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Arsenic Contamination in Soil–Water–Plant (Rice, *Oryza sativa* L.) Continuum in Central and Sub-mountainous Punjab, India

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Abstract In the present study, Arsenic (As) concentrations in underground water, soil, and plants (rice) and their inter-relationships in central and sub-mountainous Punjab, India were studied. Approximately, 32 % of the tubewell water samples had As concentrations greater than the maximum permissible limit (10 μg As L^{-1}) set by the World Health Organization (WHO) whereas in hand pump waters, As concentrations were within the safe range (i.e. <10 μg As L^{-1}). As concentrations in tubewell waters were significantly correlated with As concentrations in surface soil ($r = 0.57$; $P < 0.05$) and plant samples ($r = 0.27$–0.82; $P < 0.05$) in central and sub-mountainous Punjab. The estimated daily intake of As through human consumption in rural and urban population was 0.016 and 0.012 μg day^{-1} kg^{-1} body weight respectively.

Keywords Arsenic · Soil–water–plant continuum · Punjab · India

Arsenic (As) is one of the most hazardous metalloid present almost everywhere in the environment. Exposure to higher As concentrations can cause several diseases including Arsenicosis, cancers, hyperkeratosis, restrictive lung diseases, and ischemic heart diseases (Chakraborti et al. 2004). High concentrations of As in drinking water have been reported from almost all over the world (Ng 2005). In addition to drinking waters, food crops have also been considered as important pathways for As intake by humans (Stone 2008). Plants can uptake As present in the surface soils. Previous studies have indicated that irrigation with As contaminated waters can cause greater As retentions in soils via adsorption on soil exchange complexes (Saha and Ali 2007). In this way, As contaminated soils and irrigation waters may increase As in the food crops via plant uptake mechanisms. Interestingly, among all the agricultural crops, rice is the most susceptible crop that accumulates greater As concentrations probably due to greater requirements for irrigation water (Stone 2008). Sengupta et al. (2006) reported that rice is an important source of dietary intake of As in humans and animals in Indian sub-continent. However, information on the relationships between As concentrations in irrigation waters, and soils, and its uptake by rice crop is limited and needs further exploration.

Objectives of this study were to determine As concentrations in underground waters as well as soils and to examine their effect on the uptake of As in rice plant parts in central and sub-mountainous districts of Punjab, India. As a baseline research, this would help in estimating the daily intake levels of As through rice consumption as well as evaluating the impeding potential health risks involved. The present study area consisted of two physiographic regions namely central plain and sub-mountainous regions of Punjab, India (Fig. 1) each consisting of four districts. Sub-mountainous region consisted of districts namely Gurdaspur, Hoshiarpur, Nawanshehar, and Rupnagar. The four districts constituting the central plain region were namely Amritsar, Jalandhar, Kapurthala, and Ludhiana.

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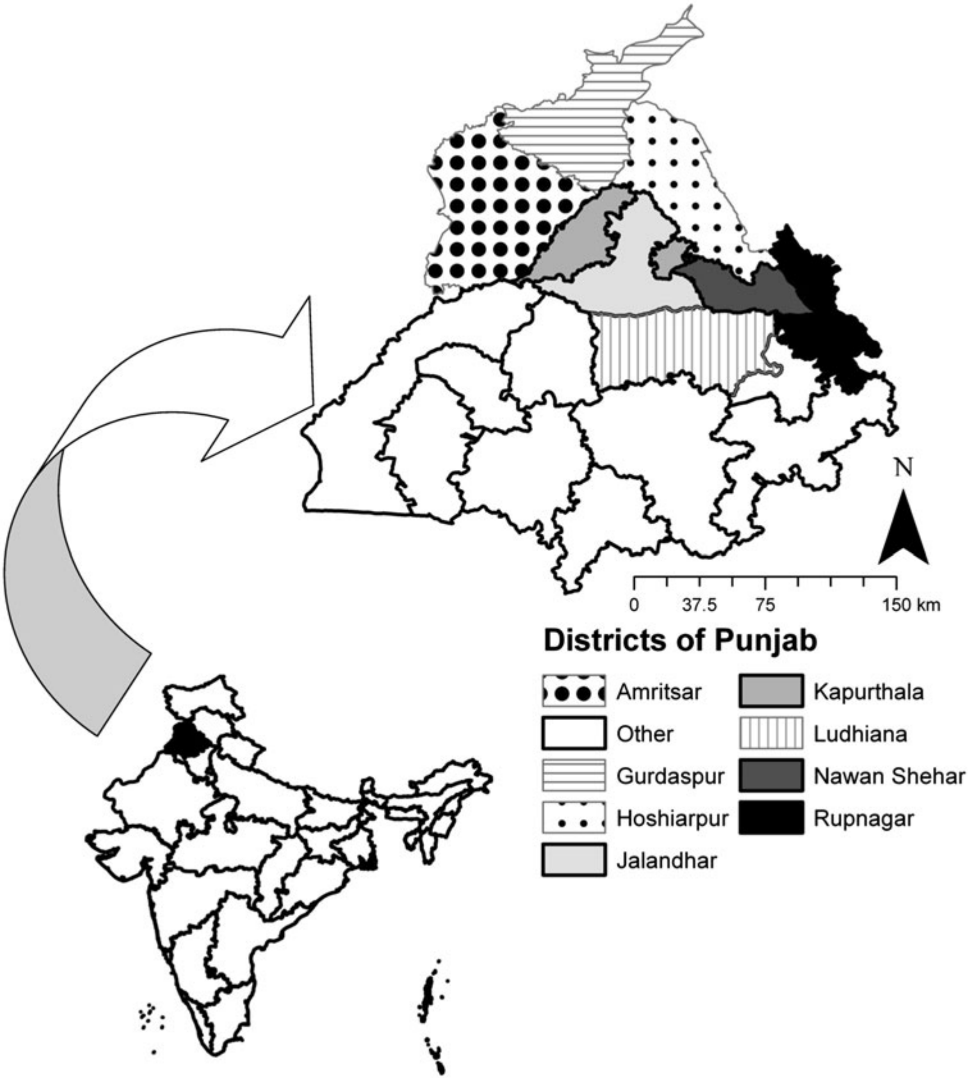

Fig. 1 Study area comprising central and sub-mountainous districts of Punjab, India

Some of the physico-chemical parameters of soils of the region have been reported in Table 1.

Materials and Methods

In each district, four villages were randomly selected for the sampling purpose. Five tube well operated farmer fields in each village were selected to study the water–soil–plant As uptake mechanism. To avoid As contamination from any other source of irrigation water, only those fields were selected where tubewell water was the only source of irrigation. Additionally, one hand pump water sample was collected from each village to examine As concentrations in shallow aquifers. Twenty tube well and four hand pump water samples from each district were collected in the month of August 2006. Immediately, after sample collection, 1 mL concentrated HCl was added to the sample bottle for sample preservation. For As analysis, 100 mL of each sample was acidified using 0.5 mL of concentrated HNO_3 and 2.5 mL of concentrated HCl. Samples were then heated on a hot plate to reduce the volume to 15–20 mL. The aliquot was filtered through Whatman No. 42 filter paper and the final volume was adjusted to 25 mL using double distilled water.

Soil samples were collected during October 2006, from the above mentioned farmer fields. Twenty (five from each village) surface (0–15 cm) and one profile sample (0–200 cm) were collected from each districts of the study area. In this way, a total of 21 surface soils (0–15 cm) and six sub-surface (15–200 cm) soil samples were collected from each districts of the study area. For As determination, one gram of soil sample was transferred to a 250 mL conical flask and 10 mL of concentrated HNO_3 was added to the sample. Covered samples were heated and refluxed for about 15 min without boiling. After cooling, 5 mL of HNO_3 was added and the sample was refluxed for another 30 min. The solution was allowed to evaporate to a volume

Table 1 Physico-chemical parameters of soils in central and sub-mountainous Punjab, India

Parameter	Range Surface soil	Average	Range Sub-surface soil	Average
pH	7.45–8.70	8.21 ± 0.22	7.68–8.70	8.20 ± 0.25
OC (%)	0.13–0.68	0.42 ± 0.10	0.09–0.56	0.24 ± 0.12
EC (μmhos cm^{-1})	0.10–0.58	0.23 ± 0.08	0.06–0.33	0.18 ± 0.06

of 5 mL without boiling. After cooling the sample, 2 mL of double distilled water and 3 mL of 30 % H_2O_2 were added to the sample. Then the beaker was covered with a plastic sheet and placed over a hot plate. During heating, 30 % H_2O_2 was added to the sample in 1 mL aliquots until the effervescence was minimized. After cooling the sample, 5 mL concentrated HCl and 10 mL water were added to the sample. Covered beaker was then placed over the hot plate for 15 min for additional refluxing without boiling. The samples were filtered through Whatman No. 42 filter paper and diluted to 25 mL volume using double distilled water.

Rice (grain and straw) samples were collected in the month of October 2006 from the same sites from where surface soil samples were collected. These samples were washed with 2 % HCl and double distilled water to remove all the impurities and dust particles. Plant samples were digested using the method outlined by Miller (1998). The As content in all the water, soil and plant samples was determined using ICP-AES (iCAP 6300) technical specifications for which have been reported in Table 2.

To validate the analytical method, a CertiPUR As (Arsenic) 1,000 ppm standard solution (traceable to SRM from NIST H_3AsO_4 in HNO_3) LOT HC625147 purchased from Merck was used. After every 30 samples, a blank and a standard 40 ppb As used for calibration were tested in order to validate the analytical work. A relative error of less than 5 % was observed between measured and Merck arsenic standard solutions. All the samples were statistically analyzed for As content using Duncan's Multiple Range Test (Duncan 1955). In this study, correlation coefficients were calculated using MS excel 2007.

Table 2 Instrumental specifications of iCAP 6300

Spectrometer	Echelle type, 52.91 grooves/mm ruled grating, ross dispersion prism
Detector	High performance CID86 chip
Detection limit for As	1.42 ppm at a wavelength 189 nm
RF source	27.12 MHz solid state, 750–1500 watts output power
Sample pump	3-channel, 12 roller peristaltic
Spray chamber	Glass cyclonic type
Plasma gas	Fixed 12 L/min, argon
Nebulizer gas	Pressure control, from 0–0.4 MPa
Auxiliary gas	4 fixed flows, 0, 0.5, 1.0 and 1.5 L/min

The provisional tolerable weekly intake (PTWI, given by Joint FAO/WHO Expert Committee on Food Additives (JECFA) (UNEP/FAO/WHO 1992) was used to assess the potential health risks. Firstly, the tolerable daily intake (TDI) was calculated as per the equation:

$$\mathrm{TDI} = \mathrm{PTWI}/\ 7\ \mathrm{TDI} = 350/7 = 50\left(\mu\mathrm{g\ day^{-1}kg^{-1}bw}\right)$$

Then using this value, estimated daily intake (EDI) of As through rice consumption for rural and urban populations in India were evaluated using the formula given by Rezaitabar et al. (2012):

$$\mathrm{EDI} = (\mathrm{C}\ \times\ Cons./Bw)\left(\mu\mathrm{g\,day^{-1}kg^{-1}bw}\right)$$

where C, *Cons*., and B*w* represent the As concentration in rice grains, daily average rice consumption and the body weight respectively. Per capita daily average rice consumption (*Cons.*) in India has been calculated to be 0.24 and 0.18 kg respectively for the rural and urban populace by Chatterjee et al. (2009). Similar to an earlier study (Rezaitabar et al. 2012), B*w* has been set to 60 kg in the present study.

Results and Discussion

In central and sub-mountainous Punjab, As concentrations ranged from 0.42 to 20.97 μg As L^{-1} in tubewell and hand pump water samples (Table 3). Tubewell As concentrations

Table 3 Arsenic concentrations in tubewell water, hand pump water, surface soil, sub-surface soil, plant grain, and straw samples in central and sub-mountainous Punjab, India

Sample	Range	Mean ± SD
Tubewell water	0.42–20.97	9.21 ± 3.24
Hand pump water	0.60–9.58	5.40 ± 2.42
Surface soil	0.68–2.33	1.46 ± 0.38
Sub-surface soil	0.26–1.85	1.23 ± 0.32
Rice straw	2.12–26.01	8.78 ± 3.75
Rice grains	1.01–13.95	3.94 ± 1.58

For water, soil and plant samples units of As concentration are μg As L^{-1}, mg As kg^{-1} and μg As kg^{-1} respectively

SD Standard deviation

Table 4 District wise distribution of arsenic concentrations in groundwater, soil, and plant samples in central and sub-mountainous Punjab, India

District	Arsenic concentration					
	Tube well water	Surface soil	Rice straw	Rice grains	Hand pump water	Sub surface soil
Amritsar	10.31c	1.35a	9.78c	4.51bc	4.52a	1.25a
Gurdaspur	9.25bc	1.36ab	5.77a	2.96a	7.31a	1.42a
Hoshiarpur	9.23bc	1.51bc	6.67a	3.42ab	6.45a	1.32a
Jalandhar	6.16a	1.34a	9.32bc	4.21bc	4.05a	1.38a
Kapurthala	10.76c	1.35a	8.92bc	3.78ab	4.41a	0.98a
Ludhiana	8.72b	1.43ab	10.07c	4.15bc	5.86a	1.12a
Nawanshehar	9.66bc	1.63 cd	7.12ab	3.53ab	3.87a	1.36a
Rupnagar	9.62bc	1.73d	12.59d	4.95c	6.73a	1.00a

**Mean followed by different letters (a, b, c, and d) denotes significant differences at ($p < 0.05$)

For water, soil and plant samples units of As concentration are μg As L^{-1}, mg As kg^{-1} and μg As kg^{-1} respectively

showed significant spatial variations in central and sub-mountainous Punjab (Table 4). For example, in two central plain districts namely, Amritsar and Kapurthala, mean tubewell As concentrations were greater (10.31 and 10.76 μg As L^{-1} respectively) than Jalandhar (6.16 μg As L^{-1}). In a previous study, Hundal et al. (2007) reported a wide range of As concentrations (3.5–42.5 μg As L^{-1}) in the central and sub-mountainous districts of Punjab.

However, Hundal et al. (2007) did not report the variations in As concentrations in shallow hand pumps and deeper tubewell waters in the central and sub-mountainous Punjab. Our results showed that 32 % of tubewell water samples were not fit for human consumption (>10 μg As L^{-1}) based on the WHO guidelines (WHO 2005) whereas all of the hand pump water samples had As concentrations lower than 10 μg As L^{-1} in central and sub-mountainous Punjab. Mean As concentration in tubewell water samples was almost two folds of that observed in the hand pump water samples (Table 3). In the study area, depth of the hand pumps is 40–50 m whereas the tubewells are usually at a depth of >200 m. Greater concentrations of As in the tubewell waters may be due to presence of As containing minerals in the deeper aquifers of central and sub-mountainous districts of Punjab (Sidhu 2007).

In Punjab, tube well water is the primary source of irrigation. Greater exploitation of the underground waters has lowered water table that exposes As containing salts/minerals to aerobic conditions in the soils. In the groundwater alluvial aquifers of Punjab, high pH (>8.0) and high concentrations of phosphate, borate, sulfate, and hydroxyl anions were responsible for the wide spread release of As concentrations from sulfide oxidation and oxyhydroxide of iron (Hundal et al. 2007). Therefore, greater concentrations of As in deep tubewells than shallow hand pump waters probably indicate the dominance of natural rather than anthropogenic sources such as application of As containing chemicals in the agricultural crops (Sidhu 2007).

Mean As concentration was greater in surface (1.46 mg As kg^{-1}) than sub-surface soils (1.23 mg As kg^{-1}) in central and sub-mountainous Punjab (Table 3). This could be due to irrigation with As contaminated water to the soils. Previous studies have reported that the top soil was mainly affected by the As inputs with irrigation water. For example, Panaullah et al. (2009) reported that approximately whole of the As added through irrigation was retained in the surface soil. Another possibility for higher As concentrations in the surface than sub-surface soils could be the result of long term use of arsenical pesticides along with irrigation with As contaminated water. However, the effect of arsenical pesticides on soil As concentrations was beyond the scope of this study.

In the present study, mean As concentration was approximately two folds greater in rice straw (8.78 μg As kg^{-1}) than grain (3.94 μg As kg^{-1}) samples (Table 3). This remarkable shielding of rice grains from the contaminated irrigation water and soil As could be attributed to the presence of rice husk on the grains. Rice husk acts as a translocation barrier for which As could not mobilize easily into the rice grains (Bhattacharya et al. 2009). The greater As concentrations in the straw samples could be a threat to the animals because they consume straw as fodder. In our study, all the plant samples in the region had As content less than permissible limit of 1.0 mg As kg^{-1}dry weight. Previously, Walsh and Keeney (1975) reported that plants seldom accumulate As at concentrations hazardous to human and animal health because phytotoxicity usually occurs much before such threshold concentrations are reached in plants. India has not yet established maximum permissible limits for As intake in human food. Therefore, such studies may act as a baseline for establishing such

Table 5 Mean estimated daily As intake by a 60 kg body weight person (µg day^{-1} kg^{-1} bw)

Mean rice grain As	TDI (As)	EDI		*% TDI	
(µg As kg^{-1} dry weight)	(µg day^{-1} kg^{-1} bw)	Rural	Urban	Rural	Urban
3.94	50	0.016	0.012	0.03	0.02

*Denotes EDI percentage of TDI

Table 6 Relationships between arsenic concentrations in tubewell water, surface soils, and rice plants in central and sub-mountainous Punjab, India

Arsenic concentrations	Correlation coefficient
Tubewell water and surface soil	0.569*
Tubewell water and plant grain	0.272*
Surface soil and rice straw	0.818*
Surface soil and rice grain	0.269*
Rice straw and rice grain	0.394*

*Denotes significant relationships at ($p < 0.05$)

limits. The estimated daily intake (EDI) of As in this study was 0.03 % and 0.02 % of TDI for rural and urban population, respectively (Table 5). This was much lower than the JECFA's safety standard limit i.e. TDI of 50 µg As day^{-1} kg^{-1} body weight set for As. Similar results have been reported in an earlier study by Munoz et al. (2005) from Chile.

In this study, As concentrations in the surface soil, tubewell water, rice grains, and straw were significantly and positively correlated with each other (r > 0.26–0.82; $p < 0.05$) (Table 6).

This indicates towards higher accumulation of As in soils irrigated with As contaminated irrigation waters which would finally result in a reduction of soil quality and crop yields. Similar, positive and significant relationships between As concentrations in irrigation water and rice plants have been reported in earlier studies also (Hossain et al. 2008). Results of this study suggest that though in the present study area, As contamination is an impending threat to human health through drinking water. But due to lower As uptake by rice crop it does not pose any threat to human population in near future via water–soil–plant-human food chain in Punjab, India. This study provided preliminary links between As concentrations in the water–soil–rice plant continuum in central and sub-mountainous Punjab. Regular monitoring and increased awareness among the policy makers and scientific community is required to assess the harmful effects of As contamination on the environment.

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