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Original Research

Trace Elements in Garden Soils, Vegetables and Apples Near a Large Cu Tailings Pond in SW Poland

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Abstract

Copper ore mining, processing, and smelting are considered important sources of soil and crop contamination with metals and metalloids. The concentration of elements in soils and selected vegetable and fruit species has been monitored around a large-scale tailings pond (located in SW Poland) to evaluate its impact on soil quality and food plant production. The total concentrations of As, Cu, Pb, and Zn in root vegetables (carrot, parsley and beetroot), potato tubers and apple fruit grown in the backyard gardens in the surroundings of the Cu tailings pond were analyzed along with element concentrations in the topsoil layers. Element concentrations in plants were found at similar levels compared to products commercially available in Poland and did not exceed legal or provisional limits. The concentrations were noticeably lower than reported from the other mining or industrial regions and argued for little impact of the tailings pond. The concentrations measured in the present study did not differ from the respective values reported previously, suggesting a stable environmental quality in the landfill surroundings. Element concentrations in edible plant parts were poorly correlated with their concentrations in soils and the distance to the tailings pond, suggesting the importance of local or individual factors.

Keywords: soil contamination, vegetables, apple, backyard gardens, trace elements

Introduction

The copper industry, particularly the mining, ore processing, and smelting stages, has been widely reported as an important source of soil and plant pollution with trace metals and metalloids [1-4] that generate various risks to human health [5-11]. Large volumes of contaminated or toxic fine-grained rock wastes (tailings) produced during ore enrichment are stored in above-ground tailing ponds [11-13] and influence the surrounding environment by infiltrating groundwater, dusting from an uncovered landfill surface, and, periodically, by catastrophic dam failures and tailing floods [14-17]. Although several means are implemented to control the present influences of the tailing landfills [18, 19], their negative impact cannot be completely eliminated, partly due to historical soil contamination [20-22]. Therefore, the present-day

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impacts and contamination levels must be permanently monitored to provide knowledge regarding the existing risk to human health and required activities, including land remediation [23, 24].

The copper industry in south-western Poland has developed in a typical agricultural and residential area and initially led to serious local soil and plant contamination [25-27]. Furthermore, real and serious threats were connected with numerous tailing ponds [28, 29]. At present, the potential impacts are minimized at all stages of ore mining and processing, and environmental monitoring has been widely implemented [30]. Many potential environmental risks have also been identified for Europe’s largest copper ore tailings pond – “Żelazny Most” – including the contamination of soils as well as consumable and fodder plants [6, 31]. The results of previous studies differ, but some of them suggest that the tailings pond, despite the colossal scale of its construction, has limited impact on the soil and plant contamination, which may result from effective protection means [32]. However, already due to the scale of impoundment and his planned enlargement, local residents are still concerned about the negative impact of the landfill on their health, either directly by air contamination or indirectly, e.g., by contaminating food plants, in particular fruit and vegetable from the backyard gardens.

Therefore, the aim of this study was (a) to assess the present concentration of trace elements in the vegetables and fruit grown in the villages neighboring the large tailing pond, in terms of legal limits and common element concentrations in the market products, and (b) to analyze the relationship between plant contamination and the distance from the landfill and the concentration of elements in garden/orchard soils.

Material and Methods

Description of the Area

The tailings impoundment – “Żelazny Most – located in south-western Poland (Lubin and Polkowice counties; N51°31’, E16°12’) has operated since 1977 on a total disposal area of ca. 1600 ha (Fig. 1). It collected ca. 560 mln m^3 of tailings (ca. 28 mln tons annually at present) that led to the dam uplifting up to 65 m above the initial ground level. Finely ground tailings are transferred from the flotation plants hydraulically in a slurry form. To ensure geotechnical stability, the external part of the landfill is free of water cover, leading to the formation of 200-m-wide dry beaches, extending on a total area of ca. 800 ha (changing seasonally). The significant elevation of the landfill

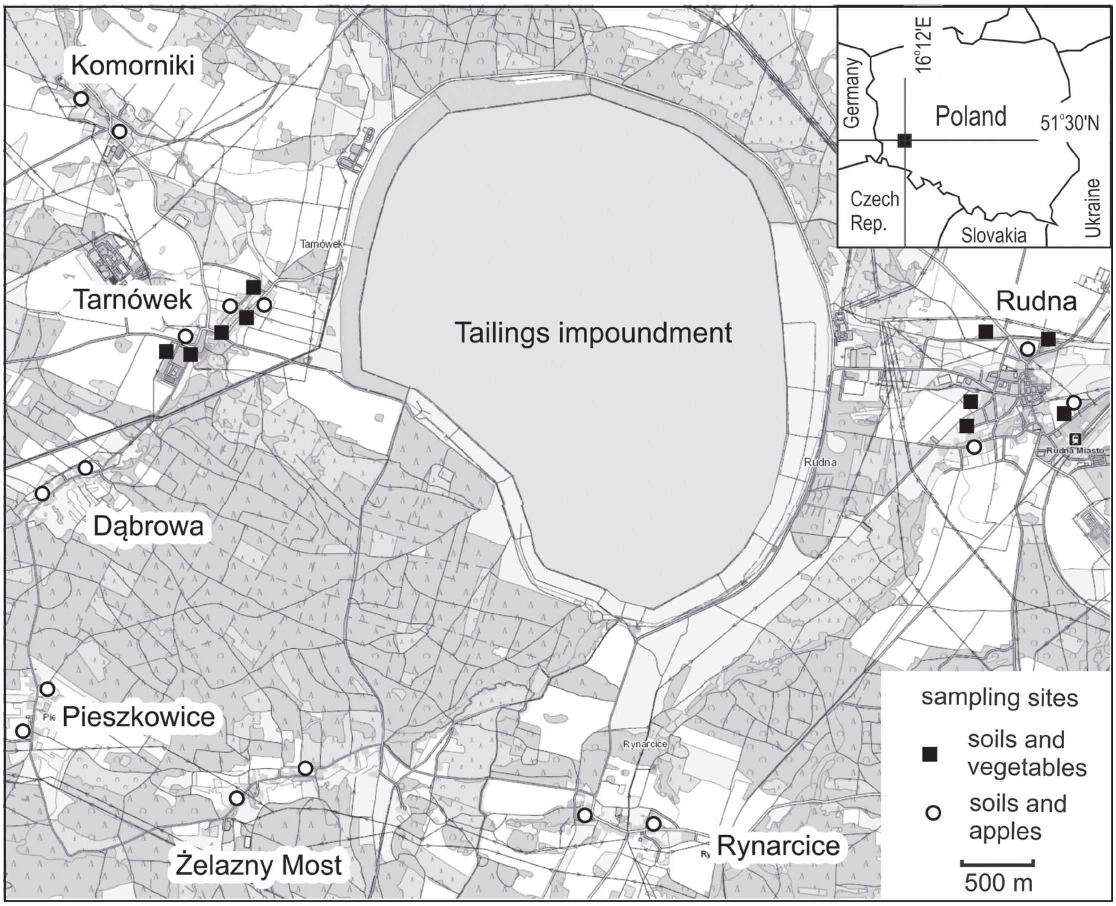

Fig. 1. Schematic situation map of the sampling sites around the tailing impoundment.

above the surroundings and the silty-sandy texture of tailings favor wind erosion on dry beaches, observed for 36-72 days per year [30]. The tailings have a neutral to slightly alkaline reaction (pH of 7.5-7.8) and contain residual amounts of trace elements, including Cu 1700-2600 mg kg^{-1}, Pb 300-500 mg kg^{-1}, As 10-90 mg kg^{-1}, and Zn 40-60 mg kg^{-1} [18]. To avoid excessive dusting with metal-bearing particles, a set of anti-erosion means was implemented, including selective deposition of sediments, covering of dry beaches with thin bitumen film, water curtains on the dams, and successive biological reclamation of the external dam walls [18]. Soil and plant monitoring began in 1995 to demonstrate the effectiveness of the implemented protection means [31-33].

The landfill is surrounded by soils developed primarily from the Pleistocene glacio-fluvial sands and moraine tills, often also cover sands over tills, classified as Brunic Arenosols, Planosols, and Luvisols [32]. The mean annual air temperature is approximately 8.0-8.5ºC, and the mean annual precipitation is 500-550 mm (temperate moist climate). Despite extensive land afforestation in the direct neighborhood of the impoundment, arable soils still predominate with rye (triticale), wheat, barley, corn, and rape being the dominant crops. Despite industrialization, traditional backyard gardening has maintained its popularity in the surrounding villages; thus, locally produced vegetables and potato still significantly contribute to the inhabitants' diet.

Collection and Analysis of Plant Samples

Vegetable and garden soil samples were collected in the years 2018-2020. The most commonly grown vegetable species (carrot root, parsley, root beetroot, and potato) were sampled in various locations, depending on their growing sites and farmer permission in particular year. Apple and orchard soil samples were collected in 2018 only. No typical orchards occur in this area, thus the word "orchard" was used in this paper as conventional term for a section of garden with apple trees. Root vegetables and potato samples (number of samples displayed in Table 4) were collected in early September from gardens in Rudna and Tarnówek, the villages closest to the landfill, taking 3-5 primary samples from each site (Fig. 1). Apple samples were collected in October, as 5 primary samples per site, from six villages neighboring the impoundment (Rudna, Tarnówek, Dąbrowa, Komorniki, Pieszkowice, and Żelazny Most). Plant samples were cleaned in distilled water, and potato tubers were peeled before analysis [34]. All plant samples from a site were cut into smaller pieces and mixed together, then the representative analytical subsample (ca. 100 g) was randomly selected. Analytical samples were dried at 40ºC to a stable weight, finely ground, and mineralized with concentrated HNO_3 (in triplicate) in a microwave oven. The total concentration of trace elements (As, Cu, Pb, and Zn) was measured in extracts using inductively coupled plasma (Thermo Scientific iCAP 7400). The reference plant materials (IAEA-V-10 Hay, CRM 279 Sea lettuce, CRM 281 Rye grass) were used as certified internal standards. The procedures were accepted if the recovery ratio was 90-105%. The dry mass of plant samples was measured after sample drying at 40 ºC to a stable weight. The selection of particular elements depended on the legal monitoring decision of the Environmental Protection Agency issued based on initially expected element emission and resulting contamination in the vicinity of the impoundment.

Collection and Analysis of Soil Aamples

Topsoil (0-25 cm) samples, 36 in gardens and 43 in orchards, were collected in spring on the same plots as the plant samples (15 primary soil samples mixed together from each plot to prepare representative and analytical soil sample, ca. 500 g each). After drying at 50ºC to a stable weight, samples were ground, sieved to pass a 2-mm mesh, and digested (in triplicate) with a mixture of concentrated HCl and HNO_3 (3:1, v/v) in a microwave oven, following ISO standard 11466. The near-total (or "aqua-regia extractable") concentration of trace elements (Cu, Zn, Pb, As) was measured in the extracts using inductively coupled plasma (Thermo Scientific iCAP 7400). The reference soil materials (RTH 912, RTH 953) were used as certified internal standards. The procedure was accepted if the recovery ratio was 90-105%. Soil and plant analyses were performed in the laboratory of environmental analysis at the University of Environmental and Life Sciences in Wroclaw, awarded by the Polish Certification Centre with a legal accreditation certificate.

Moreover, particle-size distribution of soil samples was analyzed by sieve and hydrometer method, pH in distilled water (1:2.5 v/v) was measured potentiometrically, and the content of soil organic carbon was analyzed by dry combustion method (CS Matt 500, Stroelein, Germany).

Most of the analyzed variables did not have a normal distribution (confirmed by Shapiro-Wilk test). Thus, the Spearman algorithm was preferred to calculate correlation coefficients, and the medians are displayed along with mean values. The statistical significance of differences between the mean values was demonstrated at $p<0.05$ using a post-hoc Fisher test. The analytical results below detection limit were included in the statistical calculations with a half of detection limit as their conventional value. Calculations were performed and graphs produced using Statistica 13 software.

Table 1. Particle size distribution, pH and soil organic carbon in topsoil layer of soils under study (n = 79).

Soil properties	Unit	Minimum	Maximum	Mean
Clay fraction	%	5	11	7
Silt fraction	%	11	25	15
Sand fraction	%	73	82	78
pH (water)	-	6.8	7.6	7.1
Soil organic carbon	%	1.05	1.60	1.33

Results and Discussion

Soil Properties and Element Concentrations

The ranges are mean values of basic physicochemical soil characteristics were similar for backyard gardens and orchards, thus were presented in one block (Table 1). The topsoil layers of soils under study had relatively uniform texture in loamy sand and sandy loam classes, with clay content ranging from 5 to 11%. Topsoil layers had neutral reaction and relatively high content of organic carbon (Table 1), as for post-glacial materials in Central Europe, which indicates the influence of regular liming and organic fertilization. The concentration of elements in topsoil layers varied in a broad range for all elements, and the medians were in all cases lower than mean values, indicating the strong influence of the few highest results. Among elements under study, mean As was at the lowest, while Zn at the highest concentrations (Table 2). Mean As, Zn, and Pb concentrations were higher in backyard gardens than in orchards, but the difference was insignificant in case of As. Conversely, Cu concentration was somewhat, but insignificantly higher in orchard than in garden soils. Cu concentration in topsoil layers of garden soils significantly decreased with the distance to the tailings pond, whereas Pb and As concentration, conversely, increased with the distance. Similar trends were also observed in orchard soils, but only the relationship for Pb was significant (Table 3). Element concentrations in

Table 2. Concentration of elements in the topsoil layer of soils under study.

Land use	Number of samples	Element	Minimum	Maximum	Mean mg kg^{-1}	Median	Standard deviation
Backyard gardens	36	As	2.8	14.1	4.8*a*	4.0	2.6
		Cu	12.1	40.5	20.2*a*	16.1	8.8
		Pb	21.5	42.2	30.3*a*	27.8	7.0
		Zn	38.2	206	90.9*a*	79.8	32.3
Orchards	43	As	2.2	9.5	3.5*a*	3.1	1.5
		Cu	8.0	36.5	23.2*a*	19.0	9.0
		Pb	6.5	29.0	17.3*b*	15.5	4.7
		Zn	28.5	135	65.3*b*	62.0	22.9

Letters *a* and *b* indicate homogeneous groups of means (separately for each element) checked by Fisher test (different letters confirm a difference significant at $p<0.05$).

Table 3. Spearman coefficients for the correlation between the distance to the tailings pond and element concentration in the topsoil layers.

Land use	Parameter	Cu	Zn	Pb	As
Backyard gardens	Distance	-0.51*	0.08	0.38*	0.29*
	Clay content	0.11	0.16	0.05	-0.02
	pH	0.06	0.15	0.22	0.12
	Organic carbon	0.14	0.19	0.28*	0.20
Orchards	Distance	-0.22	0.11	0.28*	0.19
	Clay content	0.15	0.10	-0.01	-0.05
	pH	0.09	0.12	0.20	0.10
	organic carbon	0.18	0.18	0.25	0.12

Explanation: * - coefficients significant at $p<0.05$; number of samples – as in Table 2.

topsoil layers did not or poorly (insignificantly) correlate with clay content and soil pH. The correlation values were relatively highest for the relation between organic carbon and elements concentration, but significant only for Pb in garden soils (Table 3).

Element Concentrations in Vegetables and Apples

As and Pb were at significantly lower concentrations than Cu and Zn in all plant species under study, both in fresh (Table 4) and dry mass (Fig. 2). The lowest (minimum) As concentrations in a fresh mass of products were at 0.01 mg kg^{-1} or below detection limit. Maximum As concentrations were clearly higher in beetroot (up to 0.18 mg kg^{-1}) than in other vegetables and apple (0.06-0.09 mg kg^{-1}), which resulted in higher mean and median As values in beetroot (Table 4). Also, the mean As concentrations calculated for dry mass of plants were significantly highest in beetroot and significantly lowest in apple (Fig. 2). The lowest Pb concentrations in a fresh mass of products, similarly to As concentrations, were at 0.01 mg kg^{-1} or below detection limit (Table 4). The highest Pb concentrations reached similar levels in all plant species, i.e., 0.08-0.11 mg kg^{-1}. Irrespectively of similar range of results, the mean and median Pb concentrations (in a fresh mass) were clearly lower in potato tubers than in root vegetables. Similarly, mean Pb concentrations calculated to dry mass were significantly lower than in root vegetables (Fig. 2). Mean Pb concentration in apple, although similar to those in potato, did not differ significantly from calculated for root vegetables (Fig. 2). Both the minimum and maximum Cu concentrations (in a fresh mass) in apple were the lowest (0.02-0.6 mg kg^{-1}), followed by potato tubers (0.2-0.8 mg kg^{-1}), whereas in the root vegetables both the minimum and maximum Cu concentration were at least two times higher than in potato (Table 4). Among the root vegetables, the highest median Cu values were found in parsley, followed by beetroot and carrot, reaching 1.30, 1.12, and 0.84 mg kg^{-1}, respectively. Whereas the mean Cu concentrations recalculated to dry mass, still the highest in parsley, did not differ significantly from those in carrot (Fig. 2). Respectively to low values in fresh mass, also the Cu concentration in dry mass of potato was significantly lower than in root vegetables and significantly lowest in apple (Fig. 2). The differences between plant species were the most evident in the case of Zn concentration. The median value of this element in beetroot (5.85 mg kg^{-1}

Table 4. Concentration of elements in fresh mass of apple, root vegetables, and potatoes grown in the surrounding of tailing impoundment.

Species	Number of samples	Element	Minimum	Maximum	Mean mg kg^{-1}	Median	Standard deviation
Carrot (root)	33	As	<0.01	0.06	0.03	0.02	0.01
		Cu	0.44	1.50	0.89	0.84	0.27
		Pb	<0.01	0.11	0.05	0.05	0.03
		Zn	1.67	7.99	3.32	3.03	1.21
Beetroot (root)	32	As	<0.01	0.18	0.05	0.04	0.04
		Cu	0.56	2.19	1.17	1.12	0.37
		Pb	0.01	0.10	0.05	0.05	0.03
		Zn	2.29	13.2	6.11	5.85	2.50
Parsley (root)	32	As	<0.01	0.09	0.03	0.03	0.02
		Cu	0.77	2.91	1.40	1.30	0.40
		Pb	<0.01	0.09	0.05	0.05	0.03
		Zn	2.43	8.39	3.97	3.54	1.47
Potato (tubers)	28	As	<0.01	0.06	0.02	0.01	0.01
		Cu	0.22	0.76	0.41	0.39	0.14
		Pb	<0.01	0.08	0.02	0.01	0.01
		Zn	0.66	2.24	1.33	1.33	0.37
Apple (fruit)	43	As	<0.01	0.06	0.02	0.01	0.01
		Cu	0.02	0.59	0.24	0.22	0.11
		Pb	0.01	0.10	0.03	0.03	0.01
		Zn	0.05	0.87	0.26	0.24	0.12

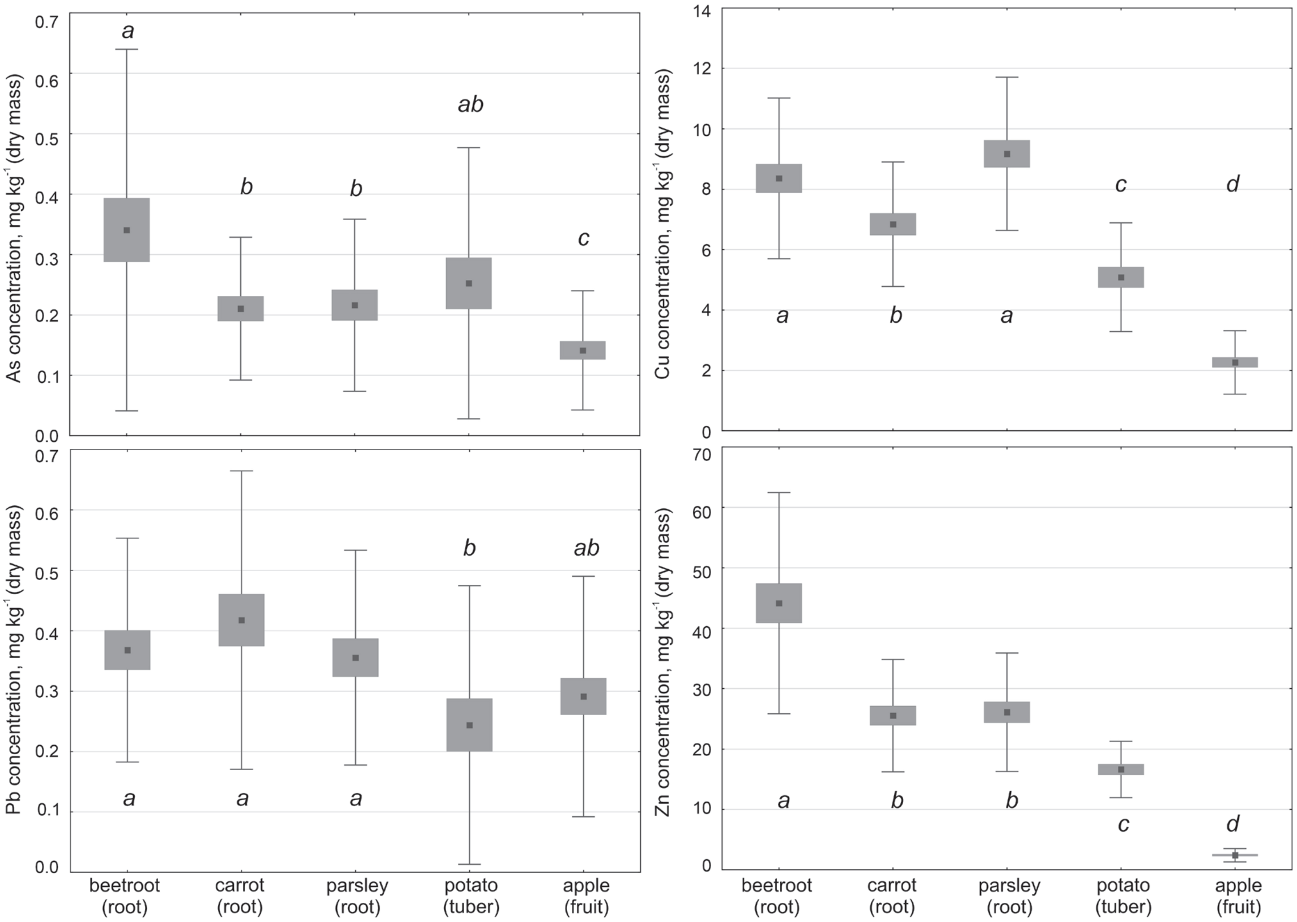

Fig. 2. Comparison of element concentration in fruit, vegetables, and potato. Central point – arithmetic mean, box – standard error, whiskers – standard deviation. Letters *a, b, c* and *d* indicate homogeneous groups of means checked by Fisher test (different letters confirm a difference significant at $p<0.05$).

in a fresh mass) was 4.5-times higher than in potato tubers and nearly 25-times higher than in apple (Table 4). The differences were also evident in a dry mass (Fig. 2) that resulted in a statistically significant differentiation of mean values: beetroot (44 mg kg^{-1}) > carrot+parsley (26 mg kg^{-1}) > potato (17 mg kg^{-1}) >> apple (2.6 mg kg^{-1}). The concentration of elements in the food plant samples generally wasn't related to the distance to the tailings pond (Table 5), excluding Pb in apple, significantly higher in the larger distance to the pond. In general, the element concentrations in vegetables and cereal grain did not correlate with the concentrations in soils (Table 6). The correlation coefficients were in many cases negative, but statistically insignificant. The only significant exceptions were identified for negative association for Zn in soil and Zn in parsley root, and the positive relationship between As in soil and As in apple (Table 6).

Table 5. Relationship (Spearman coefficients) between element concentration in vegetables and apples, and the distance to the tailings pond.

Element	Beetroot	Carrot	Parsley	Potato	Apple
As	0.10	0.02	0.10	0.10	0.16
Cu	-0.02	-0.22	-0.09	-0.11	0.00
Pb	0.15	0.01	0.02	0.04	0.38*
Zn	0.17	0.11	0.25	0.04	-0.20

Explanation: * - coefficients significant at $p<0.05$; number of samples – as in Table 4.

Table 6. Correlation (Spearman coefficients) between element concentrations in the topsoil layer and vegetables and apples.

Element	Beetroot	Carrot	Parsley	Potato	Apple
As	-0.06	0.03	0.11	0.20	0.22
Cu	-0.18	-0.07	0.10	-0.20	0.13
Pb	-0.03	-0.10	0.20	-0.02	0.30*
Zn	-0.05	0.13	-0.50*	0.03	0.05

Explanation: * - coefficients significant at $p<0.05$; number of samples – as in Table 4

The comparison of the data obtained in the present study to element concentrations reported in food plants by other authors is difficult due to methodological differences and varying presentation standards. In particular, plant tissues may be analyzed in a fresh state or after drying, and the metal concentrations may be reported in a fresh or dry mass, respectively to local or international legal limits. Mean and median Pb concentrations in root vegetables, potato tubers, and apple were in the area under study similar to Pb concentrations in respective species commercially available in Poland [35-39]. Only the maximum Pb concentrations in potato tubers were higher than that typically reported [35, 36]. However, nearly all reported Pb concentrations in vegetables under study were lower than the thresholds allowed in Poland and the EU, specified at 0.1 mg kg^{-1} [34]. The reported Pb concentrations were also noticeably lower than those obtained from contaminated arable and garden soils in Europe, China, and the USA [9, 10, 40-44]. Arsenic concentrations were similar to typical levels reported for potato and beetroot commercially offered in Poland but higher in carrot, parsley, and apple. The legal threshold for As has not been specified in Poland/EU, but all samples under study had As concentrations lower than 0.5 mg kg^{-1} of a fresh mass (the highest observed was 0.18 mg kg^{-1}) established as a limit in China [45]. The legal limits for Zn and Cu in vegetable and apple have not been established either in Europe, America, or China, as these metals are important microelements, often supplied to crops intentionally in fertilizers. However, the provisional (indicatory) maximum levels in potato and root vegetables were specified in Poland at 50 and 20 mg kg^{-1} of a dry weight for Zn and Cu, respectively [46]. The median Zn and Cu concentrations in vegetable, potato, and apple in the present study were within the ranges reported as typical for non-contaminated products offered in Poland [37-39], very close to median values calculated by Kabata-Pendias [47], and noticeably below the provisional limits [46]. Moreover, Zn and Cu concentrations in plants, even the highest found, were noticeably lower than the concentrations reported from sites contaminated by metal mining or smelting [7, 9, 21].

Element concentrations reported in root vegetables and potatoes in the present study were at similar levels (no statistically significant differences) compared to the values reported in previous studies [31, 32]. Relatively low and stable concentration of elements in food plants over longer periods confirmed relatively little impact of the tailings pond on the quality of plants grown in the gardens and orchards located in villages in the pond surroundings. The latter statement was supported by low values of correlation coefficients for element concentration in plants and the distance to the tailing pond, in general insignificant statistically (Table 5). The only significant value, for Pb in apple, was positively correlated to distance to the pond, i.e., Pb concentration was the highest in the most distant villages. However, it must be noted, that the closest vegetable/fruit gardens are located in a distance of at least 850 m from the landfill (up to 2,600 m), i.e., are absent in a direct proximity of the pond (Fig. 1). Moreover, some of villages are "isolated" from the landfill by forest complexes. Both these factors (distance and isolation) may result in a limited inflow of the metal-bearing dust from the pond to garden/orchards under study [16, 18, 30, 48].

Soil contamination has been typically considered the first potential threat for crop and vegetable quality in the mining and industrial regions [15, 16]. Median concentrations of all elements under study (Table 2) were 1.5-3 times higher than the values for arable soils of Poland, approximated at 2.8, 6.2, 11.8, and 32 mg kg^{-1} for As, Cu, Pb, and Zn, respectively [49] that suggests a negative influence of the tailing impoundment. However, the statistical relationships between element concentration and distance to the impoundment were, in general, poorly marked (Table 3). Of course, Cu concentration decreased with distance (value significant for garden soils), but Pb and As concentrations increased, indicating other than the tailing pond sources of enhanced levels of Pb and As in distant locations. Many authors [40, 43, 44, 50-52] have reported substantial variability of soil contamination in gardens on meso- and micro-scales (in the neighboring plots), closely related to individual soil management practices, as fertilization and liming history over a longer period of cultivation. Therefore, it is not clear, whether the median element concentrations in arable (agricultural) soils are appropriate reference levels for backyard garden soils, if these soils typically are featured by higher contamination with elements, as well as higher pH and higher organic matter content [43]. The concentrations of elements in garden soils, reported in present study, were similar to those reported from the so-called allotment gardens in the small cities, and noticeably lower than in the large towns [43, 44, 52]. This supports opinion about insignificant impact of the tailings pond on the quality of garden soils in the area under study and larger importance of the local contamination sources, such as road traffic and transportation, liming (using the waste lime from metal industry), or even individual (plot-related) factors, such as some kinds of organic fertilization (e.g. using the sewage sludge) and plant protection (using the heavy metal-based pesticides) [43, 52]. All these contamination sources are highly probable, in particular in the gardens/orchards located in the old villages. The mean concentrations of all elements in soils under present study were substantially below the legal intervention limits, established in Poland at 10-50, 100-300, 100-500, and 300-1000 mg kg^{-1} (depending on soil texture, pH and soil organic matter) for As, Cu, Pb, and Zn, respectively [53], arguing for a low potential risk for plant contamination [5, 7, 8, 9].

At such low soil and plant contamination levels, the correlation coefficients for element concentrations in soils and plants were statistically insignificant

(Table 6), as often reported from the other sites with limited soil contamination [10, 16, 23]. The only two significant coefficients were identified for Pb in soil and apple (positive value), and Zn in soil and parsley (negative value) (Table 6). Although adversely oriented, these two relationships may have similar explanations. Cao and Bourquin [45] reported little impact of soil contamination with Pb and As on the quality of apple fruit, due to element blocking in roots and vegetative parts. The significant correlation between Pb in soil and apple in this study may be, therefore, an apparent effect of parallel relationships of the correlations between the distance from the tailings pond and Pb content in the garden/orchard soils (Table 3) and apple fruit (Table 5). In case of Zn, its content in soil was unrelated to the distance from the pond (Table 3), whereas Zn content in parsley was larger in larger distances from the pond (Table 5). These two significant relationships, in line with prevailing insignificant ones, confirmed that Zn (and other elements) in food plants may not be closely related to element concentration in soil, if the latter is relatively low. Metal uptake by plants may depend on soil physico-chemical properties (in particular on soil pH and humus content), influencing the element solubility, mobility, and availability to plants [40, 54, 55, 56, 57]. In such conditions, the concentration of elements in food plants may stronger depend on the current element inflow from the applied fertilizers, metal-containing pesticides and local air contamination than on the content of element bound in soil [8, 41, 44, 52, 58, 59].

Conclusions

Although the large tailings pond analyzed in this study is considered a potentially serious source of soil and plant contamination, the concentrations of As, Cu, Pb, and Zn in root vegetables, potato, and apple fruit grown in its proximity were found at similar levels compared to commercially available products and did not exceed the legal and indicative limits, where specified. The measured concentrations of elements in vegetables and apple were noticeably lower than those reported from other mining or industrial regions and argue for a relatively low impact of the tailings pond on the crop quality. The median element concentrations measured in the present study did not differ significantly from the respective values reported in previous studies and indicate a stable environmental quality in the surrounding of the tailings pond. Element concentrations in vegetable, potato, and apple were poorly correlated with their concentrations in soils and with the distance to the tailings pond, confirming the limited impact of the emissions from the tailings pond on plant quality and the presence of the other local or even individual (plot-related) pollution sources.

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Conflict of Interest

The authors declare no conflict of interest.

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