

**Arsenic Accumulation Pattern in Common Vegetables  
from Successive Irrigation and  
Water - Soil - Plant Arsenic Relationship**

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***Supporting Information***

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## ***Part A: Chemical Analysis***

### ***Materials***

Chemicals used in this investigation were of high purity analytical grade. Ultrapure water, prepared by purifying distilled de-ionized water (DDW) with a Milli-Q A10 (Millipore Corporation, USA) system, was used throughout. Perkin Elmer pure atomic spectroscopy standard solutions (Perkin Elmer, USA) of arsenic (As) (CAS # 7440-38-2) was used for calibration. Standard solution of arsenic (III) ( $1000 \text{ mg L}^{-1}$ ) was prepared by dissolving 1.320 g of  $\text{As}_2\text{O}_3$  (Fluka, Switzerland) in 20.0 mL of ultrapure water containing 4.0 g of NaOH (Merck, Germany). The pH of the standard solution was adjusted to 5.5 with 2M HCl (Merck, Germany). Finally, the standard solution was diluted to 1000 mL using ultrapure water. It was stored in acid-washed glass bottle, kept refrigerated and used to prepare model water. The seeds of the vegetables were bought from regional office of BARC (Bangladesh Agricultural Research Council).

A number of standard solutions, reducing solutions, buffer solutions, ternary acid mixture and other solutions were prepared according to respective standard procedures (*I-4*). The other reagents were from Merck (Germany), Fluka (Switzerland) or Aldrich (USA). Two standard reference materials (SRM) were used to check the accuracy of the methods. Tomato Leaves (SRM 1573a) were from the U.S. National Institute of Standards and Technology (Gaithersburg, MD) and Pond Sediment (NIES 2) from the National Institute for Environmental Studies (Japan).

## ***Digestion***

Prior to elemental analysis, the water samples were subjected to mild digestion with HNO<sub>3</sub> (method 3030 E) (1) for total Fe and Mn estimation, and to vigorous digestion with HNO<sub>3</sub>-H<sub>2</sub>SO<sub>4</sub> (method 3030 G) (1) for total As and Cr. For estimations of Na, K, Mg and Ca, the water samples were not pre-treated.

Oxidation of the organic matter of plant tissue and release of arsenic was carried out through Wet Oxidation by means of HNO<sub>3</sub>-H<sub>2</sub>SO<sub>4</sub>-HClO<sub>4</sub> ternary acid mixture (2-6). Accurately weighed ( $1.0 \pm 0.005$  g) dried and powdered biotic samples were taken in a 500-mL Erlenmeyer flask. After adding 10 mL of 69% (v/v) HNO<sub>3</sub>, the flask was placed on a steam plate for 30 min. When cooled, 10 mL of the ternary acid mixture was added to it and the flask was placed on a hot plate at 180<sup>0</sup> C until nearly to dryness.

Digestion of soil samples was carried out by modified method of Small and McCants with H<sub>2</sub>SO<sub>4</sub>-HClO<sub>4</sub> acid mixture (2, 7). Accurately weighed  $0.5 \pm 0.005$  g of the soil sample was mixed with 2 mL of 98% (v/v) H<sub>2</sub>SO<sub>4</sub> and was heated in a 500-mL Erlenmeyer flask until fuming. When the organic matter had been destroyed and the soil assumed a gray color, the flask was cooled and 3 mL of 70% (v/v) HClO<sub>4</sub> was added. The mixture was boiled on a hot plate at 220<sup>0</sup> C for about 2 hours until the soil was near to dryness.

The digestion flask containing the residue of oxidized plant tissue or soil was cooled and 10 mL of 82% HCl was added. The digestate was collected in another 100-mL measuring flask. After rinsing, the digestion flask with small amounts of 6M HCl followed by ultrapure water twice, the collected digestate was diluted to make a final volume of 100 mL.

## ***Chemical Analysis***

### *Hydride Generation – Atomic Absorption Spectroscopy (HG-AAS):*

Total arsenic concentration ( $As_T$ ) of water samples as well as plant tissues and soil digestates were analyzed by HG-AAS, using an AAnalyst 200 (Perkin Elmer; Shelton, CT, U.S.A.) atomic absorption spectrometer. The system is equipped with deuterium arc background corrector and a computer with Microsoft Windows CE compatible “WinLab32™” software. The instrument is combined with a Mercury / Hydride System MHS-15 (Perkin-Elmer, U.S.A.) that comprises a reactor assembly and a quartz tube atomizer (QTA) assembly, and electrodeless discharge lamp (EDL System 2) as line source. The instrumental conditions used for As were as follows: EDL current 400 mA; wavelength 193.7 nm; spectral bandpass 0.7 nm; signal measurement peak height; flame air-acetylene; carrier gas argon, flow rate 0.7 L min<sup>-1</sup>; reducing agent 3% (w/v) NaBH<sub>4</sub> in 1% (w/v) NaOH solution, flow rate 1.5 mL min<sup>-1</sup>; measurement volume 10 mL; pre- and post-reaction purge time 50 and 40 s respectively; cell temperature 1000 °C. The calibration equation was set as linear, calculated intercept and replicates as 3. Standard concentrations for calibration were set as 5, 10, 20 and 40 µg L<sup>-1</sup>. Arsenic detection limit was 3 µg L<sup>-1</sup>.

### *Other analyses:*

Total Iron ( $Fe_T$ ), Calcium (Ca) and Magnesium (Mg) were estimated with FL (flame)-AAS technique. A 10-cm air-acetylene burner head and hollow cathode lamps were used for the purpose. The calibration curve correlation coefficient was checked to ensure that it was higher than 0.995. Potassium (K) and Sodium (Na) were measured with a Flame Photometer (CHEMITO 1020, Toshniwal Instrument Ltd., India) after proper calibration. Manganese (Mn) was estimated using USEPA approved Periodate Oxidation method and Chromium (Cr) using Alkaline Hypobromite Oxidation method by colorimetric techniques (DR/4000 U, HACH, Colorado, U.S.A.).

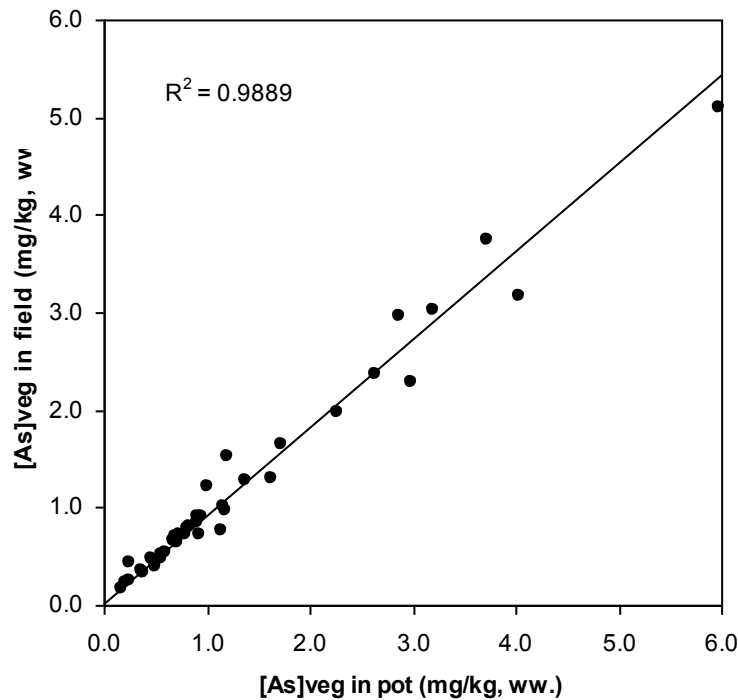
### ***Soil Characterization***

Soil pH was measured using a 1:1 soil to water ratio after 24 h equilibration (3). Soil organic matter was determined by Walkley-Black method (3) and the water content by loss of weight of dried (12 h, 105<sup>0</sup>C) samples. Particle size analyses were performed by sieve method (8). Total P was estimated according to the procedure of ISRIC (8). For reactive (extractable) Fe estimation, 25 mL of 0.2 M ammonium oxalate and 0.2 M oxalic acid buffer solution (pH 3.0) was added to 0.5 g soil and the mixture was shaken for 4 h (8). After centrifuging and diluting the clear supernatant, the aliquot was measured in AAS for reactive Fe.

## Part B: Quality Assurance

### Field Trails

Field trails were made with fertilizer amended soil (As concentration of  $9.835 \text{ mg kg}^{-1}$  on wet weight basis) for eight vegetables (Amaranth, Arum, Arum leaf, Carrot, Eggplant, Indian spinach, Okra and Radish) with natural arsenic contaminated groundwater having As concentration of 0.005, 0.044, 0.103, 0.507 and  $0.903 \text{ mg L}^{-1}$ . The detail procedure is mentioned in “Methods” section of main text. Least-square regression analysis of the data ( $n = 80$ , Table S12) yielded  $y = 0.9058x + 0.0107$  ( $r^2 = 0.9889$ ). Although field and pot results coincide at low or moderate vegetable As concentrations, at higher vegetable As concentrations pot results yielded slightly higher values than the corresponding field values ( $P = 0.0019$  by paired-samples t-test). The reason might be more deposition of arsenic in top soil in pot due to loss of moisture or less leaching of arsenic as compared to the field. This former effect is minimized by supplemented irrigation with control water in pot. In our investigation each harvesting corresponds to 10 irrigations, whereas farmers of Bangladesh generally use 6-7 irrigations for each harvesting in field.



**Figure S1.** Correlation of EPT arsenic levels in field and pot experiments.

*Part C: Tables and Figures*

**Table S1.** Analyses of shallow and semi-deep groundwater samples used in irrigation.

Sample ID <sup>†</sup>	Unit	RTW 01	RTW 02	RTW 03	RTW 04	RDP 05
Location (Upazilla)		Motiher	Motiher	Paba	Boalia	Boalia
Aquifer		Shallow	Shallow	Shallow	Shallow	Semi-deep
Well depth	m	40	43	25	30	65
Temperature	<sup>0</sup> C	26.3	27.1	26.8	25.9	27.6
pH		6.77	6.84	7.12	7.41	7.02
E <sub>h</sub>	mV	15.4	13.8	25.3	-14.2	5.1
DO	mg L <sup>-1</sup>	0.90	0.77	1.43	1.69	1.18
EC	μS cm <sup>-1</sup>	706	672	785	800	732
Fe	mg L <sup>-1</sup>	6.249	3.532	0.410	1.025	0.278
As <sub>T</sub>	mg L <sup>-1</sup>	0.903	0.507	0.103	0.044	0.005
Ca	mg L <sup>-1</sup>	85.360	79.915	67.001	90.472	49.838
Mg	mg L <sup>-1</sup>	41.112	30.312	54.295	28.413	30.092
Mn	mg L <sup>-1</sup>	0.253	0.210	1.048	0.442	0.123
Cr	mg L <sup>-1</sup>	0.02	< d. l.	< d. l.	0.01	< d. l.
Na	mg L <sup>-1</sup>	24.437	67.921	31.634	24.557	50.238
K	mg L <sup>-1</sup>	3.218	4.852	8.090	3.913	4.921

<sup>†</sup> RTW stands for Tube-Well of Rajshahi and RDP for Deep-Pump of Rajshahi.



**Table S2.** Arsenic concentration in vegetables irrigated with natural water.

Vegetables	[As] <sub>water</sub> <sup>a</sup> / mg L <sup>-1</sup>	[As] <sub>EPT</sub> <sup>b</sup> / mg kg <sup>-1</sup>		
		4 irrigation (40 days)	7 irrigation (60 days)	10 irrigation (80 days)
Okra	0.005 <sup>c</sup>	0.102 (0.618)	0.247 (1.492)	0.436 (2.701)
	0.044	0.238 (1.542)	0.502 (3.155)	0.741 (4.689)
	0.103	0.461 (2.764)	0.599 (3.780)	0.840 (5.279)
	0.507	0.316 (2.031)	0.766 (4.614)	0.981 (6.432)
	0.903	0.637 (3.802)	0.935 (6.139)	1.127 (8.015)
Radish	0.005 <sup>c</sup>	0.131 (1.109)	0.235 (2.114)	0.362 (3.238)
	0.044	0.257 (2.110)	0.412 (3.625)	0.760 (6.139)
	0.103	0.483 (3.852)	0.851 (7.298)	0.615 (5.074)
	0.507	0.705 (6.097)	0.998 (9.102)	1.100 (10.017)
	0.903	0.983 (9.008)	1.505 (13.564)	2.031 (18.071)
Aurum leaf	0.005 <sup>c</sup>	0.192 (2.114)	0.389 (4.206)	0.602 (6.603)
	0.044	0.310 (3.433)	0.851 (9.370)	0.711 (7.854)
	0.103	0.527 (5.812)	0.574 (6.095)	1.023 (10.435)
	0.507	0.885 (9.487)	1.240 (13.329)	1.505 (16.444)
	0.903	1.264 (13.301)	2.167 (21.288)	2.514 (24.937)

<sup>a</sup> 3.0 L of water of the mentioned conc. was applied to an earthen pot containing 15.0 ± 0.25 kg of soil having cross-section of base of 0.071 m<sup>2</sup>.

<sup>b</sup> EPT stands for edible plant tissue (i.e., vegetables).

<sup>c</sup> application of irrigation water having arsenic concentration of 0.005 mg L<sup>-1</sup> can be treated as control run.

<sup>d</sup> the concentrations within first bracket correspond to dry weight, while the remaining to fresh weight.

**Table S3.** Arsenic concentration in vegetables irrigated with model water.

Vegetables	[As] <sub>water</sub> <sup>a</sup> / mg L <sup>-1</sup>	[As] <sub>EPT</sub> <sup>b</sup> / mg kg <sup>-1</sup>		
		4 irrigation (40 days)	7 irrigation (60 days)	10 irrigation (80 days)
Amaranth (Lal shak)	0.005 <sup>c</sup>	0.206 (1.837)	0.420 (3.802)	0.411 (3.691)
	1.00	0.940 (8.555)	0.749 (6.513)	1.334 (10.187)
	5.00	0.975 (9.124)	1.245 (10.793)	1.529 (13.005)
	10.00	1.542 (13.361)	2.024 (16.180)	2.267 (18.112)
	25.00	2.548 (21.696)	3.439 (28.203)	4.080 (34.284)
Indian Spinach (Pui shak)	0.005 <sup>c</sup>	0.145 (1.753)	0.234 (2.801)	0.535 (6.362)
	1.00	0.447 (5.262)	0.618 (7.359)	0.761 (8.833)
	5.00	0.685 (8.117)	0.870 (10.548)	1.617 (18.231)
	10.00	1.160 (13.445)	1.291 (16.127)	2.419 (29.010)
	25.00	2.068 (25.988)	2.637 (32.260)	3.481 (41.974)
Potato (Alu)	0.005 <sup>c</sup>	0.153 (1.327)	0.284 (2.256)	0.395 (3.384)
	1.00	0.560 (4.602)	0.751 (6.443)	0.968 (8.375)
	5.00	0.832 (7.491)	1.099 (9.738)	1.513 (13.646)
	10.00	1.654 (14.006)	2.143 (16.801)	2.591 (20.162)
	25.00	2.550 (22.569)	3.267 (27.083)	3.824 (32.500)

<sup>a</sup> 3.0 L of water of the mentioned conc. was applied to an earthen pot containing 15.0 ± 0.25 kg of soil having cross-section of base of 0.071 m<sup>2</sup>.

<sup>b</sup> EPT stands for edible plant tissue (i.e., vegetables).

<sup>c</sup> application of irrigation water having arsenic concentration of 0.005 mg L<sup>-1</sup> can be treated as control run.

<sup>d</sup> the concentrations within first bracket correspond to dry weight, while the remaining to fresh weight.

**Table S4.** Concentration of arsenic (mg kg<sup>-1</sup>) in soil and vegetables for successive harvesting.

Soil / Vegetables	[As] <sub>water</sub> <sup>a</sup> / mg L <sup>-1</sup>	[As] <sub>soil</sub> <sup>b, d</sup> or [As] <sub>EPT</sub> <sup>c, d</sup> / mg kg <sup>-1</sup>			
		1 <sup>st</sup> harvesting	2 <sup>nd</sup> harvesting	3 <sup>rd</sup> harvesting	4 <sup>th</sup> harvesting
Soil	N/A <sup>e</sup>	6.802 (9.657)			
	0.005 <sup>f</sup>	9.935 (13.901)			
	0.044	11.487 (16.762)	13.087 (18.115)	16.538 (24.164)	36.794 (52.739)
	0.103	15.060 (20.567)	16.361 (21.447)	20.286 (30.507)	60.687 (86.341)
	0.507	28.189 (36.433)	71.942 (94.822)	88.120 (123.108)	125.465 (189.341)
	0.903	50.741 (67.049)	101.238 (131.675)	187.239 (263.723)	231.316 (346.889)
Okra (Dherosh)	0.005 <sup>f</sup>	0.141 (1.827)	0.178 (2.213)	0.235 (2.936)	0.343 (4.120)
	0.044	0.346 (4.485)	0.450 (5.827)	0.658 (9.809)	0.998 (13.733)
	0.103	0.392 (5.002)	0.553 (6.974)	0.836 (11.558)	1.925 (24.381)
	0.507	0.689 (7.891)	0.984 (15.760)	1.581 (28.013)	4.147 (54.932)
	0.903	1.017 (12.088)	1.280 (16.319)	2.012 (30.681)	7.035 (96.551)
	Amaranth (Lal shak)	0.005 <sup>f</sup>	0.273 (2.815)	0.315 (3.406)	0.442 (4.912)
0.044		0.657 (6.104)	0.859 (7.928)	1.463 (14.270)	2.904 (31.395)
0.103		0.800 (7.216)	0.957 (8.902)	1.832 (16.234)	4.371 (43.463)
0.507		0.914 (8.372)	1.289 (11.578)	3.108 (34.849)	8.193 (80.152)
0.903		1.127 (11.155)	1.713 (15.843)	4.524 (40.062)	14.108 (124.317)
Indian Spinach (Pui shak)		0.005 <sup>f</sup>	0.253 (1.854)	0.269 (2.008)	0.323 (2.471)
	0.044	0.547 (4.318)	0.601 (4.798)	0.731 (6.237)	0.948 (7.550)
	0.103	0.716 (5.179)	0.787 (6.103)	0.986 (7.394)	1.395 (9.607)
	0.507	0.836 (6.332)	0.992 (7.710)	1.512 (11.302)	2.436 (14.681)
	0.903	1.124 (8.129)	1.479 (10.431)	1.693 (16.730)	5.760 (34.925)
	Carrot (Gajor)	0.005 <sup>f</sup>	0.213 (1.925)	0.265 (2.364)	0.310 (2.835)
0.044		0.550 (5.022)	0.618 (5.685)	0.765 (6.877)	0.996 (8.671)
0.103		0.641 (6.128)	0.769 (7.048)	0.993 (8.518)	1.492 (11.802)
0.507		0.824 (8.019)	0.998 (8.793)	1.627 (12.425)	3.011 (23.821)
0.903		1.226 (11.473)	1.748 (15.085)	2.939 (25.810)	7.564 (56.322)

<sup>a</sup> natural water of the mentioned arsenic conc. was applied; in each irrigation 3.0 L of the water was applied to an earthen pot containing 15.0 ± 0.25 kg of soil having cross-section of base of 0.071 m<sup>2</sup>.

<sup>b</sup> surface soil of depth 0 - 3 cm is considered.

<sup>c</sup> EPT stands for edible plant tissue (i.e., vegetables).

<sup>d</sup> the concentrations within first bracket correspond to dry weight, while the remaining to fresh weight.

<sup>e</sup> the soil can be treated as blank where no irrigation water applied.

<sup>f</sup> application of irrigation water having arsenic concentration of 0.005 mg L<sup>-1</sup> can be treated as control run.

**Table S5.** Concentration of arsenic ( $\text{mg kg}^{-1}$ ) in different parts of vegetable plants irrigated with natural groundwaters.

Vegetable Plants	[As] <sub>water</sub> / $\text{mg L}^{-1}$	[As] <sub>plant tissue</sub> <sup>a</sup> / $\text{mg kg}^{-1}$		
		Root	Stem	Leaf (or Fruit)
Amaranth	0.005 <sup>b</sup>	3.088 (16.587)	0.873 (7.490)	0.501 (3.583)
	0.044	5.178 (23.459)	0.930 (9.187)	0.902 (4.876)
	0.103	8.597 (43.233)	3.075 (21.935)	0.561 (2.866)
	0.507	14.446 (90.526)	4.962 (49.340)	2.045 (13.109)
	0.903	23.248 (124.890)	7.176 (56.399)	4.484 (22.931)
Brinjal	0.005 <sup>b</sup>	1.879 (9.125)	0.980 (5.397)	0.384 (2.325)
	0.044	3.113 (15.263)	2.142 (9.947)	0.793 (3.641)
	0.103	3.426 (15.341)	3.657 (15.928)	1.060 (7.422)
	0.507	7.352 (28.087)	3.623 (20.556)	3.565 (17.730)
	0.903	15.219 (52.867)	5.045 (23.162)	4.046 (21.148)
Arum	0.005 <sup>b</sup>	1.429 (6.001)	0.991 (6.028)	0.964 (9.007)
	0.044	6.890 (28.247)	3.802 (16.599)	1.168 (11.364)
	0.103	9.239 (35.995)	5.006 (22.681)	1.747 (15.705)
	0.507	15.894 (63.561)	6.622 (31.306)	2.256 (21.942)
	0.903	27.028 (102.707)	11.559 (57.351)	3.330 (31.811)

<sup>a</sup> the concentrations within first bracket correspond to dry weight, while the other numbers to fresh weight.

<sup>b</sup> application of irrigation water having arsenic concentration of  $0.005 \text{ mg L}^{-1}$  can be treated as control run.

**Table S6.** Concentration of arsenic ( $\text{mg kg}^{-1}$ ) in different parts of vegetable plants irrigated with model water.

Vegetable Plants	[As] <sub>water</sub> / $\text{mg L}^{-1}$	[As] <sub>plant tissue</sub> <sup>a</sup> / $\text{mg kg}^{-1}$		
		Root	Stem	Leaf (or Fruit)
Indian Spinach	1.00	4.142 (37.076)	1.016 (10.519)	0.687 (8.222)
	5.00	4.557 (46.564)	2.154 (23.701)	1.480 (19.434)
	10.00	10.089 (85.153)	4.127 (46.082)	2.839 (36.915)
	25.00	28.864 (259.772)	7.447 (84.568)	6.108 (76.003)
	50.00	103.165 (920.285)	56.758 (613.343)	38.975 (465.117)
Okra	1.00	8.386 (32.508)	1.493 (8.022)	1.402 (7.705)
	5.00	13.977 (50.708)	4.592 (23.461)	2.851 (18.533)
	10.00	23.831 (81.023)	4.670 (27.103)	3.371 (21.923)
	25.00	70.138 (245.271)	13.896 (67.399)	10.014 (64.002)
	50.00	278.244 (912.047)	85.827 (429.111)	83.510 (542.819)
Radish	1.00	6.803 (54.082)	3.619 (24.704)	2.281 (21.763)
	5.00	9.720 (70.139)	3.852 (31.335)	1.762 (19.589)
	10.00	12.697 (101.597)	5.407 (45.852)	4.298 (50.477)
	25.00	44.415 (325.864)	12.605 (158.741)	3.513 (45.349)
	50.00	172.674 (1233.915)	82.991 (775.226)	10.175 (107.502)

<sup>a</sup> the concentrations within first bracket correspond to dry weight, while the other numbers to fresh weight.

**Table S7.** Average mass of different parts of the studied vegetable plants.

Vegetable Plants	Average mass <sup>a</sup> (g)		
	Root	Stem	Leaves / Fruit
Amaranth	4.73 (0.63)	19.73 (1.80)	29.54 (4.10)
Arum	11.13 (1.55)	28.73 (5.62)	44.66 (3.98)
Eggplant	17.66 (4.95)	81.90 (11.30)	48.98 (6.69)
Indian Spinach	11.40 (0.97)	58.36 (3.32)	58.26 (4.82)
Okra	65.98 (13.10)	115.53 (19.01)	29.81 (4.67)
Radish	3.01 (0.33)	52.33 (3.97)	97.76 (7.43)

<sup>a</sup> the concentrations within first bracket correspond to dry weight, while the other numbers to fresh weight.

**Table S8.** Water – Soil – Plant (edible plant tissue) profile.

[As] <sub>water</sub> / mg L <sup>-1</sup>	Total arsenic applied <sup>a</sup> (mg)	[As] <sub>soil</sub> <sup>b, f</sup> / mg L <sup>-1</sup>	[As] <sub>EPT</sub> <sup>f, i</sup> / mg kg <sup>-1</sup>							
			Amaranth <sup>c</sup>	Brinjal	Arum	Arum Leaf	Indian Spinach <sup>c</sup>	Okra	Radish	Radish Leaf
N/A	0.000	6.802 <sup>d</sup> (9.657)	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
0.005 <sup>g</sup>	0.150	9.935 <sup>e</sup> (13.901)	0.632 (5.351)	0.384 (2.325)	0.991 (6.028)	0.964 (9.007)				
0.044 <sup>g</sup>	1.320	11.487 (16.762)	0.912 (6.827)	0.793 (3.641)	3.802 (16.599)	1.168 (11.364)				
0.103 <sup>g</sup>	3.090	15.060 (20.567)	1.446 (11.495)	1.060 (7.422)	5.006 (22.681)	1.747 (15.705)				
0.507 <sup>g</sup>	15.210	23.189 (31.549)	3.072 (29.503)	3.565 (17.730)	6.622 (31.306)	2.256 (21.942)				
0.903 <sup>g</sup>	27.090	34.235 (46.092)	5.431 (38.075)	4.046 (21.148)	11.559 (57.351)	3.330 (31.811)				
1.00 <sup>h</sup>	30.00	23.282 (31.938)					0.856 (9.326)	1.402 (7.705)	3.619 (24.704)	2.281 (21.763)
5.00 <sup>h</sup>	150.00	42.755 (60.674)					1.826 (21.485)	2.851 (18.533)	3.852 (31.335)	1.762 (19.589)
10.00 <sup>h</sup>	300.00	80.516 (113.869)					3.500 (41.322)	3.371 (21.923)	5.407 (45.852)	4.298 (50.477)
25.00 <sup>h</sup>	750.00	491.834 (663.281)					6.795 (80.121)	10.014 (64.002)	12.605 (158.741)	3.513 (45.349)
50.00 <sup>h</sup>	1500.00	922.761 (1196.374)					48.094 (488.152)	83.510 (542.819)	82.991 (755.226)	10.175 (107.502)

<sup>a</sup> estimated from 10 times of irrigation, each time 3.0 L of water of the mentioned conc. was applied to a tub (earthen pot) containing 15.0 ± 0.25 kg of soil.

<sup>b</sup> surface soil of depth 0 - 3 cm is considered.

<sup>c</sup> the experimental ratios of stem and leaf for Amaranth and Indian Spinach are found to be 1: 1.21 and 1 : 1.08 respectively.

<sup>d</sup> the soil can be considered as blank soil.

<sup>e</sup> the soil can be treated as control soil.

<sup>f</sup> the concentrations within first bracket correspond to dry weight, while the remaining to fresh weight.

<sup>g</sup> irrigated with natural groundwater of the mentioned arsenic conc.

<sup>h</sup> irrigated with model water of the mentioned arsenic conc.

<sup>i</sup> EPT stands for edible plant tissue (i.e., vegetables).

**Table S9.** Water – Soil – Plant (root) profile.

[As] <sub>water</sub> / mg L <sup>-1</sup>	Total arsenic applied <sup>a</sup> (mg)	[As] <sub>soil</sub> <sup>b, f</sup> / mg L <sup>-1</sup>	[As] <sub>root</sub> <sup>e</sup> / mg kg <sup>-1</sup>					
			Amaranth	Brinjal	Aurum	Indian Spinach	Okra	Radish
N/A	0.000	6.802 <sup>c</sup> (9.657)	N/A	N/A	N/A	N/A	N/A	N/A
0.005 <sup>f</sup>	0.150	9.935 <sup>d</sup> (13.901)	3.088 (16.587)	1.879 (9.125)	1.429 (6.001)			
0.044 <sup>f</sup>	1.320	11.487 (16.762)	5.178 (23.459)	3.113 (15.263)	6.890 (28.247)			
0.103 <sup>f</sup>	3.090	15.060 (20.567)	8.597 (43.233)	3.426 (15.341)	9.239 (35.995)			
0.507 <sup>f</sup>	15.210	23.189 (31.549)	14.446 (90.526)	7.352 (28.087)	15.894 (63.561)			
0.903 <sup>f</sup>	27.090	34.235 (46.092)	23.248 (124.890)	15.219 (52.867)	27.028 (102.707)			
1.00 <sup>g</sup>	30.00	23.282 (31.938)				4.142 (37.076)	8.386 (32.508)	6.803 (54.082)
5.00 <sup>g</sup>	150.00	42.755 (60.674)				4.557 (46.564)	13.977 (50.708)	9.720 (70.139)
10.00 <sup>g</sup>	300.00	80.516 (113.869)				10.089 (85.153)	23.831 (81.023)	12.697 (101.597)
25.00 <sup>g</sup>	750.00	491.834 (663.281)				28.864 (259.772)	70.138 (245.271)	44.415 (325.864)
50.00 <sup>g</sup>	1500.00	922.761 (1196.374)				103.165 (920.285)	278.244 (912.047)	172.674 (1233.915)

<sup>a</sup> estimated from 10 times of irrigation, each time 3.0 L of water of the mentioned conc. was applied to a tub (earthen pot) containing 15.0 ± 0.25 kg of soil.

<sup>b</sup> surface soil of depth 0 - 3 cm is considered.

<sup>c</sup> the soil can be considered as blank soil.

<sup>d</sup> the soil can be treated as control soil.

<sup>e</sup> the concentrations within first bracket correspond to dry weight, while the remaining to fresh weight.

<sup>f</sup> irrigated with natural water of the mentioned arsenic conc.

<sup>g</sup> irrigated with model water of the mentioned arsenic conc.



**Table S10.** Statistical Analyses of concentrations of arsenic in vegetables irrigated with natural groundwaters (n represents no of observation and SE standard error of mean).

Vegetables	n	Concentration of arsenic in vegetables <sup>a</sup> (mg kg <sup>-1</sup> )								Dry wt. : Fresh wt.
		Control (Averaged)	Range	Geomet- ric Mean	Median	Arithmetic Mean ± SE	95% CI for mean	Standard Deviation	Variance	
Amaranth	11	0.439 (3.952)	0.657 - 5.431 (6.104-38.075)	0.985 (8.645)	0.912 (7.216)	1.425 ± 0.461 (11.873±3.414)	0.399 - 2.451 (4.266-19.480)	1.528 (11.324)	2.334 (128.224)	0.096
Arum	5	0.991 (6.028)	3.802 - 11.559 (16.599-57.351)	4.284 (20.990)	5.006 (22.681)	5.596 ± 1.752 (26.793±8.678)	0.732 - 10.460 (2.699-50.887)	3.917 (19.405)	15.346 (376.542)	0.135
Arum leaf	10	0.783 (7.805)	0.711 - 3.330 (7.854-31.811)	1.377 (13.792)	1.337 (13.535)	1.582 ± 0.279 (15.610±2.625)	0.951 - 2.213 (9.673-21.547)	0.882 (8.300)	0.777 (68.884)	0.067
Brinjal	5	0.384 (2.325)	0.793 - 4.046 (3.641-21.148)	1.360 (7.489)	1.060 (7.422)	1.970 ± 0.761 (10.453±3.801)	-0.143 - 4.082 (-0.10-21.007)	1.702 (8.500)	2.896 (72.248)	0.063
Carrot	5	0.213 (1.925)	0.550 - 1.226 (5.022-11.473)	0.597 (5.588)	0.641 (6.128)	0.691 ± 0.167 (6.513±1.586)	0.228 - 1.153 (2.111-10.916)	0.372 (3.546)	0.139 (12.573)	0.104
Indian Spinach	6	0.394 (4.108)	0.547 - 1.124 (4.318-8.128)	0.607 (4.884)	0.632 (5.756)	0.669 ± 0.122 (5.362±0.876)	0.356 - 0.981 (3.110-7.614)	0.298 (2.146)	0.089 (4.605)	0.070
Okra	10	0.289 (2.264)	0.741 - 1.127 (4.689-12.088)	0.576 (5.164)	0.715 (5.141)	0.671 ± 0.104 (5.841±0.934)	0.435-0.907 (3.729-7.953)	0.329 (2.953)	0.109 (8.718)	0.072
Radish	5	0.362 (3.238)	0.760 - 2.031 (6.139-18.070)	0.823 (7.117)	0.760 (6.139)	0.974 ± 0.290 (8.508±2.635)	0.168 - 1.779 (1.191-15.825)	0.649 (5.893)	0.421 (34.726)	0.122
All	57	0.477 (4.133)	0.547 - 11.559 (3.641-57.351)	0.996 (8.152)	0.912 (7.422)	1.550 ± 0.255 (11.204±1.376)	1.039 - 2.061 (8.447-13.960)	1.925 (10.389)	3.707 (107.924)	0.085

<sup>e</sup> the concentrations within first bracket correspond to dry weight, while the remaining to fresh weight.

**TABLE S11.** Comparison of arsenic concentrations ( $\text{mg kg}^{-1}$ , ww.) in fresh vegetables from field and pot experiments ( $n = 80$ ).

[As] <sub>water</sub> /ppm	0.005		0.044		0.103		0.507		0.903	
	Field	Pot	Field	Pot	Field	Pot	Field	Pot	Field	Pot
Vegetables										
Amaranth	0.472	0.558	0.681	0.712	0.906	0.943	1.003	1.172	1.964	2.280
Arum	0.841	0.906	2.280	2.985	3.160	4.047	5.096	5.973	10.160	10.906
Arum leaf	0.523	0.564	0.718	0.802	0.969	1.182	1.645	1.732	3.014	3.211
Carrot	0.248	0.250	0.426	0.243	0.718	0.729	0.793	0.842	1.272	1.385
Eggplant	0.476	0.460	0.690	0.697	0.775	1.142	2.950	2.871	3.758	3.730
Indian spinach	0.219	0.205	0.538	0.600	0.716	0.923	0.897	0.905	1.294	1.633
Okra	0.164	0.172	0.326	0.385	0.400	0.512	0.664	0.683	1.205	1.007
Radish	0.345	0.376	0.638	0.711	0.780	0.815	1.516	1.207	2.359	2.644

**Table S12.** Results of paired, non-parametric test (Wilcoxon Signed Ranks Test).

Data set comparison	P-value
EPT for Gr <sup>†</sup> 0.044 vs Control	< 0.05
EPT for Gr <sup>†</sup> 0.103 vs Control	< 0.05
EPT for Gr <sup>†</sup> 0.507 vs Control	< 0.05
EPT for Gr <sup>†</sup> 0.903 vs Control	< 0.05

<sup>†</sup> Gr stands for groundwater.

**Table S13.** Results of paired-sample t-test.

Data set comparison	P-value (2-tailed)
EPT for Gr <sup>†</sup> 0.044 vs Control	0.0015
EPT for Gr <sup>†</sup> 0.103 vs Control	0.0004
EPT for Gr <sup>†</sup> 0.507 vs Control	0.0001
EPT for Gr <sup>†</sup> 0.903 vs Control	0.0022

<sup>†</sup> Gr stands for groundwater.

**Table S14.** Ratio of arsenic concentration<sup>a</sup> of soil to plant organ.

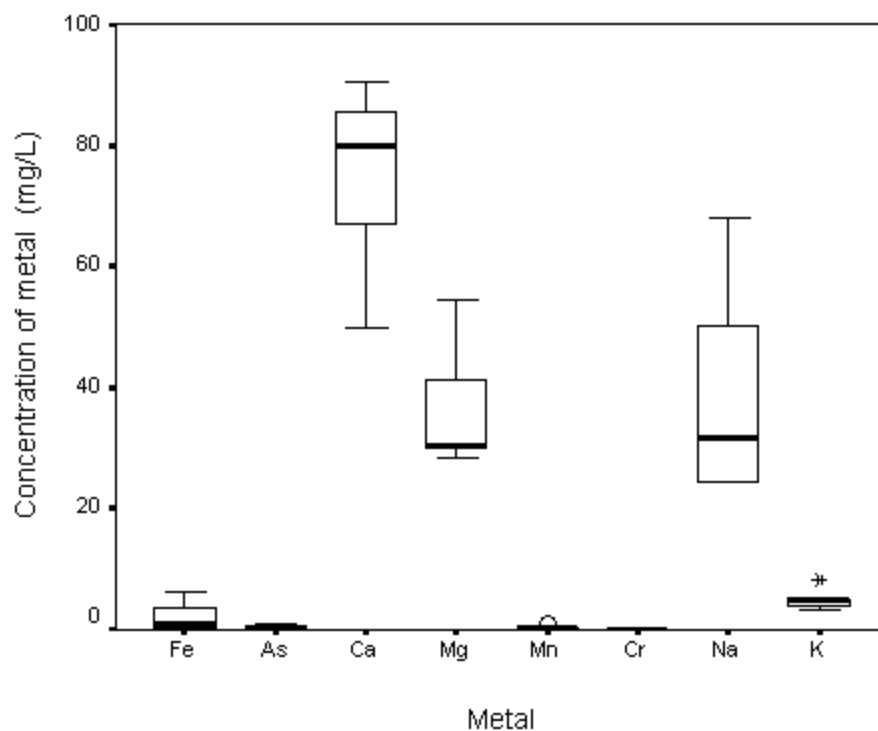
Vegetable Plant	Soil : EPT <sup>c</sup>	Soil : Root <sup>c</sup>	Soil : Stem <sup>d</sup>	Soil : Leaf <sup>d</sup>
Amaranth	10.516	2.053	4.396	9.549
Eggplant	13.906	3.755	8.041	17.467
Aurum	4.504 <sup>b</sup>	2.595	4.504 <sup>c</sup>	9.864 <sup>c</sup>
Indian Spinach	33.037	9.794	20.971	45.552
Okra	23.130	3.909	8.369	18.179
Radish	16.512 <sup>b</sup>	6.116	16.512 <sup>c</sup>	56.780

<sup>a</sup> arsenic concentrations are presented on fresh weight basis.

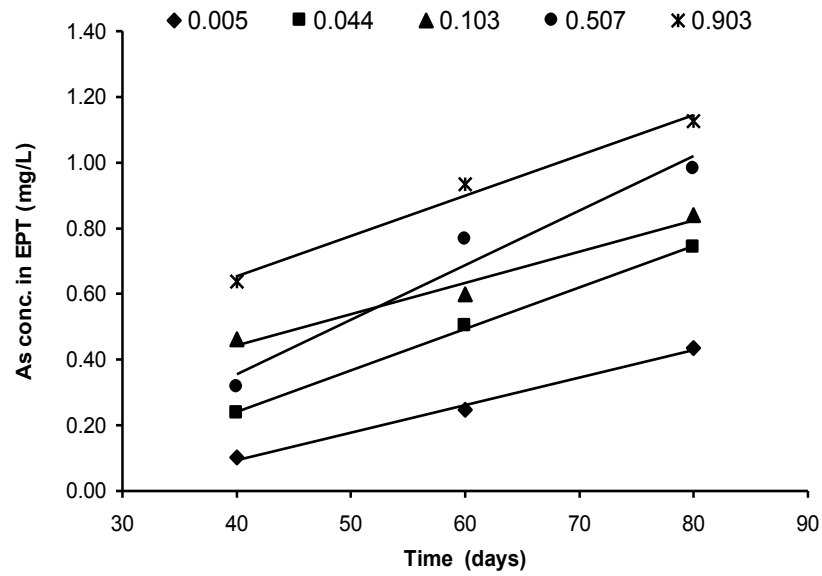
<sup>b</sup> stem was considered here as edible plant tissue.

<sup>c</sup> determined experimentally.

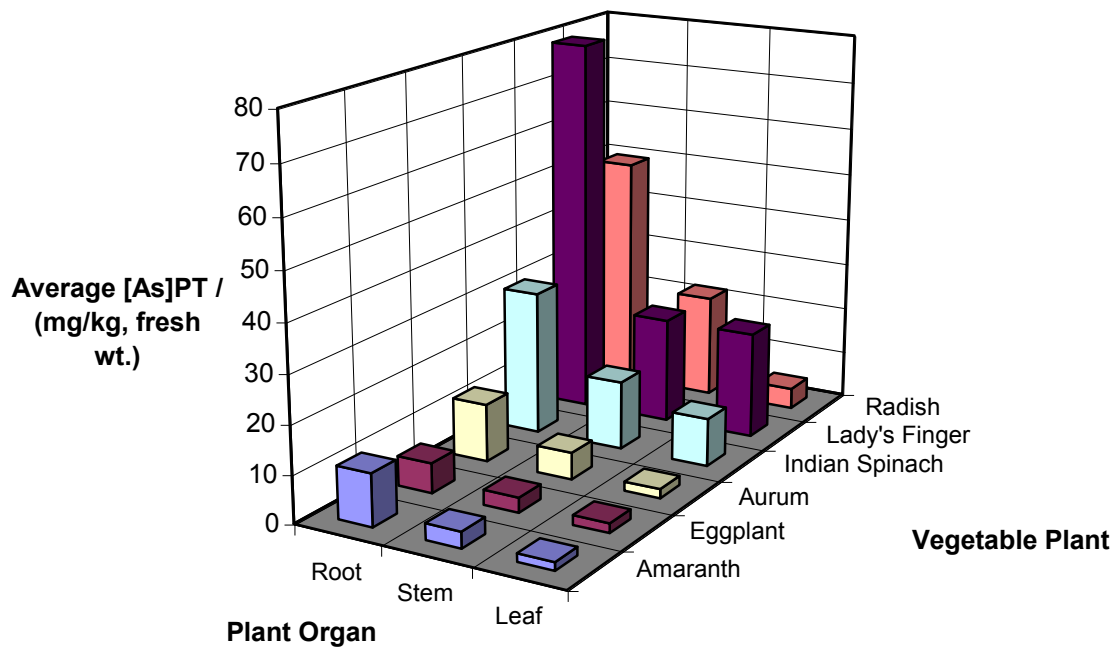
<sup>d</sup> estimated from the ratio of root:stem:leaf (or fruit) as 1:0.467:0.215.



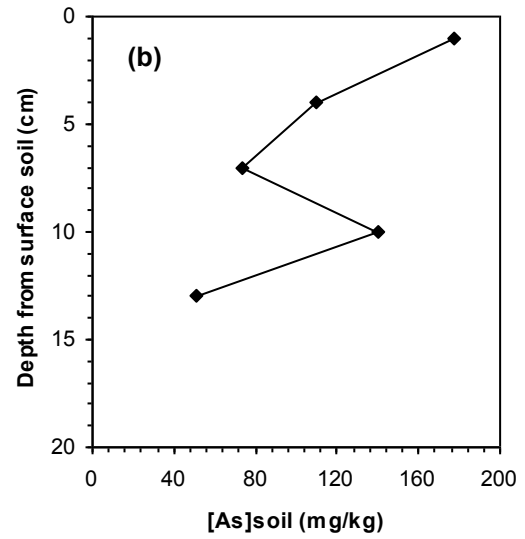
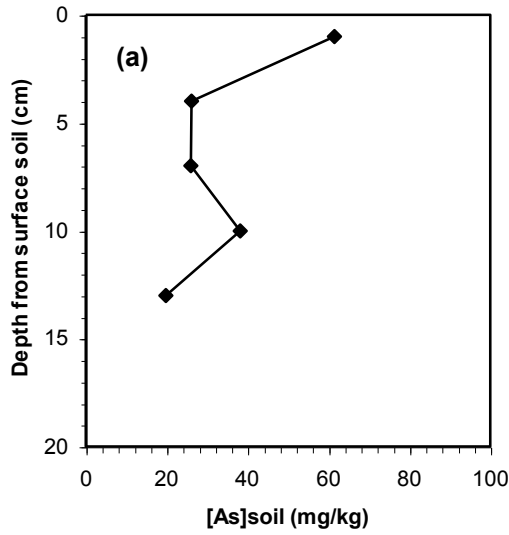
**Figure S2.** Box-and-Whisker plots for eight metal concentrations in five groundwater samples. [(—) indicates median; lower and upper box boundaries 25<sup>th</sup> and 75<sup>th</sup> percentiles of each distribution; Whiskers as vertical lines ending in horizontal lines at the largest and smallest observed values; (\*) indicates outside value and (O) far outside value.]



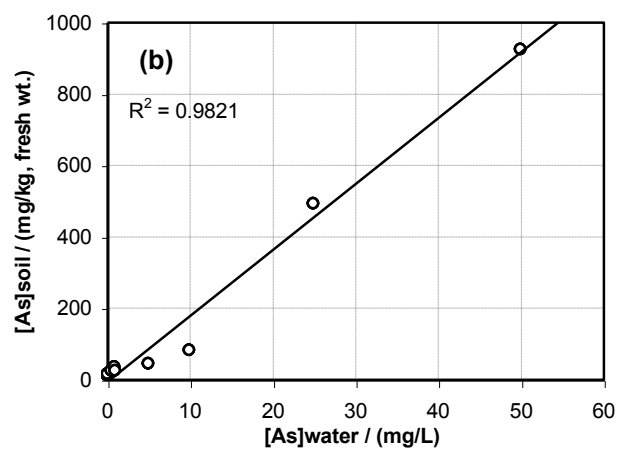
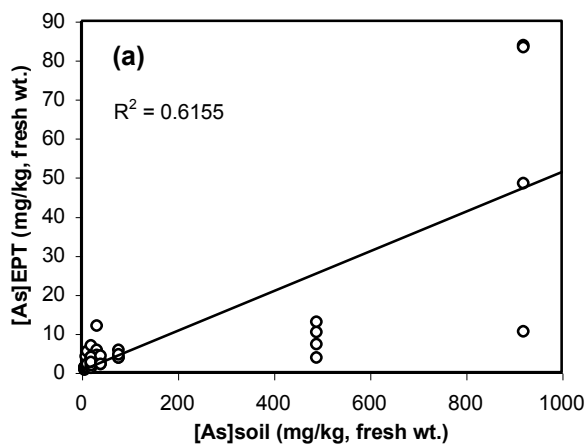
**Figure S3.** Variation in arsenic concentrations in Okra with respect to time. [The groundwater arsenic concentrations mentioned at the top are in  $\text{mg L}^{-1}$  unit.]



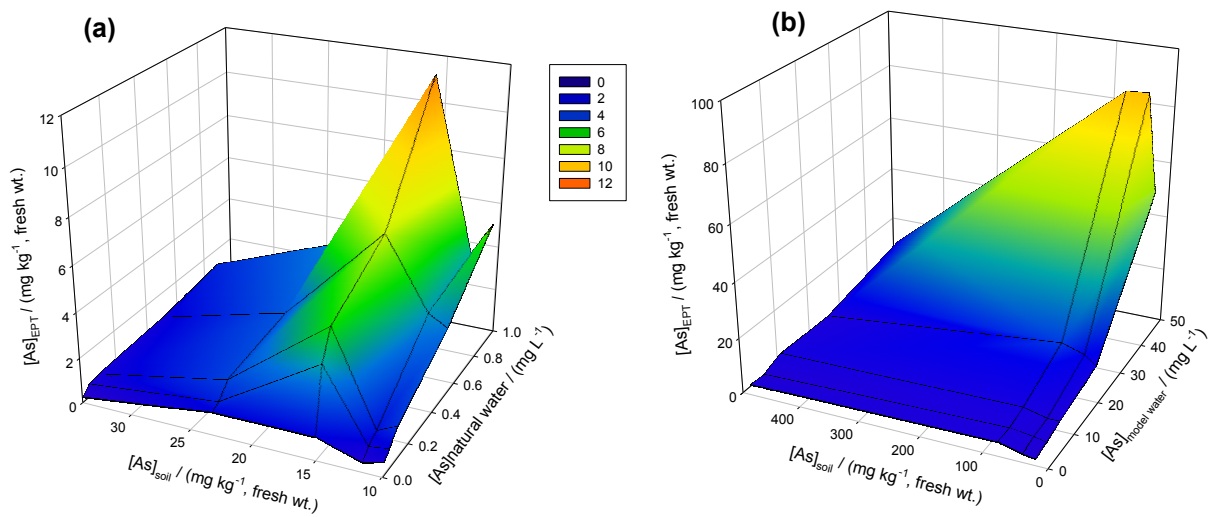
**Figure S4.** Distribution of arsenic in roots, stems and leaves/fruits of six vegetable plants.



**Figure S5.** Vertical distribution of arsenic in soil. [(a) low arsenic contaminated soil, (b) moderate arsenic contaminated soil.]

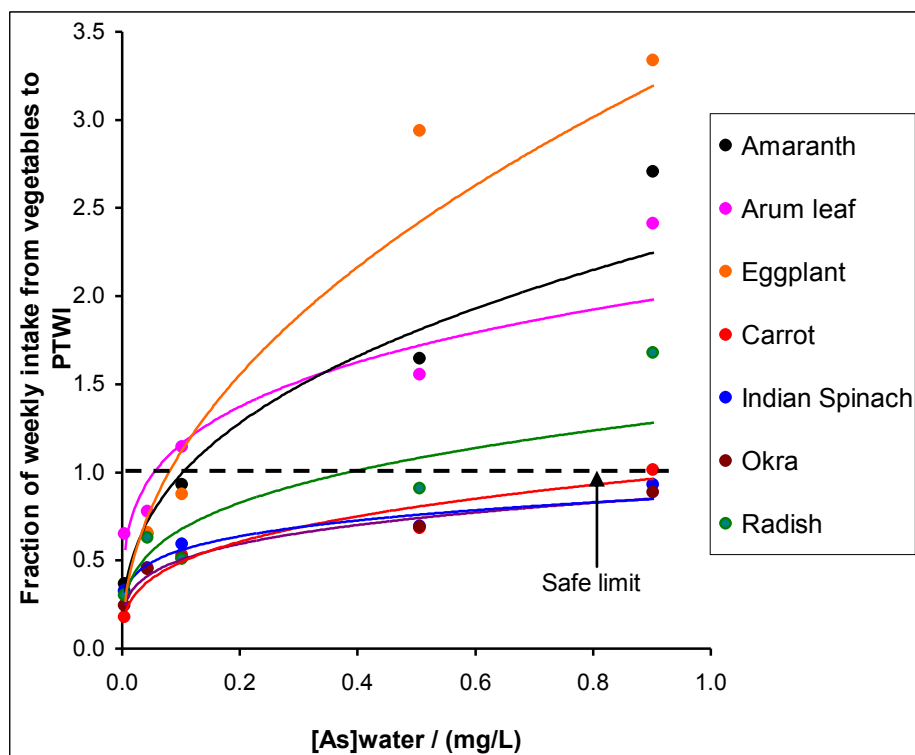


**Figure S6.** Relationship amongst arsenic concentrations of EPT, irrigation water and soil. [(a) for EPT - soil, (b) for soil - irrigation water.]



**Figure S7.** Relationship between arsenic concentrations of EPT, irrigation water and soil in combined. [(a) for natural groundwater, (b) for model water.]





**Figure S8.** Fraction of weekly intake from vegetables to PTWI for inorganic arsenic.

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