

Trend of Arsenic Exposure through the Food-Chain in Bangladesh for the Past Two Decades¹

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Abstract: Large scale use of groundwater for irrigation which is often Arsenic laden, during past two decades, has given rise to the apprehension that this may be resulting in higher Arsenic intake through the food chain. This work was undertaken to determine the trend of average Arsenic exposure of the population in Bangladesh during the period 1990-2010. Based on available data on individual food items, an average estimate of the level of both total and inorganic Arsenic intakes through food chain in Bangladesh have been made using the market basket approach; which is a widely used protocol for such work. The results have been discussed in the light of available information in literature. The results obtained show no significant change in the average intake of inorganic Arsenic during the period. The level of total Arsenic exposure does not also show statistically significant increase in uptake during the study period. Consequently, it is concluded that no restriction is needed on the use of groundwater in irrigation from Arsenic exposure risk consideration of the population. Now that there is no WHO recommended tolerable limit for intake of Arsenic, nationally acceptable limits have to be determined probably by considering cost-benefit analysis.

Keywords: Groundwater, Total Arsenic, Inorganic Arsenic, Food-chain, Market Basket

1. Introduction

The problem of Arsenic (As) ingestion through food chain in Bangladesh has been reported in the press from time to time. Such reports have given rise to the apprehension that ingestion of Arsenic through food and its consequent health impacts may be substantial. This apprehension is being reinforced by the fact that Arsenic contaminated groundwater has been extensively used for irrigation of crop lands during the past two decades, particularly the rice fields.

Dietary exposure assessments for Arsenic are needed to determine the potential health risks associated with the transfer of arsenic from groundwater used for irrigation, to soil and the food chain and then to human. It is necessary to assess the changes in the concentration of Arsenic in the food supply to determine if such changes are important as a public health issue; with the continuing use of Arsenic contaminated water for irrigation in Bangladesh. It is also necessary to assess the strength of different contributing factors in the overall human health risk assessment process, so that this knowledge can be fruitfully utilized in identifying the risk

management options in this context and developing appropriate mitigation strategies.

Data on Arsenic content of food, commonly consumed in Bangladesh, are scattered in literature with no comprehensive compilation available to date. There are also methodological limitations in the estimates of the level of Arsenic ingested through the food chain. Under such circumstances estimates of Arsenic ingestion through the daily food intake are bound to be somewhat uncertain. The present study is thus an attempt to reflect upon the status of arsenic exposure during past 20 years within such boundary conditions. The estimates of arsenic exposure through the food chain in Bangladesh available in literature are of limited use and can't be used without additional approximations for policy decisions. In the present work, the uncertainties have been explained at the appropriate context of the problem studied.

1.1. Arsenic Compounds in Water and Food

Arsenic is a ubiquitous chemical element in the biosphere and it occurs naturally in both inorganic and organic forms. The most important inorganic arsenic compounds are arsenic

1. The views expressed in this article is in no way related to World Bank.

trioxide, sodium arsenite, arsenic trichloride (i.e., trivalent forms), and arsenic pentoxide, arsenic acid and arsenates, such as lead and calcium arsenates (i.e. pentavalent forms). The common organic arsenic compounds are arsanilic acid, methylarsonic acid, dimethylarsinic acid (cacodylic acid), and arsenobetaine (AB). This latter compound (AB) is considered to be the most predominant organoarsenical in marine biota. Other organoarsenicals including arsenocholine, dimethyloxyarsylethanol, trimethylarsonium lactate, arsenic containing sugars and phospholipids have also been found in fish. Arsenites (trivalent) and arsenates (pentavalent) are found in water although methylarsonic and dimethylarsinic acids have been found at low levels. More details on the issue can be found in the literature [1].

1.2. Standards for Arsenic Intake Through Food

Arsenic intake through food was reviewed by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) recently (2010:72nd Meeting)[1]. This JECFA meeting dropped earlier provisional maximum tolerable weekly intake (PTWI) limit for ingested inorganic arsenic of 15µg/kg bw (body weight). Therefore, *there is currently no reference health standard for inorganic Arsenic ingestion*. The new inorganic arsenic lower limit on the benchmark dose for a 0.5% increased incidence of lung cancer (BMDL0.5) was determined from epidemiological studies to be 3.0 µg/kg bw per day (based on the range of 2–7 µg/kg bw per day estimated total dietary exposure) using a range of assumptions to estimate total dietary exposure to inorganic arsenic from drinking water and food. This limit is equivalent to 180 µg/day of inorganic Arsenic for 60 kg bw (i.e., body weight of Reference Asian Man as per International Commission on Radiological Protection (ICRP)) including both food and water. It is, therefore, necessary to determine not only the content of total Arsenic intake through food and water but it is also essential to find its inorganic component to verify compliance with this limit.

In the Australia -New Zealand Food Authority Report [2], PTDI (Permissible Tolerable Daily Intake) of 3µg/kg bw per day for inorganic Arsenic intake previously recommended has also been dropped following JECFA recommendation. The U.S. Food and Drug Administration (USFDA) has no recommendation on tolerance levels for arsenic in food, except for the by-product foods from animals treated with veterinary drugs. The permissible levels range from 0.5 ppm in eggs and uncooked edible tissues of chickens and turkeys to 2 ppm in certain uncooked edible by-products of swine [3].

2. Literature Review

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2.3. Dietary Arsenic Intake in Different Countries

Dietary arsenic represents the major source of arsenic exposure of most of the general population. Persons who are high consumers of fish may ingest significant amount of arsenic (primarily organic) from this food group. Although, most monitoring data are given as the concentration of total arsenic, arsenic in foods is a mixture of inorganic species and

organoarsenicals including arsenobetaine. The actual total arsenic concentrations in foodstuffs from various countries will vary widely depending on the food type, growing conditions (i.e., type of soil, water, geochemical activity, use of arsenical pesticides) and processing techniques.

Examples of mean total daily intakes of arsenic from food and beverages in different countries are given in Table 1. The

variation in dietary intake of total arsenic in adults reflects, largely in the variability in the consumption patterns of arsenic-rich food groups (fish/shellfish and meats) confirming the need to consider such regional variations in arsenic intake when assessing human health effects for arsenic.

Table 1. Estimated average dietary intake of As in various countries

Country	Sampling method/ Other Info	As intake (µg/kg bw per day)		Reference
		Total	Inorganic	
Bangladesh	Average food intake without water	3.01	0.88	[7]
	Five Scenarios (including water)	2.68-5.07	-	[8,9]
	Small community without water	-	1.68-3.00	[10]
India	Urine Analysis in As contaminated areas (DD)	-	3.32 - 12.9	[11]
	TDS(including water)	-	0.24-0.76	[10]
China	Small community without water (adult)	-	0.91	"
China (Taiwan)	TDS including water(Adult)	-	0.36 – 0.46	"
Japan	Adult (EFSA)	-	0.21-0.61	"
Europe	TDS (All without water)	-	0.29	"
Canada	TDS(Adult, water included in some studies)	-	0.08-0.20	"
USA	1-6 years	-	0.12-0.32	"
	Infant<12 Months	-	0.24 -1.19	"
	>17 yrs (MB) not per Kg bw	41	-	[12]*
Australia	13-16 yrs	37	-	"
	6-12 yrs	32	-	"
	2-5 yrs	25	-	"
	9 Months	8.8	-	"

MB - market basket survey; TDS - total diet study; DD- duplicate diet study; LOQ- limit of quantification; * Assuming (No data=LOQ)

Table 2. Arsenic Concentration in rice in Bangladesh, Japan and USA.

Sl.	Rice Type	No of Samples	Range (mg/kg)	Median/mean (mg/kg)	Comments	Ref
1	IRRI	15	0.11 - 0.55	0.20	HYV (mean value)	[13]
2	Aman	12	0.08 - 0.36	0.20	Local	[13]
3	Samata	21	0.11 -0.4	0.23	Contaminated area (Single high value of 0.94 found)	[14]
4	Market	41	0.03 - 0.34	0.1	Dhaka-Rajshahi Market	[14]
5	Japan	29	0.07 - 0.32	0.18		[14]
6	Boro	-	0.01 - 0.42	0.183	Average for Boro (14% moisture content)	[15]
7	Aman	-	-	0.117	Avg. for Aman (14% moisture content)	[15]
8	US	-	-	0.24	USA	[16]

2.4. Arsenic Concentrations in Specific Food Items

2.4.1. Arsenic in Bangladesh Rice

As the most important component of diet in Bangladesh, rice is likely to contribute the largest amount of Arsenic in the dietary uptake. It is also the crop in which the maximum amount of irrigation water is used specially in the dry season. The data on Arsenic concentration in different types of rice in Bangladesh and some other countries are given in Table 2. The data for Bangladesh sample are for a number of rice varieties and from different regions. The data also cover a considerable period of time. The earliest data available on rice are that from a study by Khan et al dates back to 1989[13]. It is interesting to note that ranges of concentration found have not changed significantly since then. It appears that a value of 0.2 mg/kg should be appropriate for rice being near median. Such contingent approach has to be used due to

the spread in the available data which, of course, are reflected in the final estimates obtained. It may also be noted that rice from Japan and US contains similar amount of total Arsenic as in Bangladesh. It may be further noted that analytical measurements for Japanese and some Bangladeshi rice samples were carried out in the same laboratory; so there are no systematic analytical uncertainties involved in these measurements. This review also shows that all the data available are due to the contribution of individual researchers mostly through one single work. In order to keep track of the time variation of Arsenic level is essential to do regular analytic studies at national or regional levels.

In choosing a concentration level for Arsenic for use in dietary intake calculations, median values have been used rather than the mean level to represent the most likely level in any given food material. The median level is a more stable central statistic and is not sensitive to skewing by chemical

detections above the normally expected range.

The median simplifies calculations for surveys containing analytical results below the limit of quantification (LOQ) because the position of the median, unlike the mean, is not dependent on the treatment of results below the LOQ. Means and medians are generally well correlated where there are few results reported below the LOQ. This is the approach that has been followed in the ANZFA study [2].

2.4.2. Arsenic Concentration Data in Other Food Items

The data on other food items required for the MB are shown in Table 3. In cases local values are unavailable, values available in literature from other countries have to be used. For all the values in this table references have been provided in column-4. The nonlocal values should be revised on the availability of Bangladesh specific data. The Arsenic concentration data do not appear to show systematic time variation. So, only single value will be used in Arsenic load calculation.

Table 3. Concentration of As in food items from literature (wet weight except rice) with the values chosen for calculation in this work.

Food groups	Total As (µg/kg)	Inorganic %	Ref.	Total Conc. (µg/kg)	Inorganic As (%) of total	Comments
Rice	-	43	[16]	200.	43	Conc. Data from Table 3.
Potato	23	10	[17]	23.0	10	
Vegetables	2.8 7.0	5 -	[17] [18]	7.0	5	
Pulses				200.	43	Same as rice assumed
Milk/milk products	12	75	[17]	12.0	75	
Milk/dairy prod	3.8	-	[18]			
Meat/poultry/eggs						
Chicken	22	41	[16]	22.0	41	
Meat and Poultry	24.3	-	[18]			
Fish						
Sole	4000	1	[16]	1662.	10	
Tuna	1100	2	[16]			
Shrimp	650	16	[16]			
Prawn	5040	-	[13]			
Fish and shell fish	1662	-	[18]			
Spices(onion, chillies & others)				7.0	5	Same as vegetables assumed
Edible oils						
Fats and oil	19.0		[18]	19.0	100	100% Inorganic assumed
Fruits	6.4	10	[17]	6.4	10	
Sugar						
Sugar/candies	10.9		[18]	10.9	43	Inorganic % same as grain assumed
Miscellaneous*						
Tea	35	26	[16]	12.5	43	Inorganic % same as grain assumed
Miscellaneous	12.5	-	[18]			

* Contingent estimates are used when no or multiple values are available.

3. Objectives

The objective of the present study is to evaluate the status of arsenic exposure of the Bangladesh population during past 20 years, in view of the use of the irrigation with Arsenic contaminated water during the period. Such assessment on time variation of arsenic exposure through the food chain in Bangladesh is needed for policy decision on the continued use of Arsenic laden irrigation water. As seen in the review above, the data on Arsenic content of food, commonly consumed in Bangladesh, are scattered in literature with no systematic time series of analytical work at national level. There is also a lack of comprehensive compilation of Arsenic data to date. Further, there are methodological limitations in the estimates of the level of Arsenic ingested through the food chain. Under such circumstances estimates of Arsenic ingestion through the daily food intake has been undertaken using the established market basket (MB) protocol. The

uncertainties in the results obtained have been carefully kept in view and explained at the appropriate context in view of the data uncertainties.

4. Methodologies for Arsenic Intake Analysis through Food

For a particular population group, the dietary intake (exposure) of Arsenic is the total Arsenic from all the food consumed per day and can be written as:

$$I = \sum_i C_i \times W_i \times M \quad (1)$$

where I = Dietary intake; C_i = Arsenic concentration in a food item, I; W_i = weight assigned for a specific food item, I; and M = Average quantity of food consumed by a specified group for which the estimate is being made.

Once the estimates for food consumption level and the composition of the food are made, these can be combined with food Arsenic concentration data to estimate the dietary

intake (exposure) of Arsenic. In a dietary exposure assessment, protocols have been established which are widely used all over the world. Two main approaches have been reported in literature for estimating dietary exposures which are briefly discussed below.

4.1. Total Diet Study (TDS)/ Market Basket (MB) Approach

The Total Diet Study (TDS), sometimes, is also called as the Market Basket (MB) Study. It is used in the USA and other countries (e.g., Australia, New Zealand) for the purpose of estimating intakes of many substances including Arsenic in representative diets of specific age-sex groups of the population. TDS approach originated in response to the public concern over radioactive contamination in the early sixties but since that time, the program has been adapted to reflect the interest in additional contaminants including Arsenic. A total of 382 different food items are represented in the MB and dietary intake of each contaminant for each of 14 age/sex groups are calculated from the analytical results and the weighted food factor for each group in the US study. These studies are conducted by the USFDA[4].

Unfortunately, an agreed definition of a MB is not yet available for Bangladesh or in the South Asia region. There are very limited concentrations data reported in literature for Arsenic in many of the food items. Information on the difference of concentration among contaminated and uncontaminated areas are even scarcer. In view of this paucity in data, it is imperative that some initiative like the Latin American Total Diet Study [5], should be undertaken in South Asia. Such a study may include toxic chemicals, heavy metals, pesticides and toxins in addition to Arsenic.

4.2. Duplicate Diet Approach

This approach is experimental and it avoids much of the analytical data needs as in the TDS/ MB method. In this method, difference in the urine Arsenic concentration between two groups of volunteers are utilized. Urine level of Arsenic is a short term estimate (one week) for arsenic exposure. Two groups of participants are selected in this approach; an intervention group (provided with the same mix of food grown with Arsenic free water) and the other is a control group (provided with a mix of food grown in an area with Arsenic containing irrigation water). The differences in the concentration of urine Arsenic between the two groups are used to obtain the information on the Arsenic intake levels through the food. One such study was carried out in the Jessore area (a contaminated area) of Bangladesh by Milton et al. [6]. This study indicated that food production with arsenic contaminated irrigation water did not add significantly to total

arsenic exposure of the population in the area.

5. Results and Discussion

5.1. Composition of Average Diet in Bangladesh

The nearest equivalent to market basket (MB) data in Bangladesh is the per capita daily food intake data from BBS (Bangladesh Bureau of Statistics) Household Income and Expenditure Survey (HIES). The available data from the latest survey report (HIES 2013)[19] for 34 food items are for 2005 and 2010. Earlier data for 1990 and 1995 are from the 1998 HIES report [20]. This approach of using per capita diet (as equivalent to MB) was included in the methodologies for the estimates of contaminants in food in the WHO documents [21]. The data on Arsenic concentration in different food items given are limited as shown in tables 3 and 4. In the light of the limited data on Arsenic the different food items, some of the food items in the HIES data have been combined (i.e., similar items, total categories reduced from 25 to 14) and resulting data during 1990-2010 are shown in table 4. It can be seen from the data in this table that total amount of dietary intake has increased over the period under consideration. The dietary mix has also changed with progressively with the reduction in the amount of rice and increase in other cereals. There is also progressive increase in the amounts of vegetables, protein items (fish, meat, poultry and egg). Such changes in food habit with time are reflected in the Arsenic intake as the quantities are different in different food items.

5.2. Estimates of Arsenic Intake Through Food

The concentration of both total and inorganic Arsenic intakes have been calculated using equation (1). The results obtained for four survey years covering 20 years are also given in table- 4 along with food intake data. In choosing a concentration level for Arsenic for use in dietary intake calculations, median values have been used rather than the mean level to represent the most likely level in any given food material as explained in 5.2.1. The median level is a more stable central statistic and is not sensitive to skewing by chemical detections above the normally expected range.

The median simplifies calculations for surveys containing analytical results below the limit of quantification (LOQ) because the position of the median, unlike the mean, is not dependent on the treatment of results below the LOQ. Means and medians are generally well correlated where there are few results reported below the LOQ. This is the approach that has been followed in the ANZFA (2010) [2] study.

Table 4. Total and inorganic Arsenic intake calculated using the BBS average food intake values and concentration and speciation estimates from literature.

SL	Food Items	Average Per Capita Per Day Food Intake (Grams)				Arsenic Concentration	
		1990	1995	2005	2010	Total As µg/kg	Inorg %
1.	Year* → CEREALS						
1.1	RICE	516.2	508.7	439.6	416	200	43
1.2	WHEAT**	-	-	12.1	26	-	-

1.3	OTHERS**	-	-	17.3	21.9	-	-
2.	POTATO	43.7	49.5	63.3	70.3	23	10
3.	VEGETABLES	137.4	152.6	157	166.1	7	5
4.	PULSES	17.9	14	14.2	14.3	200	43
5.	MILK/MILK PRODUCTS	19.1	32.3	32.4	33.7	12	75
6.	EDIBLE OILS	10.1	9.9	16.5	20.5	19	100
7.	MEAT, POULTRY, EGG	12.9	15.1	20.8	26.2	22	41
8.	FISH	34.5	43.8	42.1	49.5	1662	10
9.	CONDI & SPICES	43.5	57.2	53.4	66	7	5
10.	FRUITS	16.9	27.6	32.5	44.7	6.4	10
11.	SUGAR/GUR	8.8	9.3	8.1	8.4	10.9	43
12.	MISCELLANEOUS ITEMS*	25.2	13.8	38.2	36.5	12.5	43
	Total Diet (g/day)	886.2	933.8	947.5	1000.1	-	-
	Total Arsenic Intake ($\mu\text{g/day}$)	167.65	191.83	165.59	173.77	-	-
	Inorganic Arsenic Intake($\mu\text{g/day}$)	52.50	53.17	47.30	46.67	-	-

*Surveys are usually conducted for longer periods than one year prior to publication of reports

**Consumption of these items have significantly increased over time

As the Arsenic concentration in food in individual items have not changed much over the period under consideration. The changes in the intake values shown in table-4, basically reflect the changes in the dietary habit. It can be seen that the values obtained for total and inorganic Arsenic have not changed much over 20 years although total diet intakes have increased. The average intake levels for 20 years are 175 ± 12 and 50 ± 3.4 $\mu\text{g/day}$ respectively for total and inorganic Arsenic. With the ICRP standard body weight of 60kg assumption, the values intakes correspond to 2.92 ± 0.2 and 0.83 ± 0.06 $\mu\text{g/kg bw}$ per day for total and inorganic Arsenic respectively. The changes in inorganic Arsenic levels observed are not statistically significant (i.e., within 2 standard deviation) in the 20 years period considered. The same trend is observed for total Arsenic intake also. Apparent inorganic arsenic intake levels show slight decreasing trend with time although not statistically significant. This apparent decrease may be due to increase of imported food items in the diet which come from Arsenic free areas.

5.3. Uncertainties in the Estimates

There are uncertainties in the values of Arsenic intake estimates through food consumption as given in Table 4 above. These uncertainties arise from the data and methodological limitations. The main limitations are the following:

- Absence of an agreed Market Basket definition for different age groups;
- Limited availability Arsenic concentration data in some food items;
- Limited availability of Arsenic speciation (inorganic and organic) data in some food items;
- Limited information on the distribution in the Arsenic concentration values in the same food item;
- Presence of analytical values below LOQ in the food items in surveys; and
- Analytical uncertainty in measurements due to noncompliance with QAQC procedures in some laboratories in Bangladesh.

The issue of distribution in the values of concentrations available has already been discussed earlier. This uncertainty

have been reduced to some extent by using the median values in this work. It makes a lot of difference in the final results whether zero values or LOQ are used for concentrations below the LOQ. For example, if LOQ values are used to calculate intakes when no arsenic is detected, the estimated arsenic intake may be an order of magnitude higher than that estimated when a value of zero is used.

6. Conclusions

The values for total and inorganic Arsenic of 2.92 ± 0.2 and 0.83 ± 0.06 $\mu\text{g/kg bw}$ per day respectively for the average intake by the average Bangladesh person, are comparable with those from some Asian countries, not affected by Arsenic contamination of groundwater, such as Japan and China as shown in Table 1. Present estimates for inorganic Arsenic intake through food is lower than the $3 \mu\text{g/kg bw}$ per day values quoted in the JEFCA[1] for BMDL 0.5. It is to be noted that the data quoted in JEFCA are from very a limited study by Rahman et al. (2008)[22] and Williams et al. (2005)[23].

A single duplicate diet study in Bangladesh as discussed in Section 4.2, also reported no incremental change in the Arsenic intake through consumption of food grown in an area irrigated with Arsenic contaminated water. So, in the light of the data available and the analysis presented in this work, the following conclusions can be made as to the current status of Arsenic exposure through food chain. These estimates, of course, have to be revised time to time as more quality assured new data on Arsenic level in different food regimes become available.

- (i). As no significant change in the intake level of 'Inorganic Arsenic' through food in Bangladesh has taken place during the past 20 years, there is no need for any restriction on the use of Arsenic contaminated groundwater for crop land irrigation.
- (ii). As there is no health based international guidelines for inorganic Arsenic intake any more, an acceptable limit has to be decided nationally probably based on cost benefit analysis just as in the case of water. However, provisionally BMDL0.5 value of $3 \mu\text{g/kg bw}$ per day

from JECFA [1] may be considered as the guideline for food and water combined as the risk is considered acceptable until a new value is accepted. This value is actually the same as the earlier Australia-New Zealand guideline.

- (iii). As data limitation is the main constraint in the accurate estimation of Arsenic ingestion, systematic and regular estimate of Arsenic species (inorganic and organic) in food should be carried out periodically (probably every five years) at country level. The Arsenic ingestion level for all age and sex groups should be determined using MB approach from such analytical data. This should be extended to all food contaminants, important from food safety point of view (i.e., toxic chemicals, heavy metals, pesticides and toxins in addition to Arsenic) for different age and sex group. An agreed definition of MB should be arrived at preferably by the Government. Some initiative like the Latin American Total Diet Study [5], may be undertaken at South Asia to reduce resource requirement for country level effort. As a number of agencies (i.e., scientific/ Academic institutions) available in Bangladesh which can do such work, only a decision by the government or a professional body is needed to get the work started.

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