



Lead (Pb) Contamination in Agricultural Products and Human Health Risk Assessment in Bangladesh

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Abstract Lead (Pb) is a widely occurring heavy metal employed in industrial products and hence released into the environment, causing several environmental health risk concerns. This study comprehensively surveyed the literature on Pb contamination in different agricultural foods and food products commonly consumed by Bangladeshi inhabitants and assessed associated cancer and non-cancer health risks. Cereals (i.e., rice, wheat and maize) contained very high concentrations of Pb among the selected food items, the highest was found in wheat ($4.04 \mu\text{g g}^{-1}$), while rice and maize were 2.22 and $1.43 \mu\text{g g}^{-1}$, respectively, that exceeded the maximum allowable concentration (MAC, $0.20 \mu\text{g g}^{-1}$) for

cereals. Vegetables contained higher Pb than the MAC of $0.01 \mu\text{g g}^{-1}$, except for mushroom, green banana, cauliflower and arum. In addition, pulses also contained a moderate amount of Pb; however, fruits contained a low level of Pb, except for mangoes. When examining spatial differences in Pb contamination, most districts exhibited high Pb content in cereals; however, vegetables of the Tangail district exhibited the highest Pb concentrations ($2.17 \mu\text{g g}^{-1}$), originating from industrial operations and vehicular emissions. In terms of human health risk assessment, it was observed that consumption of rice, zucchini, tesla gourd, sponge gourd, okra, drumstick lib, chili and cabbage might pose non-cancer health risks (THQs > 1); however, fruits and pulses do not pose any non-cancer health risks to Bangladeshi residents. Most of the cereals and vegetables showed a higher value than

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10^{-6} , indicating a potential cancer risk; however, fruits and pulses showed lower risk only marginally exceeding the lower allowable limit (i.e., 10^{-6}).

Keywords Lead pollution · Human health risk assessment · Cancer and non-cancer risks · Vegetable · Rice

1 Introduction

As a hazardous heavy metal, lead (Pb) exists in a divalent state and contributes to 0.002% of the Earth's crust (Arias et al., 2010). Except for natural sources, Pb is primarily emitted from several anthropogenic sources, including mining, smelting of ores, burning of coal, effluents from storage of battery industries, automobile exhausts, metal plating, leather tanning, finishing operations, fertilizers, pesticides, and additives in pigments and gasoline. Consequently, the widespread use of Pb in many parts of the world has resulted in several environmental concerns and associated human health risks (Kushwaha et al., 2018).

Pb and its congeners remain stable for long periods in soil and are difficult to dissociate, resulting in bioaccumulation in agricultural products and subsequent trophic transfer to humans (Ogwuegbu & Muhanga, 2005). Although plants accumulate a significant portion of lead ions from the soil, most of the Pb load remains concentrated in the roots, with limited translocation to the stems and leaves (Wińska-Krysiak et al., 2015). The restricted translocation of Pb from root to stem and leaves is primarily due to barriers to transport at the root epidermis and endodermis (Kabata-Pendias & Pendias, 1984). Despite these barriers, significant transport of Pb to aerial portions of plants is observed on contaminated land, with aerial portions including leaves, fruits, and seeds being bound for human consumption (Baylock & Huang, 2000).

The direct breathing of Pb-laden dust, dermal exposure to Pb-contaminated dust and soils, oral consumption of Pb-contaminated water, and food produced in Pb-contaminated areas are the main significant pathways of Pb entering the human body (Kumar et al., 2020). Additionally, Pb is a well-known toxin that has been linked to a variety of human health problems (Bellinger, 2011). For example, Pb affects

the renal, cardiovascular, bone hematopoietic, nervous, and reproductive systems (Flora et al., 2012). Besides, Pb triggers oxidative stress and increases the sensitivity to oxidative stress leading to higher estrogen levels as an essential risk factor for breast cancer (Kasten-Jolly & Lawrence, 2017). Furthermore, elevated Pb levels in the blood affect behavior, cognitive function, postnatal development, puberty delays, and hearing ability of infants and children (Kumar et al., 2020). Additionally, brain damage, anemia, anorexia, vomiting, and disease of the circulatory and nervous systems are noteworthy examples of the adverse effect of Pb contamination (Chakraborty et al., 2020).

Given these various significant health concerns from Pb exposure, assessing Pb consumption from common agricultural foods like vegetables, cereals, pulses, and fruits, is crucial to evaluate human health risk, particularly in Bangladesh, where vegetables, cereals, and pulses are extensively consumed as the staple food. Although previously a significant amount of research has examined the effect of heavy metal contamination in different food items in Bangladesh (Islam et al., 2015; Rahman et al., 2013; Shaheen et al., 2016), very few of them have focused on Pb contamination. In addition, most of the research to date was conducted in particular districts of Bangladesh, only reflecting localized Pb contamination in foods.

Recently, a review on Pb contamination in Bangladesh reported Pb-contaminated sites and Pb concentrations in the atmosphere, water, sediments, soil, vegetables, fish, and other foods, indicating severe Pb pollution in soil from industrial influences (i.e., unauthorized lead-acid battery recycling, lead smelting, paint industry) (Majumder et al., 2021). Among the foods, the review study indicated a high level of Pb in vegetables ($0.2\text{--}22.09\ \mu\text{g g}^{-1}$) and fish ($0.018\text{--}30.8\ \mu\text{g g}^{-1}$) (Majumder et al., 2021). However, little is known about the human health risks of consuming Pb-contaminated agricultural products, nor their spatial and temporal distribution. The human health risk assessment model provided by the United States Environmental Protection Agency (USEPA) is a widely used method to estimate the nature and probability of negative health effects to humans from contaminant exposure that helps to mitigate the potential human health risk. However, no such model has been applied to date to assess the human health

risks of consumption of Pb-contaminated agricultural products in Bangladesh. Besides, information on the spatial and temporal distribution of Pb in agricultural food products in Bangladesh has not been summarized to date, enabling a comprehensive assessment of Pb hotspots across the country. Such data will be useful to policymakers or regulators to identify contaminated areas so that appropriate decisions and potential remediative action can be taken.

Thus, the present review aimed to amass Pb concentrations in vegetables, pulses, cereals, and fruits grown in Bangladesh through a comprehensive literature survey to provide a holistic view of Pb concentrations in commonly consumed plant-based foods in Bangladesh. In addition, the temporal and spatial distribution of Pb concentration in food items will be quantified to identify temporal trends and districts at risk. Finally, the associated health risks to Bangladeshi inhabitants are assessed from the consumption of Pb-contaminated vegetables, pulses, cereals, and fruits.

2 Methodology

2.1 Data Collection

A literature survey was conducted through the “Web of Science” database using a topic search that found articles in the Web of Science Core Collection, Current Contents Connect, Chinese Science Citation DatabaseSM, Derwent Innovations Index, KCI-Korean Journal Database, MEDLINE®, Russian Science Citation Index, and SciELO Citation Index. The following keywords were used as search terms such as “Lead or Pb in Bangladeshi vegetables”, “Lead or Pb in Bangladeshi cereals,” “Lead or Pb in Bangladeshi fruits, and “Lead or Pb in Bangladeshi pulses” that returned 123, 60, 50, 15, 25, 11, 31, and 5 articles, respectively. The timeframe was selected from January 2000 to December 2020 covering 20 years, due to the unavailability of Pb concentration data in agricultural products before 2001.

From the initial research articles sourced, each was read closely and a subset of 30 (thirty) relevant research articles that contained data on Pb concentrations in vegetables, cereals, fruits, and pulses were employed. Pb concentration data were collected for

34, 5, 3, and 10 commonly consumed vegetables, pulse, cereal, and fruits species, respectively. A list of the commonly consumed food items in Bangladesh with their English and scientific name and the number of studies that reported Pb concentration in them are provided in Supplementary Table S1. The major criterion of data compilation was that the Pb concentration has been reported in text or tabular format in the published articles or corresponding supplementary section; however, data presented in graphical format was excluded due to the possibilities of inaccurate measurement.

2.2 Data Analysis

Data were sorted into species of individual food items, year, and district where the products were grown. The Pb concentrations were averaged for each species, year, and district to show their respective averages along with variation. In addition, for different types of samples in the literature, the quantification of Pb was expressed in either a dry weight or fresh weight basis. The dry weight was converted to fresh weight for all types of samples as people generally prefer fresh foods to eat and the unit was expressed as $\mu\text{g g}^{-1}$ (i.e., $\mu\text{g g}^{-1}$ fresh weight, unless otherwise specified) to make an easy comparison among different food items.

The following formula was used to convert dry weight into fresh weight (Cresson et al., 2017):

$$C_w = C_d \left(\frac{100 - \% \text{ Moisture content}}{100} \right) \quad (1)$$

where C_w and C_d are concentrations on a fresh and dry weight basis, respectively. An average of 79% moisture content in each of the vegetables and fruits (Saha & Zaman, 2013), and 11% and 13% of moisture content in respective pulses and cereals were considered (Ahmmed et al., 2020; Rahman & Naidu, 2020).

2.3 Human Health Risk Assessment

Human health risks were estimated for Bangladeshi adults in terms of the target hazard quotient (THQ) and target carcinogenic risk (TCR) from consumption of Pb in vegetables, fruits, pulses, and cereals. However, health risks were not estimated for turmeric, betel quid, and tobacco due to the scarcity of data related to their accurate consumption rate.

The THQ was used to estimate non-carcinogenic risks based on the U.S. Environmental Protection Agency (USEPA) Region III's Risk-based Generic Table (US-EPA, 2021). The THQs for Pb were estimated in vegetable, fruit, pulse, and cereal consumption and the formula used for calculating the THQ is as follows (US EPA, 1989):

$$THQ = \frac{EF \times ED \times FIR \times C}{BW \times RfDo \times AT} \times 10^{-3} \quad (2)$$

where THQ is the target hazard quotient, EF is the exposure frequency (365 day year⁻¹), ED is the exposure duration (70 years), FIR is the food ingestion rate (g day⁻¹), C is the metal concentration in foods (µg g⁻¹ fresh weight), RfDo is the oral reference dose (µg g⁻¹ day⁻¹), and AT is the averaging time for non-carcinogens (365 days year⁻¹ × number of exposure years). BW is the body weight (60 kg for adults in Bangladesh) (HIES, 2011). The RfDo of Pb is 0.0035 µg g⁻¹ day⁻¹ (US-EPA, 2019a). FIRs of Bangladeshi people for vegetables, fruits, rice, wheat, other cereals, and pulses were 166.1, 44.7, 416, 26, 21.9, and 14.3 g person⁻¹ day⁻¹, respectively (HIES, 2011). The 10⁻³ was a unit conversion factor. THQ > 1 indicates a chance of non-carcinogenic health risk, while THQ ≤ 1 indicates no possible health risk (US EPA, 2001).

Furthermore, the cumulative THQ of all food items for Pb consumption, also known as total THQ (tTHQ), were calculated as follows:

$$tTHQ = \sum THQ = \sum THQ_{Cereals} + \sum THQ_{Pulses} + \sum THQ_{Fruits} + \sum THQ_{Vegetables} \quad (3)$$

Cancer risk is the probability of an individual lifetime cancer-related health risk from Pb. The TCR (lifetime cancer risk) can be calculated using the equation provided in USEPA Region III Risk-Based Concentration Table as follows (US-EPA, 2019b):

$$TCR = \frac{EF \times ED \times FIR \times C \times SF}{BW \times AT} \times 10^{-3} \quad (4)$$

where SF is the oral carcinogenic slope factor from the Integrated Risk Information System database that is 8.5 × 10⁻³ (µg g⁻¹ day⁻¹)⁻¹ for Pb

(US-EPA, 2019a). Other terms have been discussed above.

There are no absolute criteria for the acceptable number of additional cancers over a lifetime period. However, the USEPA generally adopt 1 (one) additional case of cancer in 1 (one) million (i.e., 10⁻⁶) as a management goal for the government to suggest the point at which management decisions should be considered. The cancer risk surpassing 10⁻⁴ (1 case of cancer in 10,000 populations) is considered as unacceptable (US EPA, 2005).

3 Results and Discussion

3.1 Pb in Cereals

Rice, wheat, and maize are the major cereals consumed by Bangladeshi people, and Pb concentration was only reported for these dominant cereals to date. About 74% of cultivable land in Bangladesh is used by farmers for cereal production, particularly 69% for rice production, while only 2% and 3% are for wheat and maize production, respectively (Mahmud, 2019). All cereals contained a very high average concentration of Pb in Bangladesh, among which the highest amount reported (as fresh weight) in wheat was 3.24 µg g⁻¹, whereas rice and maize were 2.22 µg g⁻¹ and 1.43 µg g⁻¹, respectively (Fig. 1; Supplementary Table S2). The average Pb content in wheat, rice, and maize was about 20, 11, and 7 times higher than the MAC of 0.20 µg g⁻¹ (FAO/WHO, 2019). In comparison to other Pb-contaminated locales, wheat and maize grown in the vicinity of a former Pb smelter in northern France contained <0.19 and <0.19 µg g⁻¹ fresh weight of Pb, respectively (Douay et al., 2013), which is a much lower than this study. In Beijing, China, Pb content in rice was 0.09 µg g⁻¹ (Wei & Cen, 2020), which is about 25 times lower than in Bangladesh. Similarly, in Australia, rice contained about 0.33 µg g⁻¹ of Pb (Rahman et al., 2014), which is also about 7 times lower than that of Bangladesh. However, the respective Pb concentration in rice, wheat, and maize in Punjab, India, was about 17.13, 16.98, and 18.28 µg g⁻¹ (Sharma et al., 2018), which are several times higher than this study. Essentially, levels of Pb were highly variable

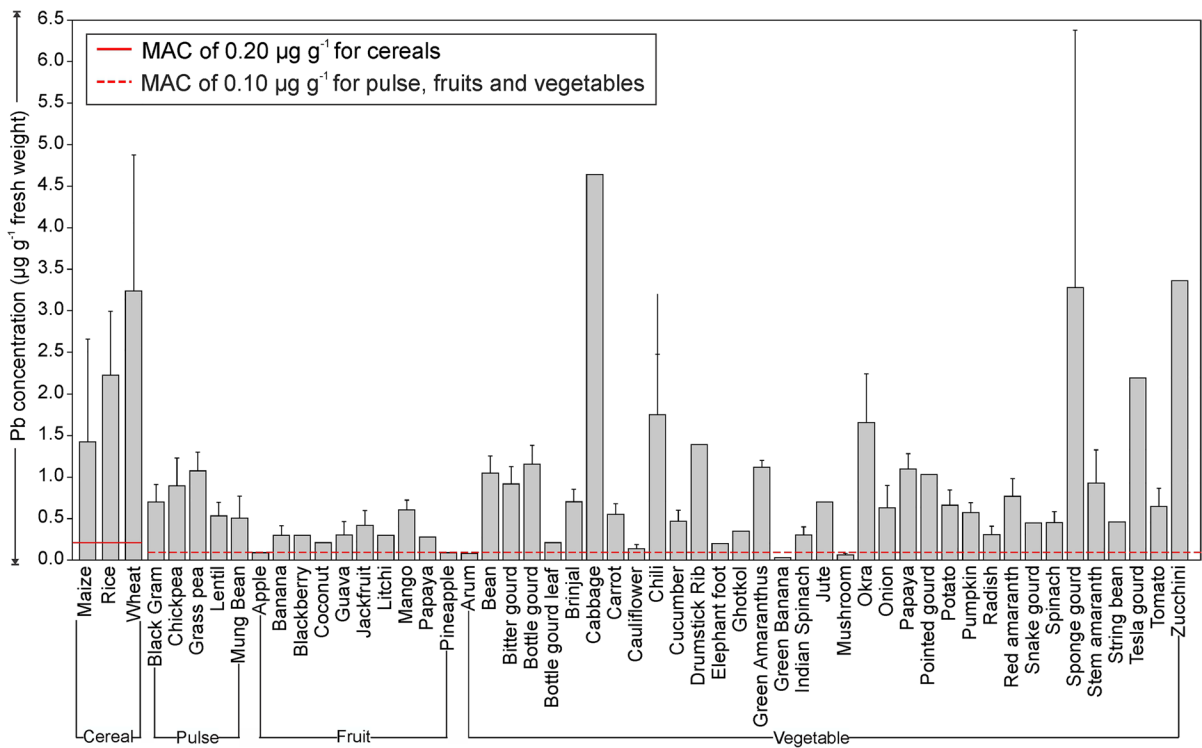


Fig. 1 Pb concentration ($\mu\text{g g}^{-1}$ fresh weight; average \pm standard deviation) in commonly consumed foods in Bangladesh. MAC for cereals ($0.20 \mu\text{g g}^{-1}$); MAC for pulse, fruits and vegetables ($0.10 \mu\text{g g}^{-1}$) (FAO/WHO, 2019)

spatially and depend on area of cultivation and contamination history.

In terms of Bangladeshi districts, the average Pb concentration in rice ranged from $0.08 \mu\text{g g}^{-1}$ (found in the Dhaka district) to $9.64 \mu\text{g g}^{-1}$ (found in the Chapai Nawabganj district) (Real et al., 2017; Saha & Zaman, 2011). The possible cause of the highest contamination in the Chapai Nawabganj district can be explained by Pb contamination of groundwater ranging from 0.06 to 0.16 mg L^{-1} , which is extensively used for irrigation purposes in that territory (Saha & Zaman, 2011). However, the second-highest Pb content in rice ($7.48 \mu\text{g g}^{-1}$) was reported in the Khulna district, which might be attributable to an abandoned lead-acid battery recycling workshop having Pb concentration in soil at $231 \mu\text{g g}^{-1}$ (Islam et al., 2019).

The average Pb concentration in wheat ranged between $<0.01 \mu\text{g g}^{-1}$ (found in the Mymensingh and the Chittagong district) and $7.78 \mu\text{g g}^{-1}$ (found in the Jhenaidah and Kushtia districts) (Kormoker et al., 2019; Zakir et al., 2021). The possible reason for the highest Pb concentration in wheat in

Jhenaidah and Kushtia districts is that the wheat samples were collected from the vicinity of metal smelting and battery industries, where Pb in soil was $19.2 \mu\text{g g}^{-1}$ (Kormoker et al., 2019). On the other hand, the average Pb in maize is relatively lower than other cereals in Bangladesh, it ranged between 0.04 and $6.35 \mu\text{g g}^{-1}$ in the Bogra and Jhenaidah-Kushtia districts respectively (Islam et al., 2014; Kormoker et al., 2019). Again, the highest values were observed in the Jhenaidah-Kushtia districts possibly attributable to the metal smelting and battery industry.

In contrast, in all three kinds of cereal, the cumulative average Pb concentration in Chapai Nawabganj was $8.07 \mu\text{g g}^{-1}$, whereas Khulna, Jashore, Jhenaidah-Kushtia, Tangail, Mymensing-Chittagong, Dhaka, Gazipur, and Bogra districts were 7.48 , 6.71 , 5.20 , 3.28 , 1.04 , 0.48 , 0.45 , and $0.12 \mu\text{g g}^{-1}$, respectively (Fig. 2; Supplementary Table S3). The highest Pb in cereals in Chapai Nawabganj district may be attributable to Pb contamination in groundwater used for irrigation purposes in that district (Saha & Zaman,

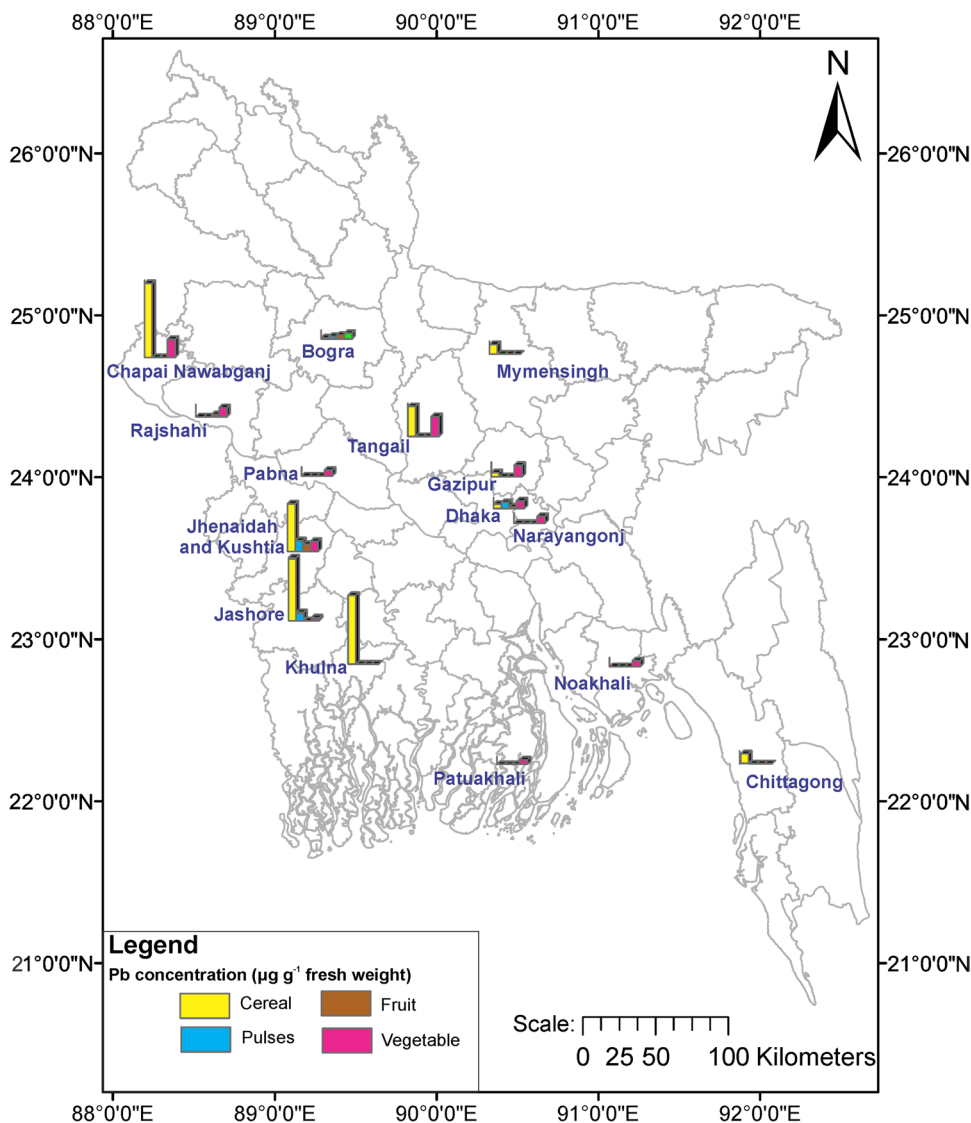


Fig. 2 Average Pb concentration ($\mu\text{g g}^{-1}$ fresh weight) in commonly consumed foods in different districts of Bangladesh

2013). However, in other areas, the increased Pb concentration in cereals might be attributed to the high level of Pb in soil due to metal smelting and other industrial activities.

3.2 Pb in Pulses

Several pulses like black gram, chickpea, grass pea, lentil, and mung bean are commonly used as foods in Bangladesh. Among these pulses, the highest Pb was found in grass peas ($1.08 \mu\text{g g}^{-1}$), while the lowest was found in mung bean ($0.51 \mu\text{g g}^{-1}$). The average

Pb in black gram, chickpea, and lentil was 0.70 , 0.90 , and $0.53 \mu\text{g g}^{-1}$, respectively (Fig. 1). However, the average Pb concentration in all of the pulses was much higher than the MAC value of $0.10 \mu\text{g g}^{-1}$ (FAO/WHO, 2019). In Yunnan, China, the Pb content in chickpea was $0.48 \mu\text{g g}^{-1}$, which is about half of Bangladesh (Wang et al., 2022); however, it was higher than the MAC value of $0.10 \mu\text{g g}^{-1}$. Similarly, a lower Pb content than Bangladesh and the MAC was reported in mung bean ($0.04 \mu\text{g g}^{-1}$) from the Swat valley of Pakistan (Khan et al., 2014).

Pb content in pulses was not commonly reported except in seven districts in Bangladesh, such as the Bogra, Dhaka, Jashore, Jhenaidah -Kushtia, and Mymensingh-Chittagong districts (Supplementary Table S3). Among these districts, the highest Pb in pulse was found in the Jhenaidah and Kushtia districts ($1.18 \mu\text{g g}^{-1}$), while the lowest was found in Mymensingh and Chittagong districts ($<0.01 \mu\text{g g}^{-1}$) (Fig. 2). Lead smelting, emission from vehicles, and other industrial activity might be responsible for higher Pb in pulses in Jhenaidah and Kushtia districts (Kormoker et al., 2019).

3.3 Pb in Fruits

The Pb concentration in fruits was relatively lower than other food crops in Bangladesh, although some fruits showed higher Pb concentrations, exceeding the MAC value of $0.10 \mu\text{g g}^{-1}$ (Fig. 1). Average Pb content in fruits ($\mu\text{g g}^{-1}$) showed the following increasing order: apple and pineapple (0.09) < coconut (0.21) < papaya (0.28) < banana, blackberry, guava, and litchi (0.30) < jackfruit (0.42) < mango (0.61) (Supplementary Table S2). Only apple and pineapple showed lower Pb concentrations than the MAC value of $0.10 \mu\text{g g}^{-1}$ (Fig. 1). The highest Pb was found in mango ($0.61 \mu\text{g g}^{-1}$), six times greater than the MAC for Pb (FAO/WHO, 2019). Among the different districts, Pb in mango ranges from 0.18 to $0.86 \mu\text{g g}^{-1}$, collected from the Rajshahi and Bogra districts, respectively (Islam et al., 2015; Saha & Zaman, 2013). However, Mangoes cultivated on Hainan Island in China showed $0.042 \mu\text{g g}^{-1}$ fresh weight of Pb (Bi et al., 2010). In Pakistan, Pb concentrations in mango (average: $0.37 \mu\text{g g}^{-1}$) and guava (average: $0.17 \mu\text{g g}^{-1}$) were much lower than Bangladesh; however, Pb in banana (average: $0.42 \mu\text{g g}^{-1}$), papaya (average: $0.64 \mu\text{g g}^{-1}$), and apple ($0.65 \mu\text{g g}^{-1}$) were several folds higher than Bangladesh (Jaffar & Masud, 2003).

Further, among the different districts in Bangladesh, Pb in fruits was only reported for the Bogra, Dhaka, Jhenaidah-Kushtia, and Rajshahi districts (Fig. 2). The highest Pb in fruits was revealed in the Jhenaidah and Kushtia districts ($0.76 \mu\text{g g}^{-1}$), while the lowest was measured in the Dhaka district ($0.22 \mu\text{g g}^{-1}$) (Supplementary Table S3). In Jhenaidah and Kushtia districts, Pb concentration was reported for banana, guava, jackfruit, and mango, where

the highest Pb was found in bananas ($0.90 \mu\text{g g}^{-1}$) (Kormoker et al., 2019). In contrast, the lowest Pb was found in guava ($0.62 \mu\text{g g}^{-1}$), even though it is about six times higher than the MAC of $0.10 \mu\text{g g}^{-1}$ (FAO/WHO, 2019). The higher concentration of Pb in food samples could perhaps be due to lead smelting, emission from vehicles, and other industrial activity in the study area (Kormoker et al., 2019).

3.4 Pb in Vegetables

Pb concentrations in different vegetables are depicted in Fig. 1. The highest Pb concentration was found in cabbage ($4.64 \mu\text{g g}^{-1}$), while the lowest was found in green banana ($0.03 \mu\text{g g}^{-1}$). In the trans-Himalayan region (seven different villages in Ladakh, Union Territory, India), Pb in cabbage was about $0.14 \mu\text{g g}^{-1}$ (Giri et al., 2021), which was slightly higher than the MAC (at $0.10 \mu\text{g g}^{-1}$, FAO/WHO, 2019), but more than 33 times lower than Bangladesh. In addition, Rehman et al. (2019) evaluated (Pb levels in cabbage, spinach, cauliflower, and carrot which were 0.021 , 0.296 , 0.023 , and $0.015 \mu\text{g g}^{-1}$ Pb respectively in Pakistan, which were again lower than in Bangladesh (Rehman et al., 2019). Moreover, the Pb concentrations in food crops, such as yam, pepper, and green amaranth, were reported as ranging from 0.27 to $8.96 \mu\text{g g}^{-1}$, at a Pb-Zn mine site in Nigeria (Bello et al., 2016). Cabbage is grown in different urban and metal smelter areas of Australia, for example, Cowra and Sydney basins contained about <0.02 and $0.041 \mu\text{g g}^{-1}$ fresh weight of Pb, respectively (Kachenko and Singh, 2006). However, most of the vegetables in Bangladesh contained higher Pb concentrations than the maximum allowable concentration (MAC) of $0.10 \mu\text{g g}^{-1}$ (FAO/WHO, 2019), except mushroom, green banana, cauliflower, and arum. The respective average Pb concentrations in mushroom, green banana, cauliflower, and arum were 0.07 , 0.03 , 0.14 , and $0.08 \mu\text{g g}^{-1}$. However, around 10–46 times higher Pb content was revealed in cabbage ($4.64 \mu\text{g g}^{-1}$), zucchini ($3.36 \mu\text{g g}^{-1}$), sponge gourd ($3.28 \mu\text{g g}^{-1}$), tesla gourd ($2.19 \mu\text{g g}^{-1}$), okra ($1.66 \mu\text{g g}^{-1}$), chili ($1.75 \mu\text{g g}^{-1}$), drumstick lib ($1.39 \mu\text{g g}^{-1}$), green amaranthus ($1.12 \mu\text{g g}^{-1}$), papaya ($1.10 \mu\text{g g}^{-1}$), and pointed gourd ($1.03 \mu\text{g g}^{-1}$) that were grown in different districts of Bangladesh (Supplementary Table S2).

In Bangladesh, only 5% of cultivable land is used for vegetable production (Mahmud, 2019). However, among different districts, the highest Pb concentration in vegetables was found in the Tangail district ($2.17 \mu\text{g g}^{-1}$), which is about 22 times higher than the MAC of $0.10 \mu\text{g g}^{-1}$ (Fig. 2). In this district, the highest Pb was found in sponge gourd ($6.38 \mu\text{g g}^{-1}$), and the second-highest was measured in chili ($5.91 \mu\text{g g}^{-1}$) (Islam et al., 2020; Proshad et al., 2020). Since there is higher traffic and industrial emissions in this district, this higher level of Pb may originate from these sources. It is mentionable that a large number of different types of industries are under operation, for example, garment manufacturing, packaging industry, dyeing, brick kiln, metal workshops, battery manufacturing industries, tanneries, textile industries, and pesticide and fertilizer industries (Proshad et al., 2020). In contrast, the lowest Pb in vegetables ($<0.01 \mu\text{g g}^{-1}$) was found in the Mymensingh and Chittagong districts, followed by the Jashore district ($0.19 \mu\text{g g}^{-1}$) (Supplementary Table S3). The lower Pb content in vegetables in these districts might be related to the light industrial activities in these areas as they are rural or a less industrialized zone in Bangladesh (Alam et al., 2003). However, in other districts, Pb content in vegetables was higher than $0.10 \mu\text{g g}^{-1}$ (Fig. 2). The increasing order of Pb in vegetables ($\mu\text{g g}^{-1}$) in different districts are as follows: Mymensingh and Chittagong (<0.01) Jashore (0.19) < Patuakhali (0.51) < Bogra (0.61) < Pabna (0.63) < Noakhali (0.65) < Narayanganj (0.78) < Dhaka (0.85) < Rajshahi (1.00) < Jhenaidah and Kushtia (1.08) < Gazipur (1.27) < Chapai Nawabganj (1.97) < Tangail (2.17) (Supplementary Table S3).

3.5 Miscellaneous

Turmeric is a common spice popularly used in cooking in almost all families in Bangladesh as a yellow colorant in food. However, very little has been reported on Pb concentration in turmeric grown in Bangladesh. The first study in 1987 reported Pb concentration in Bangladeshi grown turmeric, and it was found that the turmeric bulb matrix and cuticle contained 1.3 and $2.6 \mu\text{g g}^{-1}$ of Pb, respectively (Syed et al., 1987). Later, turmeric was identified as a potential source of Pb exposure that increases blood level Pb in Bangladeshi inhabitants (Gleason et al., 2014). This study also reported a very high level of Pb in turmeric samples (average: $80 \mu\text{g g}^{-1}$; range: $< \text{LOD}$ $483 \mu\text{g g}^{-1}$) collected from

Sirajdikhan, Munshiganj. Uptake of Pb from the soil into the turmeric and/or addition of lead chromate in turmeric was anticipated as potential sources of Pb (Gleason et al., 2014). Subsequently, a case study also reported Pb in turmeric was one of the potential sources of elevated blood lead levels among pregnant Bangladeshi women in the Mymensingh, Tangail, and Kishoreganj districts. The Pb concentration was estimated in 17 turmeric samples with a median concentration of $1.8 \mu\text{g g}^{-1}$; however, one turmeric sample contained about $265 \mu\text{g g}^{-1}$ of Pb (Forsyth et al., 2018). After that, a follow-up study reported Pb content in turmeric from 9 (nine) major and 2 (two) minimally turmeric-producing districts indicating that PbCrO_4 was added to turmeric to enhance the yellow color as consumers prefer the bright color of turmeric. Consequently, the highest Pb in turmeric reached $1152 \mu\text{g g}^{-1}$ in the Dhaka and Munshiganj districts (Forsyth et al., 2019). In raw turmeric root, average Pb content was 0.52 and $0.48 \mu\text{g g}^{-1}$ in major and minimally turmeric-producing districts, respectively. However, after polishing, the content increased several folds to 11.67 and $147 \mu\text{g g}^{-1}$ in major and minimally turmeric-producing districts, respectively (Forsyth et al., 2019). It was because of the adulteration with PbCrO_4 at the polishing stage of turmeric that Pb elevation was observed. Additionally, in loose turmeric powder, the average Pb level was 99.83 and $14.32 \mu\text{g g}^{-1}$ in major and minimally turmeric-producing districts, respectively (Forsyth et al., 2019). In Saudi Arabia, Pb concentrations in turmeric were reported at $1.0 \mu\text{g g}^{-1}$ (Seddigi et al., 2016) in Saudi Arabia; however, turmeric contained about $5.54 \mu\text{g g}^{-1}$ of Pb in Malaysia (Nordin & Selamat, 2013).

Now, it is evident that Pb in raw turmeric is relatively lower than in powdered and polished turmeric. This is because of the adulteration with PbCrO_4 at the polishing stage, which has caused a direct threat to human health by consuming turmeric. However, there is no data on how much turmeric is consumed daily by the Bangladeshi people. Therefore, we were unable to estimate probabilistic human health risks for turmeric consumption in this study. Nevertheless, due to the extreme Pb level in turmeric, its consumption may pose a severe health threat to the population.

On the other hand, a significant number of Bangladeshi people chew betel quid that is prepared from Piper betel leaves, areca nut, some slaked lime, and

flavored or raw dried tobacco (Al-Rmalli et al., 2010). It was reported that these betel quid contained a significant amount of Pb. For example, Bangladeshi produced lime, and dried tobacco leaves, which contained 0.33 ± 0.03 , 0.22 ± 0.07 , and $1.01 \pm 3.07 \mu\text{g g}^{-1}$ Pb, respectively (Al-Rmalli et al., 2010). This Pb concentration is much greater than the MAC of $0.10 \mu\text{g g}^{-1}$, indicating that betel quid consumption may pose severe health risks to the consumers (FAO/WHO, 2019). In addition, tobacco grown in southwestern Bangladesh showed 0.56 ± 0.49 and $0.54 \pm 0.37 \mu\text{g g}^{-1}$ of Pb in raw root and leaf (Saha et al., 2016). Therefore, it is evident that non-commonly consumed foods contain a high level of Pb that may pose severe health risks to the consumers.

3.6 Mechanisms of Pb Uptake by Plants and Regulating Factors

There are two major mechanisms of metal uptake in plants, namely passive uptake due to osmotic concentration gradients along cell walls or inducible substrate-specific energy-dependent uptake (Williams et al., 2000). However, Pb is a non-essential element, so uptake via specific membrane-associated transporters do not exist for Pb, but rather uptake can occur via other divalent cation transporters (e.g., Ca channels). Pb can be available as the free cationic form or bound to carboxylic groups of mucilage uronic acids (Morel et al., 1986; Sharma & Dubey, 2005). After reaching root tissue, most of the absorbed Pb remains in the root and binds to exchangeable ion sites in the cell walls and extracellular precipitation as Pb phosphate and Pb carbonate (Raskin & Ensley, 2000; Sahi et al., 2002; Sharma & Dubey, 2005). The endodermis forces all apoplastic transport to the symplastic pathway. Thus, a much reduced quantity of unbound Pb may then move through Ca channels at the endodermis allowing limited acropetal transport (Antosiewicz, 2005; Huang & Cunningham, 1996). Thus, the most common pattern of Pb uptake is significant concentrations in root tissue with limited translocation of Pb to shoots.

Further, Pb uptake is directly or indirectly influenced by several soil physiochemical factors, for example, metal concentration in soil, soil pH, redox status, organic matter content, metal hydroxides, the clay content of the soil, and soil permeability. As such, this could explain some variability across taxa

(Dong et al., 2009). It appears that cereals and some vegetables exhibit greater uptake of Pb than pulses and fruit which may be attributable to their differing physiologies and growth habit and the harvestable parts analyzed (i.e., most vegetables are root vegetables), or could be due to the way these different plants are cultivated, with cereals and vegetables requiring more soil disturbance for cultivation. Further, findings may be due to where these different crops are grown spatially across Bangladesh (Garg et al., 2014; Liu et al., 2013; Saha & Zaman, 2013).

3.7 Temporal Variation of Pb in Food Crops

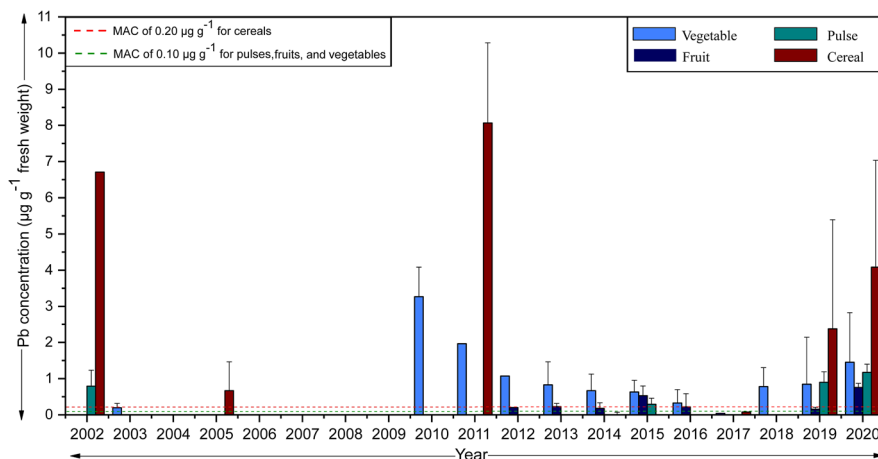
Figure 3 depicts the variation in Pb in different food crops from 2002 to 2020 in Bangladesh. Additionally, Pb concentrations in different food items in different years are provided in Supplementary Table S4. The publication year of articles was considered as the sampling year as most of the published articles did not mention the sampling period. Therefore, the temporal variation of Pb in different food items should be viewed cautiously. However, patterns clearly indicate an increasing trend in Pb in cereal food crops with time to the present. In the case of vegetables, from 2003, a sharp increase in Pb concentration was observed in 2010 and then gradually decreased until 2017. After that, Pb in vegetables again started increasing (Fig. 3).

This increasing trend might be related to the previously emitted Pb from vehicles, leaded gasoline, or recent release of Pb from lead-acid battery recycling, or industrial and vehicular emissions in Bangladesh. It is mentioned that the use of leaded gasoline in Bangladesh was phased out in the 1990s (Biswas et al., 2003), but Pb from previously combusted fuel may have accumulated in the soil and dust on roadsides. Consequently, the story of environmental Pb contamination in Bangladesh is still a matter of serious concern, and the decline is very slow due to the recent extensively used lead-acid batteries in different sectors.

3.8 Health Risks of Pb from Food Consumption

Consumption of Pb-contaminated vegetables, fruits, cereals, and pulses is associated with Human health risks that were estimated using the US EPA model. Both cancer and non-cancer health risks were

Fig. 3 Temporal variation in Pb content ($\mu\text{g g}^{-1}$ fresh weight; average \pm standard deviation) in commonly consumed foods in various districts of Bangladesh during 2002–2020. MAC for cereals ($0.20 \mu\text{g g}^{-1}$); MAC for pulse, fruits, and vegetables ($0.10 \mu\text{g g}^{-1}$) (FAO/WHO, 2019)



estimated in terms of target cancer risk (TCR) and target hazard quotient (THQ), respectively.

Figure 4 shows the THQ of each food item included in this study. In general, $\text{THQ} > 1$ indicates a chance of non-carcinogenic health risk, while $\text{THQ} \leq 1$ indicates no possible health risk (US EPA, 2001). Therefore, it is apparent that fruits and pulses do not pose any non-cancer health risks to Bangladeshi residents. However, consumption of rice and some vegetables, for example, cabbage, chili, drumstick lib, okra, sponge gourd, tesla gourd, and zucchini, may pose non-cancer health risks to the population as they showed THQ value higher than unity (Fig. 4). Among these food items, the highest THQ was observed for rice ($\text{THQ} = 4.41$), which is more than four times higher than the threshold value. The second-highest THQ was observed for cabbage ($\text{THQ} = 3.67$), while zucchini, tesla gourd, sponge gourd, okra, drumstick lib, and chili showed THQ values of 2.66, 1.73, 2.59, 1.31, 1.10, and 1.28, respectively (Supplementary Table S5). THQs of food groups indicated the highest THQ was exhibited by Pb-contaminated vegetables ($\text{THQ} = 27.2$), while the lowest was evaluated for pulse consumption ($\text{THQ} = 0.26$). The second-highest THQ was observed in cereal consumption ($\text{THQ} = 5.06$) and fruits ($\text{THQ} = 0.62$) (Supplementary Table S5). Besides, considering all these Pb-contaminated food items, the total THQ was 33.1, which is significantly higher than that of the threshold value ($\text{THQ} = 1$). Therefore, consumption of Pb through these food items is likely to pose non-carcinogenic health risks to the Bangladeshi inhabitants.

Lifetime cancer risks were also estimated based on individual food items, which are depicted in Fig. 5. Surprisingly, most of the vegetables and cereals showed values between 10^{-6} and 10^{-4} , indicating that consumption of these vegetables and cereals may pose cancer risks to the Bangladeshi population. Among the vegetables, only green bananas showed a lesser value than the lowest threshold limit of 10^{-6} . Comparatively, all of the fruits and pulses indicated a much lower value of TCR than the cereals and vegetables, indicating their lower cancer risk to the population (Supplementary Table S6). However, staple foods like rice showed a TCR value of 1.31×10^{-4} , which is almost one and a half times higher than the highest provisional safe limit of 10^{-4} , and cabbage also crossed the threshold (US EPA, 2005). Besides, most of the vegetables showed a higher level of TCR value than the lower limit of TCR value of 10^{-6} , indicating the possibility of lifetime cancer risks to Bangladeshi residents through the consumption of these vegetables.

Since cancer and non-cancer health risks of most of the food products indicated higher values than the threshold limits, it is likely to expect some of the symptoms of Pb poisoning provided in Fig. 6. In particular, the most diverse effect of Pb is neurotoxic and causes a number of health hazards in humans. The Pb alters the release of neurotransmitters (i.e., acetylcholine) from presynaptic nerve endings. The spontaneous release of neurotransmitters is enhanced due to the activation of protein kinases in the nerve endings, while the evoked release is inhibited by the blockade of voltage-dependent calcium channels

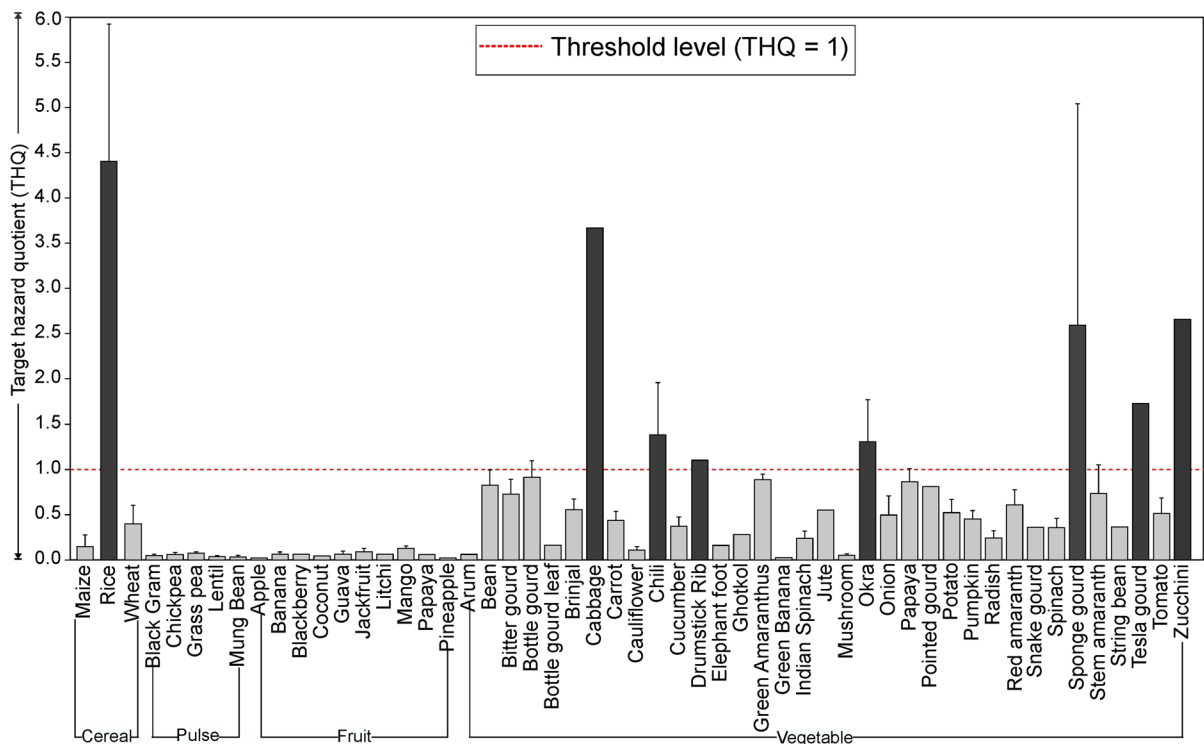


Fig. 4 Non-carcinogenic human health risk of Pb (average \pm standard deviation) from various agricultural food consumption in Bangladesh. Threshold level of target hazard quotient (THQ) is unity

(Bressler & Goldstein, 1991; Minnema et al., 1988). In the nervous system, Pb causes lipid peroxidation, excitotoxicity, alterations in neurotransmitter synthesis, storage and release, alterations in the expression and operation of receptors, interference with mitochondrial metabolism, interference with second-messenger systems, and damage to the astroglia and oligodendroglia (Lidsky & Schneider, 2003). Even the low level of Pb ($5\text{--}10 \mu\text{g dL}^{-1}$) in children's blood is associated with deficits in intelligence, visual-spatial skills, executive functions, problem solving and dispositions, and finally, IQ-adjusted academic achievement (Surkan et al., 2007).

Besides, Pb poisoning causes anemia by inhibiting heme synthesis through preventing the incorporation of Fe^{2+} into protoporphyrin (Ahamed & Siddiqui, 2007). Additionally, Pb induces oxidative stress by generating free radicals that exceed the capacity of antioxidant defence mechanisms. Oxygen radicals generated via Pb exposure induce the depletion of glutathione and protein-bound sulfhydryl groups and alter the activity of various related

antioxidant enzymes (Ercal et al., 2000). In contrast, Pb affects sperm function as it interacts with human protamine (HP2) and DNA-protamine binding disturbing chromatin decondensation in sperm (Quintanilla-Vega et al., 2000). Furthermore, renal functions are disturbed by Pb poisoning through the morphological change of the kidney. In the kidney, lead-protein complexes are formed, which is called nuclear inclusion bodies and ultrastructural changes in cellular organelles, especially the mitochondria (Goyer, 1993). Even Pb causes hepatic, renal, and hematologic injury by increasing concentrations of white blood cells, serum urea, creatinine, aspartate aminotransferase, and alanine aminotransferase and decreasing concentrations of hemoglobin and hematocrit (Nakhaee et al., 2019).

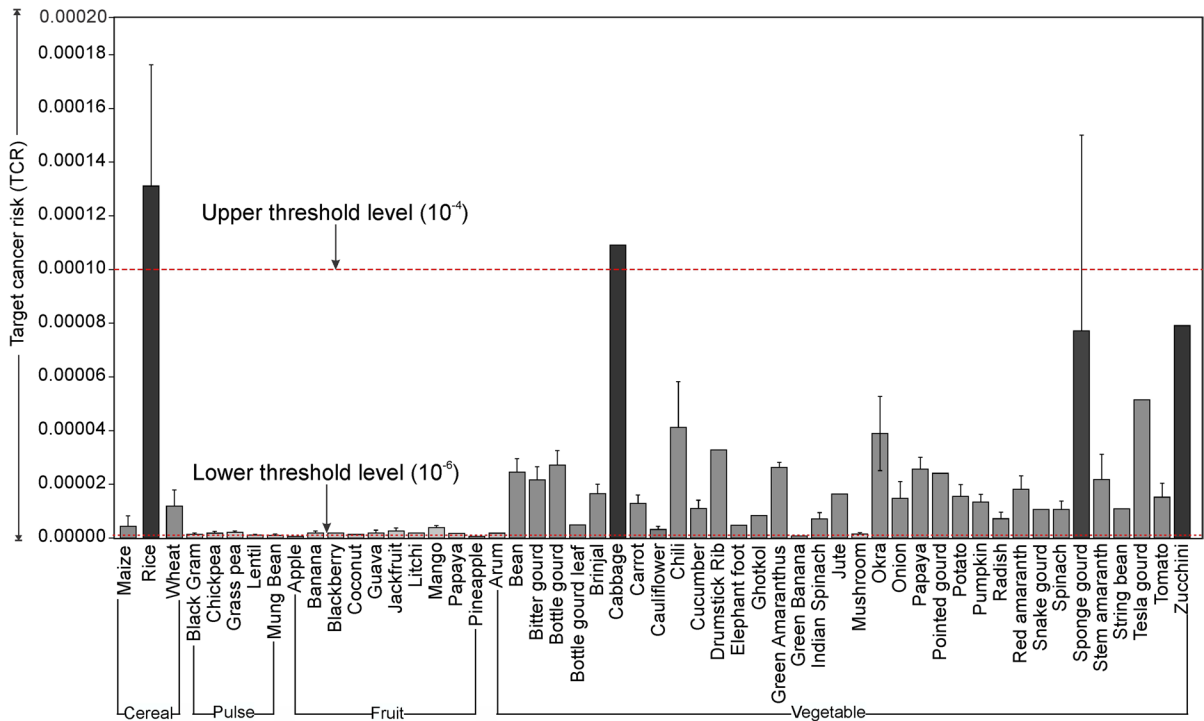


Fig. 5 Carcinogenic human health risk or target cancer risk (TCR) of Pb (average \pm standard deviation) from different agricultural food consumption in Bangladesh. Lower and upper

limits of target cancer risks are one in a million (i.e., 10^{-6}) and one in ten thousand people (i.e., 10^{-4}) respectively (US EPA, 2005)

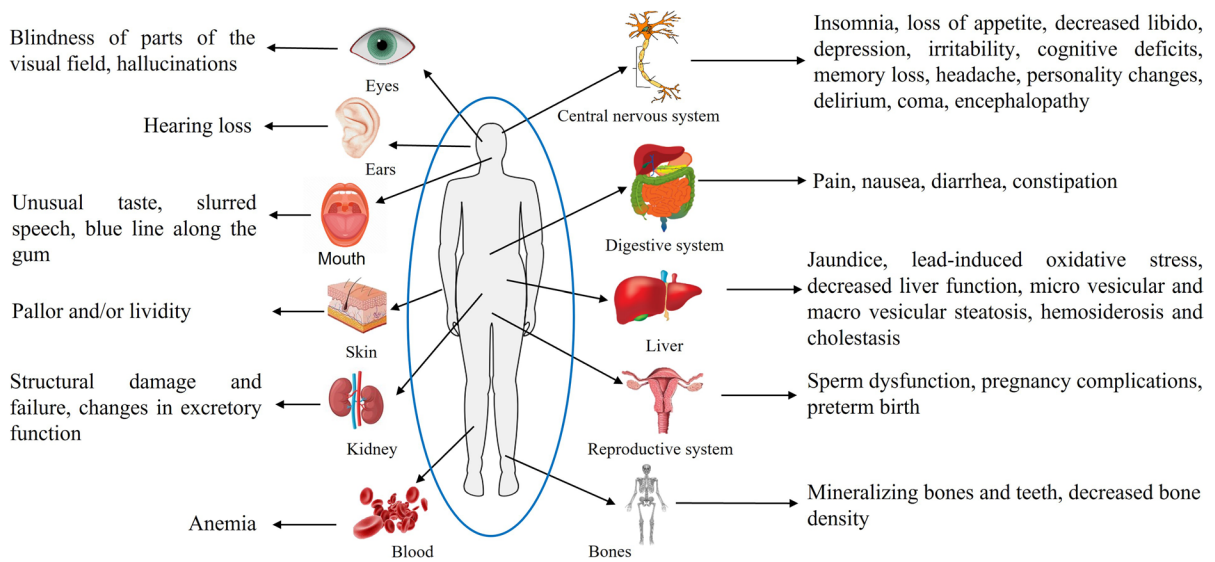


Fig. 6 Generalization of clinical symptoms of Pb poisoning in humans (modified from Kumar et al., 2020)

4 Possible Pb Sources and National Mitigation Programs in Bangladesh

4.1 Pb Sources in Bangladesh

Agricultural products in Bangladesh accumulate Pb from the soil, air particulates, and waters. The central repository for airborne Pb in the atmosphere is the plant and soil surface, contributing to dietary Pb ingestion (Farsam & Zand, 1991). Figure 7 shows the possible sources of Pb that can be accumulated in the food items. Basically, the use of synthetic fertilizers, untreated sewage disposal, and the burning of fossil fuels pollute the water, soil, and plants with Pb and other heavy metals (Nazemi & Ahmad, 2011). Besides, the recent extensively emitted Pb from vehicles used leaded gasoline as burning fuel. Further sources are lead-acid battery and their recycling, and/or other metal industrial activities in Bangladesh. Although the leaded gasoline has been phased out in Bangladesh after 1999s (Biswas et al., 2003), legacy Pb exists in curbsides and dust. The extensive use of livestock manures and composts in agricultural lands in Bangladesh is very common. Consequently, their use in agricultural practices is another potential source of Pb in agro-products. The recently

established lead-acid battery industry might be one of the highest Pb contributors to the Bangladeshi environment (Chowdhury et al., 2021; Kumar et al., 2022).

In contrast, internationally, it is well recognized that the application of fertilizer can considerably increase the content of Pb in the soil through essential elements of fertilizers contaminated with traceable amounts of Pb as impurities (Jones et al., 1981), especially the phosphate fertilizers (Davenport & Peryea, 1991). In addition, insecticides and fungicides might be another potential source of Pb, which was reported in the UK, in which 10% of the chemicals permitted for insecticides and fungicides were based on Pb compounds (Wuana & Okieimen, 2011). Thus, using fertilizer and insecticides for agricultural practices can contribute to the Pb pollution in soil. Besides, treatment of various biosolids like livestock manures, composts, and municipal sewage sludge unintentionally releases Pb that finally accumulates in soil (Basta et al., 2005).

On the other hand, mining, smelting, and fossil fuel burning cause Pb pollution in the environment. According to Wuana and Okieimen (2011), vast mining and smelting of Pb also pollute the soil. Additionally, the burning of fuel containing tetraethyl lead

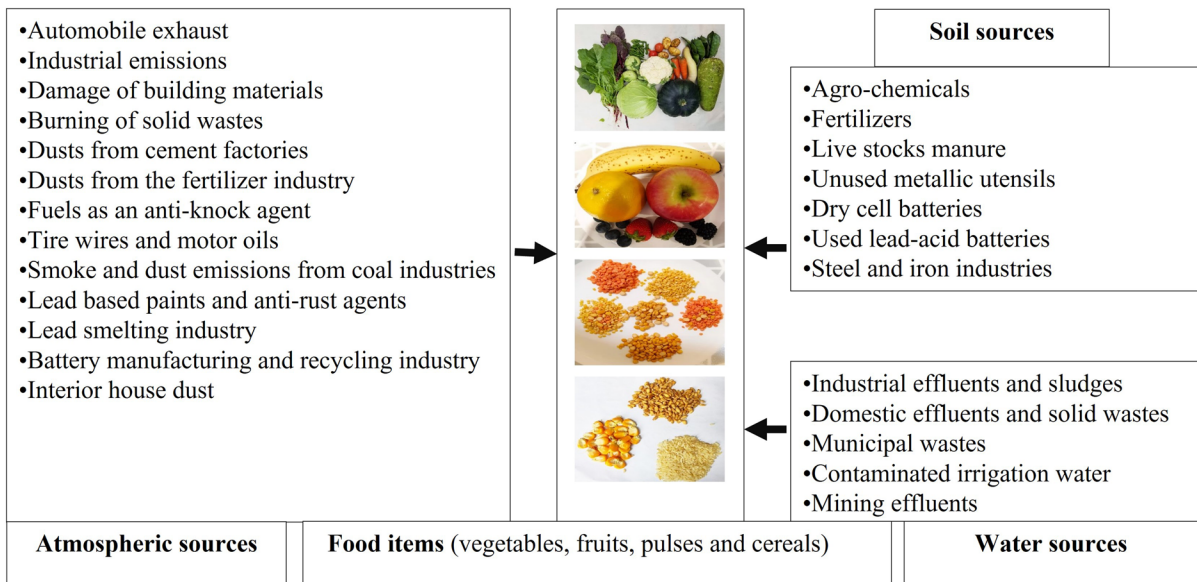


Fig. 7 Potential sources of Pb contamination in food items (modified from Majumder et al., 2021)

emits Pb in the air, which is another major cause of soil pollution (Wuana & Okieimen, 2011). Another finding indicated that emissions from vehicles and reduced rainfall are significant sources of lead poisoning in city plants (Farsam & Zand, 1991).

Thus, the Pb sources in foods can be categorized into two major groups, namely, the local sources and “hot spots” of contamination that include workshop, industry, and mine activities or heavily contaminated soil by anthropogenic activities or industrial waste dumps located close to cultivation sites; and the dispersed sources including contamination of a catchment area, large-scale soil fertilization, agricultural practices, and airborne particles dispersion.

4.2 National Pb Management and Mitigation Program

Although Pb pollution is very severe in Bangladesh, there is a scarcity of comprehensive Pb mitigation strategies to date. Currently, there are few programs and policies to control contemporary Pb sources, reduce exposures, and identify and treat Pb poisoning. However, recently, several strategies have been taken into account by Pure Earth Bangladesh in coordination with the Bangladesh Department of Environment (DoE) along with the support from the United States Agency for International Development, OAK Foundation, Swiss Agency for Development and Corporation SDC, and the Global Alliance on Health and Pollution (GAHP). The following steps were taken by the responsible authorities or policymakers to make a roadmap toward national level Pb abatement and management (Pure Earth Bangladesh, 2021):

(a) A field project at Kathgora, Dhaka has been established with finance from USAID and World Bank, Pure Earth along with other stakeholders to reduce the burden of disease from Pb pollution in Bangladesh. (b) The unauthorized lead-acid battery (ULAB) recycling factories are the foremost Pb releasing hub in Bangladesh. Therefore, the Bangladesh government has recently issued a special statutory regulatory order on ULABs; c). Bangladesh has banned Pb in gasoline and paint.

The following recommendations were proposed by the Pure earth Bangladesh (not edited) through the discussion with different authorities and

stakeholders to forward toward a national plan of Pb abatement (Pure Earth Bangladesh, 2021):

1. “A multi-stakeholder approach with the leadership of the relevant government ministries should be established to eradicate community lead exposure. The Department of Environment (DoE) should take the lead in a joint, multi-stakeholder approach to eradicating lead pollution. A coordination committee could be formed in the Ministry of Environment, Forest, and Climate Change, and a technical committee can be formed under the leadership of the Director-General of DoE.
2. The relevant government departments and ministries that should work closely on this issue are the Ministry of Environment, Forest and Climate Change, the Department of Environment, the Ministry of Health, the Ministry of Commerce, the Ministry of Industry, the Local Government Division, and the Food Safety Authority.
3. A time-bound, holistic national action plan which considers existing legislation is needed to advance progress on the issue of lead exposure. This action plan should include provisions for monitoring, reporting, and enforcement.
4. Comprehensive lead pollution studies and a national inventory of lead pollution sources are key to prioritizing effective exposure mitigation projects. This research is needed to identify polluting industries and lead-containing consumer products.
5. Effective monitoring by relevant agencies is needed to identify lead exposure sources, develop interventions, and ensure long-term success. This is needed for both industrial sources (e.g., the closure of informal ULAB sites) as well as in products (e.g., lead chromate adulteration in spices).
6. Interventions in the ULAB recycling sector should be prioritized as this is a major known source of community lead exposure. Research institutions and universities should come up with ways to shift illegal, informal ULAB recycling industries to the regulated, registered sector. Second-generation lead-acid batteries or alternatives such as lithium-ion batteries should be examined for application in Bangladesh.

7. Occupational health and safety hazards of working with lead need more attention; workers who are working directly with lead require additional education. For those workers engaged in informal lead industries, alternative livelihoods should be explored as more of the industry shifts to the formal sector.
8. Ensuring effective waste management across all sectors, especially industrial waste, is important because contaminated waste ends up affecting health through different pathways, including the agricultural pipeline.
9. Blood Pb monitoring must be established. Blood Pb data could be integrated into the MOHFW's existing routine health information system, DHIS2. Sufficient investment is needed to conduct this testing at district and division levels. Blood Pb data can be used to identify contributing sources and monitor the efficacy of interventions.
10. The capacity building of health workers and the health care sector to address lead exposure should be expanded.
11. Sensitization through print and electronic media plays a crucial role in creating public awareness about the sources and effects of lead pollution and spurring government bodies to act. There should be various training sessions and workshops to enhance the knowledge and skills of stakeholders related to lead pollution".

This research supports the recommendations of Pure Earth and further recommends reducing current sources of Pb contamination, mainly focusing on reducing emissions from industry and managing the industrial waste disposal sectors. Further, produce should be grown on uncontaminated land where possible and remediation strategies put in place for Pb-contaminated land which is employed for agricultural purposes.

5 Conclusion

The increasing problem of Pb contamination in Bangladesh from growing human activity results in hazardous impacts on plants, and ultimately humans who consume plants. This study provided a complete assessment of the health risks associated with Pb

contamination of commonly consumed cereals, vegetables, fruits, and pulses in Bangladesh using published data from the peer-reviewed literature.

Most vegetables and cereals contain higher Pb concentrations than the maximum allowable limits, and thus consumed vegetables and cereals may expose Bangladeshi citizens to cancer and other health-related risks. Although pulses and fruits contained substantial levels of Pb, intake of fruits and pulses was cooperatively lower in individuals, which made it relatively safe in terms of human health risk assessment. In contrast, Pb concentrations were relatively high in cereals (i.e., rice, wheat, and maize). Among several districts, Tangail showed the highest Pb levels in vegetables, which could be related to industrial operations and traffic emissions. However, synthetic fertilizer, livestock manures, untreated sewage water, and industrial and traffic emissions are some of the potential sources of Pb pollution in food crops in Bangladesh. In terms of temporal trends in Pb accumulation in crops, patterns clearly indicate an increasing trend in Pb in cereal food crops with time to present, while a decreasing trend in Pb in vegetable food crops.

In terms of health risks, assessment of most of the cereals and vegetables exceeded allowable limits for cancer and non-cancer health-related risks. We recommended abatement strategies to reduce Pb emissions, and growing produce on uncontaminated land or remediate land prior to cultivation of agricultural crops.

Author contribution Sazal Kumar: conceptualization, investigation, methodology, data curation, formal analysis, writing—original draft. Rafiqueel Islam: conceptualization, supervision, investigation, methodology, writing—review & editing. Pritom Bhowmik akash: data curation, writing—review & editing. Md Hafjur Rahaman Khan: data curation, writing—review & editing. Ram Prosad: data curation, writing—review & editing. Joyanto Karmoker: data curation, writing—review & editing. Geoff R. MacFarlane: conceptualization, investigation, methodology, supervision, and administration, writing—review & editing.

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Data Availability All data generated or analyzed during this study are included in this published article and its supplementary information files.

Declarations

Conflict of Interest The authors declare no competing interests.

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