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Review of organic and conventional agricultural products: Heavy metal availability, accumulation and safety

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Abstract
This paper provides a broad review of the process of heavy metal entering to food chain via different sources. A thorough study was conducted on heavy metals including possible sources, parameters control the plant availability in micro environment, mechanism of taking up by plants, translocation through the plants, extent of heavy metals in organically amended soils, impact of organic fertilizer on heavy metal availability and finally, the novel strategies available for removal of heavy metals from the environment. Metal contamination issues are becoming increasingly common in Sri Lanka and everywhere. Heavy metals such as cadmium, copper, lead, arsenic and chromium are major environmental pollutants, particularly areas in high anthropogenic activities. Agricultural activates have been identified as major source of heavy metals entering to food chain and, organic agriculture has been popularized throughout the world recent past due to safety and environmental concerns. However, limited evidence exists for comparison heavy metal content in the products of conventional and organic systems and, a thorough analysis is required to confirm the safety of organic products.

Keywords: heavy metals, organic foods, conventional foods, safety

1. Introduction

Food safety is one of the major concerns among the consumers of worldwide in recent decades due to growing incidence of health hazards. Reliable systems such as organic farming have attracted consumers in recent past due to its safety and environment friendly nature. Synthetic chemicals are avoided to use in organic agriculture and foods produced from such systems are expected to be healthier than conventional ones.

Organic farming greatly depend on recycling of variety of renewable sources and wastes, imposes restrictions on chemical usage (pesticides, fertilizer, feed additives and veterinary drugs) and genetically modified organisms which might cause detrimental effects on environment and human health (European Council, 1991) [23].

Organic farming is a sustainable system, which encompasses numerous positive effects on biosphere. The key principles and practices of organic food production aim to encourage and enhance biological cycles within the farming the farming system. Soil fertility in organic farming systems are managed by use of composted materials derived by adopting agronomic practices (such as crop rotation and planting nitrogen fixing plants), biodegradation of vegetables and animal sources, and the organic fraction of urban and agro-industrial waste. Compost enhance the soil fertility by modifying physical, chemical and biological properties of soil, and often used as an amendment agent for remediating metal polluted soils (Terzano *et al.*, 2008) [95].

Although the demand for organic foods significantly increasing day by bay, the growers are reluctant to adapt such a systems due to lower yield and higher production cost. However, producer benefits include premium prices of the products (Smith *et al*., 2004; Chavas *et al*., 2009) [83, 10], access to local and international markets, and due to the support of local, government and international bodies.

Many studies have been carried out to find out the consumer behavior for organic foods, however, only few studies are available for the comparison of safety of organic and conventional foods.

2. Heavy Metals

The term “heavy meals (HMs)” applied for group of elements including metals and metalloids with atomic density greater than 4g cm^{-3}(Hawkes, 1997) [31]. Most of the HMs toxic to human and animals even at low concentrations (Shahid *et al*., 2015) [82]. HMs are categorized in to main two groups: (i) Essential elements such as boron (B), copper (Cu), iron (Fe), molybdenum (Mo), nickel (Ni) and zinc (Zn). These elements are essential for plant growth in low concentrations and become toxic if the level exceeded the threshold limit. (ii) Non-essential elements such as arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb), can cause detrimental health effects at extremely low levels such as mutagenic, teratogenic and carcinogenic effects (Järup, 2003; Abdul *et at*., 2015) [35, 01].

Table 1: Heavy metal emission to the environment from different sources

Lithogenic Source	Anthropogenic sources
▪ Weathering soil minerals ▪ Volcanogenic particles ▪ Windblown dust ▪ Forest wild fires ▪ Vegetation ▪ Sea salt	▪ Industrial waste and activities (battery production, metal products, metal smelting, cable coating industries) ▪ Brick kiln ▪ Mining ▪ Agrochemicals (Pesticides and fertilizer) ▪ Waste water irrigation ▪ Sewage sludge ▪ Traffic emission ▪ Power plants (coal combustion)

2.1 Sources of Heavy Metals

HMs originate due to lithogenic and anthropogenic activities (Table 1 and 2) (Qishlaqi and Moore, 2007; Zhang *et al*., 2008) [73, 116]. Most of the HMs are non-degradable, persist in the environment and tend to accumulate in the biosphere along the food chain (Liu *et al*., 2007; Dong *et al*., 2013; Zhang *et al*., 2015) [53, 20, 117].

Agricultural activities are one of the major sources of entering heavy metal to food chain (Table 3) and posing an overwhelming risk to the environmental health. Continued application of chemical fertilizers in over dose may have contributed to increase the level of heavy metal in soil and ground water aquifers.

Use of waste water to irrigate agricultural fields as an alternative for waste disposal practiced by most of countries (Yadav *et al*., 2002) [111] and, heavy metals such as Cr, Ni and Zn would enter to agricultural fields via such polluted irrigated water (Yang *et al*., 2017) [112]. Khan *et al*., 2008 [45] observed that continued use of wastewater for irrigation has led to moderately enrich the soil with Cr, Cu, Ni, Pb, and Zn, and strongly enriched with Cd in Tongzhou District, Beijing, China. Furthermore, they have noticed that plants which were grown under the waste water irrigation have contaminated with those heavy metals and exceeded the permissible limits (Table 4) for vegetables set by SEPA and WHO.

Mineral fertilizers, pesticides and animal manure has been reported as predominant pathways of HMs entering to the food chain in European countries while, atmospheric deposition of HMs is in China (Shi *et al*., 2018) [85].

Although trace amounts of HMs are anticipated in chemical fertilizers available in market, excess levels of As and Cd have been reported in phosphate fertilizers (Dissanayake and Chandrajith, 2009; Jayasumana *et al*., 2015a) [19, 36]. Defarge *et al*., 2018 [17] reported that glyphosate based herbicide formulas

Table 2: Table Sources of heavy metal contamination in the environment.

Sources of heavy metals	Cd	Cu	Pb	Zn	Ni	Mn	Fe	Hg	Se	As	Cr	Co	Sn	Al	Ag	Ba	Bi	Be
Rock outcrops	+	+	+	+	+	+		+			+	+	+					
Volcanoes		+	+		+	+		+	+					+				
Wind dust (Desert)			+		+	+	+				+							
Metals smelters	+	+	+	+	+													
Metallurgy and electroplating	+		+		+	+		+		+	+				+	+	+	
Blast furnaces				+		+	+											
Electrolysis								+										
Traffic density Leaded gasoline			+	+														
Metal emission from tires				+	+													
House hold waste	+	+	+	+	+													
Sewage sludge	+	+	+	+	+						+							
Energy supply coal burning	+								+									
Petroleum refining	+		+	+			+	+		+	+			+				
Petroleum combustion					+													
High tension lines		+			+													
Food additives		+		+														
Phosphate fertilizers	+		+		+						+							
Fertilizers	+		+	+	+	+	+	+		+				+				
Pesticides		+	+					+		+								
Waste water (WW)	+	+	+	+	+			+		+	+							
Black water (toilet water)		+		+														
Mining	+		+			+		+		+				+				
Chemical industries	+		+	+			+	+		+	+		+	+		+		
Dyes and pigments	+		+				+	+		+								
Ink manufacturing					+		+					+						
Alloys			+															+
Print			+	+							+					+		
Photography	+		+		+						+				+			
Glass										+		+				+		
Paper mills								+		+	+			+				
Leather training				+			+	+		+				+				
Pharmaceuticals							+	+		+				+				
Textile	+				+		+	+		+				+		+		
Nuclear technology	+															+		

Source: Modified from Sharma & Agrawal, 2005 [84]

Table 3: Heavy metal content (µg g-1) in different agricultural sources

Metal	Agricultural amendments						
	Sewage sludge	Compost refuse	Farmyard manure	Phosphate fertilizer	Nitrate fertilizer	Lime	Pesticides
Cr	8.40-600	1.8-410	1.1-55	66-245	32-19	10-15	-
Ni	6-5, 300	0.9-279	2.1-30	7-38	7-34	10-20	-
Cu	50-8,000	13-3,580	2-172	1-300	-	2-125	-
Zn	91-49,000	82-5,894	15-556	50-1,450	1-42	10-450	-
Cd	<1-3,410	0.01-100	0.1-0.8	0.1-190	0.05-8.5	0.04-0.1	-
Pb	2-7,000	1.3-2,240	0.4-27	4-1,000	2-120	20-1,250	11-26

Source: Nagajyoti *et al*., 2010 [62]

poses a higher risk of heavy metal contamination of soil and edible plants, especially in the case of arsenic (As). Perera *et al*., 2016 [68] investigated that As and Cd concentrations in upper malwathu oya, Sri Lanka showed a significantly high level during the cultivation season of rice than in other seasons.

A study conducted by Kananke *et al*., 2016 [41] to identify the heavy metal contamination of leafy vegetables available (*Mukunuwenna*) in urban and sub urban areas of Colombo Sri Lanka, investigated that the average concentration of Ni, Cd, Cr and Pd have exceeded the recommendations given by World Health Organization (WHO) and, recommended to conduct regular heavy metal monitoring programs in order to ensure the safety of consumers.

Solid waste collection and disposal is an issue for most of the Asian countries during last decade, where dumping garbage and burning in collection sites are the most common modes of disposal. As waste separation is not well practiced, heterogeneous wastes piles can be seen in these open solid waste dump sites.

The surface water and ground water (via infiltration) at the vicinity of these sites can be polluted as the leachates of waste dumping contain trace metals. A study conducted by Dharmarathne and Gunatilake (2013) [18] in Gahagoda dump site in Sri Lanka showed that, heavy metals such as Pb, Zn, Ni, Cr, Co, Fe, Mn and Cu levels were above the standard levels of WHO for drinking water.

2.2 Effect of heavy metal on human and animal health

Continued intake of HMs in excess level may lead to accumulate in the liver and kidney of human beings, and will serve as causative agents for nervous, cardiovascular, kidney, and bone diseases (Li *et al*, 2015) [51]. Previous studies have shown the possibility for onset of stomach cancer due to the excess exposure to heavy metals such as Cd, Pb, Cu, and Cr (Türkdoğan *et al*., 2003) [98].

Agricultural activates have shown a great impact on human health risk than industrial activities (Wang *et al*., 2018) [103]. Chronic exposure to heavy metals such as As and Cd in miner concentrations may cause detrimental health effects (Williams *et al*., 2009; Roberts, 2014) [108, 76]. During recent past, enormous numbers of patients with damages in kidney were reported in north central area of Sri Lanka, where the major cultivation is rice and phosphate fertilizers have been used extensively (Jayasumana *et al*., 2013; Wimalawansa, 2014) [37, 109]. The prevalence of chronic kidney disease in affected areas of Sri Lanka is suspected to be due to continues overdose application of P fertilizers for seeking higher crop yields (Sirisena and Suriyagoda, 2018) [87].

Table 4: Maximum admissible concentrations of toxic metals (mg/kg) in soil and plant from the international and standards

Reference	Sample from	Heavy metal standards (mg/kg)						
		Cd	Pd	As	Cu	Cr	Ni	Zn
United Kingdom	Soil	3.00	300	20.0				
Union of Europe	Soil	1.00-3.00	50-300	-	140	150	75	50-300
	Plant	0.20	-	-				
United State	Soil		150					1400
Environmental Protection Agency	Soil	-	40.0	-				
World Health Organization	Soil	0.20	100	-	100	100	50	300
	Plant	0.20	0.30	0.70	40.0	2.3	4.0	100
Iran Department of Environment	Soil	5.00	75.0	40.0				
Canada	Soil	-	-	25.0				
Japan	Soil	-	-	15.0				
Iran	Plant	0.10	0.20					
Poland ministry of Environmental Protection	Soil	4.00	100	30.0				
Poland ministry of Agriculture and Rural Development	Soil	0.75-1.5	50-100	-				
Poland ministry of Health	Plant	0.1	0.2	-				
India	Soil	3-6	250-500		135-270	-	75-150	300-600
	Plant	1.5	2.5	1.5	30	20	1.5	50
Sri Lanka	Soil	4.00	20	2				

Source: Modified from Zolfaghari *et al*., 2018 [118].

2.3 Influence of soil characteristic on metal uptake by plants

Plants are exposed to heavy metals via growth media (soil), atmosphere (air), and the source of irrigation (water). HMs have largest availability in soil and aquatic ecosystems and to a relatively smaller proportion in atmosphere as particulate or vapors (Nagajyoti *et al*., 2010) [62]. HMs accumulates in ecological food chain through uptake at primary producer level and then through consumption at consumer levels (Fig 01).

Roots of plants are the primarily contact site for HMs, in aquatic systems, whole plant body is exposed to these irons. In addition to that, aerial organs of plants (leaves, fruits and flowers) also possible to absorb heavy metals directly due to particles deposited on the foliar surfaces (Bondada *et al*., 2004) [6].

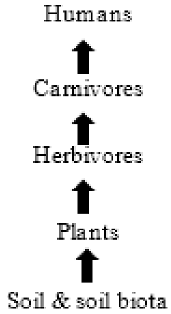


Fig 1: Heavy metal transfer through terrestrial tropic chain

Plant uptake of heavy metals depends on form of heavy metal present in the soil. Main forms of heavy metals available in soil are: dissolved in soil solution, exchangeable for organic and inorgnic matters, structural components in soil lattice and, insoluble precipitates (Zalidis *et al*., 1999)[115]. Heavy metals which are dissolved in soil solution and exchangeable form are readily available for plants. Majority of metal present in the soil are in insoluble form, bound with organic matter (humus) or with inorganic (clay) substances. To accumulate these heavy metals in plants, it is necessary to change the insoluble form to mobile in to the soil solution.

Soil properties such as pH, content of organic matter, cation exchange capacity (CEC), oxidation-reduction status (Eh), clay minerals, $CaCO_3$, Fe and Mn oxides determine the mobility and availability of heavy metals present in the soil (Kashem and Singh, 2001; Antoniadis *et al*., 2008; Usman *et al*., 2008)[43, 03, 99].

Numerous studies have emphasized that soil pH has great impact on mobility and availability of heavy metals. There is a negative co-relation between soil pH value and bioavailability of heavy metals such as Cd, Zn and Pb (Sukreeyapongse *et al*., 2002) [91]. Yang *et al*., 2014[113] studied the influence of soil type (Anthrosols and Cambosols) and land use duration on phytoavailability of heavy metals (Cd, Cu, Zn and Pb) and observed a higher Cd accumulation in leafy vegetables grown in the Anthrosols. Further, they emphasized that prolong use of land for agricultural activities may enhanced the availability of heavy metals, due to reduction of soil pH in the growing media.

Organic matter content in soil can be considered as the second major contributor for plant heavy metal availability. Organic matter facilitates soil to retain heavy metal in exchangeable form and, provide organic chemicals (chelates) that boost the heavy metal availability to the plants (McCauley *et al*., 2009) [59]. A study conducted by Hettiarachchi *et al*., 2003[32] showed that heavy metal adsorption to soil particles declined with lower organic matter content. However, Almås and Singh (2001) [02] observed a positive correlation of organic matter content and Cd uptake in rhy grass. A similar positive correlation was observed by Dai *et al*., (2004) [16] with regard to Cd, Pb and Zn.

2.4 Metal accumulation in plants

Some HMs bio-accumulate in plants to a higher extent without declining the yield or showing any visible symptoms. However, some HMs cause harmful effects in metabolism, physiological and biochemical processes such as growth reduction and lower biomass production (Nagajyoti *et al*., 2010) [62]. Metal accumulations in plant depend on plant species, growth stages, types of soil and metals (Table 5), soil conditions, weather and environment. A study conducted by Khairiah (2004) [44] indicated that potential bioavailability of heavy metals in the soil is strongly influenced by chemical form of metal in the soil such as whether it is chelated with organic matters, oxides and hydroxides form and trapped in the silicate clay.

2.4.1 Root uptake of HMs

Plants uptake heavy metals via active or passive transport. The mobility of heavy metals present in the soil can be controlled by plant roots through releasing metal chelating ions, reducing soil bound metal ions by specific plasma membrane bound metal reductase, and acidifying the soil environment by extruding protons (Raskin *et el*., 1994) [74].

Table 5: Heavy metal composition of typical uncontaminated soils and agricultural crops

Heavy metal	Range in soil (ppm d.wt)	Range in agricultural crops (ppm d.wt)
Cd	0.01-0.7	0.2-0.8
Co	1-40	0.05-0.5
Cr	5-3,000	0.2-1.0
Cu	2-100	4-15
Fe	7,000-55,000	-
Mn	100-4,000	15-100
Mo	0.2-5	1-100
Ni	10-100	1.0
Pb	2-200	0.1-10
Zn	10-300	15-200

Source: From Nagajyoti *et al*., 2010 [62]

Generally, metals absorbed to root surface and bind to the polysaccharides of the rhizodermal cell surface or carboxyl groups of mucilage uronic acid (Seregin *et al*., 2001) [80]. Absorbed metals penetrate to roots passively and diffuse through translocating water streams. Apoplastic (extracellular) and symplastic (intracellular) pathways are the major pathways available for metals to enter the plants.

Table 6: Heavy metal emission to air by different sources (tons)

Industry activity	Cd	Hg	Pb	As
Energy sector	5.72	19.8	80.9	2.06
Mineral oil and gas refineries	1.09	1.04	2.14	1.63
Thermal power stations and other combustion facilities	3.72	18.50	61.1	205
Coke ovens	1.01	0.28	17.4	-
Production and processing metals	9.66	4.77	398.3	-
Mineral industry	1.79	4.00	60.9	1
Chemical industry	0.72	6.14	2.34	-
Waste and waste water treatment	0.24	1.22	5.41	0.3
Paper and wood production processing	0.56	0.22	3.17	9
Animal and vegetable products from the food and beverage sector	0.05	-	-	-
Other activities	0.03	0.01	-	-
Total	25	56	632	219

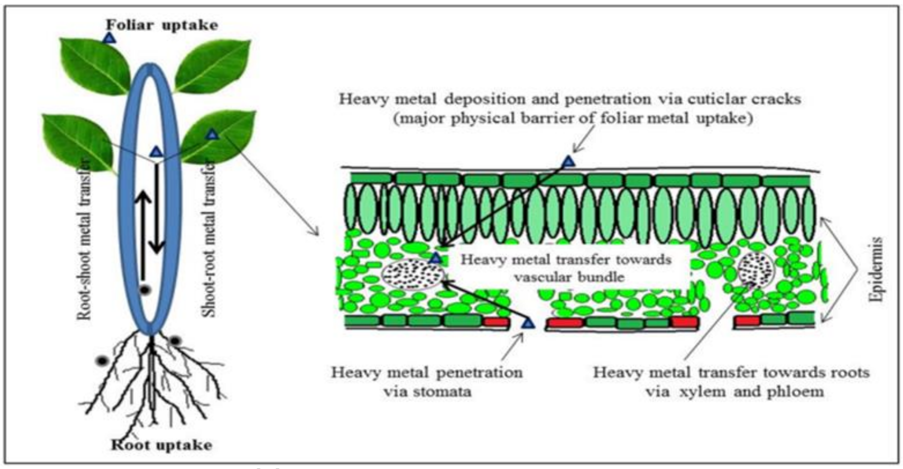
Source: Shahid *et al*., 2017 [81]

Most of the metals ions are water insoluble and immobilized in the apoplastic and symplastic compartments after forming carbonates, sulfate, or phosphate precipitates (Garbisu and Alkorta, 2001; Raskin *et at*., 1997) [27, 75]. Non-cationic metal chelates enter to plants through apoplastic movement, as cell wall needs a higher exchange capacity for cations (Raskin *et al*., 1997) [75]. Heavy metal ions enter to the xylem via root symplasm and, metal transportation in the xylem is mediated by proteins (Thakur *et al*., 2016) [96].

Zolfaghari *et al*., 2018 [118] studied the heavy metal concentration (Cd, Pb, and As) of soil and plants (roots and leaf/seed of wheat, corn, tomato) of agriculture fields situated the vicinity of industrial and rural areas in Iran. Highest metal content (Cd, Pb and As) was observed in the root part of each plant than the soil and leaf/seed.

2.4.2 Foliar uptake of HMs

Atmospheric pollutants are emitted by various sources as particulate matter (PM) or aerosols (Table: 06). Most of HMs emitted as volatile



Source: Shahid *et al*., 2017 [81]

Fig 2: Mechanism of foliar uptake of heavy metals

compounds or very fine particles and, deposit on plant surfaces, enter to plants through the penetration via stomata, cuticles/cuticular cracks, lenticels, ectodesmata, aqueous pores and finally accumulate in the underlying tissues (Fernández *et al*., 2013) [24].

Generally, foliar uptake of heavy metals occurs via: (1) adsorption and internalization via cuticle; (2) penetration of metals via stomatal pores (Schreck *et al*., 2013) [21] (figure 2). Among the various factors governing this process, stomata size and density, and leaf surface area plays a major role. Rao and Dubey (1992) [54] proposed that efficiency of dust collection by plants depend on the morphological factors such as stomatal index, trichome density and length.

Many previous studies reported that plants and vegetables growing near smelters, roadside or industries show elevated levels of foliar HMs (Shahid *et al*., 2013; Uhlig *et al*., 2001) [55, 8]. As an example, Pb in France 50-400 µg/g (Quinche *et al*., 1969) [34], Bahrain 9-420 µg/g (Madany *et al*., 1990) [33] and Canada 100-3000 µg/g (Cannon *et al*., 1962) [29]. Schreck *et al*., 2013 [21] observed a higher level of heavy metals such as Zn, As, Sn, Pb, Cu and Cd taken via leaves in vegetables grown nearby battery recycling factory in France.

2.4.3 Translocation of heavy metals in plants

Lead (Pb)

The main source of emitting lead to the environment has been identified as the burning of petrol during the last decade; however, developed countries have decreased it markedly by introducing unleaded petrol in recent past (Järup *et al*., 2003) [35]. Lead present in plant growing media normally binds to root surface and cell walls, thus make it limit the translocation to shoots and leaves (Cobb *et al*., 2000) [13]. Thus, it would be expected a higher level of Pb content in the root part of the plants.

Several studies have shown that Pb can be strongly retained in many soils, thereby hindering Pb mobility into plant tissues (Nolan *et al*., 20003) [64]. Acute exposure may cause lead poisoning. International agency for research on cancer (IARC) has classified lead as 'possible human carcinogen' based on animal data, however, the overall evidence for lead as a carcinogen being only weak (Steenland *et al*., 2000) [90].

Arsenic (As)

A widely distributed metalloid, found in rock, soil, water and air. Humans are exposed to as via food and drinking water. As accumulation in soil predominantly occurs through the application of arsenic containing pesticides and herbicides (Woolson *et al*., 1971) [22]. Arsenic can exist in plants as arsenate (AsO_4^{-3}) or arsenite (AsO_3^{-3}). Arsenate is lesser in phytotoxicity and translocation than arsenic. Arsenic behaves similar to phosphate and translocate from roots to mature leaves (Cobb *et al*., 2000) [13].

Cadmium (Cd)

Cd is a highly toxic metal and mainly used in manufacturing of re-chargeable nickel-cadmium batteries and phosphate fertilizers. Cd emission was increased in recent past as these products are recycled very rarely, and often dump together with household waste. Cigarette smoking is the major source of human exposure. Plants rapidly uptake Cd^{2+} through the roots via metal transporters.

Cadmium is a highly mobile metal and found to accumulate in plants in large amounts without showing phytotoxic symptoms. Cd has tendency to bind with organic matter and thus taken up by plant and enter to the food supply. John *et al*., 1972 [38] observed that cadmium was not immobilized in the roots, thus more accumulation would be expected in the above ground part of the plants.

Zinc (Zn)

Zinc is an essential trace nutrient for plants and humans at minor level and, toxic at higher concentrations. It plays an important role in protein synthesis and catalytic functions. Zn shows a fairly low concentration in surface water due to its restricted mobility from the place of rock weathering or from the natural sources (Nazir *et al*., 2015) [63].

Copper (Cu)

Cu is the third most widely consumed metal globally, after iron and aluminum. Copper is released into environment by mining, farming, and waste water releases into rivers and lakes. Usually copper attaches to particles of organic matter, clay, soil, or sand. Copper plays an important role in plants for CO_2 assimilation and ATP synthesis. Excess level of copper causes cytotoxic effect, includes stress and injury to plants. High concentrations of Cu may cause metal fumes fever, hair and skin discolorations, and dermatitis in human beings (Khan *et al*., 2008) [45].

Nickel (Ni)

Nickel available in natural soils at trace concentrations and, level can be elevated due to anthropogenic activities such as mining, smelters, burning coal and oil, use of phosphate fertilizers and pesticides. Exposure to nickel produces a specific form of dermatitis and may include the lining and nasal cavity cancer (Chukwu, 2011) [11].

Iron (Fe)

Iron makes up 5% of the earth's crust and is second in abundance to aluminum among the metals. Excess iron introduced to natural ecosystem in the liquid waste streams of many industries and, leachates from municipal solid waste landfills. Iron is an important mineral for all plants and provides important biological roles such as photosynthesis, chloroplast and chlorophyll biosynthesis. The U.S Recommended Daily Allowance for iron is 18mg (Table 7).

Table 7: Recommended daily intake of minerals

Mineral	Recommended Daily Intake (RDI)
Na	2400mg
Mg	350mg
K	3500mg
Ca	1000mg
Fe	15mg
Mn	5mg
Cu	2mg
Zn	75µg
Cr	120µg

Source: Withanage *et al*., 2015[110]

Chromium (Cr)

Cr mainly found in rocks, soil, plants and animals. Cr ions are bound to water very tightly and stable in sediments in water. Cr is a major factory run-off pollutant and, hexa-valent form is most toxic species of Cr (carcinogenic to humans). Chromium compounds are highly toxic to plants and are detrimental to plant growth and development (Geiger, 2010) [28].

Mercury (Hg)

Mercury mainly available as inorganic mercury or organic mercury. Metallic mercury commonly is used in thermometers, barometers, and blood pressure measuring instruments. Mercury amalgam is still used for filling teeth in many countries (WHO, 1991) [105]. Mercury in organic compounds such as methyl mercury is very stable and accumulated in the food chain. Generally, humans are primarily exposed to mercury via food, fish being a major source of methyl mercury exposure (WHO, 1990) [106]. Acute exposure of mercury may cause lung damages, kidney damages, and allergen symptoms and, nerve system damages (Langworth *et al*., 2002) [50].

3. Identification of contamination level

3.1 Translocation Factor (TF)

Translocation factor indicates the ability of plant to translocate metals from the roots to the aerial parts of the plants.

$$TF = \frac{C_{(Stems\ and\ Leaves)}}{C_{Roots}} \quad (1)$$

Where, $C_{Stem\ +Leaves}$ and C_{Roots} represent the heavy metal concentration of areal part and roots respectively. Metals that are accumulated by plants and largely stored in the roots of plants are indicated by TF value lower than 1 and, the values greater than one indicates the translocation of heavy metals to the aerial parts (Marchiol *et al*., 2004)[58].

3.2 Bio-Concentration Factor (BCF)/ Transfer Factor (TF)

Metal concentrations in plant and soil extracts are calculated on the basis of dry weight (Khan *et al*., 2008) [45]. Bio-concentration factor can be defined as the ratio of heavy metal content of crop to soil.

$$BCF = \frac{C_{Plant}}{C_{Soil}\ in\ same\ region} \quad (2)$$

Where, C_{plant} and C_{soil} represent the heavy metal concentrations of plant and soil on dry basis respectively (Cui *et al*., 2005) [15]. TF gives the extent of heavy metal accumulation in soils. Lian *et al*., 2019[52] studied about transfer of heavy metal from soil to crops and found that Cd has high mobility to transfer from soil to crops. In most crops, decreasing trend of BCF value observed as Cd>Zn>Hg>Pb. In addition to that leafy vegetables and rootstalks, and fruit vegetables showed a higher BCF's than rice, maize and soybean grains for all types of metals.

4. Impact of organically amended soils on heavy metal availability

Application of synthetic fertilizer and sludge has been restricted for organic agriculture as scientific evidences shown that synthetic fertilizer may contain hazardous heavy metals. In contrast of that, organic agriculture highly relies on organic fertilizers, animal manures, and copper based fungicides, which could result considerable level of HMs in organic agriculture (Zaccone *et al*., 2010) [114]. As a result, it might be expected a lower level of trace elements in the organic fields.

Compost based agricultural systems are booming all around the world during recent years, as a consequence of negative impact of synthetic chemicals as well as higher consumer demand on such foods. Compost generally has higher C/N ratio and which would ultimately cause extra pressure for the plants which are growing under these systems. Nevertheless, the main problem associated with compost is its high concentration of heavy metals. Secondly, some undesirable materials such as thin glass and plastics can be added to the soil, and at last pollution of the soil.

HMs tend to form complexes with organic matter in the soil (humic and fulvic acids) and affinity of each metal towards organic matter is different. In addition to that, organic matter retaining HMs in an easily exchangeable form. According to Jardao *et al*., 1989 [39] the degree of binding of heavy metals with organic matter can be observed as Cu>Zn>Pb>Cd.

In contrast of that, several researches have shown that addition of humified organic matter in the form of compost can induce the formation of stable, insoluble complexes with humic substances that reduce metal mobility and availability for plant uptake (Orlov *et al*., 1994) [65]. However, organic production of agricultural products could not always allow reducing heavy metal uptake in plants when compared to conventional farming (e.g., Ni, Pb, Zn) (Zaccone *et al*., 2010) [114].

The possibility of contaminating foodstuffs via environment and food processing is equal in all farming systems. There are still uncertainties about the chemical process of recycling of waste materials and releasing of chemical in the soil and plants (Pussemier *et al*., 2006) [72]. Thus, thorough analysis required to confirm the safety of organic products.

Limited evidence exists for comparisons of HM content in organic and conventional products. Although the mineral and trace elements (such as Ca, Fe, P, Mn, Zn, Cu and K) in organic and conventional products have gained great interest of researchers, few have reported slightly high level in organic vegetables (Schuphan, 1974; Auclair *et al*., 1995) [79, 4], majority of evidence has reviled no significant differences (Peavy & Greig, 1972; Clarke & Merrow, 1979; Lairon *et al*., 1984; Termine *et al*., 1987; Perez-Llamas *et al*., 1996; Warman & Havard, 1996; Tan *et al*., 1998; Fjelkner-Modig *et al*., 2000) [67, 12, 49, 93, 69, 104, 92, 25].

Cabrera *et al*., 1989[9] studied the influence of urban compost on heavy metal level in soil and plants, and observed a noticeable increase of total content of Cu and Zn in soil, and level of other metals were negligible. The reason they given was the application of urban compost in large doses.

Pinamonti *et al*., 1997 [70] evaluated the effect of compost on the heavy metal level in soil and plants using compost prepared by different sources such as cattle manure, sewage sludge and poplar bark, and municipal soil waste. The use of compost prepared by municipal solid waste (MSW) for an extended period (over 6 years) significantly increased the concentration of Zn, Cu, Ni, Pb, Cd, and Cr in the soil.

Gallardo-Lara *et al*., 1987 [26] showed that application of organic matter increases the level of Cu and Zn in plants. However, cow manure reduces the plant uptake of heavy metals due to its inherent ability to increase the soil pH slightly. A study undertaken in 2002 in France (Malmauret *et al*., 2002) [57] to monitor the contamination level of heavy metals (Pb, Cd, As and Hg) in organic and conventional raw materials, there was no significant evidences showed to differentiate two groups. However, the recommended levels were exceeded for Pb in organic carrot and buckwheat, and in conventional wheat; for Cd, in both methods.

Another study conducted by Vrček *et al*., 2014 [101] in Croatia for wheat flour, observed the concentrations of Al, As, Cd and Pb were significantly lower in organically produced sample. Similar results have observed by Wieteska and Drzewiṅska (1999) [107] in Poland, Ščančar *et al*., 2004 [78] in Slovenia and Müller *et al*., 1998 [61] Germany.

Hajšlová *et al*., 2005 [30] noticed that there was no consistency difference observed for toxic minerals in organic and conventional potato tubers. Jorhem and Slanina (2000) [40] studied heavy metal (Cd, Pd, Cr and Zn) accumulation in organically and conventionally cultivated wheat, rye, carrot and potatoes for two consecutive harvests. The results showed that there was no clear-cut correlation between Cd content of food stuff and cultivation system. Due to contradictory results observed, more extensive studies recommended over a prolong period of time.

Pandey *et al*., 2009 [66] studied the effect of atmospheric deposition of trace metal in organically grown vegetables in India, and suggested that atmospheric deposition was the main contributor to raise the level of heavy metals in edible parts, especially fruits and leaves. This could cause serious implications since organic farming is now increasingly being recognized as a future sustainable agriculture to obtain hazard free agricultural produce.

5. Attempts and novel strategies for remediation

There are number of research have been conducted to identify the strategies that can be used to minimize the contamination of ecosystem by heavy metals.

In this context, phytoremediation can be considered as economical and very valid option for lightly polluted areas (Kumar *et al*., 1995) [47]. Phytoremediation can be accomplished by several means including phytoextraction, phytostabilization, phytovolatilization and phytodegradation (Mellem *et al*., 2009) [60].

Bio-sorption is one of the processes which involves a solid (sorbent) and a liquid (solvent) which containing dissolved materials attracted by affinity, through several mechanisms, involving chemisorption, physisorption, complexation, ionexchange, chelation and entrapment (Côrtes *et al*., 2015; Kang *et al*., 2016; Mahajan and Sud, 2012; Wang *et al*., 2017) [14, 42, 56, 102]. Use of biosorbants is an ecofriendly technology for treating industrial and domestic waster waters (Kuppusamy *et al*., 2015) [48]. Even though, phytoremediation has been proposed for removal of heavy metals in landfill leachate, however, there are problems with this method due to low performances.

Sewage sludge is rich in organic and inorganic plant nutrients and its application in to the agricultural fields is abundantly practiced by most of the countries as most desired disposal method. However, due to the long-term irrigation with waste water may resulted in elevated levels of HMs. Thus, due to sever threat for human health, use of waste water often restricted by regulatory bodies (Sing *et al*., 2008) [86].

Tervahauta *et al*., 2014 [94] have studied the possibility of using black water for agriculture lands instead of sewage water for the areas where water is limited, and suggested that, contamination of soil/food cycle can be minimized by application of black water.

Turan *et al*., 2018 [97] conducted an experiment to evaluate the effect of chitosan and charcoal on growth and nutritional quality of brinjal plant together with in situ immobilization of heavy metals. In this experiment, they used a polluted soil and further irrigated with contaminated water and observed that, application of water in high rates together with charcoal or chitosan significantly reduce the heavy metal concentrations in the plant and thereby improvement in the growth too.

6. Analytical techniques for determination of heavy metals

6.1 Sample preparation techniques

Heavy metals naturally existing as complexes with different substances (organic or inorganic) and, releasing of HMs via digestion is very important for obtaining reliable results for the analytes (Polkowska-Motrenko., *et al* 2000) [71]. The wet and dry ashing procedures are slow and difficult to carry out operate successfully and, microwave digestion has been reported as an efficient and rapid method (Kingston *et al*., 1986) [46]. Most of the analytical techniques are principally dedicated to analyze liquid samples and there for all solid samples have to brought into solution form.

6.2 Analytical techniques

Colorimetry

Transitional metals complexes often form highly colored solutions due to electronic transition with energy light that corresponds to wavelengths in the visible region. Hence, light is passed through the solution of metal containing and, specific wavelength would be absorbed that correspond to energy of the electronic transition (Auting Peavy University) [5].

Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

The analyte elements can be analyzed by collisions with Ar+, exited argon atoms or energetic electrons. This technique is expensive but offers the advantages of good sensitivity, limits of detections (LODs), throughput, and simultaneous multi-element determination. To yield the most accurate and repeatable results, the ICPMS can be used in conjunction with an isotope dilution quantitative methodology (ID-ICP-MS).

Inductively coupled plasma atomic emission spectrometry (ICP-AES)

This method is well suited for the determination of refractory elements with high sensitivity because very high temperatures (8000 K) are attained with plasma. It has better LODs to flame atomic absorption spectrometry (FAAS) but can detect many elements simultaneously and has a much larger linear dynamic range than AAS-based techniques. It is susceptible to interferences at low concentrations (Vandecasteele, 1993) [100].

X-Ray Fluorescence (XRF)

X-ray spectroscopy is based upon the measurement of emission, absorption, scattering, fluorescence, and diffraction of electromagnetic radiation. The most used X-ray methods are the XRF and X-ray absorption for quantitative and qualitative determination of all elements in the periodic table having atomic numbers greater than that of sodium. Elements with atomic numbers in the range of 5 to 10 require special equipment (Skoog *et al*., 2005) [88].

Neutron Activation Analysis (NAA)

Measures the radioactivity that has been induced in samples by irradiation with neutrons or charged particles such as hydrogen, deuterium, or helium ions (Skoog *et al*., 2005) [88].

Anodic Stripping Voltammetry (ASV)

ASV is an electroanalytical technique with the lowest LOD for metals. Its LOD for solutions is in the range of 10-6 to 10-9 M (Rubinson *et al*., 2000) [77].

Adsorptive Stripping Methods

In this method the microelectrode, HMDE, is immersed in a stirred solution of the analyte for several minutes. The analyte is deposited by physical adsorption on the surface of the electrode rather than electrolytic deposition. The stirring is discontinued after enough of the analyte have built up. The deposited material is then determined by linear scan or pulsed voltammetry measurements. Quantitative information is based upon standard solutions treated in a similar manner to the samples

Ion Chromatography

With present method, a single separation experiment enables cations or anions to be quantified at the level of parts per billion (ppb). The retention time of the bands identify the ions and the area under the bands gives the quantity of the ion. Ions are detected by measuring the electrical conductivity of the effluent. When the bands containing ions pass through the detector, conductivity rises (Rubinson *et al*., 2000) [77].

Atomic Emission Spectroscopy (AES)

In AES analyte concentration is determined by measuring the quantity of optical emission of excited atoms. The analyte atoms are sucked into the excitation region where they are desolvated, vaporized, and atomized by a flame, discharge, or plasma. Plasma source is preferred to a flame source in that they are hotter and more homogenous in temperature than flame. The energy to excite the atoms is provided by the high temperature atomization source. The excited atoms on returning to the ground state emit light that gives emission spectra because the transition occurs between distinct atomic energy levels

Atomic Absorption Spectroscopy (AAS)

AAS is the most widely used and accepted technique capable of determining trace (µg/mL) and ultra-trace (sub µg/mL) levels of elements or metals in areas of environmental, clinical, biological, food, and geological samples with good accuracy and precision (Sneddon, 1997) [89].

Flame Atomic Absorption Spectrometry (FAAS)

The FAAS consists of six components: a radiation source, flame atom cell, sample introduction unit, monochromator, detection system, and readout. The hollow cathode lamp (HCL) is the most widely used radiation source. The HCL consists of a glass envelope that is filled with an inert gas, usually neon, argon, or helium at low pressure

Graphite Furnace Atomic Absorption Spectrometry (GF-AAS)

This method requires smaller sized samples (1-100 µL) compared to FAAS (minimum volume 1-2 mL) and is more sensitive than FAAS. It also has limit of detection (LOD) that is 10-100 times better than those of FAAS. In spite of the above, it's still unable to determine refractory elements with acceptable precision (Brown and Milton, 2005) [7].

7. Conclusion

Heavy metals are bio-accumulated and bio-transferred both by lithogenc and anthopogenic sources. The contamination of water and plants is one of the major issues faced throughout the world and, requires a close attention due to exposure over the threshold limits have created threatening effect to human and animal lives.

Continues use of chemical fertilizers and pesticides in over dose have given a great impact on rising the heavy metal concentration of fruits, vegetables and ground water aquifers. Most of countries where water is scared use waste water to irrigate their agricultural lands as an alternative for waste disposal and, it has been reported that such water mainly provides heavy metals such as chromium, nickel and zinc to the food chain. An excess level of As and Cd have been reported in phosphate fertilizers and As in glyphosate based herbicides formulas in Sri Lanka, and excess application of such ingredients have created serious health implications in the north central area of the country.

Due to safety issues reported on conventional agricultural products, a trend has been reported that most people turn towards the organic products in recent past. Even though, the term "organically grown food denotes products that have been produced in accordance with the principles and practices of organic agriculture, a thorough study should have to be conducted to analyze the safety of organically produced items.

8. References

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