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ASSESSMENT OF HEAVY METAL LEVELS ON FRUIT SURFACES: IMPLICATIONS FOR PUBLIC HEALTH

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Abstract

This study investigated the presence of heavy metal contaminants, including nickel (Ni), arsenic (As), lead (Pb), chromium (Cr), and cadmium (Cd) on the surfaces of dragon fruit and bananas obtained from Fulbari Gate Bazar in Khulna and Hazrahati Village in Kushtia, Bangladesh. The concentrations of these metals were analyzed on a dry weight basis and compared to the permissible limits established by Bangladesh and the World Health Organization (WHO). Several results indicated alarming levels of contamination, exceeding national (Bangladesh) and international (WHO) safety standards. The amount of nickel in the banana from Fulbari Gate was 2.11 mg/kg, which is higher than Bangladesh's standard of 0.1 mg/kg and the WHO standard of 0.07 mg/kg. Dragon fruits from both places contained far higher levels of arsenic (0.271 mg/kg and 0.253 mg/kg) against the Bangladesh standard of 0.01 mg/kg and the WHO standard of 0.05 mg/kg. Furthermore, the level of chromium contamination in dragon fruit from Fulbari Gate was recorded as 1.24 mg/kg, which greatly surpasses the permissible limits of 0.05 mg/kg recommended by both Bangladesh and the WHO. The HQ analysis identified arsenic and chromium as the two elements posing the highest health risks, with respective HQ values of 3.6328 and 1.781. Additionally, dragon fruit from Fulbari Gate had an HI value over 6.0, indicating a substantial overall health risk. The HPI values obtained for the local market samples were classified as "Bad" with HPI = 3.42, whereas that of organically raised fruits was classified as "Moderate". Hence, there is a need for checking, control, and reduction of heavy metal contamination to ensure public health safety.

Keywords

Fruit Safety; Heavy Metal; Hazard Quotient; Hazard Index; Heavy Metal Pollution Index

1. Introduction

For a healthy diet, fruits are essential as they provide essential nutrients and fiber. However, they can also contain harmful contaminants, such as food additives and heavy metals, which can threaten our health [1]. The term 'food additive' refers to any substance that is added to food during preparation, handling, processing, packing, shipping, or storing in order to become part of the food or change its properties. The US Food and Drug Administration or FDA defines food additives as "substances intended to be added to food, or to have an effect on its properties" [2]. Food additives can be classified according to their functions. Antimicrobial agents, such as sulfur dioxide, are used to preserve dried fruits [3]. Antioxidants, including vitamins C and E, BHA, BHT, and propyl gallate, effectively inhibit food spoilage and oxidation processes. Artificial colors enhance the visual appeal of food products by providing colors associated with specific flavors, such as red for cherry or green for lime [4]. Artificial flavors are added to give specific tastes or to enhance existing tastes, whereas flavor enhancers are those substances that tend to enhance or strengthen the flavor of food. Peroxides act as bleaching agents and are used to whiten certain foods, notably cheese and wheat flour. Chelating agents, like tartaric acid, malic acid, and citric acid, prevent discoloration, loss of flavor, and rancidity during food processing. By adding vitamins and minerals to foods, nutrition additives ensure that the food is nutritionally adequate. The release of heavy metals through human activities poses a significant environmental threat. The activities range from industrial releases to disposal of organic waste, rubbish burning, transportation, and power generation [5]. Particulate matters containing toxic metals are blown by winds over long distances and finally deposit into soils and water bodies through atmospheric deposition. Dissolved metal residues from industries, as well as drainage waters from mining and smelting, contaminate aquatic habitats [6]. Arsenic, mercury, lead, and cadmium are among the common heavy metals found in fruits. Arsenic is highly toxic and carcinogenic. Lead has various environmental and health effects. Mercury is very toxic to aquatic organisms due to its bioaccumulation nature. Cadmium, a byproduct of zinc production, contributes to oxidative stress in plants. Natural sources of heavy metals include volcanic emissions, sea salt, forest fires, biological sources, rock weathering, and suspended soil particles in the wind. Metals pollute water bodies by releasing in a variety of forms, altering water chemistry and affecting aquatic life. Long-term exposure to heavy metals such as lead, chromium, mercury, cadmium, nickel, arsenic, zinc, and copper can also be toxic to organisms even in their trace levels [7]. Fruits are a vital component of a healthy diet since they offer several of the nutrients required for proper health. However, the presence of heavy metals on the surface of fruits carries substantial health hazards, such as neurological damage, cancer, and kidney disease [5]. Therefore, it is of utmost importance to comprehend the levels of heavy metal concentrations on fruit surfaces in order to assess potential health risks and guarantee adherence to food safety regulations. This study aims to (i) detect and measure the concentrations of heavy metals on the surfaces of fruits and (ii) compare naturally ripened, preservative-free fruits with those obtained from local markets.

2. Materials and Methods

2.1 Study Area

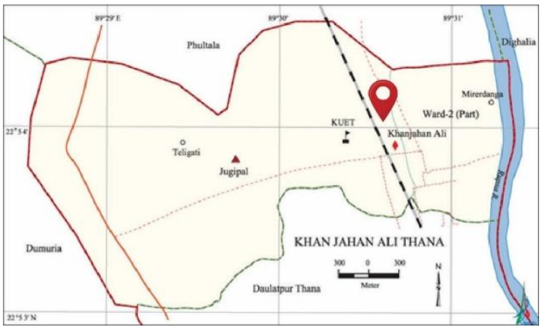

Fig. 1: Map of Khan Jahan Ali Thana and pointer shows Fulbari gate bazar [8]

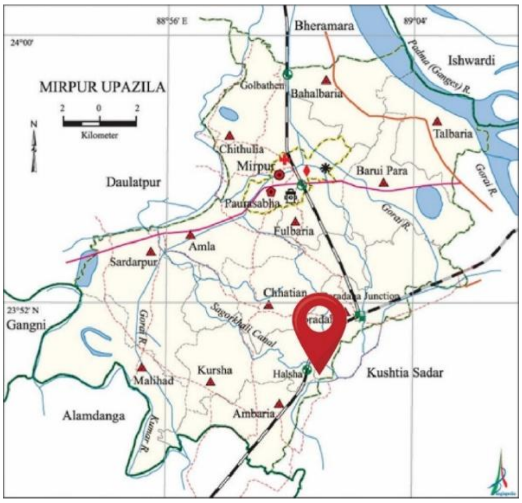

Fig. 2: Mirpur Thana and pointer shows Hazrahati village [9]

In this experiment, the fruits were collected from two different places. The fruits from the local market were collected from Fulbari Gate bazar of Khan Jahan Ali Thana. Fruits from organically cultivated fields were collected from Hazrahati village in Mirpur Thana, Kushtia.

2.2 Selection of Sampling Sites

The sampling sites were numbered sequentially from S-1 to S-4, with S-1 being the first location sampled. The locations were selected on the basis of their prominence as sources of fruits used for human food in that region.

Table 1: Table for sample collection area and sample number

Area	Fruit	Sample No.
Fulbari gate (Local market)	Banana	S-1
	Dragon fruit	S-2
Hazrahati (Organic)	Banana	S-3
	Dragon Fruit	S-4

2.3 Sample Collection

The fruits gathered from the sampling sites on November 10th, 2023, were labeled as S-1 to S-4. Two types of fruits at each sampling site were collected and packed into airtight polyethylene bags. All such bags were properly sealed to avoid contamination.

2.4 Sample Preparation

The sample preparation was performed systematically to ensure accuracy in the analysis with minimal contamination. Fresh fruit samples were collected from the field and followed by proper washing using deionized water to remove impurities on the surface of fruits. After that, washed fruits were spread out on tissue paper and then left for 2 hours for soaking so that some amount of water could be absorbed into the plant tissues. Afterwards, these samples were put into clean dry glass beakers. The samples were then dried in the electric oven at 105°C for 24-48 hours for moisture and solid content analysis. After drying and cooling, the samples were placed in a desiccator and weighed to determine moisture and solid content. After weighing, the samples were put in poly bags and stored at room temperature. To prepare the samples for analysis, they were ground using a mechanical grinder machine and sieved through a 0.2 mm screen to ensure uniform particle size.

2.5 Sample Digestion

In the present study, the digestion procedure was adopted from EPA 3050B to ensure that the preparation of the sample would occur in a uniform and reliable manner. The sample was digested according to a strict protocol to minimize errors and interference. 0.5 g to 5.0 g of the sample was transferred into a 100 ml borosilicate glass beaker, previously cleaned before use to prevent contamination. 15 ml of concentrated nitric acid HNO_3 was added to the sample then a watch glass covered the beaker and left overnight. During this step, the sample matrix broke down, and the components became solvated. On the following day, this sample was heated on a hotplate at 120°C for 4 hours until the volume reduced to 10 ml. This step is important to ensure the complete digestion of the sample and the border removal of volatile compounds. On cooling, 5 ml of concentrated nitric acid was added to the sample and heated on the hotplate at 120°C for 2 hours until the volume is reduced to 10ml and white fumes appear. This step ensures complete oxidation of the sample and removes all organic matter. 3.0 mL of 30% hydrogen peroxide and 2.0 mL of deionized water were added to the sample. This was then heated on the hotplate until it reached 120°C, or until the effervescence subsided. This oxidizes any remaining organic matter and fully digests the sample. Letting the sample to cool after which the peroxide reaction was repeated for full digestion or almost 30 minutes of heat depending on how much effervescence was present. The sample beaker was then rinsed with 2 ml of 1:9 nitric acid and deionized water. For the arsenic test, 2 ml of 1:1 hydrochloric acid should be used. The sample was filtered using Whatman-41 filter paper into a 100 ml volumetric flask. The filter paper was washed with 1 N hydrochloric acid. The residue was then rinsed with deionized water and acid-deionized water up to the 100 ml mark. This is the stock solution for metal analysis by the AAS method in soil and plant samples. Finally, the sample was shaken well and transferred to 100 ml plastic sample bottles with proper labeling. The sample was kept in a refrigerator at 4°C for preservation and storage prior to analysis.

2.6 Assessment of Human Health Risk

2.6.1 Daily Intake of Metal (DIM) and Health Risk Index (HRI)

The metal intake per day was calculated by the use of the formula to find out human exposure dose [10]:

$$\text{DIM} = \frac{D_{fruit\ intake} \times C_{metal}}{B_{average\ wait}} \qquad (1)$$

Where, C_{metal} is the concentration of heavy metals in fruits (mg/kg) and $D_{fruit\ intake}$ represents the daily intake of fruits (average fruit intake = 0.280 kg/person/day). An average body weight of 65 kg per person has been taken into consideration. The dose-response assessment was estimated by considering the oral reference dose (R_fD), and the HRI was calculated by dividing the estimated exposure of the test fruit to the oral R_fD. In health risk assessment, this index is also referred to as the Hazard Quotient (HQ).

$$HRI = DIM/R_fD \qquad (2)$$

The oral reference doses (R_fD) for Ni, As, Pb, Cr, and Cd are 0.0003, 0.001, 0.02, 0.003, and 0.004 mg/kg/day respectively. In order to assess the human health risk from exposure to multiple heavy metals, a Hazard Index was calculated using the following formula index [10]:

$$HI = \sum HQ = HQ_{Pb} + HQ_{Cd} + HQ_{Cr} + HQ_{As} + HQ_{Ni} \qquad (3)$$

HI value more than 1 means higher likelihood of non-carcinogenic health effects and this likelihood increases with increasing HI value.

2.6.2 Heavy Metal Pollution Index (HPI) Calculation

The HPI is a designed tool that enables the assessment of fruit quality with respect to its concentration of heavy metals and the potential health effect of specific trace metals. HPI uses the weighted arithmetic mean of fruit quality parameters against standards set by the WHO for heavy metals. HPI was calculated using the formula [11]:

$$HPI = \frac{\sum_{i=1}^{n} Q_i \times W_i}{\sum_{i=1}^{n} W_i} \qquad (4)$$

where W_i is the ith parameter's unit weight and $W_i = K/S_i$, where K is the proportionality constant, and n and i are the number of parameters taken into consideration.

$$K = \frac{1}{\sum_{i=1}^{n} \frac{1}{S_i}} \qquad (5)$$

Here, S_i denotes the permissible range of values, I_i is the ideal value, and Q_i is the sub-index of the ith parameter in the monitored values, calculated as:

$$Q_i = \sum_{i=1}^{n} \frac{(M_i - I_i)}{(S_i - I_i)} \times 100 \qquad (6)$$

2.6.3 Heavy Metal Evaluation Index (HEI) Calculation

The HEI was computed to assess the quality of fruits with respect to the heavy metals and trace elements present therein by using the equation [12]:

$$HEI = \sum_{i=1}^{n} \frac{M_i}{S_i} \qquad (7)$$

where Si is the standard value of ith parameter and M_i is the monitored value.

2.6.4 Quality Index (QI)

This study employs the Quality Index (QI), recognized as one of the most accurate methods for assessing pollution levels in the two types of harvested fruits. The QI determination involved several steps. First, the relative weight (Wi) of each parameter was calculated using the equation:

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i} \qquad (8)$$

where n represents the total number of parameters, and w_i denotes the weight of each parameter. Obtained results included the quality rating Q_i, and the concentration each parameter C_i in mg/kg. Q_i was evaluated using this formula:

$$Q_i = \frac{C_i \times 100}{S_i} \qquad (9)$$

Where S_i is deduced through WHO fruit quality standard. Finally, QI was calculated using the formula below [13]:

$$QI = \sum W_i \times Q_i \qquad (10)$$

Table 2: Classification of fruit quality based on QI values

Ranking	<100	100-200	200-300	300-400	>400
Fruit Quality	Excellent	Good	Poor	Very poor	Likely not suitable for eating

3. Results and Discussions

3.1 Concentration of Heavy Metal in Fruits

The concentration of some heavy metals in dragon fruit and banana sampled from different locations of the study area showed some important data on nickel, arsenic, lead, chromium, and cadmium. The metal content was determined on a dry weight basis by referring to the standards of Bangladesh and the World Health Organization.

Table 3: Heavy metal content in fruits measured by Atomic Absorption Spectrometry (AAS)

Collection point	Sample ID	Source	Conc. of Ni (mg/kg)	Conc. of As (mg/kg)	Conc. of Pb (mg/kg)	Conc. of Cr (mg/kg)	Conc. of Cd (mg/kg)
Fulbarigate, Khulna	S-1	Dragon fruit	1.98	0.271	0.012	1.24	0.003
	S-2	Banana	2.11	0.11	0.008	1.15	0.004
Hazrahati, Kushtia	S-3	Dragon fruit	0.312	0.253	0.004	0.987	0.0035
	S-4	Banana	1.37	0.183	0.009	0.786	0.0035
WHO standard (mg/kg)			0.07	0.05	0.05	0.05	0.003
Bangladesh standard (mg/kg)			0.1	0.1	0.1	0.05	0.0005

The measured nickel content exceeded the allowable limit in fruits obtained from Fulbari Gate, Khulna, and Hazrahati, Kushtia. Dragon fruit from Fulbari Gate contained 1.98 mg/kg, and in banana, it was 2.11 mg/kg, higher than the Bangladesh standard value of 0.1 mg/kg and the WHO standard value of 0.07 mg/kg. The level of arsenic in dragon fruits obtained from Fulbari Gate and Hazrahati were 0.271 mg/kg and 0.253 mg/kg, while in banana, it was 0.11 mg/kg and 0.183 mg/kg, higher compared to the Bangladesh standard value of 0.01 mg/kg and the WHO standard value of 0.05 mg/kg. Lead concentrations were lower but still exceeded the Bangladesh standard of 0.01 mg/kg, with dragon fruit from Fulbari Gate at 0.012 mg/kg and bananas at 0.008 mg/kg. Chromium levels in dragon fruit and bananas from Fulbari Gate and Hazrahati exceeded the permissible limit of 0.05 mg/kg. Cadmium concentrations in dragon fruit and bananas from both locations slightly exceeded the WHO standard of 0.003 mg/kg and significantly surpassed the Bangladesh standard of 0.0005 mg/kg. The results indicate heavy metal contamination at a significant level in both dragon fruits and bananas from the locations studied, which may pose potential health risks to the consumer.

3.2 Assessment of Public Health Risks

3.2.1 Daily Intake of Metals (DIM)

The DIM was computed to build the quantitative assessment of human exposure through the consumption of fruit to these heavy metals. The table below presents the DIM values for heavy metals such as nickel (Ni), arsenic (As), lead (Pb), chromium (Cr), and cadmium (Cd) in fruits collected from Fulbarigate Bazar in Khulna (local market) and Hazrahati in Kushtia (organic cultivation). For dragon fruit from the local market, the DIM values are as follows: 0.001344 mg/day for Ni, 0.00109 mg/day for As, 1.72E-05 mg/day for Pb, 0.004252 mg/day for Cr, and 1.51E-05 mg/day for Cd. As for bananas, the DIM values are 0.00590154 mg/day for Ni, 0.000788 mg/day for As, 3.88E-05 mg/day for Pb, 0.003386 mg/day for Cr, and 1.51E-05 mg/day for Cd. Fruits from Hazrahati show different DIM values for dragon fruit: 0.00852923 mg/day for Ni, 0.001167 mg/day for As, 5.17E-05 mg/day for Pb, 0.005342 mg/day for Cr, and 1.29E-05 mg/day for Cd. DIM values of bananas from Hazrahati are 0.00908923 mg/day for Ni, 0.000474 mg/day of As, 3.45E-05 mg/day for Pb, 0.004954 mg/day

Table 4: DIM for fruits collected from local market and Hazrahati

Heavy metals	**DIM mg/day person**				
	Ni	As	Pb	Cr	Cd
Fruits collected from local market(Fulbarigate Bazar, Khulna)					
Dragon	0.001344	0.00109	1.72E-05	0.004252	1.51E-05
Banana	0.00590154	0.000788	3.88E-05	0.003386	1.51E-05
Fruits collected from Hazrahati, Kushtia					
Dragon	0.00852923	0.001167	5.17E-05	0.005342	1.29E-05
Banana	0.00908923	0.000474	3.45E-05	0.004954	1.72E-05

for Cr, and 1.72E-05 mg/day for Cd. These results reveal higher DIM values for Ni and Cr in fruits from Hazrahati, indicating greater exposure to these heavy metals through organically cultivated fruits. Conversely, the DIM values for As, Pb, and Cd are lower in fruits from Hazrahati compared to the local market.

3.2.2 Hazard Quotient (HQ) and Hazard Index (HI) Analysis

Table 5: Calculated values of Health Quotient (HQ)

Collection Point	**Fulbarigate**		**Kushtia**	
Parameters	HQ of Dragon	HQ of Banana	HQ of Dragon	HQ of Banana
Ni	0.0672	0.426	0.295077	0.45446
As	3.6328	3.891	2.627692	1.57949
Pb	0.0043	0.013	0.009692	0.00862
Cr	1.4172	1.781	1.128615	1.65128
Cd	0.0151	0.013	0.015077	0.01723

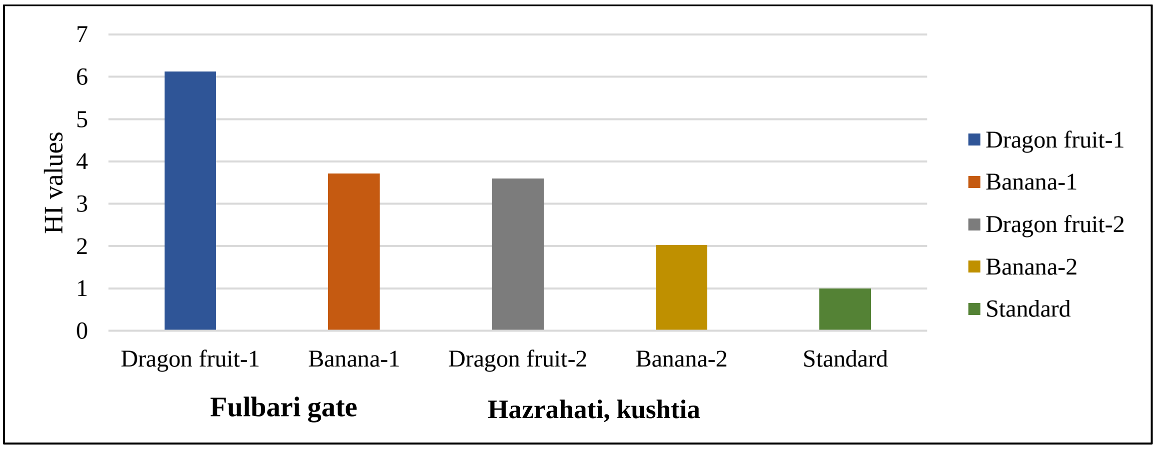

Fig. 3: Health index of various samples

As indicated in Table 5, the HQ (Hazard Quotient) values for arsenic (As) and chromium (Cr) were remarkably high in fruits obtained from Fulbari Gate and Hazrahati, Kushtia. The HQ for arsenic in the dragon fruit and banana from Fulbari Gate was represented by 3.6328 and 3.891, respectively, which may pose a potential health risk. Similarly, HQ value of chromium in these fruits were measured up to 1.4172 and 1.781, respectively. Fruits from Hazrahati also exhibited elevated HQ values, with arsenic levels of 2.627692 in dragon fruit and 1.57949 in banana, and chromium levels of 1.128615 in dragon fruit and 1.65128 in banana. Fig. 3 visually represents the Hazard Index (HI) values for various fruit samples, demonstrating that dragon fruit from Fulbari Gate poses the highest overall health risk, with an HI value exceeding 6.0. The banana from Fulbari Gate and dragon fruit from Hazrahati also have high values for HI, indicating considerable health risks. The results represent the high extent of contamination by heavy metals in the fruits available at local markets and even in organically reared ones. Based on this finding, higher HQ and HI values would indicate a greater risk to health; thus, tighter monitoring and regulation would be needed to ensure that fruits are safe to consume.

3.2.3 Heavy Metal Pollution Index (HPI) Analysis

The HPI analysis has led to the assessment of their levels of pollution and associated health risks. According to Table 6, samples S-1 and S-2 collected from Fulbari Gate local market have been classified with 'Bad' heavy metal pollution levels with an HPI value of 3.42 for both. However, samples S-3 and S-4, collected from Hazrahati Village, have their degree of contamination classified as 'Moderate' with HPI values of 2.20 and 2.62, respectively.

Table 6: Calculated fruit HPI and classifications

Sample No	S-1	S-2	S-3	S-4
Classification	Bad (3.42)	Bad (3.42)	Moderate (2.20)	Moderate (2.62)

From the above results, it could be concluded that the fruit obtained from the local market had expansively higher levels of heavy metal pollution compared to the organically cultivated counterpart. It is seen that the status averred as 'Bad' corresponded to the fruit sample procured from the local market, which corresponds to a higher risk for consumers related to heavy metal exposure. Though the organically cultivated fruits showed contamination, the levels were comparatively lower.

3.2.4 Heavy Metal Evaluation Index (HEI) Analysis

The results for the HEI analysis brought out some important findings concerning the level of heavy metal contamination. According to Table 7, samples S-1 and S-2 from Fulbari Gate local market showed a very high degree of contamination, with HEI values of 59.74 and 56.83, respectively. However, samples S-3 and S-4 from Hazrahati Village posited at a moderate degree of contamination with HEI values of 30.33 and 40.29, respectively.

Table 7: Calculated fruit HEI and classifications

Sample No.	S-1	S-2	S-3	S-4
Classification	High (59.74)	High (56.83)	Moderate (30.33)	Moderate (40.29)

These results clearly indicate that there is greater contamination of the fruits from the local market compared to organically cultivated fruits. Higher HEI values observed for samples from the local market indicated a higher risk of heavy metal exposure through such fruits. Though moderate contaminated level in organically cultivated fruits was also shown, they were relatively safer. These findings underscore the fact that monitoring and control of heavy metal contamination in fruits are necessary not only for mitigating health risks for consumers but also for improving the quality of a food item.

3.2.5 Quality Index (QI) Analysis

QI analysis represented the overall assessment of the levels of contamination in fruit samples. From Table 8, the samples S-1 and S-2 collected from Fulbari Gate local market were classified as 'Very Poor' quality with a QI value of 332.6 and 342, respectively, indicating severe pollution. In contrast, samples S-3 and S-4 from Hazrahati Village were classified as 'Poor' quality, with QI values of 220.4 and 261.19, respectively.

Table 8: Calculated fruit QI and classifications

Sample No	S-1	S-2	S-3	S-4
Classification	Very poor (332.6)	Very poor (342)	Poor (220.4)	Poor (261.19)

Though lower in values, they still denote a considerable amount of pollution, reflecting that organically grown fruits cannot also be completely free of contamination. These results have brought to the fore the fact that unless tight controlling and monitoring measures are adopted, good-quality and safe fruit for consumption may not become available.

4. Conclusion

The analysis revealed significant levels of heavy metal contamination in dragon fruit and bananas from both local markets and organic cultivation sites, highlighting the pervasive nature of this issue. Nickel, arsenic, and chromium have been the most critical contaminants that often exceeded the safety standard. HQ and HI analyses further indicated considerable health risks, mostly due to exposure to arsenic and chromium. The HPI classified the local market fruits as 'Bad' and the organic fruits as 'Moderate,' proving the advantage of organic practices in bringing down contamination. The QI classified the local market samples as 'Very Poor,' while the organic samples were under the category as 'Poor,' thus representing the inverse levels of pollution. These findings raise concerns about the proper monitoring and regulation of the heavy metal contamination of fruits to ensure public health safety. Effective strategies for enforcement of stricter environmental controls, setting up of sustainable agricultural practices, and education of consumers create the basis for ensuring food safety.

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