Antoni ROGÓŻ<sup>1</sup>, Barbara WIŚNIOWSKA-KIELIAN<sup>1</sup> and Monika  $TABAK<sup>1</sup>$ 

# **ASSESSMENT OF DEGREE OF TRACE ELEMENT POLLUTION OF SOIL AND ROOT VEGETABLES**

## **OCENA STOPNIA ZANIECZYSZCZENIA GLEB ORAZ WARZYW KORZENIOWYCH PIERWIASTKAMI ŚLADOWYMI**

**Abstract:** The investigations were focused on determining the level of trace element pollution of soil and root vegetables cultivated for human consumption in the Miechowski county. The collected soils differed in view of their floatable particles, colloidal clay and organic matter content, as well as pH value. Differences in the total content of trace elements and the content of their soluble forms extracted with  $0.1 \text{ mol} \cdot \text{dm}^{-3}$  HCl solution also occurred.

The total content of trace elements in the analysed soils and their soluble forms were determined by soil reaction. Geometric mean of total heavy metals content in the studied soils was as follows: 55.08 mg  $\text{Zn} \cdot \text{kg}^{-1}$ , 8.08 mg Cu  $\cdot \text{kg}^{-1}$ , 10.05 mg Ni  $\cdot \text{kg}^{-1}$ , 19.85 mg Pb  $\cdot \text{kg}^{-1}$  and 0.46 mg Cd  $\cdot \text{kg}^{-1}$ . The assessment of heavy metals content in the analysed soils was conducted according to the framework guidelines for agriculture elaborated by IUNG. In this respect 44 of analysed samples were classified as soils with natural heavy metals contents. The content of trace elements in cultivated root vegetables was determined by the species, analysed plant part and soil pH.

The content of these elements was much higher in plant tops than in roots. For instance, if one assumes geometric mean of cadmium and lead content in parsley roots grown in soil with  $pH \le 6.5$  as 100, cadmium concentration in leaves was higher by 64 %, and lead content was higher by 36 %. On the basis of the obtained results, content of trace elements in vegetable roots was assessed assuming the critical levels of trace element in agricultural products stated by IUNG. Taking into consideration these criteria was stated that all samples of the vegetable roots fulfilled the requirements of usefulness for human consumption in respect of copper and nickel contents, and majority of them in respect of lead content. However, 7.6 % of samples did not fulfil these requirements because of excessive content of lead ( $> 1$  mg  $\cdot$  kg<sup>-1</sup>), 52.5 % of zinc ( $> 50$  mg  $\cdot$  kg<sup>-1</sup>), and as much as 81.6 % samples because of cadmium excess (> 0.15 mg  $\cdot$  kg<sup>-1</sup>), despite their being cultivated in majority in soils with natural these metals content. Excessive content of copper and nickel was registered only in 1.3 % of tops samples, lead in 51.3 %, zinc in 79.7 %, and cadmium in 98.1 % of tops samples, in which 7.6 % of tops samples did not meet fodder requirements for zinc, and as much as 65.8 % for cadmium.

<sup>&</sup>lt;sup>1</sup> Department of Agricultural and Environmental Chemistry, University of Agriculture in Krakow, al. A. Mickiewicza 21, 31–120 Kraków, Poland, phone: +48 12 662 4348, fax: +48 12 662 43 41, email: rrrogoz@cyf-kr.edu.pl, email: rrkielia@cyf-kr.edu.pl

<sup>\*</sup> Corresponding author: rrrogoz@cyf-kr.edu.pl

Taking in to account all roots and tops samples, only 0.6 % of them did not meet criteria of usefulness for human consumption, because of copper and nickel excess, 29.4 % for lead, 66.1 % for zinc, and as much as 89.9 % of samples contained excessive amount of cadmium. Moreover, in 3.8 % of samples Zn content exceeded admissible values in fodder and in 32.9 % contained excessive amount of Cd.

**Keywords:** soil, root vegetables, content of Cu, Zn, Cd, Pb and Ni

## **Introduction**

A characteristic feature of heavy metals is their considerable ability to accumulate in soil and plants. An increase in the content of these metals in the soil-plant system poses a hazard to animals and people. Exceeding critical element content in soil has an inhibitory effect on plant growth, leading to changes in plant chemical composition [1–3]. Total contents of individual heavy metals in the soils of Poland fluctuate widely: from 0.01 mg  $\cdot$  kg<sup>-1</sup> to over a dozen, reaching even several hundred mg  $\cdot$  kg<sup>-1</sup>, depending on many factors [1]. Investigations conducted by numerous authors revealed that heavy metal uptake by plants depends on: the forms and concentrations of individual metals in the substratum, pH value, species and variety of plants, kind of applied organic and mineral fertilizers, etc. [2–5]. These factors may decide about a stimulating or limiting effect on the uptake of these metals by plants. Therefore, some agrotechnical measures aim to reduce heavy metal availability in the soil-plant system [3, 6, 7]. The content of trace metals (zinc, copper, nickel, lead, cadmium) is usually determined in crops cultivated for human consumption, animal feed or industrial use.

The presented research aimed to determine the usefulness for human consumption of root vegetables cultivated in the soils with natural content of heavy metals (zinc, copper, nickel, lead, and cadmium).

#### **Materials and methods**

In the first decade of September 2003, 44 soil samples were collected from the depth of 0–25 cm of arable land topsoil from the Miechowski county. Each of the analysed soil samples was an average of between 10 and 20 individual samples (0.5–1.0 kg of soil). The places of sample collection were described in the "Material and methods" section in the first part of the paper [8]. Basic physical and chemical properties of soils were determined in air-dried samples by means of methods commonly used in agrochemical studies: granulometric composition by Bouyoucose-Casagrande aerometric method in Proszynski' modification, pH by potentiometer in soil suspension in  $H_2O$  and in 1 mol  $\cdot$  dm<sup>-3</sup> KCl solution (1:2.5), hydrolytic acidity by Kappen's method, and organic carbon content by Tiurin's method [9].

The total content of trace elements in the analysed soils was assessed after previous mineralisation at 450 °C. Subsequently, the samples were digested in a mixture of perchloric and nitric acids  $(2:3, v/v)$  and dissolved in HCl [9]. Concentrations of trace element soluble forms were assessed in  $0.1 \text{ mol} \cdot \text{dm}^{-3}$  HCl solution. Vegetables (red beetroot, carrot, parsley and celery) at full consumption maturity were taken from the same places as the soil samples. Analysed plant sample was an average of 12 samples (about 1 kg of fresh mass). The plant material prepared for analyses was dry-mineralised. The obtained ashes were dissolved in nitric acid diluted with water at 1:2  $(v/v)$ ratio to obtain the solutions in which trace element concentrations were assessed by means of ICP AES method. The obtained results were used to compute simple correlation coefficients (r) between the content of trace elements in soils and selected physico-chemical soil properties.

## **Results and discussion**

**Contents of trace elements in soil.** The soil samples gathered in the Miechowski county varied according to their physico-chemical properties. The pH value of the analysed soils assessed in water suspension ranged from 5.14 to 7.64, and from 4.13 to 7.23 when measured in 1 mol  $\cdot$  dm<sup>-3</sup> KCl solution. The organic carbon content ranged from 6.8 to 16.1 g  $\cdot$  kg<sup>-1</sup>, at geometric mean 10.1 g  $\cdot$  kg<sup>-1</sup>, whereas the content of floatable particles fluctuated from 25 to 52 %, and the content of colloidal clay was from 1 to 23 %. The site of soil and plant samples collection as well as some physico-chemical properties of soils were presented in the first part of this paper in the "Materials and methods" section [8].

Total contents of trace elements in the analysed soils and the content of their soluble forms assessed in  $0.1 \text{ mol} \cdot \text{dm}^{-3}$  HCl solution were diversified (Table 1). Taking in to consideration texture of studied soils one may recognize as natural content of Pb, Cu and Ni and natural to increased content of Zn and Cd [9].

Table 1



Statistical parameters of trace elements concentration in soils

The share of the content of soluble forms assessed in  $0.1 \text{ mol} \cdot \text{dm}^{-3}$  HCl solution in the total heavy metal content fluctuated widely depending on the analysed element, *eg*

for copper it was on the level of 14 %, for cadmium it was on the level of 78 %. The content of trace elements in soil, and therefore their bioavailability, is determined by many factors: the bedrock, soil forming processes, pH value, and anthropogenic factors [1, 6]. In the analysed soils, the pH value determined the content of metal soluble forms (Table 2).

Table 2



Parameter of trace elements concentration in soils  $[\text{mg} \cdot \text{kg}^{-1}]$  depending on soil pH value

Total contents of the analysed metals and their soluble forms determined in 0.1 mol HCl  $\cdot$  dm<sup>-3</sup> solution were decreasing with increasing pH value. For instance, assuming geometric mean total zinc content in at least slightly acid soils ( $pH_{\text{KCl}} \leq 6.5$ ) as 100, the total content of this cation in neutral soils ( $pH_{\text{KCl}} \geq 6.6$ ) was lower by 11 %, whereas its soluble form, respectively by 37 %. Similar relationships occurred, to a bigger or lesser extent, for the other analysed elements.

On the basis of the obtained results, an assessment of the pollution degree of the analysed soils with trace elements was made, according to the guidelines of IUNG [10]. The assessment of pollution of the analysed soil considered the content of floatable particles and the pH value. The studied soils were classified to soils with natural content of heavy metals. Research conducted by Terelak et al [11] revealed that over 80 % of agricultural soils in Poland are characterised by a natural content of heavy metals in the arable layer, whereas in 17 % the content is elevated. Physico-chemical properties of the analysed soils to different extent influenced total contents of trace elements and their soluble forms, as evidenced by simple correlation coefficients (Table 3).

Table 3

Properties of soils	Zn	Cu	Ni	Ph	Cd
	Total content				
pH in KCl	$-0.1551$	0.064	$-0.2706$	$-0.3488*$	$-0.2296$
$\varnothing$ < 0.02 mm	0.2531	0.2343	$0.495**$	0.2606	$0.595***$
$\varnothing$ < 0.002 mm	$-0.390**$	$-0.1839$	0.264	0.1168	$0.457**$
Content of soluble forms					
pH in KCl	$-0.169$	$-0.420**$	$-0.602$ ***	$-0.330*$	$-0.243$
$\varnothing$ < 0.02 mm	$-0.162$	0.2143	0.037	0.076	$-0.112$
$\varnothing$ < 0.002 mm	$-0.358**$	$-0.075$	$-0.062$	$-0.3630*$	$0.355**$

Simple correlation coefficients (r) between trace element content in soils and chemical properties of soils

(r) significant at \* p = 0.05; \*\* p = 0.01; \*\*\* p = 0.001; n = 44 samples.

The analysed soil pH value ( $pH_{\text{KCl}}$ ) variously influenced total contents of copper, zinc, cadmium and nickel in soils, significantly affecting lead concentrations (negative correlation). A significantly negative correlation was determined between the pH value measured in KCl and concentrations of copper, lead and nickel in soluble forms assessed in 1 mol HCl  $\cdot$  dm<sup>-3</sup> (Table 3).

**Concentrations of trace elements in root vegetables.** Vegetables are plants accumulating in their tissues considerable amounts of heavy metals which change their chemical composition. The content of selected elements in cultivated vegetables was determined by soil pH, plant species, and analysed plant part [4, 12, 13]. Irrespective of pH value, the content of the studied elements was much higher in tops than in roots (Table 4). If one assumes *eg* geometric mean content of cadmium and lead in parsley roots grown in soils with  $pH_{KCl} \le 6.5$  as 100, then this metal content in its leaves was higher by 64 % and the lead concentration was higher by 36 %. Similar dependencies occurred in the other studied root vegetables.

Trace elements group includes microelements whose are crucial for living organisms because they fulfil some specific functions, e.g. they are catalysts of cell metabolism (Cu, Zn), but also the elements whose physiological role has not been determined yet (Cd and Pb) [14]. Average copper content in plants fluctuated from 5 to 20 mg  $\cdot$  kg<sup>-1</sup> d.m. Its deficit may occur below 5 mgCu  $\cdot$  kg<sup>-1</sup> d.m., while over 20 mg  $\cdot$  kg<sup>-1</sup> d.m. has a toxic effect on plants. Assuming copper content on the level of  $\leq 5$  mg  $\cdot$  kg<sup>-1</sup> d.m. as deficient, an about 20 % deficiency of this cation was noted in beet roots, celery and parsley roots.

Based on the obtained results, an assessment of vegetable pollution with trace elements was made, assuming the critical values of trace metal concentrations in agricultural products as suggested by IUNG [10]. The guidelines allow for human consumption of agricultural products with the following concentrations of trace elements:  $\leq 50$  mg Zn  $\cdot$  kg<sup>-1</sup>,  $\leq 20$  mg Cu  $\cdot$  kg<sup>-1</sup>,  $\leq 10$  mg Ni  $\cdot$  kg<sup>-1</sup>,  $\leq 1$  mg Pb  $\cdot$  kg<sup>-1</sup> and  $\leq 0.15$  mg Cd  $\cdot$  kg<sup>-1</sup> d.m. The permissible values for usefulness for animal feed are as follows:  $\leq 100$  mg Zn  $\cdot$  kg<sup>-1</sup>,  $\leq 25$  or 50 mg Cu  $\cdot$  kg<sup>-1</sup> (for sheep and for other animals, respectively),  $\leq 50$  mg Ni  $\cdot$  kg<sup>-1</sup>,  $\leq 10$  mg Pb  $\cdot$  kg<sup>-1</sup> and  $\leq 0.5$  mg Cd  $\cdot$  kg<sup>-1</sup> d.m. The root vegetables pollution is presented in Table 4.

The root vegetables only in case of copper and nickel fulfilled the criteria of permissible content for human consumption proposed by IUNG [10].

The permissible Zn content for human consumption was exceeded in roots in 75.6 % of beetroot, in 11.6 % of carrot, in 70.5 % of parsley and in 53.3 % of celery samples. The permissible Zn content was exceeded in tops in 70.7 % of beetroot, in 65.1 % of carrot, in 81.8 % of parsley and in 70.0 % of celery samples. The permissible Zn content for animals was exceeded in tops in 9.8 % of beetroot, in 2.3 % of carrot, in 6.8 % of parsley and in 13.3 % of celery samples.

The permissible Cu and Ni contents for human consumption and for animals were exceeded only in 93.3 % and 6.7 % of celery tops samples, respectively.

The permissible Pb content for human consumption was exceeded in roots in 2.4 % of beetroot, in 6.8 % of carrot, in 11.4 % of parsley and in 10.0 % of celery samples. The permissible Zn content was exceeded in tops in 26.8 % of beetroot, in 68.2 % of carrot, in 40.9 % of parsley and in 73.3 % of celery samples.

The permissible Cd content for human consumption was exceeded in roots in 87.8 % of beetroot, in 95.5 % of carrot, in 54.5 % of parsley and in 90.0 % of celery samples. The permissible Cd content was exceeded in tops in 19.5 % of beetroot, in 20.5 % of carrot, in 65.9 % of parsley and in 16.7 % of celery samples. The permissible Cd content for animals was exceeded in tops in 80.5 % of beetroot, in 77.3 % of carrot, in 27.3 % of parsley and in 83.3 % of celery samples.

Soil pollution with cadmium is an undesirable phenomenon both from biological and ecological perspective. The problem is more complex, because an assessment of cadmium made according to suggested criteria revealed that the cultivated root vegetables do not meet the requirements for usefulness for consumption since they contain over  $0.15 \text{ mg } \text{Cd} \cdot \text{kg}^{-1} \text{ d.m.}$ , despite their cultivation in soils with natural or slightly increased content of Cd (Table 4).

Organic and mineral fertilization aims at supplying nutrients to cultivated plants but it should also affect their chemical composition without worsening the crop yield quality. Numerous papers have demonstrated that some amounts of heavy metals which do not affect considerably their soil level are supplied with mineral fertilizers, *eg* phosphorous or multicomponent ones, which are absorbed by the root system in large quantities [7, 15]. Similarly as in Author's own studies, necessity of monitoring trace elements content in vegetables also demonstrated others scientists [16, 17]. Sharma et al [17] stated more Zn, Pb and especially Cd, and less Ni and Cu in carrot roots: 30–112.6 mgZn, 17.75–23.5 mgPb, 0.25–1.25 mgCd, 0.25–5.5 mgNi and 4–6.5 mgCu  $\cdot$  kg<sup>-1</sup> d.m.



Table 4

Assessment of Degree of Trace Element Pollution of Soil and Root Vegetables 75

\* Critical values for sheep; \*\* Critical values for other animals. \* Critical values for sheep; \*\* Critical values for other animals.

(on average 55.52 mg, 20.35 mg, 0.62 mg, 5.15 mg and 3.10 mg  $\cdot \text{ kg}^{-1}$  d.m., respectively) than in Author's own studies. In studies of Ngole [18] roots of carrot growing in control object without fertilization contained  $2 \text{ mgPb} \cdot \text{kg}^{-1}$ , and on sludge-amended soil between 2 and 3 mgPb  $\cdot$  kg<sup>-1</sup> d.m. Under those conditions carrot contained 12 mgCu  $\cdot$  kg<sup>-1</sup> and 5–6 mgCu  $\cdot$  kg<sup>-1</sup> d.m., respectively.

On the basis of their investigations, Wisniowska-Kielian and Baran [5] conducted an assessment of vegetables quality based on IUNG criteria [10] and stated that in 11 % of samples zinc concentrations were exceeded (celery and parsley leaves). They also demonstrated that 71 % and 32 % of the analysed samples meet the criteria of consumption usefulness, respectively. Leaves of the analysed vegetables accumulated much bigger quantities of metals than roots. Research conducted by Jasiewicz et al [13] shows that culinary measures such as washing or separating the flesh from the skin reduces the amount of consumed heavy metals. The authors demonstrated that cadmium concentration in radish flesh was six-fold lower than in its skin. Similar results was stated in case of beetroot [19].

The obtained research results point to a necessity to apply agrotechnical measures in the analysed region, which would reduce passing of excessive amounts of heavy metals (particularly cadmium) from soil to plants and therefore to higher organisms. Cadmium poses a hazard to animals and people, because it is toxic and easily absorbed by the organism and has a long biological half-life. Cadmium undergoes a considerable bioaccumulation, so there is a large diversification in its concentrations in animal tissues. Literature data reveal that the problem of cadmium may be solved by increasing pH due to soil liming, increasing the content of organic matter and phosphorus fertilization [3, 20]. In result of these agrotechnical measures, soil ability to bind cadmium increases and therefore its bioavailability decreases. Zaniewicz-Bajkowska et al [21] demonstrated significant decrease of cadmium content in roots of celery and in leek. However, the same Authors [22] show that not always this reduction is so easy to achievement. They found that beetroot cultivated on limed soil contained in roots from 1.08 to 1.14 mgCd  $\cdot$  kg<sup>-1</sup> d.m., and without liming from 1.19 to 1.23 mgCd  $\cdot$  kg<sup>-1</sup> d.m., whereas total content of cadmium in soil was  $1.25$  and  $1.24$  mgCd  $\cdot$  kg<sup>-1</sup> d.m., and its soluble forms  $0.114$  and  $0.126$  mg Cd  $\cdot$  kg<sup>-1</sup> d.m., respectively. It means that all root samples contained 7.2–8.2-times higher amount of cadmium than permissible in plant material destined for human consumption while soil contained up to twofold bigger amount of this metal than in Authors own study. Under those conditions leaves of beetroot contained  $1.27-1.36$  mgCd  $\cdot$  kg<sup>-1</sup> and  $1.38-1.43$  mgCd  $\cdot$  kg<sup>-1</sup>, respectively.

Kachenko and Singh [23] studied heavy metals contamination in vegetables grown in urban and metal smelter contaminated sites in Australia stated 2.89–4.23 mgZn, 0.860–1.04 mgCu,  $\lt$  0.02 mgPb and  $\lt$  0.05 mgCd  $\cdot$  kg<sup>-1</sup> f.m. of beetroot and 4.45–32.18 mgZn,  $0.626-3.85$  mgCu,  $\lt 0.02-0.630$  mgPb and  $0.005-0.141$  mgCd  $\cdot$  kg<sup>-1</sup> f.m. of parsley. After these data convertion to dry mass Zn, Cu and Pb contents in beetroot were lower than obtained in Authors' own studies, and Cd content was to high. In case of parsley only Cu content was similar to stated in these studies. In Elbagermi et al [24] studies carrot contained 3*.*61 mgZn, 5*.*00 mgCu, 0*.*21 mgNi, 0*.*21 mgPb and 0*.*12 mgCd  $\cdot$  kg<sup>-1</sup>, *ie* less than in presented studies.

## **Conclusions**

1. The analysed soils were greatly diversified in regard their pH values measured in 1 mol  $\cdot$  dm<sup>-3</sup> KCl solution (from 4.13 to 7.31), and total trace element content (5.65–18.28 mgCu  $\cdot$  kg<sup>-1</sup>, 54.6–227.6 mgZn  $\cdot$  kg<sup>-1</sup>, 0.52–2.92 mgCd  $\cdot$  kg<sup>-1</sup>).

2. Contents of the analysed trace elements in root vegetables cultivated in the Miechowski county ranged widely depending on the soil pH, content and form of elements, species, and analysed plant part.

3. Taking into consideration heavy metal content was stated that the root vegetables (both roots and tops) did not fulfil criteria of usefulness for human consumption (according to IUNG) because of excess of copper and nickel in 0.6 %, lead in 29.4 %, zinc in 66.1 %, and especially of cadmium in 89.9 % of samples, despite their being cultivated in soils with natural Cd content.

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#### **OCENA STOPNIA ZANIECZYSZCZENIA GLEB ORAZ WARZYW KORZENIOWYCH** PIERWIASTKAMI ŚLADOWYMI

Katedra Chemii Rolnej i Środowiskowej Uniwersytet Rolniczy im. Hugona Kołłątaja w Krakowie

Abstrakt: Badania dotyczyły określenia stopnia zanieczyszczenia gleb oraz warzyw korzeniowych uprawianych na cele konsumpcyjne pierwiastkami śladowymi na terenie powiatu miechowskiego. Zebrane gleby były zróżnicowane pod względem zawartości cześci spławialnych, iłu koloidalnego, materii organicznej, wartości pH, całkowitej zawartości pierwiastków śladowych oraz ich form rozpuszczalnych w roztworze 0,1 mol  $HCl \cdot dm^{-3}$ .

O całkowitej zawartości pierwiastków śladowych, jak i ich formach rozpuszczalnych w badanych glebach decydował odczyn gleb. Średnie geometryczne całkowite zawartości metali ciężkich w badanych glebach wynosiły: 55,08 mgZn  $\cdot$  kg<sup>-1</sup>, 8,08 mgCu  $\cdot$  kg<sup>-1</sup>, 10,05 mgNi  $\cdot$  kg<sup>-1</sup>, 19,05 mgPb  $\cdot$  kg<sup>-1</sup> i 0,46 mgCd  $\cdot$  kg<sup>-1</sup>. Zanieczyszczenie badanych gleb metalami ciężkimi oceniono w oparciu o ramowe wytyczne dla rolnictwa opracowane przez IUNG. Pod tym względem 44 badane próbki glebowe zaliczono do gleb o naturalnej zawartości metali ciężkich. O zawartości pierwiastków śladowych w uprawianych warzywach korzeniowych decydował gatunek, analizowana część rośliny oraz pH gleby. Zawartość tych pierwiastków była znacznie większa w częściach nadziemnych niż w korzeniach. Przyjmując np. średnią geometryczną zawartość kadmu i ołowiu w korzeniach pietruszki wyrosłych na glebach o pH<sub>KCl</sub>  $\leq$  6,5 za 100, zawartość kadmu w naci była większa o 64 %, a ołowiu o 36 %. Na podstawie uzyskanych wyników oceniono zawartości metali śladowych w warzywach korzeniowych, przyjmując ich poziomy krytyczne w płodach rolnych opracowane przez IUNG. Biorac pod uwagę te kryteria stwierdzono, że wszystkie próbki korzeni spichrzowych badanych warzyw spełniały wymagania przydatności konsumpcyjnej dla ludzi pod względem zawartości miedzi i niklu, a większość pod względem zawartości ołowiu. Natomiast 7,6% próbek nie spełniało tych wymagań z powodu nadmiernej zawartości ołowiu (> 1 mg · kg<sup>-1</sup> s.m.), 52,5% cynku (> 50 mg · kg<sup>-1</sup> s.m.), a aż 81,6 % próbek z powodu nadmiaru kadmu (> 0,15 mg ·  $kg^{-1}$  s.m.), pomimo że warzywa były w większości uprawiane na glebach o naturalnej zawartoœci tych metali. Nadmierne zawartoœci miedzi i niklu zanotowano tylko w 1,3 % próbek, o³owiu w 51,3 %, cynku w 79,7 %, a kadmu a¿ w 98,1 % próbek naci tych warzyw, w tym 7,6 % próbek nie spełniało wymagań paszowych pod względem zawartości cynku, a aż 65,8 % – kadmu.

Z ogółu próbek korzeni i naci nie spełniało wymagań konsumpcyjnych z powodu nadmiernej zawartości miedzi i niklu tylko 0,6 % próbek, ołowiu 29,4 % próbek, cynku 66,1 %, a kadmu aż 89,9 % próbek, w tym w 3,8 % próbek wykazywało przekroczenia dopuszczalnych zawartości Zn i 32,9% Cd w paszach.

Słowa kluczowe: gleba, warzywa korzeniowe, zawartość Cu, Zn, Cd, Pb, Ni