The Effect of Amendment Addition Drill Cuttings On Heavy Metals Accumulation In Soils And Plants: Experimental Study And Artificial Network Simulation

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Research

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Abstract

A greenhouse experiment was carried out to evaluate the influence of drill cuttings addition on the accumulation of heavy metals in soil, in plant biomass (Trifolium pretense L.) cultivated on soils with the addition of this type of waste. The transfer and transformation of heavy metals in the soil with drill cuttings—Trifolium pretense L were discussed.

Drilling waste in the amount of 2.5%, 5%, 10% and 15% of dry weight were added to acidic soil. The concentrations of heavy metals in the soil and plant materials were determined by an inductively coupled plasma mass spectrometry method. Results indicated that drilling wastes addition had a positive influence on the growth of Trifolium pretense L. However, the concentrations of heavy metals increased in the prepared mixtures along with the dose of drilling wastes. The drilling wastes addition also changed the metal accumulation capacity in plant parts. Nevertheless, the concentrations of heavy metals in soils and above-ground parts of plants did not exceed the permissible values in respective legal standards. The values of the heavy metals bioconcentration coefficient in Trifolium pretense L at the highest dose of drill cuttings were as follows: in the above-ground parts Cd>Cu>Ni>Cr>Pb>Zn, in roots Cd>Ni>Cr>Zn>Pb>Cu.

An artificial neural network model was developed in order to predict the concentration of heavy metals in the plants cultivated on the soils polluted with drill cuttings. The input (drill cuttings dose, pH, organic matter content) and the output data (concentration of heavy metals in the shoot cover) were simulated using an artificial neural network program. The results of this study indicate that an artificial neural network trained for experimental measurements can be successfully employed to rapidly predict the heavy metal content in clover. The artificial neural network achieved coefficients of correlation over 90%.

Introduction

Drill cuttings constitute the spoils obtained in the course of drilling, which is extracted onto the surface along with spent drilling fluid. Their composition and amount depend on numerous factors, such as: depth and structure of the well, type of drilled rocks, type of employed drill cuttings and performed actions, as well as the properties of formation water (Abbe et al. 2009). Therefore, the content of chemical compounds in these wastes may vary significantly. When considering their impact on the natural environment, the main factors taken into account include the content of heavy metals, inorganic salts, oil derivatives, radionuclides, and colloids. Variability of drilling waste compositions hinders assessing their effect on the environment as well as selecting an optimal method of their management (Ball et al. 2012)

The physical and chemical properties of drill cuttings, especially pH (~ 10), high buffer capacity, high content: carbonates, organic matter, and calcium indicate that they may be used for neutralizing acidic soils. However, there are a number of limitations. Drill cuttings contain variable amounts of certain toxic trace elements (e.g., Cd, Cr, Pb, Ni, B and Mo) and high soluble salt (Jamrozik et al. 2015), and this property may affect the the application of drill cuttings (Zvomuya et al. 2011).

It seems that the studies on bioavailability and mobility of heavy metals in the soils with drilling wastes addition are important for assessing the impact of this type of wastes on the environment and the
possibility of their addition to soils. As indicated by the studies conducted thus far, the heavy metals contained in drill cuttings occur mainly in the form which is hardly available for living organisms, which is indicated by low percentage of their leaching (Mikos-Szymańska et al. 2018; Cel et al. 2017). In the studies by Zhu et al. (2011) conducted on the drill cuttings containing the residues of oil-based drilling fluid, the highest leachability – amounting to 1.61% – was observed in the case of copper, whereas for cadmium and lead it was lower, reaching 0.51% and 0.20%, respectively (Zhu et al. 2011).

Certain researchers (Stuckman et al. 2016) conducted fractionation studies indicating that heavy metals will not be released in significant amounts, whereas others observed an increase in the available forms of metals and their mobility in a column study (Bates 1988).

The previous studies on the impact of heavy metals from wastes on soil also differ in terms of the type, amount of added wastes, and the infiltration of metals to soils and plants.

McFarland evaluated selective placement burial as an alternative technique for on-site disposal of drilling fluids in arid and semiarid areas. No migration of Ba, Cr, Cu, Ni or Zn from the drilling wastes buried in the top soil was observed, but the concentrations of Cu and Zn were higher in saltbush stems growing on soils (McFarland et al. 1994).

Bauder investigated the content of metals extracted from soils with the addition of aqueous bentonite drilling fluid. He observed that the levels of DTPA extractable Zn (in a plot with the addition of 80 MG/ha of wastes) were approximately 55-fold higher than in control soil (0.38 mg) (Bauder et al., 2005).

Yao and Naeth applied potassium silicate drilling fluid in four doses: 20, 40, 60, and 120 m$^3$/ha on two types of soil (with sand and loam texture). The wastes used in their studies had no significant influence on the increase in metal concentrations in soils. The contents of investigated metals were much lower than those stated in the provincial criteria limits for agricultural and natural land use (Alberta Environment Sustainable Resource Development 2010) (Yao and Naeth 2014)

There are few studies on the bioaccumulation of heavy metals in the plants cultivated on soils with the addition of drilling fluids. In this paper, the content of heavy metals in the soils with drilling fluid addition and clover cultivated on the prepared mixtures.

Moreover, an artificial neural network (ANN) model was devised for predicting the concentration of heavy metals in the biomass of clover cultivated on the soils contaminated with drilling wastes. The input data included: the dose of drilling wastes, organic matter content as well as the pH in the soils with drilling wastes addition.

The artificial neural network models are increasingly often employed for predicting the migration of pollutants in the environment. ANN are widely used, among others, for: predicting the hydraulic properties of soils (Minasny et al. 2004), generating digital maps of soils (Bagheri Bodaghabadi et al. 2015; McBratney et al. 2003) as well as modeling the behavior of trace metals (Jahantab et al. 2020; Kulisz et al. 2020; Hattab et al. 2013; Buszewski and Kowalkowski 2006;) Anagu et al. 2009; Gandhimathi 2012). In such cases, ANNs are trained in order to find the input-output relationships of the model, using iterative calibration process
(training phase). Moreover, the advantage of ANNs is that they do not enforce limitations on inputs and outputs, which enables their easy application for conducting reverse calculations Rafiq et al. 2001; Hambli et al. 2006).

The objectives were to: (1) investigate the effect of amendment addition drill cuttings on heavy metals accumulation in soils and plants, and (2) assess the effect of soil-amendment–plant interaction on the mobility and phytoavailability of these metals in soil. The main research tasks were to (i) estimate the metals concentrations and metals uptake by plants, (ii) assess the translocation properties and accumulation characteristics of these metals, (iii) develop an artificial neural network model.

**Materials And Methods**

2.1. *Examined materials*: The main component used in the experiment was soil, which was taken from an arable field in the vicinity of Lublin, from a depth of 10–20 cm. The soil contained 71% silt, 6% clay and 23% sand. Drilling waste in the form of drill cuttings dewatered in a chamber press was used as the second component of the prepared soil mixtures. The waste was obtained at the site of their management, in a facility located in Southeastern Poland.

2.2. *Pot experiment*: The studies were conducted on four types of mixtures designated as Z0, Z-2.5, -Z5. Z-10, and Z15, which were prepared based on different proportion of soil and drill cuttings. The share of cuttings in the soil mixture ranged from 2.5–15.0 % by weight, based on the air-dried weight (Table 1). The soil without addition of drill cuttings were used as a control sample (Z0). The doses were adjusted to be lower than the maximum amount of wastes permitted for sowing, established in Directive 050: drilling waste management. Alberta Energy Resources Conservation Board, Calgary (Directive 50 2016).
Table 1
Some physical and chemical properties of the soil used in this research (Kujawska and Pawlowska, 2020)

| Type of mixtures          | Drill cuttings | Soil          |
|---------------------------|----------------|---------------|
| pH                        | 9.6 ± 2.31     | 4.16 ± 0.02   |
| Electrical conductivity [mS/m] | 40.1 ± 2.68 | 0.33 ± 0.04   |
| Cation exchange capacity [cmol(+)/kg] | 49.65 ± 1.74 | 8.02 ± 0.12   |
| Organic matter [% of dry weight] | 11.54 ± 0.21 | 1.9 ± 0.1     |
| Na total [%]              | 0.48 ± 0.01    | 0.004 ± 0.001 |
| Ca total [%]              | 7.6 ± 0.9      | 0.11 ± 0.08   |
| Mg total [%]              | 0.4 ± 0.07     | 0.17 ± 0.01   |
| Soil texture              | -              | Clay loam     |
| Soil particles (%)        | -              | Clay slit sand|
| Silty clay sand           | 18 7 75        | 39 36 25      |
| N total [%]               | 0.04 ± 0.001   | 0.06 ± 0.001  |
| C total [%]               | 6.8 ± 0.33     | 1.09 ± 0.01   |
| P mgP₂O₅ 100g/soil       | 0.92 ± 0.03    | 5.79 ± 0.091  |
| K mgK₂O 100g/soil        | 63.1 ± 0.71    | 2.18 ± 0.24   |

The tested materials were placed in plastic pots with a capacity of 350 ml and each pot was sown with 12 clover seeds (Trifolium pratense L). The studies were carried out in three replicates for six weeks. The culture was carried out in accordance with the recommendations given in PN-EN ISO 11269–2: 2013-06. The experiment was run under controlled greenhouse conditions at 60% field water capacity and at 25°C. At the end of the experiment, the plants were harvested and the roots were gently separated from the soil by repeated rinsing. The collected plants were divided into roots and above-ground (shoots) parts. The dry biomass was weighed with an accuracy of 0.01 g.

2.3. Analysis
2.3.1. Soil pH and organic matter percentage in mixtures

The soil pH were measured in 1:5 soil:water suspension in triplicates (1997). In order to calculate the dry weight, the samples were oven-dried for four hours at 105°C to constant weight. The percentage of organic
matter (OM, %) was determined according to PN–EN 15935:2013-02 by the combustion of the samples at 500°C for four hours (2013).

2.3.2. Metal concentrations in soils, mixtures, plant biomass and water extract of mixtures

The concentration of Cd, Cr, Cu, Ni, Pb, and Zn in the examined soil and the water extracts of the soil and plant biomass were determined with ICP-OES Ultrace 238 (Jobin Yvon Horriba France) using a direct calibration method. The samples of homogenized soil (1 g) and biomass (0.1 g) were digested in an acid mixture of HNO$_3$:HCl (1:3), and the water samples (15 g) were digested in HNO$_3$ (3 ml) in a microwave system (Multiwave 3000, Anton Paar). The digestion process lasted for 45 min at 180°C and at a pressure of 18 bars.

2.4. Results elaboration

Accumulation of particular heavy metals transported from soil to plant was evaluated using the bioconcentration factor (BCF), according to Eq. (1)

\[
\text{BCF} = \frac{\text{PC}}{\text{SC}} \quad (1)
\]

where: PC is heavy metal concentration in the biomass of whole plant (root and shoot) (mg kg$^{-1}$), and SC is the concentration of metal in the soil (mg kg$^{-1}$) (Abreu et al. 2012).

The BCF values greater than 1 indicate hyperaccumulation of an element in a plant, the values ranging from 0.1 to 1 denote moderate accumulation, whereas the values from 0.01 to 0.1 correspond to its low accumulation (Netty 2013).

The ability of particular species to translocate heavy metals from the roots to the shoots was assessed on the basis of calculation of the translocation factor (TF, %) according to Eq. (2)

\[
\text{TF} = \frac{\text{LC}}{\text{RC}} \times 100\% \quad (2)
\]

where LC: is heavy metal concentration in the biomass of shoots (mg kg$^{-1}$), and RC is heavy metal concentration in the biomass of roots (mg kg$^{-1}$) (Ociepa 2011).

2.5. Statistical analysis

The data were statistically analyzed through parametrical test ANOVA (Tukey’s test) using the Statistica 13.1 software package (Lublin University of Technology license). The letter indicators given at the average value of particular parameters considered in ANOVA test indicate statistically homogeneous groups (Tukey Homogeneous Groups). The presence of the same indicator designates the lack of statistically significant difference between them.

2.5.1. Artificial Neural Network (ANN) model

The obtained experimental data were introduced into the artificial neural network (ANN) model in order to experimentally verify the concentration of heavy metals in the biomass of clover cultivated on the substrates containing drilling wastes. The input parameters for the model included: drill cuttings dose, pH, and the
organic matter content, whereas the concentration of heavy metals in plant shoot was the output neuron (a single neuron in the output layer). Schematic representation of an artificial neural network for the model is shown in Fig. 1. The simulation works were carried out with the application of the specialistic statistical software, Statistica Neural Networks, and employed two ANN types: MLP (multi-layered perceptron) and RBF (radial basis function). The former model used linear, exponential, logistic, tanh, and sinus activation functions, while for the BFGS gradient (Broyden–Fletcher–Goldfarb–Shanno), the conjugate gradient and the steepest descent training algorithm were used to train the network. The latter network’s activation functions were for hidden neurons, the Gaussian distribution, and for output neurons, the linear function, and it was trained with the RBFT algorithm (Kulisz et al. 2020).

Results And Discussion

3.1. Effects of amendments on soil pH, organic matter, and total heavy metals in soils accumulation

Studies have shown that the drilling wastes addition statistically significantly increased the organic matter content in substrates, and the value of this parameter increased along with the amount of drill cuttings in mixtures (Table 3), reaching 5–37% higher values compared to non-modified soil. The highest concentration of organic compounds (2.6%) was observed in the substrate containing the highest, 15% dose of drill cuttings.

| Soil/mixture | pH       | OM [% d.m.] |
|--------------|----------|-------------|
| Z-0          | 4.16 d 0.015 | 1.9 b ± 0.05 |
| Z-2.5        | 6.66 c ± 0.02 | 2.0 ab ± 0.05 |
| Z-5          | 6.83 c ± 0.1  | 2.2 ab ± 0.05 |
| Z-10         | 6.98 b ± 0.1  | 2.3 ab ± 0.1  |
| Z-15         | 7.09 a ± 0.05 | 2.6 a ± 0.01  |

The pH value is a predominant factor which affects the mobility and bioavailability of heavy metals in soil through governing the solid-solution equilibrium of heavy metals (Zhao and Masaihiko 2007); therefore, the changes of this parameter occurring after waste addition to soil were analyzed. In the conducted studies, the pH of substrates containing wastes increased significantly compared to the control sample, ranging from 6.66 to 7.09 and increasing with the drill cuttings dose (Table 2). An increase in pH in the soils following drill cuttings addition was observed by numerous scholars (Bauder et al. 2005; Kisic et al. 2009; Yao and Naeth 2014; Yao 2013; Gonet 2006).
3.2. Effects of drill cuttings addition on heavy metal concentrations in soils

Studies showed that drill cuttings addition statistically significantly increased the concentration of heavy metals in soil, proportionally to an increased in the share of drill cuttings in mixtures. Cadmium, the concentration of which did not increase following waste addition, and even slightly reduced instead, constituted an exception (Fig. 2).

The highest content of each heavy metal was found in the mixture containing 15% drill cuttings. However, even in this mixture, the content of one of the analyzed heavy metals did not exceed the limit concentrations.
stated in the Polish law for class II soils, which include agricultural lands (2016), which amount to: for Cd – 2 mg/kg; Cr – 150–500 mg/kg; Cu and Ni 100–300 mg/kg; Pb 100–500 mg/kg; Zn 300–1000 mg/kg.

The concentrations of Cu (0.28–6.82 mg/kg), Ni (7.65–10.26 mg/kg), Pb (10.88 – 13.27 mg/kg), Cd (0.82–1.03 mg/kg), Cr (13.9–17.19 mg/kg) and Zn (62.86–151.75 mg/kg) in the prepared substrates were within the ranges of values measured in Poland in non-contaminated soils, provided by Kabata-Pendias et al. (2010), amounting to: 10–25 mg/kg for copper; from 10 mg/kg (light soils) to 50 mg/kg (heavy soils) for nickel; from 20 mg/kg (light soils) to 60 mg/kg (heavy soils) for lead; from 0.3 mg/kg (light soils) to 2 mg/kg (heavy soils) for cadmium; from 15 mg/kg (medium soils) to 24 mg/kg (heavy soils) for chromium, and 50–100 mg/kg for zinc (Kabata-Pendias 2010).

The regulations in Canada (Directive 50 2016) permit introducing certain types of drilling wastes to soil, provided that the quality standards for these wastes and soils are met. While comparing the concentrations of metals measured in the prepared substrates to the requirements established in Canadian regulations for agricultural soils, to which drilling wastes (expressed in the ratio to dry mass) amounting to Cd – 1.4 mg/kg; Cr – 64 mg/kg; Cu – 3 mg/kg; Pb – 70 mg/kg; Ni – 50 mg/kg; Zn – 200 mg/kg, were introduced, it was noted that they are lower than determined in regulations. Similar results were obtained by Yao et al. (2014), in the studies conducted on the soils with the addition of spent potassium drill fluids (Yao and Naeth, 2014). The standards established in Canadian regulations were not exceeded also in the case of the soils to which spent oil-based frill fluids were added, although elevated mean multi-annual contents of Hg, Pb, Ni, Co, Cu, Cr, and Zn were observed in these soils, compared to the soils without additions (Kisic et al. 2009).

### 3.3. Effects of drill cuttings addition on plant growth and heavy metals accumulation and translocation in red clover biomass

Changes in the substrate conditions caused by the introduction of drilling wastes to acidic soil, affected the amount and chemical composition of the test plant – red clover (*Trifolium pretense*) (Fig. 3).

The highest amount of clover biomass was obtained in the case of a mixture with 5% drilling wastes addition; it was 2.5-fold higher in comparison with the control soil. The biomass of clover cultivated on the mixtures with 5% and 10% drill cuttings addition reached twice higher values, whereas on the mixtures with 15% drill cuttings addition, it was 1.5-fold greater than the biomass cultivated on the control soil. Tukey’s test indicated no statistically significant differences between the mass of clover roots cultivated on particular mixtures. However, differentiation occurred in the biomass of clover shoots (Kujawska and Pawłowska, 2020). The heavy metal concentration in the shoots of clover cultivated on the investigated mixtures was determined and the results are shown in Fig. 4.

The drilling wastes addition statistically significantly increased the content of cadmium and copper, whereas it reduced the content of Cr and Pb in clover shoots. No statistically significant changes in the concentrations of nickel and zinc in clover shoots cultivated on the non-modified soil and the soil with
drilling wastes addition were observed. The increasing concentrations of these elements in biomass can be attributed to a change in the pH of the substrate, which affected the mobility and bioavailability of these elements (Kgopa et al. 2017). The mobility of these elements increases already at pH 6-6.5 values, slightly acidic and very slightly alkaline, which characterized the prepared mixtures. These values are within the range measured by Reeves and Baker (2000) for plants growing in metalliferous soils (5–25 mg kg\(^{-1}\)) (Reeves and Baker 2000). Cadmium is the most mobile and easily soluble heavy metal (Akhter et al. 2014). Although drill cuttings did not significantly increase the concentration of cadmium in substrates, the clover cultivated on these substrates took up cadmium easily.

The assessment pertaining to the usefulness of the investigated biomass as animal forage was based on maximum heavy metal contents in plants, determined by Kabata-Pendias et al. (1993). They are as follows: Cd – 0.5 mg/kg, Cr – 20 mg/kg; Cu – 30 mg/kg; Ni and Pb – 10 mg/kg; Zn – 100 mg/kg (Kabata-Pendias 1993). The plants cultivated on the prepared mixtures could be used as industrial plants, due to high content of zinc, which prevent them from being used as forage.

In order to evaluate the availability of heavy metals for plants and the capacity of plants for accumulating these metals, the bioconcentration factors (BCF) in shoots and roots of clover cultivated on the prepared mixtures and translocation factors (TF) were determined. Their values were presented in Figs. 5 and 6. Studies showed higher heavy metal accumulation capacity in the roots of plants, than in shoots.

Hyperaccumulation of cadmium was observed in the shoots of the plant cultivated on the soils without waste addition (BCF > 1). The values of BCF in the case of Cr, Pb, Zn, and Cu (for the Z–2.5 and Z–5 mixtures) in the aboveground parts reached the values < 0.06, whereas Ni and Cd were much higher; they were within the ranges of 0.18–0.37 and 0.38–0.79, respectively, which indicates moderate accumulation of these elements in plant shoots. The drill cuttings addition in the amount of 10% and 15% resulted in increased BFC for Cu in plant shoots to the level of 0.15–0.34, indicating its moderate accumulation.

In the case of roots, it was observed that the drill cuttings addition statistically significantly increased the clover capacity for Zn and Ni accumulation in all mixtures. Moreover, statistically significant increase of Cd accumulation in clover roots was observed, but only for the highest, 15% drill cuttings dose. However, the values of BCF for Cd were the highest, compared to other metals, and in all collected plants, the BCF value of this element was higher than 2. Such high bioconcentration factor was observed only in the case of Pb on the control sample. In turn, the BCF values higher than 1 were observed in root biomass in the case of Ni in the plants cultivated on the substrates containing drill cuttings (Fig. 6). It was observed that clover roots accumulated (0.48–0.52), Pb (0.20–0.45), Cu (0.16–0.19) i Zn (0.29–0.34) to a moderate degree.

It was observed that the drill cuttings addition to the substrate changed the metal accumulation capacity in particular plant parts. The accumulation of metals in the below-ground parts of plants cultivated on the control soil can be presented in the following order: Cd > Ni > Pb > Zn > Cr > Cu, whereas in roots, it is slightly different: Pb > Cd > Cr > Ni > Cu > Zn. After the highest drill cuttings addition the order was as follows: Cd > Cu > Ni > Cr > Pb > Zn in the above-ground parts and Cd > Ni > Cr > Zn > Pb > Cu in roots.
Accumulation of the investigated elements in clover roots was much higher than in the above-ground parts, which indicates the usefulness of this species in phytostabilization of polluted soils. In addition, introduction of 5% drill cuttings improved the growth conditions, which increased the accumulation of metals in roots and reduced their transport to the above-ground parts, which is especially evident in the case of cadmium, nickel, and zinc.

Although BCF can be a useful tool for assessing the influence of waste addition to soil on the accumulation of elements in biomass; however, interpretation of the obtained results is not easy, since bioaccumulation of metal by plants is dependent upon numerous factors, including variable soil conditions. As it was observed by McGrath and Zhao (2003), the values of BCF generally decrease with increasing metal concentration in soil (McGrath and Zhao 2003).

The obtained translocation factor (mobility) values of heavy metals in the clover cultivated on the mixtures with drilling wastes addition decreased under their influence (Table 3). All the determined TF values were lower than 1; hence, the mobility of metals in the root–above-ground part system of clover was very low. The reason for lower translocation of metals in the substrates containing drill cuttings might be alkalinization, which causes retention of metals in the root system. This phenomenon was also described by Kumpiene (2007) relating to the soils in which alkalinization occurred as a result of external organic matter addition (Kumpiene et al. 2007).

3.4. Artificial Neural Networks

On the basis of the experimental data: drilling waste doses, pH, organic matter of the mixtures with drill fluids, 100 networks were developed. The network quality was assessed using the following indicators: quality of training, quality of validation, training error and validation error from the least squares method, in order to select the most appropriate network type – MLP or RBF. The obtained network parameters were presented in Table 5.
| Network No. | Network Name | Quality (Training, %) | Quality (Validation, %) | Error (Training) | Error (Validation) | Activation (Hidden) | Activation (Output) |
|------------|--------------|-----------------------|-------------------------|------------------|-------------------|---------------------|---------------------|
| Cr         | MLP3-10-1    | 99.26                 | 97.11                   | 0.008            | 0.002             | Tanh                | Sinus               |
|            | MPL 3-6-1    | 99.74                 | 91.57                   | 0.004            | 0.008             | Tanh                | Tanh                |
|            | RBF 3-7-1    | 98.37                 | 94.04                   | 0.001            | 0.008             | Gaussian            | Linear              |
| Ni         | MLP 3-3-1    | 99.99                 | 99.90                   | < 0.001          | 0.004             | Exponential         | Logistic            |
|            | MLP 3-6-1    | 98.13                 | 99.98                   | < 0.001          | < 0.001           | Gaussian            | Linear              |
|            | RBF 3-8-1    | 98.97                 | 99.99                   | < 0.001          | 0.001             | Tanh                | Sinus               |
| Pb         | RBF 3-9-1    | 99.71                 | 99.72                   | 0.002            | 0.001             | Gaussian            | Linear              |
|            | MLP 3-7-1    | 99.97                 | 99.84                   | 0.001            | < 0.001           | Tanh                | Logistic            |
|            | RBF 3-8-1    | 98.97                 | 99.97                   | < 0.001          | < 0.001           | Gaussian            | Linear              |
| Cd         | RBF 3-5-1    | 94.88                 | 99.96                   | 0.003            | 0.009             | Gaussian            | Linear              |
|            | RBF 3-7-1    | 99.05                 | 94.32                   | 0.002            | 0.004             | Gaussian            | Linear              |
|            | MPL 3-8-1    | 93.46                 | 91.78                   | 0.005            | 0.012             | Sinus               | Logistic            |
| Cu         | MLP 3-9-1    | 98.17                 | 97.84                   | 0.014            | 0.004             | Sinus               | Tanh                |
|            | RBF 3-6-1    | 95.06                 | 99.88                   | 0.004            | 0.009             | Gaussian            | Linear              |
Figure 7 shows a comparison of experimental data obtained via predicting the concentrations of selected metals in plants for selected networks.

Regression coefficients (R) for selected networks for training, validation, and test data assume the values over 90%. Such high regression coefficients indicate good fit of the network. As it was presented in Table 5, mean coefficients of correlation between the experimentally determined concentrations of heavy metals in plants and the values predicted by ANN reached were higher than 95%, which indicates that the ANN model was able to quickly and reliably predict the concentrations of heavy metals. Low values of errors (< 0.1) also prove high accuracy of neural networks.

In order to investigate the influence of drill cuttings addition, pH, and organic matter content of soil on the concentration of heavy metals, a sensitivity analysis (Table 6) was carried out. The network sensitivity analysis indicated the highest sensitivity to the impact of drill cuttings addition on the concentration of heavy metals in plants.
On the basis of the obtained experimental results, an artificial neural network model for predicting the metal concentrations in plants. Such models can be created using various soil additives and soil quality parameters, which facilitates predicting the impact of wastes on the accumulation of metals in plants.

The results obtained by us and other researchers showed that ANN can be employed for predicting the concentrations of metals in plants,

Haatab et al. (2013) created an artificial neural network model for predicting the chromium concentration in the leaves of laboratory-cultivated dwarf French bean, in the soils with the addition of dolomite limestone, compost of poultry manure and pine bark (CPM), as well as mixture of dolomite limestone and compost of poultry manure and pine bark. The input data used in the model are: soil amendments, soil pH, electric conductivity and dissolved organic carbon in soil, and the obtained result is the concentration of Cr in dwarf French bean. Their ANN model indicated mean coefficient of correlation between the measured and predicted chromium values in dwarf French bean equal to 0.9998 (Hattab et al., 2013).

Jahantab et al. trained the neural network for Zn and Cr heavy metals in soil and plant, with the R2 value in most cases higher than 0.9 and close to 1, indicating the applicability of neural network for over-predicting data (Jahantab et al. 2020).

Gharaibeh and Ben-Hani (2003) created an artificial neural network model for predicting phytotoxicity, dry mass accumulation and reduction depending on the concentrations of metals used for irrigation. The input (selenium and nitrate levels) and the output data (growth reduction and selenium bio-tissues uptake) were simulated using artificial neural network program. Simulated data was then used to predict the interaction between selenium and nitrate in irrigation water at different levels of both nitrates and selenium (Gharaibeh and Bani-Hani 2003).

The versatility of artificial neural network tools is the feature, which enables to account for the selected quantity and quality of the investigated soil quality parameters. Moreover, artificial neural network models may be the basic tool for managers, engineers, and decision makers, aiding in designing, managing, and making decisions pertaining to the introduction of additives to soil.

**Conclusions**

1. Drill cuttings addition to acidic soil (pH ~4.2) significantly increased the concentration of heavy metals (excluding cadmium). However, the concentrations of these metals did not exceed the permissible values established in the regulations related to the quality of agricultural lands.

2. The biomass of plants cultivated on the mixtures with drill cuttings addition exhibited the highest enrichment for Cd and Cu. The concentrations of heavy metals in the above-ground parts of the plants cultivated on the mixtures with each dose of wastes, did not exceed the values recommended for the plants used as animal forage.

3. Heavy metals were accumulated in higher concentrations in the roots of red clover than in the aboveground parts. This observation is of practical importance, because clover is commonly used as animal
forage. Shoots of the clover cultivated on the mixtures with wastes indicated hyperaccumulation of cadmium and nickel as well as moderate accumulation of chromium, lead, copper, and zinc.

4. Drilling wastes reduced the mobility of heavy metals in the root-shoot system.

5. The performed experimental studies showed the potential of developing the models for predicting the heavy metal concentrations in plants, based on artificial neural networks. This is proven by a good quality of the networks determined on the basis of high coefficient of correlation (> 0.99). The sensitivity analysis of the developed networks showed that the drilling wastes addition had the highest impact on the heavy metal content in plants, in comparison to the changes in pH and organic matter content.

6. Undoubtedly, the impact of pollutants contained in drill cuttings requires constant monitoring; therefore, it seems justified to include model studies, in addition to experimental studies.

**Declarations**

**Ethical approval** The experiments comply with the current laws of Poland.

**Compliance with ethical standards**

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Figures

![Figure 1](image)

**Figure 1**

Schematic representation of the artificial neural network (ANN) for the analysis of process parameters
Figure 2

Heavy metals concentration in the soil mixtures obtained in the experiment

Figure 3

Dry weight of red clover [g]

Different part of red clover
Average values of the dry biomass of shoot and root of red clover obtained in the pot experiment (Kujawska and Pawłowska, 2020)

Figure 4

Heavy metals concentration in the shoot biomass obtained in the experiment

Figure 5

Bioconcentration factors (BCF) of the examined metals in the red clover shoots
Figure 6

Bioconcentration factors (BCF) of the examined metals in the clover roots
Figure 7

Comparison between models and real data for metals in shoots