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Arsenic toxicity through contaminated vegetables in selected blocks of Murshidabad and N-24 Pargana district, West Bengal, India

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ABSTRACT

An investigation was conducted to study the arsenic (As) accumulation in locally grown vegetables at selected blocks of Murshidabad and N-24 Pargana district, West Bengal, India and also to evaluate the possible health hazard on consumption among inhabitants. As concentration in ground water ranged from 0.25 -0.44 mg l^{-1} in Murshidabad and 0.15 -0.29 mg l^{-1} in N-24 Pargana. The cultivated soil of studied area are highly contaminated with As, using As rich ground water for irrigation, varied from 1.50– 6.8 mg kg^{-1}. The highest As accumulation was found in Potato (0.28±0.05 mg kg^{-1}) and (0.34±0.007 mg kg^{-1}) in Domkal and Beldanga respectively, whereas lowest value was found in Hyacinth Bean (0.050±0.03 mg kg^{-1}) and Bitter gourd (0.037±0.02 mg kg^{-1}) of the same site. Alike the highest and lowest accumulation was found in Potato (0.30±0.01 mg kg^{-1}) and Ridge gourd (0.04±0.01 mg kg^{-1}) for Bira and Potato (0.30±0.01 mg kg^{-1}) and Hyacinth Bean (0.03±0.002 mg kg^{-1}) for Deganga. Estimated daily intake (EDI) of As from vegetables were calculated and it was ranged from 0.102-0.23 mg/(kg bw/d) for Murshidabad and 0.09 -0.33 mg/(kg bw/d) for N-24 Pargana for adults respectively. Similarly the Target hazardous quotient (THQ) value ranged from 0.102-1.1 of the two studied area. THQ of As through vegetable consumption was greater than 1 at Deganga, indicates higher possibility of carcinogenicity by consumption of the As contaminated food. Also in case of other selected sites the THQ value was lower than 1, indicates it was still safe for consumption.

Key words: Arsenic accumulation, Estimated daily intake, Hazard quotient, Murshidabad, N-24 pargana.

INTRODUCTION

Arsenic (As) poses a severe threat to human health through exposure pathways mainly including water and diet. Its toxicity problem is widespread in the endemic areas of Ganges delta of West Bengal, especially, in irrigated crops. The first report of Groundwater As contamination and its health effects in the Ganga plain from West Bengal was published in 1984[1]. Under different geochemical condition As can be mobilized from soil to ground water [2,3]. Regarding the epidemiological study established a report on carcinogenic effect of As on humans and have related the element with numerous other skin diseases [4,5].In Taiwan, Japan, Bangladesh, Chile, Argentina and West Bengal, a large number of populations have exposed to arsenic-contaminated water causing higher cancerous risks for skin and other organs, including melanosis (hyper pigmentation or dark spots and hypo pigmentation or white spots, hyperkeratosis hardened skin), restrictive lung disease, peripheral vascular disease (black foot disease), gangrene, diabetes mellitus, hypertension, and heart disease [6-12]. The methodology for estimation of target hazardous quotient (THQ) indicates the ratio of the potential exposure to a substance and the level at which no adverse effects are expected. If the Hazard Quotient (HQ) is calculated to be less than 1, then no adverse health effects are expected as a result of exposure. If the Hazard Quotient (HQ) is greater than 1, then adverse health effects are possible.

Based on the report of Roychowdhury et al.[13] ,studying over the arsenic affected area of Murshidabad, West Bengal, it was found arsenic accumulation in various food composites like (potato skin, leaves of vegetables, rice, wheat, cumin, turmeric powder, and cereals) ranged between <0.0004 and 0.693 mg kg-1. Vegetable is an important part of the human diet after rice, so for most of the people it is also the main route of exposure to toxic element through dietary intake [14,15]. The food crops grown using arsenic contaminated water are sold off to other places, including uncontaminated regions where the inhabitants may consume arsenic from the contaminated food. A number of study were carried out in As affected areas of Bengal delta plain [16,17,18-30].

The objectives of the present study are: (1) to determine the accumulation of arsenic concentration in water-soil-vegetables at two studied location of Murshidabad district throughout the year. (2) Estimation of the daily intake through consumption of these contaminated food. (3) To evaluate the potential risk of arsenic from contaminated vegetable consumption to human health through target hazardous quotient (THQ).

MATERIALS AND METHODS

Sampling location:
The study area Murshidabad (24 º 50´ N and 23º 43´ N and 88º 46´E and 87º 49´ E) and N-24 Pargana (22º48.467´´N88 º38.302´´E) district, West Bengal, India is highly contaminated with arsenic [31,32,13]. Two blocks, namely Beldanga, Domkal of Murshidabad district and for N-24 Pargana district, Bira, Deganga blocks has been chosen for the present study. In all these areas the level of arsenic in groundwater exceeding WHO permissible limit for drinking water (0.01 mg l^{-1})[33] and Food and Agricultural Organization permissible limit for irrigation water (0.10 mg l^{-1}**)** [34]**.**

Vegetable, Water, soil, sampling:
The vegetable samples were collected seasonally (pre monsoon, monsoon, post monsoon) from this area to analyze the level of total arsenic content. Groundwater samples have been collected from shallow tube well pumps with large diameter in 100ml bottles with replica (n=3), and preserved with 1ml/L concentrated HNO_3. Surface soil samples were collected from 0-10 cm depth and sub-surface soil samples were collected from 10-50cm depth by composite sampling from the fields and transferred it to into air-tight polythene bags.

Sample Treatment:
Plant samples were washed thoroughly with tap water followed by de-ionised water for several times. Finally the samples were dried in hot air oven at 50º-60ºc for 72h, powdered and stored in air tight polythene bags at room temperature. Water samples were filtered through 0.45µ Millipore filter paper and the filtered samples will kept in polythene bottles at 4ºc prior to analysis. The soil samples were immediately sun-dried after collection and later dried in hot air oven at 60ºc for 72h, grind and screened. Finally the samples were stored in air tight polythene bags at room temperature.

Sample Digestion:
Vegetables and soil samples were digested separately following heating block digestion procedure [24]**.** Plant samples were digested by adding perchloric acid ($HClO_4$), sulphuric acid (H_2SO_4) and nitric acid (HNO_3) at 1:1.5:4 ml ratio. The tubes were placed on a heating block at 110º-120ºc until get clear solution. The samples were cooled, diluted to 25ml with de-ionized water and filtered with Whatman No. 41 filter paper. For soil sample, concentrated H_2SO_4 was added in addition to concentrated $HClO_4$ at 3ml: 2ml ratio. Then the tubes were heated at 160ºC for about 4-5h. The heating will be stopped when the dense white fume of $HClO_4$ was emitted. The content was then cooled, diluted to 25ml with de-ionized water, and filtered through Whatman No. 42 filter paper.

Analysis of total arsenic:
The total arsenic of samples were analyzed by flow injection hydride generation atomic absorption spectrophotometer (FI-HG-AAS, Perkin Elmer AAnalyst 400) using external calibration [35]. The optimum HCl concentration was 10% v/v and 0.4% $NaBH_4$ produced the maximum sensitivity. Standard reference materials (SRM) from National Institute of Standards and Technology (NIST), USA, were analyzed in the same procedure at the start, during, and at the end of the measurements to ensure continued accuracy.

Data analysis:
Estimated Daily Intake of arsenic from vegetables:
Estimated daily intake (EDI) (µg/(kg body weight (bw.d) of arsenic from vegetable consumption was obtained by Eq. (1):

$$EDI = (C \times F_{IR})/ W_{AB} \quad (1)$$

where, C (µg/g fw) is an average weighted arsenic content in the edible portion of the vegetable, and is calculated by multiplying the mean arsenic content in vegetables with their corresponding percentage of consumption. F_{IR} (g/d/person) is a daily vegetable consumption [36].

Health Risk Assessment:
Health risk is associated through As contaminated vegetables consumption based on the ratio between the exposure and reference dose (RfD) and is used to express the risk of non-carcinogenic effects and were assessed using Target Hazardous Quotient (THQ) [38,39,40,41]. This method is available in US EPA Region III Risk-based Concentration table [40] and it is described by the following equation:

$$THQ= 10^{-3} (EF \times ED \times FIR \times C/RFD \times WAB \times TA) \quad (2)$$

Where EF is the exposure frequency (365 days/yr), ED is the exposure duration (70yrs), FIR is the food ingestion rate (g/person/day), C is the concentration of food, RFD is the oral reference dose; As = 3×10^{-4} mg/kg/day [39], WAB is the average body weight (60kg), TA is the average exposure time for non-carcinogens (365 days/ year X number of exposure years, assuming 70 yrs in this study).

RESULTS AND DISCUSSION

Arsenic concentration in irrigation water and soil:
Agriculture is the most important livelihood in the study area and is in practice throughout the year using groundwater as the main source for irrigation. Shallow tube well pumps are used for irrigation in this area (running 8h-10h per day). From this study, mean arsenic concentration of water sample ranged from 0.28– 0.38 mg lt^{-1} in Domkal and 0.25 - 0.44 mg l^{-1} in Beldanga. Likewise from Bira and Deganga block, arsenic concentration of irrigated water ranged from 0.15-0.20 mg l^{-1} and 0.20-0.29 mg l^{-1} (Table 3). Results revealed that the As content in irrigated water collected, was many fold higher than WHO/FAO permissible limit. The heavy withdrawal of ground water may be the reason of iron pyrite decomposition and release of arsenic in water [41,42].

The total As concentration in investigated soil ranged from 2.85 – 6.8 mg kg^{-1} in Domkal and 2.54 - 4.09 mg kg^{-1} Beldanga (Table 3), correspondingly in Bira and Deganga block soil As content varied from 1.50-2.13 mg kg^{-1}(Table 3). Previously it has been said that arsenic accumulation in soil is due to irrigate with arsenic-contaminated ground water in West Bengal **[**43,44]. Thus the results clearly showed that extensive use of arsenic-contaminated ground water in the study site for irrigation may increase level of arsenic in soil. The highest content of arsenic in soil was reported (19.4 mg kg-1) in West Bengal [45]. The accumulation of arsenic in soil of the study area was lower than the reported global average of 10.0 mg kg^{-1} [46] and was below the maximum acceptable limit for agricultural soil of 20.0 mg kg^{-1}, recommended by the European Community [24]. Thus there is a possibility of increasing arsenic concentration in soil in future if the trend of ground water irrigation will not restrict.

Uptake of arsenic by vegetables :
The present study were designed to monitor total arsenic content of collected vegetables from Murshidabad and N-24Parganas districts. Table 1 and Figure 1 describes the mean As content from seasonally collected vegetables of Domkal and Beldanga block, ranged from (0.05-0.28 mg kg^{-1}) i.e Hyacinth Bean and Potato, and (0.037-0.34 mg kg^{-1}) i.e, Bitter gourd and Potato. Similarly in case of Bira and Deganga block, N-24 Pargana district mean As content of collected vegetables was ranged 0.04-0.29 mg kg^{-1} i.e, Ridge gourd and Potato, and 0.03-0.30 mg kg^{-1} i.e, Hyacinth Bean and Potato ,Table 2 and Figure1. Das et al [47], from their study in Bangladesh, also found higher arsenic contents in potatoes (0.07– 1.36 mg kg-1). But, very low arsenic accumulation in potato (<0.01 mg kg-1) was reported from a study in Nepal. Food composites (potato skin, leaves of vegetables, rice, wheat, cumin, turmeric powder, and cereals, etc.) collected from As-affected areas of the Murshidabad district, West Bengal, contained 7–373µg kg^{-1}asenic [13]. The uptake of arsenic by agricultural plants depend on water requirement, soil properties [48,49,50]. Previously it was predicted that the higher concentrations of arsenic was found in potato, arum, amaranth, radish, lady's finger, cauliflower, brinjal, etc., is due to irrigated with arsenic contaminated water [51,52,13]. Thus the arsenic content in the analyzed sample of the study area is more or less equivalent comparing with the reported value of cultivated vegetables of Nadia district [53].

Food habit and arsenic intake of local inhabitants:
Rice and vegetables are the main staple food among the local people of the study area. After learning the arsenic rich exposure from food and drinking water, we have studied arsenic in vegetables. After surveying randomly 30-40 family, it has been found that, normally people of those areas eat rice, vegetables and dal three times per day (Table 4). Adult (both male and female) normally eat 150gm of rice with 100gm of mixed vegetables in morning and 250gm of rice at lunch and dinner with around 200gm and 150 gm of mixed vegetables at each meal. The average mixed vegetable taken by adults are 450 gm respectively i.e. 90gms in dry weight (about 80% moisture content in vegetables).

Table 1- Seasonal variation of arsenic concentration (mean ± SD) (mg kg^{-1}) in vegetable sample of Murshidabad district

Blocks	Type of vegetables in Pre Monsoon	As concentration (mean ± sd) (mg kg^{-1})	Type of vegetables in Pre Monsoon	As concentration (mean ± sd) (mg kg^{-1})	Type of vegetables in Pre Monsoon	As concentration (mean ± sd) (mg kg^{-1})
Domkal 24º06.323´´N 88 º32.796´´E	Beans (n=8)	0.078±0.05	Potato (n=5)	0.23±0.02	Lady's finger (n=10)	0.077±0.05
	Carrot (n=4)	0.12±0.03	Chilli (n=8)	0.074±0.02	Brinjal (n=2)	0.072±0.03
	Potato (n=6)	0.13±0.05	Hyacinth Bean (n=8)	0.050±0.03	Potato (n=7)	0.28±0.05
	Brinjal (n=3)	0.06±0.01	Red Amaranth (n=10)	0.072±0.03	Spring onion (n=12)	0.21±0.06
	Spring onion (n=10)	0.086±0.04	Lady's finger (n=7)	0.077±0.04	Onion (n=5)	0.063±0.002
	Onion (n=2)	0.10±0.002	Bottle gourd (n=3)	0.075±0.05	Cabbage (n=1)	0.17±0.04
	Cabbage (n=2)	0.20±0.03	Pumkin (n=1)	0. 106±0.01	Cauliflower (n=1)	0.23±0.03
	Cauliflower (n=4)	0.18±0.04			Carrot (n=4)	0.092±0.03
Beldanga 23º59.150´´N 88 º14.389´´E	Potato (n=4)	0.34±0.007	Red Amaranth	0.10±0.03	Ridge gourd (n=4)	0.065±0.05
	Pumpkin (n=2)	0.24±0.008	Bottle gourd (n=2)	0.056±0.008	Hyacinth Bean (n=8)	0.048±0.04
	Tomato (n=4)	0.18±0.007	Biiter gourd (n=6)	0.037±0.03	Bottle gourd (n=1)	0.11±0.05
	Spinach (n=10)	0.15±0.03	Chilli (n=12)	0.082±0.003	Pumkin (n=1)	0.083±002
	Red Amaranth (n=10)	0.085±0.08			Tomato(n=5)	0.19±0.04
	Bitter gourd (n=6)	0.053±0.08				
	Lady's finger (n=7)	0.052±0.030				

**n= No of sample analyzed*

Table 2- Seasonal variation of arsenic concentration in vegetable sample of N-24 Pargana district

Blocks	Type of vegetables in Pre Monsoon	As concentration (mean ± sd) (mg kg^{-1})	Type of vegetables in Pre Monsoon	As concentration (mean ± sd) (mg kg^{-1})	Type of vegetables in Pre Monsoon	As concentration (mean ± sd) (mg kg^{-1})
Bira 22º48.467´´N 88 º38.302´´E	Beans (n=8)	0.17±0.05	Chilli (n=10)	0.10±0.02	Brinjal (n=4)	0.09±0.02
	Carrot (n=6)	0.24±0.05	Parval (n=4)	0.08±0.05	Ridge gourd (n=6)	0.04±0.01
	Potato (n=4)	0.30±0.01	Bitter gourd (n=6)	0.05±0.03	Bottle gourd (n=6)	0.08±0.01
	Brinjal (n=5)	0.16±0.001	Pui saak (n=6)	0.06±0.03	Hyacinth bean (n=12)	0.04±0.03
	Spinach (n=8)	0.13±0.03	Radish (n=4)	0.07±0.02	Bitter gourd (n=10)	0.08±0.02
	Red amaranth (n=6)	0.08±0.05				
	Carrot (n=4)	0.29±0.01				
Deganga 22º41.100´´N 88 º39.525´´E	Tomato (n=8)	0.15±0.01	Chilli (n=12)	0.10±0.04	Pumkin (n=2)	0.10±0.01
	Brinjal (n=6)	0.17±0.001	Parval (n=6)	0.08±0.001	Tomato (n=11)	0.13±0.05
	Sweet potato (n=8)	0.26±0.01	Pumkin (n=2)	0.08±0.001	Potato (n=9)	0.24±0.04
	Cauliflower (n=4)	0.19±0.005	Lady's finger (n=10)	0.07±0.001	Ridge gourd (n=7)	0.04±0.003
	Onion (n=14)	0.29±0.002	Ridge gourd (n=4)	0.07±0.001	Hyacinth bean (n=14)	0.03±0.02
	Potato (n=8)	0.30±0.01	Bitter gourd (n=8)	0.06±0.001	Lau (n=6)	0.06±0.002
					Cauliflower (n=3)	0.20±0.03

**n= No of sample analyzed*

Table 3- Seasonal variation of arsenic concentration of water and soil samples of Murshidabad and N-24 Pargana district

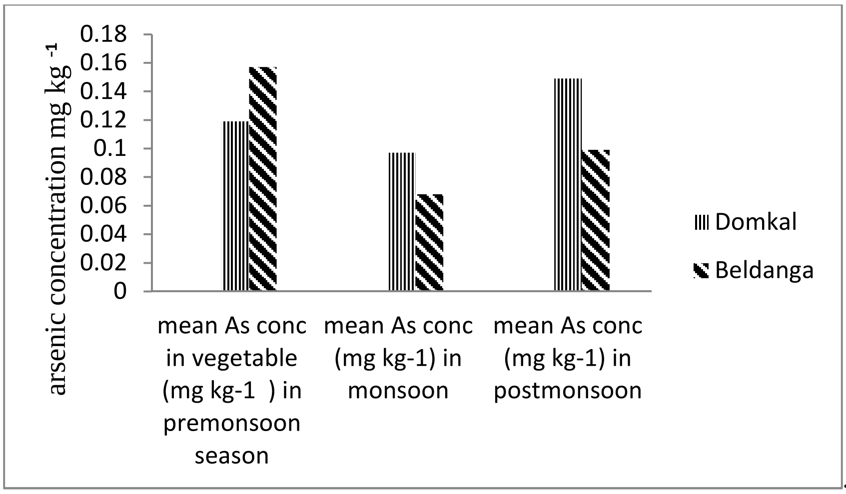
Location	Block	Average arsenic concentration (mean±sd) in Pre monsoon		Average arsenic concentration (mean±sd) in Monsoon		Average arsenic concentration (mean±sd) in Post Monsoon	
		Water (mg/l)	Soil (mg/kg)	Water (mg/l)	Soil (mg/kg)	Water (mg/l)	Soil (mg/kg)
Murshidabad	Domkal	0.38±0.10	6.8±0.22	0.28±0.06	2.85±0.05	0.33±0.05	6.44±0.05
	Beldanga	0.44±0.05	4.02±1.16	0.31±0.04	2.54±0.85	0.25±0.05	4.09±0.87
N-24 Pargana	Bira	0.19±0.06	1.93±0.07	0.15±0.06	1.50±0.04	0.20±0.03	1.87±0.18
	Deganga	0.26±0.01	2.13±0.04	0.20±0.02	1.50±0.18	0.29±0.04	1.77±.17

Table 4- Dietary components of local inhabitants of the studied area

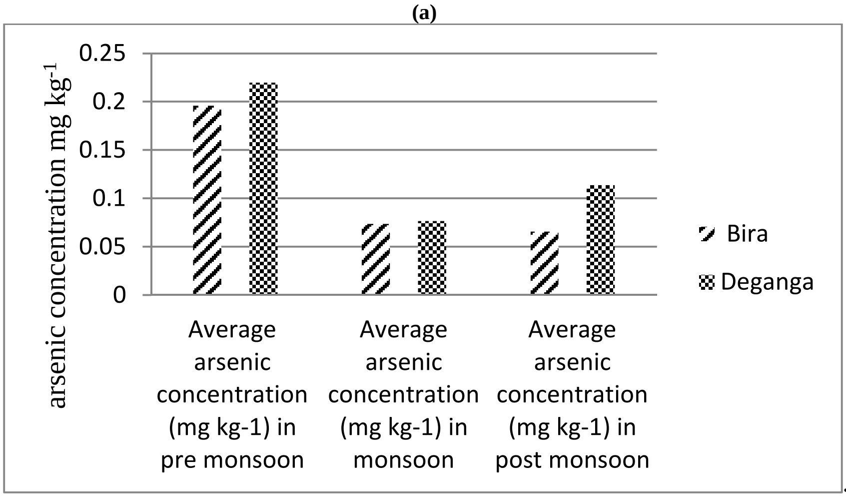
	Morning	Lunch	Dinner	Total
Adult	Rice (150g), Vegetable (100g)	Rice (250g), Vegetable (200g)	Rice (250g), Vegetable (150g)	Rice (650g), Vegetable(450g)

Table 5- Estimated daily intake (EDI) and Target hazardous quotient (THQ) of arsenic from vegetables

Location	Block	Estimated daily intake mg/(kg bw/d)			Target hazardous quotient (THQ)		
		Premonsoon	Monsoon	Post monsoon	Premonsoon	Monsoon	Post monsoon
Murshidabad	Domkal	0.178	0.145	0.223	0.59	0.49	0.74
	Beldanga	0.23	0.102	0.148	0.78	0.34	0.49
N-24 Pargana	Bira	0.28	0.110	0.09	0.98	0.37	0.33
	Denganga	0.33	0.115	0.17	1.1	0.38	0.57



(a)



(b)

Figure1. Mean arsenic concentration (mg kg^{-1}) of vegetable samples in three season of the studied area of (a) Murshidabad district and (b) N-24 Pargana district

Estimated daily intake (EDI) and Target Hazardous quotient (THQ) of arsenic from vegetables consumption:
Estimated daily intake (EDI) and target hazardous quotient (THQ) of arsenic were calculated for local people considering the average concentration of As through consumption of mixed vegetables (Table 5). Comparing the

EDI value of three season Table 5 and Figure 2, higher intake of As from mixed vegetables, was found in post monsoon, i.e, 0.223 mg/(kg bw/d) in Domkal block and 0.23 mg/(kg bw/d) for pre monsoon in Beldanga block for adult age group, which means the collected mixed vegetables for this season were accumulating a large amount of As from irrigation water. Similarly higher EDI value was found in pre monsoon at Bira and Deganga, i.e, 0.28 mg/(kg bw/d) and 0.33 mg/(kg bw/d).Incase of Domkal block the EDI value for pre monsoon and monsoon was 0.178 mg/(kg bw/d) and 0.145 mg/(kg bw/d) whereas in Beldanga block the value was 0.102 mg/(kg bw/d) and 0.148 mg/(kg bw/d) for monsoon and post monsoon. Thus the As intake from vegetable was lower than 0.463 µg/(kg bw/d) that for adults in Bangladesh [51] and higher than 0.174 µg/(kg bw/d) for adults in West Bengal, India [15]. Based on toxicological evaluation of suggested the provisional daily intake (PTDI) for inorganic arsenic was 2.1 µg/(kg bw/d) or (0.0021 mg/ kg body wt. per day). Considering the seasonal consumption of As through vegetables, it can be say that vegetables which grown in post monsoon and pre monsoon of the studied region, might be cause more risk for human health comparing two other season.

The THQ values of As for inhabitants were shown in Table 5 and Figure 2. The range of THQ value was decreasing seasonal order postmonsoon>premonsoon>monsoon (ranged 0.74-0.49) in Domkal block and premonsoon>postmonsoon>monsoon (ranged 0.78-0.34) in Beldanga block. Results revealed all the THQ values were not exceed the permissible limit (THQ > 1), so there may be a possibility of low risk exposure from arsenic contaminated vegetables of those two block. In case of Bira and Deganga block the range of THQ value was ranged from 0.98-0.33 (premonsoon>monsoon>postmonsoon) and 1.1-0.38 (premonsoon>postmonsoon>monsoon) respectively. The risk from As exposure from vegetables is higher in postmonsoon and premonsoon season of Domkal and Beldanga block.

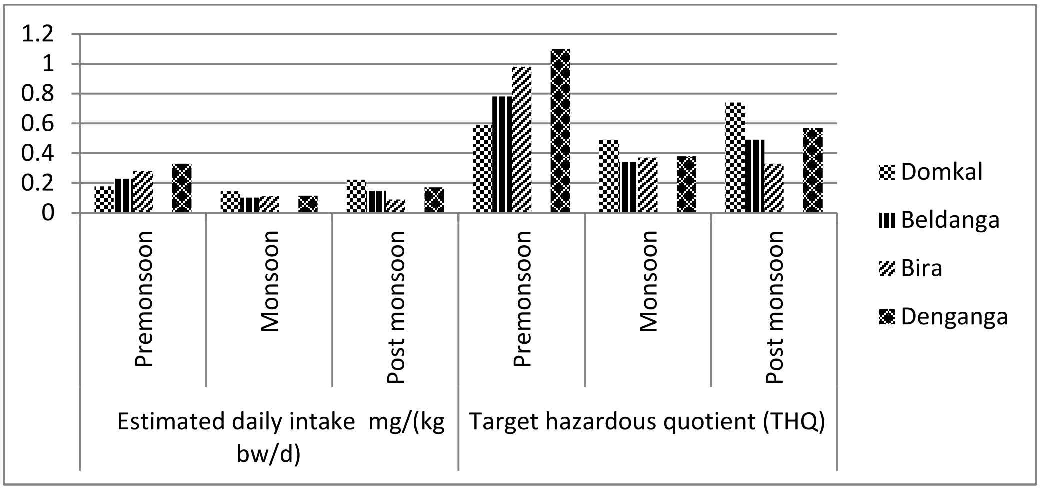


Fig 2- Study of estimated daily intake and target hazardous quotient of the study area

CONCLUSION

The results of this study revealed that due to uncertainty of rainfall, most of the area under the two studied block use primarily ground water for irrigation purposes. The ground water arsenic content was high than the permissible limit proposed by WHO. Analyzing the severity of arsenic toxicity in ground water as well as soil, their uptake capabilities, which influenced the irrigated vegetables with respect to seasonal variation. A significant As contamination was found in soil as well as irrigated vegetables using As contaminated ground water. Therefore there is a probability of entry of arsenic into the food chain through water soil vegetable pathway. Both the EDI and THQ value from As accumulation were exceed the maximum limit and as a result it may cause health risk to the communities. In conclusion frequent monitoring of arsenic contamination in vegetables grown at the irrigated area is necessary and consumption of contaminated vegetables should be avoided in order to reduce health risk caused by taking contaminated vegetables.

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