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PRESENCE AND SOURCE OF TOXIC HEAVY METALS IN CAMELLIA SINENSIS SHOOT

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ABSTRACT: Tea has been considered as a health promoting drink & stimulant. Globally 4842.1 thousand tons tea consume annually. But due to change in soil characteristics, indiscriminate use of pesticide & fertilizer lead to accumulation of toxic heavy metal like Lead(Pb), Arsenic(As), Copper(Cu), Nickel(Ni) & Cadmium(Cd) in tea leafs & bud. Tea market in India is not well regulated, an abundance of loose tea dust grade leaf very often sell in the local market without any proper labeling and regulatory license no. As this metal have high molecular weight and need more time to eliminate from our body, therefore, serious health ailment like teratogenicity, spontaneous abortion, renal failure & encephalopathy may happen if heavy metal presence in tea leafs & bud above the limit set by different regulatory authorities. In this review, we try to find out the source of heavy metal in the tea garden and differ limit for heavy metal set by major regulatory agencies. Soil, irrigation water, underground water, fertilizer, pesticide & anthropogenic factor influence the accumulation of heavy metal in tea leaf & shoot. Although all factors related heavy metal entry into the plant is not possible particularly the anthropogenic factors. But good agricultural practice can limit the entry of heavy metal into a shrub.

INTRODUCTION: Tea has been widely recognized as a health promoting aromatic beverage. Globally 4842.1 thousand tons tea consume annually 1. After water, it is the most widely consumed drink in the world ². Tea is used in folk medicine for a headache, digestion, diuretic, enhancement of immune defense, as an energizer and to prolong life. According to pharmacological & epidemiological studies, tea is considered to have beneficial effects on the prevention of many diseases, including cancer, parkinsonian disease, myocardial infraction & coronary artery disease ³.



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Tea is also prevented dental carries due to the presence of fluorine. The role of tea is well established in normalizing blood pressure, lipid depressing activity, prevention of coronary heart disease and diabetes by reducing the blood-glucose activity. Tea also poses germicidal & germ static activities against various gram positive & gram negative human pathogenic bacteria ⁴.

It's known that serious systemic health problems can develop as a result of excessive accumulation of toxic heavy metal such as Cadmium(Cd), Chromium(Cr) & Lead(Pb) in the human body **Table 2**. Heavy metals are extremely persistent in the environment: they are nonbiodegradable and nonthermodegrable and thus readily accumulate to the toxic levels. Heavy metals are elements having an atomic weight between 63.5 and 200.6 and a specific gravity greater than 5.0 ⁵.

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Uptake and accumulate the heavy metal by plant & animal as well as the potential for its propagation into the food chain exacerbate its toxic health effect

In this review article, we try to highlight the heavy metals present in tea leaf particularly in black tea variation by searching the existing research work on the heavy metal presence on tea leaf by using SCOPUS & Google Scholar as a search engine. And also try to find the sources of this heavy metal & how these individual sources influence the metal bioavailability in the tea plant.

2. Regulatory guideline for heavy metal: The main threats to human health from heavy metals are associated with exposure to lead(Pb),cadmium(Cd), mercury(Hg) & arsenic(As). This metal has been extensively studied and their effects on human health regularly reviewed by national & international bodies such as USEPA, WHO, FACCI etc. ⁷ **Table 1**.

TABLE 1: REGULATORY GUIDELINE ON HEAVY METAL

SL No	Name of Heavy Metal	Limit Set by Regulatory Authorities			
		USFDA	WHO	FASSI	SFDA
1	Arsenic (As)	10 ppm	10μg/Kg BW	0.5 ppm	≤0.3 mg/Kg*
2	Cadmium (Cd)	0.005mg/L	7µg/Kg BW	0.1 ppm	1 ppm
3	Copper (Cu)	0.05 mg/ml	1mh/m^3	150 ppm*	-
4	Lead (Pb)	0.1 ppm	0.2 ppm	10.0*	$\leq 0.5 \text{mg/Kg*}$
5	Mercury (Hg)	0.002mg/L	3.3µg/Kg BW	0.5 ppm	0.5 ppm
6.	Nickel (Ni)	70 ppm	-	1.5 ppm	-

^{*}Specific for Tea only

USFDA: United States Food & Drug Administration; WHO: World health Organization,

FASSI: Food Safety & Standard Authority in India; SFDA: State Food & Drug Administration China

TABLE 2: TOXIC EFFECT OF HEAVY METAL IN HUMAN

Sl No	Heavy Metal	Effect in Human Health	
1	Arsenic (As)	Encephalopathy, Black foot disease, Stocking Glove Syndrome, Conduction block, QT prolongation 35, 36	
2	Cadmium (Cd)	Hepatic, Pulmonary, Testicular injury, Renal & bone injury 35	
3	Copper (Cu)	Wilson Disease, Menkes' Disease 35	
4	Lead (Pb)	Teratogenic, Inhibit Hemoglobin synthesis, Kidney dysfunction, Joint & reproductive system	
		dysfunction, Damage Central nervous system & peripheral nervous system, Psychosis, poor	
		development of grey matterin child ¹³	
5	Mercury (Hg)	Spontaneous abortion, Congenital malfunction, Gestrointestinal tract & Central nervous	
		system disorder, Pulmonary edema, Haemolysis ¹³	
6.	Nickel (Ni)	Immunomodulatory & immunotoxin, Reproductive toxin class II, Kidney injury & Frank	
		haematuria, Reduce hemoglobin HB level & Neurotoxin. 37, 38	

3. Heavy metal source & accessibility: The soil, water, and air are contaminated by these heavy metals, which directly affect agricultural crops through cultivation consumable heavy metals from contaminated agricultural crops. On a long-term basis, this can accumulate in plant crop, and therefore in an animal which consumes the crops. Once accumulated in animals or crops, to decontaminated or create can be difficult ⁸. Emission of heavy metals to the environment occur via a wide range of processes and pathways, including to the air e.g. during combustion, extraction and processing, to surface waters via runoff & release from storage and transport, to the soil and hence into ground waters and crops. People may be exposed to potentially harmful

chemical, physical & biological agents in air, food, water or soil. However, exposure does not result only from the presence of a harmful agent in the environment. The key word in the definition of exposure is contact. There must be contact between the agent and the outer boundary of the human bodies, such as the airways, the skin or the mouth. Exposure is often defined as a function of concentration and time: "an event that occurs when there is contact at a boundary between a human and the environment with a contaminant of a specific concentration for an interval of time".

Generally the source of heavy metal in tea garden from Soil, Ground Water, Fertilizer, Pesticides & geographical location of the garden. Among this soil has been considered as a reach source of heavy metal in any tea garden.

3.1. Soil as a source of heavy metal: The nature of the soil is one of the most important factors in determining the heavy metal content of food plants. Heavy metal contamination of agricultural soil can pose long-term environmental problems and is not without health implication. A number of factors influence the concentration of heavy metals on and within plants. These factors include climate, atmospheric deposition, the nature of the soil on which the plant is grown and the time of harvesting ⁹. Heavy metal in soils can be sequestered into a number of fractions including the sorbed to clay, hydrous oxides, organic matter & metal within the matrix of soil minerals¹⁰.

Another probable reason behind the accumulation of heavy metal in tea is that this plant is acidophilic, and acidic soil in tea gardens are affected by an increase in heavy metal dissolution, in comparison to the neutral & alkaline soil, which increases the uptake of metals by tea leafs ¹¹.

Arsenic(As) is a crystal which is very dangerous for human health, the even low level of Arsenic, leads to carcinogenesis 12. The maximum permissible limit for Arsenic is 50µg/L USEPA & 10µg/LWHO. It has been reported that tea gardens from Karbi-Anglong, Cachar & Karimgani district India, soil contain Assam, traces Arsenic(As). The particular concentration of AsIII concentration is high in the soil of Karbi-Anglong district. AsIII is 10 times more soluble, mobile than ASV and reacts with sulphydryl group of the enzyme ¹¹. Study on contamination of tea garden soil by Arsenic revealed that traces of Arsenic 1.27- 2.54mg/Kg was found in aerial part of tea. Particularly on mature tea leaf have a tendency to accumulate Arsenic 0.03-0.08 mg/Kg whereas young shoot does not have this tendency. The concentration of Arsenic in the root is changed from 1.2 to 1.9 mg/Kg. This study hypothesized that translocation of Arsenic from root to leaf is very low because of their buffering property ¹¹.

Soil acidification & organic matter accumulation were well correlated with Lead(Pb) bioavailability. Lead in fertilizer may be another significant contributor to the annual input of Pb in soil.

A study conducted in the tea gardens at Zhejiang province in China find that total Pb concentration in soil is very high on the surface layer of soil as compared to China's own national guideline ¹⁰. The solubility of Lead increase with H⁺ ion concentration in soil, hence, the older tea gardens are less affected by Lead Pb, as the garden become older the soil pH has been increased and lead concentration decrease. Average of Lead (Pb) in tea leaf in Zhejiang province of China was 4.4mg/Kg & in soil was 27.16 to 47.99mg/Kg.

Allowable limit of Pb in tea leaf is 5mg/Kg set by Europian union & 20mg/Kg by Japan. Lead poisoning in human body leads to learning disability & Anemia ^{10, 13}. In 2005 Chong Wei Jin *et al.*, report from China that soil acidification & organic matter accumulation were well correlated with Lead(Pb) bioavailability and their study shows that tea garden soil adjacent to Hangzhou city in the Zhejiang, province of china heavily contaminated with Lead(Pb) ¹⁰.

Sushma R Ambedkar and her co-workers find a high level of Lead(Pb) concentration14.0212µg/gm in one popular tea brand in India. They collect tea leaf from the local market at Mumbai, India. The level of Nickel(Ni) is also high in all tea brand leaf collected for this study, followed by Cadmium(Cd) ⁴. A study conducted in Sonitpur district of Assam, India in the tea garden belt found that soils health in tea gardens is not in accordance with the fertility rating chart given by Indian Council of Agricultural Research ICAR. The inherent fertility of the soil in the study area is poor because of low nutrient status in soil.

The average pH of the soil is highly acidic whereas N, P, KNitrogen-Phosphorus-Potassium percentage in per hectare soil is also very high as compared to the Indian Council of Agricultural Research ICAR limit, the N-P-K percentage is also another factor which influences heavy metal accumulation in soil

Copper(Cu) which is also regarded as heavy metal & have been attributed for non-Indian childhood cirrhosis & Wilson Disease also been studied in some province in China. And it has been found that Copper(Cu) concentration in the sample of Tea leaf ranged from 8.05 to 33.50 mg/Kg.

And Copper accumulation depends on the high level of soil acidity. Therefore, soil acidification has a direct correlation with Copper accumulation in Tea leaf and tea garden soil ¹⁵.

3.2. Influence of fertilizer: Fertilizer has an important role in the growth of plants. Fertilizer also has a role to make plant healthy. Fertilizer provides some macronutrient like Nitrogen, Phosphorus, Potassium, Sulphur, Manganese, Calcium N, P, K, S, Mg, Ca & micronutrients like Manganese, Chlorine, Zinc, Molybdenum Fe, Mn, Cl, Zn, Cu, Mo to either on soil or plant itself. Nitrogen is the most important essential nutrient for improving yield & quality of tea ¹⁶. But long term use of nitrogen N fertilizer can accelerate the soil acidification ¹⁷. The high rate of N fertilizer in some tea field cause root damage or root death. A study conducted in Japan on the influence of N fertilizer on tea root growth found that in the topmost layers of white roots is reduced when nitrogen amount is high in soil ¹⁶.

Heavy metal can also accumulate in the soil due to the application of liquid & soil manure as fertilizer or inorganic fertilizer ¹⁸. Another study from Japan report that when N input has 400 Kg/Ha/Year white root active is numerous but if N input is 800Kg/Ha/Year then the white root decrease by 57%. They also report that soil will more acidic if the rate of N fertilizer application is high, the average pH is below 4.0 ¹⁹. A further acidification of soil increased the risk of heavy metal accumulation.

Cadmium(Cd) has been present in most of the fertilizer. Lime & super phosphate contain impurities of cadmium ²⁰. High fertilizer applications and acidic atmospheric deposition, combined with insufficient liming, may also cause an increase heavy metal availability, aggravate the problem of deteriorating food quality, metal leaching & impact on soil organism ²¹.

A researcher from the Iran reports the concentration of cadmium in composite fertilizer and triple super phosphate is higher than prescribed limit set by California Department of Food & Agriculture CDFA & also the concentration of Arsenic in Zinc sulfate fertilizer also two-fold higher than prescribed limit ²⁰. A surplus of heavy

metals in soil is frequently caused by using fertilizers, metal pesticides, and sewage sludge or by industrial activity & concentration of heavy metals in soils can increase by repeated & excessive fertilizer and pesticide application ²².

A group of researcher from Spain report in 1995 super phosphate contains that a higher concentration of Cadmium(Cd), Cobalt(Co), Cupper(Cu) & Zinc(Zn) as impurities. They find only Cu concentration in soil meet the limit set by Europian Union Legislation ²². In tea plantation, Ammonium Sulphate and single super phosphate have been extensively used, and ammonium sulfate caused the greatest increase in H⁺ ion concentration followed by ammonium phosphate ²³. Progressive acidification of soils resulted in the accumulation of metals in vegetation above that attributed to increasing total soil content resulting from the deposition of heavy metals from the atmosphere.

Manganese(Mn) & Cadmium(Cd) were most sensitive to increase soil acidity with effective mobilization occurring at pH 6.0-5.5 followed by Zn, Ni & Cu at pH 5.5-5.0, Lead(Pb) has been mobilized at pH < 4.524. This metal mobilization influence deposition of heavy metal in the tea plant.

3.3. Influence of Pesticides: Any substance or mixture of substance intended for preventing, destroying, repelling or mitigating any pest or weed is pesticide ²⁵. About 145 pesticides registered in India and their annual production was approximately 85000 metric tons. Aldrin, BHC, Chlordane, Endrin heptachlor, and Dieldrin are included in the list as banned pesticides and DDT and Lindane are included in the list of pesticides restricted for the use in India by central insecticides board and registration committee ²⁶.

Narita and his co-worker report in 1987 that high concentration of Cu, Pb, and As derived from such metal-based pesticides have accumulated in the surface soil ²⁷.

Soil heavy metal concentration also depends on the growth promoting bacteria present in the soil. These bacteria present in the soil. This bacteria helps to control the heavy metal migration from layers of soil to the soil solution But pesticides always kill this non-target bacteria, which adversely lead to the increase concentration of

heavy metal in the soil solution. Almost all pesticides are moderate to weakly sorb in soils, mainly by soil organic matter because most of the molecule in pesticides are dominated by polar groups *i.e.* aliphatic and/or aromatic carbon and other have only one functional group ²⁸.

Most pesticides reduce nitrogen fixation by a soil microorganism. Numerous report on the influence of pesticides on nitrogen fixation has been published ²⁹. Soil microorganisms *Rhizobacteria* have been shown to possess several traits that can alter heavy metals bioavailability, through the release of chelating substances, acidification of microenvironment & influence in redox potential. Diazotrophs may be free-living or in association with nodules on roots, which serve as a metal buffer zone, providing further protection for the plant against invading metal ion. By mechanisms pesticides, the application can influence the accumulation of heavy metal in root & shoot in the plant.

Tea garden all over the world is routinely using pesticides. One of the world's largest tea exporter country India harbor about 300 species of pests, therefore Organophosphate, Organochlorine, Carbamates and synthetic Pyrethroid insecticides have been used routinely ³⁰.

Another world largest tea & pesticide producer country China routinely use pesticides to control the pest in tea garden ³¹.

3.4. Heavy Metal From ground Water: Heavy metals occur in the earth's crust & may get solubilized in groundwater through natural processes or by a change in soil pH. Although groundwater flow characteristics are vital in influencing the transport of metal contaminants ³². Ground water move through rocks and subsurface soil, it has a lot of opportunities to dissolve substances as it move ³³.

A study was done by a researcher from Assam, India report that presence of Cadmium Cd and Manganese mg in ground water at 14 different location in tea garden belt at Assam and their range is above 0.003 ppm and 0.15 for Cadmium and Manganese ³⁴. To date, very few report has been publishing on the role of ground water in a heavy metal deposition on tea leave.

CONCLUSION: Heavy metal is not always harmful to human health, traces of metal is required as an essential nutrient for growth & development of human body. But when their concentration has been increased in our daily diet & drink, over a period of a long time we may develop different physical & psychological problem. Heavy metals have the high molecular weight, it is not easily eliminated from our body, and long-term deposition of the metal in our body, change our body's normal hemostasis & cellular functioning. As in our country tea has been unofficially regarded as the national drink, so as a brew lover we represent some of the finding reported by the different researcher, about the presence of heavy metal in tea leaf & their possible source. We hope regulatory authorities will make a comprehensive & harmonized guideline in near future only for tea to minimize the risk of human health and millions of tea lover will enjoy their morning cup of tea without thinking its harm

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