

Arsenic in soil, vegetation and water of a contaminated region

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ABSTRACT: Arsenic concentrations of surface waters, soils and plants were surveyed in three contaminated villages of Bijar County. Total arsenic in water samples (4.5 to 280 µg/L) was correlated with electrical conductivity, total dissolved solid, total hardness, alkalinity, chloride, sulphate, bicarbonate, calcium and sodium ($p < 0.001$). Total arsenic in the soils ranged from 105.4 to 1500 mg/kg. Some of the soil factors play an important role in soil arsenic content and its bioavailability for organisms. In general, the arsenic concentrations in plants were low, especially in the most common wild species. Among 13 plant species, the highest mean arsenic concentration was found in leaves of *Mentha Longifolia* (79.4 mg/kg). Arsenic levels in soils and plants were positively correlated, while the ability of the plants to accumulate the element, expressed by their biological accumulation coefficients and arsenic transfer factors, was independent of the soil arsenic concentration. Relationships between the arsenic concentrations in plants, soils and surface water and the environmental aspects of these relationships have been discussed in comparison with literature data. The accumulation of arsenic in environmental samples (soil, sediment, water, plant, etc.) poses a potential risk to human health due to the transfer of this element in aquatic media, their uptake by plants and subsequent introduction into the food chain.

Keywords: Arsenic contamination; Arsenic transfer coefficient; Biological accumulation factor; Groundwater

INTRODUCTION

Arsenic can be found naturally on earth in small concentration. It occurs in soil and minerals and it may enter air, water and land and uptake by plants (Mahzuz *et al.*, 2009; Reza and Singh, 2010). Plants vary considerably in their tolerance of arsenic and in the amount of arsenic that they can take up from soils and water (Goyal *et al.*, 2008; Feng *et al.*, 2009; Malakootian *et al.*, 2009; Urik *et al.*, 2009). Arsenic is a naturally occurring element present in the both inorganic and organic forms in different environmental and biological samples and its concentrations may be increased by anthropogenic contamination (Villa-Lojo *et al.*, 2002). Arsenic presence in water has been reported from several parts of the world, like USA, China, Chile, Bangladesh, Taiwan, Mexico, Argentina, Poland, Canada, Hungary, Japan, India, Vietnam, Nepal (Jain and Ali, 2000; Jack *et al.*, 2003) and recently from Iran (Mosafari *et al.*, 2003). Inorganic As is considered

to be the major form of As in groundwater, surface water, soil and various foods (Babel and Opiso, 2007) and have been classified as group I carcinogens based on human epidemiological data (Hughes, 2002). Millions of people have been exposed to arsenic contamination through drinking water. In some regions of Pakistan, drinking water comes from groundwater and surface water, including rivers, lakes and reservoirs (Duker *et al.*, 2005). The present free style way of disposing agricultural, industrial and domestic effluents into natural water-bodies results in serious surface and groundwater contamination (Abdel-Ghani and Elchaghaby, 2007; Okafor and Opuene, 2007; Samarghandi *et al.*, 2007; Abdel-Ghani *et al.*, 2009; Karimi *et al.*, 2010). Run-off from agricultural and saline seeps subject the most vulnerable water bodies to pollution and increased salinity, so the freshwater lakes are highly impacted. Arsenic contaminated groundwater is not only used as a source of drinking water, but also

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extensively used for irrigation in some regions. Uptake of As by plants depends on the physicochemical make up of the plant species and soil. The concentration of arsenic in cereals, vegetables and fruits is directly related to the level of arsenic in the soil (Kazia et al., 2009). Severe arsenic contamination of soils may cause arsenic toxicity in plants, animals and human (Warren et al., 2003). Remediation of arsenic contaminated soils has thus become a major environmental issue. Kurdistan, a western province of Iran, is facing the problem of arsenic con-tamination. In the northeast of the province, there are some villages in Bijar County where drinking water has been contaminated with naturally occurring arsenic. Widespread water and crop contamination in these areas are originating from the natural release of arsenic through aquifer and sedimentary rocks. Also, this element is a fundamental constituent of the sulfide mineral arsenopyrite (FeAsS), as well as the minerals lingite (FeAs), realgar (As₂S₃) and orpiment (As₂S₃); (Karimi et al., 2010). In some villages the arsenic in water supplies, exceeds 1,000 µg/L with an average con-centration of 290 µg/L (Mosaferi et al., 2009). Chronic poisoning manifestations, such as skin lesions: keratosis, pigmentation and even amputation due to gangrene, have been reported. Despite many studies on the occurrence of arsenic in drinking water in Bijar county and the resulting poisoning of the inhabitants (Mosaferi et al., 2003, 2005 and 2009), there is no information on the arsenic

concentrations in soil and plant species and on the arsenic bioaccumulation potential of plant species indigenous to this area. Therefore, the present study has been performed with the following objectives: (1) to determine the As concentrations in water, soil and leaves of plants growing in contaminated areas to explore the degrees of environmental contamination; (2) to assess the potential risk of environmental arsenic in the Bijar area and (3) to correlate the As concentrations in ground water soil and plants. To do so, three villages located in highly As-contaminated areas in Bijar County were examined from May to October 2008.

MATERIALS AND METHODS

Study area

Kurdistan province is located in the West of Iran, bordering Iraq from 34° 44'2" to 36° 30'2" North, and, 45° 31'2" to 48° 16'2" East (Fig. 1). This province is one of the most mountainous provinces of Iran and has a generally cold climate. Kurdistan province represents about 1.7 % of the area of the entire country and has more than, 450,000 inhabitants. In the Northeast of the province, there are some villages in Bijar County (a 580-km² area with an average altitude of about 1750 m above sea level) where drinking water has been contaminated with naturally-occurring arsenic (Mosaferi et al., 2005). In addition to Bijar, the region of Ghorveh has also faced a similar problem (Mosaferi et al., 2003).

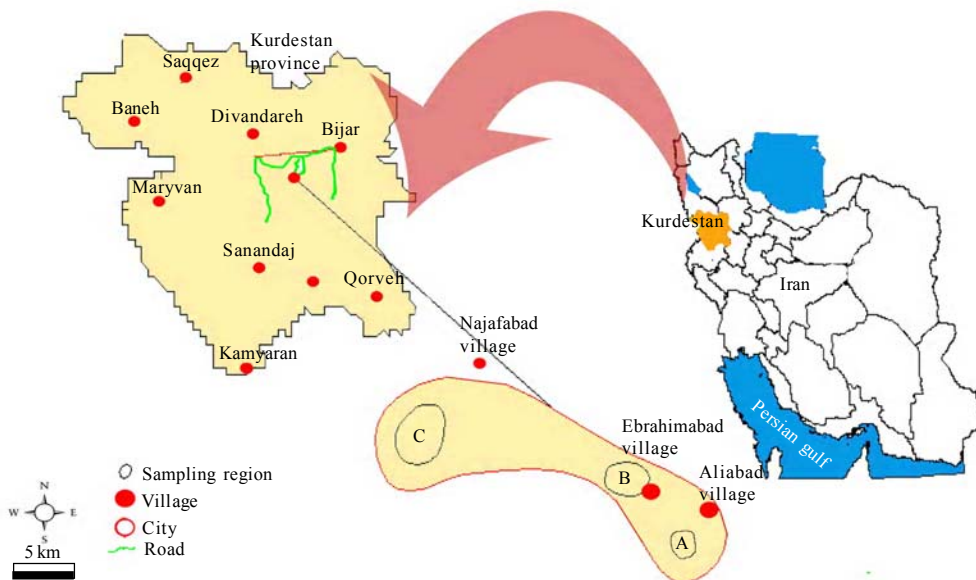


Fig. 1: Location of the study area in Bijar County, Iran

The present study was carried out in three contaminated villages of Bijar County. These villages were selected on the basis of the high arsenic contamination and the inadequate supply of safe drinking water (Mosaferi *et al.*, 2009). These are Aliabad, Ebrahimabad and Najafabad and their positions are shown in Fig. 1.

Surface water sampling and analysis

Shallow wells for sampling were selected based on a previous study regarding arsenic occurrence in drinking water of the Kurdistan province (Mosaferi *et al.*, 2003). Water samples were collected from the most contaminated shallow wells used for agricultural irrigation (one well per village). Water samples were filtered through a 0.45 μm membrane filter and divided into two portions. One portion was acidified with HNO_3 (0.2 % v/v) for the analysis of As concentrations, while the other was left un-acidified for pH, electrical conductivity (EC), total dissolved solid (TDS) and total hardness (TH) measurements. Water samples were stored in a refrigerator at 4 °C to prevent changes due to chemical or biological activity as well as loss due to evaporation. The analyses of water samples represented the total load of arsenic (dissolved plus colloid bound) in the water samples. Physical parameters like pH, TDS and EC were determined by a portable combine meter (Milwaukee, model SM802) and TH was measured by the following equation: $\text{TH} = 2.497\text{Ca} + 4.115\text{mg}$ (Vasanthavigar *et al.*, 2010). The major cations and anions including sodium, potassium, calcium, magnesium, chloride, bicarbonate and nitrate were determined according to standard specifications prescribed by APHA *et al.*, (1992). The samples were analyzed for arsenic using hydride generation atomic absorption spectrometry with a flow injection hydride generator. Standard materials for chemical analysis were purchased from Merck and the calibration curve fit (at least five standard concentrations) was of $R^2 > 0.97$ in all cases. The method's recovery of As (0.79 ± 0.08 mg/kg) from certified reference material (Beach leaves material FD8, Commission of the European Communities, Joint Research Centre ISPRA) was not significantly different from the certified reference value (0.76 ± 0.1 mg/kg). The mean As concentration in blank digests was 0.08 $\mu\text{g/L}$ and the detection limit for As in plant tissue was 0.05 $\mu\text{g/L}$.

Soil sampling

Soil was collected during the period of sampling from three As-contaminated areas (18 samples per each area). The soil samples were taken in three different ways. First, surface soil samples were collected at different distance

(0, 400 and 800 m) from the contaminated shallow well of each village to determine the extent of As contamination. Second, samples were taken from different depth (surface and subsurface). Third, soil attached to plant roots (rhizosphere soil) was sampled to determine the bioaccumulation factor of each plant species collected from the study areas.

Soil characteristics

Soil properties were determined as follows: pH was determined potentiometrically in a soil paste saturated with water and organic matter was determined by dichromate oxidation using the Tiurin method (Soon and Abboud, 1991). Total soil phosphorus (P) was measured by the Olsen method (Olsen *et al.*, 1954). The particle size distribution (sand, silt and clay) was analyzed by the hydrometer method (Ashworth *et al.*, 2001).

Soil arsenic analysis

A portion of each sample (about 5 g) was spread to remove roots and external contaminants, air-dried and oven-dried at 80 ± 5 °C for 12 h and ground to fine powder with an agate mortar and preserved in desiccators for subsequent analysis. For total As analysis, soil samples (0.5 g) were transferred to a Kjeldahl digestion tube for extraction with 10 ml of a 3:1 HCl/ HNO_3 mixture. Tubes were left overnight at room temperature and then placed in a heating block. After cooling, the digests were filtered through a moistened Whatman No. 40 filter paper and made up to 50 mL volume with distilled water. The digest was analyzed for arsenic using hydride generation-atomic absorption spectrometry as described above.

Plant sampling and analysis

The plant samples were taken from the same location as the soil samples. Only plant species which were found frequently in the study areas and/or had a high shoot biomass were collected. A total of 46 plant samples belonging to 13 different species were collected. Only the aerial parts of the plants (stems, branches and leaves) were collected; the roots were not sampled to avoid contamination with adhering soil material. Plant samples were washed with tap water, rinsed with deionized water and oven-dried at 50 – 55 °C for 72h. They were digested as described by Meharg and Jardin (2003). Analysis of As was performed by atomic absorption spectrophotometry, as described above.

RESULTS AND DISCUSSION

Arsenic in surface water

Table 1 shows the characteristics of the shallow wells used for irrigation in the villages studied. The characteristics were different and considering hardness, the water sources were classified into hard or very hard water (total hardness >150 mg/L as CaCO₃). The highest levels of hardness and TDS were observed in Aliabad, while in Najafabad the levels were the lowest. Except for arsenic, the concentrations of heavy metals were lower than the WHO guideline levels. All the shallow wells sampled in this study, had arsenic concentrations higher than the WHO drinking water standard of 10 µg/L. The electrical conductivity (EC) of the surface water varied from 500 to 927 ds/m with pH 7.38 to 7.72, indicating that TDS in water is within the limit. There were significant correlations between arsenic levels and EC, TDS, TH, alkalinity, chloride, sulphate, bicarbonate, calcium and sodium of analyzed waters (p<0.001) (Table 1). The highest correlation was observed between arsenic and TDS (R=0.867). The As concentration of the Aliabad shallow well was higher than 1200 µg/L, i.e. more than 120 times higher than the WHO drinking water standard of 10 µg/L.

Soil characteristics

The pH, organic matter and soil texture of each site, together with the total As concentrations, are given in Table 2. The pH was neutral in area A and area B, while it was alkaline area C. All the sampling sites showed very low levels of P in the soil. The soil texture analysis revealed significant differences between sites, particularly with regard to the silt and clay fractions. In area B the silt content was much higher than that of clay, whereas the opposite pattern was found in area A (Table 2). Total soil P at each sampling site was within the range normally encountered in soils (4.5–28 mg/kg) and showed no significant differences between sites.

Arsenic in soil

The highest soil As concentrations were found in the area around the shallow well in Ebrahimabad (area B; Table

2). In general, the soil arsenic concentrations in area A and B were much higher than the average toxicity threshold of 40 mg/kg established for crop plants as reported by Sheppard (1992). The mean As concentration in Najafabad was much lower than in Aliabad and Ebrahimabad (Table 2). The As concentrations of 0-20 cm and the 20-40 cm soil layers were comparable and there was no consistent decrease of the soil As concentration with increasing distance from the well (Table 2).

Arsenic in plants

A total of 13 plant species belonging to 11 families were sampled (Table 3). In area C, plants showed very low As concentrations, occasionally below the limit of instrumental detection (Table 3). Only 2 species, among which *Mentha longifolia*, had As concentration above the normal range of 0.1-0.5 mg/kg (Lombi and Nolan, 2005). In the B area, plants had much higher As concentration, usually above 5 mg/kg (10 out of 13), but only *Mentha longifolia* showed considerable accumulation, with 86.25 mg/kg of As in its leaves (Table 2). Of the 13 plant species sampled in area A 8 had more than 10 mg/kg of arsenic in their leaves, again with *Mentha longifolia* as the best As accumulator (79.34 mg/kg).

Relationship between the As concentrations in surface soils and surface water

The As concentration ranged from 180 to 1500 mg/kg in surface soil and from 4.5 to 1280 µg/L in surface water. The As concentration of surface soil tended to increase with the As concentration of the surface water with linear regression coefficients of 0.371 (P<0.001), 0.221 (P<0.001) and 0.026 (P<0.01), for Aliabad, Ebrahimabad and Najafabad respectively.

Relationship between the As concentrations in plant leaves and surface soils

There is a relationship between As concentrations of plant leaves and surface soils at the three studied sites in Bijar county (Fig. 2). Overall, shoot As concentrations increased more or less linearly with total

Table 1: Analysis of local surface water sources from three contaminated areas in Bijar County

Location	As (µg/L)	K (meq/L)	Na (meq/L)	Ca (meq/L)	Mg (meq/L)	HCO ₃ (meq/L)	Cl (meq/L)	NO ₃ (meq/L)	EC (ds/m)	pH	TDS (mg/L)	SAR	Hardness (mg/L CaCO ₃)
Aliabad (Area A)	1280.56	0.065	0.221	1.832	7.896	5.723	0.35	0.09	750.12	7.38	553.23	0.09	192.52
Ebrahimabad (Area B)	400.89	0.155	0.521	1.712	6.121	5.865	1.14	0.13	927.56	7.72	434.85	0.02	156.96
Najafabad (Area C)	4.53	0.012	0.027	3.521	2.512	3.645	1.92	0.26	500.32	7.58	302.63	0.26	120.46

Table 2: Arsenic concentration and some edaphic parameters in the three contaminated areas in Bijar County

Location	Block	Total As in soil samples (mg/kg)		Soil texture (%)			pH	EC (ds/m)	O.M (mg/kg)	Total P (mg/kg)
		0-20	20-40	Sand	Silt	Clay				
Aliabad	0	1300.36* ± 365.23	1670.56	48.21	21.02	26.01	7.11	1.177	1.12	28.21
	400 m	1180 ± 211.56	1000.54	59.52	18.01	23.06	7.59	1.596	0.965	13.02
	800 m	875.63 ± 98.65	1250.05	52.12	25.11	32.32	7.69	0.89	1.32	15.98
Ebrahimabad	0	1500.00 ± 101.87	1100.32	60.25	42.32	9.12	7.82	0.72	3.02	13.25
	400 m	1050.11 ± 113.52	1150.87	57.32	33.23	10.15	7.89	0.92	2.17	14.86
	800 m	1250.63 ± 275.63	1400.56	53.23	36.17	14.45	7.69	2.67	2.32	25.87
Najafabad	0	250.56 ± 21.32	105.45	47.11	19.03	23.45	8.25	0.85	1.56	5.81
	400 m	180.87 ± 15.56	190.32	55.12	25.04	20.52	8.15	1.05	1.62	4.53
	800 m	270.25 ± 20.52	211.12	53.52	21.32	29.52	8.32	2.20	1.89	10.02

*Mean ± SE

Table 3: Arsenic concentration in leaf samples collected from three contaminated areas in Bijar County

Family	Scientific Name	No. sample	Total As (mg/kg)		
			A	B	C
Apiaceae	<i>Falcaria scioides</i> Asch.	2	2.62 ± 0.3	9.03 ± 0.6	0.30 ± 0.01
Asteraceae	<i>Achillea millefolium</i>	6	2.31 ± 0.2	21.41 ± 1.6	0.52 ± 0.03
Brassicaceae	<i>Cardaria draba</i>	5	4.41 ± 0.05	21.32 ± 2.1	ND
Brassicaceae	<i>Descurainia sophia</i>	3	2.65 ± 0.4	4.61 ± 0.6	ND
Chenopodiaceae	<i>Chenopodium album</i>	8	25.00 ± 4.3	15.64 ± 0.2	10.41 ± 1.3
Euphorbiaceae	<i>Euphorbia spp.</i>	2	3.58 ± 0.8	10.11 ± 0.3	0.24 ± 0.02
Fabaceae	<i>Astragalus spp</i>	9	29.62 ± 2.4	23.31 ± 2.1	4.05 ± 0.03
Fabaceae	<i>Medicago sativa</i>	2	29.61 ± 2.1	23.30 ± 2.1	4.05 ± 0.06
Juncaceae	<i>Juncus spp.</i>	9	6.72 ± 0.6	6.37 ± 0.3	ND
Lamiaceae	<i>Mentha longifolia</i>	9	79.34 ± 4.0	86.25 ± 3.5	8.1 ± 0.3
Malvaceae	<i>Althaea officinalis</i>	4	7.23 ± 0.3	ND	ND
Poaceae	<i>Triticum aestivum</i>	6	24.90 ± 0.5	ND	2.52 ± 0.02
Ranunculaceae	<i>Ranunculus arvensis</i>	7	6.22 ± 0.6	9.62 ± 0.5	ND

*Mean ± SE; ND = Not Detectable; (A= Aliabad, B= Ebrahimabad, C= Najafabad)

soil As concentrations in plant species Aliabad, Ebrahimabad and Najafabad (Fig. 2), with linear regression coefficients of 0.371 (P<0.05), 0.221 (P<0.05) and 0.026 (P<0.01), respectively.

The arsenic transfer coefficient (AsTC) is defined as the shoot As (mg/kg dry wt) to total soil As (mg/kg dry wt) concentration ratio. In this study, AsTCs were in the ranges of 0.002-0.165 (Fig. 3).

The occurrence of high concentrations of arsenic (As), one of the most hazardous chemical elements in drinking water has been recognized, over the past two or three decades, as a great public health concern in several parts of the world (Mukherjee et al., 2006; Urik et al., 2009). Seventy five percent of the samples taken from the wells in the study areas contained As at concentrations higher than 100 µg/L (Table 1), greatly exceeding the maximum allowed limit for drinking waters (50 µg/L) (USEPA, 1980; WHO, 1993 and 1996).

Natural contamination of surface waters with As, has been recognized before in some villages in Bijar and Qorveh districts (Mosafari et al., 2003). The accumulation of trace elements in environmental samples (soil, sediment, water, biota, etc.) is a potential risk to human health owing to presence at excessive concentrations in drinking water, or their uptake by plants and subsequent introduction into the food chain (Davis et al., 2001).

Findings of this study (unpublished data) indicate that people in the Kurdistan Province may be overexposed not only to As, but also to NO₃⁻, Mn and S from groundwater. Adverse health effects that may manifest in the coming years are a serious concern for the local population. The data reported here is used to compare As concentration in the main plant groups that have naturally grown on As-contaminated areas. These data are important regarding assessment of the

‘risk’ that natural processes may pose to the environment in contaminated area.

The mean total As concentration in the soil ranged from 180 to 270 mg/kg in area C to 1050 to 1500 mg/kg in area B. At all the sites sampled in this study, the mean As concentrations were higher than the average toxicity threshold of 40 mg/kg established for agricultural soil (Sheppard, 1992). In soils, the bioavailability and toxicity of As and other metals are dependent on various soil parameters. Soil factors play an important role in As accumulation by plants, but their effects differ from species to species (Marin et al., 1993; Davis et al., 2001). In the areas under study here, it is the regular use of As contaminated groundwater for irrigation of agricultural land that contaminates the soil and agricultural products, potentially leading to human intake of As.

In some of the plants studied here the As concentrations are higher than the general background concentrations in plants [none detectable to 5 mg/kg dry weight] (Otte et al., 1990). The plant bioconcentration data shows that there is little relationship between the total As concentration of the soil and the soil–plant transfer coefficient.

Comparing the As concentration in the selected plants of this study with those sampled at mining sites with higher soil As concentrations (de Koe 1994; Bech et al., 1997; Madejo’n et al., 2002), the bioconcentration is found to be much lower maximum values (from less than one-half to less than one-tenth). Since As concentrations in the soils of study area were also lower, this is consistent with the idea that the soil to plant As transfer coefficient are more or less

independent of the soil As concentrations. Alternatively, since the authors of this study sampled only 13 plant species, the authors might have missed a more strongly accumulating species by chance.

It is well known that arsenate enters plant roots via the phosphate uptake system. Both ions are taken into plant roots by a common carrier; however this phosphate/arsenate plasma membrane carrier has a much greater affinity for phosphate than arsenate. Phosphate is reported to be an efficient competitive inhibitor of arsenate uptake (Meharg and Macnair, 1990). The 3 soils investigated here had normal and similar P concentrations and it seems that soil P did not significantly interfere with plant As uptake at any of these sites. In general, As uptake by plants is largely dependent on the source, chemical speciation, pedological factors (pH, Eh, organic matter and colloid contents, soil texture, minerals and drainage conditions), plant species, age and part of plants (Otte et al., 1990; Casado et al., 2007; Singh et al., 2007). Yet in this study, these factors had no apparent influence.

Concerns have been expressed regarding potential food chain transfer of soil As via plant uptake (Lombi and Nolan, 2005). Reported soil–plant transfer coefficients for As are in the range of 0.01–0.1, considerably lower than for many other elements. The arsenic transfer coefficient (AsTC) is defined as the shoot As (mg/kg dry wt) to total soil As (mg/kg dry wt) concentration ratio. It can be used to assess the As bioaccumulation capacity of plants. The AsTCs in this study ranged from 0.002 to 0.16 with the a mean value of 0.016 in area A (Fig. 3) and mean values of 0.017 and 0.025 in areas B and C respectively (Fig. 3). These values

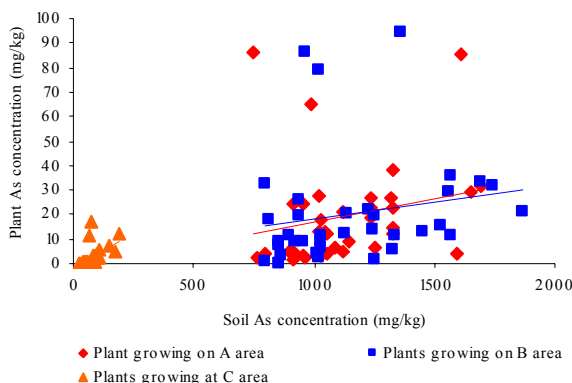


Fig. 2: Shoot As concentrations plotted against total soil As concentrations (0 – 10 cm) for samples from three contaminated areas in Bijar County

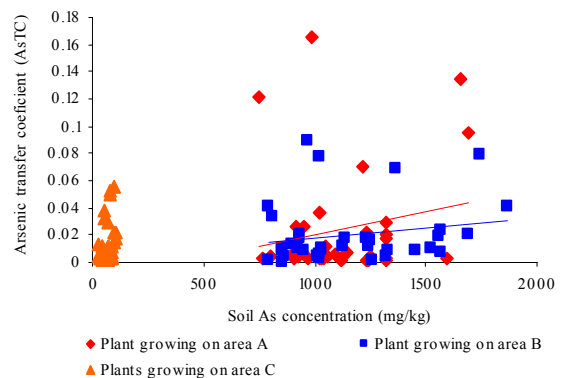


Fig. 3: Soil-plant transfer coefficients (AsTC) plotted against total soil As concentrations (0 – 10 cm) for samples from 3 contaminated areas in Bijar County

fall well within the typical range for non-polluted soils (Sheppard, 1992), despite the presence of elevated soil As.

This strongly suggests a very low soil-plant transfer of As that is independent of both soil As concentration and soil As source. This was especially evident for the most common crop species (wheat) and in common wild herbs (*Achillea millefolium*). However, there is considerable variation among species regarding their AsTC values. Among all plant samples, *Mentha longifolia* was a particularly effective foliar accumulator of As. Therefore, its potential use as a phytoremediator should be assessed.

CONCLUSION

Tissues of the 13 plant species generally exhibited an As content positively correlated to that of the soil. The soil levels of organic matter, P, pH and texture had no effect on soil As content and its bioavailability to plant. The As content in plants was always low, even in the most contaminated conditions, with three exceptions: *Mentha longifolia*, *Astragalus spp* and *Medicago sativa*. In spite of the long contamination history of the surveyed areas, there is an evident lack of effective pressure toward As tolerance by the plant species through accumulation of the element. This means that it is likely that plants play a minor role in superficial geochemical cycling of arsenic. Nevertheless, the arsenic levels above the legal limits in agricultural soils suggest that a wider survey of As contents in crops, fodders and vegetables should be carried out.

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