Accumulation of As and Pb in Vegetables Grown in Agricultural Soils Contaminated by Historical Mining in Zacatecas, Mexico

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Research Article

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Abstract

Historical mining activities are a source of environmental pollution that affects the food chain and the health of human beings. The aim of this study was assessment the accumulation of arsenic and lead in vegetables grown in agricultural soils contaminated by old mining in Zacatecas, Mexico. The concentration of arsenic and lead in agricultural soil and edible parts of carrot, garlic, and pepper was analyzed by atomic absorption spectrometry. The soil-vegetable bioconcentration factor and pollution load index were determined. The pH values of the farmland were alkaline. The concentration of arsenic in agricultural soil exceeds the permissible limit for arsenic of Mexican standards and international representing health risks. The lead content in most soil samples they were low. The arsenic and lead content in edible parts of species vegetable exceeded the national standard from various countries and the values established by the Codex Alimentarius (FAO-WHO). The highest arsenic concentration was found both in Capsicum annum and Allium sativum. The highest concentration of Pb was in pepper fruits. Among vegetable the high BCF value was for arsenic, ranging from 2.33 to 0.64, and the average for all vegetable samples was 1.01. The pollution index indicates that arsenic is the dominant pollutant accumulated in soil and vegetables grown in agricultural soils. According to the findings, the state and national agricultural and health authorities should not recommend the cultivation of vegetables in agricultural soil located in this area of historical mining activities. Likewise, preventive measures must be taken on the consumption of contaminated vegetables and certifying their safety.

Keywords Historic mining waste • Arsenic • Lead • Vegetables • Pollution load index • Bioaccumulation factor
**Introduction**

Soil contamination by heavy metals is one environmental problem raising critical concern to human health and ecosystems (Kabata-Pendias and Pendias 2011). Mining activities are well known for their damaging effects on the environment due to the deposition of large volumes of wastes on the soil (Ashraf et al. 2011). One main concern regarding historical mining activities is the control of mine wastes that contain large quantities of metals (Coob 2000). Accordingly, due to rapid industrialization and coal production, the geochemical composition of the soil is greatly changed at nearby mining areas (Du et al. 2018).

Mexico, due to its long mining history, millions of tons of tailings are abandoned and dispersed in the country, whose conditions and potential for affecting the environment are still unknown (Ramos-Arroyo and Siebe-Grabach 2006). In particular, the opencast coal mining method associated with generated millions of tons of sulfide-rich tailings, large quantities of emissions (dust and gases) (Bhattacharya et al. 2006; Masto et al. 2011).

Additionally, during mining operations, waste heaped of mines abandoned, and tailings are deposited inappropriately, with a scarce layer of the original land, without reforestation programs and lack of protective systems (Salas et al. 2017) and without any continuing management (Salari et al. 2012). These wastes usually are deposited on the ground that occupies large areas of the land surface (Conesa et al. 2007). The tailings generate spoils, effluents, and dust with outstandingly large concentrations of metal elements and metalloids (Wiegleb and Felinks 2001). In this way, extractive mining activities degrade the environment affecting the health of the inhabitants living in the surrounding vicinity. This problem is even getting more serious all over the world, especially in developing countries (Bagdatlioglu et al. 2010).

On the other hand, the spread of fine particles has caused damage to ecosystems, contamination of adjacent agricultural soils, deterioration of food chain and economic and social injury, and posed severe health risks to humans and grazing animals (Clemente et al. 2007; Martínez-Sánchez et al. 2012). The accumulation of heavy metals and metalloids in agricultural is of increasing concern nowadays (Jolly et al. 2013). Arsenic is slowly mobile in contaminated soils
and, near-surface soils can be redistributed by tilling, burrowing animals, or overland runoff (MDEP 1995; Barringer et al. 2001). While lead is generally immobile in soil; therefore, it accumulates in the top layer of soil (Berglund et al. 2000) and can remain in the soil for thousands of years (Kumar et al. 1995).

On the other hand, fresh vegetables and fruits are vital to our diet because they contain essential nutrients by the human body and for human health, such as carbohydrates, proteins, minerals, vitamins, and trace elements (Itanna 2002; Zhong et al. 2018). Also, have multiple health benefits are rich fibers, antioxidant and medicinal properties. The carrot (*Daucus carota*) name of its carotenoids originates from the massive accumulation in the root (Klein and Rodriguez-Concepcion 2015). *Capsicum annuum* is considered the second most vegetable of the world after tomato and used as spices in various cuisines (Aluko 2016). Pepper its main ingredient is capsaicinoids and source of vitamins A (ascorbic acid), B and C (carotenoids), polyphenols, phosphorus and calcium (Andrews 1995; Topuz and Ozdemir 2007). Garlic (*Allium sativum*) contains more than 70 organosulfur compounds (Randle and Lancaster 2002). Vegetables uptake heavy metals from contaminated soils and deposits on parts of the vegetables exposed to the air from polluted environments (Wang et al. 2005). Vegetables cultivated in contaminated soils uptake heavy metals in large quantities enough to cause potential health risks to the consumers (Yang et al. 2007; Sihgh et al. 2010).

In this way, ingestion of contaminated suspended tailings and soil particles, ingestion of crops grown on mine tailings contaminated soils or, deliberate ingestion of the tailings contaminated soils (geophagia) (Ngole-Jeme and Fantke 2017). The continuous addition and accumulation of heavy metals in the soil significantly increases their transfer to the food chain (Hussain et al. 2019). The food chain contamination is the main pathway of heavy metal exposure for humans (Khan et al. 2008). Heavy metals produced by mining activities represent a potential threat to the environment and can harm human health through various routes such as inhalation, dermal contact, oral ingestion, or ingestion (Komárek et al. 2008; Rout et al. 2014). It should be a highlight, toxic elements such as arsenic (first) and lead (second) stand out among the CDC rank substance priority list pose the most significant potential threat to human health (ATSDR 2021). Long term health effects of exposure to As are skin and lung cancer, kidney disease,
hypertension, cardiomyopathy, damages liver, bladder, lymphatic system and neuropathy (Vahidnia et al. 2007). While the complications related to Pb toxicity are damage to the nervous system of children, such as intellectual deficits, neurological damage, cognitive dysfunction, neurobehavioral disorders, and encephalopathy. Also, hypertension, renal impairment, abdominal colic. Probably, cancer and anemia even at relatively low blood levels and, in extreme cases, death (Lanphear et al. 2005; Patrick 2006; Flora et al. 2012).

So, it is essential to monitor food quality, given that plant uptake is one of the main pathways through which heavy metals enter the food chain (Antonious and Kochhar 2009). Nowadays, increasing food demand and security are of great concern throughout the world due to toxic metals contaminated foodstuffs and their associated health risks (Rehman et al. 2017; Nawab et al. 2018). Thus, vegetables cultivated in soil pollution without restrictions environmental and inefficient mining activities may accumulate heavy metals to levels above those normally expected. Therefore, information about toxic element concentrations in vegetables is essential for assessing the potential risks to human health and ecological systems. The aim of this study was assessment the accumulation of As and Pb in vegetables grown in agricultural soils contaminated by old mining in Zacatecas, Mexico.
Materials and Methods

Description of the study area

The study was conducted agricultural soils of four community rural El Bordo (22°54’34’’ N, 102°24’45’’ W), El Lampotal (22°54’43’’ N, 102°24’10’’ W), La Era (22°51’17’’ N, 102°25’03’’ W) and Santa Rita (22°54’42’’ N, 102°25’06’’ W) located within a region rich in natural deposits of arsenic and lead, and contaminated since colonial and postcolonial times, from Guadalupe and Vetagrande municipalities of Zacatecas in Mexico (Fig. 1). Soils in cultivated area are mainly classified as haplic xerosols are dominants in the valleys, eutric fluvisols are found along the streams, and lithosols in mountains (FAO/UNESCO 1975). This region presents a temperate arid tropical climate with an average temperature of 16.6 °C, annual precipitation that varies from 400 to 500 mm and, altitude ranges from 2,000 to 2,300 m above sea level (Medina et al. 2009).

HERE GOES FIGURE 1

Historic mining and origin of the soil pollution

In Zacatecas, after discovery of silver veins in 1548, its extraction was carried out in haciendas of benefit through of the “amalgamation method” from 1570 to 1820 (Santos-Santos et al. 2006). Mine wastes generated were placed on the shores of streams of the mountains and for centuries dragged by the rains toward channels of the “Plata arroyo” which leads into in the dam “El Pedernalillo” in community La Zacatecana. As well as those wastes dragged for centuries through the mountains towards the valley of the municipalities of Guadalupe and Vetagrande, Zacatecas. In this way, previous studies have reported that agricultural soils of this region are contaminated and have high concentrations of As, Au, Pb, Cd y Hg, and high-grade silver (Flores 2003; Ogura et al. 2003; Santos-Santos et al. 2006). Between 1920-1930, through a variation of the Russell process (Stetefelt 1895) processing activities began to recover Ag, Au
and other metals from tailing wastes. Nowadays, agricultural soil and subsoil that contain millions of tons of alluvial wastes are processed by a company. The amalgamated sediments were deposited in two abandoned tailing and a tailing still in operation, which generates dust emissions and dispersion of heavy metals and generally without any control, becoming a source of contamination around agrosystems and nearby communities with a total number population is proximally 6,500. Nowadays, these lands are mainly used for agriculture and there are no restrictions imposed by agriculture and environmental Mexican agencies.

**Soil and vegetable sampling**

To obtain a representative average sample for each crop selected, surface soil and vegetable samples were collected randomly. These vegetables are part of the food chain for the own consumption of the inhabitants, supply, and commercialization to retail and wholesale country markets. At each sampling site, soil samples were taken from the upper 0-25 cm of the profile representing the rhizosphere and arable layer, and 10 random subsamples were collected in a restricted circle about 60 cm around the species a distance of about 20 m each from the first sub-sample. A total of 9 composed samples equivalent to 5 kg thoroughly mixed collected to form a representative surface sample for analyses. The rhizosphere soil samples were isolated from the roots and vegetative organs shaking, them in a plastic bag. Soil samples brought to the laboratory were mixed and left dried naturally in clean plastic trays for a week at ambient temperature (26°C) followed by an oven-dry until constant weight obtained. Then samples stored in polyethylene bags, until used for acid digestion.

All vegetables at the stage of maturity and for fresh consumption were collected before harvest. Vegetable composed samples (about 5 Kg) were collected into polyethylene bags jointly where the soil samples. Individual crop samples include tissue collected from 4 locations within each sampling site to obtain representative samples and as well as to obtain sufficient dried tissue (0.5 g) for analysis. Vegetable samples were mixed and washed thoroughly with tap water twice; the dust and soil particles adhering to the carrots were removed but not peeled. The rest of the particles adhering to the plant surfaces were then removed with distilled water, followed by deionized water, and dried with tissue paper. The edible parts of all vegetable samples were
cut into small pieces, fresh tissue was weighed separately and recorded, and then were heated in an oven at 70°C for 48 h to a constant weight. Samples ground using a porcelain mortar and then a stainless-steel mill, mixed and sieved a 2-mm mesh and re-dried until weights again, and stored in plastic bags at ambient temperature before digestion (McBride et al. 2015).

**Physical-chemical parameters of agricultural soil**

The <2-mm fraction of composite samples soil was used to determine the physicochemical properties. Soil particle-size composition (sand, silt, and clay) was determined using the micropipette method (Miller and Miller 1987). Soil pH was measured in a soil paste saturated with deionized water at a ratio of 1:2.5 using a glass electrode (McLean 1982; McCauley et al. 2017). The electrical conductivity (EC) of the soil was measured using a conductivity meter on an extract of soil obtained by shaking soil with deionized water at a 1:1 (w/v) soil: water ratio (Janzen 1993). Organic matter content was determined by the Walkley and Black method (1934). All parameters were determined by triplicate.

**Sample preparation and analysis**

Separately, representative samples for both soil and vegetable for digestion procedures were taken. All digested samples were diluted to 50 ml with 0.5 % HNO₃ and stored at 4 °C until analysis of As and Pb concentration. Then, a sample of fine powder (0.5 g) was weighed for digestion (Cao et al. 2014) using a microwave oven Microwave 3000, Microwave Reactor System (MARS), digestion techniques following the standard Method: SW 846: 3050AB (US. EPA 1996a). The total soil Pb and As concentrations were measured in composite samples using atomic absorption spectrometer (AAS, model 800) Perkin Elmer, according to previously described EPA Method: SW 846: 3050/6010B three replicates by crop were analyzed (US. EPA 1996a). Procedures used to ensure precision and accuracy in the measurements of Pb and As in the soil and vegetable included the use of Standard Reference Materials (SRM) of certified plant and soil standards from the National Institutes of Standards and Technology (NIST) USA, were analyzed in the same procedure at the start, during analysis, and at the end of the measurements to ensure continued accuracy, as well as laboratory internal standards along with
duplicate samples and blanks in the sample set. Trace reagent analysis grade reagents were used.

**Pollution load index**

The suitability of soils for agricultural uses can be further assessed by using the pollution index which assesses the environmental risk caused by the contaminated soils. The pollution load index expressed as the single index method can be calculated as follows:

\[
P_i = \frac{C_i}{S_i}
\]

where \( P_i \) is an environmental quality index for heavy metal \( i \); \( C_i \) is the heavy metal content in a soil sample (mg kg\(^{-1}\)); and \( S_i \) is the permitted standard of the same metal (mg kg\(^{-1}\)). Where \( P_i > 1 \), the soil sample is classified as polluted, while \( P_i \leq 1 \) suggests unpolluted soil (Li et al. 2006).

**Soil-vegetable bioconcentration factor**

The bioconcentration factor (BCF) is defined as the ratio of metal concentration in shoots to that in the soil (Bui et al. 2011; Chang et al. 2014). The bioconcentration factor (BCF) was calculated as follows:

\[
BCF = \frac{\text{Heavy metal concentration in edible parts of vegetables}}{\text{Heavy metal concentration in the soil}}
\]

Where \( C_{\text{vegetable}} \) is the total concentration of a particular heavy metal contained in the vegetable (mg kg\(^{-1}\)), and \( C_{\text{soil}} \) represents heavy metal concentration in the soil habitat of the vegetable (mg kg\(^{-1}\)) on dry weight basis, respectively (Chang et al. 2014; Jolly et al. 2013).
Results and Discussion

Physicochemical parameters of agricultural soil

The results of the physicochemical analysis of the study area are given in Table 1. The values of the agricultural soil samples from all the sites were alkaline, with pH ranged in a narrow interval from 7.7 to 8.5. The EC found ranged from 1.52 to 4.94 with a mean of 3.2 dS m\(^{-1}\), while the OMC oscillated from 2.0 to 3.3 and a mean of 2.8\%. As it is observed in Table 1, the soil samples collected from the northern sites displayed the highest EC value, and south sites had the lower value. However, the OMC value in the soil samples was higher in both the northern and southern sites. Analysis of the soil textural the relative percentages of sand, loam, and clay was in the range (37-65\%) for sand, (22-38\%) for loam, and (13-39\%) for clay. According to the USDA textural triangle soil textural class reveal that in the region samples showed a certain degree of homogeneity with a predominance of clay sandy loam, sandy loam, and clay loam. In general terms, the pH of the soil analyzed was close to 8.0 at all sites. In this regard, Merry et al. (1986) stated that increasing soil pH decreased Pb concentrations in vegetable crops. Since Pb is relatively immobile and As very slowly leaches through soils (Hood 2006). In this context, Alam et al. (2003) suggest that the relatively neutral soil pH (7.6–8.5) arsenic will be immobile in the local soil profile. Therefore, the alkaline range of soil (>8.0) restricts the mobilization of heavy metals, thus reducing their uptake and transference from soil to crops (Cheng 2007; Sharma et al. 2007). The EC value indicates soil salinity. Horneck et al. (2011) reported that soil with EC values less than 1 mS cm\(^{-1}\) is suitable for crop production. Although, values of EC showed a normal range (1.52) to slightly saline (4.94) and vegetables did not show symptom by salinity.

Arsenic concentration in agricultural soil

The analysis of heavy metal concentration in agricultural soil is shown in Table 1. The elements toxic in all the sites were detected, the concentrations were in the following order: Pb> As>. In this study, a high percentage was of arsenic (100\%) of samples with a concentration above several guideline values set for agricultural soils show that the land of the region is polluted.
The concentration of As in soil from all the sites ranging from 39.02 at 165.00 mg/kg$^{-1}$, an average of 109.22 mg/kg$^{-1}$ did exceed the critical level (>20 mg/kg$^{-1}$), Mexican standard, for agricultural soil and residential is far superior (NOM-147-SEMARNAT/SSAI-2004). Likewise, the maximum acceptable limit for agricultural soil of 20.0 mg kg$^{-1}$ as recommended by the European Community (Rahman et al. 2007). Levels exceeded different guideline values for agricultural, residential, and commercial land uses U.S. EPA (1993). Also, the concentrations are higher and surpassed environmental critical limit concentrations of the WHO permissible limit for arsenic in agricultural soils is 0.5 mg/kg$^{-1}$ (WHO 2004). Accordingly, total As represents a potential source of possible risk through bioaccumulation and consumption of vegetables. Because the concentration of As in soil in the area is much higher than the reported global average of 10.0 of 10.0 mg kg$^{-1}$ (Das et al. 2002). Norton et al. (2013) found SW field survey soil in horticultural produce grown in the impacted mining region the average As concentration was 110.3 mg kg$^{-1}$.

In Mexico, previous studies found a range of arsenic similar to that of this work. For example, Santos–Santos et al. (2006) and Gonzalez et al. (2012) showed that As concentration all soil valley of the agricultural region of Guadalupe, Zacatecas, average 109 mg/kg$^{-1}$. However, Mendoza-Amezquita et al. (2006) found that As concentration contaminated soils in the old mining region of Guanajuato, Mexico, ranged from 21-36 ppm. Although, differ significantly from the concentrations reported in other studies from an old mining area in Zimapan, Hidalgo, Mexico, 2,550-14,600 ppm (Ortega-Larrocea et al. 2010). Furthermore, in the present study, As concentrations were significantly higher than those found in other studies in agricultural soils around and near sites with mining activities (Baig and Kazi 2012; Rahama et al. 2013; Alam et al. 2016; Bui et al. 2016). Consequently, our results corroborate other findings of waste mining, which reported elevated levels of heavy metals in the farmland around of mining area (Zhuang et al. 2009a).

Also, is consistent with the previous fact that highlights that historic metal ore mining is considered one of the most important sources of soil contamination (Dudka and Adriano 1997). Similarly, our results confirm comparable As values than those of literature reported in vegetables (Cao et al. 2009; Liu et al. 2010; Chang et al. 2014; Tasrina et al. 2015). However,
arsenic levels observed in horticultural crops soils in this study were lower than those found in contaminated agricultural soils (Filippi et al. 2004; Chakraborti et al. 2013).

**HERE GOES TABLE 1**

**Lead concentration in agricultural soil**

Likewise, lead concentrations of agricultural soils in this study were low in most of the soil samples from four Communities, except at El Bordo and La Era (Table 1). These levels show that contamination by Pb was not very extensive. However, maximum and minimum concentration for Pb was 1206 and 25 mg/kg$^{-1}$, respectively, and mean concentration was 355.4 mg/kg$^{-1}$. Besides, not exceed the acceptable levels for Pb in the agricultural soil of Mexico (400 mg/kg$^{-1}$) (NOM-147-SEMARNAT/SSAI-2004). Nevertheless, this value is comparable to the maximum permissible Pb concentration in soils for agricultural purposes, being 375 mg/kg$^{-1}$ (OECD 1993). Although exceeds standard values ÖNORM L 1075 (Austrian standard L 1075) 100 mg/kg$^{-1}$ defined as the limit for agricultural and grassland soils (ÖNORM 1990). Also, exceed 200 considered ecological risks but is below 700 that represent health risks (Ministry of Environment of Finland 2007; Tóth et al. 2016). Likewise (Kabata-Pendias 2011) reported that the range and the mean world content of lead in soil are 3–90 and 27 mg kg$^{-1}$, respectively. Also, that to name a contaminated soil Pb this must exceed 100 mg/kg.

In this regard, the concentration of Pb was two times higher than the permitted standards in El Bordo (1201.4 mg/kg$^{-1}$) and La Era (1205.8 mg/kg$^{-1}$) samples exceeded the maximum limits of 400 mg/kg (WHO 1993). Besides, no evidence of phytotoxicity was observed from the levels of Pb in the sites studied. Therefore, the soil is unsuitable for agricultural use. In Mexico, previous studies different of lead concentrations were reported in diverse crops soils of Valle of the Mezquital, Mexico (22.86 mg/kg$^{-1}$) (Prieto-Garcia et al. 2007); in agricultural soils near mining regions of Guadalupe, Zacatecas, Mexico (100 and 400 mg/kg$^{-1}$), respectively (Santos–Santos et al. 2006; Gonzalez et al. 2012); Yaqui and Mayo agricultural valleys, Sonora, Mexico...
(10-56 mg/kg⁻¹) (Meza-Montenegro et al. 2012) and rural community in Fresnillo, Zacatecas, Mexico (4940 mg/kg⁻¹) (Salas and Vega 2016). On the other hand, in this study, Pb concentrations were significantly higher than those reported in contaminated soils (Sharma et al. 2007; Saint-Laurent et al. 2010; Gałuszka et al. 2015; Kumar and Maiti 2015). Thereby, our results corroborate other studies on mining activities, which also reported that elevated level of lead in soil was ubiquitous in the vicinities of mines (Zhuang et al. 2009b; Luo et al. 2011). Likewise, levels of Pb in vegetables found in this study were comparable to those found by Codling et al. (2015), Eissa and Negim (2018); Ćwielag-Drabek et al. (2020). However, in this study, Pb concentrations were significantly lower than those found in agricultural soils near mining areas (Koleli and Halisdemir 2005; Gisbert et al. 2006; Girisha and Ragavendra 2009; Chu et al. 2019).

In this way, concentrations of As and Pb in the agricultural soil showed heterogeneity and irregular distribution, indicating contamination, not uniform, and a strong influence by historical mining in the selected sites. These findings could have caused variability from layers of soil enriched with metals of different concentrations accumulated in the sediments originated by the historical mining that formed the topography of the agricultural valley. Because due to redistribution of the heavy metals in farmland during centuries by wind and water erosion processes. Resulting in a variable zone of enrichment and contamination of As in most of the farmland soils and lower risk of lead in sites studied. Therefore, our results agree with those reported by Ha et al. (2011) reported that high heavy metal concentrations soil has been continuous dispersal downstream from the tailings mining. Renshaw et al. (2006) showed that erosion of contaminated soil is significant in the dispersion of As and metals within drainage basins. According to Nedelescu et al. (2017), a detailed analysis of the distribution of metals in a site requires a higher number of locations and samples in order to understand the possible patterns of this large heterogeneity.

**Arsenic and lead concentration in vegetables**

In 1993, the World Health Organization reduced its recommended criteria for drinking water from 50 to 10 mg of arsenic/L (WHO 1984; 2006). Nevertheless, the WHO evaluates its quality
and provides international norms for water quality used as the basis for regulation and standard settings targeted for the protection of human health. These guidelines are adopted by many countries as national guidelines, even if they are not necessarily enforceable by law (UNEP 2008). Also, the regulations related to arsenic levels in food are complex, which varies in each country and over time (Henke 2009). Likewise, this is reflected in the values established by the Codex Alimentarius for various products (FAO-WHO 2019). Although WHO and Codex Alimentarius are not regulators the power to set standards and enforce them, its recommended criteria regarding arsenic are included into the regulations of various countries (WHO 2006), among them Mexico.

**HERE GOES FIGURE 2**

The average concentration of total arsenic and lead (mg kg\(^{-1}\) dw) in the selected vegetables: *Allium sativum* L., *Capsicum annuum* L. and *Daucus carota* L. are listed in Table 2. According to their edible parts, vegetables were classified into two categories: root vegetable (carrot and garlic) and fruit vegetable (pepper). The results show variable levels of As and Pb in each vegetable. Also showed accumulation factor and agricultural soil degree of contamination. Arsenic concentration between root vegetable and fruit vegetable they were similar, fruit vegetable average was higher (95.66 mg kg\(^{-1}\) dw) and root vegetable (92.33 mg kg\(^{-1}\) dw). Whereas lead concentration between root vegetable and fruit vegetable varied, fruit vegetable average was higher (9.6 mg kg\(^{-1}\)). Although in two samples were not detected. While lead concentration in roots vegetable was 4.8 mg kg\(^{-1}\) but in one sample was not detected. Between the garlic, carrot, and pepper, we find highest arsenic concentration in pepper was 111 mg kg\(^{-1}\) site 9, and lead concentration was the highest carrot 9.9 mg kg\(^{-1}\) site 5.

Arsenic showed with greater clarity high concentrations in the samples collected inside the contaminated soil. In this context, all edible parts of species vegetable As concentrations far exceeded the maximum limits permitted of Latin American countries. For example, the maximum level allowed by Chilean legislation for cereals, legumes, and leguminous plants (1
µg g\(^{-1}\) of ww) and the food group “other solid products” (Muñoz et al. 2002). The limits established by Argentine Food Code (1 mg/kg) (CAA 2020). The maximum limits for inorganic contaminants in food proposed by the Brazilian Ministry of Health (limit: 1 mg kg\(^{-1}\)) (MS 2013). Likewise, the limit of 1.0 mg kg\(^{-1}\) (ww) for the classification of the concentrations for edible plants in Mexico (Osuna-Martínez et al. 2021). Also, our results on the analyzed concentrations of As in vegetable were significantly higher than maximum levels set in EU legislation, as well as by Codex Alimentarius-FAO/WHO standards for cereals and vegetables (FAO/WHO 2001). Likewise, our results far exceeded the maximum limits to Europe permissible limits for As in fodder crops is generally between 2 and 4 mg of As per kg (moisture content 12%) (Gulz et al. 2005); for food plants, the statutory limit for total arsenic in edible fruits and vegetables is set up at 1 mg As kg\(^{-1}\) fresh weight exists, e.g., in Spain (BOE 1978; Lario et al. 2002) and other European countries (Feldmann et al. 2000). Australian and New Zealand Food Standards Code (FSANZ 2019) includes to MPL level in cereal grains and milled cereal products of 1.0 mg kg\(^{-1}\) total As. The China food safety quality standard for a maximum of contaminants in foods (GB 2762-2012), exceed the Chinese standards for total As 0.5 mg kg\(^{-1}\) (f.w.) in vegetables (MHPRC 2013). The Centre for Food Safety (2018) of the Hong Kong Government has established an MPL of 1.4 mg kg\(^{-1}\) (ww) total As in all foods. Based on these maximum limits permitted of arsenic concentrations vegetables, in our study were significantly higher to found in vegetables grown in an area highly contaminated with As (Bhattacharya et al. 2010). Likewise, Bui et al. (2016) As concentrations reported in vegetables grown near Bac Kan mining area in Northern Vietnam. As well as Li et al. (2017) arsenic the level in leaf vegetables collected from a nearby mining area in China. Also, our results were higher than average As concentration vegetables in a severely As-contaminated area in Bangladesh (Rahman et al. 2013).

On the other hand, Pb concentration in all edible parts of species vegetable showed levels with ranges from 2.06 – 9.82 mg/kg, this was above the maximum permissible limit recommended for the comparison of Pb (standard level of 0.1 mg/kg in root and tuber vegetables) by EU (2006) and Food and Agricultural Organization (FAO)/World Health Organization (WHO) CODEX (2011). As well as the maximum permissible levels to Pb by the Chinese Ministry of Health (CMH 2005). In the same way, exceed the maximum limits acceptable Chilean standards
food as 0.5 μg g⁻¹ (Gonzalez 1998) and maximum limits established (0.1 mg/kg) by MERCOSUR (2014). In Mexico, although it exists an official standard NOM-117-SSa1-1994, it does not mention the permissible limits take as reference to the standards established by the Codex Alimentarius and the European Union our results show high concentrations of lead in the vegetable. Therefore, Pb concentration in vegetables grown in the area impacted by mining activities was higher than those reported in various countries, for example study realized by Alam et al. (2003), Lăcătușu and Lăcătușu (2008), Harmanescu et al. (2011), Zhuang et al. (2009b) and Islam et al. (2016). Therefore, Pb concentration in vegetables grown in the area impacted by mining activities was higher than those reported in various countries, for example, studies realized by Eissa and Negim (2018), and Roba et al. (2016). In this respect, Kabata-Pendias and Pendia (2011) highlighted that forms of the anthropogenic trace metals, availability to plants are higher than those of natural origin. Likewise, the standard concentration of Pb in plants is 0.1-10 mg/kg. Nevertheless, exceeded diverse food safety limit and vegetable safety limits from different countries. In the present work, differences observed in As and Pb concentrations could be due to variable uptake capacity of the crops and accumulation of toxic elements (Pandey and Pandey 2009).

**Arsenic accumulation in vegetables**

The arsenic concentration in vegetable species are shown in Table 2. In general terms, As concentration in vegetable recorded similar concentration from the nine sites of the four Communities studied. These varied between 89 ± 9.7, 91 ± 8.7, 90 ± 9.1, 105 ± 12.5 mg kg⁻¹ dw, respectively. The arsenic concentration average of vegetables was 93.44 mg/kg⁻¹ value was higher than the maximum limits established by international standards (1.0 mg/kg⁻¹), those permitted in many countries, including accepted in Mexico (Osuna-Martínez et al. 2021). The highest As concentration were found both in *Capsicum annum* and *Allium sativum* (111 ± 11.97 and 100 ± 13.04, respectively, site 9 from Community La Era). The Communities of Santa Rita, El Bordo, and Lampotal registered similar concentrations, with an average of 90 ± 9.09 mg kg⁻¹ dw, in all the sites.
The As concentration soil samples average of 109.22 mg/kg\(^{-1}\) which did exceed the critical level (22 mg/kg\(^{-1}\)) Mexican Standard for agricultural soil (NOM-147-SEMARNAT/SSAI-2004) and permissible limit for arsenic in agricultural soils 0.5 mg/kg according to WHO (2004). These results indicated that vegetables accumulate arsenic. Therefore, this implied that the vegetables that are grown in these soils absorb more arsenic. Plant arsenic concentration tends to increase with increasing soil arsenic and then stabilize at some maximal value at higher concentrations in soil (Tasrina et al. 2015). Thereby, McBride et al. (2015) indicated that vegetable As concentration increase with increasing soil total As. Arsenic (As) uptake by vegetable is related to its concentration in the respective soil (Ramirez-Andreotta et al. 2013; Khan et al. 2018). Elevated concentrations of As, were higher than the FAO/WHO permissible limits, shower this element toxic might represent risk by consumption these vegetable species.

**Allium sativum:** In the present study, the As concentration observed in garlic bulbs was 87 mg/kg, and all vegetables were the lowest concentration. These results showed that all vegetable samples exceeded the maximum concentration of arsenic recommended by EU (2006) and WHO/FAO (2011). Additionally, our results revealed that As concentration was higher than those found in previous studies on As in garlic bulbs. For example, Muñoz et al. (2002) in Northern Chile (0.238 µg g\(^{-1}\)); Bhattacharya et al. (2010) in West Bengal, India (0.126 mg/kg); Alam et al. (2016) in India (0.25 mg/kg) and Rehman et al. (2016) in Pakistan (0.04 mg/kg) and Aguilar et al. (2018) central Chile under the limit of detection. **Daucus carota:** Our study average arsenic concentration observed in carrots was 93.4 mg/kg. These results showed that exceeded the maximum concentration of arsenic recommended by EU (2006) and CODEX (2011). Thereby, As concentration was higher than those found in previous studies on As in carrots. For example, Muñoz et al (2002) in Northern Chile (0.132 ± 0.001 µg g\(^{-1}\)); Garcia-Rico et al. (2012) in Mexico (0.041 mg kg\(^{-1}\)); Rahaman et al. (2013) in West Bengal, India (0.235 ± 0.004 mg/kg); Tasrina et al. (2015) in Bangladesh (<0.1 mg/kg) and Zhou et al. (2016) in China (0.188 ± 0.030 mg/kg). **Capsicum annuum:** According to the results obtained average arsenic concentration observed in pepper fruits was 95.6 mg/kg\(^{-1}\). This result showed that exceed the
maximum concentration of arsenic recommended by EU (2006) and CODEX (2011). This value indicates a comparatively higher value than other studies on As in fruits pepper reported by Castro-Larragoitia et al. (1997) in San Luis Potosí, Mexico (3.0 mg/kg); Roychowdhury et al. (2003) in West Bengal, India (68.24 µg/kg); Prieto-García et al. (2005) in Zimapán mining areas, Mexico (6.26 mg/kg); Ramirez-Andreotta et al. (2013) in Arizona, USA Solanaceae (bell pepper, green chili, jalapeno, and tomato) range from 0.00132–0.0100 mg kg\(^{-1}\). Similarly, in this work, As concentration between root vegetable and fruit vegetable were comparable. However, our results differ from those reported by Cao and Ma (2004) suggested that direct soil contact by root vegetables leads to higher concentrations compared to leafy vegetables, which must translocate As from roots to shoots. Therefore, we found that As concentrations in garlic and carrot roots tissues and pepper fruits were positively proportional to the levels of As in the soil.

**Lead accumulation in vegetables**

*Allium sativum*: The found mean concentration for Pb in garlic was 3.0 mg/kg. Permissible limit concentration to lead in bulb vegetables is 0.1 mg/kg (FAO/WHO 2014). Therefore, this value revealed that was higher than those found in previous studies (Song et al. 2009). Also, confirm comparable Pb values than those of literature reported in garlic (Guerra et al. 2012; Rehman et al. 2016; Roba et al. 2016). However, in our study were low that those found by Türkdogan et al. (2003), Maleki and Zarasvand (2008) and Senila (2014). *Daucus carota*: The mean concentration found of Pb to carrot was 5.0 mg/kg. The permissible limit concentration of lead in carrot is 0.1 mg/kg (EU 2006; FAO/WHO 2014). Therefore, this value showed that exceed maximum limits in carrots (Knapp et al. 2013; Islam et al. 2016; Rehman et al. 2016; Shaheen et al. 2016; Zhou et al. 2016). Also, were comparable with the found by Banerjee et al. (2010) and Pančevski et al. (2014). However, concentration was lower than the studied by Senila (2014). *Capsicum annuum*: The found mean concentration for Pb in pepper fruits was 9.6 mg/kg\(^{-1}\). These findings indicate that Pb levels were higher than those reported in other research Antonious and Kochhar (2009), Guerra et al. (2012), Islam and Hoque (2014), Mirecki et al. (2015), Islam et al. (2016) and Antoine et al. (2017). However, concentration was comparable with Ahmad and Goni (2010).
Regarding Pb concentration in this survey pH was close to 8.0. These results corroborate with findings by Merry et al. (1986) stated that increasing soil pH decreased Pb concentrations in the plants, an effect that was more remarkable in highly contaminated soils. In this way, Singh et al. (2010) affirmed that Pb concentration varied among vegetables reflect the difference in their uptake capabilities and their further translocation to the edible portion of the plants. Thus, the content of Pb in various plant organs can vary with plant species (Sharma and Dubey 2005). Therefore, our results agree with Alexander et al. (2006) and McBride (2013) who highlight the general observation on the barrier to the translocation of Pb from the stem to the fruits in highly contaminated soil. In particular, *C. annuum* samples where Pb concentration no was detected. Also, our results corroborate with findings by Cai et al. (2020), who found *C. annuum* has lead tolerance ability indicating that it can effectively absorb lead in soil.

In general terms, according to Manzoor et al. (2018) presence of heavy metals in vegetables might be due to the higher uptake from soil. The uptake of As and Pb in plants are regulated by chemical speciation, biogeochemical characteristics, other physic-chemical parameters of the soil; microbial activity by mycorrhization in soils, as well as plant factors (Davies 1995; Feleafel and Mirdad 2013; Abbas et al. 2018). Thereby, apart from heavy metal content in the soil, the physic-chemical properties of soil also affect the degree of contamination of vegetable crop (Singh and Kumar 2006). Possibly, spatial difference in the contamination load of As and Pb in plants was due to different levels of contamination, as well as to the previously mentioned in the literature (parameters of soil, biogeochemical, microbial, and plant factors). Therefore, the plants absorbed As and Pb from the contaminated soil and possibly, those deposited in the parts of the plants exposed to dust from the polluted environment (Kachenko and Singh 2006; Singh and Kumar 2006).

**Pollution load index**

The use of a pollution index indicates whether soil quality is suitable for agricultural use and whether vegetables grown can be consumed eaten safely. Therefore, the soil of agricultural use should be further assessed to determines the environmental risk caused by contaminated soils (Li et al. 2006). The environmental quality indices of the As and Pb in the vegetables are
presented (Table 2). The order of the indices was As>Pb. The pollution index to As followed a similar trend in all soils. These results demonstrate that As are the dominant metal pollutants accumulated in soil and vegetables grown in agricultural soils in the region. The indices of all soil samples of As and two soil samples of Pb were greater than 1. However, the PLI values for As was lower than those reported by Khan et al. (2017b). Therefore, showing that soils are classified as polluted and indicated that the study area is highly contaminated (Li et al. 2006). It also showed that the contamination index by As is high and persists a greater possibility of causing health and food safety problems (Khan et al. 2017a).

The elevated As and Pb concentration in farmland are attributed to the geogenic origin and anthropogenic by wastes of historical mining activities. Also, other pollution matrices as tailing abandoned, the processing company, and new tailing ponds not considered in the study. The tailing ponds or impoundments are excavations of highly variable sizes located within farmland, where local processing companies dig and extract the historical mining waste that was dragged through the streams. Subsequently, tailings ponds were filled with processed waste have a high level of heavy metals that seep over time and migrate through the soil profile to the deeper layer. Nowadays, in these new tailing ponds are grow various crops. The new mine ponds are recklessly managed and do not exist mechanisms to oblige the tailings processing company to obey the environmental laws and regulations (González et al. 2012).

Also, new tailings ponds found inside farmland probably represent a risk of contamination of the aquifer and a grave health hazard. In this regard, previous studies have found high levels of As and Pb in groundwater extraction wells that supply Guadalupe municipality (Castro et al 2003; Gonzalez 2011). Also, heavy metal contaminated crops could aggravate human health risks when consumed along heavy metal contaminated drinking water (Brammer and Ravenscroft 2009; Zavala and Duxbury 2008). Regarding, a survey conducted among the population of the area 18.48% of the respondents reported at least one household member with dark spots on the hand palms (arsenicosis symptom) (Gonzalez (2013). The results of this study support the proposal that setting arsenic standards for food is urgent and beneficial in combination with existing arsenic water standards for protecting public health (Peralta-Videa et al. 2009; Meharg and Raab 2010) and fulfill the commitments on agri-food safety standards.
with WHO. Therefore, high pollution index shows the need to perform permanent monitoring because As and Pb are potentially toxic elements and could be bioaccumulated to human beings through the food chain, and avoid potential problems that could be dangerous to the population. It is recommended that community members living around sources of pollution take steps to prevent public health. Also, the other routes of exposure existing in the study area should be the subject of future studies to assess their specific contribution to the food chain and the agrosystems.

**Soil-vegetable bioaccumulation factor**

The bioconcentration factor (BCF) is one of the key components of human exposure to metals through the food chain. Also, is a criterion to assess global human health concerns (Woldetsadik et al. 2017). The BCF values of As and Pb carrot, garlic and pepper are presented in Table 2. These values varied between vegetable species and sites. The trend of BCF was similar to the order of the As>Pb. Among vegetables, the highest BCF value was for As, ranging from 2.33 to 0.64, and the average for all samples was 1.01. This value is a clear indication that the As has a higher capacity for As accumulation in the edible parts in root vegetables (carrot and garlic) and fruit vegetable (pepper) compared with Pb. Likewise, root vegetables show the highest BCF. The BCF value less than one or equal to one shows the plant can only absorb but not accumulate heavy metals. If BCF is more than one, it indicates that the plant can absorb and accumulate metal (Liu et al. 2009). According BCF values of plants are characterized as excluder (<1.0), and hyperaccumulator (> 1.0 - 10.0), respectively (Ma et al. 2001).

The highest BCFs were recorded in garlic (2.33), carrot (1.22) and pepper (1.21). These might be due to higher mobility of the As with a natural occurrence in soil (Alam et al. 2003), and its lower retention in the soil than other toxic cations (Zurera et al 1987). Therefore, a high BCF value indicates a higher accumulation potential of metals in the vegetable (Cui et al. 2004). According to these data implicated that vegetables are hyperaccumulators (>1.0), and those with values close to 1.0 like accumulators of arsenic. While the lead that recorded the lowest values BCF in all vegetables showed a relatively low potential for Pb accumulation in agricultural soil studied. Although reduced uptake of heavy metals is one of the plant's
adaptation strategies to avoid metal toxicity (Baker and Walker 1990). Therefore, these results agree with Bui et al. (2016) who found that soil pH slightly alkaline was very similar across sites, we rule out pH as a significant driver of BCF differences in our study. In addition, the results of this study have revealed that the responses of vegetables to exposure to As and Pb are complex due to the heterogeneous tolerance, and the various relationships between the concentration of As and Pb in the soil and the plants (Kabata-Pendais et al. 1993).

**Conclusions**

This study brings recent evidence on the assessment and accumulation of As and Pb in vegetables cultivated in soils contaminated by old mining activities. According to results found the most relevant findings are agricultural soils are highly contaminated with As and remain available under conditions alkaline. Thus, substantial concentrations of As compared to the maximum permissible limits in soils for agricultural use in other areas close to mining activities were registered. The average concentration of As in fruit vegetables were 95.66 mg kg$^{-1}$ and root vegetable 92.33 mg kg$^{-1}$. The edible parts of vegetables bioaccumulate As and Pb at levels higher. The average As concentration found garlic, carrot, and pepper exceeded their permissible limit levels of health and food safety the national, various countries, and various international standards. Among vegetables, root vegetables show the highest bioconcentration factor value. These data proved that these vegetables have attributes as hyperaccumulators of arsenic. The soil pollution load indices of all soil samples of As and two soil samples of Pb were higher than 1. These results are showed that As is the dominant pollutants accumulated in soil and vegetables grown in agricultural soils in the region.
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The study was conducted in agricultural soils of four community rural El Bordo (22°54’34” N, 102°24’45” W), El Lampotal (22°54’43” N, 102°24’10” W), La Era (22°51’17” N, 102°25’03” W) and Santa Rita (22°54’42” N, 102°25’06” W) located within a region rich in natural deposits of arsenic and lead, and contaminated since colonial and postcolonial times, from Guadalupe and Vetagrande municipalities of Zacatecas in Mexico (Fig. 1). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.
Figure 2

please see the manuscript file for the full caption