



# Characterization and risk assessment of arsenic contamination in soil–plant (vegetable) system and its mitigation through water harvesting and organic amendment

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**Abstract** Field experiments with vegetables [cauliflower (*Brassica oleracea* var. *botrytis*), tomato (*Solanum lycopersicum*) and spinach (*Spinacia oleracea*)] were conducted at geogenically arsenic-contaminated Ghentugachi village in West Bengal, India, for two consecutive years to study arsenic accumulation by the selected vegetables and to explore the efficiencies of use of harvested/harnessed water and organic amendments (Mustard Cake, Vermicompost and Farm Yard Manure) in reducing arsenic load in soil–plant system. Results revealed that arsenic accumulations in the cauliflower head, spinach leaf and tomato fruit were in the range of 0.15–0.17, 2.73–3.00 and 0.08 mg kg<sup>-1</sup>. Organic amendment and pond

water irrigation when applied either separately or together were found to be effective in reducing arsenic contamination in soil–plant system compared to irrigation with shallow tube well-drafted underground water. Vermicompost remained most successful among the organic amendments. Conjunctive use of surface (pond) and ground water also significantly reduced the level of arsenic in the system. The risk of dietary intake of arsenic through the selected vegetables was computed through % Provisional Tolerable Weekly Intake, Hazard Quotient and Target Cancer Risk. Cauliflower and tomato were found safe in the individual contribution to food chain, while consumption of spinach leaf (possessing 10.4–22.6% more arsenic than maximum tolerable limits) remained unsafe in all dietary risk measures.

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## Introduction

As a toxic, ubiquitous metalloid, arsenic (As) has been a major contaminant of soil and water in many countries (Chen et al. 2017; Bhattacharyya and Sengupta 2020). Its carcinogenic ability accompanied by long-time toxic persistence has emerged as a global concern (Shakoor et al. 2019). While As

contamination in drinking water has been the major concern, the menace of As contamination through consumed food and water with a high proportion of As transfer through soil-crop-food chain has been recently reported (Mondal et al. 2018; Mandal et al. 2019a). The transfer of As from soil to the edible parts of plant imparts dietary dilemma and adds risk to human health (Meharg et al. 2009; Zhao et al. 2010; Wang et al. 2018).

The characters of the metalloid As with a density greater than  $5 \text{ g cc}^{-1}$  contemplate with its categorization as ‘heavy metals/transition/post-transition metals’ (Fowler 1993; Hawkes 1997; Wu et al. 2016). Similar to other heavy metals and their associated toxicity mechanisms, As in the living body is also highly correlated with toxicity mechanisms like production of reactive oxygen species, replacement of essential elements in molecular structures of proteins and enzymes, blocking of functional groups and thereby leads to malfunctioning of biochemical and physiological processes and ultimately leads to severe oxidative stress, cancer and death of species (Wang and Fowler 2008; Tchounwou et al. 2012; Engwa et al. 2019; Fu and Xi 2020).

The present study area of Chakdah block in Nadia district is an important vegetable agri export zone (AEZ) (<http://agriexchange.apeda.gov.in>), which has accrued a major setback due to presence of high As in the marketable commodity often exceeding the export threshold guidelines (Das et al. 2016). High As accumulation in daily consumed vegetables in the contaminated area as evident from several studies cannot be denied as well (Meharg and Rahman 2003; Biswas et al. 2012). Significant accumulation of arsenic ranging between 0.01 and  $0.69 \text{ mg kg}^{-1}$  was reported in potato skins, vegetable leaves and some spices and cereals (Roychowdhury et al. 2002). In separate experiments from adjoining belt, the total mean arsenic content in vegetables was found to vary between 0.03 and  $0.41 \text{ mg kg}^{-1}$  (Rahaman et al. 2013) and 0.03 and  $0.65 \text{ mg kg}^{-1}$  (Bhattacharya et al. 2010), respectively. Vegetables, owing to substantial presence of arsenic, significantly victimize the community as a major dietary component of the area under study (Santra et al. 2013; Biswas et al. 2019).

Harvested rain water in ponds contains less arsenic than STW-pumped groundwater. Mondal et al. (2015) reported crops irrigated with pond water had lower As content. Usefulness of organic amendments to reduce

As entry into soil solution and plants through organo-As chelation has also been reported (Kumpiene et al. 2008; Ahmad et al. 2014; Khan et al. 2015; de Oliveira et al. 2017; Mandal et al. 2019a).

An alarming finding was reported by Guha Mazumder et al (2013) showing As transfer to humans even in some low-level contaminated areas via soil-crop-dietary route where As in drinking water was within the WHO permissible level. The human dietary risk to As through food items and water has been expressed by % PTWI, hazard quotient (HQ) and target cancer risk (TCR) (Datta and Young 2005; Rattan et al. 2009; Singh and Ghosh 2012; Chang et al. 2014; Antoine et al. 2017; Golui et al. 2014, 2017; Biswas et al. 2018; Mandal et al. 2019a).

Highly precise prediction of the free ion activity of As present in soil solution can predict its entry into the plant (Mandal et al. 2019b). The adoption of simple solubility-free ion activity model can be used as a good tool for predictability of As movement from soil to plant (Golui et al. 2014, 2017; Mandal et al. 2019b). Based on simple soil physicochemical properties, this model has already been used for As mobility from soil to rice grain collected from 28 different rice cultivating families of Malda, West Bengal, India (Golui et al. 2017); however, its usefulness for irrigated vegetables with organic amendments has not been tested before.

The present study focuses on (i) assessing As load in cauliflower, tomato and spinach, (ii) adopting water harvesting and organic amendments to minimize As load in soil-plant system and (iii) calculating the dietary risk of the As through consumption of the selected vegetables.

## Materials and methods

### Site selection and experimental details

The experiment was conducted in a farmer’s field ( $23^{\circ} 02' \text{ N}$ ,  $88^{\circ} 34' \text{ E}$ ) at Ghentugachi village of Chakdah block in Nadia of West Bengal, India. The study area has a typical sub-tropical climate with average annual rainfall diverging from 1200 to 1500 mm, relative humidity from 36 to 85%, and the maximum and minimum temperature by average is about  $37.5^{\circ} \text{ C}$  and  $12^{\circ} \text{ C}$ . The soil is a new alluvial (*Inceptisol*), characterized as silty clay with neutral soil pH, moderate in

available N, K and high in available P. Elevated levels of As in soil and water, especially in shallow tube well (STW) water in comparison with pond water (PW) as obtained in the study, has similarity with the findings of Das et al. (2016) for the same area. Local popular varieties of the selected winter vegetables *e.g.*, cauliflower (*Brassica oleracea* var. *botrytis* cv. Snow ball), tomato (*Solanum lycopersicum* cv. PAN 1286) and spinach (*Spinacia oleracea* cv. Bengal King), were grown in a thrice replicated factorial randomized block design, where one factor is source of water (three levels, as,  $I_1$  = Shallow Tube Well;  $I_2$  = 50% Pond + 50% Shallow Tube Well irrigation and  $I_3$  = Pond water) and the other factor was organic amendment (four levels,  $F_1$  = Control;  $F_2$  = Vermicompost @ 3 t ha<sup>-1</sup>;  $F_3$  = FYM @ 10 t ha<sup>-1</sup>;  $F_4$  = Mustard Cake @ 1 t ha<sup>-1</sup>).

#### Agronomic practices of the crops

The site for carrying out the experiment has been selected in such a way that there is no chance of water stagnation. In total, 36 plots, each of 3 m × 4 m size, were selected and the treatments were randomized in thrice replicated blocks. Soils were repeatedly plowed and pulverized to a fine tilth. Separate water channels for pond, STW water and combined treatments were prepared. The organic amendments in the corresponding plots (Vermicompost @ 3 t ha<sup>-1</sup>; FYM @ 10 t ha<sup>-1</sup> and Mustard Cake @ 1 t ha<sup>-1</sup>) were applied during final land preparation and mixed with the soil. In nursery bed, cauliflower and tomato seeds were sown in middle of October and thereafter transplanted in the experimental field in mid-November in 60 cm × 45 cm spacing. The spinach seeds were directly sown in experimental field in early to mid-November in 30 cm × 5 cm spacing. The same protocol was practiced for both the study years of 2016–2017 and 2017–2018. N, P and K fertilizers were applied at their recommended dose (125:75:60 kg ha<sup>-1</sup> for cauliflower, 70:50:30 kg ha<sup>-1</sup> for spinach and 100:50:50 kg ha<sup>-1</sup> for tomato). 1/3 N and full P and K were applied as basal dose just before transplanting, while remaining 2/3 N was applied in two split doses at intervals of one month. A pre-sowing or transplanting irrigation was given to ensure sufficient moisture for germination and crop stand. Thereafter irrigations were provided at 7–10-day interval (based on climate). The As-contaminated pond and

STW water and their combinations were allocated in the respective plots for irrigating the crops. Fields were kept weed free by frequent weeding, hoeing and earthing up without any chemical herbicide application. Chemical pesticides were primarily restricted in the cultivation protocol. Integrated pest management strategies involving erecting of yellow sticky traps, pheromone traps, application of neem oil (5 ml per liter of water in conjunction with sticker) and 4% neem seed kernel extract (NSKE) have been employed based on the pest incidence. The harvesting of the crops was initiated in early January for spinach, end of January for cauliflower and early to mid-February for tomato and continued at staggered dates, to ensure continuous harvest over extended period. The harvested plants and root zone soils were collected from each plot leaving the plants at the edge to reduce the border effect. The edible portion of plants was separated for As analysis.

#### Sample collection and preparation

Post-harvest (PH) top-soil (0–15 cm) samples were collected, air-dried, ground, sieved (2-mm sieve) and finally stored in pre-marked airtight polythene packets. Soil pH was measured in 1:2 soil:water suspension (Datta et al. 1997) along with the electrical conductivity (Jackson 1973). Soil organic C was determined by Walkley and Black (1934) method, while soil N, P and K were determined by the standard methods of Subbiah and Asija (1956), Olsen and Sommers (1982) and Knudsen et al (1982), respectively. Hydrometer method was employed for clay content determination (Bouyoucos, 1962). Soil available As content was determined by Olsen (NaHCO<sub>3</sub>) extractable As (Johnston and Barnard 1979), while the total As content of soil and water samples was determined by the method of Sparks et al. (2006).

The edible plant samples (cauliflower head, tomato fruit and spinach leaves) were collected, thoroughly washed with tap water followed by dilute acid solution and finally by double-distilled water. They were appropriately labeled, chopped and dried in an air-oven at 105 °C for 24 h, cooled to room temperature, ground and acid digested using a mixture of HNO<sub>3</sub>, HClO<sub>4</sub> and H<sub>2</sub>SO<sub>4</sub> @ 10:4:1 (v/v) as in Jackson (1973). After digestion samples were filtered for chemical analysis.

## Instrumental analysis

The plant digest and soil extract were diluted with distilled water, reacted with concentrated HCl, KI and ascorbic acid for 45 min and then analyzed by atomic absorption spectrophotometer with the standard instrumental procedures as described by Sparks et al. (2006) and Giri et al. (2012).

The methodology of As determination was validated using standard reference materials of rice (SRM1568a), prepared by National Institute of Standards and Technology (NIST). The certified value of SRM1568a is  $290 \pm 30 \mu\text{g kg}^{-1}$ . Analysis of standard material in the current PerkinElmer AAnalyst 200 Atomic Absorption Spectrophotometer (AAS) attached with Flow Injection for Atomic Spectroscopy (FIAS) Systems at  $\lambda_{\text{max}} = 193.7 \text{ nm}$  was carried out in triplicate, and As content was recorded as  $287 \pm 8.1 \mu\text{g kg}^{-1}$ . The observed value of tri-acid digest, determined through PerkinElmer FIAS 400 attached AAnalyst 200 AAS, was thus in good agreement with the certified value of the material. In every batch of 30 samples, two blank reagents and one standard reference material were analyzed for validating the accuracy.

## Predicting As content in selected vegetables

A theoretical approach of simulating the As content in edible parts of vegetable can be assessed by the solubility-free ion activity model without actually measuring As or rather even when the crop is not grown as previously reported by Hough et al. (2005), Datta and Young (2005) and Mandal et al. (2019a). The model is based upon consideration of some simple physico-chemical parameters of the soil. The free ion activity of As in soil solution is considered as a function of labile soil extractable As and pH with the assumption that entire amount of metalloid is adsorbed on humus (Meena et al. 2016).

A simple pH-dependent Freundlich equation is used in the model to predict the As uptake in plant based on transfer factor (TF), the quotient of metalloid concentration in plant ( $M_{\text{plant}}$ ) and its content in soil pore water ( $M^{n-}$ ) (Jopony and Young 1994; Mirecki et al 2015).

$$\text{TF} = \frac{M_{\text{plant}}}{M^{n-}} \quad (1)$$

$$p(M^{n-}) = \{p[M_c] + k_1 + k_2pH\}/nF \quad (2)$$

where  $M_c$  is the labile As pool adsorbed on humus ( $\text{mol kg}^{-1}$  carbon);  $k_1$ ,  $k_2$  and  $nF$  forms the Freundlich equation's empirical constants and power term. Combining Eqs. (1) and (2), the equation for As uptake becomes:

$$p[M_{\text{plant}}] = C + \beta_1 p[M_c] + \beta_2 pH \quad (3)$$

where

$$C = k_1/nF - \log\text{TF}; \beta_1 = 1/nF \text{ and } \beta_2 = k_2/nF$$

$C$ ,  $\beta_1$  and  $\beta_2$  are being plant and metal(loid)-based constants computed by 'SOLVER' facilities of *Microsoft Excel* through evading of nonlinear errors (Golui et al. 2017). The parameter  $C$  is specific to As, while  $\beta_1$  and  $\beta_2$  are crop specific and thereby can easily predict As hazard by knowing soil pH and organic carbon of the site, integrating soil-crop metal transfer pathway.

Risk assessment of dietary exposure to As through selected vegetables

## % Provisional tolerable weekly intake (PTWI)

Vegetables grown in As-contaminated soils result in substantial uptake leading to dietary risks. The weekly As intake from each vegetable (mg per week) is simply the product of As concentration ( $\text{mg kg}^{-1}$ ) and the rate of human dietary consumption (kg per week) of that vegetable category based on their wet weight. Relating the Joint FAO/WHO Expert Committee on Food Additives (JECFA) values of PTWI with the National Nutritional Monitoring Bureau (NNMB) dataset, a factor of 0.9 was divided to get actual PTWI (Guha Mazumder et al. 2013).

$$\text{PTWI}\% = [(\text{As conc.} \times \text{weekly consumption}^* \times 100) / \text{JECFA, FAO - WHO PTWI}]$$

(\*Data obtained from National Nutritional Monitoring Bureau- NNMB 2012).

*Target cancer risk (TCR)*

Assessment of carcinogenic risks to a human throughout his life can be computed by target cancer risk (TCR) (Antoine et al. 2017). The equation for TCR is:

$$TCR = \frac{Efr \times Ed \times Fir \times C \times CPSo}{BWa \times ATc} * 10^{-3}$$

where Efr is the frequency of As exposure, Ed is the duration of exposure as 70 yrs, Fir is the ingestion of each food category in gram per day, C is the As concentration in the food item, CPSo is the oral slope factor for cancer occurrence, 1.5 mg As kg<sup>-1</sup> day<sup>-1</sup>, BWa is the average human body weight (68 kg), ATc is the average time of carcinogenic exposure as product of 365 days\*70 yrs and 10<sup>-3</sup> being the factor for conversion of units. The acceptable range of TCR is between 10<sup>-4</sup> and 10<sup>-6</sup> (1 in 10,000 to 1 in 1,000,000) (Antoine et al. 2017).

*Hazard quotient (HQ)*

The US Environmental Protection Agency (USEPA) has used HQ as another index of computing human dietary risk (IRIS 2020). It is the ratio of average daily dose (ADD) and reference dose of the contaminant (RfD). Thereby, HQ values provide an indication for maximum tolerable limit of As avoiding human health consequences (mg As.kg body wt<sup>-1</sup> day<sup>-1</sup>) (Pierzynski et al. 2000):

$$HQ = \frac{ADD}{RfD}$$

If HQ > 1, then ADD > RfD, indicating toxicity of As. With the assumptions that 2.1 µg As kg body wt<sup>-1</sup> day<sup>-1</sup> as the provisional maximum tolerable daily intake (PMTDI) (WHO 1996), daily intake of cauliflower, spinach and tomato is 0.08, 0.06 and 0.05 kg day<sup>-1</sup>, respectively (NNMB 2012), and average body weight for an adult is 68 kg (Rahaman et al. 2013); the final equation for HQ computation becomes:

$$HQ = \frac{M_{plant} \times W}{RfD \times 68}$$

where M<sub>plant</sub> is As content (mg kg<sup>-1</sup>) and W is the daily intake of selected vegetables, the product of which may be categorized as ADD.

Statistical analysis

Statistical computations like Duncan’s multiple range test, simple descriptive statistics, prediction modeling of As content as well as risk assessment of arsenic through diet, the associated hazard quotient as well as the target cancer risk were performed using *Microsoft Excel 2016* and Statistical Product and Service Solutions (*SPSS version 23.0 (SPSS, Inc.)*).

**Results and discussion**

Characteristics of experimental site including As content in the selected vegetables grown nearby

The average physico-chemical properties of initial soils (Table 1) showed neutral pH (6.94), low soluble salt content with electrical conductivity (EC) of 0.35 dS m<sup>-1</sup>, medium organic carbon content (0.55%), silty clay texture with 48.5% clay, low available nitrogen (258 kg ha<sup>-1</sup>) and available potassium (199 kg ha<sup>-1</sup>), while the available phosphorus content was high (33.5 kg ha<sup>-1</sup>). The total As content of the top soil (0–15 cm) was 19.4 mg kg<sup>-1</sup> with relatively high available As content (2.57 mg kg<sup>-1</sup>). The shallow tube well (STW) water had substantially high As (0.32 ± 0.11 mg l<sup>-1</sup>) compared to the pond water (PW) (0.03 ± 0.00 mg l<sup>-1</sup>). As concentrations in

**Table 1** Initial status of soil and water used in field experiment

Parameters	Value
<i>Soil characterization</i>	
pH	6.94
EC (dS m <sup>-1</sup> )	0.35
Clay content (%)	48.5
Organic carbon (%)	0.55
Available N (kg ha <sup>-1</sup> )	258
Available P (kg ha <sup>-1</sup> )	33.5
Available K (kg ha <sup>-1</sup> )	199
Available As (mg kg <sup>-1</sup> )	2.57
Total As (mg kg <sup>-1</sup> )	19.4
<i>Irrigation water</i>	
Pond water (mg L <sup>-1</sup> )	0.03 ± 0.00
Shallow tube well water (mg L <sup>-1</sup> )	0.32 ± 0.11

cauliflower head, spinach leaf and tomato fruit were 0.15–0.17, 2.73–3.0 and 0.08 mg kg<sup>-1</sup>, respectively.

Carbonell-Barrachina et al. (2009) reported gradual decrease in As concentration from plant roots (e.g., potatoes, carrots, radishes, turnips) followed by vegetable shoots (e.g., spinach and grasses) and finally to edible seeds or the fruits. High As presence in vegetables in As-affected areas of West Bengal, India, has been widely reported (Roychowdhury et al. 2002; Signes et al. 2008; Halder et al. 2013; Santra et al. 2013; Das et al. 2016).

#### As accumulation in experimental vegetables

As accumulation was significantly less when crops were irrigated either with pond water or with conjunctive use of 50% pond + 50% STW water as compared to sole STW water. As described in Tables 2 and 3, irrigation with pond water managed As accumulation in the ranges of 0.05–0.06 mg kg<sup>-1</sup> in cauliflower head, 1.46–1.54 mg kg<sup>-1</sup> in spinach leaf

and 0.05 mg kg<sup>-1</sup> in tomato fruit which are significantly lower than As accumulated through STW irrigation, i.e., 0.08–0.1 mg kg<sup>-1</sup>, 1.94–2.11 mg kg<sup>-1</sup> and 0.06–0.07 mg kg<sup>-1</sup>. Reduction in As accumulation in vegetables irrigated with pond water over the combination of STW and pond water is shown in Fig. 1. A substantial reduction in pond water is obtained, while even the use of STW and pond water in conjunction can ensure reduction of As in the crop edibles to some extent.

Mondal et al. (2015) reported low As content in pond water over STW which possibly results in low accumulation by the crops. Sinha and Bhattacharyya (2014) also suggested the use of low contaminated surface water as a possible alternative of irrigating the crop over STW water. However, the easy access to STW accompanied by the scarcity of water bodies in high-intensity arable land reduces the scope of surface water utilization in irrigation. During the winter season when large-scale STW water is used due to lack of rainfall, the surface water bodies are often

**Table 2** Effect of irrigation sources and organic intervention on arsenic (mg/kg) in selected vegetables and postharvest Olsen extractable soils (Rabi 2016–2017)

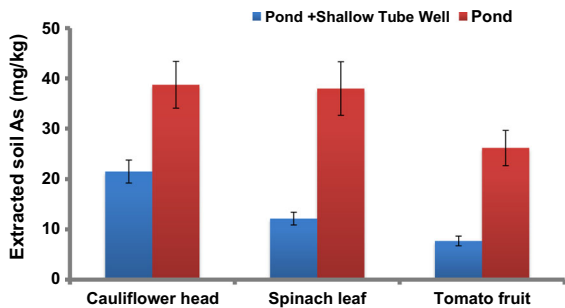
	Cauliflower		Spinach		Tomato	
	Head As	Soil As	Leaf As	Soil As	Fruit As	Soil As
<i>Source of irrigation</i>						
<i>I</i> <sub>1</sub>	0.08 <sup>a</sup>	2.74 <sup>a</sup>	2.11 <sup>a</sup>	3.01 <sup>a</sup>	0.07 <sup>a</sup>	1.96 <sup>a</sup>
<i>I</i> <sub>2</sub>	0.07 <sup>b</sup>	2.46 <sup>b</sup>	1.81 <sup>b</sup>	2.49 <sup>b</sup>	0.06 <sup>ab</sup>	1.51 <sup>b</sup>
<i>I</i> <sub>3</sub>	0.06 <sup>c</sup>	2.01 <sup>c</sup>	1.54 <sup>c</sup>	1.92 <sup>c</sup>	0.05 <sup>b</sup>	1.07 <sup>c</sup>
<i>Organic amendment</i>						
<i>F</i> <sub>1</sub>	0.11 <sup>a</sup>	2.54 <sup>a</sup>	2.30 <sup>a</sup>	2.81 <sup>ab</sup>	0.08 <sup>a</sup>	1.45 <sup>a</sup>
<i>F</i> <sub>2</sub>	0.02 <sup>d</sup>	1.94 <sup>b</sup>	1.31 <sup>d</sup>	2.02 <sup>b</sup>	0.05 <sup>c</sup>	1.20 <sup>bc</sup>
<i>F</i> <sub>3</sub>	0.09 <sup>b</sup>	2.65 <sup>a</sup>	2.01 <sup>b</sup>	2.88 <sup>ab</sup>	0.06 <sup>b</sup>	1.27 <sup>b</sup>
<i>F</i> <sub>4</sub>	0.05 <sup>c</sup>	2.51 <sup>a</sup>	1.50 <sup>c</sup>	3.12 <sup>a</sup>	0.05 <sup>d</sup>	1.15 <sup>c</sup>
<i>Interaction</i>						
<i>I</i> <sub>1</sub> <i>F</i> <sub>1</sub>	0.15 <sup>a</sup>	3.16 <sup>a</sup>	3.00 <sup>a</sup>	3.72 <sup>a</sup>	0.08 <sup>a</sup>	2.31 <sup>a</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>2</sub>	0.03 <sup>e</sup>	2.32 <sup>cd</sup>	1.41 <sup>e</sup>	2.16 <sup>f</sup>	0.05 <sup>c</sup>	1.44 <sup>d</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>3</sub>	0.10 <sup>bc</sup>	2.78 <sup>b</sup>	2.20 <sup>bc</sup>	2.34 <sup>e</sup>	0.08 <sup>a</sup>	1.48 <sup>cd</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>4</sub>	0.08 <sup>d</sup>	2.53 <sup>bc</sup>	1.65 <sup>d</sup>	2.51 <sup>d</sup>	0.06 <sup>b</sup>	1.51 <sup>c</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>1</sub>	0.12 <sup>b</sup>	2.16 <sup>d</sup>	2.32 <sup>b</sup>	2.96 <sup>c</sup>	0.08 <sup>a</sup>	1.81 <sup>b</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>2</sub>	0.03 <sup>ef</sup>	2.01 <sup>ef</sup>	1.24 <sup>f</sup>	2.13 <sup>f</sup>	0.04 <sup>c</sup>	1.21 <sup>f</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>3</sub>	0.09 <sup>c</sup>	1.98 <sup>f</sup>	2.13 <sup>bc</sup>	2.41 <sup>d</sup>	0.07 <sup>b</sup>	1.50 <sup>cd</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>4</sub>	0.06 <sup>de</sup>	2.32 <sup>cd</sup>	1.83 <sup>c</sup>	2.26 <sup>e</sup>	0.06 <sup>bc</sup>	1.24 <sup>f</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>1</sub>	0.11 <sup>bc</sup>	2.51 <sup>c</sup>	1.93 <sup>c</sup>	3.00 <sup>b</sup>	0.06 <sup>bc</sup>	1.81 <sup>b</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>2</sub>	0.01 <sup>f</sup>	1.96 <sup>f</sup>	1.22 <sup>f</sup>	2.06 <sup>g</sup>	0.04 <sup>d</sup>	0.91 <sup>g</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>3</sub>	0.08 <sup>d</sup>	2.04 <sup>e</sup>	1.65 <sup>d</sup>	3.01 <sup>b</sup>	0.06 <sup>bc</sup>	1.52 <sup>c</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>4</sub>	0.03 <sup>e</sup>	2.03 <sup>e</sup>	1.43 <sup>e</sup>	2.81 <sup>c</sup>	0.04 <sup>d</sup>	1.33 <sup>e</sup>

*I*<sub>1</sub> = Shallow tube well; *I*<sub>2</sub> = Pond + STW; *I*<sub>3</sub> = Pond; and *F*<sub>1</sub> = Control; *F*<sub>2</sub> = Vermicompost; *F*<sub>3</sub> = FYM; *F*<sub>4</sub> = Mustard Cake. Means followed by a different letter are significantly different (otherwise statistically at par) at *P* < 0.05 by Duncan's multiple range test

**Table 3** Effect of irrigation sources and organic intervention on arsenic (mg/kg) in selected vegetables and postharvest Olsen extractable soils (Rabi 2017–2018)

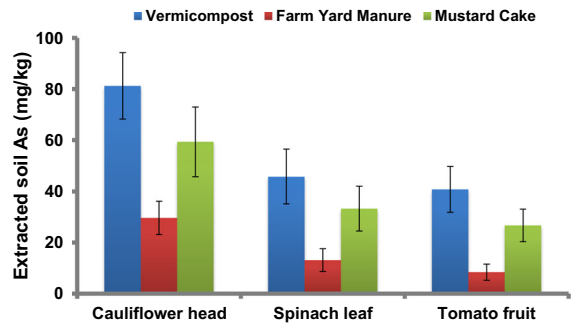
	Cauliflower		Spinach		Tomato	
	Head As	Soil As	Leaf As	Soil As	Fruit As	Soil As
<i>Source of irrigation</i>						
<i>I</i> <sub>1</sub>	0.10 <sup>a</sup>	2.96 <sup>a</sup>	1.94 <sup>a</sup>	2.97 <sup>a</sup>	0.06 <sup>a</sup>	1.92 <sup>a</sup>
<i>I</i> <sub>2</sub>	0.08 <sup>b</sup>	2.58 <sup>b</sup>	1.75 <sup>b</sup>	2.31 <sup>b</sup>	0.06 <sup>a</sup>	1.55 <sup>b</sup>
<i>I</i> <sub>3</sub>	0.06 <sup>c</sup>	1.81 <sup>c</sup>	1.461 <sup>c</sup>	1.79 <sup>c</sup>	0.05 <sup>b</sup>	1.11 <sup>c</sup>
<i>Organic amendment</i>						
<i>F</i> <sub>1</sub>	0.14 <sup>a</sup>	2.82 <sup>a</sup>	2.30 <sup>a</sup>	2.63 <sup>ab</sup>	0.07 <sup>a</sup>	1.40 <sup>a</sup>
<i>F</i> <sub>2</sub>	0.03 <sup>d</sup>	2.04 <sup>c</sup>	1.18 <sup>d</sup>	2.03 <sup>b</sup>	0.04 <sup>c</sup>	1.18 <sup>d</sup>
<i>F</i> <sub>3</sub>	0.09 <sup>b</sup>	2.59 <sup>b</sup>	1.98 <sup>b</sup>	2.68 <sup>ab</sup>	0.07 <sup>a</sup>	1.21 <sup>b</sup>
<i>F</i> <sub>4</sub>	0.06 <sup>c</sup>	2.47 <sup>b</sup>	1.57 <sup>c</sup>	2.81 <sup>a</sup>	0.05 <sup>b</sup>	1.13 <sup>c</sup>
<i>Interaction</i>						
<i>I</i> <sub>1</sub> <i>F</i> <sub>1</sub>	0.17 <sup>a</sup>	3.10 <sup>a</sup>	2.73 <sup>a</sup>	3.59 <sup>a</sup>	0.08 <sup>a</sup>	2.29 <sup>a</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>2</sub>	0.04 <sup>e</sup>	2.22 <sup>d</sup>	1.41 <sup>e</sup>	2.17 <sup>f</sup>	0.05 <sup>c</sup>	1.44 <sup>d</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>3</sub>	0.11 <sup>b</sup>	2.72 <sup>b</sup>	2.24 <sup>b</sup>	2.27 <sup>e</sup>	0.06 <sup>b</sup>	1.44 <sup>d</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>4</sub>	0.06 <sup>d</sup>	2.51 <sup>c</sup>	1.57 <sup>de</sup>	2.58 <sup>cd</sup>	0.06 <sup>c</sup>	1.45 <sup>cd</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>1</sub>	0.11 <sup>b</sup>	2.08 <sup>e</sup>	2.11 <sup>b</sup>	2.64 <sup>c</sup>	0.08 <sup>a</sup>	1.75 <sup>b</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>2</sub>	0.01 <sup>f</sup>	2.07 <sup>e</sup>	1.10 <sup>g</sup>	2.15 <sup>f</sup>	0.04 <sup>d</sup>	1.06 <sup>g</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>3</sub>	0.10 <sup>c</sup>	2.06 <sup>e</sup>	1.99 <sup>c</sup>	2.39 <sup>d</sup>	0.06 <sup>b</sup>	1.39 <sup>e</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>4</sub>	0.05 <sup>de</sup>	2.30 <sup>d</sup>	1.52 <sup>de</sup>	2.25 <sup>e</sup>	0.06 <sup>c</sup>	1.25 <sup>f</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>1</sub>	0.11 <sup>b</sup>	2.47 <sup>c</sup>	1.71 <sup>d</sup>	2.88 <sup>b</sup>	0.06 <sup>c</sup>	1.65 <sup>bc</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>2</sub>	0.01 <sup>g</sup>	1.88 <sup>g</sup>	1.09 <sup>h</sup>	1.99 <sup>g</sup>	0.03 <sup>e</sup>	0.90 <sup>h</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>3</sub>	0.07 <sup>d</sup>	1.98 <sup>f</sup>	1.78 <sup>d</sup>	2.95 <sup>b</sup>	0.06 <sup>c</sup>	1.52 <sup>c</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>4</sub>	0.04 <sup>e</sup>	1.99 <sup>f</sup>	1.21 <sup>f</sup>	2.61 <sup>c</sup>	0.03 <sup>e</sup>	1.31 <sup>e</sup>

*I*<sub>1</sub> = Shallow tube well; *I*<sub>2</sub> = Pond + STW; *I*<sub>3</sub> = Pond; and *F*<sub>1</sub> = Control; *F*<sub>2</sub> = Vermicompost; *F*<sub>3</sub> = FYM; *F*<sub>4</sub> = Mustard Cake. Means followed by a different letter are significantly different (otherwise statistically at par) at *P* < 0.05 by Duncan’s multiple range test



**Fig. 1** Percent reduction in arsenic recoveries through pond water over STW irrigation (pooled data)

contaminated by seepage and lateral movement of As-contaminated STW water. However, the precipitation of As in the bottom of the water bodies is a silver lining as the upper surface water becomes decontaminated and fit for irrigating the crops. Even the combined application of pond and STW water can reduce the As uptake by crops and human health risks thereof.



**Fig. 2** Percent reduction in arsenic recoveries through organic amendments over no-manure situations (pooled data)

As shown in Tables 2 and 3 and Fig. 2, the reduction of As accumulation in all vegetables was noticed under organic amendment application. The order of reduction from maximum to minimum was vermicompost followed by mustard cake followed by FYM treatments. Organic manure is known for its vital roles including immobilization, adsorption, chelation and co-precipitation of As, thereby regulating mobility

and subsequent entry into plants and dietary system (Sinha and Bhattacharyya 2011). In particular, humic and fulvic acid fractions were reported to act as scavengers of As (Ghosh et al. 2012). Higher the stability of the As and HA/FA complex, lesser will be the availability of As (Sinha and Bhattacharyya 2011).

The interaction of irrigation source and organic amendment produced significant changes in As accumulation in vegetables and postharvest soils over the control (Tables 2 and 3). The least accumulation of As in vegetables and postharvest soils was seen in plot facilitated by vermicompost amendment and irrigated with pond water. Similar findings were also reported by Das et al. (2016) for some other vegetable crops where vermicompost application and irrigating the crop with pond water made such reductions in As accumulation over other organic materials and STW irrigation.

Interestingly, conjunctive use of 50% pond + 50% STW water for irrigation together with vermicompost resulted in 85% reduction in vegetable As content over control treatment. Given the scarcity of pond water in the winter season, a compromise of 50:50 pond water: STW can be a viable alternative to ensure reduced arsenic load in vegetable-based diet.

#### Assessing the selected vegetables intake-based dietary As risk to human

The growing threat of entry of As via foods grown in highly As-contaminated regions, to other noncontaminated regions has indeed necessitated a thorough investigation of As buildup in the agricultural produce and its associated dietary risks.

Rural West Bengal in India and Bangladesh communities consume a distinct yet similar food (Roychowdhury et al. 2002; Alam et al. 2003; Signes-Pastor et al. 2008) including rice as a staple food with vegetables about thrice a day. Other grains, fruits and animal proteins (eggs, fish, chicken, mutton, etc.) are generally taken in negligible amounts (Halder et al. 2013).

The possible entry of As via the food chain in the greater Bengal basin has already been reported for rice as well as vegetables by Santra et al. (2013), noting that higher water content in vegetable grown in highly contaminated area was found to lower the risk of As exposure to human (Das et al. 2016).

#### *Provisional tolerable weekly intake (PTWI) of As due to consumption of selected vegetables*

Dietary risk of As from vegetables was computed by %PTWI (Table 4 and supplementary Table S1). The estimated %PTWI for the selected vegetables was found to vary and ranged from 10.0 to 0.75, 122 to 49.4 and 3.03 to 1.36 for cauliflower, spinach and tomato, respectively. Elevated levels of As in leafy vegetables and associated dietary risk have been reported in earlier studies (Roychowdhury et al. 2002; Williams et al. 2006; Halder et al. 2013) in affected areas of West Bengal, India, and Bangladesh.

Analyzing the results of %PTWI, it was found that dietary risk of As through consumption of cauliflower and tomato was far below the tolerance limit ( $\cong 100$ ), while for spinach it crossed the limit. The high water content and succulency increased the uptake of As in spinach. The significance of the study for cauliflower and tomato lies with the consumption of cooked food materials using As-contaminated water of the locality. The combined diet may have a PTWI in excess of 100% and thus become lethal.

The current study involving the use of surface water for irrigation and use of organic amendment reduced As level in edible portions of the vegetables, and subsequently, the composite diet of cooked food materials became safer. Combined effect of irrigating the crop using pond water and application of vermicompost reduced the %PTWI to 0.75, 49.4 and 1.36 for cauliflower, spinach and tomato. Even using pond and STW water in 50:50 proportion curtailed risk to As exposure significantly.

#### *Target cancer risk (TCR) due to As consumption via selected vegetables*

The TCR for As through consumption of the selected vegetables is given in Table 5 and in details in supplementary Table S2. The estimated TCR for the selected vegetables varied considerably based on their As loads. The TCR for cauliflower, spinach and tomato ranged from ( $2.84 \times 10^{-4}$  to  $2.12 \times 10^{-5}$ ), ( $3.48 \times 10^{-3}$  to  $1.40 \times 10^{-3}$ ) and ( $8.6 \times 10^{-5}$  to  $3.86 \times 10^{-5}$ ), respectively. The TCR computation generally includes oral cancer slope for inorganic arsenic. Our evaluation based on total As in this study may yield lower estimate (Antoine et al. 2017). The



**Table 4** Dietary risk (PTWI) of arsenic through the pooled data of contaminated vegetables with most efficient treatment combinations

Treatment	As concentration (mg/kg)	Average weekly consumption (kg)	Arsenic ingestion in a week (mg)	PTWI (%)
<i>Cauliflower head</i>				
$I_1F_1$	0.16	0.56	0.09	10.0
$I_2F_2$	0.02	0.56	0.01	1.49
$I_3F_2$	0.01	0.56	0.01	0.75
<i>Spinach leaf</i>				
$I_1F_1$	2.87	0.39	1.10	122
$I_2F_2$	1.17	0.39	0.45	50.1
$I_3F_2$	1.16	0.39	0.45	49.4
<i>Tomato fruit</i>				
$I_1F_1$	0.08	0.35	0.03	3.03
$I_2F_2$	0.04	0.35	0.01	1.56
$I_3F_2$	0.04	0.35	0.01	1.36

$I_1$  = Shallow Tube Well;  $I_2$  = Pond + STW;  $I_3$  = Pond; and  $F_1$  = Control;  $F_2$  = Vermicompost;  $F_3$  = FYM;  $F_4$  = Mustard Cake  
 $I_1F_1$  corresponds to control or least effective treatment.  $I_3F_2$  is the most effective, while conjunctive use of pond and STW has been compared through  $I_2F_2$

**Table 5** Target cancer risk (TCR) of arsenic through the pooled data of contaminated vegetables with most efficient treatment combinations

Treatment	Efr (days)	Ed (years)	Fir (g/day)	C (mg/kg)	CPSo (mg/kg day <sup>-1</sup> )	BWa (kg)	ATc (days)	TCR
<i>Cauliflower head</i>								
$I_1F_1$	365	70	80	0.16	1.5	68	25,550	$2.84 \times 10^{-4}$
$I_2F_2$	365	70	80	0.02	1.5	68	25,550	$4.24 \times 10^{-5}$
$I_3F_2$	365	70	80	0.01	1.5	68	25,550	$2.12 \times 10^{-5}$
<i>Spinach leaf</i>								
$I_1F_1$	365	70	55	2.87	1.5	68	25,550	$3.48 \times 10^{-3}$
$I_2F_2$	365	70	55	1.17	1.5	68	25,550	$1.42 \times 10^{-3}$
$I_3F_2$	365	70	55	1.16	1.5	68	25,550	$1.40 \times 10^{-3}$
<i>Tomato fruit</i>								
$I_1F_1$	365	70	50	0.08	1.5	68	25,550	$8.60 \times 10^{-5}$
$I_2F_2$	365	70	50	0.04	1.5	68	25,550	$4.41 \times 10^{-5}$
$I_3F_2$	365	70	50	0.04	1.5	68	25,550	$3.86 \times 10^{-5}$

$I_1$  = Shallow Tube Well;  $I_2$  = Pond + STW;  $I_3$  = Pond; and  $F_1$  = Control;  $F_2$  = Vermicompost;  $F_3$  = FYM;  $F_4$  = Mustard Cake  
 $I_1F_1$  corresponds to control or least effective treatment.  $I_3F_2$  is the most effective, while conjunctive use of pond and STW has been compared through  $I_2F_2$

level of  $10^{-4}$  has been accepted as threshold level for cancer development (Shaheen et al. 2016).

Analyzing TCR values for all three vegetables, tomato was found safe regarding exposure to the

carcinogen. Pond water irrigation and vermicompost amendment together resulted in reduction of TCR for tomato to  $3.86 \times 10^{-5}$  and therefore had low risk. In contrast, consumption of cauliflower head poses a

TCR threat of  $2.84 \times 10^{-4}$ , which is unsafe. Replacing STW water with pond water for irrigation together with vermicompost application reduced the TCR for cauliflower to the safe limit ( $2.12 \times 10^{-5}$ ).

However, great concern existed in case of spinach leaf. It accumulated a huge load of As resulting in a high TCR of  $3.48 \times 10^{-3}$ . Both pond water irrigation and organic amendment failed to bring down the TCR within safe limit ( $10^{-4}$  to  $10^{-6}$ ). A similar report by Bhatti et al. (2013) revealed As movement to plant shoots might possibly be the reason of higher levels of TCR in spinach leaves. It is a serious issue and warrants a further thorough investigation.

#### *Hazard quotient (HQ) due to As consumption via selected vegetables*

The estimate of HQ is presented in Table 6 and in details in supplementary Table S3 with values for cauliflower and tomato in the range of 0.09 to 0.01 and 0.03 to 0.01, respectively, which is well below the safe limit ( $\cong 1.0$ ).

Analyzing the HQ for the consumption of spinach, a maximum value of 1.104 was reported, much higher than the safe limit of HQ for As. Golui et al. (2014) have put forward an important consideration that As

may enter the human body not only from vegetable-based diet but also intake of water and inhalation of dust. So consideration of safe limit of HQ for As as 0.5 seems to be more accurate. In the current study, use of pond water for irrigation and application of vermicompost was found to reduce the HQ values to 0.01, 0.01 and 0.45 for consumption of cauliflower, tomato and spinach, respectively, which is within the safe limit of HQ ( $< 0.5$ ). Similar to TCR, as we have computed HQ based on total As data, an overestimation cannot be denied as against inorganic As, similar to the findings of Antoine et al. (2017).

#### *Prediction of As content and HQ in selected vegetables by solubility-free ion activity model (FIAM)*

In some As endemic areas cultivation of crop may be seemingly difficult due to various reasons; however, the knowledge of the extent of the uptake of As under measured soil pH and organic C along with extractable soil As content can give a clear insight into the dietary risk of exposure upon consumption of the said crops. The fundamental of this model is that any treatment which results in a negative association between soil pH, organic carbon and extractable soil

**Table 6** Dietary risk (HQ) of arsenic through the pooled data of contaminated vegetables with most efficient treatment combinations

Treatment	As concentration (mg/kg)	Average daily intake (kg/day)	Reference dose (RfD) (mg/kg bodyweight/day)	Average body weight (kg)	HQ
<i>Cauliflower head</i>					
$I_1F_1$	0.16	0.08	0.002	68	0.09
$I_2F_2$	0.02	0.08	0.002	68	0.01
$I_3F_2$	0.01	0.08	0.002	68	0.01
<i>Spinach leaf</i>					
$I_1F_1$	2.87	0.06	0.002	68	1.10
$I_2F_2$	1.17	0.06	0.002	68	0.45
$I_3F_2$	1.16	0.06	0.002	68	0.45
<i>Tomato fruit</i>					
$I_1F_1$	0.08	0.05	0.002	68	0.03
$I_2F_2$	0.04	0.05	0.002	68	0.01
$I_3F_2$	0.04	0.05	0.002	68	0.01

$I_1$  = Shallow Tube Well;  $I_2$  = Pond + STW;  $I_3$  = Pond; and  $F_1$  = Control;  $F_2$  = Vermicompost;  $F_3$  = FYM;  $F_4$  = Mustard Cake  
 $I_1F_1$  corresponds to control or least effective treatment.  $I_3F_2$  is the most effective, while conjunctive use of pond and STW has been compared through  $I_2F_2$

As, can be efficient in mitigation of the As problem. In the current experiment, the combined use of pond water for irrigation and use of organic amendments resulted in an increase in the soil pH and organic carbon content in all the cultivated soils of the vegetables (Table 7) and thus emerge as a viable option for As mitigation.

Employing the solubility-FIAM model for predicting the uptake of As in the selected vegetables, the crop and metalloid-specific constants were determined (Table 8). The solubility FIAM-based values of such constants as C,  $\beta_1$  and  $\beta_2$  for cauliflower were - 6.7, 1.03 and 2.96, respectively, with 59% predictability. In case of spinach, model parameters were - 9.18 for C, 0.96 for  $\beta_1$  and 0.92 for  $\beta_2$  with 69% variability explained. In tomato, the model parameters are -6.35 for C, 0.73 for  $\beta_1$  and 0.75 for  $\beta_2$  with 72% predictability. The positive values of the model parameters  $\beta_1$  and  $\beta_2$  signify the negative association between As uptake and pH and organic carbon. Therefore, a considerable relationship between the

**Table 8** Prediction of arsenic content in cauliflower head, spinach leaf and tomato fruit by solubility-free ion activity model

Crop	Model parameters			
	C	$\beta_1$	$\beta_2$	R <sup>2</sup>
Cauliflower	- 6.70	1.03	2.96	0.59 <sup>a</sup>
Spinach	- 9.18	0.96	0.92	0.69 <sup>a</sup>
Tomato	- 6.35	0.73	0.75	0.72 <sup>a</sup>

<sup>a</sup>Values of R<sup>2</sup> > 0.20 are significant at 5% probability level

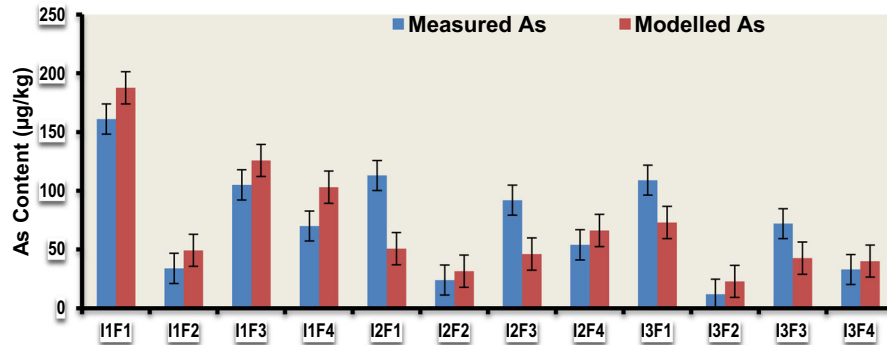
measured As content and the solubility FIAM-mediated modeled As (as shown in Figs. 3, 4 and 5) was observed for the selected vegetables.

This finding conforms several other studies relating pH, organic carbon and As phytoavailability (Datta and Young 2005; Meena et al. 2016; Golui et al. 2014, 2017; Mandal et al. 2019b). As can easily form ligand exchange-mediated stable complexes with

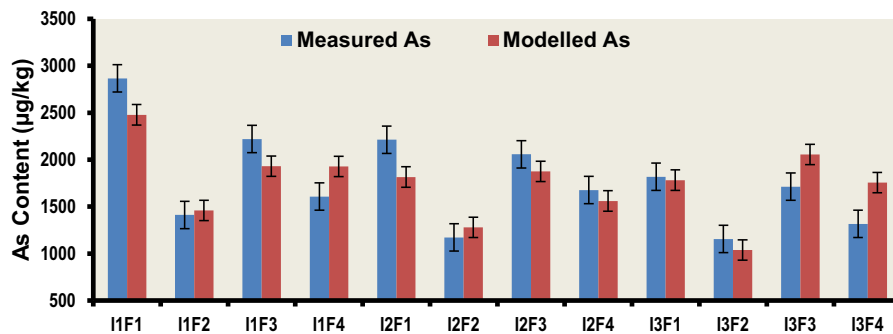
**Table 7** Effect of organic amendments and irrigation sources on pH and organic carbon of soil under cultivation of selected vegetables (pooled for two years)

	Cauliflower		Spinach		Tomato	
	pH	OC (%)	pH	OC (%)	pH	OC (%)
<i>Source of irrigation</i>						
<i>I</i> <sub>1</sub>	6.89 <sup>c</sup>	0.53 <sup>b</sup>	6.90 <sup>c</sup>	0.53 <sup>b</sup>	6.89 <sup>c</sup>	0.52 <sup>b</sup>
<i>I</i> <sub>2</sub>	6.94 <sup>b</sup>	0.53 <sup>a</sup>	6.95 <sup>b</sup>	0.53 <sup>b</sup>	6.94 <sup>a</sup>	0.52 <sup>a</sup>
<i>I</i> <sub>3</sub>	6.97 <sup>a</sup>	0.53 <sup>a</sup>	6.98 <sup>a</sup>	0.54 <sup>a</sup>	6.98 <sup>a</sup>	0.53 <sup>a</sup>
<i>Organic amendment</i>						
<i>F</i> <sub>1</sub>	6.97 <sup>a</sup>	0.50 <sup>d</sup>	6.98 <sup>a</sup>	0.51 <sup>d</sup>	6.97 <sup>a</sup>	0.50 <sup>d</sup>
<i>F</i> <sub>2</sub>	6.96 <sup>b</sup>	0.58 <sup>a</sup>	6.97 <sup>a</sup>	0.59 <sup>a</sup>	6.95 <sup>b</sup>	0.55 <sup>a</sup>
<i>F</i> <sub>3</sub>	6.88 <sup>d</sup>	0.53 <sup>b</sup>	6.89 <sup>c</sup>	0.53 <sup>b</sup>	6.88 <sup>d</sup>	0.53 <sup>b</sup>
<i>F</i> <sub>4</sub>	6.93 <sup>c</sup>	0.51 <sup>c</sup>	6.94 <sup>b</sup>	0.52 <sup>c</sup>	6.93 <sup>c</sup>	0.52 <sup>c</sup>
<i>Interaction</i>						
<i>I</i> <sub>1</sub> <i>F</i> <sub>1</sub>	6.93 <sup>e</sup>	0.49 <sup>f</sup>	6.95 <sup>e</sup>	0.51 <sup>e</sup>	6.92 <sup>e</sup>	0.49 <sup>e</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>2</sub>	6.92 <sup>f</sup>	0.57 <sup>b</sup>	6.92 <sup>f</sup>	0.57 <sup>b</sup>	6.91 <sup>e</sup>	0.54 <sup>b</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>3</sub>	6.84 <sup>h</sup>	0.53 <sup>c</sup>	6.85 <sup>i</sup>	0.53 <sup>c</sup>	6.83 <sup>g</sup>	0.52 <sup>c</sup>
<i>I</i> <sub>1</sub> <i>F</i> <sub>4</sub>	6.88 <sup>g</sup>	0.51 <sup>de</sup>	6.90 <sup>g</sup>	0.52 <sup>cd</sup>	6.89 <sup>f</sup>	0.51 <sup>d</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>1</sub>	6.97 <sup>c</sup>	0.50 <sup>e</sup>	6.98 <sup>c</sup>	0.51 <sup>e</sup>	6.98 <sup>b</sup>	0.49 <sup>e</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>2</sub>	6.96 <sup>d</sup>	0.57 <sup>b</sup>	6.97 <sup>cd</sup>	0.58 <sup>b</sup>	6.96 <sup>c</sup>	0.55 <sup>b</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>3</sub>	6.87 <sup>g</sup>	0.54 <sup>c</sup>	6.88 <sup>h</sup>	0.53 <sup>c</sup>	6.88 <sup>f</sup>	0.54 <sup>b</sup>
<i>I</i> <sub>2</sub> <i>F</i> <sub>4</sub>	6.94 <sup>e</sup>	0.51 <sup>de</sup>	6.95 <sup>e</sup>	0.52 <sup>de</sup>	6.94 <sup>d</sup>	0.52 <sup>c</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>1</sub>	7.01 <sup>a</sup>	0.51 <sup>de</sup>	7.00 <sup>b</sup>	0.52 <sup>de</sup>	7.01 <sup>a</sup>	0.51 <sup>cd</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>2</sub>	6.99 <sup>b</sup>	0.59 <sup>a</sup>	7.02 <sup>a</sup>	0.61 <sup>a</sup>	6.99 <sup>b</sup>	0.57 <sup>a</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>3</sub>	6.92 <sup>f</sup>	0.53 <sup>c</sup>	6.93 <sup>f</sup>	0.53 <sup>cd</sup>	6.94 <sup>d</sup>	0.52 <sup>cd</sup>
<i>I</i> <sub>3</sub> <i>F</i> <sub>4</sub>	6.97 <sup>c</sup>	0.52 <sup>d</sup>	6.97 <sup>d</sup>	0.52 <sup>c-e</sup>	6.96 <sup>c</sup>	0.52 <sup>cd</sup>

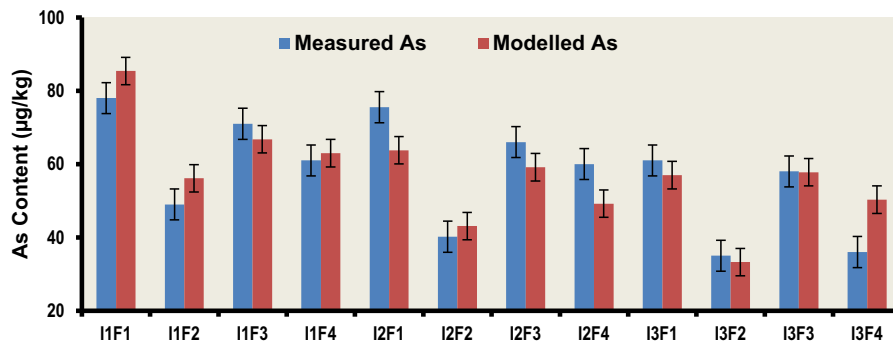
*I*<sub>1</sub> = Shallow Tube Well; *I*<sub>2</sub> = Pond + STW; *I*<sub>3</sub> = Pond; and *F*<sub>1</sub> = Control; *F*<sub>2</sub> = Vermicompost; *F*<sub>3</sub> = FYM; *F*<sub>4</sub> = Mustard Cake. Means followed by a different letter are significantly different (otherwise statistically at par) at *P* < 0.05 by Duncan's multiple range test



**Fig. 3** Comparison of observed and modeled\* As content of cauliflower head



**Fig. 4** Comparison of observed and modeled\* As content of spinach leaf



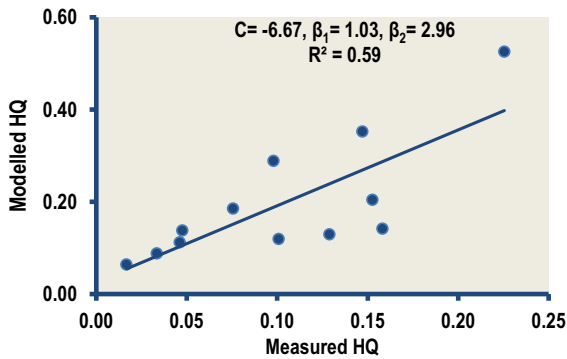
**Fig. 5** Comparison of observed and modeled\* As content of tomato fruit. {\*As content was predicted by solubility-free ion activity model based on pH and Mc (extractable metal) assumed to be adsorbed on Walkley and Black organic carbon; based on

pooled data} where I1 = Shallow Tube Well; I2 = Pond+STW; I3 = Pond; and F1 = Control; F2 = Vermicompost; F3 = FYM; F4 = Mustard Cake.

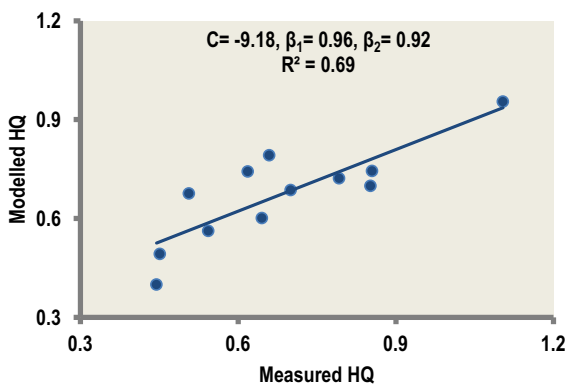
organic matter, thereby restricting its bioavailability (Paikaray et al. 2005).

Apart from this, the predicted hazard quotient has been compared with observed As-mediated HQ for selected vegetable on 1:1 line fit (Figs. 6, 7 and 8), showing reasonable line of fit values. Further development in this predictive ability and building of

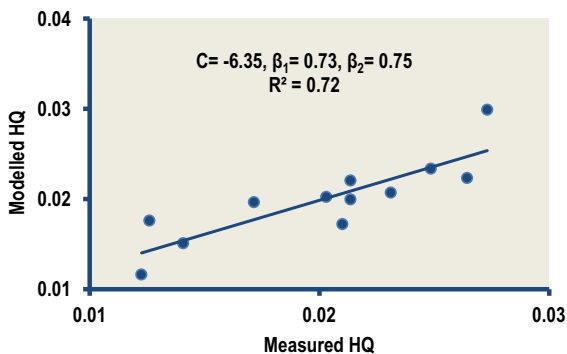
database for varied crops can be used to inform future decision making in selection of crops to be grown in the As-contaminated area in order to minimize As risk through human dietary exposure.



**Fig. 6** Comparison of hazard quotient (HQ) values based on actual (measured) and modeled\* As content in cauliflower head on 1:1 line



**Fig. 7** Comparison of hazard quotient (HQ) values based on actual (measured) and modeled\* As content in spinach leaf on 1:1 line



**Fig. 8** Comparison of hazard quotient (HQ) values based on actual (measured) and modeled\* As content in tomato fruit on 1:1 line. {\*As content in plant was predicted by solubility-free ion activity based on extractable metal, Walkley–Black organic carbon and pH; based on pooled data}

**Conclusion**

The problem of As contamination has been an emerging threat to the human community through contaminated food material consumption grown in the polluted belts. Rice and vegetable-based diet are the primary staple food for rural people; thus, mitigation of As in these materials deserves special significance. Analysis of the datasets regarding the soil–water-crop transfer of As through the predominantly cultivated and traded vegetables (cauliflower, spinach and tomato) and associated dietary risk parameters and modeling has very little documentation in earlier studies and provides novel insights into the situation. In the current study, use of surface water for irrigation and application of organic amendments like vermicompost emerged as the best suitable option for reducing As load in the vegetable edibles. The dietary risk parameters (% PTWI, HQ, TCR) were not alarming for cauliflower and tomato, but they really posed threat for spinach consumption. Vermicompost and irrigation through pond water alone or in conjunction to STW rendered many of the dietary risks to be benign. Modeling the As uptake in vegetables through solubility-FIAM can enable prediction of As load even in absence of crop cultivation. Reliability of the forecasting based on simple soil pH and organic carbon can ensure database generation and future selection of crop for augmenting sustainable agricultural productivity in the As-contaminated areas of West Bengal, India, through proper policy generation and enactment.

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**Compliance with ethical standards**

**Conflict of interest** The authors declare that they have no conflict of interest.

**Human and animals rights** This article does not contain any studies with human participants or animals performed by any of the authors.

**Informed consent** Informed consent was obtained from all individual participants included in the study.

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