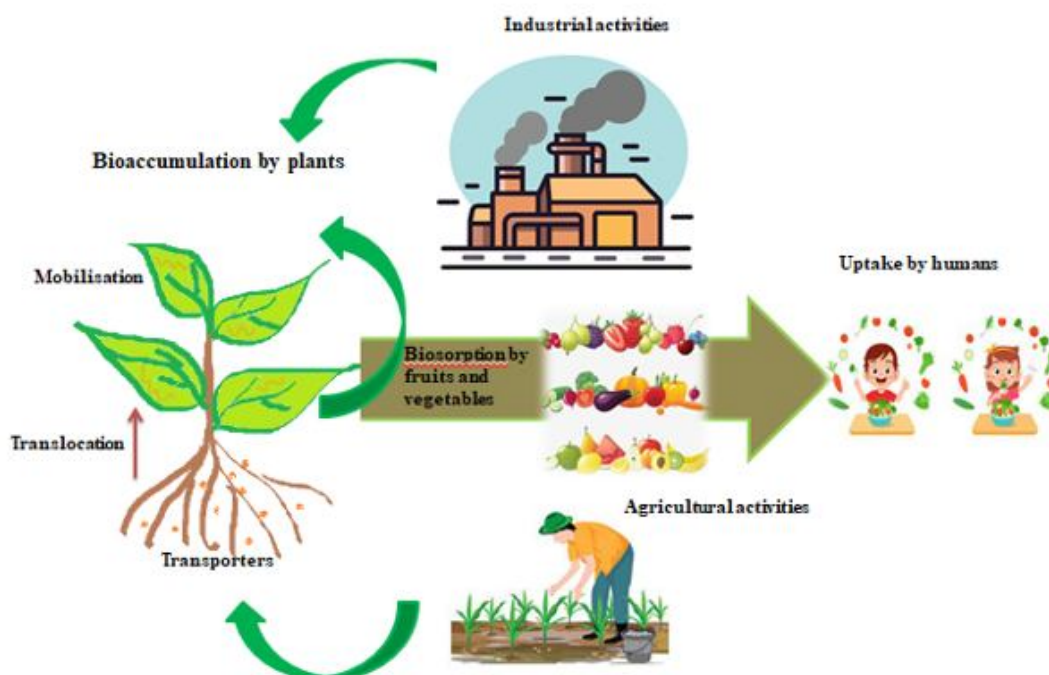


Original Research Article

Investigation of heavy metal contamination in fruits and vegetables from local markets of Chennai, Tamil Nadu

Abstract

Certain areas of Chennai such as red hills, Minjur, T. Nagar, Thiruvanmyur, Ambatur have been found to be particularly susceptible to high pollution levels due to severe traffic congestions, the ignition of coal, cement industries, SIPCOT and other chemical industries etc., which pose threat to the environment. The population besides these industries areas also increasing due to the employment opportunities and other activities. Considering the persistent nature, cumulative behaviour, and probability toxicity of the heavy metals from these industries, it can enter the food chain linking the plant system to the soil (fruits & vegetables) and transformed to human body. The heavy metals, Chromium, Cadmium, Nickel and Lead, when exceed their permissible limit, it causes metabolic malfunctions to human beings. At this at the background, a study was attempted on the levels of metal deposits from industries in the fruits and vegetables from the surrounding local markets in the designated locations. Although there have been numerous research studies conducted worldwide on the subject, this is the first of its kind study in Chennai district. Vegetable and fruit samples collected from the local markets near the industrial zones and in congested areas were analysed and compared for the levels against the global safety standards to ensure that fruits and vegetables can be consumed without the risk of heavy metal contamination. The study's findings demonstrated that all of the fruits and vegetables from the local markets in the study had levels of contaminants below detectable thresholds and within WHO and FSSAI's allowed limits. This indicates there may be no possible health concerns linked to the concentrations of heavy metals in fruits and vegetables from the markets near the highly polluted industrial zones or in the areas with severe traffic congestion.



Graphical abstract

Key words: Industrial areas, Heavy metals, Fruits and vegetables, Chennai market

1. Introduction

Population increases, urbanisation, and industrial activity lead to contamination and a detrimental influence on land, water, and atmosphere (Dev et al., 2023). Due to their long-term toxicity effects, heavy metals are considered the soil pollutants originating via natural and artificial sources. They are the primary focus of recent investigations due to the contaminating effect Wang et al., 2021. World over, the safety of food is a key societal concern. The increased demand for food safety over the past few decades has prompted research on the hazards related to consuming foods contaminated by toxic substances, Lebelo et al., 2021. In regions with high industrial activity, metal concentration in soils tends to increase. These places have a few times more metal deposition than uncontaminated sites. The fact that pollutants may move via plants in soil to reach the food

chain. usage or direct intake by animals that consume them is the most significant effect of pollution on environmental health (Uddin et al., 2021)

Traditional channels and anthropogenic mechanisms are both ways that heavy metals enter the environment (Fig. 3). They can be associated with causes, such as mining operations, deterioration of the earth's crust, and soil deterioration, liquid waste, sewage from cities, and insecticides (Parida et al., 2023). Long-term use of pesticide use, untreated municipal effluents in agriculture fields, sludge waste with metal contamination are the main contributing factors to the accumulation of heavy metal in productive soils (Lebelo et al., 2020). Their movement from soil to plant parts are significantly impacted by a number of soil and plant characteristics that control their bioavailability. Lower improvement of the plant matter, produce, and inadequate caloric value are the results of metal exposure (Taqeer et al., 2022). These substances have the potential to contaminate the atmosphere, streams, and soils from both point and non-point sources (Gavrilescu et al., 2020). Vegetable and fruits contamination with heavy metal draws increasing attention as understanding the problem in relation to population (Mawari et al., 2022). Vegetables and fruits accumulate heavy metals as a result of air and soil contamination. The interaction with contaminated soils or air pollution can cause natural food contamination (Xiang et al., 2021). Fruits and vegetables constitute 90% of the human diet are the most common way that people are exposed to this contamination the remaining 10% is ingested through skin contact and contaminated dust inhalation (Najmi et al., 2023).

Copper, iron, nickel, cadmium, manganese, and zinc are examples of heavy metals that are necessary for humans and play significant role in several metabolic processes. When their consumption approaches specific threshold levels, these metals can be harmful to both people and other living things (Mitra et al., 2022). Even at low quantities, non-essential substances including lead, cadmium, mercury, and arsenic is harmful for humans. Vegetables can accumulate heavy metals due to land and atmosphere pollution (Bansal et al., 2021). Bioaccumulation in food causes serious health risks to people. The human body absorbs these metals through food and breathing. Soil movement and availability contribute to this contamination (Alengebawy et al., 2021). Every person should eat 300 g of greens every day, including tubers, roots, and green leafy vegetables, according to Indian Council of Medical Research Expert Committee. People consume a variety of

vegetables each day, depending on the produce's growing region, the type of irrigation source utilised, and its consumer age (Kumar et al., 2019). In humans, in all forms of deposition of heavy metals (HMs) results in a variety of difficulties. including renal failure, peripheral neuropathy, lung cancer, skin cancer, osteoporosis, bone thinning, and prostate cancer. Heavy metal soil additions have detrimental consequences on food, crop growth, and environmental safety (Tafera et al., 2021)

In Tamil Nadu, cash crops, vegetable crops and fruits crops are cultivated, 10% of all fruits and vegetables are produced in India are grown in Tamil Nadu; these include bananas, mangoes, pineapple, guava, citrus fruits, grapes, and other vegetables. No investigation has found evidence of heavy metal build up in fruits or vegetables; instead, previous studies have mostly concentrated on a limited number of specific vegetable varieties (Ganesh et al., 2022) and on the exposure to contaminant from industrial regions throughout local available markets throughout the world (Table 2) but no significant investigation is being conducted in the Chennai district. So, the main objective of the research is aimed at the heavy metal accumulation (Lead, Cadmium, Chromium and Nickel) in the fruits and vegetables from the local markets near the industrial sites and localities with severe traffic congestion namely red hills, Minjur, T. Nagar, Thiruvanmyur, Ambatur in Chennai district. The findings of these trace elements were compared with standards and also investigated the everyday intake and risks of metal exposure to human health, so that vegetables and fruits can be consumed without any harm to human body.

2. Materials and methods

2.1. Location of the research

The examination was conducted in the Chennai district of Tamil Nadu, in five different parts of the city: Red Hills, Minjur, T. Nagar, Thiruvanmyur, and Ambatur. Leading industries in the region include SIPCOT, the cement, coal, chemical, automotive, and polymer industries, among others. The city has a climate that is both wet and dry. Maps displaying sampling location relative positions (Table 1) (Fig.2). Along with the dense population, industrial production activities have been operating in this area since the late 1970s that influence the surrounding environment. The air pollution levels are also

high. While being transported from the production site to the market, these fruits and vegetables are covered with polyethylene sheets; nevertheless, when they are being marketed, they are exposed to the urban environment.

2.1. Sample collection

From the areas of Thiruvanniyur, T. Nagar, Ambattur, Red hills, Minjur of Chennai districts, same type of vegetable and fruit specimens have been purchased through several nearby marketplaces (Table 2). Four samples were purchased from each market and combined at each sampling location. The collected sample (1 district x 5 local markets x 11 samples) were tagged, placed in PP containers, and shipped to the testing facility. The samples were properly cleaned with deionized water then dried for one day at 60 degrees Celsius in an oven, glass mortar was used to ground the dried samples into a homogeneous mixture, and a 2 mm nylon screen was used to filter out any coarse particles. The specimens with botanical characterizations are in Table 2.

2.3. Analysis of fruits and vegetable

For the analysis of vegetables and fruits, a 100 ml acid-washed beaker containing one gram of material was filled with 15 ml of a di-acid mixture (Allen et al., 1986). Then digested at 80 degrees Celsius until a transparent solution was obtained. After cooling, the digested samples were filtered through Whatman No. 42 filter paper, and the filtrate was subsequently diluted to make 50 millilitres using deionized water in a volumetric flask. Using AAS, the heavy metals were found in the vegetable filtrate and air deposition. Using the standard solutions of varied concentrations of the appropriate heavy metals, the instrument was calibrated. Air served as the support and acetylene gas served as the fuel. Each instance involved the work of an oxidising flame, which reveals various settings for the instrument depending on the metal.

2.4. Method validation

The analytical process was validated using various metrics, such as accuracy, precision, and limit of detection.

2.5. Daily Intake of Heavy Metals through Fruits and Vegetables

The daily intake of heavy metals through the consumption according to the equation (Cui et al., 2004).

Daily intake of heavy metals $\mu\text{g day} = [\text{Daily fruit or vegetable consumption} \times \text{fruit or Vegetable heavy metal concentration}]$

3. Results and discussion

As a result of their toxicity, heavy metals are regarded as the most significant environmental pollutant. Table (4) illustrates the number of heavy metals in a few fruits and vegetables from five different Chennai, Tamil Nadu marketplaces, along with a comparison to the WHO's permitted guideline (Table 5) for heavy metals (Fig. 4 & Fig. 5)

3.1. Lead concentration in fruits and vegetables

Lead could enter the human system through food, drink, and air. No known safe level of exposure exists. threshold because it has adverse effects on human metabolism, is dangerous even at low doses, and has no known beneficial effects. It is mostly stored in both human and livers and kidneys. The toxicity of daily lead intakes is very low at the determined concentration levels (Ara et al., 2015).

According to dry weight of the sample, the amounts of Pb were estimated. The Pb concentrations results with below detectable limits (BDL) for all the vegetable and fruit samples of all the sites (Table 4) which is well within the 0.3 mg/kg permissible range (Table 5) .The levels of lead identified in a variety of greens collected from markets in Tanzania, Saudi Arabia, Nigeria, and Serbia were less than the highest amounts allowed (0.30 and 0.10 mg kg⁻¹ for greens) (Hussain et al., 2019) and also with the vegetable field Nigerian urban wastewater that was used for irrigation contained a permissible amount of lead level within an acceptable threshold (Ogunkunle et al., 2014). Air, water, and food all have pathways for lead to get into the human circulatory system. But this decreased level of lead in the study area is because, Pb is the least transportable element from the soil, and soil Pb concentrations are highly correlated with minerals, oxides of manganese, hydroxyls ions of iron and aluminium, and organic matter in soils (Li et al., 2022). Whereas the soils

near the industrial areas could not be rich in organic matter and hence the insufficient level of Pb (Stefanowicz et al., 2020). The daily intake of Pb is also at BDL from the study area of the selected vegetables and fruits. Also, the lead concentration in the mango fruit was less than the instrument detection limit, indicating that either no lead was present in the sample or only a trace amount was (Pant et al 2020). According to numerous reports, using whole fruits-including their peels-carries a risk of consuming heavy metals that may be found in the peel. (Czech, et al., 2021). The evaluation of health risks is within the hazard quotient level with less than 1 as per Zhang et al. (2018).

3.2. Nickel concentration in fruits and vegetables

Based on the collected samples of vegetables and fruits, nickel concentration shown in (Table.4) but all the concentrations were below the permissible limit of Ni (67.9 mg/Kg) in all the samples from the study area. This corroborated with the findings of (Bukhari et al., 2013) where the Nickel concentration of in Pakistan had 3-12 mg Ni kg⁻¹ and also the Ni levels varied between 0.05 and 0.24 mg/Kg in Jews mallow and highest observed in fluted pumpkin plant 0.01 - 0.1 to 0.23 and 0.25. However, Ni level of 0.067 mg/Kg for Indian Basil have been reported by Divrikli et al., 2006 which is within the range of values obtained from this study. Wastewater irrigation of vegetables in Morocco also resulted in 85 mg Ni kg⁻¹ (Al-Jaboobi et al., 2014). In India, vegetables grown in peri-urban areas and irrigated with untreated wastewaters had Ni concentration as high as 506 mg Ni kg⁻¹ (Yadav et al., 2013). Nickel in the soil at less than pH 6.7 is in soluble form and most Ni exists as insoluble hydroxides. So, the Ni concentration would be in insoluble form so that the uptake is less (Bhalerao et al., 2015).

3.3. Cadmium concentration in fruits and vegetables

Extended cadmium accumulation in humans can result in several health problems, including kidney damage, renal disorders, and human carcinogens. Due to cadmium buildup in the body, prolonged ingestion of several of these fruits and vegetables causes health issues (Rahimzadeh et al., 2017). Even at low concentrations, Cadmium is an extremely hazardous metal which forms up in internal organs and could result in fatal conditions (Genchi et al., 2020). The level of Cd is determined to be BDL in all the

vegetables and fruits from the sites taken for the study area (Table 5) which falls in the acceptable range of 0.2 mg/Kg (Table 4) which correlates with the findings of Ezeilo et al., 2020 that the majority of the three regions fruits and vegetables had cadmium concentrations that were within the acceptable range of 0.2 mg/kg. (Ezeilo et al., 2020) and also the vegetables and fruits spices analysed for the Cd in Nigeria were found to be within the limit (Inam et al., 2013). The results are in line with the research undertaken in different parts of the world. (Table 3). Fruits often have lower levels of pollutants in their edible sections than vegetables. The only explanation for this would be that vegetable fields tend to be more contaminated than orchards. Furthermore, this is a result of physiological barriers that prevent harmful heavy metal(loid)s from entering phloem, being loaded or unloaded, or accumulating in fruits (Nazir et al., 2015) were found to be under the WHO/FAO and Indian acceptable limits (Mawari et al., 2022) and (Table 3). Another important criterion for low Cd transportation into the produce is certain soil characteristics, including soil pH and organic matter that can have an impact on the sequences of Cd speciation in soils (Li et al., 2021). According to Wang et al. (2021) increased pH result in decreased concentration of Cd in plants due to decreased Cd availability in soils. According to Adriano et al. (2004), organic matter in alkaline industrial waste water intruded soils easily forms soluble Cd complexes and prevents exchange of ions and transfer to roots because most of the industries of study area were observed to let alkaline effluents into the soils and water nearby.

3.4. Chromium Concentration in fruits and vegetables

Cr (VI) and its metabolites, especially chromate, enter the human body through different ways (inhalation, ingestion and skin contact), which will cause pathological changes to the human organs and systems (respiratory tract, skin, gastrointestinal tract and so on) and even increase the incidence rate and mortality of many cancers (Christou et al., 2021). Prolonged human exposure can cause gastrointestinal upset, respiratory problems, kidney and liver damage, and altered genetic material, among other conditions (Ahmed et al., 2021). At modest levels, Cr (VI) can be acutely hazardous and is recognised as a known human respiratory carcinogen.

Within the selected fruits, the Cr concentrations were found to be BDL in the study area and also within the permissible limit (Table 5) was correlated with from the study of Dhaka and Chattogram 2021, Pomelo and star fruit had the highest chromium values, at 0.217 mg kg⁻¹ respectively but Mangoes from the Chattogram region and jackfruit from both regions showed no signs of chromium. Therefore, the local population is safe and no such urgent measurements are needed (Islam et al., 2022) because of even at low concentrations the Cr is toxic in nature exposure to Cr can cause lung, kidney, or liver damage. The uptake is BDL in the selected vegetables and fruits may be due to the chromium behaviour in ion exchange capacity in soil. Since Cr is less mobile, due to its adsorption rate, Cr⁺⁶ reduces as pH rises (Bamigboye et al., 2021).

3.5. Daily intake of vegetables and fruits

The provisional tolerated daily intake (PTDI) is typically used to express consumer exposure and the associated health hazards. The World Health Organisation has established a heavy metal intake threshold determined by the typical adult's body weight, is 60 kg. Vegetable and fruit intake per person per day is 98 and 78 grams, respectively. From the results of the study, all the samples were found to have below detectable limit and do not pose any threat to humans. When compared with the permissible limits. For an average person, the calories of fruit contribute 36.89 g, 5.54 g, 0.205 mg, 0.288 mg, 45.24 g, and 0.116 mg of (Pb, Cd, Zn, Cu, Co, and Ni) heavy metal intake, respectively. In case of vegetables, the equivalent shown daily intake will be 24.8 g, 13.3 g, 0.8 mg, 0.33 mg, 49.7 g, and 0.0231 mg, if Pb, Cd, Zn, Cu, Co, and Ni intakes are 0.25, 0.14, 8.15, 3.36, and 0.51 mg/kg, and 0.24 mg/kg Elbagerm et al., 2012. Therefore, heavy metal contamination in industrial area is more likely than food crops cultivated in isolated locations. Fruits and vegetables that had been treated had lower heavy metal contents (Chen et al., 2021). Since the heavy metal pose detrimental influence on human health as well as the nutritional properties of agricultural goods; they must comply within WHO's specified safety criteria and there must be sufficient control over the levels in food (Abdurrahman et al., 2022).

4. Conclusion

The findings of the research revealed that concentrations of toxic heavy metals were below detectable levels of Cd, Ni, Cr and Pb in all the fruits and vegetables samples collected from the study area. This study indicated that heavy metals did not enter the produce from the research zone through polluted air/ smoke and dust emanating from the industries and vehicular emissions and dust due to severe traffic congestion near the market sites. The daily intake of the metals from the produces were also analysed to be below detectable limit. The hazards related to toxicity of contaminants in vegetables and fruits should be taken into account for the local people, as these vegetables and fruits are among the most popular nourishment consumed in the study area. Further research may be carried out extensively and periodically in the markets near the other industrially polluted areas of Tamil Nadu to ascertain the food safety levels as per the norms laid down by the Food Safety and Standards Authority of India.

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Table.1. Features of the Chennai market sample sites

S.No.	Study sites	Type of markets	Land uses
1.	Thiruvanmyur	Local	HT
2.	T. Nagar	Local	HT
3.	Ambattur	Local	NIA
4.	Red Hills	Local	NIA
5.	Minjur	Local	NIA

HT- Heavy traffic; NIA- Near Industrial Area

Table 2. Botanical description of the vegetable and fruits used in the study

S.No.	Local name	Botanical description
1.	Grapefruit	<i>Vitis vinifera</i>
2.	Apple	<i>Malus pumila</i>
3.	Guava	<i>Psidium guajava</i>
4.	Orange	<i>Citrus sinensis,</i>
5.	Amla	<i>Phyllanthus emblica</i>
6.	Lemon	<i>Citrus limon</i>
7.	Beans	<i>Phaseolus vulgaris</i>
8.	Tomato	<i>Solanum lycopersicum</i>
9.	Potato	<i>Solanum tuberosum</i>
10.	Carrot	<i>Daucus carota</i>
11.	Cucumber	<i>Cucumis sativus</i>

Table 3. Observations on heavy metals in fruits and vegetables

S.No.	Study area	Fruit & vegetable	Ni	Cd	Cr	Pb	References
1.	Netherlands	Apple, apricot, lemon, orange	-	-	-	0.020 mg kg ⁻¹ (dry weight)	Namiesnik et al., 2012
2.	India	Apple,	-	-	-	224.4	Muhammad Rahim

		Grapes, Orange				ug l-1 (juice)	et al., 2020
3.	Maharashtra	mango	-	-	-	-	Ivan Siric et al., 2022
4.	Uttar Pradesh	Tomato	0.89	0.14	-	0.46	Gupta et al., 2021
5.	Vellore	onion		0.073	-	-	Bharani et al., 2015
6.	Nigeria		8.90	3.08		8.90	Sobukola et al., 2010

Table 4. Limit of heavy metals permitted in fruits and vegetables

S.No.	Element	FAO/WHO (mg/Kg)
1.	Cd	0.2
2.	Pb	0.3
3.	Ni	67.9
4.	Cr	-

*FAO: Food and Agriculture Organization *WHO: World Health Organization

Table.5 Concentration of Cr in vegetables

Vegetables & fruits	Location																			
	T. Nagar				Ambattur				Red Hills				Minjur				Tiruvanmyur			
	Ni	Cr	Cd	Pb	Ni	Cr	Cd	Pb	Ni	Cr	Cd	Pb	Ni	Cr	Cd	Pb	Ni	Cr	Cd	Pb
Beans	1.8 6	BD L	BD L	B DL	2.2 4	B DL	BD L	BD L	3.1 4	BD L	BD L	BD L	3.45	BD L	BDL	BD L	1.5 7	BD L	BD L	BD L
Tomato	13. 1	BD L	BD L	B DL	16. 2	B DL	BD L	BD L	16. 32	BD L	BD L	BD L	15.2 2	BD L	BDL	BD L	10. 3	BD L	BD L	BD L
Potato	18. 3	BD L	BD L	B DL	21. 5	B DL	BD L	BD L	20. 2	BD L	BD L	BD L	26.1 4	BD L	BDL	BD L	11. 6	BD L	BD L	BD L
Carrot	14. 2	BD L	BD L	B DL	16. 3	B DL	BD L	BD L	17. 25	BD L	BD L	BD L	1.12	BD L	BDL	BD L	1.3	BD L	BD L	BD L
Cucumber	2.6 1	BD L	BD L	B DL	2.1 9	B DL	BD L	BD L	2.0 5	BD L	BD L	BD L	1.47	BD L	BDL	BD L	1.4 9	BD L	BD L	BD L
Grapes	1.8 7	BD L	BD L	B DL	2.1 3	B DL	BD L	BD L	1.4 4	BD L	BD L	BD L	3.12	BD L	BDL	BD L	1.6 3	BD L	BD L	BD L
Apple	1.5 4	BD L	BD L	B DL	1.3 3	B DL	BD L	BD L	1.3 8	BD L	BD L	BD L	1.02	BD L	BDL	BD L	2.0 1	BD L	BD L	BD L
Guava	1.8 8	BD L	BD L	B DL	1.4 2	B DL	BD L	BD L	1.4 7	BD L	BD L	BD L	1.33	BD L	BDL	BD L	1.2 4	BD L	BD L	BD L
Orange	2.2 6	BD L	BD L	B DL	3.5 2	B DL	BD L	BD L	0.8 7	BD L	BD L	BD L	3.28	BD L	BDL	BD L	BD L	BD L	BD L	BD L
Amla	1.2 2	BD L	BD L	B DL	1.1 7	B DL	BD L	BD L	0.9 8	BD L	BD L	BD L	1.74	BD L	BDL	BD L	BD L	BD L	BD L	BD L
Lemon	2.3 3	BD L	BD L	B DL	1.9 7	B DL	BD L	BD L	1.2 4	BD L	BD L	BD L	0.68	BD L	BDL	BD L	BD L	BD L	BD L	BD L

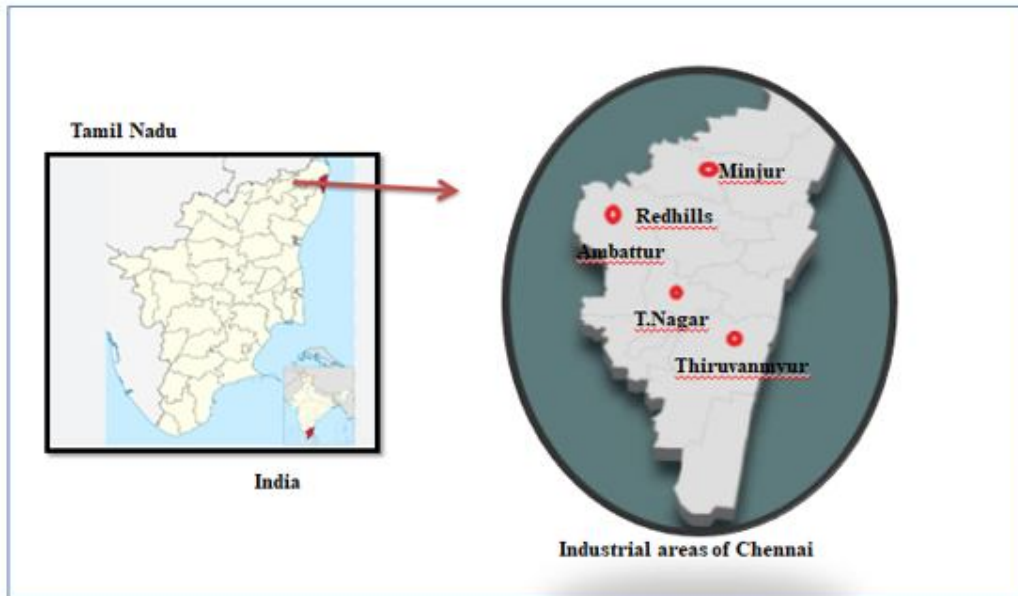


Fig.1.Study area

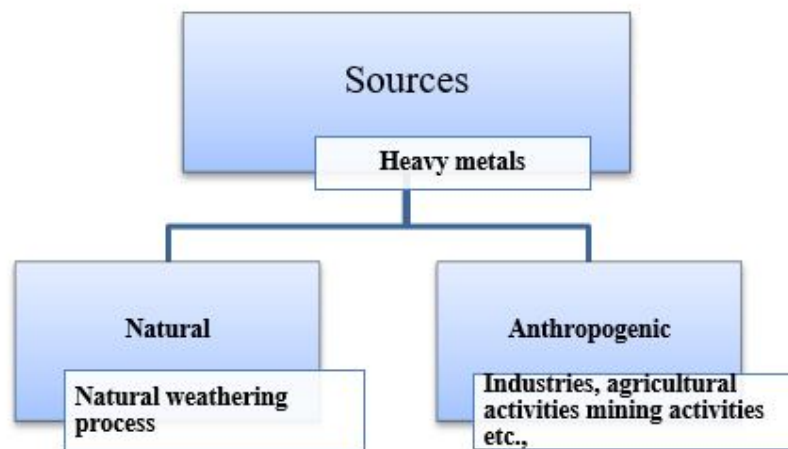


Fig.2. Sources of heavy metals

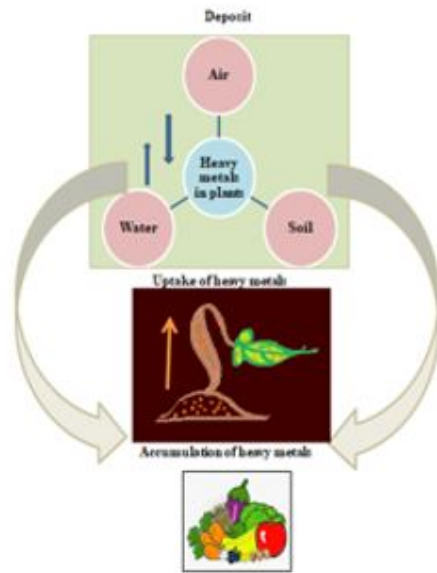


Fig.3. Heavy metal accumulation in fruits and vegetables from the environment

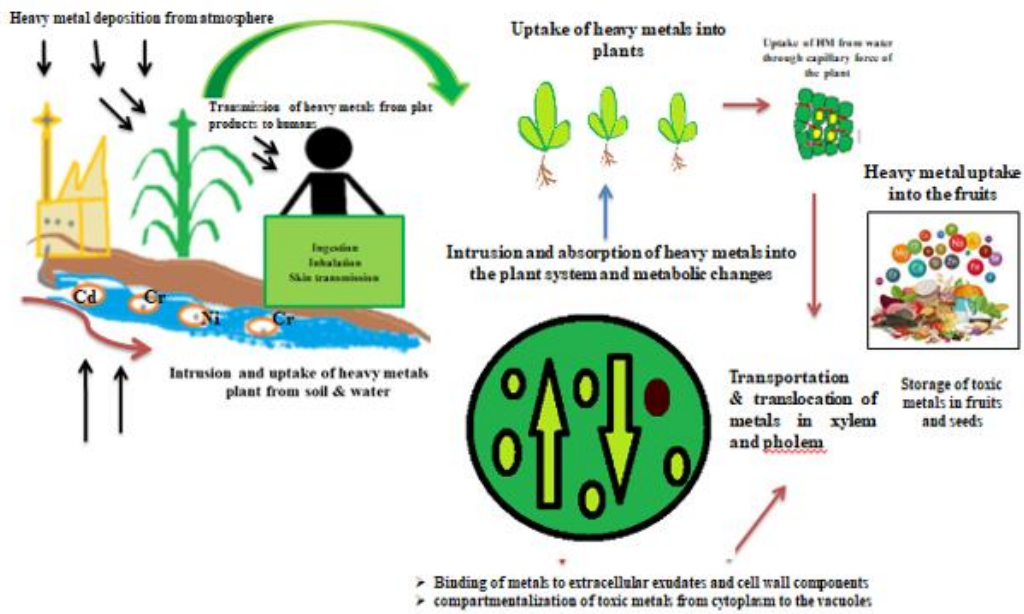


Fig.4. Heavy metal absorption in plants and transportation to fruits and vegetables

UNDER PEER REVIEW