BIOACCUMULATION AND TRANSLOCATION OF HEAVY METALS IN DIFFERENT FRUIT PLANTS WITH CORRESPONDING HEALTH RISK ASSESSMENTS

By

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CANDIDATE’S DECLARATION

It is hereby declared that this thesis or any part of it has not been submitted elsewhere for the award of any degree or diploma.

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Certification of Thesis

A Thesis on

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Dedicated
To
My Beloved
Parents

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Abstract

The contamination of crops, vegetables, and fruits with heavy and toxic metals has become one of the most severe environmental pollution problems nowadays in many developing countries like Bangladesh. Heavy metals can be released into terrestrial environment from natural and anthropogenic activities and accumulated in waterbodies and soil from which they can transfer into fruit plants through various pathways and thus can adversely affect human health. In the present study, accumulation and mobilization status of various heavy metals and metalloids have been determined in water, arable soil, and different fruit plants grown in and around Dhaka Export Processing Zone (DEPZ) located in Savar, Dhaka. Total of ten metals (Pb, Cd, Cr, Cu, Fe, Zn, Ca, Co, Ni, Mn) and 2 metalloids, (As and Se) have been analyzed in different parts (root, stem, leaves and edible parts) of three common fruits plants such as Guava (*Psidium guajava*), Pomelo (*Citrus maxima*), Banana (*Musa paradisiaca*) as well as in their growth media (water and soil). All metals and metalloids contents in the different environmental samples (fruits, soil, and water) were determined by using Atomic Absorption Spectroscopy (AAS) as well as Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The distribution and transfer of heavy metals and metalloids from water and soil into different parts of various fruit plants were investigated accordingly. Different pollution indices such as Heavy metal transfer factor (TF), Estimated daily intake (EDI), Target hazard quotient (THQ), Hazard index (HI), Carcinogenic risk (CR) factor, Food pollution index (FPI), Soil threshold values and food safety (STV), Geoaccumulation index (Igeo), and Enrichment factor (EF) were analyzed to assess twelve heavy metals and metalloids contamination, their distribution and mobilization in irrigation water, arable soil, and in different parts of fruits plants. Pearson's correlation analysis was applied to identify corresponding distributions of heavy and toxic metals in different fruit plants. Metals and metalloids concentrations found in the respective environmental samples were analyzed statistically by using Minitab software. Average concentrations of twelve different elements in water and soil were found to be in the order of: Ca > Fe > Zn > Cu > Mn > Cr > As > Pb > Ni > Co > Se > Cd and Fe > Ca > Mn > Zn > Pb > Cr > Ni > Cu > Co > As > Se > Cd respectively. Heavy metals contents found in water and soil were exceeded the permissible levels of WHO and USEPA suggesting metals pollution in the respective environment components. Different fruits plants showed variation in the uptake and deposition of heavy metals and metalloids and the average concentrations of different elements found in various fruits species followed the order of Ca > Fe > Mn > Cu > Zn > Pb > Cr > Ni > Se > Co > Cd > As. Evaluation of the mobilization of metals and metalloids in different parts of various fruits plants bodies revealed the occurrence of the highest accumulation of the elements in leaves whereas the lowest uptake of the respective metals was observed in the edible parts irrespective of the plant bodies. The extents of different heavy metals found in the edible parts of various fruits plants was relatively higher than the tolerable limits recommended by FAO and WHO. Out of three sources, heavy metals and metalloids contents in soil were much higher than those observed in water and four parts of each of the three fruits plants bodies. The extents of Pb, Cr and Mn in three fruit species might cause higher non-carcinogenic risks as evidenced from the risks assessment data. Carcinogenic risks (CR) analysis data showed the range as E-04 < CR > E-06 which suggest that Cd, Cr and Ni enriched fruits could pose a considerable cancer risk for both adults and children The significant findings of this research work suggest that the study area is highly unsafe for growing fruits, crops, and vegetables as they accumulate and deposit excessive quantities of heavy metals and other elements which severely affect food safety and thus poses potential health risks to millions of people in Bangladesh.

KEYWORDS: Fruits, Plant parts, Soil, Heavy Metals, Food pollution index, Transfer factor, Health Risks, DEPZ.

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2. Inductively coupled plasma mass spectrometry (ICP-MS)
3. Lead (Pb)
4. Cadmium (Cd)
5. Chromium (Cr)
6. Copper (Cu)
7. Zinc (Zn)
8. Iron (Fe)
9. Calcium (Ca)
10. Manganese (Mn)
11. Cobalt (Co)
12. Nikel (Ni)
13. Aresenic (As)
14. Selenium (Se)
15. World Health Organization (WHO)
16. Department of Environment (DoE)
17. Canadian Council of Ministers of the Environment (CCME)
18. United States Environmental Protection Agency (USEPA)
19. Toxicity Reference Value (TRV)
20. Lowest Effect Level (LEL)
21. Heavy metal transfer factor (TF)
22. Estimated daily intake (EDI)
23. Target hazard quotient (THQ)
24. Hazard index (HI)
25. Carcinogenic risk (CR) factor
26. Food pollution index (FPI)
27. Soil threshold values and food safety (STV)
28. Geoaccumulation index (Igeo)
29. Enrichment factor (EF)

CHAPTER-1

Introduction

1. Introduction

1.1. General remarks

Food safety is a major public concern worldwide and food consumption has been identified as the major pathway for human exposure to certain environmental contaminants, accounting for > 90% of intake compared to inhalation or dermal routes of exposure. About 30% of human cancer are caused by low exposure to initiating carcinogenic contaminants in the diet. During the last decades, the increasing demand for food safety has stimulated research regarding the risk associated with the consumption of food contaminated by pesticides heavy metals, or toxins [1]. Globally, human health and the environment are currently at high risk from food contaminated with heavy metals and other related sources. Heavy metals enter the food chain by natural contamination or because of human activities. The heavy metals accumulate in humans and animals through bio-magnification effects, thus slowly causing toxicity and resulting in serious health problems. Long-term exposure may cause progressive neurological and muscular degeneration that may result in Parkinson's, muscular dystrophy, Alzheimer's, and multiple sclerosis [2]. Heavy and toxic metal contamination in fruits, soil, and water has drawn significant attention in recent years because of toxicity, abundance, persistence, and subsequent excessive accumulation property [3]. Fruits are commonly consumed by people of all ages all over the world especially in tropical countries as a routine part of daily diet [4]. Fresh vegetables and fruits play an important role in human nutrition due to their high nutrient content of vitamins, such as vitamins B, C, K, and minerals such as calcium, potassium, and magnesium, as well as dietary fibre [5]. *Psidium Guajava*, *Citrus Maxima*, and *Musa Paradisiaca* are the most common and popular fruits grown in Bangladesh. Due to the scarcity of available agricultural lands, varieties of fruit species are grown in and around different industrial areas in Bangladesh. Fruit plants could readily uptake heavy and toxic metals over a wide range of quantities from the growth mediums through their root systems, leading to the accumulation of toxic metals in plant tissues if they are cultivated in a polluted environment. Fresh fruits and vegetables provide a healthy and balanced diet and can prevent chronic diseases such as heart disease, cancer, diabetics, and obesity including several micronutrient deficiencies especially in developing countries [6]. From Plants fruits contain both essential and toxic metals over a wide range of concentrations. Trace metals have been found to play both positive and negative roles in human health. They can be classified as toxic (arsenic, cadmium, lead, mercury, nickel, etc.), probably essential (vanadium), and essential (copper, zinc, iron, manganese, selenium, and cobalt) metals. However, toxic effects of the last two classes of metals have also been identified when the intake is excessively high. The average per capita daily intake of fruits in Bangladesh is 44.7 g, respectively [7]. Heavy metals are non-biodegradable and could persist for a long time in the environment. When fruits are cultivated in polluted environments, they could readily absorb heavy metals through the leaves or roots, leading to the accumulation of toxic metals in plant tissues. The entry of heavy metals into the food chain not only inhibits the normal physiological functions of the human body, but it also affects the growth, nutrient uptake, nitrogen fixation, and metabolism of plants [8]. Due to the properties of heavy metals like high bio accumulative potential, persistence, and toxicity, heavy metal pollution is a major environmental concern. Trace amounts of some metals are required for the growth and functioning of living organisms, but if consumed in higher quantities, they can be toxic for humans and aquatic life. A high concentration of toxic metals in groundwater enters the food chain and causes substantial risk. Exposure to heavy metals causes severe human health implications such as infertility, neurotoxicity, and cardiovascular and skeletal diseases in human beings.

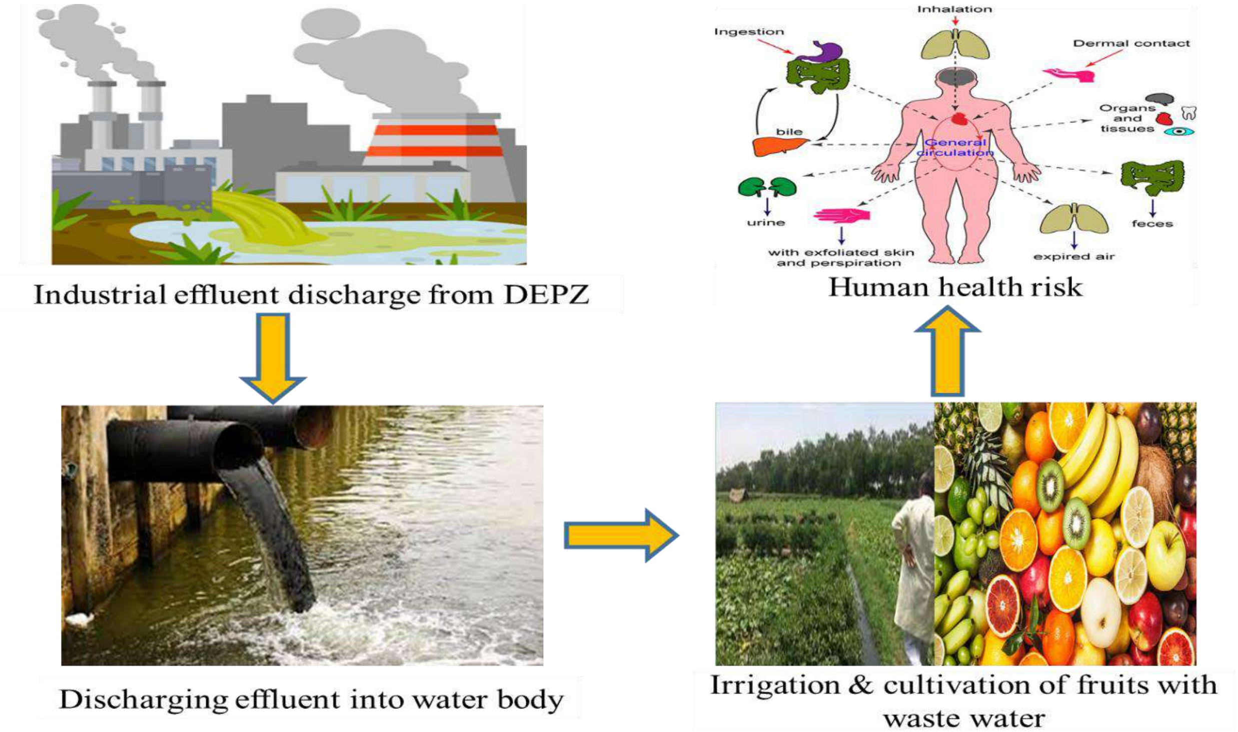
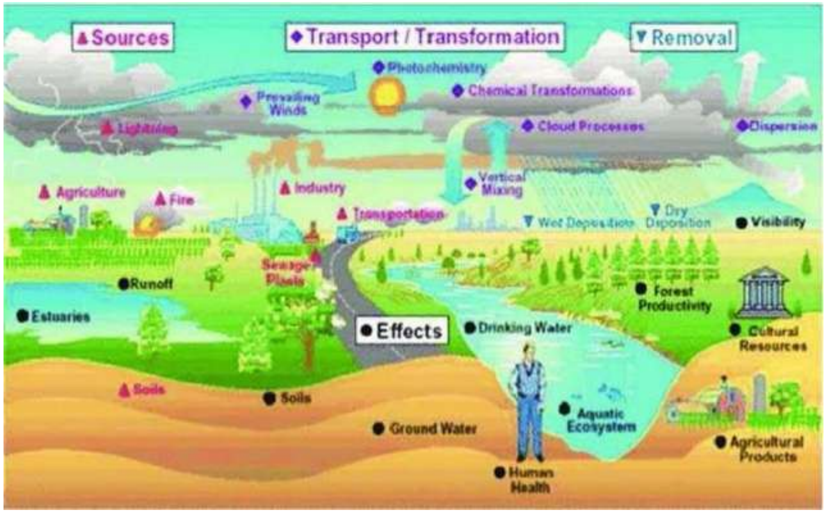


Figure 1.1: Mobilization of heavy and toxic metals to the human body

Moreover, a genotoxic carcinogen is a group of toxicants that causes liver and kidney disorders in humans due to exposure to heavy metals [9].

1.2. Environmental pollution

Environment is referred to as someone's surroundings that consist of atmosphere, hydrosphere, and lithosphere, having resources for life sustenance [10]. Environmental quality and sustainability are the two major indicators of sustaining the healthy human life in the planet that we live in. An environment can be ideal and highly sustainable for the survival of human beings or can be polluted or contaminated. Environmental pollution is the undesirable and unwanted change of our surroundings caused totally or largely by human activities. Direct or indirect effects of the changes in energy pattern, radiation levels, chemical and physical constitution and abundance of organisms are the main cause for environmental pollution. Due to having severe long-term consequences, environmental pollution has now become a global problem including both developed as well as developing countries, and so is drawing a significant attention in recent years [11]. Environmental pollution can be caused by the foreign substances or even by its own component when present above the natural background level [12]. The substance in this case that causes contamination is called contaminant or pollutant



[Figure reference: S. Karim et al. *Arab. J. Chem. Environ. Res.* 04, (2017), 01-17]

Figure 1.2: Environmental pollution (sources, transportation, transformation, removal, and effects)

1.2.1 Contamination of atmosphere (air pollution)

The release of various gases, finely divided solids, or finely dispersed liquid (aerosols) into the atmosphere at a rate responsible for exceeding the natural capacity of the environment to dissipate and dilute or absorb them is referred to as air pollution. Such a high concentration of these substances may cause undesirable health, economic, or aesthetic effects. Major Air Pollutants are classified into different categories: **Criteria Pollutants, Fine Particulates**, and **Air Toxics**. The criteria pollutants are the substances whose concentrations in the atmosphere indicate overall air quality. According to U.S. Environmental Protection Agency (USEPA) there are six major air pollutants that have been considered as criteria pollutants which are carbon monoxide (CO), nitrogen oxides (NO and NO2), sulfur dioxide (SO2), ozone (O3), particulate matter and lead (Pb). Fine particulates generally refer to very small fragments of solid materials or liquid droplets suspended in air. Rather than by chemical composition, they are usually characterized on the basis of their sizes and phases (solid or liquid) except for airborne lead, as it is treated as a separate category. For example, solid particulates between roughly 1 and 100 μm in diameter are called dust particles, whereas airborne solids less than 1 μm in diameter are called fumes. Air toxics are substances which are hazardous even when present in trace amounts in the air. Some of them cause different health problems, such as adverse effects on brain tissue or fetal development while many of them cause genetic mutations or cancer.

Although, the air toxics are comparable to the criteria pollutants, these pollutants can pose an immediate health risk to exposed individuals and can cause other environmental problems. Major air toxics are organic chemicals, carbon containing molecules, hydrogen, and other atoms. Volatile organic compounds (VOCs) are another major source of air toxic. VOCs include pure hydrocarbons, partially oxidized hydrocarbons, and organic compounds containing chlorine, sulfur, or nitrogen. Some examples of air toxics are arsenic, asbestos, benzene, chlorine, coke oven emissions, cyanide compounds, mercury compounds, radionuclides (radon, radium, uranium), selenium compounds, vinyl chloride etc. [13].

1.2.2 Contamination of hydrosphere (water pollution)

Water pollution is the release of substances (that interferes with beneficial use of the water or with the natural functioning of ecosystems) into subsurface groundwater or into lakes, streams, rivers, and oceans. In addition, water pollution may also be caused by the release of energy, in the form of radioactivity or heat, into bodies of water. A wide variety of substances can cause pollution to water bodies, such as pathogenic microorganisms, putrescible organic waste, plant nutrients, toxic chemicals, sediments, heat, petroleum (oil), and radioactive substances. Different types of water pollutants generated by human activities are Domestic sewage, Toxic waste, Sediment, Thermal pollution and Petroleum (oil) pollution. Domestic sewage is the primary source of pathogens (disease-causing microorganisms) and putrescible organic substances. Pathogens exert a direct threat to public health and putrescible organic matters present a different sort of threat to water quality. Decomposition of theses organics include consumption of dissolved oxygen and this content of the water is depleted which endangers the fish and other aquatic organisms as high levels of oxygen are required for their subsistence. When the waste is poisonous, radioactive, explosive, carcinogenic, mutagenic, teratogenic (causing birth defects), or bio accumulative, then it is considered as a toxic waste. Discharging of industrial effluents without proper treatment and surface runoff containing pesticides used on agricultural areas are the main sources of toxic wastes. Suspended sediments interfere with the penetration of sunlight into different layers of water and interrupts the ecological balance of water body. Sediment resulting from soil erosion can be carried into water bodies by surface runoff. Heat decreases the capacity of water to hold dissolved oxygen in solution, and it increases the rate of metabolism of fish that’s why, heat is considered to be a water pollutant. Discharging of cooling water from power plants into rivers is a major source of thermal pollution. Accidental oil spills and surface runoff from roads and parking lots into water bodies are the major sources of Petroleum (oil) pollution [14].

1.2.3 Contamination of lithosphere (soil pollution)

Soil is the thin layer of organic and inorganic materials that covers the Earth's rocky surface. The organic portion (derived from the decayed remains of plants and animal) is concentrated in the dark uppermost topsoil and inorganic portion (made up of rock fragments) was formed over thousands of years by physical and chemical weathering of bedrock. Consistence of toxic compounds, chemicals, salts, radioactive materials, or disease-causing agents (having adverse effects on plant growth and animal health) in the soil is defined as Soil pollution. Petroleum hydrocarbons, heavy metals, pesticides, solvents are the most common soil polluting chemicals. Different ways for soil pollution are seepage from a landfill, discharge of industrial waste into the soil, penetration of contaminated water into the soil, rupture of underground storage tanks, excess application of pesticides, herbicides, fertilizer and solid waste seepage [15].

1.3 Types of pollutants

Environmental pollution become most alarming global concern in recent years and is one of the great challenges faced by the global community. We encounter a variety of pollutants in our daily life which can be classified into different categories, namely biological, organic and inorganic. Regardless of different categories, all the pollutants receive considerable attention as they possess adversary impacts. The relationship between environmental pollution and world population has become an inarguable directly proportional relationship as it is being observed that the amount of potentially toxic substances released into the environment is continuously increasing with the dramatic rise of global population [16]. As we are much concern about water and soil pollution in this case due to their direct contribution in this research so, will discuss water and soil pollutants more specifically.

1.3.1 Biological pollutants

Biological pollutants are described as contaminants which exist as a result of human activities and which pose negative impacts on the quality of aquatic and terrestrial environment. This type of pollutants include bacteria, viruses, parasites, molds, mildew, animal dander and cat saliva, house dust, mites, cockroaches and pollens [17]. Studies have reported different sources of these types of pollutants, including pollens originating from plants; viruses that are being transmitted by people and animals; bacteria which are carried out by people, animals, soil and plant debris.

1.3.2 Organic pollutants

Organic pollution can be briefly defined as biodegradable contaminants in an environment [18]. Many insecticides and herbicides that that are generally used in pest control and agriculture respectively are included into organic pollutants. Dichlorodiphenyltrichloroethane (DDT) is a pesticide, highly effective in controlling mosquitos, which was banned in the USA in 1972 due to having toxic effect. Although the common sources of organic pollutant are natural (caused by the environment), but to fulfill the human needs anthropogenic activities also have contribution to production of organic pollutants. Some of the common organic pollutants which have been noted to be of special concern are human waste, food waste, polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), polycyclic aromatic hydrocarbons (PAHs), pesticides, petroleum and organochlorine pesticides (OCPs). Organic pollutants have drawn considerable attention in recent years because of having their pernicious effects on the environment. Properties of organic pollutants may include high lipid solubility, stability, lipophilicity, and hydrophobicity which have recently made organic pollutants to be recognized as persistent pollutants [19, 20]. These properties provide organic pollutants the ability to easily bioaccumulate in the different spheres of the environment, thus causing toxicological effects.

1.3.3 Inorganic pollutants

Inorganic pollutants are usually substances of mineral origin, with metals, and salts. In addition, inorganic pollutants are one of the major classes of pollutants discharged by chemical and allied industries such as refineries, fertilizers, and pharmaceuticals. Trace elements, mineral acids, metals, metal compounds, inorganic salts, metals with organic compounds are the common inorganic pollutants.

Inorganic pollutants may be non-biodegradable and they persist in the surrounding environment. Many inorganic pollutants have disruptive effect on public health. Inorganic pollutants enter into the environment through different anthropogenic activities such as sewerage drainage, mine drainage, smelting, metallurgical and chemical processes, as well as natural processes [21, 22]. As this study is based on the heavy metal concentration, we should have a clear idea about this pollutant and so, we will discuss little more about heavy metal.

1.4 Heavy metals

Any criterion-based definition of a heavy metal is not widely agreed. For different context the term can reflect different meanings. We can define metal based on chemical behavior as a chemist would likely be more concerned with chemical behavior. Density criteria ranges from above 3.5

g/cm^3 to above 7 g/cm^3 and weight ranges from greater than sodium (atomic weight 22.98); greater than 40 (excluding s- and f-block metals, hence starting with scandium); or more than 200(from mercury onwards) [23]. The term "heavy metals" is often used as a group name for metals and semimetals (metalloids) that have been associated with contamination and potential toxicity or ecotoxicity. Very recently, a broader definition for the term have been proposed, and heavy metals have been defined as "naturally occurring metals having atomic number greater than 20 and an elemental density greater than 5 g/cm^3 [24]. Among environmental pollutants heavy metals are the most investigated one. Regarding the roles of heavy metals in biological systems, they can be classified as essential and nonessential. Essential heavy metals are important for living organisms while nonessential heavy metals have no known biological role in living organisms. Examples of essential heavy metals are Mn, Fe, Cu, and Zn, while the heavy metals Cd, Pb, and Hg are toxic and are regarded as biologically nonessential [25]. The heavy metals Mn, Fe, Co, Ni, Cu, Zn, and Mo are micronutrients or trace elements for plants. They are essential for growth and stress resistance as well as for biosynthesis and function of different biomolecules such as carbohydrates, chlorophyll, nucleic acids, growth chemicals, and secondary metabolites [26]. Either deficiency or excess of an essential heavy metal leads to diseases or abnormal conditions. However, the lists of essential heavy metals may be different for different groups of organisms such as plants, animals, and microorganisms. It means a heavy metal may be essential for a given group of organisms but nonessential for another one. The interactions of heavy metals with different organism groups are much complex [27].

Almost every heavy metal and metalloid may be potentially toxic to biota depending upon the dose and duration of exposure. Many elements are classified into the category of heavy metals, but some are relevant in the environmental context. List of the environmentally relevant most toxic heavy metals and metalloids contains Cr, Ni, Cu, Zn, Cd, Pb, Hg, and As [28]. Heavy metal pollutants most common in the environment are Cr, Mn, Ni, Cu, Zn, Cd, and Pb. Some other heavy metals are also hazardous to living organisms depending upon dose and duration of exposure. For example, (Mansouri et al. 2012) have found Ag as more toxic than Hg to a freshwater fish [29].

1.4.1 Sources of Heavy Metals in the Environment

Sources of heavy metals in the environment can be both natural/geogenic/lithogenic and anthropogenic. The natural or geological sources of heavy metals in the environment include weathering of metal-bearing rocks and volcanic eruptions. The global trends of industrialization and urbanization on earth have led to an increase in the anthropogenic share of heavy metals in the environment [30].

1.4.1.1 Anthropogenic Sources

The anthropogenic sources of heavy metals in the environment include mining, industrial and agricultural activities. These metals (heavy metals) are released during the mining and extraction of different elements from their respective ores. Heavy metals released into the atmosphere during mining, smelting, and other industrial processes can readily return to the land through dry and wet deposition. Discharge of wastewaters such as industrial effluents and domestic sewage add heavy metals to the environment. Application of chemical fertilizers and combustion of fossil fuels also contribute to the anthropogenic input of heavy metals in the environment. Regarding contents of heavy metals in commercial chemical fertilizers, phosphate fertilizers are particularly important. In general, phosphate fertilizers are produced from phosphate rock (PR) by acidulation. In the acidulation of single superphosphate (SSP), sulfuric acid is used, while in acidulation of triple superphosphate (TSP), phosphoric acid is used. The final product contains all of the heavy metals present as constituents in the phosphate rock. Commercial inorganic fertilizers, particularly phosphate fertilizers, can potentially contribute to the global transport of heavy metals.

Heavy metals added to agricultural soils through inorganic fertilizers may leach into groundwater and contaminate it. The anthropogenic sources of Cr include electroplating industries, leather tanneries, textile industries, and steel industries [31]. Globally, about 50,000 t/year of Cr may be emitted from coal combustion, wood burning, and refuse incineration.

Industrial waste disposal

Use of fertilizer, pesticides, insecticides

Mining and mill waste disposal

Smelting waste disposal

[Figure reference: Sarker, A. et al. *Environmental Science and Pollution Research*. 29(3), (2021), 3230-3245]

Figure 1.3: Anthropogenic sources of heavy metals

1.4.1.2 Natural Sources

The presence of heavy metals in the environment leads to several adverse impacts. Such impacts affect all spheres of the environment, that is, hydrosphere, lithosphere, biosphere and atmosphere. Until the impacts are dealt with, health and mortality problems break out, as well as the disturbance of food chains. Many studies have documented different natural sources of heavy metals. Under different and certain environmental conditions, natural emissions of heavy metals occur. Such emissions include volcanic eruptions, sea-salt sprays, forest fires, rock weathering, biogenic sources and wind-borne soil particles. Natural weathering processes can lead to the release of metals from their endemic spheres to different environmental compartments. Heavy metals can be found in the form of hydroxides, oxides, sulphides, sulphates, phosphates, silicates, and organic compounds. The most common heavy metals are lead (Pb), nickel (Ni), chromium (Cr), cadmium (Cd), arsenic (As), mercury (Hg), zinc (Zn), and copper (Cu). Although the aforementioned heavy metals can be found in traces, they still cause serious health problems to human and other mammals.

Petroleum seep

Volcanic eruption

Metal containing rocks

Surface mineralization

[Figure reference: Sarker, A. et al. *Environmental Science and Pollution Research*. 29(3), (2021), 3230-3245]

Figure 1.4: Natural sources of heavy metals

1.4.2 Effect of heavy metal contamination

Heavy and toxic metals contamination have already become a serious issue of concern around the world as it has gained momentum due to the increase in the use and processing of heavy metals during various activities to meet the needs of the rapidly growing population. Soil, water and air are the major environmental compartments which are affected by heavy metal pollution [32].

1.4.2.1 Effect of heavy metals on soil

Most of the heavy metals do not undergo microbial or chemical degradation because they are non-degradable, and consequently their total concentrations last for a long time after being released into the environment. The accumulation and deposition of heavy metals in soils is a serious issue due to their mobilization and transportation into the food chains which severely impact the entire ecosystem. As much as organic pollutants can be biodegradable, their biodegradation rate, however, is decreased by the presence of heavy and toxic metals in the environment, and this in turn doubles the environmental pollution with the aspects of the presence of both organic pollutants and heavy metals. There are various ways through which heavy metals pose risks to humans, animals, plants, and ecosystems as a whole. Such ways include direct ingestion, accumulation by plants, food chains, consumption of contaminated water and alteration of soil pH, porosity, color, and its natural chemistry which in turn impact on the soil quality [32].

1.4.2.2 Effects of heavy metals on water

Heavy metals can be found in traces amount in water sources and still be very toxic and impose serious health problems to humans and other ecosystems. This is because the toxicity level of a metal depends on factors such as the organisms which are exposed to it, its nature, its biological role and the period at which the organisms are exposed to the metals. Food chains and food webs symbolize the relationships among the organisms. Therefore, the contamination of water by heavy and toxic metals actually affects all organisms. Humans, an example of organism feeding at the highest level, are more prone to serious health problems because the extents of heavy and toxic metals have dramatically increase in the food chain in recent years [33].

1.4.2.3 Effects of heavy metals on air

Increase in industrialization and urbanization, due to the rapid growth of world population have recently caused the air pollution as a major environmental problem around the world. Air pollution has been accelerated by the presence of dust and particulate matters (PMs) in the environment particularly fine particles such as PM2.5 and PM10 which are being released through natural and anthropogenic processes. Natural processes which release particulate matters into air include dust storms, soil erosion, volcanic eruptions and rock weathering, while anthropogenic activities are more industrial and transportation related. Particulate matters are important and require special attention as they can lead to serious health problems such as skin and eyes irritation, respiratory infections, premature mortality and cardiovascular diseases. These pollutants also cause deterioration of infrastructures, corrosion, formation of acid rain, eutrophication and haze. Amongst others, heavy metals such as group 1 metals (Cu, Cd, Pb), group 2 metals (Cr, Mn, Ni, V and Zn) and group 3 metals (Na, K, Ca, Ti, Al, Mg, Fe) originate from industrial areas, traffic and natural sources, respectively [34].

1.5 Sources of heavy metal exposure to humans

Heavy and toxic metals exposure to humans may occur in a variety of ways. During the mining activities, heavy metals are being released from the ores and scattered in the open environment; deposited in the soil, transported by air and water to other areas. Moreover, when these heavy metals are used in the industries for various industrial purposes, some of these elements are released into the air during combustion or into the soil or water bodies as effluents. In addition, different industrial products such as paints, cosmetics, pesticides, and herbicides also serve as sources of heavy and toxic metals. Heavy metals could also be transported through erosion, runoff or acid rain to different locations in soils and water bodies. This type of pollution can be recognized as long range contamination. The heavy metals most commonly associated with poisoning of humans are lead, mercury, arsenic and cadmium. The exposures of heavy and toxic metals are varied depending on their sources which are described below:

1.5.1 Sources of iron exposure

Iron is an attractive transition metal for various biological redox processes due to its interconversion between ferrous (Fe^{2+}) and ferric (Fe^{3+}) ions. The sources of iron in surface water are anthropogenic and are related to different mining activities. The production of sulfuric acid as well as the discharge of ferrous ion (Fe^{2+}) takes place due to the oxidation of iron pyrites (FeS_2) that are commonly found in coal seams [35].

1.5.2 Sources of nickel exposure

Nickel, a known heavy metal is found at very low levels in the environment. Nickel is available in all soil types and meteorites and also erupts from volcanic emissions. In the environment, nickel is principally bound with oxygen or sulfur and forms oxides or sulfides in earth crust. The vast industrial use of nickel during its production, recycling and disposal has led to widespread environmental pollution. Nickel is discharged into the atmosphere either from nickel mining or by various industrial processes, such as power plants or incinerators, rubber and plastic industries, nickel-cadmium battery industries, nickel-plated jewelry industries, steel manufacturing industries, and electroplating industries. The extensive use of nickel in various industries or its occupational exposure is definitely a matter of serious threat on human health [36].

1.5.3 Sources of manganese exposure

There are many environmental sources of Mn, which include eroded rocks, soils, and decomposed plants. Ocean spray, forest fires, vegetation, and volcanic activities are other major natural atmospheric sources of manganese. The major anthropogenic sources of environmental manganese include emission from manganese ore mining, manganese alloy production, welding, coke ovens, dry alkaline battery manufacturing, and manganese salt production. Its widespread applications in ceramics production and in the manufacture of glass, aluminum cans, and electronic components must also be noticeable. Some additional sources of Mn included the fungicides, medical imaging contrasting agents, and water purification agents. Additionally, several countries including the USA, Canada, Argentina, Australia, Bulgaria, France, Russia, New Zealand, China, and the European Union have approved use of the fuel additive methylcyclopentadienyl manganese tricarbonyl (MMT). Combustion of gasoline containing MMT releases Mn phosphates, sulfates, and oxides into the air, especially where there is high traffic density releasing particles within the respirable size range. Mn containing emissions contaminate soil, dust, and plants near roadways, which introduces additional Mn to the environment [37].

1.5.4 Sources of lead exposure

Lead is a slightly bluish, bright silvery metal. Natural lead pollution occurs from volcanic explosions and forest fires [38]. The main sources of lead exposure include drinking water, food, cigarette, industrial processes and domestic sources. The industrial sources of lead pollution include gasoline combustion, house painting and plumbing of lead pipes, uses of lead bullets, lead storage batteries, pewter pitchers, toys and faucets. Lead is released into the atmosphere from industrial processes as well as from vehicle exhausts. Human exposure to lead and its compounds occurs mostly in lead related occupations with various sources like leaded gasoline, industrial processes such as smelting of lead and its combustion, pottery, boat building, lead based painting, lead containing pipes, battery recycling, grids, arm industry, pigments, printing of books, etc. [39].

1.5.5 Sources of Copper exposure

Heavy metal, Copper is used in the industries to produce copper pipes, cables, wires, copper cook wares, plating, rayon, electrical and electronic tools, pesticides, paints, and pigments. Copper is also used in textile industries. Copper contents in foodstuff vary according to the local conditions. Copper concentration in soil, slurry/manure spreading, use of copper compounds as bactericides or fungicides on many crops and copper emissions from melting and casting industries may affect the copper contents in cereals, fruits, and vegetables and to a lesser extent in meat and animal products [40].

1.5.6 Sources of Cadmium exposure

Cadmium is regularly found in ores together with zinc, copper and lead. Therefore, volcanic activity is one of the natural sources for having a temporary increase in environmental cadmium concentrations. Cadmium is widely used in different industrial processes, e.g.: as an anticorrosive agent, as a stabilizer in PVC products, as a color pigment, a neutron-absorber in nuclear power plants, and in the fabrication of nickel-cadmium batteries. Phosphate fertilizers also contribute to have higher cadmium load in agricultural soil. Although some cadmium containing products can be recycled, a large percentage of the general cadmium pollution is caused by dumping and incinerating cadmium-contaminated wastes [41]. Cadmium is emitted from various industrial processes and from cadmium smelters into sewage sludge, fertilizers, and groundwater which could remain in soils and sediments for several decades and consequently taken up by plants. The possible pathway of human exposure to Cd is through the food chain. Cd is a common contaminant found in most of the human foodstuffs due to the high metal transfer properties of the respective plants. The bioaccumulation of Cd from soil to the foodstuffs makes diet a primary source of Cd exposure among nonsmoking, non-occupationally exposed populations. Certain foods such as shellfish, kidney, liver, mushrooms and root crops contain high levels of cadmium [42].

1.5.7 Sources of Arsenic exposure

Arsenic associated with arsenide of copper, lead, gold, iron hydroxides and sulfides, is stored in geological bedrocks (sedimentary rocks) or in the arsenic-rich aquifer matrices in many regions of the world such as Bangladesh, Australia, Canada, India, Vietnam, and Latin America [43]. The inorganic forms of arsenic such as arsenite and arsenate compounds are lethal to humans and other organisms in the environment. Humans get in contact with arsenic through several means which include industrial sources such as smelting and microelectronic industries, Phosphate fertilizers, Paints materials, Textile and Pharmaceutical products, Pesticides, Smelting of gold, lead, copper, nickel, and others [44].

1.6 Effects of heavy and toxic metals on the human health

The heavy and toxic metals such as Cd, Pb, Hg, and As deplete the major antioxidants of cells, particularly antioxidants and enzymes having the thiol group (—SH). Such metals may increase the generation of reactive oxygen species (ROS) like hydroxyl radical ($HO^{\cdot}$), superoxide radical ($O^{2\cdot-}$), and hydrogen peroxide (H_2O_2). Increased generation of ROS can devastate the inherent antioxidant defenses of cells and lead to a condition called “oxidative stress” [45]. Heavy metals, including Cd, Pb, and Hg, are nephrotoxic, especially in the renal cortex. The chemical form of the heavy metals is important in their toxicity. Mercury toxicity largely depends on Hg speciation [46]. Relatively higher concentrations of toxic heavy metals, i.e., Cr, Cd, and Pb, and comparatively lower concentrations of the antioxidant element Se have been found in the patients suffering from cancer and diabetes in the Lahore city, Pakistan [47]. More specifically, heavy metals could also cause several serious health problems in humans, affecting the nervous system, kidney, liver, and respiratory functions. Most trace metal elements (MTEs) are strongly carcinogenic. MTEs could also cause the delays in the human growth and development, and disruption of bioregulatory systems responsible for functional or psychosomatic disorders, like chronic fatigue syndrome, and neurodegenerative pathologies, such as the Parkinson’s and Alzheimer’s diseases. Intoxication by some heavy metals, such as mercury and lead, could also lead to autoimmunity phenomena, in which the immune system of the patient attacks his own cells. This could cause the development of joint diseases, such as rheumatoid arthritis, and kidney, circulatory, or nervous problems in the human bodies. Figure 1.6 shows the development of different diseases in human bodies for the long-time exposure of heavy and toxic metals to humans from various sources [48]. **Acute poisoning:** This happens if someone get a high dose at one time and the symptoms usually appear on quickly.

Go numb Weak or malformed bones Kidney damage

memory loss Liver damage Abdominal pain

Pass out Feeling sick Feel confused

[Figure reference: Mitra, S. et al. *Journal of King Saud University - Science*. 34(3), (2022), 101865]

Figure 1.5: Some acute poisoning of heavy metals

Chronic poisoning: Results are observed after contact with a low dose over a long time and the symptoms come on slowly.

Headache

Achy joints and muscles

Weakness and tiredness

Constipation

[Figure reference: Mitra, S. et al. *Journal of King Saud University - Science*. 34(3), (2022), 101865]

Figure 1.6: Some chronic poisoning of heavy metals

1.6.1 Effects of Iron on human health

Iron toxicity is classified as corrosive or cellular. Ingested iron could cause direct caustic injury to the gastrointestinal mucosa, resulting in nausea, vomiting, abdominal pain, and diarrhea. Significant fluid and blood loss can lead to hypovolemia. Hemorrhagic necrosis of gastrointestinal mucosa can lead to hematemesis, perforation, and peritonitis. At the cellular level, iron impairs cellular metabolism in the heart, liver, and central nervous system. Free iron enters into the cells and concentrates in the mitochondria. This disrupts oxidative phosphorylation, catalyzes lipid peroxidation, forms free radicals, and ultimately leads to cell death [49].

1.6.2 Effects of Nickel on human health

Acute toxicity of Nickel in human occurred from the absorption through the gastrointestinal tract or by inhalation through lungs. Nickel carbonyl inhalation causes two kinds of acute toxic effects: instant and delayed. The symptoms of acute toxicities include nausea, vomiting, vertigo, irritation, etc. These symptoms last for a few hours to a couple of days. Instant symptoms are followed by delayed symptoms like stiffness of the chest, constant cough, dyspnea, cyanosis, tachycardia, palpitations, sweating, visual disturbances and weakness etc. Death due to cardiac arrest has been reported in a 2 ½ year old girl, who consumed nickel sulfate accidentally [36].

[Figure reference: Zambelli, B. et al. *Biochimica Et Biophysica Acta - Proteins and Proteomics.* 1864(12), (2016), 1714-1731]

Figure 1.7: Effects of nickel poisoning

Chronic inhalation and exposure to nickel dusts and aerosols contribute to all the types of respiratory disorders, including asthma, bronchitis, etc. Another study reported that nickel refinery workers were displaying higher incidences of pulmonary and nasal cancer [36].

1.6.3 Effects of Manganese on human health

In a human study utilizing magnetic resonance imaging and spectroscopy (MRI/S) to investigate changes in neurochemistry of smelting workers, increases in GABA and decreases in myoinositol were seen in the thalamus. Changes in thalamic GABA were associated with reduced fine motor performance [50]. Mn exposure alters neurotransmitter and metabolite levels [51]. Mn exposure inhibits myocardial contraction, dilates blood vessels, and induces hypotension, suggesting that Mn exposure has a significant effect on cardiac function. Mn has a direct effect on mitochondrial function resulting in a reduced myocardial contraction, and causes vasodilation, leading to a decreased blood pressure following acute exposure. Gender may also be a contributing factor to developing cardiovascular toxicity after Mn exposure. In a study on male and female smelters exposed to Mn, female smelters had significantly shorter P–R intervals compared to controls, and there was no difference in males. QRS and T waves were also significantly different for female smelters. An increase in the mortality of infants born in Bangladesh was observed in their first year of life who are being exposed to Mn concentrations at or above the WHO’s standard of 400 μg Mn/L compared to the unexposed infants [52].

1.6.4 Effects of Lead on human health

Lead exposure occurs through various ways like inhalation, ingestion or skin contact. Direct contact of lead or lead-based compounds occurs through the mouth, nose, eyes, and through the crack in of the skin may also increase lead levels. Lead disrupts the maintenance of the cell membrane, red blood cells with a damaged membrane become more fragile, which results in anemia. Lead is also speculated to alter the permeability of blood vessels and collagen synthesis. Lead could damage the activity of cells of the immune system, such as polymorphonuclear leukocytes, resulted in decreasing the immune activity. Chronic lead nephropathy occurred due to years of lead exposure manifested in kidney biopsy by moderate focal atrophy, loss of proximal tubules and interstitial fibrosis. Low level environmental lead exposure may accelerate renal insufficiency in patients without diabetes who have chronic renal disease. The previous studies also showed that repeated chelation therapy may improve renal function and slow down the progression of renal insufficiency [38]. The reproductive systems of both males and females are affected by lead poisoning. In males sperm count is reduced and other changes occur in the volume of sperm when blood lead levels exceed 40 μg/dL. Activities like motility and the general morphology of sperm are also affected at this level. The problems with the reproductivity of females due to lead exposure are more severe. Toxic levels of Pb can lead to miscarriages, prematurity, low birth weight, and problems with development during childhood. The brain is the most sensitive organ to lead exposure. In a child's developing brain, synapse formation is greatly affected in the cerebral cortex by lead pollution.

Lead also interferes with the development of neurochemicals, including neurotransmitters, and organizing of ion channels. Lead poisoning also causes the loss of neuron myelin sheath, reduction in the number of neurons. It interferes with neurotransmission and decreases neuronal growth. The brain of adults exposed to increased lead levels during their childhood also shows a decreased volume, especially in the prefrontal cortex [38].

1.6.5 Effects of Copper on human health

Acute and chronic exposures to excess copper could cause some sever health effects on humans bodies. The emphasis is placed on acute exposure effects of copper on the gastrointestinal (GI) system. The effects include GI mucosal ulcerations and bleeding, acute hemolysis and hemoglobinuria, hepatic necrosis with jaundice, nephropathy with azotemia and oliguria, cardiotoxicity with hypotension, tachycardia and tachypnea, and central-nervous-system (CNS) manifestations, including dizziness, headache, convulsions, lethargy, stupor, and coma. A major target of chronic copper toxicity is the liver. Liver toxicity is usually seen in specific populations, such as individuals with Wilson disease and children with various cirrhosis syndromes. Hemolytic anemia due to high concentrations of circulating copper could also occur. Small amounts of copper from intrauterine devices can prevent embryogenesis by blocking implantation and blastocyst development. Genotoxicity, mutagenicity, and carcinogenicity of copper are also documented [40].

1.6.6 Effects of Cadmium on human health

Generally, there are three possible ways of cadmium resorption: Gastrointestinal, pulmonary and dermal. The respiratory system is severely affected by the inhalation of cadmium-contaminated air. Shortness of breathing, lung edema and destruction of mucous membranes are the parts of cadmium-induced pneumonitis. Cadmium-contaminated foods cause acute gastrointestinal effects, such as vomiting and diarrhea. Kidney damage has long since been described to be the main problem for patients chronically exposed to cadmium. Cadmium could find its way to the kidney in the form of cadmium-metallothionein (Cd-MT). Cd-MT is being filtrated in the glomerulus, and subsequently reabsorbed in the proximal tubules. It is then remained in the tubule cells and makes up the major part of the cadmium enriched body. The amount of cadmium in the kidney tubule cells can increase during the life span of every person. Effects of cadmium are also observed in the reproductive biology. The most harmful effects are realized in the productions of progesterone and testosterone. Cadmium intoxication is also highly connected to the bone damage, e.g. in workers exposed to cadmium-polluted fume and dust. Cadmium associated renal cancer in humans was confirmed by clinical studies [53].

1.6.7 Effects of Arsenic on human health

Integumentary system is the largest organ of the body which can easily be affected by arsenic poisoning. Skin abnormalities hold the hallmark of chronic arsenic exposure in adults. Moreover, men are likely to develop arsenic induced skin disorders compared to women. Arsenic attacks dermal system (skin lesions), cardiovascular system (black foot disease), renal system (proximal tubule degeneration, papillary and cortical necrosis), nervous system (peripheral neuropathy, encephalopathy), hepatic system (hepatomegaly, cirrhosis, altered hemi metabolism), endocrine system (diabetes), and hematological system (bone marrow depression) etc. [54].

[Figure reference: Tchounwou, P. B. et al. *Molecular and Cellular Biochemistry*. 255(1/2), (2004), 47-55]

Figure 1.8: Effects of arsenic poisoning

Objectives with specific aims:

The main purpose of this research work is to investigate the bioaccumulation and mobilization of heavy and toxic metals in different parts of various fruits plant bodies from contaminated soil and water. The main objectives of the present research are:

1) To examine the metal contents in waterbodies used for irrigation in the study areas.
2) To assess heavy and toxic metals contamination in soil irrigated with surface waterbodies which are being polluted with toxic industrial effluents.
3) To investigate the uptake, mobilization, and translocation of heavy metals from contaminated soil into different parts of fruit plants bodies such as roots, stems, leaves, and edible parts.
4) To estimate daily intake of different heavy metals due to the consumption of polluted fruits items.
5) To evaluate the potential health risks associated with the intake of heavy metals contaminated different fruit species.
6) To evaluate potential non-carcinogenic risks for both adults and children from the consumption of metals contaminated various fruits items.
7) To examine potential carcinogenic risks associated with intake of metals polluted fruits species by both adults and children.

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CHAPTER-2

STUDY AREA

2 General information

Bangladesh is an agricultural as well as a developing country. The economy of this country is growing fast because of the dramatic rapid increase in industrialization and urbanization in recent years [1]. However, the industrialization has been occurring without any proper plans and guidelines. Most of the industrial establishments occur in and around agricultural lands. The industries are emitting their toxic effluents without any prior treatment or with a partial treatment into nearby surrounding environments and thus contaminating air, water, soil, crops, vegetables, fruits, and other environmental resources. Savar upazila area within Dhaka district selected to perform environmental pollution studies. This upazila is located only about 35 kilometer away from Dhaka city. Dhaka is the capital of Bangladesh, and a great variety of industries found their location around the Dhaka city. A large number of industries are located in Savar upazila and it is considered as highly contaminated area. These industries are continuously discharging their toxic effluents to the surrounding environments and consequently polluting surface water, soil, crops, vegetables and fruits grown in and around industrial zones. **Table 2.1** highlights the different types of industries located in Savar area and possible sources of various metals from them.

Table 2.1: List of various industries located in Savar area, Dhaka, and the possible sources of metals and metalloids pollution.

Metals	Sources
Arsenic	Phosphate fertilizer, paint, textile, pharmaceutical, pesticide, metal hardening and other industries.
Cadmium	Fertilizer, electronics, paint, battery, PVC and other industries.
Iron	Iron and steel, sulfuric acid industry and other industries.
Chromium	Metal plating, rubber, photography, tanning, leather industry, textile industry, paints and other industries.
Nickel	Electroplating, semiconductor goods, iron and steel, battery and other industries.
Manganese	Dry-cell, fertilizer, brick, and other industries.
Lead	Gasoline (refinery), paint, plumbing pipe, lead bullet, battery, toy and other industries.
Copper	Plating, Rayon, electronics, Pesticide, Paints and pigment industry, Textile industry and other industries.

2.1. Geographical locations of the study area

2.1.1. Geography

Savar is located at 23.8583°N 90.2667°E. It has 66,956 units of household and a total area of 280.13 square kilometers (108.16 sq. mi.). It is bounded by Kaliakair and Gazipur Sadar upazila on the north, Keraniganj upazila on the south, Mirpur, Mohammadpur, Pallabi and Uttara Thana of Dhaka City on the east, and Dhamrai and Singair upazila on the west. The study area links with Dhaka city with comparatively higher traffic density and has significant industrial influences [1].

Table 2.2: Location of different sampling sites of the study area

Sampling site	Latitude	Longitude
S-1	23.9519	90.2620
S-2	23.9539	90.2533
S-3	23.9489	90.2407
S-4	23.9385	90.2432
S-5	23.9434	90.2539

The present study was carried out in March 2022 around Dhaka Export Processing Zone (DEPZ), which is a large industrial area comprised of a good number of local and foreign industries like fabric printing and dyeing, food processing, textiles, electric cables, pharmaceutical, chemical, etc. It is about 35 km from Dhaka City and 25 km from Hazrat Shahjalal International Airport in the NW direction and represents a limited extent of the landscape yet has distinct morphological features. The topography of this area comprises irregular elevated land blocks on which people live and surrounding low-lying areas which are mostly cultivation lands and water bodies. There is, however, moderately elevated paddy land on the western fringe of the study area that is cut by the canal connecting the waste lagoon and Bansi River. On the way through the connecting canal to reach Bansi River, the effluent pools in the depressed area next to the EPZ boundary forming an unexpected waste beel whose perimeter grows larger by excess precipitation plus runoff during Rainy Season. At that time contaminated water spills and inundate the elevated lands too, thus polluting them as well. Most industries discharge their effluents without any prior treatment, through the open drain and contaminate water, soil, and fruits in the adjacent areas. Different kinds of fruits such as Guava *(Psidium guajava),* Pomelo *(Citrus maxima),* Banana *(Musa paradisiaca)* etc. are grown in the area throughout the year and are used for home consumption and mainly for selling to residential areas of Dhaka.

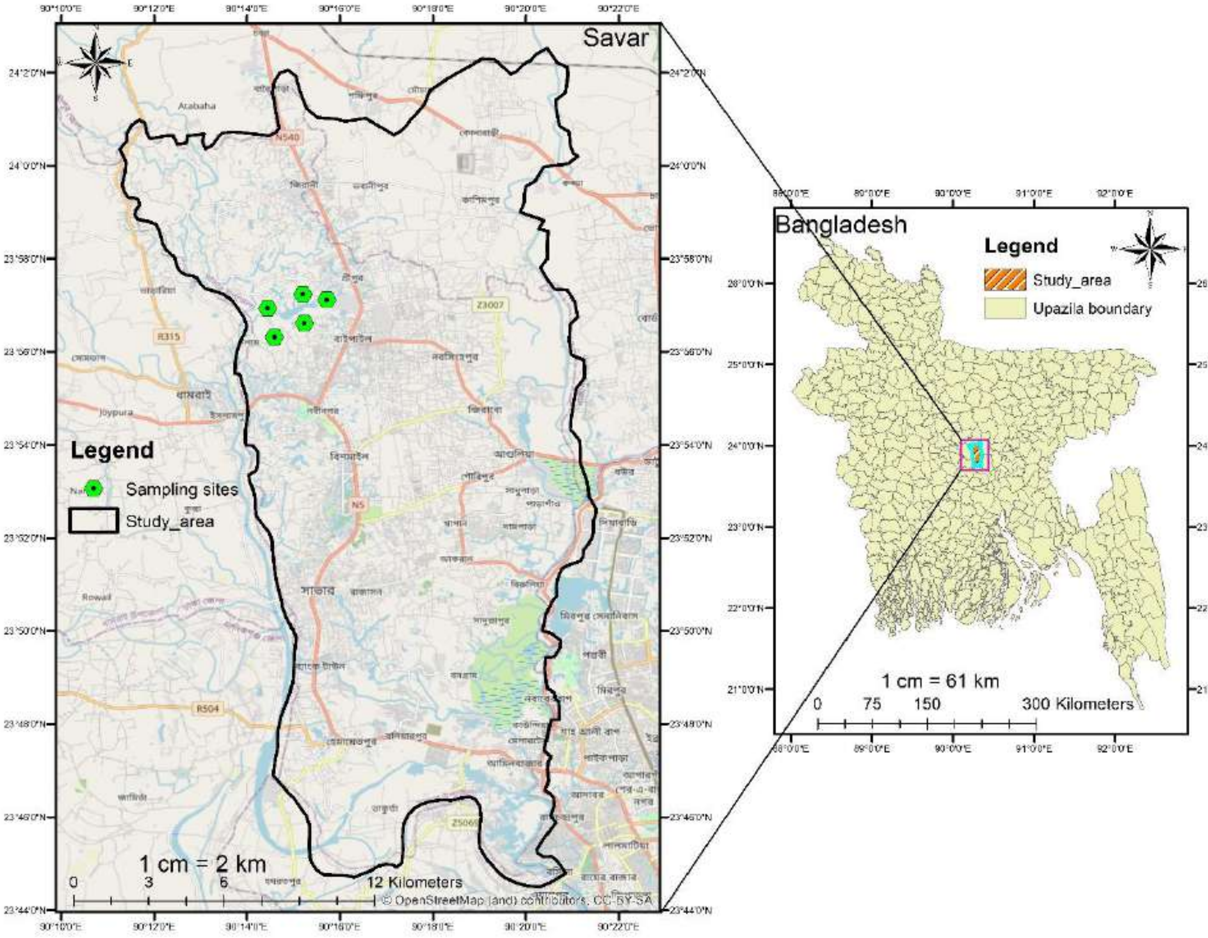

Figure 2.1: Map of the study area

2.1.2. Industries and Economy of the study area

Agricultural activities and industrial manufacturing are the two major economic sectors in Savar area. The major crop grown in this area is Paddy and common vegetables are ladies' finger, garlic, chili, spinach, and other vegetables. The extinct or nearly extinct crops in the region are Aous paddy, Asha Kumari paddy, Linseed, Kalimator, Randhuni saj, Mitha saj, Kaun and Mas kalai (local names of the crops). The common fruits grown in this area are guava, pomelo, papaya, banana, wood apple etc. There are 181 combined fisheries, dairies, and poultries dairy, 5 hatcheries, 209 poultries, and 1319 fisheries establishment are currently available in this area. The manufacturing facilities include ceramic industry, beverage industry, press and publications industry, garments industry, foot ware, jute mills, textile mills, printing and dying factory, transformer industry, automobile industry, biscuit and bread factory, pharmaceutical industry, soap factory, brick field, welding etc. [2, 3]. The following chart shows the contributions of different sectors in the total economy of the study area [2, 3].

Agriculture %	Industry %	Service %
23.6	59.6	16.8

2.1.3. Temperature

The hot season lasts for 3.5 months, from March 12 to June 26, with an average daily high temperature above 89°F in the study area. The hottest day of the year is April 15, with an average high temperature of 93°F and low temperature of 77°F. The cool season lasts for 1.5 months from December 13 to January 31, with an average daily high temperature below 78°F. The coldest day of the year is January 12, with an average low temperature of 57°F and high temperature of 75°F.

2.1.4. Rainfall

The long-term trend of annual rainfall in Dhaka shows no significant change, however, the trend in the seasonal rainfall appears to be erratic and variable. To show the variation within the months and not just the monthly totals, we show the rainfall accumulated over a sliding 31-day period centered on each day of the year. The capital city Dhaka and its surrounding areas such as Savar experiences extreme seasonal variation in monthly rainfall [4].

Figure 2.2: Discharging of industrial effluents into the nearby water body of Savar area

The rainy season of a year lasts for 9.5 months from February 13 to November 29 with a sliding 31-day rainfall of at least 0.5 inches. The most rain falls observed during the 31 days periods on July 3, with an average total accumulation of 9.9 inches. The rainless period of a year lasts for 2.5 months, from November 29 to February 13. The least rain falls is being realized around January 8 with an average total accumulation of 0.2 inches.

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CHAPTER-3

SAMPLING, MATERIALS, AND METHODOLOGY

3.1. Survey Form

We provided some questionnaire to the people living around industrial zones of Savar area. We tried to know about the maximum number of young people that were playing a key role in earning money and managing the family in the study area. We wanted to know how much of fruits they buy from the market in each week, what kind of fruits they take, how many times they get fruits and how much of them they eat in a day. Besides, we visited 20 houses where 15-16 years old children are used to live and learned from their mother about how much of fruits the children eat every day, what types of fruits they eat in every day. Food ingestion rate was also being addressed by providing questionnaire survey to local people of the study area. A total of 20 households, minimum 3 from each sampling site, were surveyed and finally the data were used to calculate the average fruits ingestion rate for adults and children. We also make sure that the fishes which were available to local people to buy and to eat collected from industrial contaminated sites of the study area.

3.2 Chemicals and reagents

The chemicals and reagents used in this research were in analytical grade and used without any further purification. Stock solutions of 1000 ppm were prepared from their corresponding salts for the selected heavy metals and metalloids (Pb, Cd, Cr, Cu, Zn, Fe, Ca, Mn, Co, Ni, As and Se). Double distilled and deionized water was used in all solution preparation and dilution purposes throughout the experimental procedures. The list of chemicals and reagents are given below:

Chemicals	Sources	Purity (W/W basis)
Sulfuric Acid	Aldrich	69%
Nitric Acid	Aldrich	98%
Hydrogen peroxide	Aldrich	30%
Per chloric Acid	BDH	70%
Hydrochloric Acid	Merck Germany	37%
Ethyl alcohol	Merck India	99.5%
Potassium Iodide	Merck Germany	Pure
Sodium Borohydride	BDH	pure

3.3 Instruments

Analyses of the samples were performed using the following instruments:

1. Oven (Lab Tech, LDO-030E)
2. Digital Balance (AB 265/S/SACT METTLER, Toleto, Switzerland)

3. Hot plate (RC- 1887/166, Velp Scientific, Italy)
4. Grinder
5. Atomic Absorption Spectrophotometer (SHIMADZU AA-7000)
6. Inductively Coupled Plasma-mass Spectrometry (ICP-MS)

3.4 Sample collection

Different parts (roots, stems, leaves, and edible parts) of three fruits such as Guava *(Psidium guajava)*, Pomelo *(Citrus maxima) and* Banana *(Musa paradisiaca)* were collected randomly in triplicate from effluent-contaminated sites located beside Dhalai beel (a lake in which all the complex industrial effluents from DEPZ are disposed of). Simultaneously soil samples were also collected from the same site for each plant. Dhalai beel water is used for the cultivation and irrigation of these trees and during the rainy season, the water from the beel goes up to the roots of the trees. For that reason, we also collected water samples from different points of Dhalai beel. Root samples were taken at a depth of 10-15 cm while the stems, leaves, and edible parts of fruit samples were collected using a pre-cleaned acid-washed stainless knife and washed again after each sampling to avoid cross-contamination. The samples were carefully packed into polyethylene bags for further analysis. Soil samples were collected from six random points along the drip line of each tree, at a depth of 15–20 cm, with the use of a plastic hand shovel. These were thoroughly mixed in a clean plastic bucket to achieve homogeneity, thus forming the composite sample. The samples of soil were collected and stored in sealed plastic bags whereas water samples were stored in autoclaved bottles to avoid any contamination for further analysis[1].

Local name: Guava
Scientific name: *Psidium guajava*

Local name: Pomelo
Scientific name: *Citrus maxima*

Local name: Banana
Scientific name: *Musa paradisiaca*

Figure 3.1. Different fruits plants studied for heavy and toxic metals and metalloids contamination in the present investigation

Table 3.1. List of the fruits plants and their different parts studied for heavy metals and metalloids pollution

Types of Fruits Plants **(Local name)**	**Scientific name**	**Studied plants parts**	**Investigated Elements**	
Guava	*Psidium guajava*	Root	Metal	Metalloid
		Stem	Pb Cd Cr Cu Zn Fe Ca Mn Co Ni	As Se
		Leaves		
		Edible part		
Pomelo	*Citrus maxima*	Root		
		Stem		
		Leaves		
		Edible part		
Banana	*Musa paradisiaca*	Root		
		Stem		
		Leaves		
		Edible part		

3.5 Sample Preparation

The collected fruit samples were washed thoroughly with distilled water to remove extraneous matter. The samples were then sliced into small pieces with a stainless-steel knife and oven dried at 65℃ for 48 hours. Dried fruit samples were crushed using a food processor and then stored in a refrigerator in sealed plastic bags until analyzed. A representative soil sample was taken from each site and was dried in an oven at 105℃ then passed through a 2 mm mesh sieve to remove organic matter and gravel. Some of this soil was crushed with a mortar and pestle to reduce the particle size for microwave digestion. Samples were stored in sealed plastic bags and kept in a refrigerator until analyzed. For water sampling, three replicate polyethylene bottles (acid washed) of capacity 100 mL were immersed at an interval of 15 s into the water of Dhalai beel, and immediately after filling, 1 mL of concentrated HNO_3 was added to the water, and bottles were brought back to the analytical laboratory and digestion was completed within a week.

3.5.1 Fruit and Soil sample digestion procedure

To estimate metal accumulation in fruit and soil samples, 0.5 g each were weighed to which 10 mL of HNO_3 was added, and the mixture was allowed to stand overnight. During digestion, the mixture was heated for 4 h at 180℃ for fruit samples and 200℃ for soil samples on a hot plate until a transparent solution was obtained [1, 2]. The solutions were cooled for 5 – 10 min, then 5 mL of $HClO_4$ was added to each sample and digested one more time. Strong acid mixture used for completely digest the sample. After that, the cooled solutions were filtered through Whatman No. 42 filter paper and made into a 50 mL solution with deionized water and kept in the marked plastic bottle. A blank sample was also prepared in the same way. All samples were stored at – 4℃ temperature before analysis.

3.5.2 Water samples digestion procedure

50 mL of acid-treated each of the water samples was heated with 10 mL of conc. HNO_3 on a hot plate at 90℃ and boiled until reduced to a final volume of 5 mL. After that, the cooled solutions were filtered through Whatman No. 42 filter paper, diluted to 50 mL with deionized water and kept in the marked plastic bottles. All samples were stored at – 4°C temperature before analysis.

3.6 Heavy metals and metalloid analysis

3.6.1 Atomic Absorption Spectrophotometer (AAS)

Atomic absorption spectrophotometer (AA-7000, Shimadzu, Japan) equipped with a single element hollow cathode lamp was used for the determination of seven heavy metals such as Pb, Cd, Cr, Cu, Fe, Zn and Ca. In this process, fruit samples were dried in a hot air oven where digestion done by a fume hood (EFD-481, ESCO, USA). For this experiment, all operating parameters considered for the quantification of each metal and their recovery percentages are listed in Table 3.2 and 3.3. Calibration graph of the correlation coefficient of respective elements were prepared to calculate standard deviation. The accuracy of data was validated for all the elements by certified reference material supplied by Sigma-Aldrich, Germany. To assure the accuracy, sensitivity, and precision of the analytical procedure of AAS, SRM 2976-Muscle tissue was analyzed as certified reference material from the national institute of standards (NIST). The limits of detection (LOD) of AAS system for Cd, Pb, Zn, Cu, Fe and Cr were 0.003, 0.01, 0.02, 0.01, 0.060 and 0.01 ppm, respectively.

Table 3.2: Operating parameters of atomic absorption spectrometer (AAS) and recovery percentages of elements

Heavy metals	Wave length (nm)	Lamp intensity (mA)	Slit intensity (nm)	Recovery percentage (%)
Cd	228.8	4	0.5	98
Pb	217.3	10	1	109
Zn	310	4	0.5	98
Cu	324.8	4	0.5	106
Cr	357.9	7	0.2	101
Fe	248.30	10.0	0.2	93

Table 3.3: Concentrations of metals found in Certified Reference Materials determined by AAS

Elements(ppm)	Certified value	Measured value	Deviation (%)	Recovery (%)
Cd	0.20±0.02	0.19±0.02	0.75	95.00
Cr	4.00±0.06	3.90±0.06	1.00	99.75
Cu	2.00±0.02	2.00±0.07	1.20	100.00
Fe	1.00±0.10	0.93±0.06	0.96	93.00
Pb	4.00±0.05	3.98±0.01	1.77	99.50

3.6.2 Inductively Coupled Plasma-mass Spectrometry (ICP-MS)

All digested samples (Fruit, soil, and water) were analyzed for Pb, Cd, Cr, Cu, Zn, Mn, Co, Ni, As and Se by inductively coupled plasma-mass spectrometry (ICP-MS) due to its multi-element determination capability, high dynamic linear range, and sensitivity. In order to satisfy the defined internal quality controls (IQCs), each sample was made to run, including blank and certified reference materials (CRM), to validate the internal standards. For excluding batch-specific errors, each sample was analyzed in triplicate. Standard stock solutions containing 10 mg/L of each element (Pb, Cd, Cr and Cu) and internal standard solutions containing 1.0 mg/L of indium (In), yttrium (Y), beryllium (Be), tellurium (Te), cobalt (Co), and titanium (Ti) (Spex CertiPrep®, USA) were prepared. The standard curve was established by using a multielement standard solution. Relative standard deviation (RSD < 5%) was inspected by a tuning solution purchased from Agilent Company. Analytical wavelengths were selected based on minimum spectral interferences and maximum analytical performance. Initially, the three most sensitive lines were chosen. From these lines, the line with no interfering elements was selected.

3.7 Quality assurance and quality control

For performing the internal calibration, standard solutions having 1.0 mg/L of indium, yttrium, beryllium, tellurium, cobalt, and thallium were obtained from Spex CertiPrepVR (Metuchen, NJ, USA). During the analysis, 10 mg/L internal standard solution was ready from the primary standard stock solution. Quality assurance and quality control were confirmed and maintained by running blank solution periodically, drawing the calibration curves, using spiked samples, and with the midpoint standard checks. All the calibration procedures were assessed based on their corresponding correlation coefficients (R2) of the corresponding calibration curves. The calibration curves were guaranteed with the correlation coefficient (R2), where, Pb-0.9992, Cr-9999, Cu-9996, Fe-0.9988 and Cd-0.9994. Mid-point checks for the metals lie in the range of 0.25 to 5.5%. Spike recoveries ranged from 96.54 to 98.85%.

3.8 Statistical analysis

Different reliable statistical software was used to analyze the concentrations of heavy and toxic metals in the water, soil, crops, and vegetables samples of the study area. Microsoft excel-2016 was used to calculate the mean, standard deviation, and other health related parameters. Pearson's correlation package SPSS (version 16.0) was also used for source analysis of heavy metals found in the samples studied.

3.9 Soil Quality Index

3.9.1 Geo-accumulation Index (Igeo)

Pollution characteristics due to the heavy metal contamination in sediments can be evaluated using the geo-accumulation index. Geo-accumulation index, proposed by Muller (1979), is used to determine metals contamination in sediments, by comparing the present concentrations with that of pre-industrial period using the following formula[3]:

$$I_{geo} = \log 2 \frac{C_n}{1.5B_n}$$

Where, C_n is the heavy metal concentration in the soil samples, and B_n is the geochemical background value in the average shale of the heavy metal element. Factor 1.5 is used to reflect the possible fluctuation of the element in the background value as well as minimal anthropogenic influences or input. Based on the I_{geo} indices, it can be classified into seven classes as shown in [4].

Table 3.4: The degree of metal pollution based on Geo-accumulation (I_{geo}) classification

I_{geo} value	Class	Designation of soil quality
$I_{geo} \leq 0$	0	Unpolluted
$0 \leq I_{geo} \leq 1$	1	Unpolluted to moderately polluted
$1 \leq I_{geo} \leq 2$	2	Moderately polluted
$2 \leq I_{geo} \leq 3$	3	Moderately to strongly polluted
$3 \leq I_{geo} \leq 4$	4	Strongly polluted
$4 \leq I_{geo} \leq 5$	5	Strongly to extremely polluted
$I_{geo} > 6$	6	Extremely polluted

3.9.2 Enrichment Factor (EF):

The EF of heavy metals has been commonly used to determine the status of anthropogenic contamination. Element Fe was chosen as the normalizing element in this study for identifying anomalous heavy metal contributions. (EF values can be calculated using the following equation:

$$EF = \frac{\frac{C}{Fe}(sample)}{\frac{C}{Fe}(Background)}$$

Where, C/Fe (sample) and C/Fe (background) represent the heavy metal-to-Fe ratios in the present study and in the background, respectively. Iron was chosen as the element of normalization because natural sources (1.5%) extensively dominate with its input [5]. A crucial step in evaluating the impact of sediment pollution is to establish a reference background or baseline sample of known metal composition [6].

Table 3.5: The degree of metals pollution based on seven enrichment factor classes (Taylor, 1964)

EF value	Designation of sediment quality
50	Extremely severe enrichment
25 - < 50	Very severe enrichment
10 - < 25	Severe enrichment
5 - < 10	Moderately severe enrichment
3 - < 5	Moderate enrichment
1 - < 3	Minor enrichment
< 1	No enrichment

3.9.3 Soil threshold values

To produce vegetables that are safe for consumption, it is essential to establish the soil heavy metal threshold levels by examining the transfer of heavy metals from soil to crops. The soil threshold levels were calculated using THQ, maximum permissible limits (MPL) and TF [7]. The STVs were obtained using the following equations:

$$STV1 = \frac{C_{food}\ (THQ = 1)}{TF} = \frac{BW \times AT \times RfD}{IR_{foood} \times EF \times ED \times TF}$$

$$STV2 = \frac{MPL}{TF} = \frac{MPL}{\frac{C_{food}}{C_{soil}}} = \frac{C_{soil}}{FPI}$$

C_{food} (THQ=1) is the heavy metal concentration in fruits (mg kg^{-1}), if THQ=1. Other notations in Eq. (7) were defined earlier. STV1 was calculated based on the HM Reference Doses (RfD) specified by the US EPA. At the core of the STV1 method is the impact on human health; that is, whether the heavy metal content of soil is within the acceptable range for human health. STV2 was back-calculated based on government-defined MPLs for greenhouse vegetable production [8], given the relationship between MPL and STV2.

3.10 Health risk assessment factor

3.10.1 Food pollution index

The food pollution index (FPI) and the collective food pollution index (CFPI) represent contamination by the selected elements [9, 10]:

$$\mathbf{FPI} = \frac{\mathbf{C_{food}}}{\mathbf{MPL}}$$

$$\mathbf{CFPI} = \sum_{\mathbf{i=1}}^{\mathbf{6}} \mathbf{FPI}$$

$\mathbf{C_{food}}$ indicates the heavy metal concentration (mg kg^{-1}, fresh weight) in the edible part of a fruit plant. MPL is the maximum permissible level in fruit suggested by CHM and FAO/WHO [11, 12]. An FPI or CFPI >1 indicates serious pollution.

3.10.2 Bioaccumulation factor or Heavy metal transfer factor

The heavy metal transfer factor (TF) is an essential parameter that is typically used to identify the rate of heavy metal transfer from soil to various types of fruits. Soil quality and heavy metal mobility control the concentration and number of heavy metals taken up by fruit plants [13, 14]. The transfer factor quantifies the possible transfer of heavy metals from different type of soils to cultivated fruit plants.

$$\mathrm{TF} = \frac{\mathrm{C_{food}}}{\mathrm{C_{soil}}}$$

$\mathrm{C_{soil}}$ is the concentration of heavy metals in the soil sample, which the fruits samples were collected from.

3.10.3 Estimated Daily Intake

The EDI was estimated using the following Equation [15]

$$EDI = \frac{C_{fruit} \times IR_{fruit} \times EF \times ED}{BW \times AT}$$

In this equation, C_{fruit} is the metal concentration of fruits, $\mathrm{IR_{fruit}}$ represents daily fruit consumption, EF (days/year) is the exposure frequency (365 days/year), ED (year) is the exposure duration (children = 6 years and adults = 30 years), BW (kg) is the bodyweight (children = 15 kg and adults = 70 kg), and AT is the average duration of exposure to heavy metals (for noncarcinogenic risk, it is 2,190 days in children and 10,950 days in adults, and for carcinogenic risk, it is 25,550 days in children and 25,550 days in adults). A survey was conducted on 27 families (3 families from each sampling site) to find the $\mathrm{IR_{fruit}}$ values for different samples.

3.10.4 Non-Carcinogenic risks

The health risk of heavy metal contaminated fruits was investigated according to the USEPA Health Risk Handbook [16]. The health risk associated with the ingestion of heavy metals is often assessed using this technique. The non-carcinogenic health risks of heavy metals are measured by calculating the target hazard quotients (THQ).

The following equations were used to calculate THQ, TTHQ and HI:

$$\mathrm{THQ} = \frac{\mathrm{C_{fruit}} \times \mathrm{IR_{fruit}} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT} \times \mathrm{RfD}}$$

RfD is reference daily allowed dose of heavy metals [17]. The reference daily doses (RfD) of Pb, Cd, As, Cr, Cu, Fe, Zn, Mn, Co and Ni are 0.004, 0.001, 0.0003, 0.003, 0.04, 0.7, 0.3, 0.014, 0.03 and 0.02 mg kg^{-1} d^{-1}, respectively [18]. The acceptable daily doses for Pb and Cu are 0.004 and

0.04 mg kg^{-1} d^{-1}, respectively, according to the China Ministry of Ecology and Environment. THQ ≥ 1 means that adverse non-carcinogenic impact can occur while THQ < 1 means that adverse non-carcinogenic impact cannot occur. After that the total target hazard quotient (TTHQ) for individual fruit was estimated by this equation[19].

$$TTHQ_{individual\ fruit} = THQ_{Pb} + THQ_{Cd} + THQ_{Cr} + THQ_{Cu} + THQ_{Fe} + THQ_{Zn}$$

$$+THQ_{Ca} + THQ_{Mn} + THQ_{Co} + THQ_{Ni} + THQ_{As} + THQ_{Se}$$

Then the overall potential non-carcinogenic impact of all heavy metals was determined by hazard index (HI) [19]. $HI = \sum TTHQ$

$$= TTHQ_{Pomelo} + TTHQ_{Guava} + TTHQ_{Banana}$$

HI < 1 is in a safe range, and 1 < HI > 5 is not in a safe range[20].

3.10.5 Carcinogenic risks

Carcinogenic risk (CR) of heavy metals was also evaluated by this equation.

$$CR = EDI \times CSF$$

CSF is the cancer slope factor (kg. day/mg). CR indicates the possibility of increasing cancer risk through exposure to $a\ specific$ compound in lifespan. CSF values are 1.5, 0.0085, 0.5, 0.38 and 0.84 for As, Pb, Cr, Cd, Ni respectively [21-23]. CR > E-04 means a carcinogenic risk. Also, E-04 < CR > E-06 means that carcinogenic risk is acceptable [24].

In addition, cumulative cancer risk (CCR) was evaluated for individual fruit by this equation[20, 25].

$$CCR_{individual\ fruit} = CR_{Pb} + CR_{Cd} + CR_{Cr} + CR_{Ni} + CR_{As}$$

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CHAPTER-4

RESULTS AND DISCUSSIONS

4 General Information

In the present study, water, soil, and different fruits plants samples were collected from Savar Upazila, Dhaka district. Savar area has been highly recognized for agricultural activities for long time. Various types of fruits have been grown in and around of Savar Upazila for years. However, rapid industrialization and growing of urbanization activities in the Savar region have greatly affected the agricultural activities in this area in recent years. A large variety of industries have been established in Savar area which are continuously discharging untreated or partially treated toxic industrial effluents into surrounding environment and thus continuously contaminating water bodies, sediments, lakes, canals, rivers, and agricultural lands in the respective areas with different types of toxic metallic, non-metallic, and organic substances. The present research works have concentrated on the determination and evaluation of heavy and toxic metals pollution in water bodies, arable soil, as well as fruits cultivated in the Savar area. The study has also determined the mobilization and translocation of heavy metals and metalloids in different parts of various fruits plant bodies and subsequent potential ecological impacts and health risks of people who are highly exposed to industrial contaminated water bodies and fruits.

4.1. Heavy metals and metalloids concentrations in irrigation water, soil, and fruits

4.1.1 Heavy metals and metalloids contents in irrigation water

Concentrations of various metals (Pb, Cd, Cr, Cu, Zn, Fe, and Ca) measured in all water samples collected from different sampling sites around DEPZ, Savar, Dhaka are presented in Table 4.1.

Table 4.1. Different metals and metalloids concentrations (mg/L) measured in irrigation water by AAS (Mean ± S.D.)

Sampling site	Pb	Cd	Cr	Cu	Zn	Fe	Ca
S - 1	0.1747 ± 0.0400	0.0010 ± 0.0003	0.2677 ± 0.0841	2.0189 ± 0.0135	5.4596 ± 0.3197	7.5094 ± 0.3987	5.1701 ± 0.2084
S - 2	0.0756 ± 0.0502	0.0009 ± 0.0001	0.0973 ± 0.0085	0.6701 ± 0.0169	1.8789 ± 0.0807	5.5426 ± 0.1596	4.6884 ± 0.0417
S - 3	0.0284 ± 0.0161	0.0003 ± 0.0002	0.0320 ± 0.0108	0.2054 ± 0.0100	1.3935 ± 0.0738	2.2576 ± 0.1146	4.4837 ± 0.0481
S - 4	0.0095 ± 0.0072	0	0.0231 ± 0.0098	0.1557 ± 0.0220	1.0133 ± 0.0165	1.7358 ± 0.0905	4.4554 ± 0.0175
S - 5	0.0015 ± 0.0017	0	0.0218 ± 0.0096	0.0687 ± 0.0222	0.4447 ± 0.0285	1.1989 ± 0.0167	3.9156 ± 0.0111
Mean	0.0579	0.0004	0.0884	0.6237	2.0380	3.6488	4.5426
Minimum	0.0015	0	0.0218	0.0687	0.4447	1.1989	3.9156

Maximum	0.1747	0.0010	0.2677	2.0189	5.4596	7.5094	5.1701
Safe Limit^{a}/ Permissible Value^{a} WHO (ppm)	_	0.03	0.05	2	3	_	_
Safe Limit^{a}/ Permissible Value^{a} NSDWQ (ppm)	__	0.01	0.05	2	5.0	___	____
Safe Limit^{b}/ Permissible Value^{b} Safe Limit of India (mg kg^{-1})	0.1	0.01	0.05	0.05	5.0	0.03	_
Safe Limit^{c}/ Permissible Value^{c} Water (mg L^{-1})	0.5	0.01	0.1	0.2	2.0	2.0	_

a,b,c are the references from where the permissible values or safe limits will be taken or adopted.

[a]Yaqub et al. (2021), [b]Awashthi (2000); [c]Safe limit of toxic heavy metals in irrigation water for agricultural purpose (Pescod 1992).

The permissible limits of some metals were not available in literatures, and they were denoted as blank in the respective places.

Concentrations of different metals and metalloids in irrigation water (mgL^{-1}) of the study area were ranged from 0.0015 to 0.1747 mgL^{-1} for Pb, 0 to 0.0010 mgL^{-1} for Cd, 0.0218 to 0.2677 mgL^{-1} for Cr, 0.0687 to 2.0189 mgL^{-1} for Cu, 0.4447 to 5.4596 mgL^{-1} for Zn, 1.1989 to 7.5094 mgL^{-1} for Fe and 3.9156 to 5.1701 mgL^{-1} for Ca (Table 4.1). Out of seven elements examined in effluent-contaminated waterbodies in the vicinity of DEPZ area, Ca concentration was observed to be the highest (4.5426 mgL^{-1}) and the lowest metal content was realized with Cd (0.0004 mgL^{-1}). Mean metals contents in irrigation water from the study area were found to be lower than NSDWQ [1], World Health Organization (WHO) [1] and safe limit of India [2] except Cr which showed higher quantity than the permissible level of NSDWQ, WHO. Comparison with the metals tolerable levels reported from India also showed the excessive presence of Cr, Cu, and Fe in experimental waterbody. Cu, Zn, and Fe contents were exceeded the tolerable limits of heavy metals in irrigation water used for agricultural purposes (Pescod, 1992) [3]. Metals contents measured in all water samples were highly varied and mean concentrations of different metals followed the order of Ca > Fe > Zn > Cu > Cr > Pb > Cd. Higher levels of Fe in water might be due to its long-term use in the production of machine tools, paints, pigments, and alloys in various industries within the study area which might have contaminated surface water bodies and changed the water quality and thus making it less suitable for cultivation [4].

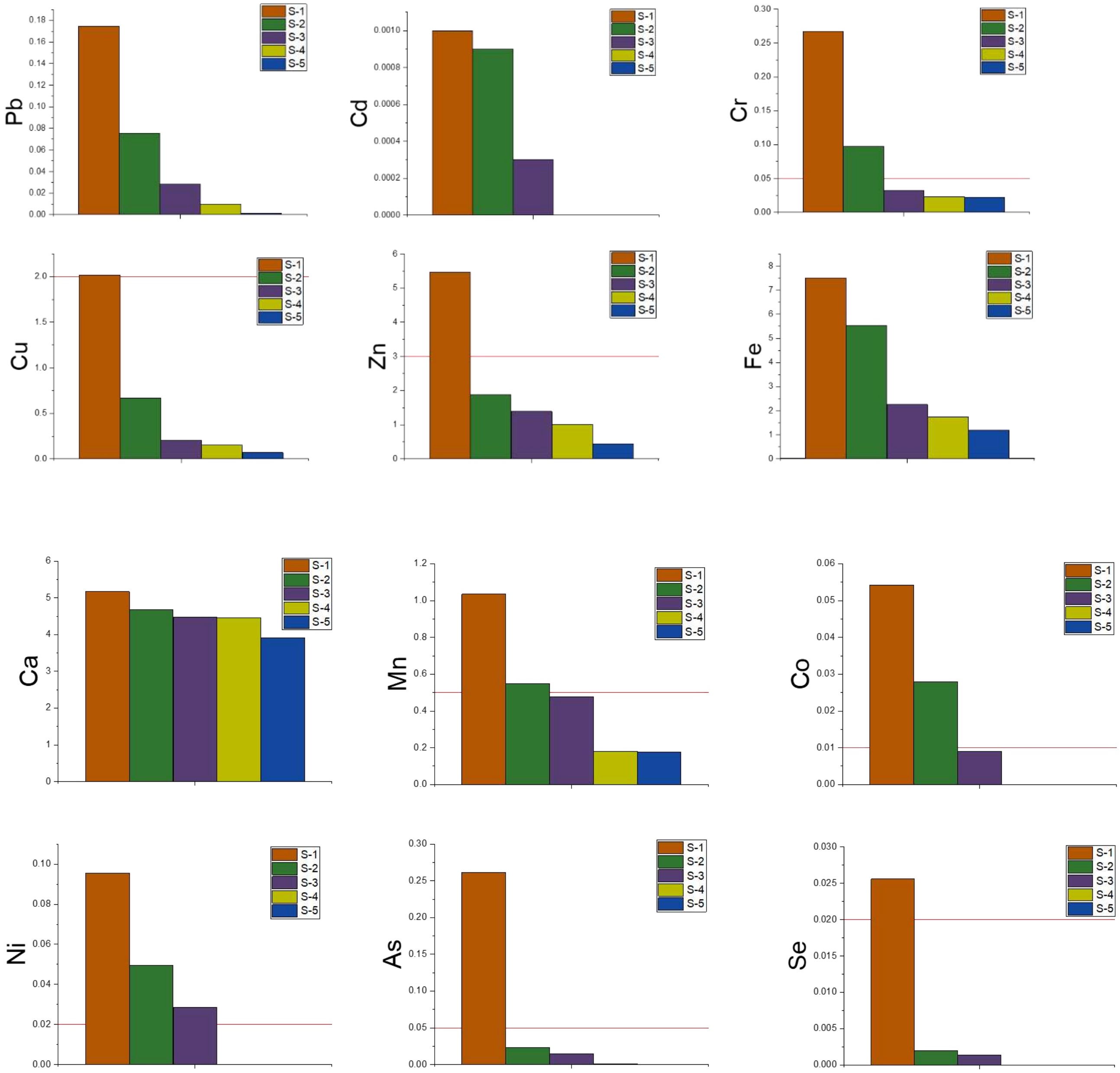


Figure 4.1. Different metals and metalloids concentrations in irrigation water

Table 4.2. Different metals and metalloids concentrations (Mean ± S.D.) in irrigation water (mg/L) determined by ICP-MS

Sample Sites	Mn	Co	Ni	As	Se
S - 1	1.0353 ± 0.0283	0.0542 ± 0.0311	0.0956 ± 0.0075	0.2617 ± 0.0257	0.0256 ± 0.0270
S - 2	0.5501 ± 0.0274	0.0280 ± 0.0255	0.0494 ± 0.0057	0.0233 ± 0.0012	0.0020 ± 0.0005
S - 3	0.4776 ± 0.0107	0.0090 ± 0.0086	0.0285 ± 0.0156	0.0148 ± 0.0076	0.0014 ± 0.0002
S - 4	0.1811 ± 0.0030	0	0	0.0011 ± 0.0008	0
S - 5	0.1781 ± 0.0179	0	0	BDL	0
Mean	0.4844	0.0182	0.0347	0.0602	0.0058
Minimum	0.1781	0	0	0	0
Maximum	1.0353	0.0542	0.0956	0.2617	0.0256
Safe Limit[a]/ Permissible Value[a] [water] WHO (ppm)	0.5	0.01	0.02	0.05	0.02
Safe Limit[a]/ Permissible Value[a] NSDWQ	0.5	_	0.02	_	_
Safe Limit[b]/ Permissible Value[e] [water]	0.02	_	0.2	_	_

a,b are the references from where the permissible values or safe limits will be taken or adopted.

[a] Yaqub et al. (2021), [b] Safe limit of toxic heavy metals in irrigation water for agricultural purpose (Pescod, 1992).

The permissible limits of some metals were not available in literatures, and they were denoted as blank in the respective places.

Mean concentrations of Mn, Co, Ni, As, As, and Se were 0.4844, 0.0182, 0.0347, 0.0602, and 0.0058 mgL^{-1} respectively and they followed the order of Mn > As > Ni > Co > Se (Table 2). Ni and As contents in water were observed to be above the maximum permissible limits [1, 3]. Among all the metals analyzed by both AAS and ICP-MS, the extent of Cadmium (Cd) was observed to be the lowest and Calcium (Ca) was found highest in each of the water samples. The succession of concentrations levels of twelve elements studied in water followed the order of: Ca > Fe > Zn > Cu > Mn > Cr > As > Pb > Ni > Co > Se > Cd. Nowadays, the application of treated wastewater in irrigation and other agricultural activities has become a very common practice in most of the developing countries due to the scarcity of enough fresh water. Small and progressive farmers often use the treated wastewater in irrigation to produce crops and vegetables all over the world [1, 2].

Correlation analysis among metals and metalloids measured in irrigation water:

Table 4.3. Correlation among metals concentrations found in irrigation water

	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Pb	1											
Cd	-0.688	1										
Cr	0.985^{**}	-0.565	1									
Cu	0.991^{**}	-0.612	0.998^{**}	1								
Zn	0.342	-0.299	0.254	0.240	1							
Fe	-0.136	-0.572	-0.302	-0.255	0.304	1						
Ca	0.799	-0.766	0.756	0.783	0.026	0.183	1					
Mn	0.940^{**}	-0.857	0.894^{*}	0.920^{*}	0.181	0.092	0.900^{*}	1				

Co	0.757	-0.697	0.751	0.782	-0.282	-0.032	0.891*	0.890*	1			
Ni	0.230	-0.042	0.190	0.162	0.918*	0.013	-0.259	0.001	-0.439	1		
As	-0.175	-0.569	-0.301	-0.240	-0.215	0.826	0.225	0.156	0.253	-0.432	1	
Se	0.921*	-0.492	0.963**	0.964**	-0.006	-0.421	0.733	0.864*	0.836	-0.025	-0.266	1

* Correlation is significant at the 0.05 level (2-tailed);**Correlation is significant at the 0.01 level (2-tailed)

Table 4.3 shows the results of statistical analysis of different metals concentrations in surface water collected from the vicinity of DEPZ area. Metal-to-metal correlation matrices have been described in terms of linear correlation coefficient (r) with the corresponding magnitudes (significant at 0.05 and 0.01). The magnitudes of r revealed the high degree of positive correlations and significant linear relationship between various pairs of metals as well as reflect their simultaneous release and identical sources from which metals could transport and accumulation in surface waterbodies. Different metal concentrations in water which were correlated with the correlation coefficient of $p < 0.01$ showed their relationship as: Pb-Cr, Pb-Cu, Pb-Mn, Cr-Cu, Cr-Se, and Cu-Se. The metal pairs which displayed correlations by their extents in water with a correlation coefficient of $p < 0.05$ were: Pb-Se, Cr-Mn, Cu-Mn, Zn-Ni, Ca-Mn, Ca-Co, Mn-Co, and Mn-Se. From the metal's correlation analyses data, it has been observed that Pb is positively correlated with Cr, Cu, Mn, Se. Similarly, Cr are positively correlated with Pb, Cu, Se, and Mn as well as Cu is positively correlated with Pb, Cr, Se and Mn respectively. On the hand, Zn is positively correlated with Ni and Ca is positively correlated with Mn and Co as well as Mn showed positive correlation with Ca, Co, Cu, Pb, Cr, and Se respectively. Different metals and metalloid concentrations determined in all water samples were found to be strongly correlated to each other which demonstrated their common sources and similar pattern of release into respective environmental sources.

4.1.2 Heavy metals and metalloids contamination in soil

The results of different metal concentrations (Pb, Cd, Cr, Cu, Zn, Fe, and Ca) measured in all soil samples collected from various fruits plants fields around DEPZ have been displayed in the Table 4.4.

Table 4.4: Different metals and metalloids concentrations (mgkg^{-1} dry soil) in soil determined by AAS

Soil samples collected from different fruits plants fields	Pb	Cd	Cr	Cu	Zn	Fe	Ca

Psidium guajava (Guava)	42.5431 ± 0.0031	0.2971 ± 0.0130	44.6425 ± 0.0027	28.2927 ± 0.0033	131.7092 ± 0.1245	26783.5215 ± 0.0162	1745.2961 ± 0.0583
Citrus maxima (Pomelo)	47.2005 ± 0.0005	0.3598 ± 0.0231	38.4194 ± 0.0085	27.3431 ± 0.0081	150.0609 ± 0.0241	26531.0381 ± 0.0124	18023.8219 ± 0.0063
Musa paradisiaca (Banana)	36.6696 ± 0.0028	0.3527* ± 0.0171	41.9643 ± 0.0031	26.5446 ± 0.0070	113.0357 ± 0.0222	24439.2857 ± 0.0220	2272.7679 ± 0.0201
Mean	42.1377	0.3365	41.6754	27.3935	131.6019	25917.9484	7347.2953
Minimum	36.6696	0.2971	38.4194	26.5446	113.0357	24439.2857	1745.2961
Maximum	47.2005	0.3598	44.6425	28.2927	150.0609	26783.5215	18023.8219
Safe Limit[a]/ Permissible Value[a] WHO (ppm)	85	0.8	100	36	50	–	–
Safe Limit[b]/ Permissible Value[b] World limit (mg kg^{-1})	35	0.35	70	30	90	1000	–
Safe Limit[c]/ Permissible Value[c] MAC of elements in agricultural soil	350	0.6	200	100	300	–	–

in China(mg kg^{-1})							
Safe Limit^{c}/ Permissible Value^{c} Threshold of elements in natural background soil in China(mg kg^{-1})	35	0.20	90	35	100	–	–
Safe Limit^{d}/ Permissible Value^{d} Safe Limit of India (mg kg^{-1})	250-500	3-6	–	135-270	300-600	–	–

a,b,c,d are the references from where the permissible values or safe limits will be taken or adopted.

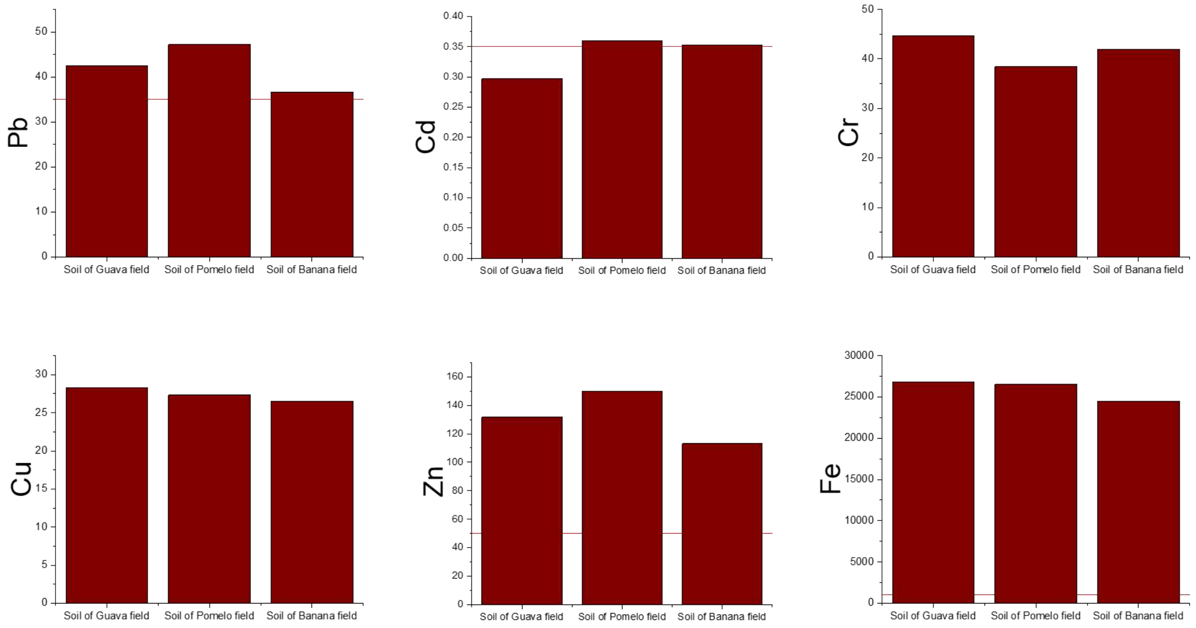
[a]Yaqub et al. (2021), [b]Coskun et al. (2006); [c]National Environmental Protection Agency of China, GB 15618 (1995); [d]Awashthi (2000)

The permissible limits of some metals were not available in literatures, and they were denoted as blank in the respective places.

* Indicates that the data were determined by ICP-MS. These metals concentrations were found as BDL (Below the Detection Limit) in AAS measurements.

All metals and metalloid concentrations (mg kg^{-1} dry soil) determined in various soil samples of three fruits plants were ranged from 36.6696 to 47.2005 for Pb, 0.2971 to 0.3598 for Cd, 38.4194 to 44.6425 for Cr, 26.5446 to 28.2927 for Cu, 113.0357 to 150.0609 for Zn, 24439.2857 to 26783.5215 for Fe and 1745.2961 to 18023.8219 for Ca (Table 4.4). The highest concentration of Pb was 47.2005 mg kg^{-1} found in soil from *Citrus maxima* field, Cd was 0.3598 mg kg^{-1} observed in *Psidium Citrus maxima* field, Cr was 44.6425 mg kg^{-1} measured in *Psidium guajava* (Guava) field, Cu was 28.2927 mg kg^{-1} also found in *Psidium guajava* (Guava) field, Zn was 150.0609 mg kg^{-1} found in *Citrus maxima* (Pomelo) field, Fe was 26783.5215 mg kg^{-1} found in *Psidium guajava* (Guava) field , and Ca found in *Citrus maxima* (Pomelo) field was 18023.8219 mgkg^{-1}. With the exception of Ca and Fe, the mean highest concentration of metal in soil was recorded for Zn followed by Pb, Cd, Cr, and Cu and the minimum concentration was observed for Cd which were comparable with results of similar study reported previously by Singh et al. (2011) [5].

The appearance of the highest Fe and Ca deposition in the studied soil from different fruits fields might probably be due to its long-term use in the production of machines, paints, and pigments in various industries built in that area. Except for Cd, Cr and Cu the metal levels found in different soil samples from three fruits plant fields exceeded the world limit [6]. Mean metals contents measured in soil in the study area were compared with the maximum allowable concentration (MAC) of elements in the agricultural soil of China, the Threshold of elements in natural background soil in China, WHO, and the safe Limit of India. Except Zn, all metals concentrations were observed to be below the safe limit of WHO [1]. According to metal threshold levels reported from China, concentrations of Cr and Cu were found to be below the permissible limits [2]. However, all metals and metalloids concentrations in soil from different fruits plants were within safe limit of India [3]. The trend of different metals contents in experimental soil system followed the order of Ca > Fe > Zn > Pb > Cr > Cu > Cd. Average concentrations of Fe, Pb, Cd, Cr and Zn in soil of three fruits fields were higher than those reported from similar studies conducted previously in Bangladesh as well as in Northwest China [7, 8, 9]. These variations of the results might be occurred due to the changes in metal concentrations in irrigation water and other agronomic practices in the respective areas [10]. The higher standard deviation reveals higher variations in metal distributions from point sources of emission to the adjacent areas. The low concentrations of metals in soil might be ascribed to its continuous removal by crops, vegetables, and fruits grown in the designated areas. Among the different metals and metalloids examined in soil from different fruits fields, concentrations of Fe, Zn, and Pb were the maximum, and variations in their concentrations were several times higher than those reported by Kisku et al. (2000) [11]. The highest deposition of Fe in the soil might be due to its long-term use in the production of machine tools, paints, pigments, and alloying in various industries of the study area which might have resulted the contamination of arable soil as well as changed the soil structure and thus making it risky for subsequent use in cultivation [12].



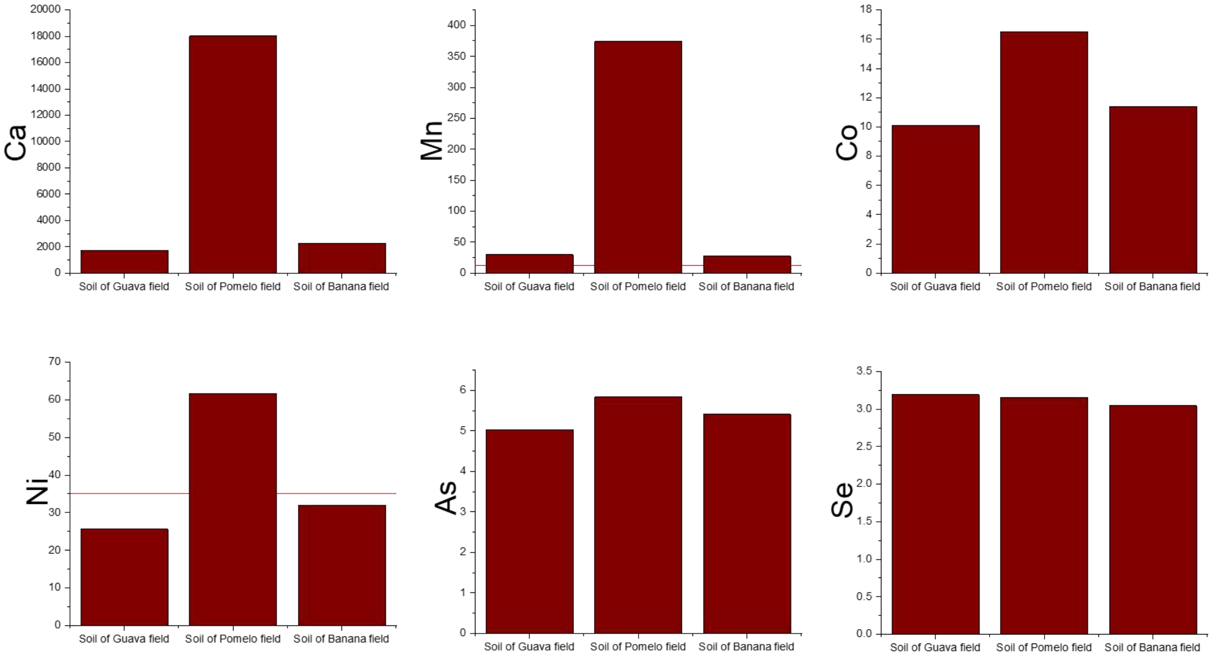


Figure 4.2. Different metals and metalloids concentrations in soil from three different fruits plants fields

Table 4.5. Different metals and metalloids concentrations in soil (mg kg^{-1} dry soil) from different fruits plants fields determined by ICP-MS (Mean ± S.D.)

Fruits name (Soil)	Mn	Co	Ni	As	Se
Psidium guajava (Guava)	29.7257 ± 0.0121	10.1000 ± 0.0258	25.6585 ± 0.0318	5.0347 ± 0.0201	3.1917 ± 0.0328
Citrus maxima (Pomelo)	373.8480 ± 5.5024	16.5189 ± 0.3724	61.7284 ± 1.5630	5.8412 ± 0.0278	3.1568 ± 0.0323
Musa paradisiaca (Banana)	27.4579 ± 0.3223	11.4080 ± 0.1935	31.9801 ± 0.5935	5.4054 ± 0.0467	3.0469 ± 0.0335
Mean	143.6772	12.6756	39.7890	5.4271	3.1318
Minimum	27.4579	10.1000	25.6585	5.0347	3.0469
Maximum	373.848	16.5189	61.7284	5.8412	3.1917
Safe Limit[a]/	12	20	35	_	_

Permissible Value[a] WHO (ppm)					
Safe Limit[b]/ Permissible Value[b] World limit (mg kg^{-1})	_	_	50	_	_
Safe Limit[c]/ Permissible Value[c] MAC of elements in agricultural soil in China (mg kg^{-1})	_	_	60	_	_
Safe Limit[c]/ Permissible Value[c] Threshold of elements in natural background soil in China (mg kg^{-1})	_	_	40	_	_
Safe Limit[d]/ Permissible Value[d] Safe Limit of India (mg kg^{-1})	_	_	75-100	_	_

a,b,c,d are the references from where the permissible values or safe limits will be taken or adopted.

[a] Yaqub et al. (2021), [b] Coskun et al. (2006); [c] National Environmental Protection Agency of China, GB 15618 (1995); [d] Awashthi (2000).

The permissible limits of some metals were not available in literatures, and they were denoted as blank in the respective places.

Metals accumulations in soil were found to be varied highly and followed the order of Mn > Ni > Co > As > Se. The average concentrations of Mn, Co, Ni, As and Se in soil (0-20 cm depth) were 143.6772, 12.6756, 39.7890, 5.4271, 3.1318 mg/kg respectively (Table 4.5). Compared with other

Mn and Ni contents in soil from the present study were observed to be higher than those reported in literature from similar study conducted by Kisku *et al.* (2000) [11]. Except for Co, concentrations of all metals and metalloids in soil were found to be higher than the WHO standard limits set for arable soils. However, the extents of Mn in all soil samples were below the permissible levels reported as the world limit, MAC of elements in agricultural soil in China, threshold of elements in natural background soil in China and safe limit of India recommendation level. Comparison of all metals and metalloids concentrations measured in soil by both AAS and ICP-MS techniques, the extent of iron (Fe) was observed to be higher, and cadmium (Cd) was found to be lower in each of the soil samples collected from three different fruits plants fields. The average concentrations of twelve elements determined in all soil samples followed the descending order of: Fe > Ca > Mn > Zn > Pb > Cr > Ni > Cu > Co > As > Se > Cd. From the analyses of all metals and metalloids in soil from the present study, it has been apparent that arable soil from *Citrus maxima* (Pomelo) plant field accumulated more metals whereas the relative quantities of all twelve elements were found lower in soil of *Musa paradisiaca* (Banana) plant field.

Correlation analysis of different metals in soil:

Pearson correlation analysis was performed among the metals determined in all soil samples and the results have been shown in Table 4.6. between all the variables. The level of significance ($p \leq 0.05$ and $p \leq 0.01$) of multi-element correlation for soil samples was determined and the results are given in Table 4.6. The magnitudes of r with $p \leq 0.05$ and $p \leq 0.01$ indicated the high degree of positive correlations and significant linear relationship between various pairs of metals indicate that they might have originated from common sources and showed similar pattern of release into different environmental sources. Ni in soil was found to be strongly correlated with Co ($p < 0.01$). Similarly, Pb in soil was positively correlated with Zn ($p < 0.05$) and Ca showed positive correlation with Mn ($p < 0.05$).

Table 4.6. Correlation among metals concentrations measured in soil from three fruits fields

	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Pb	1											
Cd	0.037	1										
Cr	-0.512	-0.877	1									
Cu	0.514	-0.838	0.473	1								
Zn	0.998^*	0.098	-0.564	0.461	1							

Fe	0.850	-0.495	0.018	0.889	0.816	1						
Ca	0.815	0.610	-0.915	-0.078	0.849	0.386	1					
Mn	0.834	0.582	-0.901	-0.044	0.866	0.418	0.999*	1				
Co	0.708	0.732	-0.969	-0.241	0.750	0.229	0.986	0.980	1			
Ni	0.728	0.712	-0.961	-0.213	0.769	0.258	0.991	0.985	1.000**	1		
As	0.483	0.893	-0.999*	-0.503	0.536	-0.052	0.901	0.886	0.960	0.952	1	
Se	0.771	-0.608	0.152	0.943	0.731	0.991	0.259	0.292	0.096	0.125	-0.185	1

* Correlation is significant at the 0.05 level (2-tailed);**Correlation is significant at the 0.01 level (2-tailed)

4.1.3 Heavy metals and metalloids contents in different parts of three fruits plants bodies

Table 4.7. Various metal concentrations(mg/kg) in different parts of three fruits plants bodies determined by AAS. (Mean ± S.D.)

Type of Fruits Plants	Plant parts	Metals contents (ppm) (Mean± SD)						
		Pb	Cd	Cr	Cu	Zn	Fe	Ca
Psidium guajava (Guava)	Root	41.0290 ± 0.0115	0.3297 ± 0.0102	4.0959 ± 0.0062	9.9900 ± 0.0026	81.0490 ± 0.0232	1278.8212 ± 0.0055	10896.1039 ± 0.0026
	Stem	4.0362 ± 0.0104	0.3133 ± 0.0022	5.4680 ± 0.0093	10.3000 ± 0.0206	36.6812 ± 0.0423	126.7610 ± 0.0805	4246.2953 ± 0.1868

	Leaves	22.4814 ± 0.0026	0.7046 ± 0.0023	5.7132* ± 0.3711	25.3380 ± 0.0106	64.9495 ± 0.0317	420.5294 ± 0.0125	20936.0122 ± 0.0024
	Edible part	3.9967 ± 0.1235	0.3221* ± 0.0086	2.9537 ± 0.0039	8.0949 ± 0.0085	5.5104 ± 0.0404	15.8852 ± 0.0115	508.4918 ± 0.0520
Citrus maxima (Pomelo)	Root	15.0452* ± 0.0802	0.3324 ± 0.0078	1.6522 ± 0.0027	4.1351 ± 0.0105	32.7857 ± 0.0131	470.8326 ± 0.0210	33490.8621 ± 0.0085
	Stem	BDL	0.1549* ± 0.0171	2.0916 ± 0.0047	3.2191 ± 0.0065	24.9354 ± 0.0032	174.5309 ± 0.0032	38065.9322 ± 0.0200
	Leaves	9.5129 ± 0.0040	0.3836 ± 0.0087	9.5723* ± 0.0185	7.0579 ± 0.0185	20.0326 ± 0.0147	265.7748 ± 0.0439	79909.8581 ± 0.1212
	Edible part	15.6711 ± 0.0013	0.3005* ± 0.0456	18.8053 ± 0.7410	5.4296 ± 0.0097	13.7998 ± 0.0611	24.7972 ± 0.0213	2933.9049 ± 0.0692
Musa paradisiaca (Banana)	Root	81.6932 ± 0.0100	0.3274 ± 0.0062	4.1908 ± 0.0044	11.1880 ± 0.0106	40.3461 ± 0.0193	1591.3938 ± 0.0043	21302.1515 ± 0.0353
	Stem	1.3776 ± 0.0086	0.3011 ± 0.0010	19.7027* ± 0.5465	1.7874 ± 0.0082	41.0433 ± 0.0154	198.5137 ± 0.0074	23798.3291 ± 0.0156
	Leaves	39.2186 ± 0.0062	1.3146 ± 0.0058	1.0498 ± 0.0100	4.2542 ± 0.0075	47.2430 ± 0.0540	4616.8523 ± 0.0154	33267.2996 ± 0.0108

	Edible part	0.9403 ± 0.0118	0.2447* ± 0.0291	16.5236 ± 0.3880	6.2169 ± 0.0064	14.2322 ± 0.0144	36.7446 ± 0.0188	1577.9624 ± 0.0343
Mean	Root	28.0371	0.3311	2.8741	7.0626	56.9174	874.8269	22193.4830
Mean	Stem	4.0362	0.3133	5.4680	10.3000	36.6812	126.761	42462.5024
Mean	Leaves	22.4814	0.7046	5.7132	25.3380	64.9495	420.5294	20936.0122
Mean	Edible part	3.9967	0.3221	2.9537	8.0949	5.5104	15.8852	508.4918
Safe Limit[a]/ Permissible Value[a]	WHO (1996) (mg/kg)	2	0.02	1.30	10	0.6	–	–
Safe Limit[b]/ Permissible Value[b]	MAC (FAO/ WHO, 2002) (mg/kg)	0.1	0.05	1.0	4.5		–	–
Safe Limit[c]/ Permissible Value[c]	WHO (ppm)	–	0.02	1.3	10	60	–	–
Safe Limit[d]/ Permissible Value[d]	FAO/WHO, 1989 $mg.kg^{-1}$	–	–	–	–	60	450.0	–

a,b,c,d are the references from where the permissible values or safe limits will be taken or adopted.

[a]Emile et al. (2020); [b]Shaheen et al. (2016); [c]Yaqub et al. (2021); [d]Dhar et al. (2019)

The permissible limits of some metals were not available in literatures, and they were denoted as blank in the respective places.

* Indicates that the data were determined by ICP-MS. These metals concentrations were found as BDL (Below the Detection Limit) in AAS measurements.

Different heavy metals and metalloids contents have been determined in various parts of three fruits plants bodies and their results have been shown in table 4.7. All metals and metalloids concentrations were found to be highly varied among various parts of three fruits plant bodies. Variations in metals and metalloids concentrations were also realized with the type and nature of fruit's plants. Among the eight elements studied in different fruits plants, the highest concentrations were found for Ca and Fe, followed by Cu > Zn > Pb > Cr > Cd (Table 4.7). Among four different parts of three fruits plants bodies, accumulation and deposition of all metals and metalloids were much higher in roots in comparison to stems, leaves, and edible parts. The lowest uptake of metals and metalloids were observed in the edible parts and their concentrations in different parts of three fruits plants showed the order of roots > leaves > stems > edible parts (Table 4.7). All metals and metalloids concentrations determined in the edible parts of the three fruits plants were compared with the permissible levels of WHO and FAO. The results indicated that average concentrations of Pb, Cd, Cr, and Zn in the edible part of three fruits plants were higher than the maximum tolerable levels of WHO (1996) [13]. The average concentrations of Fe, Pd, Cd, Cu, and Cr in the fruits were observed to be higher than maximum allowable concentrations (FAO/WHO, 2002; Yaqub et al. 2021; Dhar et al. 2019) [14, 15]. Among three fruits plants studied in the present investigation, accumulation of metals and metalloids were found highest in *Citrus maxima* (Pomelo) and their concentrations in the edible parts of different fruits plants followed the order of *Citrus maxima* (Pomelo) > *Musa paradisiaca* (Banana) > *Psidium guajava* (Guava). Variations in metals and metalloids uptake and deposition in the fruit's plants might be attributed to the type and nature of plants, differences in their metal's accumulation capacities, and characteristics of their growth media such as soil and water properties [16].

Apart from its function as a biocatalyst, Cu is necessary for body pigmentation, prevention of anemia, and maintenance of healthy central nervous system as well as it is interrelated with the functions of Zn and Fe in the body [17]. Zn is also an essential element for plants and animals, but only a small increase in its level may interfere the physiological processes. Zn is airborne pollutant and it might accumulate in fruits plants from the atmospheric deposition from the emission of surrounding metal industries as well as vehicle exhaust in the study areas [20]. Usually, Fe participates in chlorophyll synthesis and photosynthesis and green plants may accumulate and retain higher quantity of Fe [18]. However, excess uptake of Fe in human body from the consumption of iron enriched food items may play a role in etiology of heart disease and type 2 diabetes [19]. Out of three different sources studied in the present investigation, soil from three fruits fields accumulated highest quantities of all metals and metalloids and their corresponding concentrations were followed the order of soil > fruits > water. Soil system could work as sink of metals and metalloids which could accumulate and deposit inorganic, organic and other species from various sources. The main sources of metals and metalloids in plants are their growth media from which they can uptake, accumulate, and distribute these inorganic species in roots, stems, leaves, and edible parts. Excessive presence of any metal, metalloid, mineral, and other chemical species in irrigation waterbodies and arable soil can severely impact the uptake and distribution of nutrients contents in plants bodies which ultimately caused the appearance of higher quantities of metallic and other species in different organs such as edible parts, leaves, stems, and roots of the respective plant bodies.

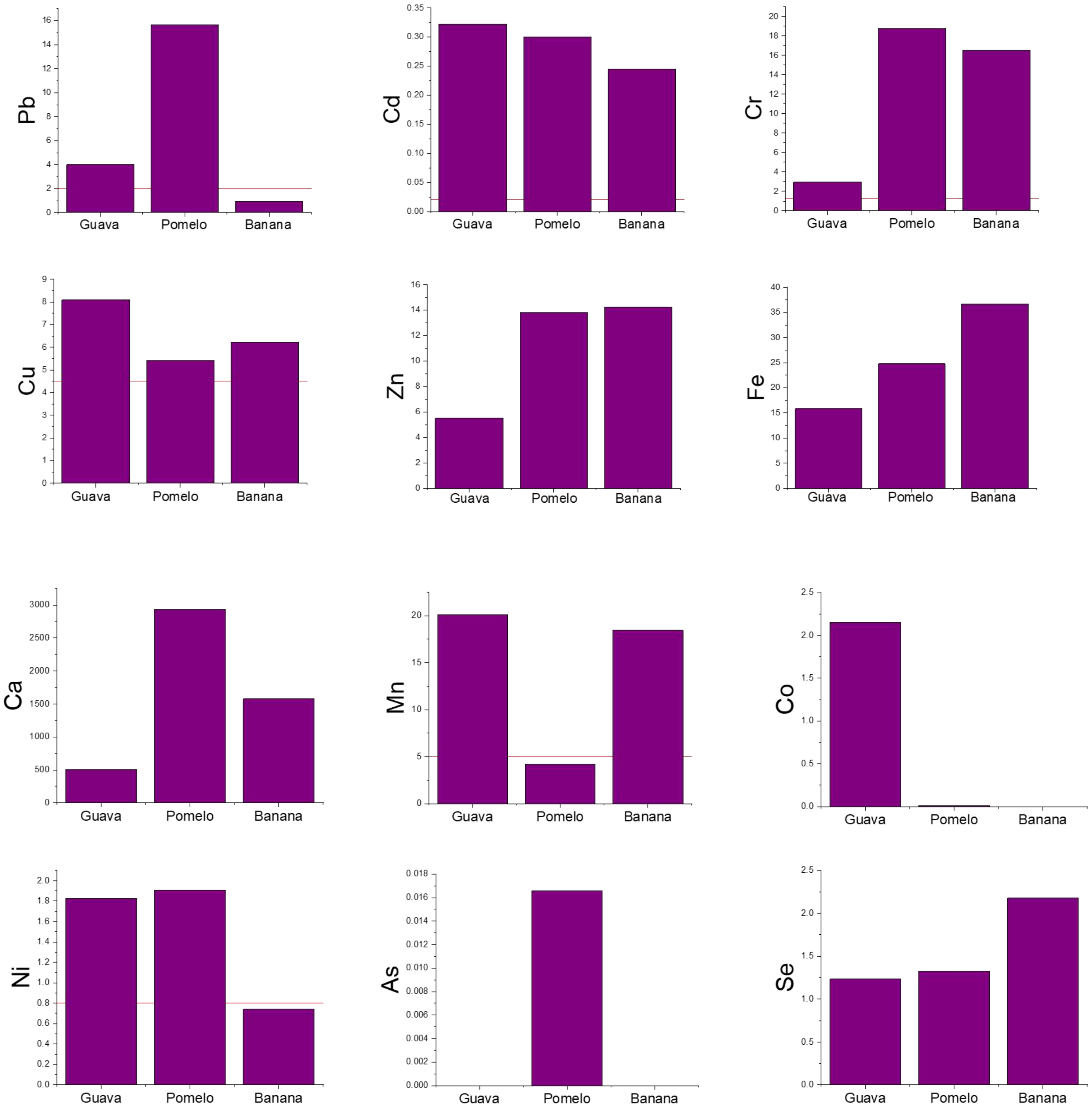


Figure 4.3. Different metals and metalloids concentrations in the edible parts of three fruits plants

Table 4.8: Different heavy metals and metalloids concentrations (mg/kg) in various parts of three fruits plants bodies determined by ICP-MS. (Mean ± S.D.)

Type of Fruits Plants	Plant parts	Metals contents (ppm) (Mean± SD)				
		Mn	Co	Ni	As	Se
Psidium guajava (Guava)	Root	BDL	1.9291 ± 0.0132	7.4765 ± 0.0298	1.2268 ± 0.0123	1.3626 ± 0.0248
	Stem	BDL	1.7125 ± 0.0051	6.0423 ± 0.0165	0.1927 ± 0.0079	0.3000 ± 0.0106
	Leaves	104.2981 ± 2.6732	0.2333 ± 0.0079	26.6616 ± 0.4962	0.5140 ± 0.0108	1.5707 ± 0.0351
	Edible part	20.0905 ± 0.0437	2.1506 ± 0.0138	1.8248 ± 0.0102	BDL	1.2341 ± 0.0185
Citrus maxima (Pomelo)	Root	29.0899 ± 0.1247	1.1049 ± 0.0517	38.7668 ± 0.5201	0.8355 ± 0.0302	1.6330 ± 0.0185
	Stem	13.9654 ± 0.0175	0.0757 ± 0.0087	2.0830 ± 0.0163	0.2298 ± 0.0152	0.3142 ± 0.0119
	Leaves	28.2806 ± 0.0208	0.1266 ± 0.0142	4.2578 ± 0.0201	0.3951 ± 0.0594	1.3982 ± 0.0416
	Edible part	4.1676 ± 0.0013	0.0129 ± 0.0018	1.9073 ± 0.0095	0.0166 ± 0.0063	1.3293 ± 0.0136
Musa paradisiaca (Banana)	Root	33.0627 ± 0.0258	0.7119 ± 0.0041	2.8120 ± 0.0116	0.5510 ± 0.0132	1.6529 ± 0.0152
	Stem	1.0686 ± 0.0100	0.6824 ± 0.0072	6.9531 ± 0.0386	0.1778 ± 0.0032	0.9476 ± 0.0263
	Leaves	62.1946 ± 0.0378	0.4546 ± 0.0279	4.7115 ± 0.0838	0.9850 ± 0.0070	1.4835 ± 0.0246
	Edible part	18.4462 ± 0.0173	BDL	0.7404 ± 0.0258	BDL	2.1782 ± 0.0283

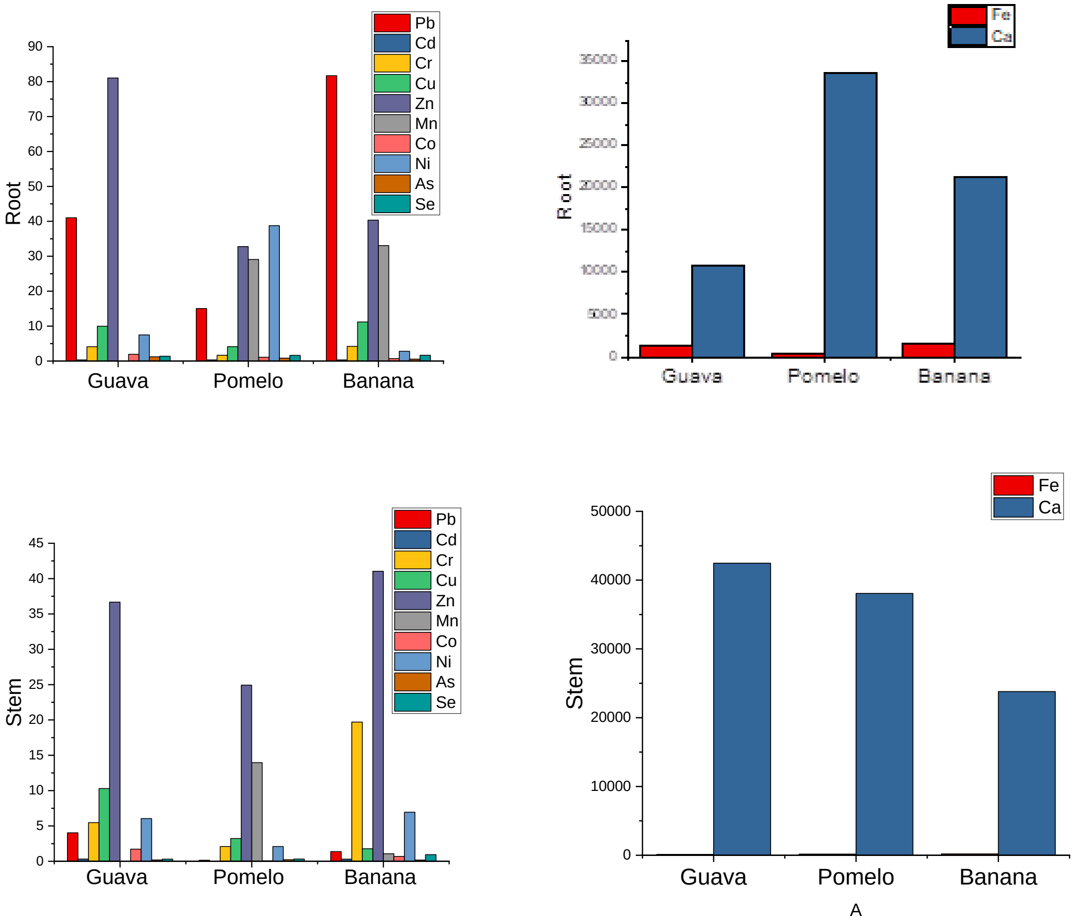
Mean	Root	20.7175	1.2486	16.3518	0.8711	1.5495
Mean	Stem	5.0113	0.8235	5.0261	0.2001	0.5206
Mean	Leaves	64.9244	0.2715	11.8770	0.6314	1.4841
Mean	Edible part	12.1291	1.0818	1.8661	0.0083	1.2817
Safe Limit[a]/ Permissible Value[a]	WHO (1996) (mg/kg)	_	_	10	_	_
Safe Limit[b]/ Permissible Value[b]	MAC (FAO/WHO, 2002) (mg/kg)	_	_	0.8	1	_
Safe Limit[c]/ Permissible Value[c]	WHO (ppm)	5	_	10	_	_
Safe Limit[d]/ Permissible Value[d]	FAO/WHO ,1989 $mgkg^{-1}$	500	_	1.0	0.10	_

a,b,c,d are the references from where the permissible values or safe limits have been adopted.

[a]Emile et al. (2020); [b]Shaheen et al. (2016); [c]Yaqub et al. (2021); [d]Dhar et al. (2019).

The permissible limits of some metals were not available in literatures, and they were denoted as blank in the respective places.

The results ICP-MS analysis of different elements such as Mn, Co, Ni, As, and Se in various parts of three fruits plant bodies have been summarized in Table 4.8 and Figures 4.3. All elemental concentrations in fruits determined by ICP-MS were based on samples dry weight. The ICP-MS data showed the average metals and metalloid contents (mg/kg dry weight) in the edible parts of three fruits plants as: 12.1291 for Mn, 1.0818 for Co, 1.8661 for Ni, 0.0083 for As and 1.2817 for Se respectively. Among the selected three fruit plants analyzed by ICP-MS, the highest concentrations of Mn, Co, Ni and Se were noticed in *Psidium guajava* (Guava) followed by *Citrus maxima* (Pomelo) and *Musa paradisiaca* (Banana). The higher levels of heavy metals and metalloids found in different fruit samples could be closely related to the presence of the corresponding pollutants in irrigation water, farm soil, and pesticides or alternatively could be due to pollution from traffic on the nearby highways [21]. According to guidelines of WHO (1996) Ni concentrations in all fruits samples were observed to be below the safe limit whereas Mn contents were higher than the respective threshold values [1,13]. However, following the maximum allowable concentrations (MAC) reported by FAO/WHO (2002), Ni contents in experiment fruits items were higher whereas As concentrations were found to be lower than the recommended threshold values [14]. Irrespective of the types of fruits plants studied, the data obtained from ICP-MS analysis were highly varied depending on the elements examined and the extents of different heavy metals and metalloids contents were found to be in the order of: Mn > Ni > Se > Co > As. The edible part of Guava plants showed much higher metals and metalloids contents which were determined by ICP-MS and their overall concentrations in different edible parts of the three fruits plants were found to be varied in the order of Guava > Pomelo > Banana. Roots of all three fruits plants showed much higher accumulation of Mn, Ni, Se, Co, As, and the distribution of the concentrations of these elements with other plants organs showed the order of root > leaves > stem > fruit (edible part) (Table 4.8). The variations in metals and metalloid contents in different organs of various fruits plants bodies might be ascribed to the physicochemical properties of water sources used for irrigation, nature of metals, fruits types, soil properties, and atmospheric deposition of HMs [22-25]. Among twelve elements studied in various parts of three fruits plants bodies by using AAS and ICP-MS, the extent of calcium (Ca) was found higher, and the minimum concentration was realized with arsenic (As) irrespective of the plant parts and fruits plants types. Concentrations of twelve different elements measured in the three fruits plants followed a descending order of Ca > Fe > Mn > Cu > Zn > Pb > Cr > Ni > Se > Co > Cd > As. Different heavy metals and metalloid concentrations determined by ICP-MS in various fruits also compared with the permissible levels set by WHO (1996), MAC (FAO/WHO, 2002), WHO and FAO/WHO, 1989 and the respective results have been shown in the table 4.7 and table 4.8.



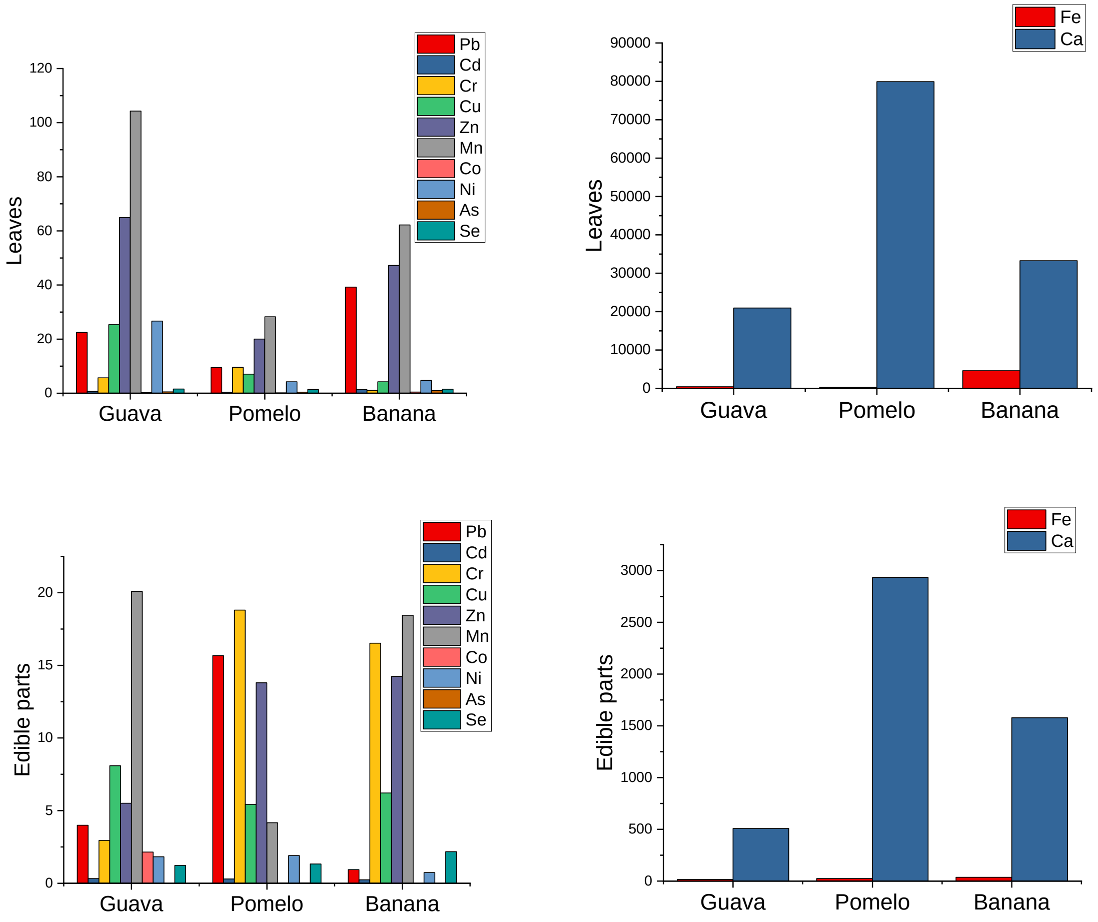


Figure 4.4. Different metals and metalloids concentrations in various parts (root, stem, leaves, edible parts) of Guava, Pomelo and Banana plants

N.B. Fe and Ca contents in various parts of the three fruits plants have been shown in a separate figure as their concentrations were found much higher than the extents of other elements observed in the respective plants' species.

Correlation analysis among heavy metals and metalloids contents measured in the fruits

Table 4.9 showed the statistical analysis of metal-to-metal correlation matrix in terms of linear correlation coefficient (r) magnitudes (significant at 0.05 and 0.01) in different parts of the fruits plant bodies. The estimated r values reveal the degree of positive correlations and significant linear relationship between various pairs of metals as well as reflect their simultaneous releasing patterns and identical sources. The metallic pairs which showed positive correlations with $p < 0.01$ in their accumulations in different parts of fruits plants bodies were: Zn-As for stem, Pb-Cr for leaves and Pb-Ni for edible part. On the other hand, the pairs of elements which indicated positive correlations with $p < 0.05$ in their uptake and translocations within the different parts of three fruits plant bodies were: Cr-Ni, Cu-Ni, Mn-Se for root, Pb-Co, Cd-Mn, Zn-Ni for stem, Fe-Se for leaves and Pb-As, Cu-Co for edible parts. The correlation analysis data shows that in the roots of three fruits plants: Cr is positively correlated with Ni, Cu is positively correlated with Ni and Mn is positively correlated with Se. Among the metal's contents found in stems: Zn is positively correlated with As, Pb is positively correlated with Co, Cd is positively correlated with Mn and Zn is positively correlated with Ni. However, for different metals contents observed in leaves: Pb is positively correlated with Cr and Fe is positively correlated with Se. In the edible parts of three fruits plants: Pb is positively correlated with Ni and As, Cu is positively correlated with Co.

Table 4.9. Correlations among metals contents found in various parts of the fruits plants bodies

	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Roots:												
Pb	1											
Cd	-0.985	1										
Cr	0.816	-0.903	1									
Cu	0.882	-0.950	0.992	1								
Zn	0.020	-0.191	0.595	0.488	1							
Fe	0.930	-0.979	0.971	0.993	0.386	1						
Ca	-0.428	0.577	-0.872	-0.804	-0.912	-0.730	1					
Mn	0.235	-0.064	-0.371	-0.251	-0.967	-0.139	0.778	1				
Co	-0.433	0.272	0.168	0.042	0.892	-0.072	-0.629	-0.978	1			
Ni	-0.863	0.937	-0.996*	-0.999*	-0.523	-0.988	0.827	0.290	-0.082	1		

As	-0.530	0.377	0.058	-0.068	0.837	-0.182	-0.539	-0.948	0.994	0.029	1	
Se	0.187	-0.015	-0.416	-0.298	-0.979	-0.187	0.808	0.999*	-0.966	0.336	-0.932	1

Stems:

Pb	1											
Cd	0.765											
Cr	-0.029	0.622	1									
Cu	0.891	0.390	-0.479	1								
Zn	0.541	0.956	0.825	0.101	1							
Fe	-0.798	-0.222	0.626	-0.984	0.076	1						
Ca	0.424	-0.259	-0.918	0.789	-0.532	-0.884	1					
Mn	-0.787	-0.999*	-0.594	-0.421	-0.945	0.255	0.226	1				
Co	0.998*	0.804	0.033	0.861	0.592	-0.758	0.367	-0.824	1			
Ni	0.613	0.978	0.772	0.188	0.996*	-0.012	-0.456	-0.970	0.661	1		
As	-0.527	-0.950	-0.834	-0.084	-1.000**	-0.093	0.547	0.939	-0.579	-0.994	1	
Se	-0.228	0.453	0.980	-0.645	0.696	0.769	-0.978	-0.422	-0.167	0.630	-0.708	1

Leaves:

Pb	1											
Cd	0.995	1										
Cr	-1.000**	-0.992	1									
Cu	-0.195	-0.296	0.176	1								
Zn	0.541	0.451	-0.557	0.719	1							
Fe	0.913	0.951	-0.906	-0.577	0.152	1						
Ca	-0.699	-0.621	0.713	-0.565	-0.980	-0.348	1					
Mn	-0.787	-0.718	0.798	-0.453	-0.945	-0.467	0.991	1				
Co	0.298	0.197	-0.315	0.878	0.964	-0.117	-0.891	-0.824	1			
Ni	0.913	0.866	-0.921	0.222	0.837	0.668	-0.930	-0.970	0.661	1		
As	-0.951	-0.913	0.956	-0.119	-0.775	-0.743	0.886	0.939	-0.579	-0.994	1	
Se	0.892	0.934	-0.883	-0.617	0.102	0.999*	-0.300	-0.422	-0.167	0.630	-0.708	1

Edible parts:

Pb	1

Cd	0.433	1										
Cr	0.443	-0.616	1									
Cu	-0.579	0.485	-0.988	1								
Zn	0.278	-0.746	0.984	-0.944	1							
Fe	-0.278	-0.986	0.738	-0.622	0.845	1						
Ca	0.794	-0.204	0.897	-0.955	0.805	0.363	1					
Mn	0.965	0.180	0.664	-0.773	0.521	-0.015	0.926	1				
Co	-0.645	0.410	-0.971	0.996*	-0.913	-0.555	-0.977	-0.824	1			
Ni	-1.000**	-0.414	-0.462	0.596	-0.298	0.258	-0.807	-0.970	0.661	1		
As	0.996*	0.507	0.366	-0.508	0.196	-0.358	0.740	0.939	-0.579	-0.994	1	
Se	-0.646	-0.968	0.398	-0.249	0.554	0.913	-0.049	-0.422	-0.167	0.630	-0.708	1

* Correlation is significant at the 0.05 level (2-tailed); **Correlation is significant at the 0.01 level (2-tailed)

Heavy metals and metalloids concentrations in different parts of *Psidium guajava* (Guava) plant:

Concentrations of different heavy metals and metalloids determined in various parts of *Psidium guajava* (Guava) plants have been shown in the Table 4.7 and Table 4.8. Among four plant parts, root of Guava plant accumulated highest amount of Pb 42.5431 mg kg^{-1} and the lowest amount of Pb was 3.9967 mg kg^{-1} found in the edible parts. Cd was mostly deposited in the leaves which was 0.7046 mg kg^{-1} and the minimum amount of Cd was 0.3133 mg kg^{-1} determined in the stems of Guava plants. The leaves of Guava plants accumulated maximum amount of Cr which was 5.7132 mg kg^{-1} whereas the edible showed the lowest uptake of Cr (2.9537 mg kg^{-1}). The leaves of Guava plants deposited maximum amount of Cu which was 25.3380 mg kg^{-1} and the least amount, 8.0949 mg kg^{-1} was measured in the edible part. Uptake of Zn was found highest in root which was 81.0490 mgkg^{-1} whereas the minimum Zn content was 5.5104 mgkg^{-1} measured in the edible parts. Similarly, the roots of Guava plants absorbed maximum quantity of Fe (1278.8212 mg kg^{-1}) and the lowest Fe content (15.8852 mg kg^{-1}) was observed in the edible part. However, in the case of Ca, the leaves of Guava plants showed the maximum uptake (20936.0122 mgkg^{-1}) whereas the edible part displayed the least quantity of Ca (508.4918 mgkg^{-1}). Uptake of Mn was also found maximum in the leaves of Guava plant which was 104.2981 mgkg^{-1} and the lowest Mn content was observed in root followed by stem. The highest concentration of Co was 2.1506 mgkg^{-1} measured in the edible part whereas the lowest Co content was 0.2333 mgkg^{-1} found in the leaves of Guava plants. The leaves of Guava fruits accumulated maximum amount of Ni (26.6616 mgkg^{-1}) whereas the edible part contain relatively lower quantity of the respective metal which was 1.8248 mgkg^{-1}. Most of the amount of As was accumulated in the root which was 1.2268 mgkg^{-1} whereas the least amount was observed in the edible parts. Se concentration was found highest in leaves which was 1.5707 mg kg^{-1} whereas the lowest amount of Se was 0.3000 mg kg^{-1} measured in the stem. It has been apparent that leaves accumulated more heavy metals and metalloids in comparison to those observed in root, stem, and edible of Guava plants. Plants leaves can accumulate heavy metals and other elements through various processes including air pollution, soil contamination, and direct deposition of metal-containing particles onto their surfaces.

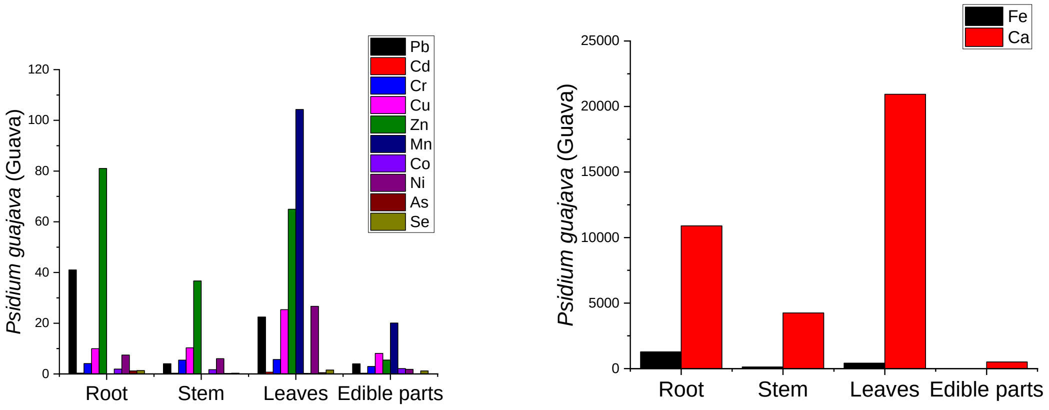


Figure 4.5. Different metals and metalloids concentrations in different parts of *Psidium guajava* (Guava) plant. Concentrations of Fe and Ca are shown in separate figures

Heavy metals and metalloids concentrations in different parts of Pomelo fruit plant

Different metals and metalloids contents determined in various parts of *Citrus maxima (Pomelo)* plant have been shown in the table 4.7 and table 4.8. The edible part accumulated highest amount of Pb, 15.671 mgkg^{-1} whereas the Pb content measured in the stem of Guava plant was found below the detection limit. Cd was mostly absorbed in the leaves (0.3836 mg kg^{-1}) and the least amount of Cd was 0.1549 mg kg^{-1} measured in the stem of Pomelo plant. The edible parts of Pomelo plant accumulated 18.8053 mgkg^{-1} of Cr whereas the minimum amount was 1.6522 mg kg^{-1} determined in the root. Cu was mostly absorbed in the leaves (7.0579 mg kg^{-1}) and the least amount of Cu was measured in stem which was 3.2191 mgkg^{-1}. Accumulation of Zn was found maximum in root (32.7857 mgkg^{-1}) whereas the edible part contained the least amount of Zn which was 13.7998 mgkg^{-1}. The uptake of Fe was found highest (470.8326 mgkg^{-1}) in the root whereas the minimum Fe content was 24.7972 mgkg^{-1} determined in the edible parts of Pomelo fruit plants. In the case of Pomelo plant, maximum concentration of Ca was 79909.8581 mgkg^{-1} determined in leaves and the minimum content was 2933.9049 mgkg^{-1} measured in the edible part. Absorption of Mn was found highest in the root which was 29.0899 mgkg^{-1} and the lowest Mn content was 4.1676 mg kg^{-1} determined in the edible part. Co metal is mostly absorbed in the root (1.1049 mgkg^{-1}) and the lowest concentration of Co was 0.0129 mg kg^{-1} measured in the edible part of Pomelo plant. In comparison to the quantities of Ni and As measured in the roots which were 38.7668 mgkg^{-1} and 0.8355 mgkg^{-1} respectively, very smaller extents of the respective elements, 1.9073 and 0.0166 mgkg^{-1} were found in the edible part of Pomelo plants which suggested that mobilization and translocation of Ni and As from root to the shoot system were not substantial and the two elements mostly retained in the root after their uptake from the respective growth media. Se metalloid content was found mostly in the root 1.6330 mg kg^{-1} whereas the lowest amount was detected in the stem which was 0.3142 mgkg^{-1}. In most cases, root of Pomelo fruit plant showed higher accumulation of different heavy metals and metalloid in comparison to other parts such as stem, leaves, and edible part.

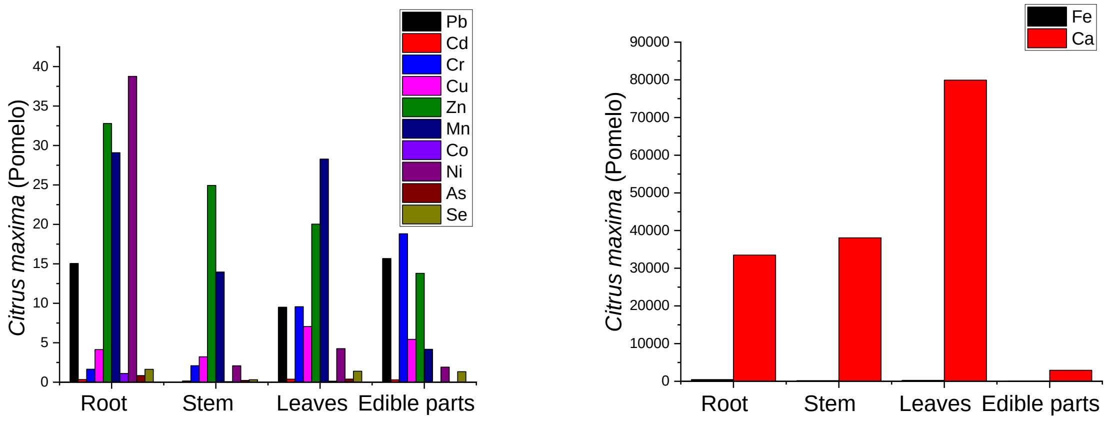


Figure 4.6. Different metals and metalloids concentrations in various parts of *Citrus maxima (Pomelo)* plant. Concentrations of Fe and Ca have been shown in separate figures.

Heavy metals and metalloids concentrations in different parts of Banana plant

The results of different metals and metalloids analysis in various parts of *Musa paradisiaca (Banana)* plant have been shown in the table 4.7 and table 4.8. Root of Banana plant accumulated highest amount of Pb which was 81.6932 mgkg^{-1} and the lowest amount was 0.9403 mg kg^{-1} measured in the edible part. Cd content was found higher in leaves (1.3146 mg kg^{-1}) and the lower quantity of Cd was measured in the edible parts (0.2447 mg kg^{-1}) of Banana plant. However, in the case of Cr, maximum accumulation was realized in the stem which was 19.7027 mgkg^{-1} whereas the lowest concentration of Cr was 1.0498 mgkg^{-1} measured in leaves of Banana plant. Uptake of Cu was observed to be highest in root (11.1880 mgkg^{-1}) and the least amount of Cu was 1.7874 mg kg^{-1} measured in the stem. However, Zn concentration was much higher in the leaves (47.2430 mg kg^{-1}) in comparison to its content determined in the edible part (14.2322 mgkg^{-1}). The leaves of Banana plant showed the maximum accumulation of Fe which was 4616.8523 mg kg^{-1} whereas the edible part displayed the lowest accumulation of Fe (36.7446 mgkg^{-1}). The highest concentration of Ca was 33267.2996 mgkg^{-1} found in the leaves and the lowest amount of Ca was 1577.9624 mg kg^{-1}. Similarly, Mn content was found much higher in the leaves (62.1946 mgkg^{-1}) whereas the lowest amount of Mn was observed in stem (1.0686 mgkg^{-1}). The maximum uptake of Co was found in the root metal (0.7119 mg kg^{-1}). However, Co concentration in the edible part was not significant and found below the detection limit. In the case of Ni, the maximum accumulation (6.9531 mg kg^{-1}) was observed in the stem of Banana and the lowest Ni concentration was 0.7404 mg kg^{-1} observed in the edible. As concentration was found maximum in leaves which was 0.9850 mgkg^{-1} and the least amount of As was detected in the edible part of Banana plant. On the other hand, Se concentration was much higher in the edible parts (1.6330 mgkg^{-1}) whereas relatively lower quantity of Se was measured in the stem which was 0.9476 mgkg^{-1}. The extents of twelve elements determined in various parts of Banana plant were compared and the results showed that accumulation of different metals and metalloids were much higher in leaves than root, stem, and edible part. Uptake of different heavy metals and metalloids in various parts of Banana plants were found to be much higher than the respective various elements concentrations observed in the Pomelo plants parts. Plants can be differed in their abilities to accumulate, mobilize, and distribute metallic and other species in different organs of their bodies.

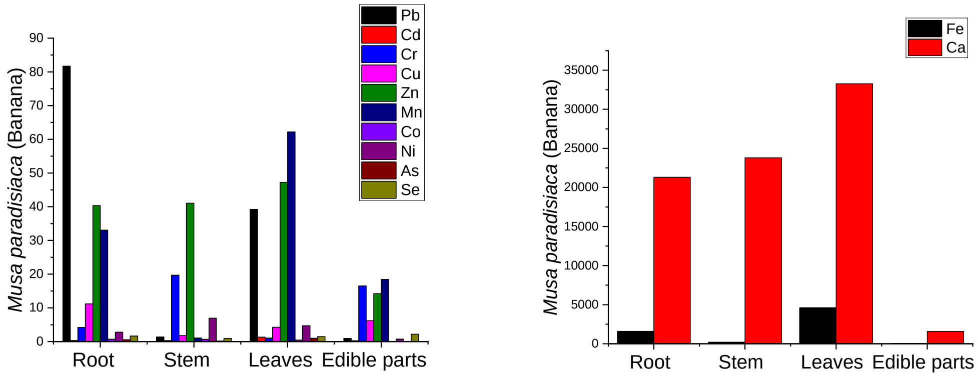


Figure 4.7. Different heavy metals and metalloids concentrations in various parts of *Musa paradisiaca (Banana)* plant. Concentrations of Fe and Ca have been shown in separate figures

4.2 Heavy metals and metalloids transfer from soil into different parts of fruits plants bodies

Generally, the food chain (soil–plant–human) pathway is being identified as one of the major pathways for human exposure to soil contamination. Soil-to-plant transfer is one of the key components of human exposure to metals through the food chain. Our data showed that HMTF values differed significantly among different plant species. The difference in HMTFs may be related to soil nutrient management and soil properties. Figure 4.4 shows the transfer of heavy metals in soils from contaminated waterbody. Evaluation of MTF is important for understanding the human exposure to HMs via the food chain [22, 26]. A heavy metal with a high transfer factor indicates the ability of that element to transfer easily from soil to the edible parts of fruits which might cause serious health risks when these contaminated fruits items are consumed by humans and animals [24, 26, 27]. The results of different metals and metalloids transfer from contaminated soil to various fruits have been shown in Fig. 4.8 and Table 4.10.

Table 4.10: Different heavy metals and metalloids transfer from soil to various parts of three fruits plants

Metals and metalloids transfer from soil into plants parts	Fruits name	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Root	Psidium guajava (Guava)	0.9644	1.1097	0.0917	0.3531	0.6154	0.0477	6.2431	0.0000	0.1910	0.2914	0.2437	0.4269
	Citrus maxima (Pomelo)	0.3188	0.9238	0.0430	0.1512	0.2185	0.0177	1.8581	0.0778	0.0669	0.6280	0.1430	0.5173
	Musa paradisiaca (Banana)	2.2278	0.9283	0.0999	0.4215	0.3569	0.0651	9.3728	1.2041	0.0624	0.0879	0.1019	0.5425
Stem	Psidium guajava (Guava)	0.0949	1.0545	0.1225	0.3641	0.2785	0.0047	24.3297	0.0000	0.1696	0.2355	0.0383	0.0940
	Citrus maxima (Pomelo)	0.0033	0.0000	0.0544	0.1177	0.1662	0.0066	2.1120	0.0374	0.0046	0.0337	0.0393	0.0995
	Musa paradisiaca (Banana)	0.0376	0.8537	0.4695	0.0673	0.3631	0.0081	10.4711	0.0389	0.0598	0.2174	0.0329	0.3110
	Psidium guajava (Guava)	0.5284	2.3716	0.1280	0.8956	0.4931	0.0157	11.9957	3.5087	0.0231	1.0391	0.1021	0.4921
	Citrus maxima (Pomelo)	0.2015	1.0661	0.2492	0.2581	0.1335	0.0100	4.4336	0.0756	0.0077	0.0690	0.0676	0.4429

Leaves	*Musa paradisiaca* (Banana)	1.0695	3.7272	0.0250	0.1603	0.4179	0.1889	14.6374	2.2651	0.0398	0.1473	0.1822	0.4869
Edible parts	Psidium guajava (Guava)	0.0939	1.0841	0.0662	0.2861	0.0418	0.0006	0.2913	0.6759	0.2129	0.0711	0.0000	0.3867
	Citrus maxima (Pomelo)	0.3320	0.8352	0.4895	0.1986	0.0920	0.0009	0.1628	0.0111	0.0008	0.0309	0.0028	0.4211
	Musa paradisiaca (Banana)	0.0256	0.6938	0.3938	0.2342	0.1259	0.0015	0.6943	0.6718	0.0000	0.0232	0.0000	0.7149

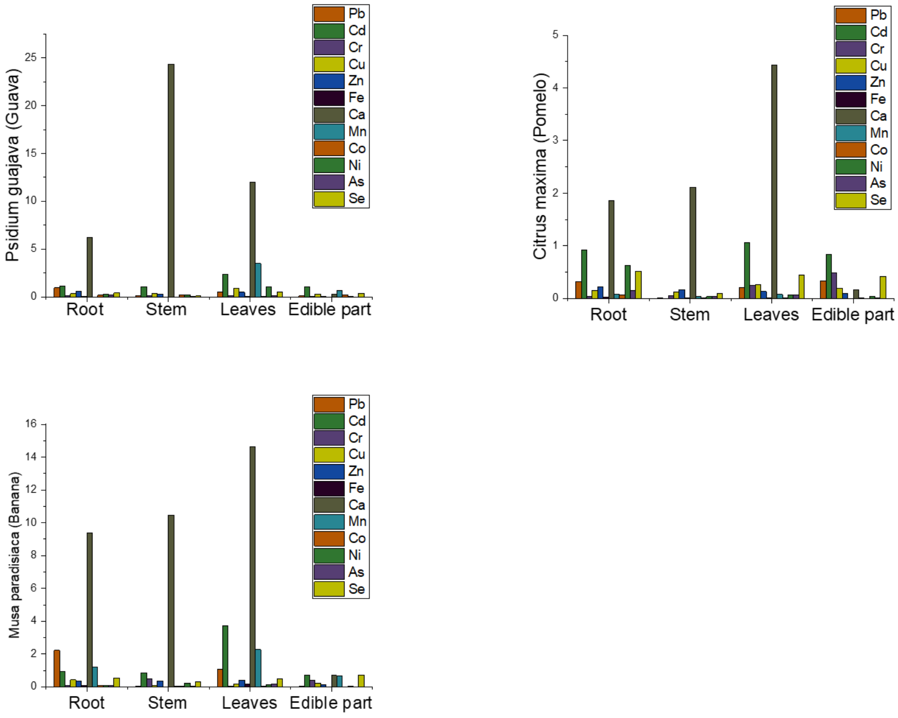


Figure 4.8. Different metals and metalloids transfer from contaminated soil to various organs of three fruits plants

The transfer factors varied significantly with the type of fruit plants as well as the natures and characteristics of metals. Among the studied fruits species, the maximum Pb transfer from soil to the root of *Musa paradisiaca* (Banana) plant was 2.2278 and the minimum Pb transfer from soil was 0.0033 observed in the stem of *Citrus maxima* (Pomelo) plant. It was found that the maximum amount of Cd transfer from soil (3.7272) was occurred to the leaves of *Musa paradisiaca* (Banana) whereas the transfer of Cr was higher (0.4895) in the edible part *Citrus maxima* (Pomelo) plants. However, the leaves of Musa *paradisiaca* showed the lower transfer of Cr which was 0.0250. Maximum transfer of Cu (0.8956) was found in the leaves of *Psidium guajava* (Guava) and the lowest transfer factor of Cu was 0.0673 determined in the stem of *Musa paradisiaca* (Banana) plant. Transfer of Zn was highest (0.6154) in the root of *Psidium guajava* whereas the minimum transfer factor for Zn was 0.0418 observed with the edible part of *Psidium guajava* (Guava) plant. The maximum transfer factor for Fe was 0.1889 measured in the leaves of *Musa paradisiaca* (Banana) plant whereas the lowest transfer of Fe was realized in the edible parts of *Psidium guajava* (Guava) which was 0.0006. Relatively higher Ca transfer was occurred from soil to the stem of *Psidium guajava* (Guava) plant as indicated by the transfer factor of 24.3297. Comparatively lower transfer of Ca (0.1628) was found in the edible parts of *Citrus maxima* (Pomelo) plant. Translocation of Mn from soil was found maximum in the leaves of *Psidium guajava* (Guava) plant (3.5087) whereas the transfer factor for Co was observed to be highest (0.2129) in the edible part of *Psidium guajava (Guava)* plant. Surprisingly, mobilization of Mn from soil to the root and stem of *Psidium guajava (Guava)* was highly insignificant as the respective metal transfer factors were found zero which indicated that most of the Mn contents absorbed from contaminated soil by root system of Guava plant mobilized and translocated into the edible part. The maximum amount of Ni (TF1.0391) was transferred from soil to the leaves of *Psidium guajava* (Guava) plant whereas the lowest transfer of Ni was found in the edible parts of *Musa paradisiaca* (Banana) plant as suggested from the magnitude of TF (0.0232). The extent of TF for As was observed to be the highest in root of *Psidium guajava* (Guava) plant which was 0.2437. Translocation of As from soil and root into the edible parts of Psidium *guajava* (Guava) and *Musa paradisiaca* (Banana) plants were insignificant as revealed from the estimation of corresponding TF which were observed to be zero in both cases. Se was transferred significantly from soil to the edible part of *Musa paradisiaca* (Banana) plant which was confirmed by the estimation of the highest TF value of 0.7149 mg kg^{-1} in the respective plant part whereas the lowest transfer of Se was realized with the TF value of 0.0940 measured in the stem of *Psidium guajava* (Guava) plant.

Heavy metals transfer factors from soil into edible parts of three fruit plants were highly varied and they followed the order of: Cd > Se > Mn > Ca > Cr > Cu > Pb > Zn > Co > Ni > Fe > As, whereas the transfer factors of heavy metals and metalloids in the leaves of three fruit plants followed the order of : Ca > Cd > Mn > Pb > Se > Cu > Ni > Zn > Cr > As > Fe > Co. Similarly, the order of transfer factors of different heavy metals and metalloid in the stems of three fruit plants was: Ca > Cd > Zn > Cr > Cu > Se > Ni > Co > Pb > As > Mn > Fe as well as the magnitudes of transfer factors for different metals and metalloids in the roots of three fruit plants showed the order of: Ca > Pb > Cd > Se > Mn > Zn > Ni > Cu > As > Co > Cr > Fe. The results of transfer factors analyses on heavy metals and metalloids translocation from soil to various parts of three fruits plants bodies indicated that there was higher mobility of Cd from contaminated soil to edible part of the fruit plants and relatively stronger mobility of Ca from soil to roots, stems, and leaves of the respective fruit plants bodies. The mean TF values showed that Ca, Cd, and Mn accumulations in the leaves were greater than those observed in edible parts, roots, and stems of three fruits plants bodies. These differences in TFs might be due to variations in the soil physicochemical characteristics, water sources used for irrigating fruits, types of plant species, or amount and types of HMs [23-25, 28-31].

4.3 Food Pollution Index

The food pollution index (FPI) and the collective food pollution index (CFPI) data show contamination by the selected elements in our study fruits. We can see from table 4.11 that for Pb, Cd, Cr, Zn and Mn the FPI data is greater than 1 and we know that an FPI or CFPI >1 indicates serious pollution. The trend of FPI were follow in the order Zn > Cd > Cr > Pb > Mn > Cu > Ni > Fe > As. The FPI values range are 0.4702 - 7.8356 for Pb, 12.2350 - 16.1050 for Cd, 2.2721 - 14.4656 for Cr, 0.5430 - 0.8095 for Cu, 9.1840 - 23.7203 for Zn, 0.0353 - 0.0817 for Fe, 0.8335 - 4.0181 for Mn, 0.07404 - 0.19073 for Ni and 0 - 0.0166 for As. We see that the CFPI values also greater than 1 that means serious food pollution occurs in the study area.

Table 4.11. Food pollution index of different studied metal and metalloid

FPI	Pb	Cd	Cr	Cu	Zn	Fe	Mn	Ni	As	CFPI
Psidium guajava (Guava)	1.9984	16.1050	2.2721	0.8095	9.1840	0.0353	4.0181	0.18248	0	34.6048
Citrus maxima (Pomelo)	7.8356	15.0250	14.4656	0.5430	22.9997	0.0551	0.8335	0.19073	0.0166	61.9647
Musa paradisiaca (Banana)	0.4702	12.2350	12.7105	0.6217	23.7203	0.0817	3.6892	0.07404	0	53.6026

4.4 Dietary intake of heavy metals and metalloids and target hazard quotients

The results of the estimated dietary intakes (EDI) of heavy metals and metalloids for children and adult via the consumption of fruits have been presented in Table 4.12 and Table 4.13. The daily intake of these heavy metals and metalloids depends on the concentrations of the respective elements as well as the amounts of fruits items consumed.

Table 4.12. Estimated daily intake of different metals and metalloids by Children from the consumption of contaminated fruits

EDI	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Psidium guajava (Guava)	0.0011	0.0004	0.0039	0.0108	0.0073	0.0212	0.6780	0.0268	0.0029	0.0024	0	0.0016

Citrus maxima (Pomelo)	0.0042	0.0004	0.0251	0.0072	0.0184	0.0331	3.9119	0.0056	0.0000	0.0025	0.0000	0.0018
Musa paradisiaca (Banana)	0.0013	0.0003	0.0220	0.0083	0.0190	0.0490	2.1039	0.0246	0.0000	0.0010	0	0.0029
Mean	0.0022	0.0004	0.0170	0.0088	0.0149	0.0344	2.2313	0.0190	0.0010	0.0020	0.0000	0.0021
Minimum	0.0011	0.0003	0.0039	0.0072	0.0073	0.0212	0.6780	0.0056	0.0000	0.0010	0.0000	0.0016
Maximum	0.0042	0.0004	0.0251	0.0108	0.0190	0.0490	3.9119	0.0268	0.0029	0.0025	0.0000	0.0029

Table 4.13. Estimated daily intake of different metals and metalloids by Adults from the consumption of contaminated fruits

EDI	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Psidium guajava (Guava)	0.0021	0.0002	0.0015	0.0042	0.0029	0.0083	0.2664	0.0105	0.0011	0.0010	0	0.0006
Citrus maxima (Pomelo)	0.0082	0.0002	0.0099	0.0028	0.0072	0.0130	1.5368	0.0022	0.0000	0.0010	0	0.0007
Musa paradisiaca (Banana)	0.0005	0.0001	0.0087	0.0033	0.0075	0.0192	0.8266	0.0097	0.0000	0.0004	0	0.0011
Mean	0.0036	0.0002	0.0067	0.0034	0.0059	0.0135	0.8766	0.0075	0.0004	0.0008	0.0000	0.0008

Minimum	0.0005	0.0001	0.0015	0.0028	0.0029	0.0083	0.2664	0.0022	0.0000	0.0004	0.0000	0.0006
Maximum	0.0082	0.0002	0.0099	0.0042	0.0075	0.0192	1.5368	0.0105	0.0011	0.0010	0.0000	0.0011

The EDIs of different heavy metals for children from the consumption of contaminated fruits followed the order of Ca > Fe > Mn > Cr > Zn > Cu > Pb > Se > Ni > Co > Cd > As. The highest EDIs of different metals and metalloids for children were found as: Pb (0.0042 Kg d^{-1}), Cd (0.0004 Kg d^{-1}), Cr (0.0251 Kg d^{-1}), Cu (0.0108 Kg d^{-1}), Zn (0.0190 Kg d^{-1}), Fe (0.0490 Kg d^{-1}), Ca (3.9119 Kg d^{-1}), Mn (0.0268 Kg d^{-1}), Co (0.0029 Kg d^{-1}), Ni (0.0025 Kg d^{-1}), As (0.0000 Kg d^{-1}) and Se (0.0029 Kg d^{-1}) (Table 4.12). The maximum EDI values associated with adult for different metals and metalloids such as Pb, Cd, Cr, Cu, Zn, Fe, Ca, Mn, Co, Ni, As and Se through the consumption of various contaminated fruit items were: 0.0082, 0.0002, 0.0099, 0.0042, 0.0075, 0.0192, 1.5368, 0.0105, 0.0011, 0.0010, 0.0000 and 0.0011 Kgd^{-1} respectively (Table 4.13). The trend of EDI for adults with the exposures of different elements through the contaminated fruits was found as: Ca > Fe > Mn > Zn > Cr > Pb > Cu > Ni > Se > Co > Cd > As.

The target hazard quotient (THQ) of individual heavy metal and metalloid through the consumption of different fruit species were estimated for both adults and children in the study area and the results have been presented in Table 4.13 and Table 4.14. The target hazard quotients (THQs) were calculated based on the daily dietary habits of residents who are highly exposed to contaminated different fruits species grown in the study area. Although heavy metals contamination could occur due to the ingestion, dermal contact, and inhalation. The THQ has been recognized as a useful parameter for evaluation of risks associated with the consumption of metal contaminated foods such as crops, vegetables, and fruits [32, 33]. However, Horiguchi et al. (2004) [34] suggested that the ingested dose of heavy metals may not be equal to the absorbed pollutant dose in reality and a fraction of the ingested heavy metals may be excreted, with the remainder accumulated in body tissues where they affect human health.

The THQ values of different elements associated with health risks of children from the intake of contaminated fruits grown in the study area were followed the order of: Cr > Pb > Mn > Cd > Cu > Ni > Zn > Fe > Co > As. All THQ values were lower than 1, except for Cr in all three fruits, Pb in *Psidium guajava (Guava)* and *Citrus maxima* (Pomelo) and Mn in *Psidium guajava (Guava)* and Musa paradisiaca (Banana) respectively (Fig. 4.9).

For Adults, the target hazard quotients (THQs) of different metals through consumption of all three fruits were decreased in the order of Cr > Pb > Mn > Cd > Cu > Ni > Zn > Fe > Co > As. The metals THQ through consumption fruits are given in Fig. 4.5. The magnitudes of THQs for adult associated with the exposure of polluted fruit items were varied from 0.1231- 2.0522 for Pb, 0.1282- 0.1687 for Cd, 0.5157- 3.2835 for Cr, 0.0711- 0.1060 for Cu, 0.0096- 0.0248 for Zn, 0.0119- 0.0275 for Fe, 0.1559- 0.7517 for Mn, 0.0000- 0.0376 for Co, 0.0194- 0.0500 for Ni and 0.0000- 0.0290 for As (Fig. 4.5). Chromium (Cr) exhibited relatively higher THQ in all fruits compared to those observed with all other metals. The maximum magnitude of THQ for Cr was

3.2835 associated with the consumption of *Citrus maxima* (Pomelo). The THQ value for Pb in the *Citrus maxima* (Pomelo) were more than 1. These THQs data indicates that *Citrus maxima* (Pomelo) and *Musa paradisiaca* (Banana) fruits pose a higher potential health risk than *Psidium guajava* (Guava). For both adult and children, the TTHQ values for all elements in the three fruits species were above 1 (Fig. 4.9), indicating the adverse effects of metal toxicities on human health from the excessive consumption of contaminated fruits species.

Table 4.14: Total Hazard Quotients (THQ) of different elements for Children associated with the consumption of polluted fruits items

THQ for Children	Pb	Cd	Cr	Cu	Zn	Fe	Mn	Co	Ni	As	TTHQ[a]
Psidium guajava (Guava)	1.3322	0.4295	1.3128	0.2698	0.0245	0.0303	1.9134	0.0956	0.1217	0.0000	5.5297
Citrus maxima (Pomelo)	5.2237	0.4007	8.3579	0.1810	0.0613	0.0472	0.3969	0.0006	0.1272	0.0738	14.8702
Musa paradisiaca (Banana)	0.3134	0.3263	7.3438	0.2072	0.0633	0.0700	1.7568	0.0000	0.0494	0.0000	10.1301
TDHQ[b]	6.8694	1.1564	17.0145	0.6580	0.1491	0.1475	4.0671	0.0962	0.2982	0.0738	30.5300 (HI)[c]

[a] The total metals THQs (TTHQ sum of individual metal THQs).

[b] TDHQ: the total diet THQ of each metal (total diet i.e., the sum of Psidium guajava (Guava), Citrus maxima (Pomelo), Musa paradisiaca (Banana)).

[c] HI: The sum of THQs values of heavy metals due to consuming the diet.

Table 4.15. Total Hazard Quotients (THQ) of different elements for Adult associated with the consumption of contaminated fruits species

THQ for Adult	Pb	Cd	Cr	Cu	Zn	Fe	Mn	Co	Ni	As	TTHQ[a]
Psidium guajava (Guava)	0.5234	0.1687	0.5157	0.1060	0.0096	0.0119	0.7517	0.0376	0.0478	0.0000	2.1724

Citrus maxima (Pomelo)	2.0522	0.1574	3.2835	0.0711	0.0241	0.0186	0.1559	0.0002	0.0500	0.0290	5.8419
Musa paradisiaca (Banana)	0.1231	0.1282	2.8851	0.0814	0.0248	0.0275	0.6902	0.0000	0.0194	0.0000	3.9797
TDHQ[b]	2.6987	0.4543	6.6843	0.2585	0.0586	0.0579	1.5978	0.0378	0.1171	0.0290	11.9939 (HI)[c]

[a] The total metals THQs (TTHQ sum of individual metal THQs).

[b] TDHQ: the total diet THQ of each metal (total diet i.e., the sum of Psidium guajava (Guava), Citrus maxima (Pomelo), Musa paradisiaca (Banana)).

[c] HI: The sum of THQs values of heavy metals due to consuming the diet.

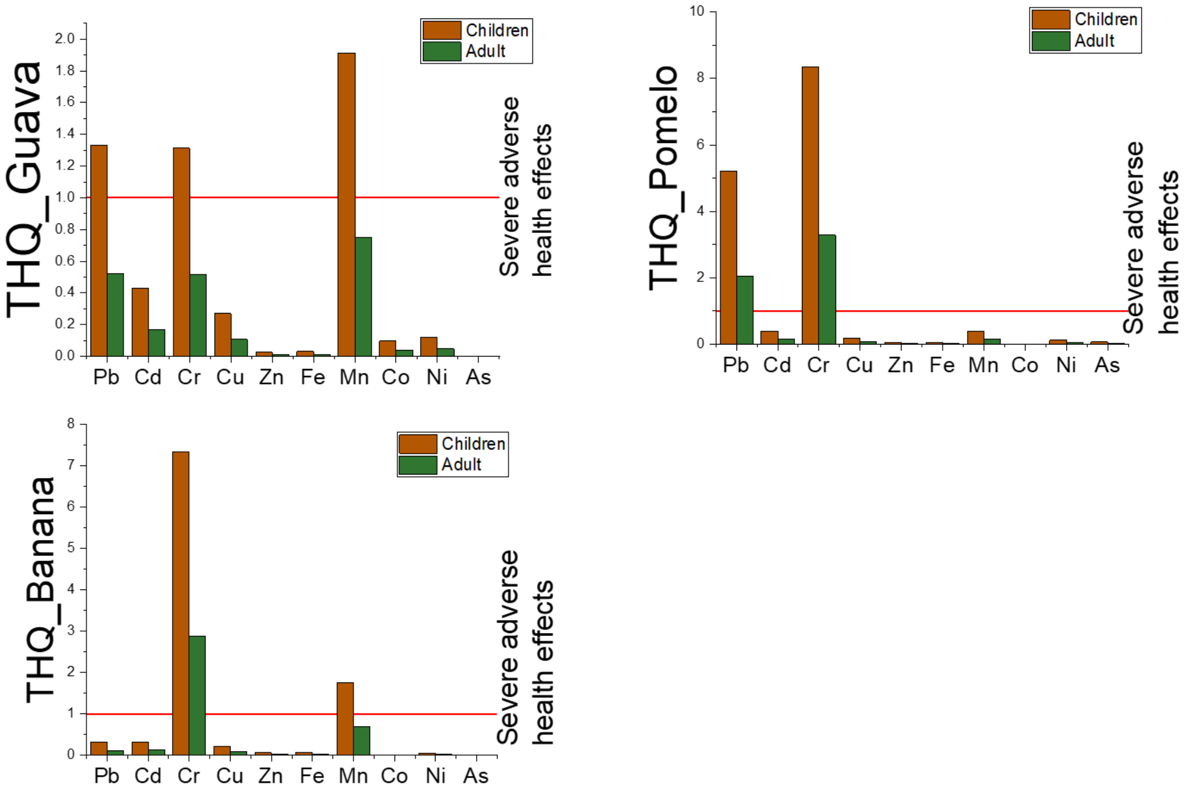


Figure 4.9. Target Hazard Quotient (THQ) for both Children and Adults from the consumption of contaminated fruits species

4.5 Cancer risks for Adults and Children from the consumption of metals contaminated different fruits species

Pb, Cd, Cr, Ni and As are classified as carcinogenic agents by the guidelines of International Agency for Research on Cancer (IARC) [35]. Chronic exposure to low doses of Pb, Cd, Cr, Ni and As could therefore result into many types of cancers [36]. Cancer risks (CR) associated with Pb, Cd, Cr, Ni and As for both Children and Adults from the consumption of contaminated fruits were calculated and the results have been presented in Table 4.16.

Table 4.16: Carcinogenic Risks (CR) data for adults and children from the consumption of metal and metalloid contaminated different fruits items

Carcinogenic Risks (CR)	Fruits	Pb	Cd	Cr	Ni	As	Cumulative cancer risk (CCR)
Children	Psidium guajava (Guava)	0.0000	0.0002	0.0020	0.0020	0.0000	0.0042
	Citrus maxima (Pomelo)	0.0000	0.0002	0.0125	0.0021	0.0000	0.0149
	Musa paradisiaca (Banana)	0.0000	0.0001	0.0110	0.0008	0.0000	0.0120
Adult	Psidium guajava (Guava)	0.0000	0.0001	0.0008	0.0008	0.0000	0.0017
	Citrus maxima (Pomelo)	0.0001	0.0001	0.0049	0.0008	0.0000	0.0059
	Musa paradisiaca (Banana)	0.0000	0.0000	0.0043	0.0003	0.0000	0.0047

US-EPA recommended the safe limit for cancer risk which is below about 1 chance in 1,000,000 lifetime exposure ($CR < 10^{-6}$) and threshold risk limit ($CR > 10^{-4}$) for the chance of cancer is above 1 in 10,000 exposure where the remedial measures are considerable and moderate risk level ($CR > 10^{-3}$) is above 1 in 1,000 where public health safety consideration is more important [37, 38].

The magnitudes of CR for Children associated with Cd, Cr and Ni exposures violated the threshold risk limit ($>10^{-4}$) in all three fruits studied in the present investigation. The trend of risk for developing cancer in children as a result of the consumption of three different types of fruits was found as: Citrus maxima (Pomelo) > Musa paradisiaca (Banana) > Psidium guajava (Guava).

For adults, the extents of CR for Cr and Ni were exceeded the threshold risk limit (>10^{-4}) in all fruit's species examined in the present study. The trend of risk for developing cancer in adults as a result of the consumption of polluted fruits items was followed as: Citrus maxima (Pomelo) > Musa paradisiaca (Banana) > Psidium guajava (Guava).

Moreover, cumulative cancer risks (CCR) associated with the intake of all three fruits species grown in the contaminated study area were exceeded the recommended threshold risk limit (>10^{-4}) (Figure 4.10). Furthermore, among all the fruits studied, *Citrus maxima* (Pomelo) has displayed the highest chances of cancer risks for both children (CCR 0.0149) and adult (CCR 0.0059) and the lowest chances of cancer risks for both children (CCR 0.0042) and adult (CCR 0.0017) was observed with the consumption of *Psidium guajava* (Guava). Therefore, consumption of *Citrus maxima* (Pomelo) grown the polluted study area is most risky which highly susceptible to cancer risk for both adult and children. Necessary actions should be needed to control the excess use of heavy metal-based fertilizers and pesticides and the discharge of toxic wastewater mixed with heavy metals from different industries as well as automobiles emissions which may save crops, vegetables, fruits, and other food plants from heavy metals and metalloids pollution and thus could save the urban population from cancer risk associated with the intake of contaminated different food species.

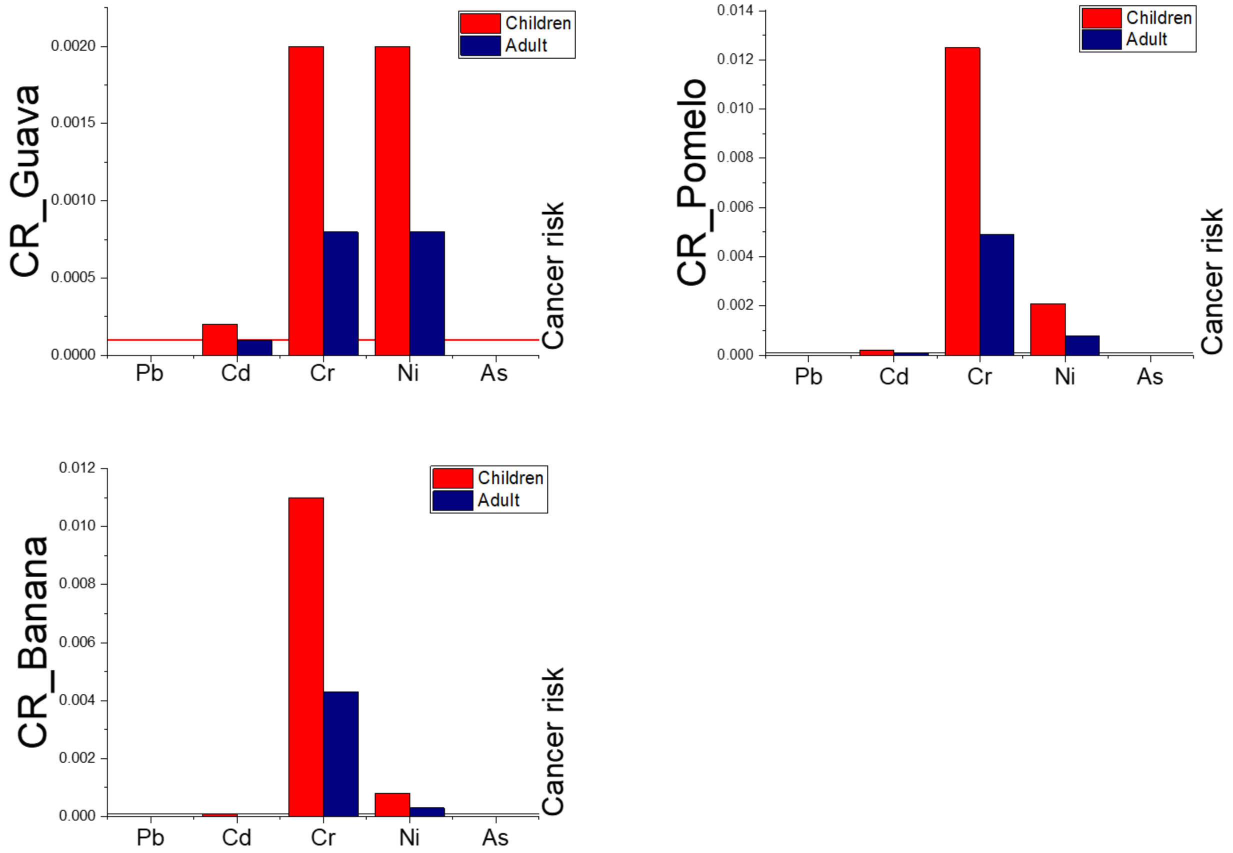


Figure 4.10. Carcinogenic Risks (CR) analyses different heavy metals for children and adults from the consumption of contaminated fruits species

4.6 Soil threshold values and food safety

Evaluation of food chain for any pollution can be considered as very significant tool to assess the risks of transferring heavy and toxic metals to humans. Generally, heavy and toxic metals and metalloids contaminations occur in food plants from their growth media such as waterbodies and soil. Literature studies have shown that setting up soil threshold values (STV) based on the types of plants and their ability to take up different elements from soil has become increasingly important to examine health risks associated with the consumption contaminated different food items including crops, vegetables, fruits etc. [39, 40].

Soil threshold values (STV) could be calculated using the method of food pollution index (FPI) based on each plant's ability to take up elements from soil as well as by applying the degrees THQ for the elements studied [41, 42]. In the present research, soil threshold values (STV) have been estimated based on the target hazard quotients (THQs) as well as food pollution index (FPI) obtained for all heavy metals and metalloids in three different fruits plants [41, 42]. The STV of Cu, Fe, Ni and As based on THQs were found to be lower than the MPL, which might be due to the higher transfer of Cu, Fe, Ni and As from soil to the edible part of fruits plants (Table 4.17). However, the magnitudes of STV for Pb, Cd, Cr, Zn and Mn based on THQs were observed to be higher than the MPL except Pb in *Musa paradisiaca* (Banana) and Mn in *Citrus maxima* (Pomelo), which might be due to the lower transfer of Pb, Cd, Cr, Zn and Mn from soil to the edible part of the respective fruit's plants (Table 4.17).

Table 4.17 Soil threshold values (STV) of different heavy metals and metalloids

STV	Soil from fruits plants fields	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
STV1[a]	*Psidium guajava* (Guava)	42.5431	0.2971	44.6425	28.2927	131.7092	26783.5215	1745.2961	29.7257	10.1000	25.6585	5.0347	3.1917
	Citrus maxima (Pomelo)	47.2005	0.3598	38.4194	27.3431	150.0609	26531.0381	18023.8219	373.848	16.5189	61.7284	5.8412	3.1568
	Musa paradisiaca (Banana)	36.6696	0.3527	41.9643	26.5446	113.0357	24439.2857	2272.7679	27.4579	11.4080	31.9801	5.4054	3.0469
STV2[b]	*Psidium guajava* (Guava)	21.2891	0.0184	19.6483	34.9512	14.3411	758730.433	–	7.3979	–	140.6099	∞	–

	Citrus maxima (Pomelo)	6.0238	0.0239	2.6559	50.3593	6.5244	481464.3244	–	448.5171	–	323.6428	351.8795	–
	Musa paradisiaca (Banana)	77.9955	0.0288	3.3015	42.6974	4.7653	299300.5384	–	7.4426	–	431.9300	∞	–

[a] Based-on target hazard quotient (THQ)

[b] Based on food pollution index (FPI)

4.7 Geoaccumulation Index (Igeo) of different heavy metals and metalloids in soil

The index of geoaccumulation (Igeo) for each heavy metal and metalloid was assessed based on the guidelines and procedures proposed by Müller (1969). The extents of Igeo of different heavy metals and metalloids in soil from three fruits plant fields were varied significantly and followed the order of: Pb > As > Cd > Zn > Ni > Fe > Co > Cu > Cr > Mn. The highest geoaccumulation index for Pb, Cd, Cr, Cu, Zn, Fe, Ca, Mn, Co, Ni, As and Se in soil of three different fruits plant fields were 0.7286 in Pomelo, 0.3610 in Pomelo, 0.0896 in Guava, 0.1032 in Guava, 0.4302 in Pomelo, 0.1075 in Guava, 0.1295 in Guava, 0.0790 in Pomelo, 0.1326 in Pomelo, 0.1652 in Pomelo, 0.6512 in Pomelo and 0.0291 in Guava (Table 4.17). According to the Muller classification (Muller, 1981) and the nature of the Igeo calculation, which involves the logarithmic function and a background multiplication factor of 1.5, the estimated Igeo values for different heavy metals and metalloids indicated uncontaminated to moderate pollution in soil of three fruits plants fields examined in the present study (Table 4.18 and Figure 4.11).

Table 4.18 The magnitudes of Geoaccumulation Index (Igeo) for different heavy metals and metalloids in soil from three different fruits plants fields

Soil from fruits plants fields	Pb	Cd	Cr	Cu	Zn	Fe	Ca	Mn	Co	Ni	As	Se
Psidium guajava (Guava)	0.6567	0.2981	0.0896	0.1032	0.3776	0.1075	0.1295	0.0063	0.0811	0.0687	0.5613	0.0291
Citrus maxima (Pomelo)	0.7286	0.3610	0.0771	0.0998	0.4302	0.1065	0.1283	0.0790	0.1326	0.1652	0.6512	0.0288
Musa paradisiaca (Banana)	0.5660	0.3539	0.0842	0.0968	0.3240	0.0981	0.1182	0.0058	0.0916	0.0856	0.6026	0.0278

Mean	0.6504	0.3377	0.0836	0.0999	0.3773	0.1040	0.1253	0.0303	0.1017	0.1065	0.6050	0.0286
Minimum	0.5660	0.2981	0.0771	0.0968	0.3240	0.0981	0.1182	0.0058	0.0811	0.0687	0.5613	0.0278
Maximum	0.7286	0.3610	0.0896	0.1032	0.4302	0.1075	0.1295	0.0790	0.1326	0.1652	0.6512	0.0291

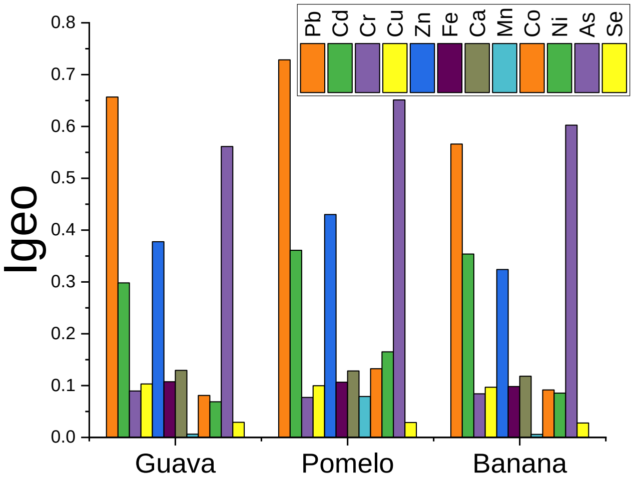


Figure 4.11: Geoaccumulation indexes of (Igeo) for different heavy metals and metalloids in soil from three fruits plants fields

4.8 Enrichment Factor (EF)

The enrichment factor (EF) is a convenient way of determining the geochemical trends and metal pollution in arable soil. [43]. The value of unity for EF denotes either metal enrichment in soil nor the depletion relative to the Earth's crust.

Table 4.19: Enrichment Factors (EF) of different heavy metals and metalloids in soil from three fruits plants fields

Soil from different fruits plants fields	Pb	Cd	Cr	Cu	Zn	Ca	Mn	Co	Ni	As	Se
Psidium guajava (Guava)	6.1093	2.7732	0.8334	0.9603	3.5125	0.0785	0.0584	0.7542	0.6387	5.2216	0.2708
Citrus maxima (Pomelo)	6.8426	3.3904	0.7240	0.9369	4.0400	0.8185	0.7416	1.2453	1.5511	6.1157	0.2704
Musa paradisiaca (Banana)	5.7709	3.6079	0.8585	0.9874	3.3037	0.1120	0.0591	0.9336	0.8724	6.1438	0.2833

The enrichment factors (EF) of twelve elements have been determined in soil of three fruits plants fields and the results are listed in Table 4.19. According to Taylor (1964) classification, the EF values of Cr, Cu, Ca, Mn, Co (except in soil from Pomelo plant field), Ni (except in Pomelo plant field) and Se were below one (EF< 1), which indicated that there were no significant enrichments by these metals in soil of different fruits plants fields in the respective study area.

The magnitudes of EF values for Pb and As in all soil from three fruits plants fields were higher than five (EF: 5 < 10) which suggested that soil from the respective fruits plants field was severely enriched with Pb and As. The higher Pb and As contents indicated that soil system in the study area were severely contaminated with the excessive presence of the respective heavy metals which might have occurred due to the anthropogenic activities in and around the study area. The EF values for Cd and Zn in most of the soil samples were between 3 and 5 (EF: 3 < 5) which confirmed moderate enrichment of these metals in soil of three fruits plants fields. However, exception was observed for Cd in soil of Guava plant field. The highest enrichment factor for Pb, Cd, Cr, Cu, Zn, Ca, Mn, Co, Ni, As and Se in soil from different fruits plants fields in the study area were 6.8426 in Pomelo, 3.6079 in Banana, 0.8585 in Banana, 0.9874 in Banana, 4.0400 in Pomelo, 0.8185 in Pomelo, 0.7416 in Pomelo, 1.2453 in Pomelo, 1.5511 in Pomelo, 6.1438 in Guava and 0.2833 in Banana plant field respectively (Table 4.19).

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CHAPTER-5

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusion

Contamination of various foodstuffs such as crops, vegetables, and fruits by different heavy and toxic metals due to the industrial and other activities has become a severe environmental pollution problem now a days in most of the developing countries like Bangladesh. The present study has investigated the accumulations, distributions, and mobilization of twelve potential heavy metals and metalloids (Pb, Cd, Cr, Cu, Fe, Zn, Ca, Co, Ni, Mn, As, and Se) in different organs of various fruits plants grown in and around Dhaka Export Processing Zone (DEPZ) in Savar, Dhaka. The study has also examined the concentrations of the respective twelve elements in plants growth media such as soil and water. In addition, the study has evaluated the potential health risks associated with the consumption of metals and metalloids contaminated fruits items grown in the respective polluted areas. Comparison of twelve different elements concentrations found in fruits, soil, and water bodies with the permissible levels of WHO, USEPA, China, and India showed that the environmental samples including fruits and soil in the respective area have been polluted with excessive quantities of heavy and toxic metals. Concentrations of Pb, Cd, Cr, Zn and Mn in fruits, Fe and Cr in water as well as Pb, Zn, Fe and Mn in soil have been found significantly higher than the permissible levels of WHO, USEPA, India and China. Among twelve significant elements including heavy metals and metalloids studied in the present investigation, the highest concentration was found for Ca measured in the leaves of Citrus maxima (Pomelo) which was 79909.8581 mg/kg. However, the extents of different metals and metalloid contents in the environmental samples from three different sources was observed to be in the order of soils > fruits > waterbodies. Accumulation and distribution of metals and metalloids from soils into different fruits species were found to be much higher irrespective of the elements studied. Among different metals and metalloids contents measured in various fruits samples, Ca showed the highest concentration which was 2933.9049 mg/kg found in *Citrus maxima* (Pomelo) whereas the lowest concentration was observed for As determined in *Psidium guajava* (Guava) as well as for As and Co measured in Musa *paradisiaca* (Banana) where their quantities were found below the detection limit. The average metals and metalloids concentrations in various fruits species have been found to be in the order of: Ca > Fe > Mn > Cu > Zn > Pb > Cr > Ni > Se > Co > Cd > As. Different heavy metals and metalloids concentrations determined in the various parts of three fruits plants were observed to be varied significantly. The larger quantities of heavy metals and metalloids were accumulated in the leaves in comparison to roots, stems, and edible parts which might have occurred through various mechanisms, including air pollution, soil contamination, and direct deposition of metal-containing particles onto their surfaces. The geoaccumulation index (Igeo) analysis data for different heavy metals and metalloids indicated uncontaminated to moderate pollution in soil of three fruits plants fields. The maximum Igeo for twelve elements including Pb, Cd, Cr, Cu, Zn, Fe, Ca, Mn, Co, Ni, As and Se in soil of three different fruits plant fields were 0.7286 in Pomelo, 0.3610 in Pomelo, 0.0896 in Guava, 0.1032 in Guava, 0.4302 in Pomelo, 0.1075 in Guava, 0.1295 in Guava, 0.0790 in Pomelo, 0.1326 in Pomelo, 0.1652 in Pomelo, 0.6512 in Pomelo and 0.0291 in Guava respectively. Heavy metals and metalloids enrichment factor study showed the magnitudes of EF for Cr, Cu, Ca, Mn, and Se below one (EF< 1), which indicated that there were no significant enrichments by these metals in soil of different fruits plants fields. However, the extents of EF for Pb and As in all soil from three fruits plants fields were higher than five (EF: 5 < 10) suggesting that soil from the respective fruits plants fields are severely polluted with Pb and As. Again, the EF values for Cd and Zn in most of the soil samples were between 2 and 5 (EF: 3 < 5) indicating the moderate enrichment of these metals in soil of three fruits plants fields.

Among all fruits studied during the present study, the highest estimated daily intake (EDI) was 3.9119 found for Ca in *Citrus maxima* (Pomelo). Transfer factors (TF) and target hazard quotients (THQ) were analyzed to assess the potential health risks associated with the consumption of metals contaminated different fruits species. Assessments of THQ and TTHQ for twelve different metals and metalloids in the three fruits provided significant information on hazard index for human health. Both adults and children have been at considerable noncarcinogenic (THQ and TTHQ > 1) risks due to the continuous consumption of metals contaminated different fruits species. However, considering the multiple metals contaminations in the study areas, the THQs of all elements for the three different fruits species were found to be increased in the following order: *Citrus maxima* (Pomelo) > *Musa paradisiaca* (Banana) > *Psidium guajava* (Guava). The highest THQ was 3.2835 found for Cr in *Citrus maxima* (Pomelo) and the lowest THQ was found for As in *Psidium guajava* (Guava). The maximum TTHQ was 5.8419 observed in the *Citrus maxima* (Pomelo). Carcinogenic risks analysis for different metals in various fruits species showed the data which were above the safety range (CR > 1.00×10^{-6}) for Cd, Cr, and Ni and thus imposed a substantial threat of developing cancer in children and adult. Among all the fruits studied, *Citrus maxima* (Pomelo) showed the higher cancer risks for both children (CR 0.0149) and adult (CR 0.0059) because of the excessive accumulations of heavy metals such as Cd, Cr, and Ni whereas *Psidium guajava* (Guava) displayed the lowest cancer risks for both children (CR 0.0042) and adult (CR 0.0017).

5.2 Future recommendations

The following necessary measures are highly recommended to mitigate and control the heavy metals and metalloids contamination in fruits and other food plants grown in different areas of Bangladesh:

1. Government should provide available opportunities to farmers to tests arable soils free of costs to ensure soil qualities and characteristics prior to the cultivation of any agricultural food stuffs.

2. The fruits plants should not be grown in metals contaminated soil and waterbodies. The lands and waterbodies in the vicinities of industrial areas should not used for cultivation of fruits, crops, and vegetables plants. Sometimes it may not be possible for small farmers due to the limited land resources in our country. In that case, farmers can grow certain types of fruits plants at such sites which show least accumulation of metals in their edible parts in comparison to other plant organs such as leaf, stem and root.

3. Effluents from various industries should be regularly monitored for heavy metals and other elements concentrations prior to discharge into nearby waterbodies and lands.

4. People awareness about the health risks due to the consumption of metal contaminated fruits should be increased.

5. Continuous monitoring of industrial activities should be performed by the Department of Environment (DoE) to minimize the discharge of untreated industrial effluents into fresh waterbodies and arable lands.

6. Government of Bangladesh should take proper actions for making new national and regional policies and appropriate preventive measures based on the metals assessment data prior to further deterioration of water quality, soil quality, and food chain contamination in the respective region.

7. Efficient methodologies for removing of toxic heavy metals and other elements from sewage wastes and industrial effluents are urgently needed before releasing the toxic effluents from various industries into srrounding environment to avoid and mitigate environmental pollution.

8. Finally, all the necessary steps should be taken to minimize the heavy and toxic metals pollution in different environmental components within the study areas.