



Heavy metals, definition, sources of food contamination, incidence, impacts and remediation A literature review with recent updates

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Abstract

Heavy metals are any metallic element that has a proportionally high density and is poisonous at low concentrations. Also, heavy metals are non-biodegradable in natural environments and categorized in two groups as toxic metals (*i.e.* Pb, Cd and As *etc.*) and essential metals (*i.e.* Cu, Zn, Mn, Fe, Ni and Cr *etc.*). The releasing sources of heavy metals are divided to the natural and anthropogenic sources. Increase of urbanization and industrialization lead to increase the metal contamination of food. The toxic metals can be translocated to different food through different sources such as irrigation water, agricultural soils, agricultural practices, air pollution, animal feed and packaging materials. The toxic metals are non-biodegradable, non-thermo degradable and extremely persistent in the environment; thus readily accumulate to the different food. The metal contamination of different food such as agricultural crops, meat, fish, milk and egg threatens both food safety and security. Contamination of agricultural soils, irrigation water, plants and animals with toxic metals cause their incorporation into the food chain and cause a great threat to human health. Most metals have toxic effects on human health and accumulate in the different organs such as skeleton, liver, spleen, and kidney. Metals have negative effects on plant and animal production. So, application of different remediation techniques became necessity to reduce the toxic heavy metals pathway to the food chain and human body. Metal nanoparticles used in benefits application, but may be have some risks.

Keywords: Heavy metals, toxicity, food safety, metal contamination, contamination sources, remediation.

1. Introduction

Food contamination with heavy metals is one of the major problems (nationally and globally) that causing a great risk to human health. Toxic metals enter the food through various sources either naturally or by human activities, then can accumulate in the human organs and causing serious problems due to their toxicity. Also, heavy metals can accumulate to human body through inhalation [1]. Most toxic substances such as veterinary drugs are usually degraded before or after their uptake in the human body, then metabolized to harmless metabolite. In contrast, some toxic substances such as heavy metals are persistent, not metabolized and remain in the food, then irreversibly bound to body organs, such as Pb in bone and Cd in kidneys [2].

The agricultural soils receive many of toxic metals from both natural and anthropogenic sources [3]. Use

of wastewater for irrigation of agricultural crops led to increasing the levels of toxic metals in the food chain and subsequently transferred to human and animal causing potential health risk [4]. The contaminated water and soil with metals are requiring some treatments to reduction of metals bioavailability in soil and subsequent decrease transfer of toxic metals in cultivated crops [5]. Biochar, zeolite, yeast and bacteria have functional groups that can adsorb the toxic metals from soil and water according to the nature of their surface charge [6-9]. Household treatments were applied to reduce the level of metals in the different food [10-12]. Nanoparticles of metals have a broad range of technological and environmental applications such as water and soil treatments [13].

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2. Definition of heavy metals

The term "heavy metals", have been generally used to describe metals with atomic weight greater than iron (56). Also, the term "heavy metals", have been used to describe metals having density greater than 5 g cm^{-3} [14]. The actual definition of heavy metals is any metallic element that has a proportionally high density and is poisonous at low concentrations such as As, Pb, Cd, Hg and Cr [15]. Generally, heavy metals are non-biodegradable in natural environment and are categorized in two groups. The first group is toxic metals (*i.e.* Pb, Cd and As *etc.*) which are undesirable, don't have biological benefits for human health and toxic at any concentrations. The second group is essential metals (*i.e.* Cu, Zn, Mn, Fe, Ni and Cr *etc.*) which are desirable and have biological benefits for human health at low concentrations, but it become toxic at high concentrations [16].

3. Sources of heavy metals

The releasing sources of heavy metals to the different environmental media such as soil, air and water are divided to the natural and anthropogenic sources [17]. The natural sources such as volcanic eruptions, sea-salt sprays, forest fires, rock weathering, biogenic sources and wind-borne soil particles. The anthropogenic sources such as industries processes, agriculture processes, discharge of wastewater, mining processes, metallurgical processes, as well as emissions of chimneys and motors. The main sources of metals are pesticides, paint productions, printing of textiles, earth's crust, hair dyes and Polyvinyl chloride (PVC) pipes for lead (Pb); rechargeable batteries, tobacco, phosphate fertilizers, pigments, industrial wastes and agrochemical wastes for cadmium (Cd); stainless steel, alloy production, wood saving, textile dyes, leather tanning and electroplating for chromium (Cr); kitchen utensils, machine parts, batteries, earth's crust, cigarette and stainless steel for nickel (Ni); volcanic activities, pyrolysis of biomass, mining, paint of ships, wastewater discharges and forest fires for mercury (Hg); volcanic activities, disposal of arsenical chemicals, mining of gold and copper, arsenical pesticides, smelting and fertilizers for arsenic (As) [18].

4. Sources of food contamination by toxic metals

4.1. Irrigation water

Expanding of population, food demand and lack of irrigation freshwater in some developing countries lead to irrigation of agricultural crops with contaminated water. So, contamination of irrigation water with heavy metals has become a local and global problem. The regular use of wastewater for irrigation of agricultural crops leads to increase the heavy metals concentrations in crops and subsequently transferred through the food chain to human and animal causing potential health risk at the long term [4]. Contamination of food such as fruits, vegetables and agricultural crops by heavy metals may happen due to release of industrial wastewater and sewage wastewater that contaminates the irrigation water sources such as canals, nearby streams and rivers. The agricultural, sanitary and industrial waste-water is reuse in plant irrigation by mixing it with the Nile River water for agricultural expansion and reclamation projects [19].

Also, Darwish and Abdel-Razek [20] reported that the agricultural, industrial and municipal sewage waste-water are the main sources of metal contamination that recklessly dumped into the Nile River. Therefore, use of this irrigation water (untreated or partially treated wastewater) leads to metal contamination of irrigation water, then accumulate in agricultural crops. In addition, Yadav et al. [21] reported that, industrial wastewater is an important source for metal contamination of irrigation water which used in cultivation of agricultural crops. They added that the different industries use great quantities of water for the processing, and then can contaminate waterways through the discharge of their wastes into streams, rivers and other water sources.

Use of contaminated water in agricultural uses requires the knowledge of the levels and types of water contaminants, especially toxic metals to protect the environment and public health. Heavy metals are the most important chemical contaminants that threaten water quality for different uses. Monitoring of metals levels in irrigation water is necessary to protect ecological and human health due to its toxic effects and its persistence [22]. For example, Abdel-Razek [19] was monitored the levels of some heavy metals in irrigation water samples collected from Bahr El-Baqar. He noticed that the levels of Pb, Cd,

Ni, Cu, Mn, Zn and Co in irrigation water were 36.6, 14.7, 73.2, 65.7, 59.0, 90.6 and 89.7 mg l⁻¹, respectively.

Also, Mahmoud and Ghoneim [23] determined the metal contamination of irrigation water collected from El-Mahla El-Kobra. They revealed that the levels of Cu, Cd, Pb and Ni were 386, 33, 92 and 164 mg l⁻¹, consecutively. In addition, EL-Hassanin et al. [24] reported that the heavy metals levels in crops cultivated in the soils irrigated with low quality water were higher than those cultivated in the soils irrigated with freshwater. Nassr et al. [25] investigated the impact of irrigation water quality for long-term on heavy metal contents in wheat grains. The levels of Cd, Pb, Ni and Cu in wheat grains irrigated with Nile water were 0.07, 0.09, 0.22 and 1.04 mg kg⁻¹, respectively. But, wheat grains irrigated with drainage water recorded higher levels of Cd, Pb, Ni and Cu as 0.09, 1.18, 0.84 and 1.55 mg kg⁻¹, respectively.

4.2. Agricultural soils

Agricultural soils are not only source of nutrients for plant life, but also transfer many contaminants such as heavy metals to cultivated plants through their roots. Contamination of agricultural soils with heavy metals, such as Pb, Cd, Cu, and Ni increased dramatically during the last years [23]. The agricultural soils receive many of toxic metals from both natural and anthropogenic sources. Accumulation of heavy metals in agricultural soil may be by through emissions from the industrial activities, petrochemicals, wastewater irrigation, atmospheric deposition and agricultural activities such as fertilizers and pesticides [3].

Abdel-Lateef et al. [26] reported that, the Gabal El Asfar farm which is a fruit plantation irrigated with sewage wastewater for over 90 years and contaminated by potentially toxic heavy metals. So, they found that the mean levels of Zn, Cu, Ni, Cd, Pb and Cr in the agricultural soil were 331, 84, 22, 3, 23 and 154 mg kg⁻¹, respectively. Also, El-Gammal et al. [27] noticed that the concentration of Ni, Cu, Zn and Fe in the soil samples collected from Damietta governorate were 15.9, 33.9, 258.7 and 19.2 mg kg⁻¹, consecutively. Moreover, El-Alfy et al. [28] reported that the levels of Cd, Pb, Ni and Cr in agricultural soils collected from El-Gharbia governorate were 292.3, 413.5, 1403.4 and 726.9 mg kg⁻¹, respectively. As well as, Abuzaid [29] reported that, the

concentration of Cd, Cr, Cu, Ni, and Pb in soil samples collected from Al-Qalyubia Governorate were 0.5, 78.7, 73.2, 63.1, and 56.3 mg kg⁻¹, respectively.

Heavy metals can accumulate from contaminated soil to the different parts of food chain [30]. In this respect, Baranowska et al. [31] noticed that the content of Cd and Pb in agricultural soil ranged from 0.13 to 5.89 and from 0.57 to 151.5 mg kg⁻¹, respectively. The accumulated metals in grass, milk, cereals, eggs, and fruits were determined. Levels of Cd ranged from 0.11 to 0.63 mg kg⁻¹ in grass, from 0.009 to 0.060 mg kg⁻¹ in milk, from 0.20 to 0.31 mg kg⁻¹ in cereals, from 0.11 to 0.15 mg kg⁻¹ in eggs and from 0.23 to 0.59 mg kg⁻¹ in fruits. Meanwhile, the content of lead ranged from 0.16 to 136.57, from 1.16 to 3.74, from 1.05 to 5.47, from 5.79 to 55.87 and from 21.00 to 87.36 mg kg⁻¹ in the grass, milk, cereals, eggs, and fruits, consecutively.

4.3. Agricultural practices

The metal contamination of agricultural crops may be due to the use of fertilizer or metal-based pesticides during agricultural practices, as well as the metal contamination may be due to the harvesting procedure, transport or storage [32]. The agricultural practices such as fertilizers, manures and sewage sludge are important sources of heavy metals. In this respect, De Miguel et al. [33] revealed that the sewage sludge is use as a main source of plant nutrients and organic matter, but it has harmful substances such as heavy metals. Also, the phosphatic fertilizers such as super phosphate are important sources of Ni and Pb in soils, as well as have a considerable acidifying effect on soils and hence increase the mobilization and plant uptake of the metals which increase the accumulation of toxic metals in agricultural crops [34].

The agrochemicals such as phosphate fertilizers applied to sustainability of conventional agriculture. In this respect, Carnelo et al. [35] reported that rock phosphate, diammonium phosphate and superphosphate had high levels of Cd, Cr, Cu, Zn, Ni and Pb as 10.1, 29.7, 29.2, 89, 17.9 and 12.2 ppm, respectively. Roberts [36] revealed that phosphate rock has variable levels of inherent Cd. Also, Cd may transfer to fertilizer products during the manufacturing process. Moreover, Salem et al. [37] disclosed that application of phosphate and urea fertilizers increased the levels of metals (Cr, Cu, Cd,

Mn, Zn, Ni and Pb) in agricultural soil. Herbicide used in agriculture might also play a role to increase the level of Cu in the plant.

Atafar et al. [38] indicated that the levels of Cd, Pb and As were increased in the soil and cultivated plant due to application of fertilizer and manure at long-term. For example, the mean levels of Cd, Pb and As in the soil (before cultivation) were 1.3, 3.6 and 4.1 mg kg⁻¹, consecutively. Meanwhile, their mean levels in the soil (after harvesting) reached to 1.4, 6.8 and 9.1 mg kg⁻¹, at the same order. Furthermore, AlKhader [39] studied the effect of phosphorus fertilizers on accumulation of metals in lettuce plant. They indicated that the levels of Cd, Pb and As in lettuce were 0.03, 0.12 and 0.013 mg kg⁻¹, respectively.

4.4. Air pollution

Awofolu [40] reported that the principal source of air pollution by toxic metals is automobile exhausts, especially air pollution with Pb in sites surrounding the highway (Organic lead compounds *i.e.* tetra ethyl lead and tetra methyl lead are adding to petrol). The airborne metals are precipitated on soils surrounding highways, then accumulate in cultivated plants. Hashim et al. [41] studied the accumulation of some heavy metals in plants grown nearby Cairo-Alexandria agricultural highway. They reported that the levels of Pb, Ni, Co and Cd in citrus and cabbage were decreased with increasing the distance from the agricultural highway.

Also, Elnazer et al. [3] determined the levels of Pb and Cd in agricultural soil collected from Alexandria-Marsa Matruh Highway. They noticed that the Pb and Cd levels ranged from 29.15 to 50.6 and from 1.25 to 3.15 mg kg⁻¹, respectively and their levels in soil decreased with increasing the distance from the road. In addition, Shendi et al. [42] estimated the concentration of Pb in soil collected from the Fayoum roadside and ranged from 22.94 to 38.65 mg kg⁻¹. Also, the mean level of Pb in soil collected from El-Tabbin industrial area was 33.3 mg kg⁻¹ as recorded by Melegy et al. [43].

Fruits and vegetables growing at the roadside may be accumulating the toxic metals, especially from vehicle emissions as suggested by Feng et al. [44]. Also, Shahid et al. [45] disclosed that airborne heavy metals may be deposited and absorbed on the leafy parts of the different plants. As well as, Sulaiman and Hamzah [46] reported that the uptake and

translocation of some metals (Cd, Cu, Fe, and Pb) in different parts of roadside plants were higher than those grown in uncontaminated site.

4.5. Animal feed

Metal contamination of animal feed and its ingredients represent a major problem for animal health and the accumulation of toxic metals in the human food chain such as meat, egg and milk. Elliott et al. [47] studied the ratios of metal contamination in different animal feed. They indicated that about 19, 24 and 15% of poultry feed samples were contaminated with As, Cd and Pb, respectively. Meanwhile, the contamination ratios of ruminant feed samples with As, Cd and Pb were 27, 21 and 16%, consecutively. Kabeer et al. [48] studied the correlation between heavy metals in poultry feed and their accumulation in egg. The levels of Pb, Cr and Se in feed were 2.59, 1.30 and 0.94 ppm, respectively. Meanwhile, levels of accumulated Pb, Cr and Se in egg white were 0.658, 0.180 and 0.216 ppm, respectively. Higher levels of accumulated Pb, Cr and Se were recorded in egg yolk as 0.701, 0.262 and 0.266 ppm, respectively.

Also, study by Zhang et al. [49] estimated the levels of some metals in cattle and poultry feeds. The detected mean levels of metals in cattle feed were 26.0, 114.9, 0.8, < d.l. and 2.0 mg kg⁻¹ for Cu, Zn, As, Cr and Cd, respectively. Meanwhile, the mean levels of the same metals in poultry feed were 21.9, 103.7, 1.22, 25.7 and 1.03 mg kg⁻¹ at the same order. Moreover, Leontopoulos et al. [50] studied the transfer of some heavy metals (Cr, Cu, Pb, Cd, Zn and Ni) from livestock feeds to cows and sheep organs such as muscle tissues, liver and kidney. The higher accumulation of Cu, Zn and Cd recorded in liver, muscle tissues and kidney, respectively. Meanwhile, levels of Cr, Pb and Ni were below 0.02 mg kg⁻¹ in all animal organs.

4.6. Cans, packages and equipment materials

The food industrializations such as canning process lead to the metal contamination of canned foods. For example, the main source of food contamination by Pb is solder used in the manufacture of cans [51]. Kocak et al. [52] demonstrated that the levels of Pb in some canned vegetables were 0.524, 0.372 and 0.838 mg kg⁻¹ for tin canned okra, tin canned sliced tomato and packaging tomato salsa, respectively. Also, Al-

Thagafi et al. [53] revealed that toxic metals (Cr, Pb and Cd) had relatively high concentrations in canned food (tuna, corned beef, sardines and tomato paste) than those of the corresponding fresh food. Oniya et al. [54] observed that the milling equipment released Pb, Ni and Fe into the foodstuff such as wheat flour, soya beans flour, maize flour and cassava flour.

Also, migration of Pb, Cd and Zn to hot foods stored in plastic bags and containers was observed by Abu-Almaaly [55]. The sources for Cd and Pb present in plastic packaging materials are inorganic pigments and stabilizers. Some metals such as Pb and Cd are used as stabilizers of PVC. Paper is inexpensive sources of food packaging and has high quality standards of safety compare with plastic. Also, food packaging papers such as sweet box, pizza box, coffee cup and pastry box had variable levels of heavy metals as reported by Sood and Sharma [56]. For example, levels of Cr, Cu, Fe, Mn, Ni and Pb in pizza box were 0.45, 0.83, 37.21, 0.44, 0.20 and 0.34 ppm, respectively.

Whitt et al. [57] reported that the recycled polyethylene terephthalate used for food packaging may possibly lead to leaching of Cr, Pb and Ni to pre-cut fruits and vegetables, as well as high-acid foods (salad dressing and sodas). Moreover, Khan and Khan [58] noticed that heavy metals such as Zn, Ni, Cu, Cr, Mn and Pb were migrated from plastic food packaging containers to 3% acetic acid and 0.9% NaCl. In future, equipment and materials containing the toxic metals such as Pb, Cd, Cr and Hg must be avoided when choosing food contact materials.

5. Incidence of heavy metals in food

5.1. Milk and milk Products

Milk and milk products are a complete food as healthy diet. However, presence of toxic metals in milk or milk products can be dangerous and detrimental for the human health. So, the safety of milk or milk products decreases with increasing of metal levels [59]. Presence of toxic metals in milk and milk products has a particular risk because milk is widely consumed by infants and children [60]. Cashman [61] revealed that the levels of heavy metals in milk and dairy products depend on the genetic factors of animal, stage of lactation, metal contamination from the equipment during production, nutritional type of the animal, environmental factors

and manufacturing practices. Also, Ajai et al. [62] and Anetta et al. [63] reported that the milk is a typical diet and has high nutrients such as minerals (Ca, K and P), vitamins (D, A, B-12, niacin and riboflavin), essential fatty acids, proteins and lactose. But, metal contamination of milk as a result increases the industrial and agricultural activities can lead to health hazards. The different plants are absorbed of heavy metals, then consequent accumulated in their tissues. Dairy animals that graze on the contaminated plants accumulate the toxic metals in their tissues, as well as milk if lactating [64].

Main sources of Cu contamination in milk or milk products is animal feed, increase of Cu levels in water and Cu alloys used in different equipment. Also, presence of Pb in milk may be return to industrial air pollution at areas of dairy farms [60]. So, heavy metals are the common contaminant that found in milk. In this respect, Enb et al. [65] determined the heavy metals in Buffalo's and cow's milk collected from Egyptian animal farms. They noticed that, the mean levels of some essential metals *i.e.* Fe, Cu, Mn and Zn in buffalo's milk as 0.98, 0.21, 0.08, and 4.37 mg kg⁻¹, respectively were higher than its levels in cow's milk as 0.79, 0.19, 0.05, and 3.97 mg kg⁻¹, respectively. Also, the levels of toxic metals such as Pb and Cd were highly in buffalo's milk (0.08 and 0.12 mg kg⁻¹, consecutively) as compare with cow's milk (0.04 and 0.09 mg kg⁻¹, consecutively).

The main sources of heavy metals in animal systems are (1) consumption of contaminated water and feed, (2) polluted air, (3) soil, (4) contaminated equipment's and (5) improper manufacturing practices [66]. In this respect, El-Bassiony et al. [67] studied the correlation between the Pb and Cd concentrations in consumption water and its levels in milk samples collected from New Valley, Egypt. They found that the mean levels of Pb and Cd in water were 0.18 and 0.05 ppm, respectively. Meanwhile, the mean levels of Pb and Cd in milk were 0.27 and 0.07 ppm, consecutively.

The most variance of metal contamination in milk could be attributed to the environmental condition. El-Sayed et al. [68] concluded that the levels of some heavy metals in milk samples varied according to the sampling sites. For example, the levels of Pb in milk samples collected from Helwan, Tanash, Shubra, Menofia, Mansoura, Awseem and Gharbia were 0.278, 0.412, 0.577, 0.614, 0.135, 0.138 and 0.141 mg kg⁻¹, respectively. They added that the levels of metals varied according season of production, for

example levels of Pb in milk samples from Helwan area collected during January-February, May-June and September-October were 0.47, 0.20 and 0.18 mg kg⁻¹, respectively. Also, levels of metal in milk increased with increasing the animal age [69]. They added that the animal body acts as an effective biological filter and accumulates the metals brought by the feed into the bone tissue rather than into the milk. Malhat et al. [60] investigated the residues levels of some metals in cow milk collected from Benha, Kaha, Shebin El-Kanater, Tokh and Kafr Shokr at El-Qaliubiya governorate, Egypt. They noticed that the levels of Pb, Cd, Cu, Zn and Fe in milk samples ranged from 1.85 to 4.40, from 0.20 to 0.229, from 0.90 to 2.84, from 4.77 to 10.75 and from 10.95 to 16.38, mg kg⁻¹, respectively.

Abdel-Hameed and El-Zamkan [70] reported that the recorded levels of Al, As, Cd and Pb in banana flavored milk samples collected from Qena city, Egypt were 0.96, 0.005, 0.022 and 0.11 mg l⁻¹, respectively. Meanwhile, Hg level in banana flavored milk samples was below the detection limits. Meshref et al. [71] estimated the levels of some metals in kareish cheese and butter samples collected from farms, individual farmers and dairy shops in Beni-Suef governorate, Egypt. They reported that the mean levels of Pb, Cd, Zn, Cu and Fe in kareish cheese samples were 0.43, 0.09, 8.59, 0.09 and 3.93 ppm, consecutively. Meanwhile, the mean levels of the same metals in butter samples were 0.49, 0.06, 5.98, 0.60 and 6.69 ppm, at the same order.

5.2. Fish

Fish meat is a desirable source of nutritional substances such as vitamins, minerals and high quality protein. Many environmental pollutants such as toxic metals are considered the main sources of metal contamination of water during discharges of industrial and agricultural wastes such as pesticides, coal and oil combustion, plastics and phosphate fertilizers. The different fish accumulated of toxic metals from water either through direct consumption of water or by uptake through the gills, skin and digestive tract [72]. Hamada et al. [73] investigated the levels of Hg, Pb and Cd in Nile tilapia fillet samples collected from Menoufia, Egypt. They reported that the levels of Hg, Pb and Cd were 0.73, 0.34 and 0.10 mg kg⁻¹, respectively in small size of wild tilapia, and reached to 1.18, 0.54 and 0.15 mg kg⁻¹, respectively in large size of wild tilapia.

Meanwhile, lower levels of Hg, Pb and Cd were recorded in small size of farmed tilapia as 0.45, 0.25 and 0.05 mg kg⁻¹, respectively, and reached to 0.94, 0.29 and 0.08 mg kg⁻¹, respectively in large size of farmed tilapia.

Emara et al. [74] measured the levels of Mn, Fe, Cu, Zn, Cd and Pb in muscle of Egyptian Nile tilapia fish collected from Al-Abbassa farm (irrigated with agriculture drainage water) and Shader Azzam (irrigated with sewage drainage water). The muscle tissues of Shader Azzam fish farm recoded the highest mean levels of Cu, Zn, Cd and Pb as 7.26, 120.8, 0.29 and 0.61 mg kg⁻¹, respectively. Meanwhile, the muscle tissues of Al-Abbassa fish farm recoded the highest mean levels of Fe as 109.2 mg kg⁻¹ and equal level of Mn in the muscle tissues of two fish farms as 1.21 mg kg⁻¹. On the other hand, lower concentrations of metals were recorded in tilapia fish collected from the River Nile at Damietta governorate, Egypt as 93.7, 5.5, 66.4, 0.01, 0.21, 0.29 and 0.09 mg kg⁻¹ for Fe, Cu, Zn, Cd, Pb, As and Hg, respectively [75].

The fish organs can accumulate the toxic metals several times higher than their levels in water as reported by Abdel-Mohsien and Mahmoud [76]. They revealed that the levels of Cr, Pb and Cd in fish samples collected from the Nile River were several times higher than their concentration in water and the bioaccumulation factor ranged between 8 and 122. Also, the accumulate rate of metals depended on the type of fish as reported by Helmy et al. [77] who revealed that the *Claris gariepinus* recorded the highest levels of Hg, Pb and Cd compared with *Oreochromis niloticus* and *Mugil cephalus*. Moreover, the accumulate rate of metals depended on the type of metal as reported by Abdelhamid et al. [78] who noticed that the bioaccumulated Pb in fish muscles higher than Cu, Fe, and Zn. The levels of metals in fish were affected by conditions of growth and nature of water. In this respect, Shokr et al. [79] studied the levels of Hg and Pb in samples of freshwater fish (*Clarias gariepinus* and *Oreochromis niloticus*) and marine water fish (*Sardina pilchardus* and *Pagrus pagrus*). They disclosed that the levels of Hg and Cd in freshwater fish (0.89-1.10 mg Kg⁻¹ for Hg and 0.49-0.64 mg Kg⁻¹ for Pb) were higher than their levels in marine water fish (0.57-0.72 mg Kg⁻¹ for Hg and 0.27-0.33 mg Kg⁻¹ for Pb).

5.3. Meat

In Egypt, offal of animals such as heart, kidneys, liver, lungs, rumen, spleen intestine and tongue are widely consumed as food source. But, content of toxic metals in offal are rarely determined in Egypt. So, estimating heavy metal in animal muscle and offal is necessary for increase the food safety and maintain the human health. The levels of metals in meat depended on the animal age as reported by Darwish et al. [80] who noticed that the water and protein contents of meat were decreased with increasing the animal age, while fat and ash contents were increased with increasing the animal age and this lead to increase of metal levels in meat. Darwish et al. [81] studied the levels of Cd and Pb in muscles, liver and kidneys of cattle and sheep collected from Zagazig Abattoir, Sharkia. The highest levels of Cd and Pb were recorded in liver and kidneys of both cattle and sheep, while the lowest values of metals were recorded in their muscles. Also, the studied metals in cattle organs were higher than those detected in sheep organs. They added that levels of Cd and Pb in organs of aged animals were higher than those detected in organs of young animals.

Also, Morshdy et al. [82] studied the levels of Cd, Pb, Hg, Cu and Zn in different organs (muscles, liver and kidneys) of different animals (cattle, buffalo, camel and sheep). They noticed that levels of metals varied according to the tissue type and animal type. Hassouba et al. [83] investigated the concentrations of Cd, Pb and Hg in frozen beef, frozen minced beef and frozen chicken samples collected from markets located in Luxor city. The sample of frozen chicken had the higher levels of Pb and Hg as 0.035 and 0.085 mg kg⁻¹, respectively. Meanwhile, the sample of frozen minced beef recorded the highest level of Cd as 0.012 mg kg⁻¹.

Food served to patients at hospitals should be free from toxic metals. El-Wehedy et al. [84] determined the toxic metals levels in meats served at Egyptian hospitals such as cooked meat, cooked chicken, raw meat and raw chicken. The cooked chicken recorded the highest mean levels of As, Cd and Pb as 0.122, 0.202 and 0.421 mg kg⁻¹, respectively. They added that, no significant difference in metal levels between beef and chicken samples. But, the cooked samples had a significant increase in metal levels compare with raw samples and this may be return to the evaporation and loss of water in the cooked tissue. Maky et al. [85] evaluated the levels of Cd and Pb in

some Egyptian meat products. The detected mean levels of Pb as ascending order were 0.08, 0.34 and 0.81 ppm for sausage, luncheon and pastirma, respectively. Meanwhile, the determined mean levels of Cd were lowered as 0.073, 0.029 and 0.016 ppm for sausage, pastirma and luncheon, respectively as descending order.

5.4. Egg

Egg is economical food and has the most nutritious that necessary for human health [86]. But, some toxic metals can accumulate in egg as reported by Hussien and Nosir [10] who revealed that the mean concentrations of some metals in egg samples collected from El-Monefia Governorate were 0.70, 0.31, 2.12 and 1.61 mg kg⁻¹ for Pb, Cd, Cr and Cu, respectively. The residual levels of As, Cd, Cu, Fe, and Pb in brown shell egg samples collected from Mansoura city were determined by AL-Ashmawy [87]. They noticed that the mean levels of Cu, Fe, and Pb in egg samples were 0.78, 30.04 and 0.19 mg kg⁻¹, respectively, while As and Cd not detected in all tested egg samples. Meanwhile, the mean levels of Pb, Cd and Cu in egg sample collected from Assiut governorate were 0.59, 0.02 and 0.57 mg kg⁻¹, respectively [88].

Also, Hashish et al. [89] estimated the metal levels in egg samples collected from four sectors representing different geographic areas in Egypt as North Delta (Alexandria governorate), Middle Delta (Menoufia governorate), South Delta (Cairo governorate) and Upper Egypt (Sohag governorate). They reported that the highest levels of Zn, Cu, Mn, Pb and As were recorded in egg samples of Menoufia governorate as 64.3, 6.7, 1.59, 0.303 and 0.033 mg kg⁻¹, respectively. Meanwhile, the highest levels of Fe and Cd were recorded in egg samples of Alexandria governorate as 86.1 and 0.015 mg kg⁻¹, respectively.

5.5. Vegetables, fruits and cereals

Abd El-Rahman et al. [90] evaluated the heavy metal contamination of vegetable samples (tomatoes, cucumber, lettuce, watercress and potatoes) collected from some Egyptian governorates such as Cairo, Giza, Alexandria, Menoufia, Kalyopia and Sohag. The highest accumulation of toxic metals (Cd, Pb, Al and As) in leafy vegetables (lettuce and watercress) and tuber vegetables (potato) compared with fruit vegetables (tomatoes, cucumber), was the important

observation of this study. In addition, Eissa and Negim [91] studied the translocation of some heavy metals (Zn Cu Pb Cd Ni) from a metal-contaminated soil to lettuce and spinach. They observed that the accumulated heavy metals in roots of lettuce and spinach were higher than those accumulated in their shoots.

Radwan and Salama [92] determined the levels of Pb, Cd, Cu and Zn in the different fruits (apple, banana, melon, date, grapefruit, peach, orange, strawberries and watermelon) collected from Alexandria city, Egypt. The detected metals were ranged from 0.05 to 0.87 mg kg⁻¹ for Pb, from < 0.002 to 0.05 mg kg⁻¹ for Cd, from 1.2 to 18.3 mg kg⁻¹ for Cu and from 1.36 to 10.5 mg kg⁻¹ for Zn. Also, Kandil et al. [93] determined the levels of 10 metals in fruit samples collected from Giza governorates. They disclosed that the levels of metals varied according to metal type and fruit type. For example, the maximum levels of Pb were 0.05, 0.12 and 0.22 mg kg⁻¹ in orange, pomegranate and strawberry, respectively. Meanwhile, the maximum levels of Cu in orange, pomegranate and strawberry were 1.9, 5.5 and 3.5 mg kg⁻¹, respectively.

With regard to cereals, Salama and Radwan [94] determined the levels of some metals *i.e.* Cd, Pb, Cu and Zn in wheat, rice, maize and barley samples collected from markets in Alexandria. The higher levels of Cd and Zn were recorded in maize samples as 0.14 and 15.45 mg kg⁻¹, respectively. Meanwhile, the highest levels of Pb (0.40 mg kg⁻¹) and Cu (1.96 mg kg⁻¹) were detected in wheat and barley samples, consecutively. Also, El-Hassanin et al. [24] studied the effect of irrigation water source on Pb and Cd levels in maize grains. They observed that levels of Pb and Cd in maize grains irrigated with freshwater ranged from < 0.001 to 0.04 and from < 0.001 to 0.01 mg kg⁻¹, respectively. But, maize grains irrigated with low-quality water had higher levels of toxic metals which ranged from 0.26 to 0.55 mg kg⁻¹ for Pb and from 0.038 to 0.112 mg kg⁻¹ for Cd.

6. Impact of heavy metals on human health

Contamination of agricultural soils, irrigation water, plants and animals with toxic metals cause their incorporation into the food chain and cause a great threat to human health (Figure 1).

6.1. Impacts of lead

Size of Pb²⁺ ions is similar with Ca²⁺ ions. So, Pb ions deposits in the bone and replaces Ca [95]; disrupts the region of hippocampus that specific for memory acquisition and lead to limitation of learning skills [96]; disorder of the central nervous system [97]; interferes with the normal function of enzymes [98]; causing carcinogenesis, mutagenesis and kidney damage [18]; effect on hemoglobin formation and lead to anemia [99]; inhibition of the synthesis of red blood cells and thus of the vital transport of oxygen [98].

6.2. Impacts of cadmium

Oxidation states of Cd are similar with Zn, hence Cd can replace Zn present in metallothionein, thereby inhibiting its activity as a free radical scavenger within the cell [18]; inhibition of some enzymes such as alkaline phosphatase and ATPases [100]; inhibition of DNA repairs processes causing the lung, prostate, pancreas and kidney cancers [101]; effect on the proximal renal tubular function causing the kidney failure [102]; causes softening of the bones and bone deformities [103]; causes the liver fibrosis [104]; induce oxidative stress in the brain cells [105].

6.3. Impacts of mercury

The toxicity of Hg depends on the mercuric form ingested as reported by Karri et al. [106] who noticed that the organic mercury such as methyl mercury (MeHg) is extremely more toxic than the inorganic mercury. The kidney is the critical target organ for inorganic mercury, while the MeHg is more toxic to the nervous system. Mercury ions disturb the metabolism of amino acid and the breakdown of membrane phospholipid [17]; release of uncontrolled Ca ions from mitochondria and disturb the mitochondria activity [107]; inhibition of glutathione peroxidase activity and increase of lipid peroxidation [106]; leads to Minamata disorder [108]; leads to neurodegenerative diseases, such as Alzheimer's [109]; binds to the sulfhydryl and disulfide groups in amino acids and cause inactivation of related enzymes and cofactors hormones [18]; effect on the nervous system, renal system and immune system [110].

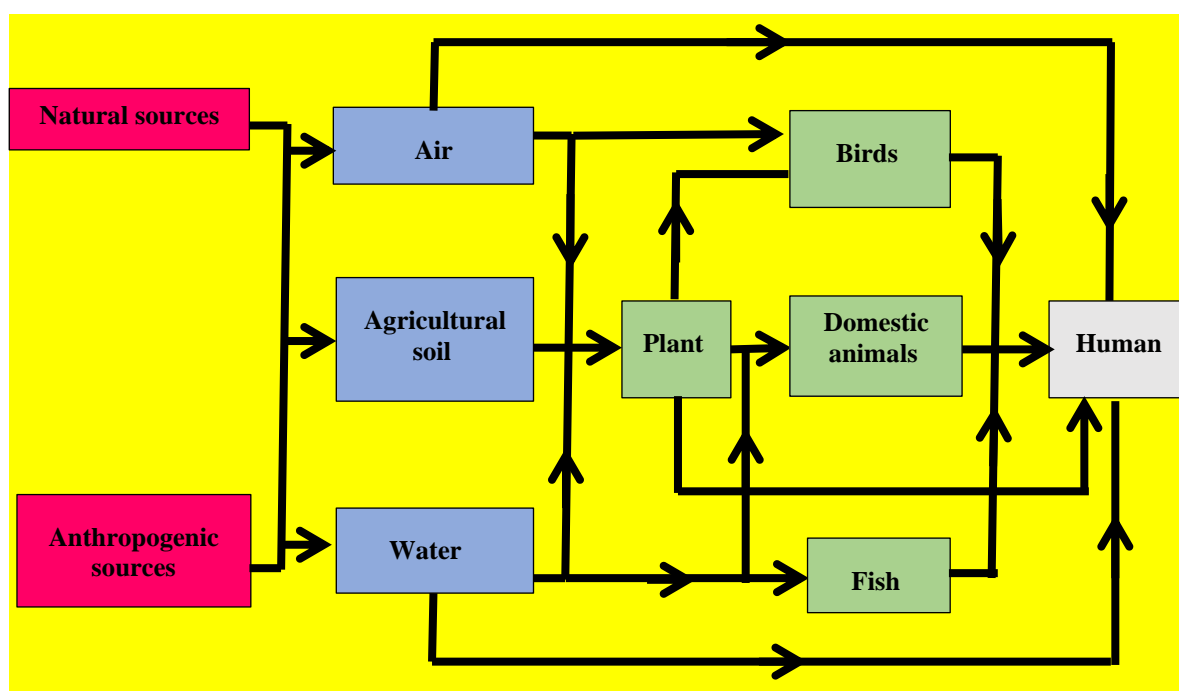


Fig. 1. Transfer of heavy metals from environmental sources to food chain and human organs.

6.4. Impacts of arsenic

Inorganic forms of As are highly toxic compared with organic forms [111]; inhalation of inorganic arsenic causing nausea and skin irritation [17]; causes internal cancers such as lung, bladder, liver and renal cancer [112]; causes external cancers such as skin cancer and skin lesions [113]; effect on the metabolism of nerve-impulse transmitters such as acetylcholine [114].

6.5. Impacts of chromium

Trivalent chromium Cr^{+3} ions are harmless due to its feeble membrane permeability, while hexavalent chromium Cr^{+6} ions are more active in penetrating the cell membrane and strong oxidizing agent [115]; Cr^{+6} ions have mutagenic properties and are categorized as a group 1 human carcinogen [18]; human exposure to Cr leads to dermatitis, allergies and ulcers. Also, Cr can effect on the respiratory system, gastrointestinal system and neurologic system [116]; Cr causes reproductive and cancer [117].

6.6. Impacts of nickel

Chronic exposure of Ni can lead to lung fibrous, cardiovascular and kidney diseases [118]; degenerative changes in heart muscle and in brain, lung, liver and kidney tissues [100]; causes carcinoma of respiratory tract and lungs [119]; causes Sarcoma of bone, connective tissue and muscles [120].

7. Impact of heavy metals on plant and animal production

The main physiological effects of metal toxicity in plants include: (1) inhibition of seed germination, (2) major reductions in growth rates, (3) changes in photosynthetic efficiency, respiration and transpiration, (4) changes in Mn, K, Mg, and Ca uptake rates [121]. Also, Kanwal et al. [122] reported that increase of Pb in the irrigation water lead to decrease of wheat germination, plant height, root weight, shoot weight, photosynthetic rate, total chlorophyll and yield parameters. Hassanein et al. [123] revealed that the soil contaminated with heavy metals had a negative effect on growth, yield and physiological responses of turnip and lettuce. Ozkay et al. [124] studied the effects of Pb, Cu, Cd and Zn levels in irrigation water on growth parameters of

eggplant. They concluded that the yield (g), plant height (cm), root length (cm), root weight (g) and leaf area (cm²) were decreased with increasing of metals in the irrigation water. Also, Wani et al. [125] investigated the effect of Cd and Pb levels in soil on chickpea production. The toxic metals decreased the plant growth, chlorophyll content, seed yield, nodulation and levels of nitrogen in root and shoot. As well as, yield and fruit quality of tomato were decreased in response to an increase of Ni levels [126].

Laying performance and egg quality of laying hens were negatively affected by toxic metals such as Pb and Hg [127]. Ibrahim et al. [128] studied the histopathological impact of toxic metals on muscles of *Clarias gariepinus* collected from El-Rahawy drain, Egypt. The observed impacts were degeneration, edema and splitting of fish muscle. Also, Zaki et al. [129] noticed that the toxic metals may affect the immune system of the fish and reduce the fish immunity to parasites. Moreover, Enb et al. [65] noticed that toxic metals can accumulate in dairy animals and cause negative effects on their health and milk production. Iron (Fe) has a many technological problems in dairy products such as its catalytic effect on lipids oxidation as well as bounding proteins and lipoproteins of milk [130].

8. Remediation of metal contamination

8.1. Remediation of heavy metals in agricultural soil

Once metals are introduced into and contaminate the agricultural soil, they may persist for a long time depending on the metal type and soil type. The contaminated soil with metals is requiring some treatments to reduction of metals bioavailability in soil and subsequent decrease transfer of toxic metals in cultivated crops. Remediation of contaminated soils has become increasingly more important in recent years to obtain safe food. In the last few years many authors studied new techniques to remove heavy metals from contaminated soils [5].

Biochar has functional groups that can adsorb the soil metals according to the nature of their surface charge [131]. Sayyadian et al. [6] studied the role of biochar on reduction of Pb, Cd and Ni bioavailability in maize. They reported that the concentrations of studied metals decreased in maize with increasing the biochar application. Moreover, Alaboudi et al. [132] concluded that the solubility of Pb and Cd was

decreased with addition of biochar in contaminated soil. Also, Khan et al. [133] investigated the effect of soil treatment with biochar on Cd accumulation in cabbage. They reported that biochar were decreased the Cd bioavailability in soil and its phytoavailability for cabbage.

The yeast such as *Saccharomyces cerevisiae* has ability to degrade toxic metals or also capable to accumulate of metals in their cells. So, the yeast biomass can be applied for adsorption of heavy metals in contaminated soil. In this respect, Damodaran et al. [8] concluded that, *S. cerevisiae* had ability to adsorption of Pb and Cd from the contaminated soil through its biosorption mechanism. Also, bacteria such as *Pseudomonas aeruginosa* is suitable biosorbent for the removal of heavy metals from contaminated soil as reported by many of studies [9, 134]. Fang et al. [135] reported that the extracellular polymeric substances extracted from *Pseudomonas putida* had binding effect for soil metals.

Wahba et al. [7] studied the effect of some clay minerals such as zeolite and bentonite to eliminate the heavy metals in contaminated soils. They concluded that, the zeolite had high ability for adsorption of heavy metals compared with bentonite. This is related to the specific molecular structure of zeolite as high cation exchange capacity and large surface area. Moreover, Azough et al. [136] investigated the influence of contaminated soil treated with natural zeolite on accumulation of Cd and Ni in wheat. They revealed that, the zeolite stabilized heavy metals in contaminated soil and reduced the negative effect of toxic metals on the wheat plant. Also, application of zeolite caused a significant reduction of Cd and Ni levels in shoots and roots of wheat.

8.2. Remediation of heavy metals in contaminated water

The plant wastes were applied for removal of heavy metals from contaminated water. The extracted biosorbents from *Jatropha* plant had affinity to adsorption of metals such as Cu and Zn from contaminated water [137]. Also, Mohammad et al. [138] applied the *Jatropha* seed hull as an adsorbent for Cd removal from the contaminated water. The synthesized activated carbon from *Jatropha* seed hull and impregnated with ZnCl₂ had high affinity for the removal of Cr⁺⁶ from industrial wastewater [139]. In addition, the waste of *Jatropha* was used for removal

of Cd from contaminated water [140]. The other techniques which used for removal of toxic metals from contaminated water such as removal of Cd and Pb from industrial wastewater using wastes of Jojoba plant [141] and olive stones [142].

Also, the biological techniques were used for metals adsorption from aqueous solutions. Farhan and Khadom [143] evaluated the ability of the yeast *Saccharomyces cerevisiae* to remove Pb, Zn, Cr, Co, Cd and Cu ions from aqueous solutions. Performance of yeast cells to remove metals from solutions were varied according to metal type, and the order of adsorbed metals is $Pb > Zn > Cr > Co > Cd > Cu$. The bacteria cells of *Pseudomonas* species such as *P. fluorescens* and *P. putida* had capability to adsorb Cd, Cr, Ni and Cu ions from wastewater as reported by Hussein et al. [144]. As well as, algae biomass was used as wastewater treatment method to eliminate Cu, Pb, Cd, Zn ions [145]. They concluded that, the effectiveness of algae biomass to adsorb toxic metals from wastewater return to its functional groups such as amine, carboxylic and hydroxyl groups.

Regarding the physical techniques, the agricultural residues converted to biochar used as adsorb material for aqueous toxic metals according to the previous literatures studies [146]. The biochar has functional groups, charged surface and high surface area that increase its adsorption efficiency for metal removal. Levels of Cu and Zn in industrial wastewaters were decreased using biochar as adsorb material [147]. Moreover, biochar had high ability for bind of Cd, Cu, Pb and Zn from aqueous phase [148]. Also, zeolite is widely used for adsorption of metal ions from wastewater such as removal of Pb and Cd from wastewater according to study of Sayed and Khater [149]. Zeolite adsorbed about 75, 32, 99, 28 and 59% of Cu, Cd, Pb, Ni and Zn, respectively from aqueous solution as determined by Shaheen et al. [150]. The effectiveness of heavy metal removal from wastewater was enhanced with decrease the particle size of zeolite [151].

8.3. Effect of household treatments on metal levels in food

Hussien and Nosir [10] evaluated the efficiency of *Enterococcus faecium* on the Cd and Pb removal from the milk. The reduction of Cd ranged from 79.4 to 82.8%, while Pb had lower reduction rate (from 72 to 75%). They concluded that *E. faecium* had higher ability for Cd binding as compare with Pb. Also, the

reduction rates of Pb and Cd in the minced meat suspended with *E. faecium* were 66.4 and 59.0%, respectively after 8 h. of incubation, then increased to 82.6 and 71.0% at the same order after 24 h. of incubation [152].

Elsharawy [153] studied the effect of grilling, marination and simmering processes on Cd, Cu, Fe and Pb levels in chicken meat. Most of cooking processes had a little effect on metal levels in the cooked meat. For example, the level of Pb in the raw chicken meat (0.30 mg kg^{-1}) was decreased to 0.25, 0.18 and 0.10 mg kg^{-1} in the grilled chicken, marinated chicken and simmered chicken meats, respectively. The aqueous solutions extracted from garlic, pomegranate peel and *Coriandrum sativum* were used for metal reduction in Grey Mullet fish [12]. The percentages of reduction for Pb, Cd and Hg in the fish tissues soaked in coriander agent were 76, 89 and 86%, respectively. Soaking of the fish tissues in the garlic agent lead to reduction of metals as 72, 100 and 96% at the same order. Levels of Pb, Cd and Hg in the fish tissues were decreased by 99, 92 and 58%, respectively after soaking in pomegranate agent.

Amir et al. [154] studied the effect of tap water and different solutions (5 and 10%) of lemon extract, sodium carbonate, citric acid and hydrogen peroxide on metal levels in spinach (soaked in solution for 10 min.). Citric acid (10%) recorded the highest reductions of Hg, Pb, As and Zn as 23, 28, 22 and 54%, respectively, while the lowest reductions were 7, 7, 6 and 15, consecutively in spinach washed with tap water. Another study estimated the effect of household processes on metal levels in potato, tomato and cucumber [11, 155]. The washing process decreased the levels of Cd, Cu, Pb, Cr and Ni in all vegetables, then more decreased after soaked in vinegar (5% for 5 min.). Sattar et al. [156] revealed that washing of vegetables by acetic acid (10%) was more efficiency for removal of metals than tap water and sodium chloride (10%). The next study of Sattar et al. [157] noticed that the vegetables washed with ginger solution (8%) recoded the maximum removal for Cd, Cr and Pb compared with tap water and radish solution (8%).

8.4. Metal nanoparticles

Nanomaterials have a broad range of technological and environmental applications such as water treatments, soil treatments, medicine and catalysis. In this respect, Anusa et al. [13] studied the effect of

nano-ZnO on adsorption of Cu, Pb and Cd ions from industrial wastewater. Reduction ratios of Cu, Pb and Cd were 99, 72 and 87, respectively at pH 2, then increased to 100, 77 and 98, respectively at pH 10. Also, Cu oxide nanoparticles were tested for adsorption of Ni and Cr from aqueous solutions [158]. Also, nanoparticles of Fe oxide are greatly used for metal adsorption due to their low toxicity and easy separation from aqueous solutions [159]. Another application was estimated the effectiveness of Fe⁺³ oxide nanoparticles stabilized with polyacrylic acid on Cd removal from contaminated soil [160]. The removal efficiency of Fe⁺³ oxide nanoparticles varied according to chemical fractionation of Cd in soil. The highest removal (100%) was recorded for exchangeable Cd, while the lowest removal (82.4%) was recorded for residual Cd.

The metal nanoparticles such as nanosilver, nanogold, and nano-TiO₂ are used in food, cosmetics, drug products and interaction with biomolecules and microorganisms. The silver nanoparticles bind the host cells of virus and lead to inhibit the virus [161]. Although, many of benefits and uses of the metal nanoparticles, but may be have risks on human health. Inhalation of metallic nanoparticles with size less than 30 nm pass rapidly into the circulatory system. The nanometals such as 30 nm (Au), 22 nm (TiO₂) and 15 nm (Ag) have a short-term or rapid translocation from lungs into the circulation and the different organs such as liver, kidney, spleen, brain and heart [162]. So, the use of nanoparticles metals and metal oxides should be conducted under maximum precautions because some reports aroused doubts about its safety. Fu et al. [163] reported that the use of nanoparticles metals and metal oxides induce genotoxicity, oxidative stress, inflammation and have been identified as a possible human carcinogen.

9. Conclusions

Heavy metals are toxic at lower concentrations and non-biodegradable. Metals released to the different environments from either natural or anthropogenic sources. The toxic metals can transfer from agricultural soil, contaminated water, polluted air, cans and packaging materials to food chain, then accumulate in the human organs causing negative impacts on human health. Toxic metals can be found in the different food such as milk, fish, meat, egg and agricultural crops. The different applications can be used for reduce the transferred metals to food chain

such as biochar, zeolite, yeast, bacteria, Jatropha plant, Jojoba plant and household treatments.

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11. References

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