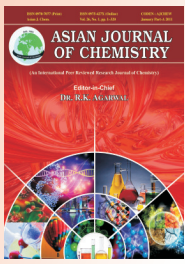
Asian Journal of Chemistry; Vol. 26, No. 20 (2014), 6761-6768

ASIAN JOURNAL OF CHEMISTRY

http://dx.doi.org/10.14233/ajchem.2014.16727



Distribution of Arsenic Forms in Decomposed Solidwaste

SHAH RUKH[1,*], MOHAMMAD SALEEM AKHTAR[1], MEHRUINSA MEMON[2], AYAZ MEHMOOD[1] and MUHAMMAD IMRAN[1]

[1]PMAS Arid Agriculture University, Rawalpindi, Pakistan
[2]Sindh Agriculture University, Tando Jam, Pakistan

*Corresponding author: Email: shahrukhshah86@hotmail.com

Received: 20 November 2013; *Accepted*: 25 February 2014; *Published online*: 25 September 2014; AJC-16006

Arsenic forms distribution helps to understand its chemical processes and bioavailability. Total and distribution of arsenic fractions in decomposed solidwastes measured by an atomic absorption spectrophotometer with flame heated quartz adsorption cell coupled to a hydride generation device, are reported. The system had detection limit 2.42 µg L^{-1} for total arsenic. The decomposed solidwaste was collected from historic dumpsites of three cities and analyzed for total by digestion in three acids and for the arsenic forms using sequential Wenzel *et al.* extraction scheme. Lower than reported for contaminated soil and solidwaste, total arsenic in the solid waste ranged 2.5 to 7 mg kg^{-1} which occurred dominantly as specifically-sorbed fraction (675 to 3070 µg kg^{-1}) positively correlated with extractable iron oxides and dissolved organic carbon. Overall the arsenic forms followed the order amorphous and poorly-crystalline metal oxides sorbed arsenic > residual phases > well crystalline metal oxides sorbed arsenic > non-specifically sorbed arsenic fraction. This study concludes that arsenic primarily occurred as specifically sorbed fraction to iron oxides and correlation with dithionite extractable iron implied that a reducing condition (*eg*., prolonged submergence) may release arsenic to the environment.

Keywords: Arsenic, Arsenic forms, Sequential extraction, Decomposed solid waste.

INTRODUCTION

Solidwaste disposal in open landfills and wastewater irrigation in the country may cause environmental contamination and risk human health. Several anthropogenic sources contribute arsenic to solidwaste which is redistributed during decomposition and mineralization[1]. Mineralization of organic matter and cyclic oxidation-reduction release arsenic in soluble form into the soil immediately after addition[2]. Distribution of arsenic fractions also changes during decomposition of organic waste. Exchangeable and reducible/oxidizable fractions increase as the mineralized arsenic is partly re-adsorbed on soil particles and the residual fractions decrease[3-6]. Solidwaste dumpsite soils differ in distribution of arsenic forms under dry and rainy season[7]. The crystalline iron oxyhydroxides' associated arsenic becomes the largest forms in arable soils. The residual arsenic form dominates in non-soil material *e.g.*, fresh and combusted coal waste or even industrial contaminated non-field soils. Once mineralized or decomposed further transformation of arsenic(V) to arsenic(III) is mediated by bacterial activity and later methylated to less toxic organic species *i.e.* monomethylarsonic acid (MMA) and dimethylarsenic acid (DMA)[8-11]. In contaminated soils, arsenic(V) species are predominant over arsenic(III) and organic bound arsenic may be upto 40 % of total arsenic content[7]. Level and forms of arsenic needs thorough assessment to determine contamination of soil and water through emanating from solidwaste[2,3].

The sequential extraction schemes are well-accepted methods to assess the trace element pools for relative liability in sediment, soils and decomposed solidwaste[13,14]. Quantitative measurement of various arsenic forms including non-specifically surface sorbed, specifically-sorbed, amorphous and poorly-crystalline metals oxide sorbed, well-crystalline iron and aluminum oxides sorbed and the residual arsenic phases are analyzed by using Wenzel *et al.*[15] sequential extraction scheme.

Distribution of arsenic forms in the decomposed solidwaste in Pakistan is not reported previously. Total arsenic and distribution of its forms in historic solidwaste dumping sites from three cities are presented. The correct estimation of arsenic forms in solidwaste will help in identifying the bioavailabe forms, potential for movement to underground water and explaining geochemical behavior and extent of anthropogenic contribution from sampled areas[13].

EXPERIMENTAL

Site description and sampling: Decomposed solidwaste samples were taken from historic dumpsites of Faisalabad, Lahore and Islamabad cities of Pakistan occurring in semi arid to sub humid climate zone. Islamabad occurs in subhumid subtropical climate with hot summers and cold winters with

an average rainfall of 1150 mm per annum and means annual temperature range -3.9 to 46.6 °C. The H-10 sector of Islamabad has a dumpsite where solidwaste was dumped in open 30 to 25 years before sampling. Lahore occurs in a hot semi-arid climate with an average annual rainfall of 490 mm with mean annual temperature ranges from -2.2 to 48.3 °C. Seven hundred tons of solidwaste per day is dumped for the last 15 years. Faisalabad occurs in an arid climate with average annual rainfall of 300 mm and mean annual temperature range -1 to 48 °C. As arsenic one of open dumpsite, the City dumped solidwaste along Pharang drain during past 20 years. The City also manages sewerage water treatment ponds at Chowkera village where nearby crop land is being irrigated with untreated wastewater. Fresh pits were dug and sampled from each location at the depths where variation in colour and texture was observed. The three pits from Islamabad had two to five horizons: 0-10, 10-21, 21-44, 44-69 and 69$^+$ cm (pit-1), 0-17 and 17-35 cm (pit-2) and 0-15, 15-35 and 35-50$^+$ cm (pit-3). The Lahore pit had three horizons at 0-20, 20-40 and 40$^+$ cm (pit-4) and the Faisalabad pit had two horizons at 0-15 and 15-35 (pit-5) cm depth. Samples from soil under wastewater irrigation were taken at 0 to 15 and 15 to 35 cm depth from crop land under wastewater irrigation. All the samples were air-dried, ground to pass through a 2 mm sieve and stored in refrigerator.

Solidwaste and soil characterization: All samples were analyzed for pH in 1:5 solid-water suspension Cole Parmer (Model-6071N) pH meter[35], organic matter by wet digestion in 1N potassium dichromate ($K_2Cr_2O_7$) titrated against 0.5 N $Fe(NH_4)_2(SO_4)_2$ solution[36], dissolved organic carbon by extraction with 0.5 M K_2SO_4[37] and for extractable iron and aluminum by citrate bicarbonate dithionate (CBD)[38].

Total arsenic assay: For total arsenic, 1 g sample was digested in HNO_3-H_2SO_4-$HClO_4$ acid mixture of 6:3:1 ratio[34]. The digestion initiated by first keeping the mixture overnight at room temperature and then further carried out by raising the temperature to 50, 100 and 150 °C at one hourly interval and to 200 °C for 6 h in a digestion block (RKC T400) under a fume hood. Total arsenic was measured by atomic absorption spectrophotometer equipped with a hydride generation assembly[39]. Arsines (AsH_3) produced using 0.4 % $NaBH_4$ (Alfa Aesar) in 0.5 % NaOH (Riedel-de Haen) with 5 M HCl (Riedel-de Haen) solution[40] pumped through the reaction coil at 1 mL min^{-1} was to the flame by N_2 gas at pressure rate of 0.32 MPa. The measuring conditions were optimized for maximum absorption signal. Use of high purity acetylene (99.9 %) yielded standard curve having r^2 value 0.998. The detection limit (mean of 10 blanks+ 3 σ) was 2.42 µg L^{-1}.

Arsenic fractionation: One gram sample was placed into 50 mL centrifugation tube and sequentially partitioned into various fractions *viz* non-specifically surface sorbed arsenic, specifically-sorbed arsenic, amorphous and poorly-crystalline metal oxides sorbed arsenic, well-crystalline metal oxides sorbed arsenic and the residual arsenic phases using Wenzel sequential extraction scheme[15] (Table-1). In all steps, shaking of solidwaste samples was carried out on an end to end shaker at 40 rpm. The centrifugation following each shaking was done at 2000 rpm for 15 min using Hermile Model-Z513 K centrifuge machine. Arsenic concentration in supernatant was measured using Hydride Vapor Generation (HVG-1, Shimadzu) assembly coupled with Atomic Absorption Spectrometer (AA-6300, Shimadzu) as described above. The stock solutions were kept in refrigerator at 4 °C to prevent any change in speciation. Working standards were prepared daily from the stock solutions in distilled deionized water. The sequential extraction scheme adopted provides reproducible results[5,12].

RESULTS AND DISCUSSION

Characteristics of decomposed solidwaste: The collected solidwaste dumpsite samples had pH 6.8 to 7.7, organic matter content from 2 to 21 %, CBD-extractable iron between 0.3 and 0.9 g kg^{-1}, aluminum between 0.1 and 1 g kg^{-1}, dissolved organic carbon (DOC) between 23 and 390 mg kg^{-1} and arsenic between 2.3 and 7.5 mg kg^{-1} (Table-2). Most samples had near neutral pH range. The pH in 1:2.5 solid water suspensions slightly decreased with depth in Islamabad dumpsite pit 1. Organic matter content increased an order of magnitude with depth which is in line with the work reported by Pietri and Brooke[16]. In few pits higher organic matter at the surface than the subsurface indicates surface sedimentation of particulate organic matter[17]. Most of solidwaste surface samples had higher DOC than the subsurface sample the same way as organic matter. Most samples had CBD-extractable iron

TABLE-1
SEQUENTIAL EXTRACTION PROCEDURE ADOPTED FOR FRACTIONATION OF ARSENIC IN THE SOLIDWASTE SAMPLES

Fraction	Arsenic form$	Procedure
As_{NES}	Non-specifically sorbed on surface of soil colloids, is completely exchangeable	1 g soil with 25 mL 0.05 M$(NH_4)_2SO_4$ was shaken for 4 h at 20° C then centrifuged, and decanted supernatant was filtered through 0.45 µm membrane and analyzed for arsenic.
As_{ES}	Inner-sphere surface complexes	25 mL of 0.05 M $(NH_4)H_2PO_4$ was added to the residue, the suspension was shaken for 16 h at 20° C then centrifuged, and decanted supernatant was filtered through 0.45 µm membrane and analyzed for arsenic.
As_{AM}	Amorphous colloids and poorly crystalline iron and aluminum oxides sorbed	25 mL of 0.2 M NH_4-oxalate buffer (pH 3.25) was added to the residues and shaken for 4 h in the dark at 20° C then centrifuged, and decanted supernatant was filtered through 0.45 µm membrane and analyzed for arsenic.
As_{CR}	Crystalline iron and aluminum oxides sorbed	The residues were washed with 0.2 M NH_4-oxalate solution, kept in a water bath at 96 ± 3° C for 0.5 h after adding NH_4 oxalate buffer (0.2 M) + 0.1 M ascorbic acid (pH 3.25), entrifuged, decanted and supernatant was filtered through 0.45 µm membrane and analyzed for arsenic.
As_{RS}	Present in primary minerals *e.g.*, arseno-pyrite, and secondary phases *e.g.*, hydrous secondary arsenates	The residues were washed with 0.2 NH_4-oxalate (pH 3.25) and microwave digestion was carried out after adding HNO_3/H_2O_2, centrifuged, decanted and supernatant was filtered through 0.45 µm membrane, diluted up to 50 mL and analyzed for arsenic.

$ These forms are operationally defined arsenic pools and do not represent exact phases

content close to 0.5 g kg^{-1}, with higher content in the subsurface samples than surface samples indicating iron oxides leaching and accumulation. CBD-extractable aluminum content in most samples was in the range of 0.8 to 1 g kg^{-1}.

Total arsenic content: Total arsenic concentration varied between 2.5 to 7 mg kg^{-1} in most solidwaste and soil samples. The pits 1 and 2 at Islamabad dumpsite had generally greater arsenic content than Lahore and Faisalabad dumpsites, especially for the surface samples (Fig. 1) which may be due to greater mineralization under relatively wet climate at Islamabad. The soil under wastewater irrigation at Faisalabad had total arsenic content slightly increased with depth indicating subsurface accumulation. Arsenic may have leached with irrigation and accumulated below the surface as shown by several authors[18,19]. Total arsenic concentrations were lower than those previously published values[15]; and values greater than 1000 mg kg^{-1} have been reported frequently for contaminated sites[13,20,21]. None of the solidwaste had total arsenic greater than 50 mg kg^{-1} as the solidwastes examined had household origin. Total arsenic correlated with CBD-extractable iron (Fig. 2a) and aluminum (Fig. 2b) indicating contribution of the oxides' surface adsorbed arsenic to total arsenic[22]. The pH range (Table-2) excludes the possibility of occurrence of crystalline aluminum oxides in the system. The extracted aluminum may have been contributed by exposed edges of organic matter[23]. Total arsenic correlated with DOC (Fig. 2c), however, the surface samples and the samples from deeper depths created a spread lowering correlation coefficient. Organic matter mobilizes arsenic oxyanions forming organo-arsenic complexes[24,25]. Large concentrations of DOC in solidwaste may enhance arsenic mobility by displacing arsenic sorbed to iron oxide surfaces[26]. Dissolved organic carbon reduces arsenic adsorption onto soil surfaces by preferentially surface-adsorption competing with arsenic or by forming soluble organometallic complexes and hence increasing its mobility[22].

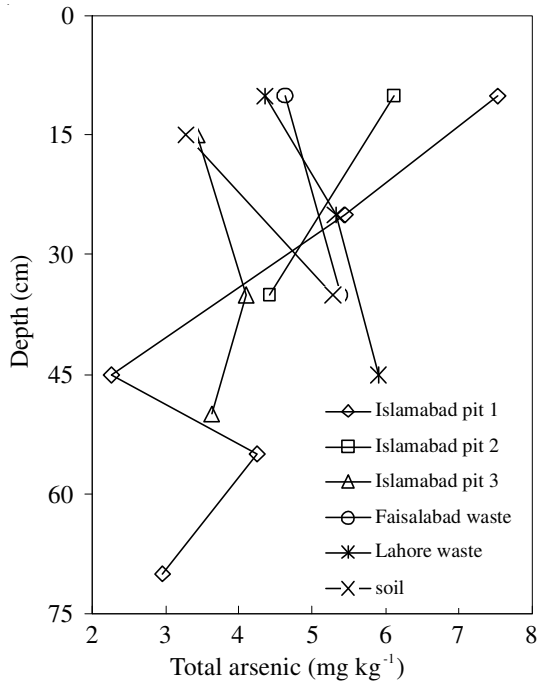


Fig. 1. Total arsenic in the solidwastes and the soil under irrigation with wastewater determined by digestion with HNO_3-H_2SO_4-$HClO_4$ acid mixture

Distribution of arsenic forms

Easily desorbable arsenic: Non-specifically surface adsorbed arsenic fraction (As_{NES}) extracted with $(NH_4)_2SO_4$ ranged from 160 to 340 µg kg^{-1} in the solidwastes (Fig. 3a) and contributed 2 to 10 % to the total arsenic content (Table-3). This fraction of arsenic is loosely attached to soil colloids, strongly correlates with soil solution arsenic and is completely labile[7]. It occurs normally as a smaller quantity than any other fraction[7,15]. In all the solidwaste profiles, it remained unchanged with depth (Fig. 3a). Previous studies by Gao *et al.*[4] and Fedotov *et al.*[3] reported non-specifically surface sorbed arsenic determined by employing the same extraction procedure to range 1 to 5 % of total arsenic. Poultry litter contains almost 50 % non-specifically sorbed arsenic[27]. Islamabad (130 to 339 µg kg^{-1}) and Lahore (198 to 258 µg kg^{-1}) dumpsites had greater arsenic-sorbed on non-specific sites compared to Faisalabad dumpsite (196 to 229 µg kg^{-1}) suggesting lower release due to relatively dryer climate. Non-specifically adsorbed arsenic in decomposed waste pit increased with depth suggesting potential for further leaching.

The specifically adsorbed arsenic (As_{ES}) extracted with $(NH_4)HPO_4$ ranged 675 to 3070 µg kg^{-1} in the solidwaste profiles with a little evidence for depth related trend (Fig. 3b). This fraction contributed 30 to 60 % to the total arsenic content in the solidewastes, close the values reported for solidwaste dumpsite soils[7]. Using the same extraction procedure, this fraction representing 10 %[15] and less than 4 %[12] of total arsenic in soils and up to 11 % in sludge samples[4] has been reported which is much smaller contribution than in decomposed

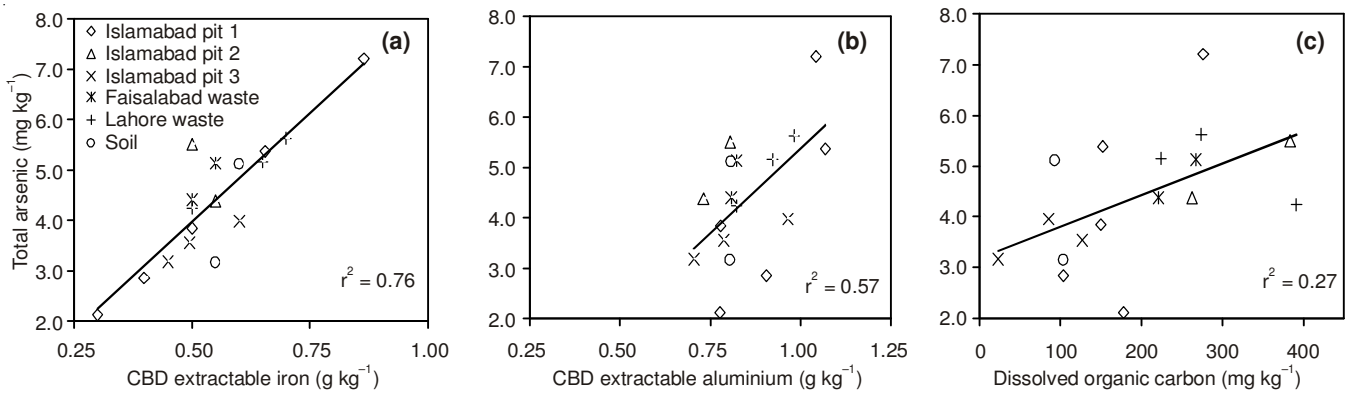


Fig. 2. Correlation between total arsenic and citrate bicarbonate dithionate extractable iron (a), aluminum (b) and dissolved organic carbon (c)

TABLE-2
CHARACTERISTICS OF SOLIDWASTE SAMPLES

Location	Depth (cm)	pH	OM (g/kg^{1})	Fe_d (g/kg^{1})	Al_d (g/kg^{1})	DOC (g/kg^{1})	$As_{(T)}$ (mg kg^{-1})
Islamabad pit 1	0-10	7.08	25	0.87	1.05	280	7.50
	10-21	7.4	16	0.66	1.07	150	5.45
	21-44	6.84	175	0.29	0.80	180	2.30
	44-69	6.54	210	0.52	0.80	150	4.25
	69^{+}	7.25	5	0.36	0.90	103	3.40
Islamabad pit 2	0-17	7.30	195	0.52	0.80	385	6.10
	17-35^{+}	7.7	80	0.53	0.70	260	4.40
Islamabad pit 3	0-15	7.11	70	0.46	0.70	23	3.40
	15-35	7.53	75	0.58	1.00	86	4.10
	35-50^{+}	7.25	70	0.50	0.80	126	3.60
Faisalabad (pit 4)	0-20	7.22	26	0.47	0.80	220	4.70
	20-35	7.30	40	0.56	0.80	270	5.40
Lahore (pit 5)	0-20	7.35	70	0.51	0.80	390	4.35
	20-40	7.5	47	0.66	0.90	225	5.30
	40^{+}	7.7	28	0.71	1.00	275	5.90
Soil (ww-irrigated)	0-15	7.89	15	0.55	0.81	103	3.30
	15^{+}	8.23	10	0.57	0.81	93	5.25

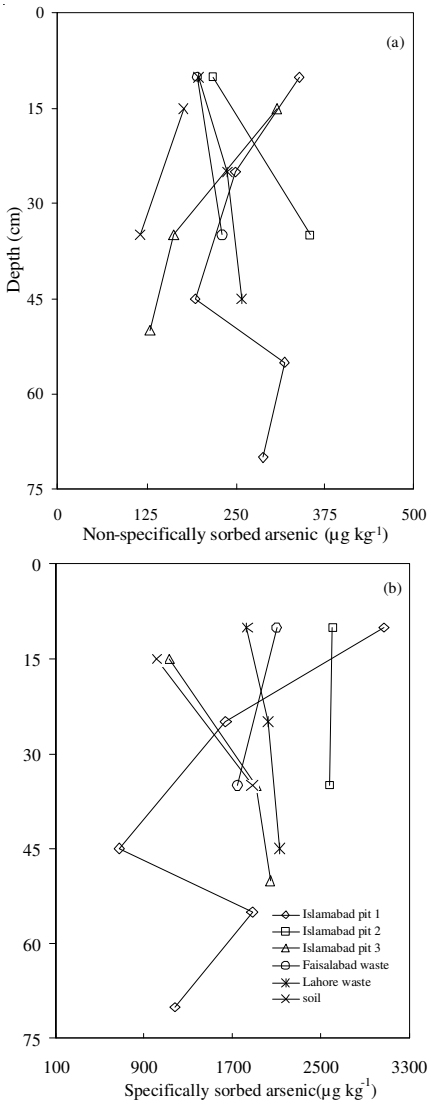


Fig. 3. Distribution of easily desorbable arsenic: (a) Non-specifically sorbed arsenic extracted by $(NH_4)_2SO_4$ shows similar behavior in all solidwaste sites (b) Specifically sorbed arsenic in more in surface sample of Islamabad pit 1, 2 and Faisalabad waste whereas in Islamabad pit 3, Lahore waste and soil under wastewater irrigation had higher in subsurface samples

solidewaste of this study. Specifically-sorbed exhibited moderate correlation with dithionite extractable iron (r^2 0.67) and with DOC (r^2 0.57) indicating their contribution as adsorbents (Fig. 4). Hydrous oxides of iron are the principle sorbents for several oxyanions and the role organic molecules for affecting arsenic adsorption on iron oxides differ with the nature of arsenic anions[22]. Arsenic forms inner-sphere complexation, involves in the formation of a coordinative complex with the mineral surface and result in exchangeable adsorption[28] however the arsenic form may be potentially mobilized due to change in pH or phosphorus addition, therefore, constitutes a risk to environmental health[12].

Metal oxides associated arsenic: Arsenic adsorbed on amorphous and poorly-crystalline metal oxides (As_{AM}) (NH_4-oxalate extracted fraction) ranged between 500 and 1250 µg kg^{-1} and was greater in the Islamabad and Lahore solidwastes compared to the Faisalabad solidwaste. The fraction contributed 11 to 35 % of the total arsenic (Table-3) which is similar to the range reported by Wenzel *et al.*[15] but much wider than the range of 24 to 30 reported for acid soils[12]. A decrease with sampling depth (Fig. 5a) was visible at least in the two profiles. Although amorphous and poorly crystalline iron and aluminum oxides have great binding capacity but the As_{AM} poorly correlated with the extractable metal oxides content (data not present). Similar poor correlation has been previous reported for the soils[12]. Probably presence of dissolved organic molecules in the same medium created some level of complexity.

Well-crystalline iron and aluminum oxides adsorbed arsenic (AS_{CR}) was in the range of 100 to 800 µg kg^{-1} in the solidwastes with a little change with depth (Fig. 5b), the fraction contributed 4 to 21 % which is similar to the values reported for a range of soils[4]. Arsenic associated with this fraction was less than the arsenic associated with the amorphous and poorly crystalline iron and aluminum oxides (Table-3). This result was in line with Wenzel *et al.*[15] ascribed to the greater surface area in the latter case. However, Novoa-Munoz *et al.*[12] reported greater contribution of As_{CR} than As_{AM} for the acid soils. Hartley *et al.*[29] shows 40 % arsenic was associated fraction with the iron and aluminum. The arsenic fraction had

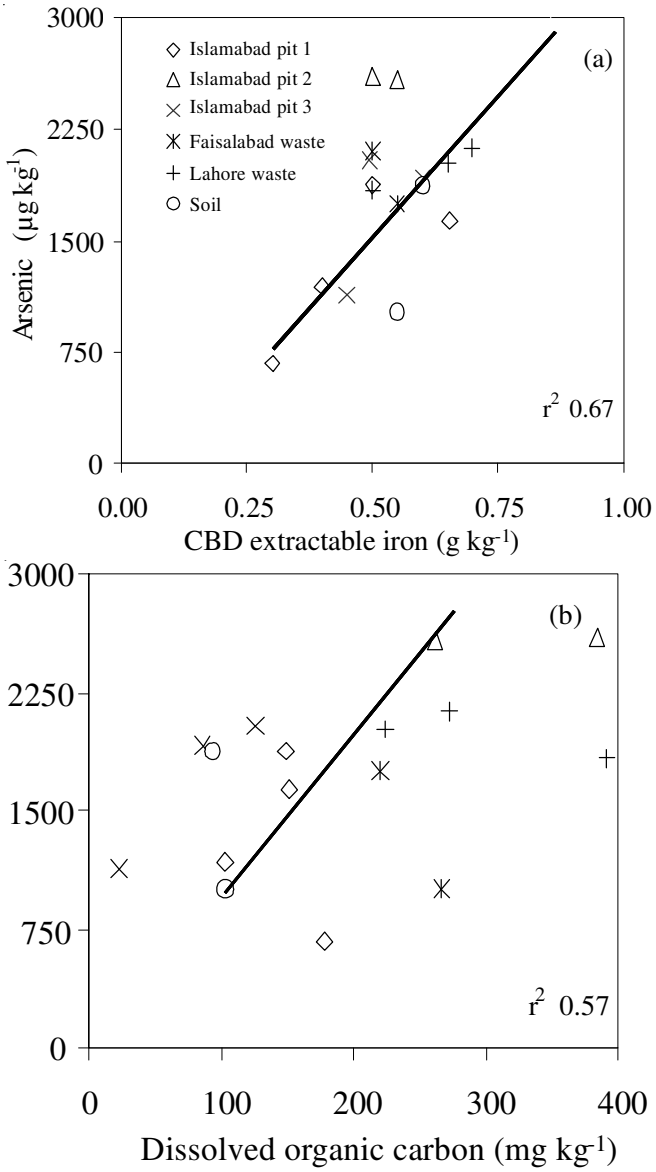


Fig. 4. Depicts strong relationship between specifically sorbed arsenic with citrate bicarbonate dithionate extractable iron (a) and positive correlation with dissolved organic carbon (b)

strong relationship with extractable iron (r^2 0.80) and relative weak relation with extractable aluminum (Fig. 6). Presence of dissolve organic carbon results in lower surface coverage of weaker bond strength between arsenic oxyanions and the metal oxide surface[22]. Also, crystalline iron oxides differ in arsenic adsorption and adsorption kinetics[30]. Iron oxides play key role in arsenic geochemistry[31] which makes it necessary to consider this fractions when arsenic is more tightly retained. Iron and aluminum oxides facilitated arsenic oxyanions' chemisorption is believed to be common mechanism for arsenic retention in soils and sediments[32].

Residual arsenic and extraction efficiency: Arsenic in the residue (As_{RS}) remaining after the surface adsorbed and metal oxide bound extraction is considered tightly held. It is co-precipitated with refractory minerals and hence remains un-extracted during the previous procedures[33]. In the decomposed solidwastes As_{RS} ranged from 400 to 1600 µg kg^{-1}. The surface samples of solidwaste from Islamabad had higher AS_{RS} while at Faisalabad and Lahore the subsurface had high values associated with total arsenic (Fig. 7a). The fraction contributed 8 to 32 % to the total arsenic content. The residual fraction in agriculture soil and sludge may range 20 to 50 % of the total arsenic content[12,3] and up to 85 % in combusted coal waste[4]. Conforming with previous studies[12,15], the residual fraction had significant positive relation with Na-dithionite-citrate extractable iron and aluminum (Fig. 8). The samples from Lahore and Faisalabad sites lied above the correlation line indicating contribution from phases other than iron oxides.

The efficiency of the sequential extraction procedure was phenomenal as indicated by r^2 0.99 when sum of five fractions was plotted against total arsenic independently measured in a three acids mixture digestion (Fig. 7b). Three acids mixture (HNO_3-H_2SO_4-$HClO_4$) of 6:3:1 ratio is more effective than the single acid digestion[34]. Wenzel *et al.*[15] used single acid digestion, reported the recovery of sequential extraction 12 % below the total arsenic and ascribed the difference to some artifacts in centrifugation and filtration. The recovery of sequential extraction in the study remained between 1 to 10 % below the total arsenic independent of arsenic content in the material.

TABLE-3
RELATIVE CONTRIBUTION OF THE EXTRACTED Arsenic FORMS TO THE TOTAL Arsenic

Samples	Depth	Arsenic (NEA)	Arsenic (EA)	Arsenic (AM)	Arsenic (CR)	Arsenic (RD)
Islamabad	(cm)			(%)		
Pit 1	0-10	4.71	42.67	15.81	12.21	24.59
	10-21	4.65	30.39	23.20	16.73	25.03
	21-44	9.13	32.03	34.80	4.88	19.15
	44-69	8.29	48.79	14.75	2.71	25.46
	69+	10.11	41.45	18.05	6.27	24.12
Pit 2	0-17	3.94	47.24	10.67	9.14	29.01
	17-35+	8.10	59.03	17.05	8.32	7.51
Pit 3	0-15	9.71	35.65	37.71	4.06	12.86
	05-35	4.08	48.14	14.75	10.29	22.75
	35-50+	3.66	57.60	11.48	11.19	16.06
Faisalabad (Pit 4)	0-20	4.45	47.85	15.55	10.41	21.74
	20-35	4.48	34.12	14.89	15.48	31.03
Lahore (Pit 5)	0-20	4.66	43.21	18.42	13.75	19.95
	20-40	4.62	39.20	25.28	16.72	14.18
	40+	4.59	37.84	13.37	15.14	29.06
Soil (ww-Irrigated)	0-15	5.58	32.07	20.41	20.45	21.49
	15-35	2.27	36.69	17.61	11.34	32.09

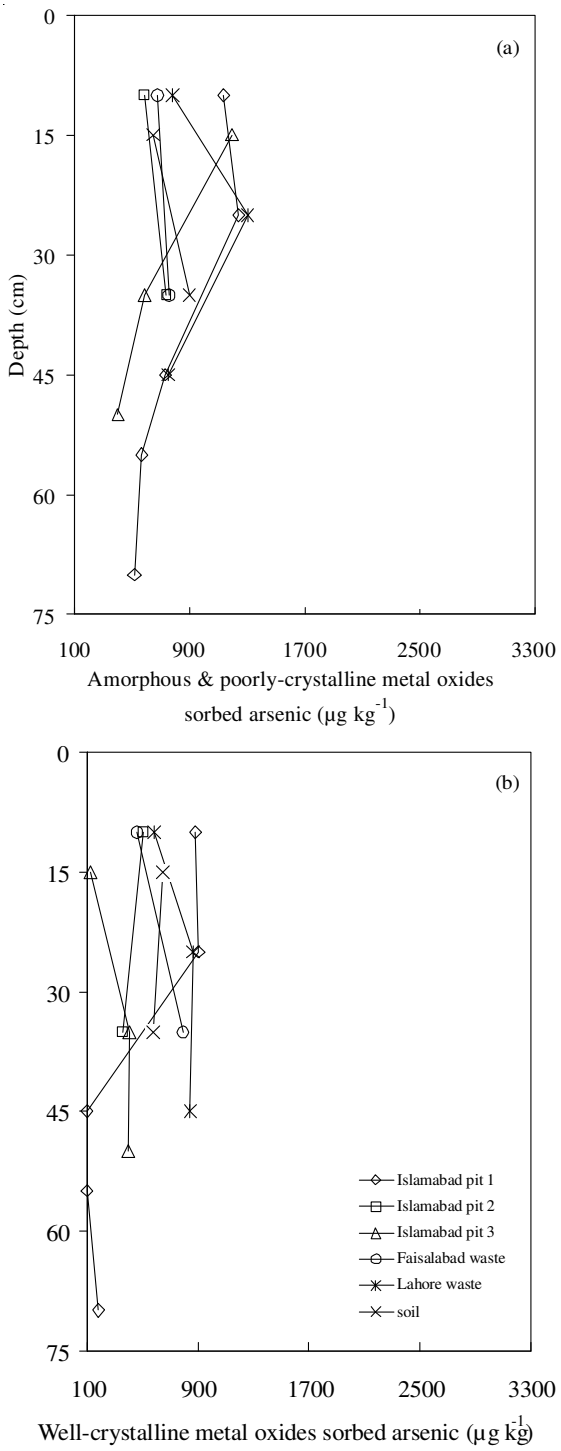


Fig. 5. Distribution of arsenic bound to iron and aluminum oxides (a) Amorphous and poorly-crystalline metal oxide arsenic extracted by NH_4-oxalate buffer in most solidwaste samples decreases with depth and in soil under wastewater irrigation increases with depth (b) Well-crystalline iron and aluminum oxides sorbed arsenic extracted by NH_4-oxalate buffer + ascorbic acid decreases with depth in Islamabad pit 1, 2 and soil under wastewater irrigation whereas increases in Islamabad pit 3, Lahore solidwaste and Faisalabad solidwaste

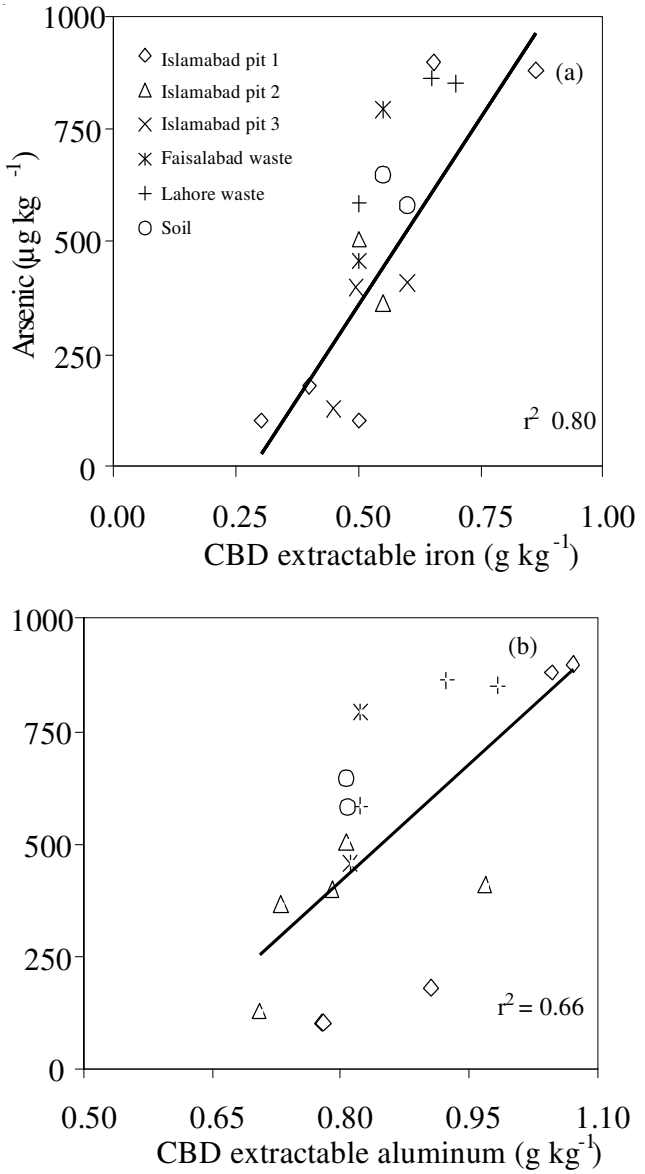


Fig. 6. Depicts strong relation between arsenic sorbed on crystalline iron and aluminum oxides with citrate bicarbonate dithionate extractable metals: (a) iron and (b) aluminum

Conclusion

Total arsenic content and distribution of forms help understand lability, potential for movement to underground water and toxicity. Arsenic in decomposed solidwaste was fractionated using a sequential extraction and measuring concentration with an atomic absorption equipped with hydride generation system. Within the inherent limitations of chemical fractionation, the reagents of the adopted sequential extraction procedure are fairly specific and selective for the target forms of arsenic. The study concludes that arsenic content in the decomposed solidwastes is below the values reported for contaminated soil and sediments. Arsenic in the solidwaste existed primarily as exchangeable and metal oxides bound forms which may constitutes a risk to environmental. Difference in relative distribution of arsenic forms may relate to rainfall. At Isalamabad greater mineralization resulted in larger labile form and at Faisalabad and Lahore dry environment caused built up of recalcitrant arsenic. Arsenic fractions existed in the decreasing order as amorphous and poorly-crystalline metal oxides sorbed arsenic, residual phases, well crystalline metal oxides sorbed arsenic and non-specifically sorbed arsenic fractions although none of the solidwaste and soil had total arsenic greater than 50 mg kg^{-1}.

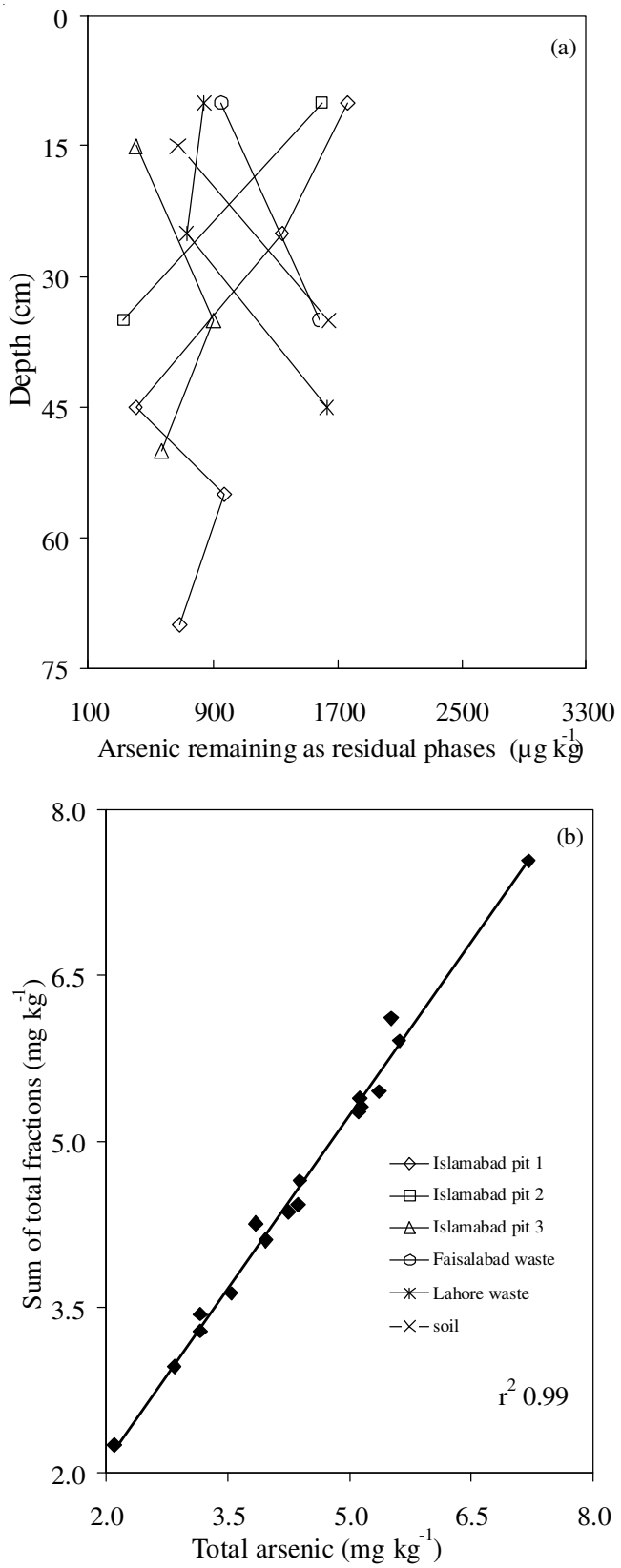


Fig. 7. (a) Arsenic remained in residual phases extracted by HNO_3/H_2O_2 in Islamabad pit 1 and 2 decreases with depth whereas in Islamabad pit 3, Lahore solidwaste, Faisalabad solidwaste and soil under wastewater irrigation increases with depth (b) Depicts the accuracy of adopted sequential extraction procedure by comparing the sum of the five fractions against the total arsenic concentrations independently measured in a single acid digest

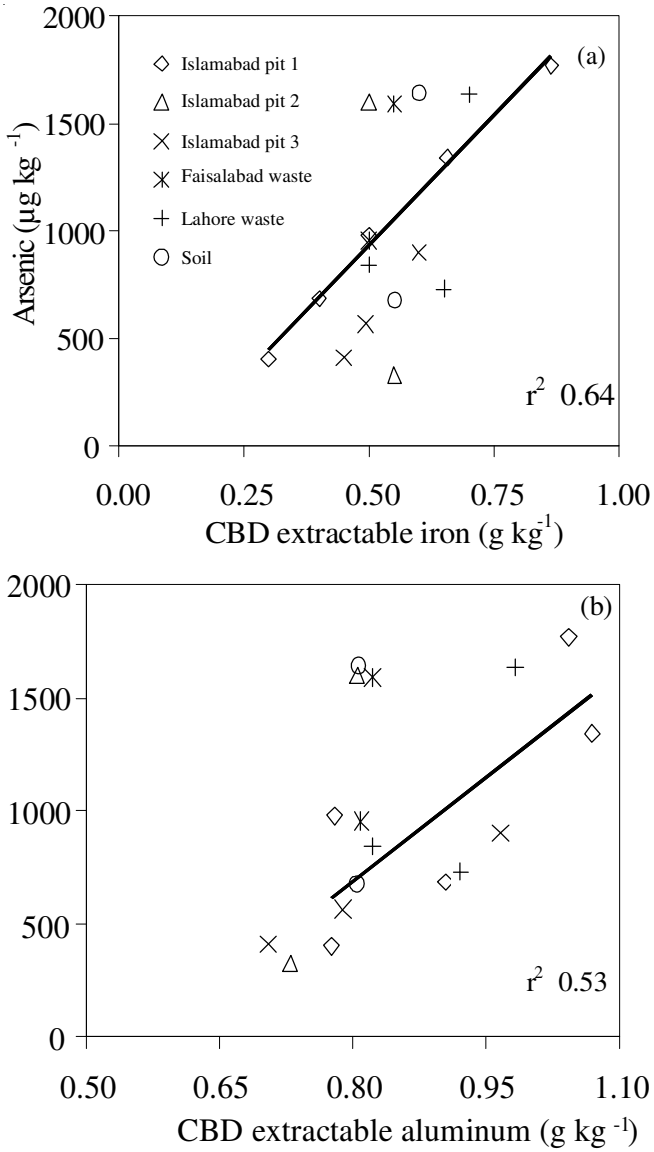


Fig. 8. Depicts positive relationship between arsenic retained in residual phase and citrate bicarbonate dithionate extractable metals: (a) iron and (b) aluminum

ACKNOWLEDGEMENTS

The authors thank to Higher Education Commission for financial support for carrying out this research.

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