



Determination of Selected Heavy Metals in Food and Water Consumed by Households

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Abstract

Environmental pollution is a worldwide problem and heavy metals belong to the most important of all pollutants. Nowadays it became an alarming issue. The main threats to human health from heavy metals are associated with exposure to mercury, cadmium, lead, copper, and arsenic. These metals have extensive effects on human health which has reviewed by international bodies such as the WHO. Exposure to heavy metals is increasing in some parts of the world, specifically in less developed countries. In the present study, the assessment of selected heavy metals such as mercury, cadmium, lead and copper will be carried out in water and food consumed by households. Detection and estimation of heavy metals will be performed by using Atomic Absorption Spectrophotometer. Here 96% of nitric acid has used to remove organic material by decomposing them into carbon dioxide and make the metals soluble. This research will investigate the exposure to heavy metals in the average diet as well as the level of contaminations in water and food in Bangladesh. This will provide information on the dietary study for taking necessary actions against potential sources of contamination. Research findings will provide the necessary evidence to mobilize support for implementing national policies that commit the Government to reduce heavy metal contamination of food and water.

Keywords: heavy metal; mercury; cadmium; lead; copper and arsenic.

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1. Introduction

Heavy metals are those elements that have a density of more than 5 g/cm³, atomic weight 63.546 to 200.590 and a specific gravity greater than 4.0 [1]. These metals include mercury, arsenic, cadmium, chromium, copper, lead, nickel, zinc, molybdenum and vanadium. Living organisms normally require some of these heavy metals up to certain limits and in case of excess accumulation, it will lead to severe detrimental effects [2]. Environmental pollution by heavy metals can occur in many different ways, either directly or indirectly. The main routes to transfer metals throughout the environment are the atmosphere and flowing waters. Heavy metals may enter the food chain as a result of their uptake by edible plants, thus, the determination of heavy metals in environmental samples is very important.

Heavy metals in air, soil, and water are global problems that are a growing threat to the environment. There are many sources of heavy metal pollution, including the coal, natural gas, paper, and industries [3]. Extreme accumulation of heavy metals in agricultural soils through wastewater irrigation, may not only result in soil contamination but also affect food quality and safety. Heavy metal accumulation in soils and plants is of great concern because of the potential human health risks. Absorption of metals from soil water to soil particles is the most important chemical determinant that limits mobility in soils [4]. Heavy metals contamination of soil is widespread due to metal processing industries, tannery, combustion of wood, coal and mineral oil, traffic, and plant protection. Heavy metals may reach and contaminate plants, vegetables, fruits and canned foods through air, water, and soil during cultivation [5]. Vegetables cultivated in wastewater-irrigated soils take up heavy metals in large enough quantities to cause potential health risks to the consumers. Again the inhalation and ingestion of heavy metals may cause various diseases such as anemia, neuropsychological effects, liver diseases, gastrointestinal pathologies implications [6]. Hence, it is necessary to identify the potential of risk agents that come into contact with the human-environment and also needed to take action to prevent and control contamination of the food chain by heavy metals and trace elements in Bangladesh

The risk to health from certain elements in food can be assessed by comparing estimates of dietary exposures with the Provisional Tolerable Weekly Intakes (PTWIs) and Provisional Maximum Tolerable Daily Intakes (PMTDIs) recommended by the Joint Expert Committee on Food Additives (JECFA) of Food and Agriculture Organization (FAO) and World Health Organization (WHO) programmers on chemical safety. The National Food Policy Plan of Action (2008-2015) (area of intervention 3.6) [7], as well as other national policies, recognize the importance of reducing food contamination in Bangladesh to improve human health.

The main objectives of this study are as follows:

- To detect the presence of heavy metals (mercury, cadmium, lead, copper, and arsenic) in water and food consumed by households.
- To estimate the level of selected heavy metals (mercury, cadmium, lead, copper, and arsenic) in water and food consumed by households
- To determine the bioaccumulation of heavy metals in the water sample.
- To determine the toxicity of each metal in comparison with the standard label of toxicity of heavy

metal.

2. Materials and Methods

2.1. Analysis of Elements

For screening heavy metals, the principal methods used are energy dispersive X-Ray fluorescence (Albaiges and his colleagues) [8], neutron activation analysis (NAA), (Jamieson and Hancock and his colleagues) [9], mass spectrometry (MS), (Swart and his colleagues) [10], flame atomic absorption spectroscopy (AAS) (Taylor and his colleagues) [11] and flame atomic emission spectrometry (AES) (Taylor and his colleagues 2006) [8] chemical polarography (Joseph and his colleagues) [12], voltammetric methods (Melluci and Locatelli and his colleagues) [13], Infrared Spectroscopy (IR), Nuclear Magnetic Resonance Spectroscopy (NMR), anodic stripping voltammetry (Salisbury and his colleagues) [14] and High-performance liquid chromatography (HPLC) (Stolker and his colleagues) [15]. However, in this study, the atomic absorption spectroscopy (AAS) has been used.

2.1.1 Atomic Absorption Spectroscopy

Atomic absorption spectrometry was first used as an analytical technique, and the underlying principles were established in the second half of the 19th century by Robert Wilhelm Bunsen and Gustav Robert Kirchhoff, both professors at the University of Heidelberg, Germany. The modern form of AAS was largely developed during the 1950s by a team of Australian chemists. They were led by Sir Alan Walsh at the Commonwealth Scientific and Industrial Research Organization (CSIRO), Division of Chemical Physics, in Melbourne, Australia.

Nowadays, atomic absorption spectrometry (AAS) systems are common element selective detectors. It is a very delicate and useful method of elemental analysis which allows us to determine metals in a variety of samples at the picogram level. It has many uses in different areas of chemistry with having thousands of applications involving a wide diversity of samples. It is a comparatively cheap method for analyzing metals in biological fluids and tissues as well as in water sources.

2.1.2 Principles

Absorption spectrometry uses a simple technique to assess the concentration of an analyte in a sample. It requires standards with known analytic content to establish the relation between the measured absorbance and the analytic concentration and relies therefore on the Beer-Lambert Law.

In short, the electrons of the atoms in the atomizer can be promoted to higher orbital's (excited state) for a short period (nanoseconds) by absorbing a defined quantity of energy (radiation of a given wavelength). This amount of energy, i.e., wavelength, is specific to a particular electron transition in a particular element. In general, each wavelength corresponds to only one element, and the width of an absorption line is only of the order of a few picometers (pm), which gives the technique its elemental selectivity. The radiation flux without a sample and with a sample in the atomizer is measured using a detector, and the ratio between the two values (the absorbance) is converted to analytic concentration or mass using the Beer-Lambert Law. Radiation flux is a

measure of the flow of radiation from a given radioactive source. $\Phi = L/4\pi r^2$ is the radiation flux, L is the Luminosity (measured in Watts) and r is the distance from the radiation source. Radiation flux density is a related measure that adds area dimensions to the above definition.

2.1.3 Instrumentation

In order to analyze a sample for its atomic constituents, it has to be atomized. The atomizers most commonly used nowadays are flames and electro thermal (graphite tube) atomizers. The atoms should then be irradiated by optical radiation, and the radiation source could be an element-specific line radiation source or a continuum radiation source. The radiation then passes through a monochromator in order to separate the element-specific radiation from any other radiation emitted by the radiation source, which is finally measured by a detector.

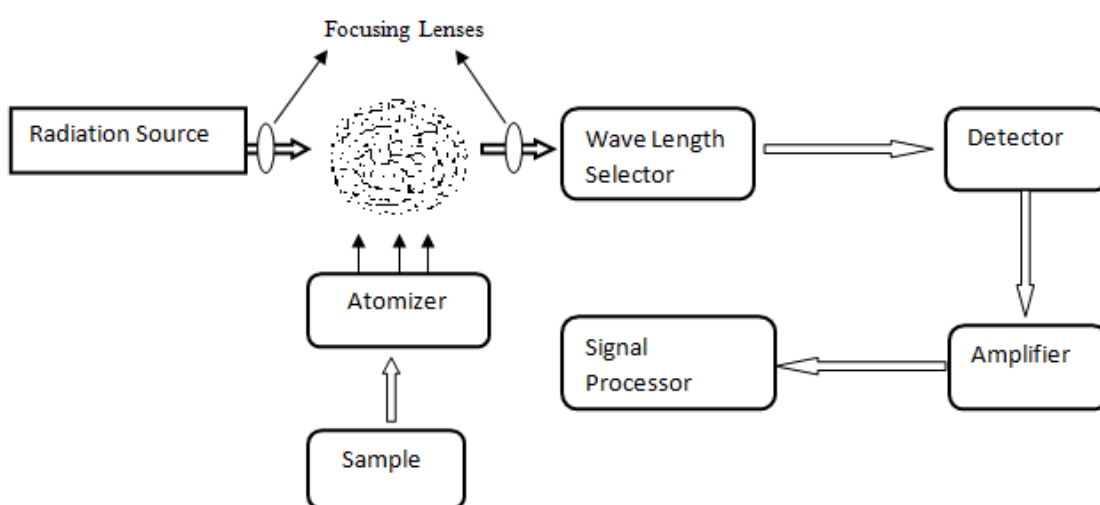


Figure 1: Schematic diagram of AAS equipment

2.1.3. a) Radiation Source (Hollow Cathode Lamp)

This is the source of analytical light line for the element of interest and gives a constant and intense beam of that analytical line.

2.1.3. b) Atomizer (Flame)

The atomizer will destroy any analyte ion and break complexes to create atoms of the element of interest.

2.1.3. c) Wavelength selector (Monochromator)

A wavelength selector isolates analytical line photons passing through the flame and remove scattered light of the other wavelength from the flame. This only impinges a narrow line on the photomultiplier tube.

2.1.3. d) Detector (Photomultiplier tube (PMT))

It determines the intensity of the analytical line exiting the monochromator. The PMT is the most commonly used detector for AAS.

2.2 Experimental

2.2.1 Apparatus

Thermo Electron Corporation Solar M6–Mk II atomic absorption spectrometer with GF 95 graphite furnace, FS95 furnace auto sampler and Zeeman background correction system. Thermo Electron Corporation hollow cathode lamps and Thermo Elemental pyrolytically coated graphite tubes (extended lifetime) were used for all measurements. Metter Delta 340 pH-meter, Microwave mineralizer, Plazmatronika UniClever TM II was used for mineralization process of samples. Glass columns of 4 mm in internal diameter filled with 0.2 g of sorbent conditioned with a solution of pH 4. All glassware was cleaned by soaking for 12 h in 2% solution of alkaline detergent, next rinsed with water and leached for 24 hours in 10% nitric acid and finally washed with water.

2.2.2 Reagents

The water of resistivity 18 MΩcm⁻¹ purified by Water Purification System–Millipore, nitric and hydrochloric acids purified by sub-boiling point distillation, hydrofluoric acid 40% supra pure (Merck), dithizone(Merck), resin Diaion HP–2MG (Supelko). Dithizone sorbent was prepared as follows: 1 g Diaion resin (polymetacrylic ester) was placed in a 50 mL separator funnel and shaken with 25 mL of 0.04 mol L⁻¹ dithizone solution in chloroform for 1 h. The sorbent was separated by filtration, allowed to dry in air for 12 hours and stored in dark in a refrigerator at 4°C. Standard stock solutions: 1 mg L⁻¹ of Cd, Pb, and Cu were prepared by dissolution of the suitable. reagents in suitable acids. Working standard solutions were obtained by dilution. Palladium-magnesium modifier solution 100 µg mL⁻¹ of Pd and Mg in 0.5 mol L⁻¹ HNO₃.

2.3 Analytical procedure

2.3.1 Preparation of samples

Table 1: Conditions for sample mineralization. Sample 0.2-0.5gm., maximum volume of solution 8 ml.

Materials	Active Reagents	Stages	Process Parameters		
			I	II	III
Soil and street dust	HNO ₃ : HF= 1:2	Time, min	5	5	10
Plant materials	HNO ₃ : HF= 4:1	Power of generator, %	60	80	100
Fishes	HNO ₃ : H ₂ O ₂ =2:1	Pressure, atm	17-20	27-30	42-45
Poultry liver	HNO ₃	Temperature, °C			-310

Food products and plant materials containing a large amount of organic matter were crumbled, homogenized in the small food mill and then dried at the temperature of 70°C to a constant weight. The samples that agglutinated during the drying process were powdered in the ag mortar. The content of water was determined in the parallel samples to enable the calculation of the obtained results for original fresh material. 0.2 g to 0.5 g of the dried samples and a maximum of 8 mL of mineralizing solution were put into microwave min under the conditions presented in Table 1.

2.3.2 Separation and preconcentration

The solution after digestion was transferred to the Teflon evaporating dish and evaporated to wet salts. The residue was dissolved in 5 mL 2 molL⁻¹ HCl which then filled with water and transferred to 50 mL beaker, adjusted to pH about 4 by ammonium hydroxide solution using pH meter and passed through the column (previously conditioned with the solution of pH = 4) at the flow rate 5 mL min⁻¹. Next, the column was washed twice with the solution used for conditioning and the metals were desorbed by 10 mL 2 molL⁻¹ HNO₃. The effluent was collected in a 20mL volumetric flask and fulfilled up to the mark.

2.3.3 Determination

The separated metals were determined by graphite furnace atomic absorption spectrometry. 2 µL mL of the effluent was injected into a pyrolytic coated graphite tube. The atomization conditions for all determining elements are presented in Tables 2 and 3. Copper was atomized without modifier whereas Cadmium and Lead in the presence of 20 µL of palladium magnesium modifier solution pyrolyzed preliminary in the tube at 800 °C temperature, before addition of the sample. The analytical signals were measured as the integrated absorbance and evaluated based on properly diluted standard solutions in 1 molL⁻¹ HNO₃.

Table 2: Spectral parameters

Element	Cd	Pb	Cu
Wave length, nm	228.8	224.0	217.8
Band pass, nm	0.5	0.3	0.5
Lamp current, mA	10	15	10
Background correction	Zeeman	Zeeman	Zeeman
Integration time, s	3	3	3

GF AAS determination of cadmium, lead, and copper in environmental materials and food

Table 3: Temperature and time programs

Operation	Cd			Pb			Cu		
	T	Ramp	Time	T	Ramp	Time	T	Ramp	Time
	°C	°Cs ⁻¹	s	°C	°Cs ⁻¹	s	°C	°Cs ⁻¹	s
Drying	100	10	30	100	10	30	100	10	30
Pyrolysis	800	150	20	800	150	20	800	150	20
Atomization	1700	0	3	1900	0	3	2100	0	3
Cleaning	2400	0	2	2600	0	2	2700	0	2

The blanks were processed in the identical way simultaneously with the samples and their values, if significant, were deducted from analytical signals.

3. Results and discussion

The samples of food products and plant materials contain mostly organic mater and many of them are very difficult to mineralize. Moreover, some products upon their disintegration can form during the mineralization of volatile compounds with the metals to be determined and cause their losses. Therefore, the mineralization process should be carried out in a closed system under severe conditions. On the other hand, the strongly oxidizing agents as perchloric or sulfuric acid must be avoided since they can interfere with the separation process on the column. Organic samples evolve during mineralization a large amount of gaseous products and the mass of the samples processed in a closed system is limited and must be carefully chosen [16]. The optimum mineralization conditions selected for various types of analytical samples are presented in Table 1.

The separation and preconcentration of copper, cadmium and lead on various sorbents were investigated in previous work and on this basis, the optimum separation condition, applied in this work and presented in the section "Recommended analytical procedure", was chosen[17].

For centuries, mercury was an essential part of many different medicines, such as diuretics, antibacterial agents, antiseptics, and laxatives. In the late 18th century, ant syphilitic agents contained mercury. It was during the 1800s that the phrase "mad as a hatter" was coined, owing to the effects of chronic mercury exposure in the hat-making industry, where the metal was used in the manufacturing process. In 1889, Charcot, in his Clinical Lectures on Diseases of the Nervous System, attributed some rapid oscillatory tremors to mercury exposure [18].

In Wilson's classic textbook of neurology, published in 1940, Wilson concurred with Charcot's attribution of tremors to mercury poisoning but also described mercury-induced cognitive impairments, such as inattention, excitement [19].

In 1961, researchers in Japan correlated elevated urinary mercury levels with the features of the previously mysterious Minamata disease. Before the etiology of Minamata disease was discovered, it plagued the residents around Minamata Bay in Japan with tremors, sensory loss, ataxia, and visual field constriction [20].

Organic mercury compounds, particularly methylmercury, are concentrated in the food chain. Fish from contaminated waters are the most common culprits. Industrial mercury pollution is often in the inorganic form, but aquatic organisms and vegetation in waterways such as rivers, lakes, and bays convert it to deadly methylmercury. Fish eat contaminated vegetation, and the mercury becomes biomagnified in the fish. The fish protein binds more than 90% of the consumed methylmercury so tightly that even the most vigorous cooking methods (eg, deep-frying, boiling, baking, pan-frying) cannot remove it.

Fish is widely consumed in many parts of the world by humans because it has high protein content, low saturated fat and also contains calcium, phosphorus, iron, trace elements like copper, and a fair proportion of the B-vitamins known to support good health [21]. Many reports on the contamination of fish by chemicals in the environment were reported [22]. Heavy metals are considered the most important constituents of pollution from the aquatic environment and the sea due to toxicity and accumulation by marine organisms.

All the aquatic samples collected from the sites contained detectable amounts of the elements studied (Cadmium, Mercury, Lead). These elements were present in all the fish samples and at varying concentrations. It must be noted that varying concentrations of the heavy metals were measured in the sampled fishes with some fishes reporting very high concentrations whilst other samples measured relatively lower concentrations. Cadmium is classified as chemical hazards and maximum residual has been prescribed for human [23]. Cadmium tended to be the least concentrated in the fish as compared to other elements measured. Concentrations of cadmium varied from 0.50 ± 0.01 mg/Kg (dry wt.) which is high compared to the permissible level of 0.01 mg/Kg. Exposure to heavy metals such as cadmium is of immediate environmental concern. A direct relationship between heavy metal poisoning and thyroid dysfunction was reported in rabbits by [24].

According to the Third National Report on Human Exposure to Environmental Chemicals (NHANES), Cd exposure is widespread in the general population. No standards exist correlating blood or urine Cd measurements with clinical toxicity; The kidney is considered the critical target organ for the general population as well as for occupationally exposed populations. The accumulation of cadmium in the kidney leads to renal dysfunction. Chronic obstructive airway disease is associated with long-term high-level occupational exposure by inhalation. The lead levels recorded in all the species exceeded the permissible limits of WHO which mentioned that the lead level should not be more than 2 mg/Kg [25]. The bioaccumulation of these metals may pose a great hazard to the health of humans. Chronic lead poisoning is characterized by neurological defects, renal tubular dysfunction, and anemia.

EDTA is approved by the FDA for lead and other heavy metals and has a long history of safe use. It should not be given faster than one gram per hour nor in a dosage greater than three grams per session. Sessions should be at least five days apart, and the replacement of essential minerals should be done orally between sessions. Several effective protocols exist implementing these principles [26].

In an earlier evaluation, the Joint FAO/WHO Committee on Food Additives (JECFA) reaffirmed a previous tentative evaluation of a maximum daily load of 0.5 mg/kg body weight as a Provisional Maximal Tolerable Intake (PMTDI) for man from all sources, which amounts to 30 mg/day for a 60 kg person [27]. The Scientific Committee on Food (SCF) has established a Population Reference Intake (PRI) of 1.1 mg copper/day [28]. The Tolerable Upper Intake Level (UL) for copper set by the SCF is 5 mg/day for adults, which would correspond to 6.34 mg/day of copper (II) oxide and 1-4 mg/day for children, depending on age.

Copper salts have moderate acute toxicity, with soluble salts being more toxic than insoluble ones. In short-term (2 weeks) repeated dose toxicity tests in rats and mice, copper salts are associated with adverse effects such as gastrointestinal irritation and liver and kidney toxicity. Reported NOAELs are in the range of 23-104 mg/kg BW/day copper, but kidney effects have been shown in male rats at levels as low as 10 copper mg/kg BW/day.

Acute toxicity of copper in humans is rare. Average daily consumption of 1.64 liters of drinking water containing 3 mg/L ionized copper (II) was associated with nausea, abdominal pain, or vomiting. In a human study, both copper sulfate (a soluble compound) and copper (II) oxide (an insoluble compound) showed comparable effects, implying that the ionic copper present in the stomach is responsible for the induction of gastrointestinal manifestations. Twenty subjects presented gastrointestinal disturbances at least once during the study, suffering diarrhea (with or without abdominal pain and vomiting), and the other eleven subjects reported abdominal pain, nausea, or vomiting. Preliminary studies in different parts of Bangladesh indicate that the food chain is exposed to contamination by heavy metals and trace elements. Islam and his colleagues found that industrial sludge, often used as a soil conditioner or fertilizer has high concentrations of heavy metals [29]. Similarly, high levels of heavy metals were found in soils in the Sundarban [30]. When these metals are absorbed by crops and animals they enter the food chain and constitute a serious health hazard. An analysis of heavy metal concentrations in vegetables in Jessore shows that all of the vegetables commonly consumed in diets contain dangerously high concentrations of heavy metals [31].

4. Conclusion

Contamination of metals in the environment and human diet represents a persistent problem that will continue to be a burden on human health. The purpose of this study will be focused on the concentration of selected metals in water and food. The results of this study will be compared with those of other studies that revealed similar levels of metals in water and food. However, the number of analyzed heavy metals and sample size were limited in our study due to small financial support. There are also other methods which are also really convenient for the determination of heavy metals. Such as in case of ICP-OES method, the calibration function has a wide dynamic range and the number of measurable elements are also high ensuring much accurate data. Concededly, the metals were found enough to cause potential health hazard. People who will continuously consume this type of contaminated food and water are nearly under threat to cancer in the long run.

5. Recommendations

Chemical modifiers may be applied to avoid some chemical interference. Standard conditions as well as the

nature and amount of a chemical modifier for the selected elements should be observed carefully before applying. For some cases, pre-concentration stage might be necessary to check the deductibility of the trace elements. Future studies should focus on the ability of lactobacilli to bind an array of heavy metals at human physiologically relevant concentrations and assess in humans the extent to which levels can be reduced over time. If such interventions can encompass locally produced foods, such as yogurt made in the home or community, this may potentially provide an affordable option for billions of people around the world who are consuming these toxic metals inadvertently on a daily basis.

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