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HEAVY METALS CONTAMINATION OF SOIL AND VEGETABLES IN THREE REGIONS FROM ROMANIA: A REVIEW

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INTRODUCTION

Heavy metals pollution can pose a danger to both human and environmental health. Heavy metals are inorganic pollutants that can be extracted from the soil and accumulated in plants (Manea et al. 2020), especially in vegetables of economic interest. The Carpathian Mountains have played an important role as a source of ferrous and non-ferrous metals since ancient times. Romania has numerous sites with a long history of mining, industrial and agricultural pollution, and consequently of soil pollution with heavy metals from such activities. For example, the long-term accumulation of heavy metals in arable ecosystems negatively affects soil fertility and the quality of cultivated vegetables. Sustainable management of heavy metals in agricultural ecosystems ensures that the soil performs its ecosystem functions. An analysis of heavy metals in agricultural ecosystems and accumulation in plants of economic interest, in particular vegetables, must be carried out to define strategies to ensure the sustainable management of these metals in agricultural systems (Olimpia Vrinceanu et al. 2015).

In total there are 14 areas with a severe degree of contamination with toxic elements, mainly heavy metals. The studies carried out by Romanian researchers (Muntean et al. 2013, Nimirciag 2012, Damian et al. 2018, Senila et al. 2011, Lăcătuşu et al. 1996, Lăcătuşu and Lăcătuşu 2008, Dumitru et al. 2018, Dumitru et al. 2000) have identified areas with contamination risks in Copşa Mică, Baia Mare, Zlatna, Ploieşti-Brazi, Oneşti, Bacău, Suceava, Piteşti, Târgu Mureş, Turnu Măgurele, Tulcea, Işalniţa, Braşov, and Govora. However, there is little information about the dangerous effects of vegetables metal-contaminated and the effect of consuming contaminated vegetables by the inhabitants of those areas (Damian et al. 2010, Harmanescu et al. 2011, Ispas et al. 2018, Damian et al. 2018, Nedelescu et al. 2017).

This review of the literature from the last ten years presents the current state of heavy metal pollution of the soil in three regions of Romania (Crişana, Transylvania, and Moldova), the degree of accumulation of these metals in vegetables and the long-term effects of ingestion of contaminated vegetables on human health.

SOURCES OF HEAVY METALS

In order to characterize the soil, the physical and chemical properties of the soil (texture, pH, carbonate content, base saturation, nitrogen content and movable forms of phosphorus and potassium) must be determined (Lăcătuşu 2014). The content of heavy metals in soil is represented by the metal elements of the base rock on which the soil layer developed and the various anthropic contamination sources (Alloway 2013). Although heavy metals are usually found in low concentrations in soil, in recent years a number of anthropogenic sources have had a strong impact on the environment (Cocârţă et al. 2016) and pose significant human health risks (Luo et al. 2015). Anthropogenic sources such as industrialization (mining, metallurgy, burning of fossil fuels) and rapid urbanization (traffic, solid municipal waste) led to the accumulation of large amounts of heavy metals in soil (Luo et al. 2015, Masindi and Muedi 2018, Sodango et al. 2018, Lacatusu 2000). For example, Dr. Anca-Rovena Lăcătuşu and Prof. Dr. Radu Lăcătuşu present two examples of cities in Romania, Copşa Mica and Baia Mare, where, in the urban soils of these two locations, the total Cd levels exceeding, on average, were determined, 10 times the maximum allowable limit in the soil from Copşa Mică and 3.6 times in those from Baia Mare (Lăcătuşu and Lăcătuşu 2018).

In Romania, Baia Mare, is situated in the Crişana region, and it was one of the most important areas for the exploitation of metals. Therefore the associated mining sites with non-ferrous deposits are now an important source of heavy metal pollution of soil and sediments (Chira et al. 2014). Thirteen soil types have been identified in the Baia Mare basin and the quality of these soils have been affected over time due to extraction activities, the preparation of non-

ferrous ore, and non-ferrous metallurgy. Although some mines were closed, behind them remained the settling ponds and the mine-settling pits. Another example is water from underground springs that now passes through the mining galleries and reaches the surface contaminated. In this area, about 25,000 ha is polluted with heavy metals (Hg, Cu, Zn, Cd, Ni, Co, Mn, Cr)(Coman and Bianca 2010). Studies already carried out on the level of soil pollution in urban areas, claim that in the Ferneziu district of Baia Mare, there are high concentrations of As, Pb, Cu, Zn, and Cd. This district from north-eastern part of Baia Mare Municipality, is around the lead smelters Romplumb, which has been in operation since 1844 (Mihali et al. 2012, Roba et al. 2015, Donici et al. 2018).Therefore, in this area, the main sources of soil pollution with heavy metals are mine openings, ore processing plants, metallurgical plants, and settling ponds (Mihali et al. 2013). In Baia Mare, the highest concentrations of heavy metals were found in the eastern part and around the industrial area (Chira et al. 2014, Mihali et al. 2013).

Another region in Romania, Transylvania, was famous for its mining activities. For example, the towns of Zlatna, Brad, Caraciu, and Deva were important sources for non-ferrous minerals, such as Cu, Zn, Pb, Au, Ag (Manu et al. 2016). The Copşa Mică area is one of the most polluted anthropic sites in Romania (Damian et al. 2018). Dorina Maria Popa et al. (2018) presented the results of the health status of the population exposed to heavy metal pollution in Transylvania and this study is focused on three contaminated areas: Baia Mare, Copsa Mică, and Aghires. They identified in soil samples from three areas where factories still operated, through 4 methods of analysis, high level of toxic trace elements, especially Hg, Cd and Cu (Popa et al. 2020). A study was done in twelve grassland ecosystems, situated in the Zlatna Depression within the Trascau Mountains, Transylvania – Romania. According to national legislation (M.O. 756/03.11.1997), all heavy metal concentrations have exceeded the reference values on the studied grasslands, except for Mn, and in few areas the soil concentrations were lower than legal-reference values (Bodescu and Onete 2016, Dumitru et al. 2018).

In the Moldova region, the sources of pollution have been classified as in figure 1. The level of soil pollution with heavy metals, in the vicinity of Iron and Steel Integrated Works (ISIW) at Galati, Romania, was measured. The results for the collected soil samples highlighted the presence of the following chemical elements: Ca, Fe, K, Mn, Ti, As, Cr, Cu, Hg, Ni, Pb, Rb, Sc, Sr, V, and Zn. In general, the concentrations of heavy metals in the soil decrease as the distance from the ISIW plants increases and are higher than the levels detected in the control samples (Ene et al. 2010). In Roman city, the high concentrations of heavy metals in the soil, resulting from pollution, determines their absorption in plants. Sometimes the toxic levels reach negative effects on plant growth and crop development, as well as with undesirable consequences for other environmental factors (Chitimus et al. 2018).

FACTORS INFLUENCING THE UPTAKE OF HEAVY METALS BY VEGETABLES

In the world, the problem of heavy metal contamination of soil in particular and in the environment in general is widespread (Zwolak et al. 2019). Harmful to organisms, heavy metals can bioaccumulate and persist in the environment. The accumulation and translocation of metals in vegetables are largely dependent on environmental factors, plant genotypes and local anthropogenic activities. In a given situation, the influence of heavy metals in vegetables is limited by different categories of vegetable species and varieties (Gan et al. 2017). The degree of pollution and the impact of heavy metals is assessed taking into account the distance from the source of pollution, but also the cumulative influence of other environmental variables (Manu et al. 2016). Extracted by plants from the soil, heavy metals can easily enter the food chain, and therefore people can be exposed to various elements and doses of metals. Exposure of animal organisms including humans to different doses of heavy metals as a result of the consumption of contaminated vegetables can be a serious problem. For example, exposure to Cd, Pb and Hg compounds is very dangerous especially during the fetus development, but also in the first years of a child's life, because this aspect can cause irreversible changes in the central nervous system. In human body, Cd, can cause skeletal disorders, liver damage, cardiovascular diseases, dysfunction of the sex glands, it is also carcinogenic, nephrotoxic and neurotoxic. It also causes and disrupts the body's mineral balance. Mercury and especially the methyl mercury compounds, accumulates mainly in the brain tissue, causing damage to the central nervous system, especially during the development of fetal brain (Zwolak et al. 2019, Branca et al. 2018, Budnik and Casteleyn 2019).

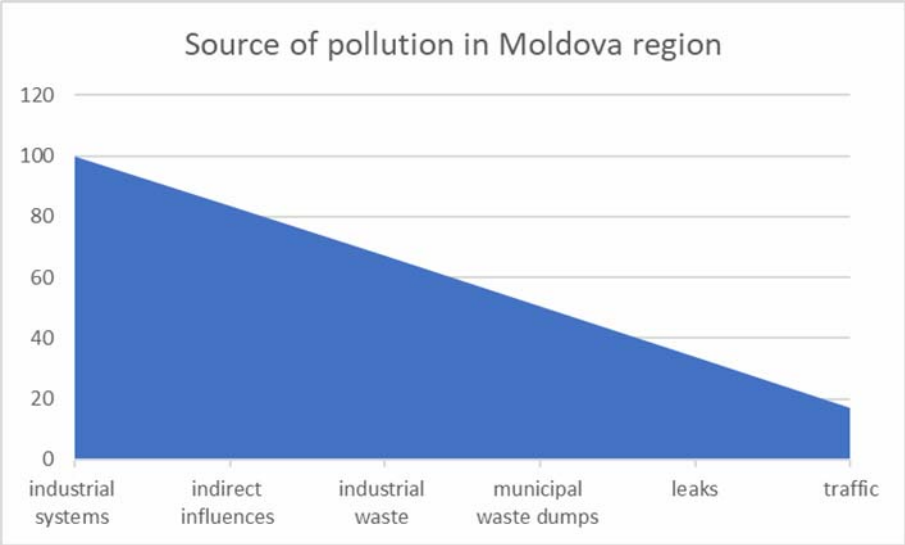

Figure 1 Source heavy metal pollution in Moldova region (Faciu et al. 2012)

CONTAMINATION OF SOIL AND VEGETABLES IN ROMANIA

Vegetables are an essential part of the human diet and are consumed both fresh, cooked and processed. They act as buffer agents for acid generation during digestion and some metals contained by vegetables are even important from a biochemical and physiological point of view for maintaining a healthy body (Jolly et al. 2013). Several studies have been carried out to assess potentially toxic elements content in vegetables grown on polluted soils from Romania (Nedelescu et al. 2015). The amount of metals in vegetables depends on their concentration in the soil and the physico-chemical properties of the soil (pH, electrical conductivity, organic matter content, etc.) since these factors affect the mobility of metals in the soil and their bioavailability to plants (Nedelescu et al. 2015, Qureshi et al. 2016, Balkhair and Ashraf 2016). The nutritional quality of vegetables depends on the physico-chemical properties but also on the perception of consumers (Kyriacou and Rouphael 2018). The influence of these factors on the safety of vegetables can't be determined by observation and necessitates monitoring to determine the influence of these factors on food safety (Grover et al. 2016).

CRIŞANA REGION

Results of a study targeting the heavy metal content of fruits and vegetables grown in the Baia Mare mining area showed that heavy metals have a greater potential to accumulate in vegetables than in fruits (Roba et al. 2016). Another study in Ferneziu, a mining/smelting influenced area, showed that investigated vegetables had high levels of heavy metals, exceeding in some cases the safe limits imposed by the national and international legislation. The concentration order of heavy metals in the analyzed vegetable samples was Zn > Pb > Cu > Ni > Cd > Cr. Lettuce, parsley, and carrot have been shown to have a large amount of heavy metals in their edible mass, indicating that the cultivation and consumption of these vegetables should be restricted in the investigated area (Roba et al. 2015). In the Baia Mare the level of pollution of soil and plants with Cu and Zn area has been studied. Mobility of Cu and Zn in some plants, including vegetables, have also been studied using soil as a transfer factor to plants. The highest value for Cu was recorded in the Sasar area, both for soil and for plants, while the highest index for Zn was recorded in the Ferneziu area. Zn's high mobility, compared to Cu, was highlighted. Depending on the high content of Zn, it is assumed that vegetables grown in the soil of the Ferneziu area pose a potential risk to the health of consumers (Cristina et al.).

TRANSYLVANIA REGION

As we showed before, factors, like pH, electrical conductivity, organic matter content, etc., affect the mobility of metals in the soil and their bioavailability in plants. But the amount of heavy metals in vegetables also depends on their concentration in the soil. Large quantities of heavy metals, in particular Pb and Cd, were identified in the edible parts of the root vegetables harvested from Copşa Mică (Nedelescu et al. 2017, Damian et al. 2018). In Târnăveni, a pilot investigation was carried out, and high concentrations for total Cr (in the range of 15.6–525.8 mg/kg), Pb (in the range 25.4– 559.5 mg/kg), and Mn (in the range 363.1 to 1389.6 mg/kg) was reported in soil samples. Significantly higher levels of heavy metals were recorded closer to a

former industrial chemical platform that was also the source of contamination for soil and cultivated vegetables. Large metal concentrations were also measured in all samples of vegetables collected from the area (Mihaileanu et al. 2018). The metal concentrations in the Rusca Montana mining area were above the values measured in the unpolluted reference area, regardless of the metal analyzed and the type of sample (soil, plant products). Among the metals investigated, Pb and Cu were prevalent in the area. The values measured in soils exceeded the normal limits, reaching the intervention threshold values for Romania. These metals were also found at high levels in vegetables collected from that area, among them root vegetables have accumulated the highest levels of metals, followed by leafy vegetables and fruity vegetables. It has also been found that the consumption of root vegetables (carrot, onion, potatoes) and leafy vegetables (parsley leaves, cabbage, lettuce) from Rusca Montana is not without risks related to human health. The local populations are exposed to dangerous levels of metals, in particular Pb and Cu, by eating the vegetables (Manea et al. 2020).

MOLDOVA REGION

Researches were carried out in industrially polluted areas to determine the level of accumulation of heavy metals (Pb, Cd, Zn, Cu) in the tissues of some vegetable species. The results revealed that the heavy metal content in vegetable species used as raw materials in the canning industry (carrots, parsnips, parsley, tomatoes, cucumbers, peppers, etc.) is different depending on the areas in which they are grown. Thus, the results showed that raw materials from industrially polluted areas have high heavy metal content, often exceeding the maximum limits allowed by the national legislation (Buculei et al. 2016).

TOXICITY OF HEAVY METALS

Heavy metals are transported up to a few kilometers away from their sources (Shahid et al. 2017), and experimental studies have revealed that combination patterns in toxicity depend on the heavy metal mixture (Lin et al. 2016). Plants are under a continual threat not only because heavy metal contamination is a major environmental concern but they can restrict plant growth, can accumulate in plant parts resulting in decreased crop productivity and threats to animal and human health (Mustafa and Komatsu 2016, Shahid et al. 2015, Asati et al. 2016). Because metals cannot be decomposed, when the concentrations exceed the optimal levels in the plant, they negatively affect the plant and can even inhibit cytoplasmic enzymes and destroy cellular structures due to oxidative stress. The negative influence of heavy metals on the activities of soil microorganisms also indirectly affects plant growth. The toxic effects of heavy metals can lead to plant death (Asati et al. 2016). Easily, heavy metals can enter the food chain if there are extracted by vegetables from the soil and thus people exposed to different doses can get intoxicated. Heavy metals in food, even in small quantities, can pose a threat to human health, so various regulations have been adopted regulating acceptable levels for heavy metals content in food(Zwolak et al. 2019). Multiple studies have shown a positive association of Pb to high blood pressure and cardiovascular disease. The studies also revealed that Zn has a benefic role in the detoxification of heavy metal ions, such as Cd (Popa et al. 2020).

HUMAN HEALTH

Heavy metals occupy an important position on the list of hazardous substances. The toxicity arising from exposure to heavy metals usually affects several organ systems, the severity of reaction for human health depends on the type and shape of the element, the route, and duration of exposure (Jan et al. 2015).The amount of metals in the soil is an indicator of environmental pollution, and the increased absorption of metals by vegetables grown on such contaminated soils is reported in specialized studies around the world and from Romania (Nedelescu et al. 2012, Balkhair and Ashraf 2016, Buculei et al. 2016, Harmanescu et al. 2011, Zwolak et al. 2019). The toxicity of heavy metals for humans can lead to damage, or reduction of nerve cell functions, damage to the composition of the blood, lungs, kidneys, liver, and other vital systems and organs. Long-term exposure can lead to degenerative muscle diseases, slow physical development, or even neurological diseases like Alzheimer's, Parkinson's, muscular dystrophy, and multiple sclerosis (Pandey and Madhuri 2014, Morais et al. 2012, Nedelescu et al. 2012). Small amounts of essential metals like Mn, Cu, and Zn are necessary to avoid body's failure since their role as metalloenzyme is to act for the proper functioning of the body. Meanwhile, plumb and cadmium are toxic even at low concentrations. Pb can induce kidney tumors, reduce cognitive development, and increase blood pressure thus increasing the risk of cardiovascular disease in adults. Cadmium can induce kidney dysfunction, increase urinary excretion of Ca, P, and Pb and reproductive dysfunction. Also, Cu in high doses has been associated with liver damage, Zn reduces immunity and high density lipoprotein levels (Nedelescu et al. 2012).

Below we have extracted from the literature the effects that heavy metals accumulated in vegetables, from contaminated agricultural soils, can have on human health. They are described in Table 1.

Table 1. Various toxic heavy metals, their application and the effect on human health

Heavy metal	Applications	Effects on human health	Authors
Arsenic (As)	pesticides, treated wood product, coloring agent in textiles, mining, wallpaper, toy-making industries, insecticide, poison for mice, ashes from coal combustion, herbicides	visceral cancers such as those of the liver, kidney, lung, bladder, and skin, skin damage circulatory problems	(Mishra et al. 2019, Zhang et al. 2019, Mensah et al. 2020, Yin et al. 2019)
Cadmium (Cd)	fertilizer, metal industries, spoiled and wasted food, cigarettes, plastic industry	kidney damage, prostate dysfunction, bone diseases, cancer, lung damage, affect calcium regulation in biological systems	(Mishra et al. 2019, Shahriar et al. 2020, Suwatvitayakorn et al. 2020)
Chromium (Cr)	leather tanning, metal refining, textile dying, pharmaceutical drugs, inks and pigment, refractories, as a wood preserver, fungicide	cancer, nephritis, ulceration, hair loss, diabetes, nausea, headache, genetic damage	(Mishra et al. 2019, Yang et al. 2019, Zeinali et al. 2019, Manoj et al. 2020)
Cobalt (Co)	industrial mining, wood preservative metal and graphics industry, electronics and healthcare	diarrhea, low blood pressure, paralysis, impaired neuromuscular functions of skeletal, smooth, and cardiac muscle, muscular weakness, mental confusion	(Mishra et al. 2019, Khan et al. 2019b, van den Brink et al. 2020, Julander et al. 2020)
Copper (Cu)	pesticides, treated wood products, insecticide	brain and kidney damage, elevated levels result in liver cirrhosis, chronic anemia, stomach and intestine irritation	(Mishra et al. 2019, Radke et al. 2019)
Plumb (Pb)	Plastic, vehicles (auto exhaust), hair dyes, paints, glazing of pottery, pipe, batteries, gasoline, and enamelware	decrease in intelligent quotient, memory loss, infertility, mood swing, sterility, and risk of cardiovascular disease	(Mishra et al. 2019, Du et al. 2020, Huang et al. 2020)
Mercury (Hg)	thermometers and barometers, in instruments used to measure blood pressure, in amalgam for dental restoration, in fluorescent lighting, in production of caustic soda, in preservation of pharmaceutical products, in nuclear reactors, and antifungal agents for wood processing	produce gastrointestinal toxicity, neurotoxicity, nephrotoxicity, depression, drowsiness, fatigue, hair loss, insomnia, loss of memory, restlessness, disturbance of vision, tremors, temper outbursts, brain damage, lung and kidney failure	(Mishra et al. 2019, Khan et al. 2019a, Gyamfi et al. 2020)
Nickel (Ni)	plating industries, combustion of fossil fuels, Ni mining, tobacco smoke, jewelry, shampoos, detergents, coins, naturally occurs in food products and water and concentrations may increase by pollution	carcinogenic, embryo toxin teratogenic,	(Mishra et al. 2019, Francisco et al. 2019)
Zinc (Zn)	fertilizer, paints, rubber, cosmetics, pharmaceuticals, plastics, inks, soaps, batteries, textiles and electrical equipment	Dizziness, fatigue, vomiting, renal damage, and cramps	(Mishra et al. 2019, Du et al. 2020, Chen et al. 2020)

CONCLUSIONS

- It is essential to get a good understanding not only about the content and variability of heavy metals in soil profiles but also about sources of heavy metals in order to be able to monitor these areas and to limit the contamination of neighboring areas.
- In Romania many areas are polluted with heavy metals and these metals can bioaccumulate in vegetables of economic importance and they can pass into the food chain. It is necessary to develop a monitoring system for these areas, as well as a method for a more accurate assessment of the effects of these metals on human health.
- For humans, contaminated vegetables are one of the main sources of exposure to heavy metals, and an increased dietary intake of heavy metals can contribute to the development of various disorders. It is necessary to monitor the levels of heavy metals in foods of plant origin and their presence in human body.
- Long-term accumulation of heavy metals in soils leads to contamination of food crops and studies have shown that vegetables contaminated with heavy metals may contain higher levels than the values recommended by current legislation.
- Local vegetables varieties available in different parts of Romania have the ability to accumulate heavy metals and this is important for human health and food safety because a high level of heavy metals can present a toxicological risk to consumers.
- In the Moldova region, studies on the degree of vegetables contamination with heavy metals are few. Therefore, studies on the level of heavy metal contamination in vegetables are needed in areas affected by heavy metal pollution.
- Because the possibility of assimilation of heavy metals by vegetables is high, the competent

authorities in the field of food safety must determine new risk parameters regarding the amount of heavy metals in vegetables.

ABSTRACT

Heavy metal soil pollution is an important global environmental problem for many ecosystems. Romania is a country rich in mineral resources and has a long history in the field of mineral exploitation. Thus, the pollution with heavy metals of agricultural soils is a major environmental problem that can affect food quality and human health. Heavy metal contamination of vegetables is a problem that can lead to toxicity and disease in humans and animals due to their nutritional importance. The high level of soil contamination with heavy metals is dangerous because vegetables absorb and accumulate metallic elements in different tissues. In Romania, there are different sources of heavy metals contamination, especially from anthropogenic activities. The study's results from the last ten years show that both, the soil and the vegetables are affected by this type of pollution. Vegetables grown for consumption in Romania are sometimes contaminated with heavy metals, and this is associated with health problems that occur in the communities from those areas. This paper reviews the soil contents in heavy metals from three regions of Romania (Crişana, Transylvania, and Moldova). Therefore, the consumption of vegetables in areas affected by heavy metal pollution should be limited, and it is also necessary to regularly monitor areas where heavy metal pollution problems are identified.

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