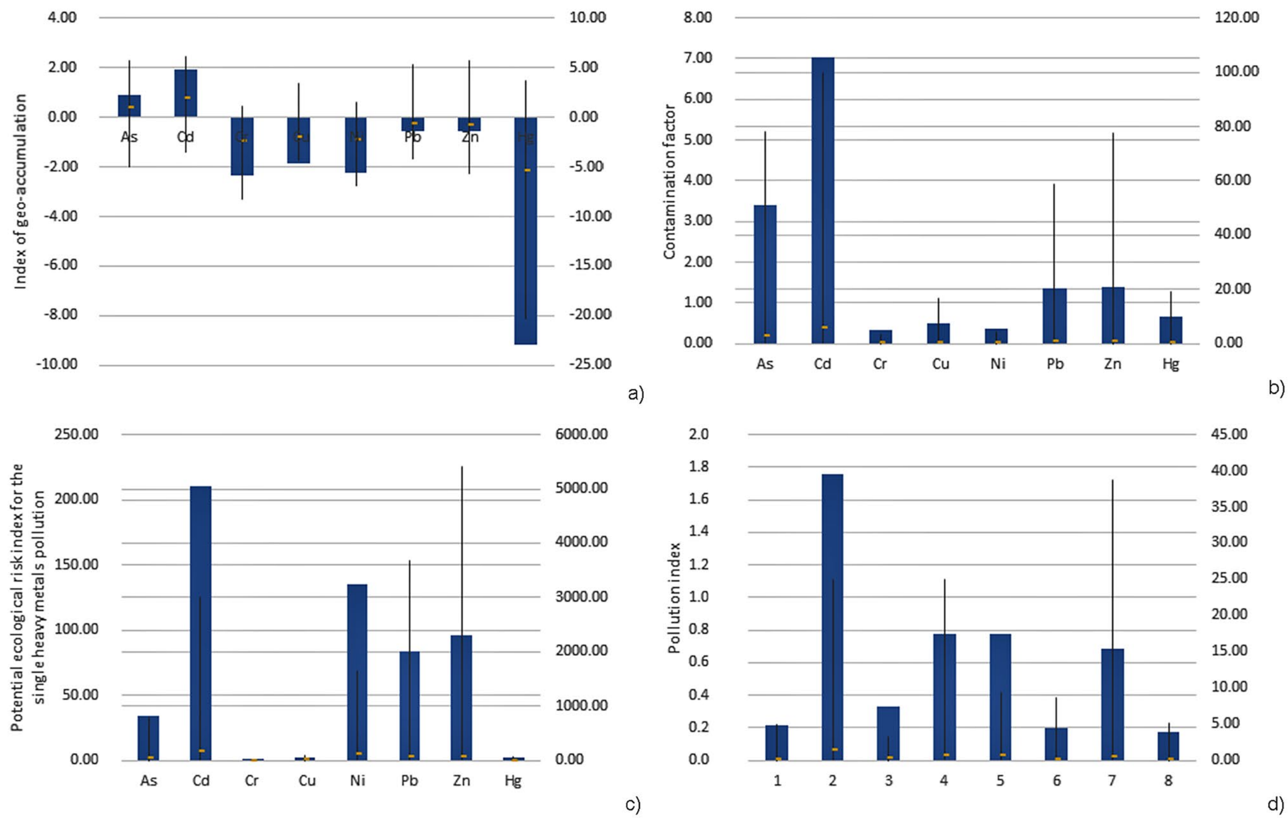


Fig. 3 The I_{geo}, C_f^i, E_r^i, and PI values for heavy metals in soil near illegal landfills in the vicinity of agricultural areas. Vertical lines at the top and bottom of the box correspond to the minimum and maximum values, and the pentagram in the box plots the average value

3.4 Assessment of the impact of illegal landfills on human health

To quantify both carcinogenic and non-carcinogenic risks of heavy metal concentrations in the soil near illegal landfills in the vicinity of agricultural areas to humans via three exposure pathways, ingestion, dermal contact, and inhalation, a health risk assessment was used. The possible health effects using the average daily doses ($ADDs$) (mg kg^{-1} day)$^{-1}$) through inhalation ($ADDinh$), ingestion (ADD_{ing}), and dermal contact ($ADDderm$), for children and adults were estimated according to the Exposure Factors Handbook (U.S. Environmental Protection Agency 1989, 1997, 2001). The values of ADD_{ing}, $ADDderm$, and $ADDinh$ can be calculated by Eqs. (9)–(11):

$$ADD_{ing} = \frac{C_i \times IngR \times EF \times ED}{BW \times AT \times 10^6} \tag{9}$$

$$ADDderm = \frac{C_i \times SA \times AF \times ABS \times EF \times ED}{BW \times AT \times 10^6} \tag{10}$$

$$ADDinh = \frac{C_i \times APM \times InhR \times EF \times ED}{BW \times AT \times 10^6} \tag{11}$$

In Table 2 are given the symbol, description, and values of each parameter.

Non-carcinogenic hazards are typically characterized by the hazard quotient (HQ). The HQ can be calculated by Eq. (12) (USEPA 1989):

$$HQ = \frac{ADD}{RfD} \tag{12}$$

where RfD is the chronic reference dose for the chemical (mg kg^{-1} day^{-1}). If $HQ < 1$, a population is located in a safe area, whereas potential non-carcinogenic effects would occur in case $HQ > 1$ (Wang et al. 2022).

The potential non-carcinogenic effect was evaluated based on a hazard index (HI) (USEPA 1989), and carcinogenic risk (CR) was estimated by calculating the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen (USEPA 1989). The values of HI and CR can be calculated by Eqs. (13) and (14):

Table 2 Input parameters for the characterization of the ADD values

Parameter	Unit	Description	Value		Reference
			Adults	Children	
IngR	mg day^{-1}	Ingestion rate per unit time	100	200	USEPA (2001)
EF	days $year^{-1}$	Exposure frequency	350	350	Liu et al. (2018)
ED	years	Exposure duration	24	6	USEPA (2001)
BW	kg	Body weight	56.8	15.9	Liu et al. (2018)
AT	days	Average time-non cancer risk	ED × 365	ED × 365	USEPA (1989)
InhR	m^3 day^{-1}	Inhalation rate of soil	20	20	USEPA (1997)
SA	cm^2	Exposure skin area	5700	2800	USEPA (2002)
AF	mg $(cm^2\ day)^{-1}$	Skin adherence factor	0.2	0.2	USEPA (2002)
ABS	Unitless	Dermal absorption factor	0.001	0.001	USEPA (2002)
APM	mg $(m^3)^{-1}$	Ambient particulate matter	0.0651	0.0651	Zhou et al. (2016)

$$HI = \sum_{i=1}^{n} HQ_i = \sum_{i=1}^{n} \frac{ADD_i}{RfD_i} \tag{13}$$

$$CR = ADD \times SF \tag{14}$$

where SF is the carcinogenicity slope factor (kg day^{-1} mg^{-1}). If HI value is less than 1, the population will not experience significant adverse health effects; otherwise, adverse health effects may occur (Liu et al. 2021). CR is defined as $< 1 \times 10^{-6}$ representing no significant effect, between 1×10^{-4} and 1×10^{-6} for a generally acceptable range, and $> 1 \times 10^{-4}$ for an unacceptable risk (Liu et al. 2021).

Table 3 lists the average values of HQ, HI, and CR for ingestion, dermal, and inhalation exposure for As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg in the soil near illegal landfills in the vicinity of agricultural areas. The average HQ values were lower than the safe level, which indicated little noncarcinogenic risk from these metals. The ingestion is the major pathway of human exposure to heavy metals, which agreed with previous reports (Xiao et al. 2015; Liu et al. 2021). The average HI values of heavy metals for adults and children decreased in the following order: As > Pb > Cr > Cd > Ni > Cu > Zn > Hg. The HI values were less than 1 for all metals, suggesting noncarcinogenic risk in the study areas.

The CR values for the heavy metals decreased in the following order: Pb > Cr > Ni > As > Cd. The average CR value of Cd in the studied soils was less than 10^{-6}, indicating no considerable carcinogenic risk for adults and children. Acceptable carcinogenic risk level suggest As for adults and children, and Ni for adults since the average CR values were in the range of 10^{-6}, and 10^{-5}, respectively. However, Cr and Pb posed a significant carcinogenic risk for adults and children, and Ni for children as CR values were more than 10^{-4}. According to HI and CR values, the health risks for children were higher than those for adults (Liu et al. 2021). The biologically effective concentrations of Cr, Pb, and Ni were probably lower than the total environmental concentration. Health risk assessments could overestimate the risk of heavy metals in soils, not considering bioaccessibility (Luo et al. 2012).

Soil pollution by heavy metals is still not adequately addressed and continues to pose a threat to human health (Niu et al. 2013). Toxicity and transformation abilities are related to the chemical speciation of heavy metals (Yu et al. 2021). Furthermore, the chemical speciation of heavy metals is related to soil physicochemical properties such as pH value, cation exchange capacity, redox potential, and microbial and enzyme activities (Chia et al. 2022). However,

Table 3 Health risk of heavy metals in soil near illegal landfills in the vicinity of agricultural areas

	HQing		HQder		HQinh		HI		CR	
	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children
As	3.4E−02	2.4E−01	9.5E−04	1.6E−03	3.14E−2	1.12E−1	6.6E−02	3.5E−01	6.9E−06	4.9E−05
Cd	2.3E−03	1.7E−02	1.0E−03	2.8−03	1.08E−2	5.78E−2	1.4E−02	7.7E−02	4.9E−09	2.6E−08
Cr	1.8E−02	1.3E−01	2.1E−04	3.7E−04			1.8−02	1.3E−01	1.1E−04	8.1E−04
Cu	1.1E−03	8.4E−03	4.5E−05	7.8E−05			1.2E−03	8.5E−03		
Ni	2.3E−03	1.6E−02	9.6E−05	1.6E−04			2.3E−03	1.6E−02	2.7E−05	1.9E−04
Pb	2.0E−02	1.4E−01	6.1E−04	1.0E−03			2.0E−02	1.4E−01	3.3E−03	2.4E−02
Zn	5.4E−04	3.8E−03	3.0E−05	5.4E−05			5.7E−04	3.9E−03		
Hg	1.7E−04	1.2E−03	2.7E−05	4.7E−05			1.7E−04	1.0E−03		

various pollutants that enter the soil can affect its condition, affecting the properties and deterioration of the soil environment (Wang et al. 2022). Among new other entities, microplastic pollution crosses planetary boundaries (Persson et al. 2022). Awareness has grown about the need to research microplastics, primarily their fate, transport, distribution, chemical and physical properties (Domercq et al. 2022), impact on living organisms and ecosystems (Tekman et al. 2020), as well as its integration in environmental impact assessment systems (Woods et al. 2021). Considering the fact that microplastic is a solid pollutant very similar to soil particles, it occupies soil space affecting its environment (Zhang et al. 2019). Several researchers investigated the effect of microplastics on heavy metals in soil and found that microplastics can affect the physicochemical properties of soil (e.g., pH, water holding capacity, organic matter (OM), and bulk density) (Zhou et al. 2020; Dong et al. 2021; Medyńska-Juraszek and Jadhav 2022); the abundance, diversity, and activity of microorganisms (Gao et al. 2021); and plant growth by altering the contents of available C, N, and P (Ren et al. 2020). Unexpectedly, Choi et al. (2021) found the largest amount of microplastics in upland soil (3440 pieces kg^{-1}) with the mean abundance of microplastics in agricultural soil being 664 pieces kg^{-1}. The highest abundance was at orchard sites, followed by the upland, greenhouse, and then paddy field sites (Choi et al. 2021).

3.5 Assessment of the impact of landfill size, municipality size, the number of residents, plant species, soil, and landforms types on the heavy metal concentrations in the soil

A multi-layer perceptron (MLP) structure, including three layers (input, hidden, and output), was used for modeling eight ANN models for the prediction of As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg concentrations in the soil near illegal landfills in the vicinity of agricultural areas based on landfill size, municipality size, the number of residents, plant species, soil, and landforms types. According to the known references, the ANN models were confirmed as completely fitted to evaluating nonlinear functions (Ćurčić et al. 2022; Ruškić et al. 2022). Input data were continuously introduced to the network; however, before calculation, the input and output database was normalized according to the min–max formula to enhance the behavior of the ANN models (Rajković et al. 2022). For solving unconstrained nonlinear equations throughout the ANN modeling, the Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm was used as an iterative method.

The experimental database for ANN was randomly divided into training, cross-validation, and testing data (with 70%, 15%, and 15% of experimental data, respectively). It was assumed that successful training was achieved when learning and cross-validation curves approached 0. The coefficients associated with the hidden layer, comprising weights and biases, were partitioned into matrices W_1 and B_1. In addition, the coefficients linked to the output layer were amalgamated into matrices W_2 and B_2 (Eq. 15) (Vojnov et al. 2022):

$$Y = f_1(W_2 \cdot f_2(W_1 \cdot X + B_1) + B_2) \tag{15}$$

In matrix representation, Y represents the matrix of output variables. The transfer functions for the hidden and output layers are denoted as f_1 and f_2, respectively, while X signifies the matrix of input variables. The elements of matrices were estimated in the course of calculation in the ANN models development. Their values were adjusted by applying optimization techniques to minimize the error between the networks and experimental outputs (Pezo et al. 2022) according to the sum of squares (*SOS*).

The obtained ANN models exhibited a good generalization power for the testing data and could accurately predict the output parameters of the soil samples for the observed

Table 4 ANN models summary (performance and errors)

		As	Cd	Cr	Cu	Ni	Pb	Zn	Hg
Net name		MLP 42-10-1	MLP 42-8-1	MLP 42-11-1	MLP 42-8-1	MLP 42-10-1	MLP 42-5-1	MLP 42-11-1	MLP 42-11-1
r^2	**Train**	0.93	0.92	0.89	0.89	0.93	0.92	0.91	0.89
	Test	0.94	0.95	0.96	0.93	0.97	0.94	0.95	0.96
	Verification	0.93	0.94	0.94	0.93	0.95	0.94	0.95	0.95
Error	**Train**	2.99	0.07	42.5	225	16.5	249	1013	0.001
	Test	11.2	0.26	70.9	354	20.5	104	20,334	0.001
	Verification	16.3	0.11	39.7	110	106	125	896	0.002
		BFGS	BFGS	BFGS	BFGS	BFGS	BFGS	BFGS	BFGS
	Train. algor.	10,000	4032	10,000	10,000	10,000	10,000	10,000	10,000
	Error funct.	SOS	SOS	SOS	SOS	SOS	SOS	SOS	SOS
	Hidden activation	Tanh	Tanh	Tanh	Tanh	Tanh	Tanh	Tanh	Tanh
	Output activation	Logistic	Tanh	Identity	Exp	Exp	Ident	Tanh	Tanh

input parameters. According to the ANN performance, the optimal number of neurons in the hidden layer for the As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg content was 10 (network MLP 42-10-1), 8 (network MLP 42-8-1), 11 (network MLP 42-11-1), 8 (network MLP 42-8-1), 10 (network MLP 42-10-1), 5 (network MLP 42-5-1), 11 (network MLP 42-11-1), and 11 (network MLP 42-11-1), respectively. With a focus on reaching a high value of the coefficient of determination r^2 (0.926; 0.917; 0.897; 0.886; 0.930; 0.915; 0.905, and 0.892 for ANN throughout the training period) and lower values of *SOS* (Table 4). Figure 4 illustrates the experimentally evaluated and ANN model predicted values, suggesting that the ANN models correctly predicted experimental variables. Also, *SOS* achieved by the ANN models is of the exact order of magnitude as experimental errors. The predicted values approached the select values regarding the ANN models' r^2 values.

In the study by Mosaffaei et al. (2020), a predictive model was developed, using ANN to assess soil and plant degradation. Soil sampling was conducted at four depths, and the

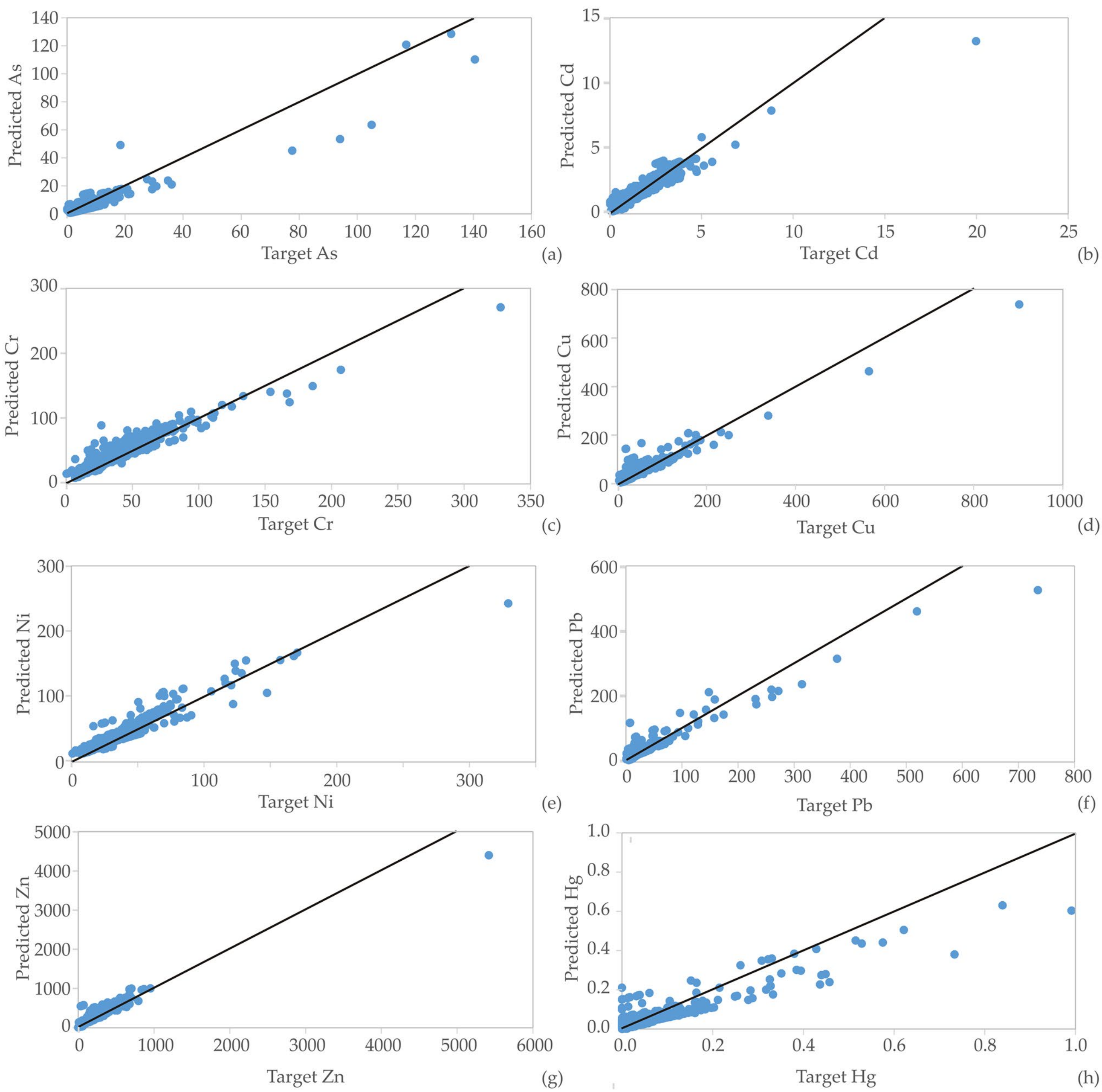


Fig. 4 Comparison between experimentally obtained and ANN model predicted values of (**a**) As, (**b**) Cd, (**c**) Cr, (**d**) Cu, (**e**) Ni, (**f**) Pb, (**g**) Zn, and (**h**) Hg concentrations

soil texture was examined using the hydrometer method. Data from 600 vegetation sample plots and 480 soil profiles, along with physical properties of soil and various human and ecological factors, were used in the model. Among the proposed models, the highest r^2 values for soil texture and biodiversity index were found for clay (0.484), sand (0.320), silt (0.349), and Margalef index (0.292).

Similar approach employing ANN model was demonstrated in the study by Adeleke et al. (2012), in which the best combinations of network architecture, training algorithm, and activation functions for accurately predicting physical waste fractions based on meteorological parameters using ANN were investigated. The optimal architectures for predicting organic, paper, plastics, and textile waste materials in soil achieved r^2 values of 0.839, 0.743, 0.696, and 0.682 during the testing phase. In the study by Jianying et al. (2021), the optimized back propagation network model outperformed the single back propagation neural network in grape sustainable supply chain risk assessment. It showed higher accuracy, fewer evaluation errors, and a larger r^2 reaching 0.932.

3.6 The accuracy of the models and the residual analysis

The computational validation of the created nonlinear models was studied by applying standard statistical tests, including the coefficient of determination (r^2), reduced chi-square (χ^2), mean bias error (*MBE*), root mean square error (*RMSE*), mean percentage error (*MPE*), a sum of squared errors (*SSE*), and average absolute relative deviation (AARD). The following Eqs. (16)–(21) describe these statistical test evaluations (Voća et al. 2022):

$$\chi^2 = \frac{\sum_{i=1}^{N} (x_{\exp,i} - x_{pre,i})^2}{N - n} \tag{16}$$

$$RMSE = \left[\frac{1}{N} \cdot \sum_{i=1}^{N} (x_{pre,i} - x_{\exp,i})^2 \right]^{1/2} \tag{17}$$

Fig. 5 The relative importance of the variables on the (**a**) As, (**b**) Cd, (**c**) Cr, (**d**) Cu, (**e**) Ni, (**f**) Pb, (**g**) Zn, and (**h**) Hg concentrations, determined using Yoon's interpretation method ▸

$$MBE = \frac{1}{N} \cdot \sum_{i=1}^{N} (x_{pre,i} - x_{\exp,i}) \tag{18}$$

$$MPE = \frac{100}{N} \cdot \sum_{i=1}^{N} \left(\frac{\left| x_{pre,i} - x_{\exp,i} \right|}{x_{\exp,i}} \right) \tag{19}$$

$$SSE = \sum_{i=1}^{N} (x_{pre,i} - x_{\exp,i})^2 \tag{20}$$

$$AARD = \frac{1}{N} \cdot \sum_{i=1}^{N} \left| \frac{x_{\exp,i} - x_{pre,i}}{x_{\exp,i}} \right| \tag{21}$$

where $x_{exp,i}$ is experimental value; $x_{pre,i}$ is the model predicted value; N and n are the number of observations and constants, accordingly.

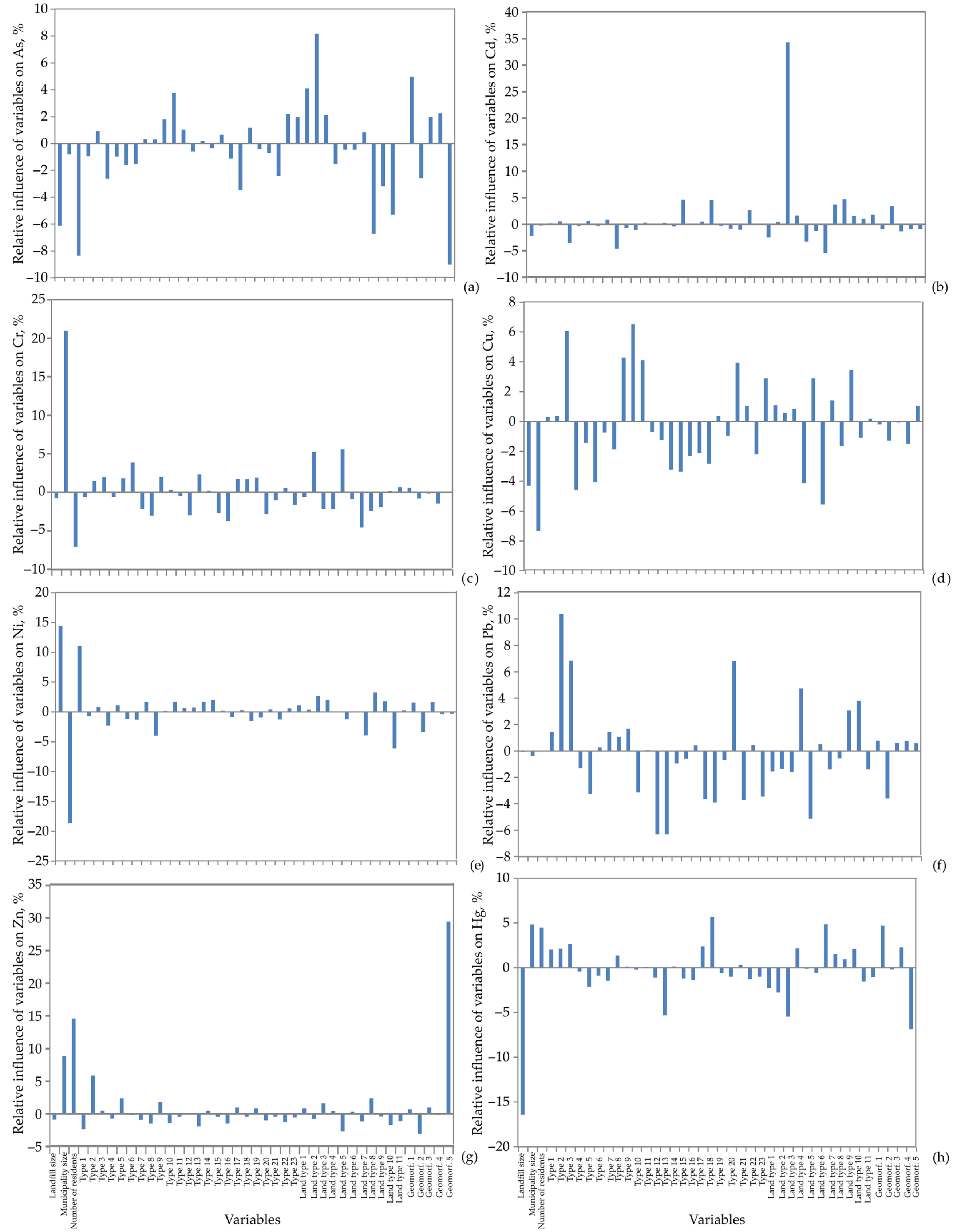
The results indicate that the ANN models had a minor lack of fit tests, which suggests that the models sufficiently predicted the values of the investigated parameters (Table 5).

3.7 Global sensitivity analysis—Yoon's interpretation method

Yoon's interpretation method was used to define the relative impact of monitored input parameters (landfill size, municipality size, the number of residents, plant species, soil, and landforms types) on the As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg concentrations in the soil near illegal landfills in the vicinity of agricultural areas. This method was used according to the acquired weight coefficients of the developed ANN models. StatSoft Statistica, ver. 10.0, Palo Alto, CA, USA, was used to calculate ANN models. The following Eq. (22) was employed for Yoon's global sensitivity method calculation:

Table 5 The "goodness-of-fit" tests for the developed ANN models

	X^2	RMSE	MBE	MPE	SSE	AARD	r^2	Skew	Kurt	Mean	StDev	Var
As	7.32	2.71	0.37	35.01	$1.2 \cdot 10^4$	$2.4 \cdot 10^3$	0.88	5.59	98.9	0.37	2.68	7.41
Cd	0.12	0.35	0.00	24.9	$2.0 \cdot 10^2$	$3.2 \cdot 10^2$	0.90	7.26	168	0.00	0.35	0.12
Cr	47.8	6.92	0.00	14.9	$7.8 \cdot 10^4$	$8.4 \cdot 10^3$	0.90	3.73	47.2	0.00	6.92	49.0
Cu	228	15.1	0.91	25.3	$3.7 \cdot 10^5$	$1.7 \cdot 10^4$	0.88	10.0	199	0.91	15.1	240
Ni	35.6	5.97	0.03	14.3	$5.8 \cdot 10^4$	$9.4 \cdot 10^3$	0.90	7.31	142	0.03	5.97	36.2
Pb	195	13.9	0.00	33.1	$3.2 \cdot 10^5$	$7.2 \cdot 10^3$	0.90	12.2	264	0.00	13.9	194
Zn	5002	70.7	0.00	24.1	$8.2 \cdot 10^6$	$4.4 \cdot 10^4$	0.89	22.2	701	0.00	70.7	5303
Hg	0.00	0.04	0.00	75.1	$2.1 \cdot 10^0$	$3.7 \cdot 10^1$	0.89	6.91	91.9	0.00	0.04	0.00


Relative influence of variables on As, %
(a)
Relative influence of variables on Cd, %
(b)
Relative influence of variables on Cr, %
(c)
Relative influence of variables on Cu, %
(d)
Relative influence of variables on Ni, %
(e)
Relative influence of variables on Pb, %
(f)
Relative influence of variables on Zn, %
(g)
Relative influence of variables on Hg, %
(h)
Landfill size
Municipality size
Number of residents
Type 1
Type 2
Type 3
Type 4
Type 5
Type 6
Type 7
Type 8
Type 9
Type 10
Type 11
Type 12
Type 13
Type 14
Type 15
Type 16
Type 17
Type 18
Type 19
Type 20
Type 21
Type 22
Type 23
Land type 1
Land type 2
Land type 3
Land type 4
Land type 5
Land type 6
Land type 7
Land type 8
Land type 9
Land type 10
Land type 11
Geomorf. 1
Geomorf. 2
Geomorf. 3
Geomorf. 4
Geomorf. 5
Variables

$$RI_{ij}(\%) = \frac{\sum_{k=0}^{n} (w_{ik} \cdot w_{kj})}{\sum_{i=0}^{m} \left| \sum_{k=0}^{n} (w_{ik} \cdot w_{kj}) \right|} \cdot 100\% \tag{22}$$

where w denotes the weighting factor in the ANN model; i input variable; j output variable; k hidden neuron; n number of hidden neurons; and m number of inputs.

According to Fig. 5, the concentration of Hg in the soil was positively influenced by the size of the municipality and the number of inhabitants, while the size of the landfill and the remoteness of the mountainous area had a negative influence. The size of the municipality had a positive effect on the concentration of Cr. Cu concentration was positively influenced by the proximity of the urban zone, and negatively by the size of the municipality. The Ni concentration was positively influenced by the number of inhabitants and the size of the landfill, and negatively by the size of the municipality. The proximity of the urban area and the proximity of the landfill had a positive influence on the concentration of Pb. As and Cd concentrations were positively influenced by the type of soil and higher concentrations were found on the fluvic cambisol type, which indicates a potential natural source of this element.

Previous research has shown that the type of landfill is one of the most important factors affecting the heavy metal concentrations in soil (Wang et al. 2022). In landfill soil, Cr was found to be the dominant heavy metal because it is widely used in manufacturing (Liu et al. 2013). Ni is a well-known element that is found in many hazardous waste sites (Haque 2016). A variety of Hg-containing products, such as batteries, fluorescent lamps, and thermometers, are discarded as household waste, thus can contribute significantly to Hg emissions (Cheng and Hu 2012). Landfill age is another factor that influences pollutant concentrations (Wang et al. 2022). The element migration in the landfill soil is also affected by rainfall (Wang et al. 2022). In addition, the element background value should be taken into account since their distribution in different soil types clearly reflects the parent material distribution affected often by the occurrence of geochemical anomalies with anthropogenic inputs (Kubier et al. 2019; Kobza 2021). Especially, in mountain area parent rock is the most important factor affecting the heavy metal accumulation in soil (Ciarkowska and Miechówka 2022). The sensitivity analysis in the study Adeleke et al. (2012) revealed that variables such as distance from the road, slope, and slope direction were the most influential factors in predicting soil and plant degradation. Similarly, distance from the road, soil moisture content, and slope direction were identified as significant variables in the plant degradation model.

4 Conclusion

Based on the obtained results, it can be concluded that illegal landfills in the vicinity of agricultural areas have a significant impact on the soil ecosystem and people's health. The average I_{geo} and C_f^i values for Cd were in the moderately contaminated and very high contamination categories, and *PI* values for Cd show high pollution status. The mean E_r^i(Ni) and E_r^i(Cd) values suggested a very high potential ecological risk level. According to the defined *RI* classes, the average *RI* showed considerable potential ecological risk. It should be noted that Cd, Cr, and Zn contributed 74% of the total *NIPI* value. The *HQ* values were lower than the safe level. However, Cr and Pb posed a significant carcinogenic risk for adults and children, and Ni for children. The obtained ANN models exhibited a good generalization power for the testing data such as landfill size, municipality size, the number of residents, plant species, soil, and landforms types and could accurately predict the output parameters of the soil samples with a high value of the coefficient of determination r^2 (0.926, 0.917, 0.897, 0.886, 0.930, 0.915, 0.905, and 0.892 for As, Cd, Cr, Cu, Ni, Pb, Zn, and Hg content, respectively). Since developed ANN models exhibited a good generalization, they should become a part of environmental risk management strategies, especially from the aspect to anticipate the impact of the illegal landfills in the vicinity of agricultural areas on heavy metal concentrations in soil, according to the before mentioned input parameters.

Author contribution Snežana Štrbac: investigation, data curation, methodology, writing—original draft, and visualization; Nataša Stojić: investigation, data curation, and methodology; Biljana Lončar: data curation, writing—original draft, and visualization; Lato Pezo: data curation and visualization; Ljiljana Ćurčić: data curation and visualization; Dunja Prokić: investigation and visualization; Mira Pucarević: investigation, methodology, and supervision.

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Availability of data Data will be made available on request.

Declarations

Competing interests The authors declare no competing interests.

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