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Environmental metal contamination and health impact assessment in two industrial regions of Romania

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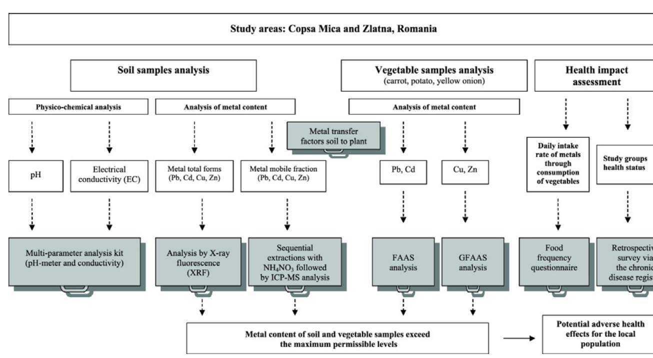
HIGHLIGHTS

- The pollution with heavy metals in two Romanian industrial areas is still alarming.
- High levels of Pb, Cd, Cu and Zn in soil and vegetables samples are revealed.
- Transfer factors show a great bioaccumulation of heavy metal in vegetables.
- Daily intake of Pb and Cd in resident population exceeds the EFSA limits.
- The need of immediate soil remediation actions is highlighted.

GRAPHICAL ABSTRACT



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ABSTRACT

We investigated two Romanian industrial regions– Copşa Mică and Zlatna, to assess the current situation of soil pollution and bioaccumulation of Pb, Cd, Cu and Zn in different vegetable species and possible risks to consumers. Both total and mobile forms of the metals were determined in soil samples, and metal content in the edible parts of root vegetable samples was also assessed.

The concentrations of Pb and Zn in soil were higher in Copşa Mică than in Zlatna (566 mg/kg vs 271 mg/kg for Pb and 1143 mg/kg vs 368 mg/kg for Zn)·The metal mobility in soil from Copsa Mica decreases in the order Zn > Cu > Cd > Pb (1.88 mg/kg, 0.40 mg/kg, 0.22 mg/kg, 0.16 mg/kg, respectively), while in Zlatna, the order was Cu > Zn > Pb > Cd (0.88 mg/kg, 0.29 mg/kg, 0.04 mg/kg, 0.01 mg/kg, respectively), apparently depending on metal and soil conditions. In Copsa Mica, the amount of Pb and Cd in vegetable samples exceeded the maximum permissible limits in carrots (median concentration 0.32 mg/kg for Pb and Cd) and in yellow onions (median concentration 0.24 mg/kg for Cd). In Zlatna region, the content of Cd exceeded the maximum limits in

Abbreviations: DIR, daily intake rate of metals; EC, electrical conductivity; Eh, redox potential; Hb, hemoglobin; FAAS, flame atomic absorption spectrometry; GFAAS, graphite furnace atomic absorption spectrometry; ICP-MS, mass spectrometry with inductively coupled plasma; PTDI, provisional tolerable weekly intake; PTWI, provisional tolerable weekly intake; rpm, rotations per minute; TF, metal transfer factor soil to plant.

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Transfer factor
Vegetables
Environment
Contamination

yellow onions (median concentration 0.11 mg/kg). The amount of Pb was higher than the maximum acceptable level in carrots from the Zlatna region (median concentration 0.12 mg/kg). Cu and Zn levels were within the normal range in all vegetable samples. In the Zlatna region, the transfer factors for Pb and Cd were higher in carrots (median values of 9.9 for Pb and 21.0 for Cd) compared to carrots harvested in Copsa Mica (median values of 4.0 for Pb and 2.0 for Cd). Daily intake rates of metals through local vegetable consumption exceeded the limit values established by the European Food Safety Authority for Pb (1.2 to 2.4 times) and Cd (5.5 to 9.1 times) in both regions, with potential adverse health effects for the local population. The results highlight the need for total soil remediation action before fruit and vegetables produced in these polluted areas can be safely consumed.

1. Introduction

Potentially toxic elements accumulate in the environment and affect the regional ecosystem (plants and animals, and through the food chain, humans) of the historically polluted industrial regions. High concentrations of metals in the environment may lead to the accumulation of these potentially toxic substances in the body, thus increasing the risk of chronic diseases such as cancer and neurodegenerative diseases (Jarup, 2003; Chang et al., 2014). Recent studies link exposure to these pollutants with lung and gastric cancer, Alzheimer and Parkinson's disease (Coon et al., 2006; Bakulski et al., 2012; Lee et al., 2012; Zhao et al., 2014). The concern has been primarily for the case of multi-pollutant exposure, with reports on the additive and sub-additive effects of exposure to Pb and Cd (Varaksin et al., 2014). Particularly vulnerable to metal exposure are children (Hough et al., 2004; Papanikolaou et al., 2005), especially the risk of neurological impairments.

In polluted areas, metal concentrations in different environments reflect the amount of historical pollution (Singh et al., 2010; Bekteshi, 2014; Golokhvast et al., 2015), and this is particularly true for contamination of soil and agricultural products in industrial regions (Cao et al., 2010). In such areas, soils tend to contain considerable amounts of metals even after the cessation of metallurgic activity, due to the persistence of these contaminants in the environment, including plants and locally grown food products and fish populations in aquatic environments (Violante et al., 2010; Sfakianakis et al., 2015). Hot-spots of contamination may occur even at distances of tens of km from the point source in the geomorphological setting of river corridors having large natural floodplains able to buffer the fluxes of highly contaminated soil and sediment fine particles originating from the industrial areas and transported downstream by surface water (Iordache et al., 2012).

Several studies have been carried out to assess potentially toxic elements content in vegetables and maize grown on polluted soils (Lacatusu and Lacatusu AR, 2008; Popa et al., 2011; Iordache et al., 2012). The amount of metals in vegetables depends on their concentration in soil and on the physicochemical 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 (Rodriguez et al., 2009; Kabata Pedias, 2011). The bioaccumulation of potentially toxic elements in plants is controlled by the type of plant-root system, and also by the plant response to potentially toxic metals as a function of the seasonal cycles (Neagoe et al., 2012).

In order to assess the impact of potentially toxic element pollution on population health it is important to quantify the amount of metals ingested from different sources. The amount of potentially toxic elements in the atmospheric air of historically polluted regions significantly decreased as a result of the efforts to reduce the emissions and the measures taken to close the factories (National Environmental Protection Agency of Romania, NEPA, 2010–2014). The current diet based on locally produced food from both vegetable and animal sources, represents the major pathway of population exposure. Consumption of foods of vegetal origin, in particular, represents the main exposure route to metals due to their bioconcentration from soil to plants (Sharma et al., 2008; Kabata Pedias, 2011; Beccaloni et al., 2013). The metal transfer and its bioavailability from soil to plants and through the food chain to the exposed population is an important key factor for the health impact assessment of these substances. The understanding of these processes contributes to approaches taken to establish recommendations for reducing the consumption of local vegetables in the historically polluted industrial areas pending a complete remediation (Adamo et al., 2014).

Polluted sites across Europe are the subject of large-scale integrated research designed to support their management (Levonton et al., 2016). Zlatna and Copşa Mică are two such sites in Romania where there is transnational research (Iordache et al., 2012, Weindorf et al., 2013, Paulette et al., 2015). Beside their importance as model systems at ecosystem scale, both sites are also relevant to potential transboundary transport of metal fluxes.

In this context, our study aim was to determine the concentrations of four potentially toxic elements (Pb, Cd, Cu and Zn) in soil and vegetables grown on polluted soils in these two regions, and to assess the health risk associated with exposure to these elements.

2. Methods

2.1. Description of the studied areas

Copşa Mică is a small town located in the North West part of Sibiu County (geographical coordinates 46°6′45″ N and 24°13′5″ E). This town was considered "one of the five most polluted industrial sites of the communist world", as reported in the Atlas of Our Changing Environment (United Nations Environment Programme, 2005). Zlatna is a city located in Alba County (geographical coordinates 46°09′32″ N and 23°13′16″ E). Both regions are located in the Transylvania region of Romania, and suffered intense environmental metal pollution as a result of the extraction and processing of the non-ferrous ore industry. Zlatna was well known since Roman times for its importance in the extraction of Cu from the Metalliferous Mountains in the Apuseni mountain chain. In Zlatna, a large smelter worked from 1747 until 2004, when it was closed. This metallurgical plant was originally equipped with six furnaces for extracting Cu and Pb from ores rich in gold and silver. During the Revolution of 1848, the smelter was destroyed, but after 1850 the factory was rebuilt, though the process was modified by applying the chlorinated roasting sulphide ores. In the mid-twentieth century, the platform from Zlatna worked at its maximum capacity and discharged into the atmosphere considerable amounts of potentially toxic elements (Pb, Zn, Cu, Cd and Sb). The Copşa Mică metallurgical company was established in 1939–1940 and included equipment for Zn and Pb extraction, recovery of Cd and other metals such as Bi, Au, Ag and Sb. In 1998 the factory entered a phase of ecological rehabilitation, and currently the activity is carried out only for the recovery of metals from the equipment.

Both study areas were affected by the operation of the two industrial units, which induced extensive environmental contamination (air, water and soil pollution). The soils from these areas belong to the Aluviosol type according to Damian et al. (Damian et al., 2008) and the FAO/UNESCO classification. These soils are physically and chemically characterized by the same authors as can be seen in Table 1.

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