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Accumulation of Arsenic, Copper and Iron in Common Medicinal Plants of Murshidabad district, West Bengal, India

Soume Pyne[1]* and Subhas Chandra Santra[1]

[1]Department of Environmental Science, University of Kalyani, Nadia-741235, West Bengal

***Corresponding Author:** pyneenvs2010@gmail.com

Abstract

Medicinal plants are the most important source of life saving drugs for the majority of the world's population. Human knowledge about medicinal value of the plants date back probably for more than five thousand years. Murshidabad is one of the severe arsenic contaminated districts in West Bengal. This study evaluated the potential of 27 medicinal plants, grown & collected from the contaminated site of Murshidabad district. Plant, soil and water samples were collected and analyzed for total Arsenic (As), Copper (Cu), Iron (Fe) concentration. The plant samples were collected from side of agricultural field and roadside area. Results revealed that arsenic-contaminated irrigation water ranged from (0.20– 0.37 mg l-1) and soil (2.23–5.31 mg kg^{1}) considerably influenced in the accumulation of arsenic in vegetables, crops, rice, pulses, as well as medicinal plants in the study area. Copper and Iron which are also an essential nutrients both for plant growth and to complete plant lifecycle. Thus it reveals from the present survey that the medicinal plants, grown in the studied area are probably not safe for using as medicinal purposes and there is a probability of entry of arsenic to human through the use of medicinal plants grown in this region. To the best of our knowledge this is the initial work reporting on the useful baseline data on the heavy metal content in common medicinal plants in arsenic prone areas of rural West Bengal.

Keywords: Arsenic, copper, iron, medicinal plant, Murshidabad.

Introduction

Plants accumulate arsenic from arsenic contaminated water and soils. Besides providing nutrition, the plant material always formed some important chemical compounds, which can be used for medicinal purposes. Medicinal plants are considered to be an important source of natural products and traditionally occupy an important position in the socio cultural, spiritual and health arena of rural and tribal lives of India (Pala et al., 2010). In recent years, there has been increasing commercialization of medicinal plants. It has been reported that about 64% of the total global population remains dependent on traditional medicine and medicinal plants for provision of their health-care needs (Cotton, 1996). The use of aromatic medicinal herbs to relieve and treat many human diseases has been increased in world because of their mild features and low side effects (Abu-Darwish, 2009). There is a common concept among people that herbal medicines have no side effects and that 'being natural in origin and safe'. The assimilation of

heavy metals in plants is obvious because of widespread heavy metals in the soil due to geo-climatic conditions (Ali khan et al, 2007).

There are 9 districts out of 19 districts in West Bengal were identified as arsenic affected areas. Among those Nadia, Murshidabad, 24-Parganas are severely affected (Santra et al., 2013; Samal, 2005; Bhattacharya et al., 2009; Chakraborty, 2001 ; Roychowdhury et al., 2003; Roychowdhury et al., 2002; Alam et al., 2003). Chakraborti et al., (2009) based on their 20 years study, classified West Bengal into three zones: highly arsenic affected (9 districts mainly in eastern side of Bhagirathi River), mildly affected (5 districts in northern part) and unaffected (5 districts in western part).

Arsenic in ground water is a major health concern in Asia and the risk from using shallow tube wells for irrigation purposes as well as drinking purposes. It is well known that metals and metalloids can easily absorbed by producers and then transfer to food chain and finally bio-accumulated to a great extent in higher tropic order of ecosystem (Norra et al., 2005). Irrigation of agricultural soils with As contaminated groundwater in Bangladesh, India, China, and other countries in South-East Asia has caused the accumulation of As in food crops and enter into the food chain, which poses long term risks to human health (Williams et al., 2005, Tuli et al., 2010). According to the reported work of Mandal and Suzuki (2002); Roychowdhury et al., (2002); Liao et al., (2005); Dahal et al., (2008), Bhattacharya et al, (2005, 2010) the arsenic concentration in crop plants varied from 0.007 to about 7.50 mg kg^{-1}.

There is necessary to monitor the level of toxic metals in medicinal plants due to the increasing trend of environmental contamination and physiological properties of the plants to accumulate toxic metals (De Smet, 1992; Ali, 1983). It is necessary to improve quality standards for herbal medicines by examining and revising the maximum allowable values of heavy metals in medicinal plants, using research based on medicinal plants. Medicinal herbs can be easily contaminated with heavy metals from the environment (soil, water, or air) during growth and manufacturing processes when the ready-made products are produced (Al-Eisawi, 1982).

In West Bengal, a huge number of medicinal plants have been found to grow. There are several medicinal plants are available in our study area of Murshidabad district (Table 1). Thus the aim of this study is to investigate the arsenic, iron and copper contents in the useful part of the commonly used medicinal plants for generating a first time baseline study in West Bengal.

Materials and Methods

Study site

Four blocks of Murshidabad district (Beldanga I & Beldanga II, Domkal, Hariharpara) has been chosen for the present study (Fig. 1). According to the previous report, in all these areas the level of arsenic in groundwater exceeding WHO permissible limit for drinking water (0.01 mg l^{-1} WHO 2001) and Food and Agricultural Organization permissible limit for irrigation water (0.10 mg l^{-1}; FAO, 1985).

Sample collection

In the study area, different commonly available medicinal and aromatic plants like Akanda, Nayantara, Basak, Kalmegh, Bhui amla, Neem, Satamuli, Iswarmuli, Hinche saak, Harjora, Swet berala, Aloe vera, Aswagandha, Thankuni, Gandal, Kesut, Bel, Gulancha, Chatim, Telakachu, Apang, Arjun etc were collected for the present study (Table 1). The useful parts of these medicinal plants were collected in a substantial amount from this area in polythene bag to analyse the level of arsenic and other heavy metal content.

Groundwater samples has been collected in 100ml bottles with replica (n=3) from the shallow tube well pumps used for irrigation in the study area and preserved with 1ml/L concentrated HNO_3.

Surface soil samples were collected from 0-10 cm depth and sub-surface soil samples were collected from 10-50cm depth in a $2m^2$ area by composite sampling from the fields and transferred it to into air-tight polythene bags.

Sample Treatment

The plant samples were washed thoroughly with tap water followed by de-ionized water for several times. Finally the samples were dried in hot air oven at 50^0C-60^0C for 72h, powdered and stored in air tight polythene bags at room temperature with proper labeling.

The water samples were filtered through 0.45µ Millipore filter paper and the filtered samples will kept in polythene bottles at 4^0C prior to analysis.

The soil samples were immediately sun-dried after collection and later dried in hot air oven at 60^0C for 72h, grind and screened through 2.0mm sieve. Finally the samples were stored in airtight polythene bags at room temperature.

Sample Digestion

Plant and soil samples were digested separately following heating block digestion procedure (Das et al., 2004; Rahaman et al., 2007). 0.2-0.5gm of plant samples were taken in clean, dry digestion tubes. The plant samples were digested by adding 1.0ml perchloric acid ($HClO_4$), 1.5 ml of sulphuric acid (H_2SO_4) and 4.0ml nitric acid (HNO_3). The mixture was allowed to stand overnight under fume hood. In the following day the tubes were placed on a heating block at 110^0 -120^0c and finally get clear solution. The samples were cooled, diluted to 25ml with de-ionized water and filtered with Whatman No. 41 filter paper.

For soil sample, 3ml of concentrated H_2SO_4 was added in addition to 2ml of concentrated $HClO_4$. Then the tubes were heated at 160ºC for about 4-5h. The heating will be stopped when the dense white fume of $HClO_4$ was emitted. The content was then cooled, diluted to 25ml with de-ionized water, and filtered through Whatman No. 42 filter paper.

Sample Analysis

The total arsenic of samples was analyzed by flow injection hydride generation atomic absorption spectrophotometer (FI-HG-AAS, Perkin Elmer A Analyst 400) using external calibration **(**Welsch et al., 1990). The optimum HCl concentration was 10% v/v and 0.4% $NaBH_4$ produced the maximum sensitivity. For each sample of the digested soil, rice, pulse and vegetable, and irrigation water, three replicates were taken and the mean values were obtained on the basis of calculation of those three replicates. Standard reference materials (SRM) from National Institute of Standards and Technology (NIST), USA, were analyzed in the same procedure at the start, during, and at the end of the measurements to ensure continued accuracy.

Analytical Quality Control Data

The observed arsenic concentrations (mg/kg dry weight) of SRM from NIST, USA, were as follows: rice flour (SRM 1568A), 0.26 ± 0.09 (certified value 0.029 ± 0.03), San Joaquin soil (SRM 2709A) 16.1 ± 0.9 (certified value, 17.7 ± 0.8).

Result and Discussion

Arsenic content in soil and water

The range of arsenic content in the soil of selected study site was ranged from 2.74-5.28 (mg/kg) at Domkal block, 2.23-3.44 (mg/kg) at

Table 1. Collection of different medicinal plants and their useful properties.

Plant species	Family	Medicinal uses	Parts used
Aristolochia indica	Aristolochiaceae	Cholera, diarrhea, gastro-intestinal disorder, allergy, asthma	Root, leaf, seed & whole plant
Paederia scandens	Rubiaceae	Asthma, diarrhea, blood dysentery, night blindness, piles, blood purification	Root & leaf
Aloe vera	Liliaceae / Aloaceae	Skin disease, dysentery, diarrhea	Leaf
Tinospora cordifolia	Menispermaceae	Chronic diarrhea, chronic dysentery, chronic fever, ulcer, jaundice, anemia, skin disease	Root, shoot, leaf, fruit
Calotropis gigantea	Asclepiadaceae	Inflammation, skin infection, eczemas, leucorrhea	Leaf, fresh root, flower
Centella asiatica	Mackinlayaceae	Headache, body ache, insanity asthma,leprosy, ulcer, eczemas, wound healing, antitumor	Leaf
Withania somnifera	Solanaceae	Antifever, leucoderma, dropsi, fistula, leprosy	Root, leaf, fruit
Datura stramonium	Solanaceae	Sedative, antihairfall, anti dandruff, Parkinson's disease, breathing trouble	Leaf, fruit, seed
Aegle marmelos	Rutaceae	Brain problem, liver problem, abdominal pain, diarrhea, dysentery, renal problem	Root, leaf, fruit, flower
Sida cordifolia	Malvaceae	Urinal problem, diabetes, asthma, nerve problem, dysentery, paralysis	Root, leaf, fruit
Phyllanthus fraternus	Euphorbiaceae	Bronchitis, problem of renal tube, anemia, gonorrhea, blood dysentery	Whole plant
Justicia adhatoda	Acanthceae	Cough & cold, leucoderma, indigestion, asthma, chronic bronchitis, diarrhea	Leaf, bark, root, flower
Coccinia grandis	Cucurbitaceae	Vomiting, diabetes, gonorrhea, jaundice, cough & cold, asthma	Root, leaf, fruit
Alstonia scholaris	Apocynaceae	Beriberi, liver disorder, chest pain, leprosy, tumor, body ache	Root, bark, leaf
Eclipta prostrata	Compositae / Asteraceae	Hair treatment, ulcer, eye trouble, antiseptic, fever, skin disease, bronchitis, asthma	Leaf, root & whole plant
Andrographis paniculata	Acanthaceae	Influenza, ulcer, skin disease, liver problem, cough, dysentery	Whole plant, leaf, root
Asparagus racemosus	Liliaceae / Asparagaceae	Night blindness, blood dysentery, cough & cold, sperm disorderness	Root & leaf
Enydra fluctuens	Compositae	Chiken pox,bronchitis, liver problem, leucoderma, gonorrhea, skin disease	Shoot & leaf
Cissus quadrangularis	Vitaceae	Repairing & joining of bone, headache, scurvy, asthma	Leaf & shoot
Terminalia arjuna	Combretaceae	High blood pressure, ulcer, hernia, cough & cold, menorezia, blood dysentery	Bark, leaf, fruit
Azadirachta indica	Meliaceae	Leprosy, urinal problem, blood purifier, skin disease, eczemas, ulcer, diabetes	Root, shoot, bark, leaf, fruit, seed
Vitex negundo	Verbenaceae	Treatment & growth of hair, eye trouble, dandruff, abdominal pain, hair loss, increase of spleen, leucoderma, acne, urinal problem	Root, shoot, bark, leaf, fruit, flower seed
Achyranthes aspera	Amaranthaceae	Dropsy, abortion, allergy, rabies, gonorrhea, pneumonia, acidity	Whole plant, root
Marseilea minuta	Marsileaceae	Blood pressure, cough & cold, memory loss, eyesight problem, skin disease	Leaf and whole plant
Nyctanthes arbortristis	Verbenaceae	High fever, malaria, gastro-intestinal problem	Root, bark, leaf, seed

Table 2. Arsenic and others Heavy metals content in commonly used Medicinal plants of Murshidabad District.

Location	Parts of plants used		No. of sampl	Arsenic content in ppm (mean ± sd)	Fe content in ppm (mean ± sd)	Cu content in ppm (mean ± sd)
	Local name	Scientific name				
A*	Iswarmuli	*Aristolochia indica*	2	0.177±0.006	0.533±0.032	0.126±0.01
	Gandal	*Paederia scandens*	3	0.070±0.003	0.424±0.033	0.098±0.006
	Alovera	*Aloe vera*	2	0.143±0.011	0.387±0.035	0.073±0.006
	Gulancha	*Tinospora cordifolia*	3	0.072±0.002	0.235±0.018	0.255±0.007
	Lohachur	-	4	0.095±0.007	0.237±0.016	0.058±0.004
	Akanda	*Calotropis gigantea*	5	0.133±0.011	0.288±0.025	0.139±0.001
	Bel	*Aegle marmelos*	6	0.126±0.008	0.279±0.016	0.164±0.01
	Bhui amla	*Phyllanthus fraternus*	3	0.215±0.027	0.518±0.042	0.050±0.001
	Nayantara	*Catheranthus roseurs*	3	0.141±0.004	0.176±0.01	0.097±0.001
B	Basak	*Justicia adhatoda*	4	0.053±0.004	0.824±0.052	0.124±0.001
	Kesut	*Eclipta alba*	5	0.065±0.004	0.217±0.014	0.304±0.023
	Kalmegh	*Andrographis paniculata*	3	0.115±0.016	0.456±0.034	0.118±0.01
	Satamuli	*Asparagus racemosus*	5	0.093±0.003	0.237±0.019	0.037±0.003
C	Bhui amla	*Phyllanthus fraternus*	3	0.165±0.011	0.467±0.009	0.055±0.004
	Neem	*Azadirachta indica*	3	0.073±0.005	0.288±0.015	0.159±0.009
	Kanak dhutra	-	3	0.095±0.005	0.296±0.018	0.079±0.006
	Chatim	*Alstonia scholaris*	5	0.027±0.007	0.195±0.013	0.199±0.016
	Harjora	*Cissus quadrangularis*	5	0.221±0.016	0.484±0.035	0.047±0.004
	Telakachu	*Coccinia grandis* -	3	0.073±0.005	0.235±0.018	0.223±0.017
D	Nishenda	*Vitex negundo*	2	0.134±0.004	0.256±0.023	0.097±0.006
	Arjun	*Terminalia arjuna*	5	0.166±0.011	0.392±0.03	0.035±0.002
	Apang	*Achyranthes aspera*	6	0.099±0.012	0.565±0.04	0.183±0.018
	Telakachu	*Coccinia grandis*	3	0.107±0.013	0.204±0.017	0.182±0.002
	Swet berala	*Sida cordifolia*	3	0.152±0.013	0.351±0.026	0.052±0.004
	Susni saak	*Marseilea minuta*	4	0.243±0.008	1.352±0.116	0.185±0.016
	Akanda	*Calotropis gigantea*	5	0.043±0.007	0.486±0.035	0.118±0.008
E	Gandal	*Paederia scandens*	4	0.298±0.021	0.585±0.021	0.138±0.001
	Siuli	*Nyctanthes arbortristis*	5	0.034±0.008	1.41±0.014	0.175±0.001
	Hinche saak	*Enydra fluctuens*	3	0.078±0.001	1.476±0.107	0.106±0.008
	White dhutra	*Datura stramonium*	4	0.064±000	0.177±0.016	0.162±0.016
	Aswagandha	*Withania somnifera*	5	0.043±0.004	0.222±0.018	0.182±0.012
	Thankuni	*Centella asiatica*	6	0.130±0.013	0.465±0.03	0.181±0.018
***A = Beldanga 2; B = Beldanga 1; C = Domkal ; D = Doulatabad; E = Hariharpara**						

Table 3. Correlation between different metals in common medicinal plants.

	As	Fe	Cu
As	1	-	-
Fe	0.136	1	-
Cu	-0.374*	-0.015	1
*correlation is significant at the 0.05 level (two tailed)			

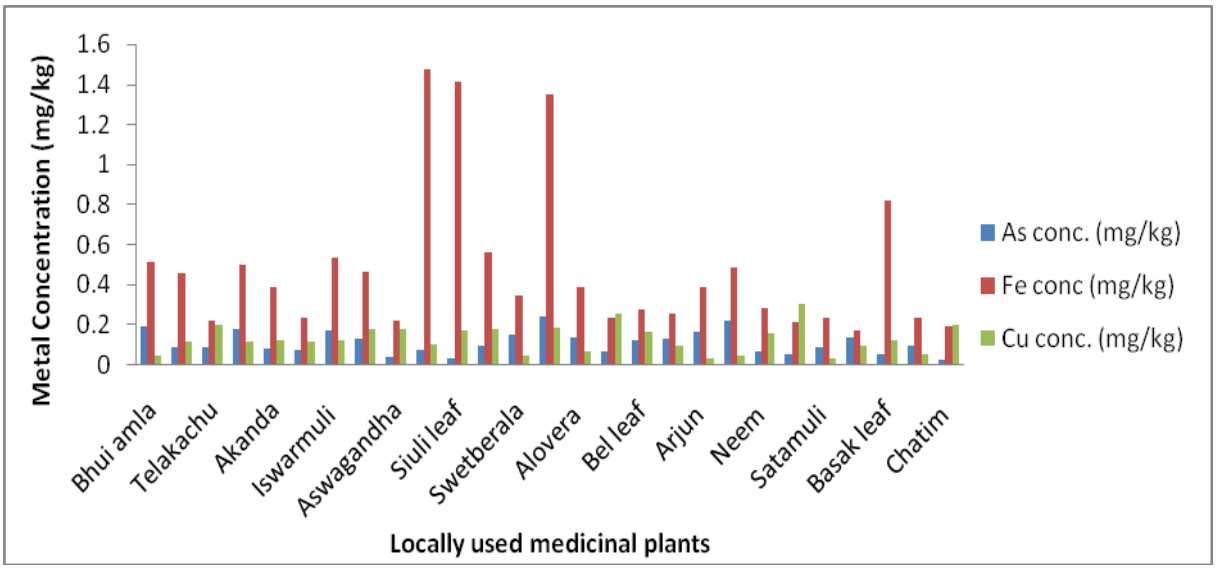

Figure 1. Distribution of arsenic, iron, copper in common medicinal plants.

Hariharpara block and 3.37-5.31 (mg/kg) at Beldanga block. The total arsenic content in water sample was ranged from 0.23-0.34 (mg/l), 0.20-0.33 (mg/lt), 0.22-0.37 (mg/lt) respectively. But, the accumulation of arsenic in soil in the study area was lower than the reported global average of 10.0 mg kg-1 and was much below the maximum acceptable limit for agricultural soil of 20.0 mg kg-1 as recommended by the European Community (Das et al., 2002). collectively which is higher than WHO (WHO, 1993) permissible limit 0.01 mg $lt^{-1}$for drinking water (WHO 1993; Rahaman et al., 2007) and FAO permissible limit for irrigation water (0.10 mg l-1; FAO 1985) (JECFA 1993).

Arsenic, Fe and Cu content in common medicinal plants

The commonly available medicinal plants with their medicinal use of the study area are given in table 1. There are about 27 species of different medicinal plants collected for the study. The total arsenic content in the commonly used medicinal plant, growing in the arsenic affected Murshidabad district, West Bengal were shown in Table 2. From this study results showed the highest and lowest mean arsenic concentration was found in Gandal shoot (*Paederia scandens)*, 0.298 mg/kg and Chatim (*Alstonea scholaris*) 0.027 mg/kg respectively (Figure 1).

Medicinal plants were also processed and analyzed for the determination of Fe and Cu (table 2). It has been seen that iron uptake is

much higher than arsenic in this site and it is ranged from 0.176 to 1.47 mg/kg, in Nayantara and Hinche saak respectively. Whereas, copper accumulation is much lower than iron and which is ranged from 0.035 to 0.255 mg/kg, in Arjun and Gulancha respectively. A site specific response was also observed in uptake of these elements. From our study the order of accumulation of arsenic was found to follow the ascending order Chatim leaf < Siuli leaf < Aswagandha root < Basak leaf < White dhutra < Kesut leaf < Neem leaf < Gulancha shoot< Telakachu leaf< Hinche saak < Satamuli shoot<Kanak dhutra, Lohachur < Apang shoot< Telakachu leaf< Kalmegh shoot < Bel leaf < Thankuni shoot < Akanda shoot < Nishenda < Nayantara < Aloe vera leaf < Swet berala root < Bhui amla < Arjun chal < Iswarmuli shoot < Bhui amla < Harjora shoot < Susni saak < Gandal shoot.

Correlation between arsenic, iron and copper in common medicinal plants

To investigate the correlation between the heavy metals in medicinal plant body, it was found that As & Fe content is positively correlated but statistically not significant. Whereas it is found that As & Cu is correlated negatively at 0.05 level of significance (Table 3). The overall results indicated that significant As accumulation was found in all medicinal plants. Soil and water of this area are enriched with arsenic, showed arsenic level beyond the WHO permissible limit, which cause a significant accumulation in medicinal plants. From this study it can be concluded that, chronic intakes of As in medicinal plants may impart health on human beings and other animals. From the results it is observe that As & Fe content of these medicinal plants is positively correlated where the *p* value is not significant. Whereas As & Cu content of these medicinal plants is negatively correlate & the *p* value is significant. Thus it reveals from the present study that the medicinal plants, grown in the studied area are probably not safe for using as medicinal purposes.

Conflict of interest:

Authors declare that there is no conflict of interest.

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