Enrichment of arsenic in the Quaternary sediments from Ankaleshwar industrial area, Gujarat, India: an anthropogenic influence

K. D. Shirke · N. J. Pawar

Abstract Arsenic (As) contamination of sediments and waters is known from Bengal and other parts of eastern and central India. However, there is paucity of reports that document occurrence of As in Quaternary sediments from western India. In this paper, we report the enrichment of As in the Quaternary sediments of Ankaleshwar area in the western Indian state of Gujarat. Twenty-five surface and five profile samples were analyzed that indicated spatial and vertical distribution of As in the sediments. The As content of fine size fractions (<63 μm) compared with upper continental crust (UCC) and standard shale indicates anomalous As enrichment that is supported by moderate to high geo-accumulation index (I_{geo}) as well as pollution index (Pi). Enrichment of As is seen in surface sediments from the industrial and urban land use regions, followed by oil field and agricultural land suggesting strong influence of anthropogenic factors. Although the arsenic concentration decreases with depth, the relatively higher values of As coupled with I_{geo} and Pi index values suggest downward migration of metal that is likely to contaminate groundwater. This calls for urgent remediation so that the ill effects of As contamination can be minimized.

Keywords Ankaleshwar · Arsenic (As) · Contamination · Sediments · Anthropogenic factors

K. D. Shirke (✉) · N. J. Pawar
Department of Geology, Savitribai Phule Pune University, Pune 411007, India
e-mail: krushnathshirke@gmail.com

Introduction

Large-scale industrialization in the western Indian state of Gujarat epitomizes regional pollution of soil and water resources (Suyash et al. 2008). The incidence of high levels of toxic metals ensuing from inadequately treated liquid and solid waste supplied to the soil has severely affected environs of various industrial complexes (Fakoyade and Onianwa 2002; Fakoyade and Olu-Owolabi 2003). The pervasiveness of heavy metals in general and augmentation of arsenic in particular beyond their ordinary allocation in sediments characteristically reflects the contamination around the industrial areas.

After the alarming discovery of arsenic toxicity from groundwater and sediments of the Ganges delta in India and Bangladesh (Alam et al. 2002; Raymahashay and Khare 2003), the subsequent studies on central and eastern India reported contamination of Quaternary sediments and groundwater by arsenic (Pandey et al. 2002). However, information on incidence of arsenic from western parts of India is sparse. Further, the earlier studies have pointed out lithogenic derivation of arsenic, and scarcely there are any reports on anthropogenic source of As in the sediments. Therefore, an attempt is made to study Quaternary sediments of Narmada River from Ankaleshwar area of southern Gujarat as there is no information available on occurrence of arsenic. The primary objectives of the present study were (1) to estimate the concentrations of arsenic in the Quaternary sediments and (2) to appraise the levels of enrichment and assess the degree of pollution besides identifying the sources.

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Study area

The Ankaleshwar Industrial Estate (AIE) is located in the western Indian State of Gujarat. It is characterized by large-scale industrial and urban sprawl that was developed at the cost of reclaiming a large extent of fertile alluvial flood plain of Narmada River (Fig. 1). AIE is a chemical domain amid the industrial "Golden Corridor" housing 171 industrial estates created by the Gujarat Industrial Development Corporation (GIDC). It includes an array of chemical, fertilizer, paint, dyes, glass, pharmaceutical, and other allied industries, which have greatly impacted the overall environment. The discovery of hydrocarbons in the area has further hastened the pace of urbanization and industrialization with relative increase in environmental problems (Suyash and Pawar 2010).

A small third-order stream, Amla Khadi, with area 240 km^2 originates in the lower reaches of Narmada and drains the alluvial flood plain (Fig. 1). AIE occurs in the upper and middle reaches of the stream and Ankaleshwar oil field is spread over in the middle-lower part. This part of the catchment and the areas along the river banks are known for intensive farming.

In the lower part, the stream carries industrial effluents with toxic heavy metal concentrations several times above the permissible limits (Suyash et al. 2008). During monsoon, the stream overflows its banks inundating the surrounding plains with polluted waters (Santillo et al. 1996). The AIE includes two industrial zones: (1) Ankaleshwar industrial area (AIA) and (2) Panoli industrial area (PIA) together comprising over 3000 industrial units that are parallel to the National Highway (Mumbai–New Delhi) and Western Railways. AIE generates between 250 and 270 million liters/day of liquid waste and ~50,000 t of solid waste annually (Bruno 1995). Of this, 58 % arises from the manufacture of dyes and dye intermediates, 19 % from drugs and pharmaceuticals, and 5 % from inorganic chemicals (CPCB 1996). Since As is an important constituent in these industries, the setup in the basin provides an opportunity to examine the sources of occurrence and distribution of arsenic in the Quaternary sediments in relation to land use and other anthropogenic factors.

Geologically, the area is covered by thick Quaternary alluvium that is categorized on the basis of environment
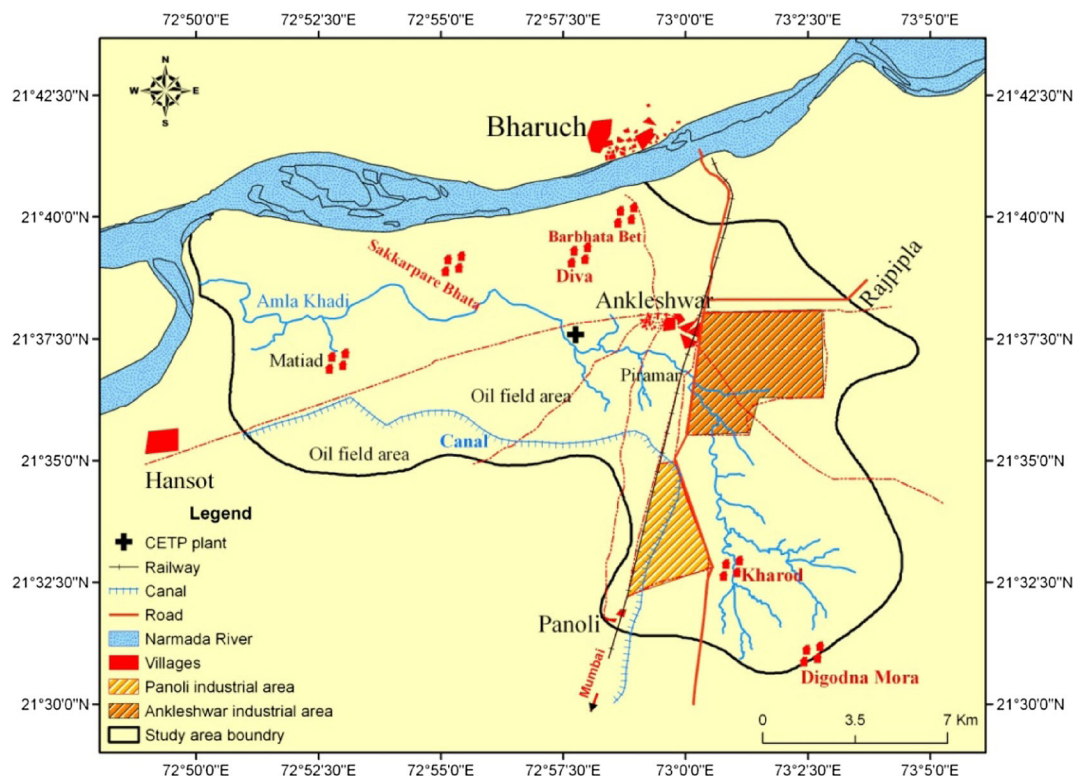

Fig. 1 Location map of Ankaleshwar area, Gujarat, India

of deposition by Geological Survey of India (GSI 2002). Older tidal flat and tidal marsh deposits belong to Rann Clay Formation and younger tidal flat deposits to Mahuva Formation both representing marine environment. AIE is established on flood plain deposits of Katpur Formation representing fluvial depositional environment (GSI 2002). Low-lying flood plain exhibits poorly developed natural drainage with very low relief. It forms a part of regional alluvial deposit of Late Pleistocene to Holocene in age (Bhandari et al. 2001; Chamyal et al. 2002) through which the stream flows (Fig. 2). The saline wastelands and swampy areas of broad estuarine mouth bar of Narmada and older tidal flat/marsh deposits mark the western boundary of this plain. Toward south and east, the plain is bound by uplands comprising Tertiary rocks and Deccan Volcanics (Chamyal et al. 2002).

Material and methods

The focal themes of the present study were to identify presence of arsenic in sediments, identify spatial and depth-wise variations, and to ascertain its possible lithogenic or anthropogenic sources. All 25 randomly selected sampling sites (Fig. 3) representing industrial, urban, agricultural, and oil field areas were chosen based on variation in the land use pattern, and samples were collected in March 2006. Since anthropogenic sources contaminate the upper layer of sediments, the samples from top 15 cm were collected. Profile samples representing industrial (PS-33, PS-38), urban (PS-3), mixed (agricultural + oil field) and polluted lower-middle reach (PS-8), and relatively unpolluted agricultural river bank (PS-19) were collected from different depths. The soils were collected in polythene self-locking bags and stored. Plastic spatula was used for sample collection to evade metal tools.

The samples were kept in darkness at 4 °C until subjected for analysis. Sieved (<0.002 mm) fine-size fraction was subjected to analysis by X-ray fluorescence technique using powder method to determine the elemental concentrations. Spectro Xepos–EDXRF (detection limit 0.5 ppm = mg/kg) manufactured by Ametek was used for the analysis. The analytical data was normalized by using Tertiary shale as a reference standard and upper continental crust (UCC) values to generate graphs (Xianjun Xie
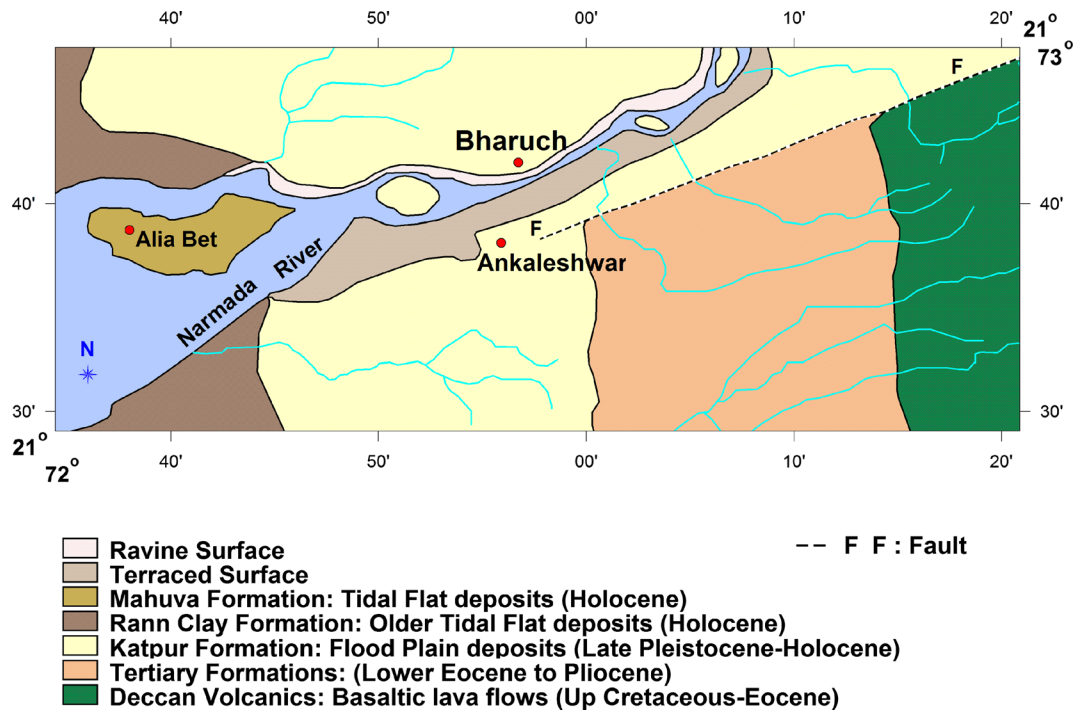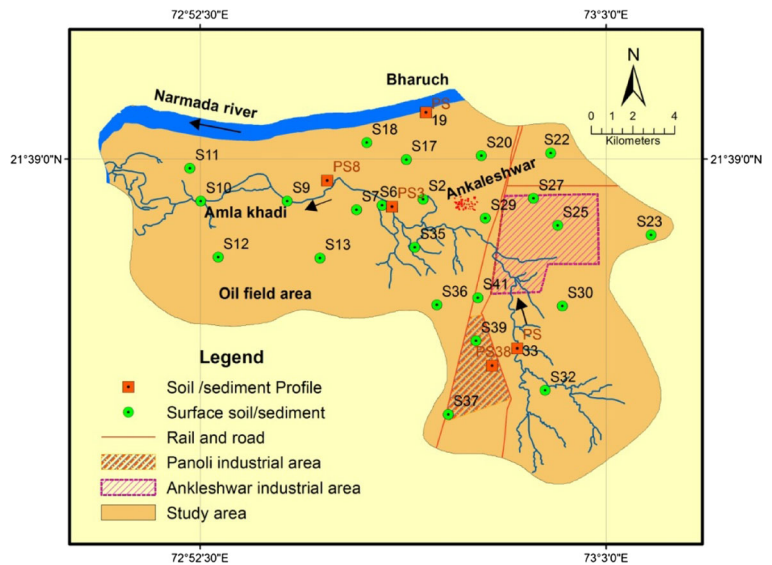

Fig. 2 Geological map of the study area (after GSI 2002; Chamyal et al. 2002)

Fig. 3 Sediment sampling locations in Ankaleshwar area, India

et al. 2008). Enrichment of As in the sediments was ascertained on the basis of index of geo-accumulation [I_{geo}=log2 ($C_n/1.5B_n$)] given by Muller (1969). In this expression, C_n is a measured concentration, B_n is background value (average (av.) shale), and 1.5 is the background matrix correction factor. Sevenfold classification based on I_{geo} (ranging from 0=unpolluted, 1=unpolluted to moderately polluted, 2=moderately polluted, 3=moderately to highly polluted, 4=highly polluted, 5=highly to very highly polluted, to 6= very highly polluted) was adopted. Similarly, Birch and Taylor (1999) have proposed a ratio of a metal to its background as an index (Pi=Ci/Si) to evaluate geo-chemical imbalance to be attributable to anthropogenic activities. Pi is a number indicating pollution, Ci=arsenic concentration, and Si=background value. This index is similar to enrichment factor specified by Sutherland (2000) that is used in classification and nomenclature of contamination categories.

Results and discussion

The summary of the statistics for the analytical values of As in the surface sediments has been shown in Table 1. It is seen that min. value of <0.5 mg/kg is observed in the sample S-18 from the agricultural land use. While, max. value of 18.87 mg/kg in S-23 coincides with that in the industrial area; the moderate values of As in general

coincide with those in urban, semi-urban, and oil field areas. It is interesting to note that all along the course of Amla Khadi stream, the sediments depict moderate arsenic concentration. This is indicative of control of the small effluent laden stream on the distribution pattern of As unlike heavy metals in the AIE (Suyash et al. 2008). The Amla Khadi watershed represents a multifaceted setup in context to anthropogenic activities. Industrial, agricultural, urban, and oil field developments, besides highways and rail routes, have reclaimed a large extent of the flood plains in the region (Suyash and Pawar 2008), and the impact of this is clearly visible in As distribution pattern. In general, the average value of As=8.37 mg/kg suggests that it is a much higher concentration than Tertiary shale (1.2 mg/kg) from the study area and standard sediment composition (2.7–6.5 mg/kg), while it is slightly lower than upper continental crust (UCC; 10 mg/kg). Based on the values of As, the sample locations have been divided into four parts for understanding spatial distribution pattern. These are (1) As=<2 mg/kg, (2) As=2 to 5 mg/kg, (3) As=5 to 10 mg/kg, and (4) As=>10 mg/kg. Only one sample (S-18) belongs to <2 mg/kg class, four locations (S10, S17, S19, S22) belong to As=2–5 mg/kg type, 12 locations (S6, S7, S9, S11, S12, S13, S20, S29, S30, S35, S36, S37) to 5 to 10 mg/kg type, and remaining eight (S2, S23, S25, S27, S32, S33, S39, S41) to >10 mg/kg part. This gives an idea about spatial spread of arsenic in the area that can be further related to land use.

Land use type	Sample nos.	As	I_{geo}	Pi
Urban, semi-urban, and oilfield (middle and lower stretch)	S-2	11.90	3.25	9.92
	S-6	8.78	2.81	7.32
	S-7	6.24	2.32	5.20
	S-9	7.57	2.60	6.31
	S-10	2.68	1.10	2.23
	S-11	6.22	2.31	5.18
	S-12	6.54	2.39	5.45
	S-13	6.38	2.35	5.32
Narmada Bank (agriculture area)	S-17	3.52	1.49	2.93
	S-18	0.5	−1.8	0.41
	S-19	4.68	1.90	3.90
	S-20	7.13	2.51	5.94
	S-22	2.17	0.80	1.81
Ankaleshwar Industrial Area	S-23	18.87	3.92	15.72
	S-25	11.15	3.16	9.29
	S-27	12.12	3.28	10.10
	S-29	9.95	2.99	8.29
Industrial area (upper stretch)	S-30	8.94	2.84	7.45
	S-32	14.16	3.50	11.80
	S-33	13.68	2.26	11.40
	S-35	5.99	2.85	4.99
Panoli Industrial Area	S-36	7.95	3.45	6.63
	S-37	9.01	2.67	7.51
	S-39	11.75	3.23	9.79
	S-41	11.86	3.25	9.88
	Min.	0.5	−1.8	0.41
	Max.	18.87	3.92	15.72
	Average	8.37	2.38	7.01

## Spatial distribution of arsenic: land use controls

To enable comparison between above parts and to draw graphically the spatial distribution pattern of As (using values in mg/kg), the terminology used in the classification by Sutherland (2000) was adopted. The spatial distribution pattern of As is shown in Fig. 4. It is observed that four zones depicting diverse precincts indicating *minimal As enrichment* to *more significant As enrichment* are discernible. Of these, the *more significant As enrichment class* coincides with the AIA and PIA and its surrounding milieu. The possible industrial sources include use of As in glass production and wood preservation industry where chromated copper arsenate is used in the treatment (Barringer and Reilly 2013). Manufacturing of metals and alloys, petroleum refining, and pharmaceutical manufacturing are additional potential sources of As (Ning 2002). The highest concentration of As is observed at S-23 in the vicinity of brick kiln where coal is used. This is followed by *less significant As enrichment* zone corresponding with urban, semi-urban, and oil field areas that cover greater area than other zones. It is possible that combustion of municipal solid waste and application of arsenical pesticides, herbicides, fungicides, and insecticides are likely causes of increase in As concentration in this zone (Matera and LeHecho 2001). The third type, *moderate As enrichment*, is largely coinciding with agricultural land use along river bank. This channel margin region is inundated every year by flood waters where the original deposition of contaminant may have been washed out of the sediments (Casper et al. 2004). This region can, however, be considered as a control zone as the values of As are broadly comparable with the standard sediment composition in the range of 2.7 to 6.5 mg/kg. Therefore, levels of As in the other two zones can be considered indicative of contamination of sediments from anthropogenic sources. The higher concentration of As in industrial land use region suggests deposition from industrial activity possibly by aerial mode coupled with addition from solid waste dumps and effluents. The use of lead arsenate pesticides and addition of phosphate fertilizers in agriculture are also potential sources of As (Barringer and Reilly 2013). Further, the chicken manure contains As because an organic arsenical (4-hydroxy-3-nitrophenylarsonic acid) is used as growth additive in the chicken feed (Hileman 2007). However, less As values in agricultural zone suggest negligible input of chicken manure and other sources.

Spatial distribution of arsenic: depth control

In the study area, profile samples were collected based on diversity in the land use pattern. The locations of profile represent industrial (PS-33 and PS-38), urban (PS-3), mixed (PS-8: oil field + agricultural), and agricultural (PS-19) land use types. Levels of As obtained from various depths at these sampling sites are given in Table 2. Distinct difference in the values of As can be seen in each profile. The lowest levels of average As (4.81 mg/kg) have been observed for samples of PS-19

Fig. 4 Spatial distribution of Arsenic (mg/kg) in surface sediments, AIE, India
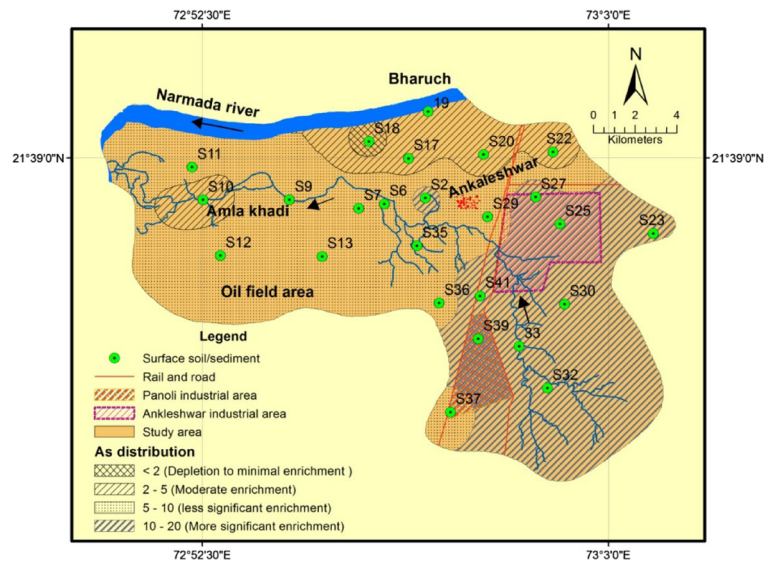
profile from agricultural land and highest av. value of As=14.62 mg/kg for PS-3 in the urban area in the middle of Ankaleshwar town. The As concentrations from industrial setup are comparable with urban area and display av. values from 9.10 mg/kg (PS-38) to 12.08 mg/kg (PS-33). Interestingly, As in mixed land use region (PS-8) depicts higher values than that in agricultural and less than urban sites. In a nutshell, on spatial scale, diverse land use in the area has exhibited control over the distribution of arsenic in the surface as well as profile samples. This suggests that arsenic in the area is primarily derived from anthropogenic sources and there is negligible lithogenic input.

Depth-wise variation in As from varied profiles also brings out different patterns of distribution of arsenic (Fig. 5). In the agricultural division (PS-19), the surface layer sediments show negligible values of arsenic (<0.5 mg/kg) confirming non-lithogenic sources. The PS-19 signifies channel margin region of river Narmada prone to frequent flooding causing washing out effect (Casper et al. 2004). From top layer to the depth of 1 m, the As shows less variation (Fig. 5c). Compared to this, PS-3 from urban area (Fig. 5a) depicts contrasting pattern. Here the highest values of As in the top layer (34.27 mg/kg) and lowest at 2-m depth (10.05 mg/kg) can be noticed (Table 2). This reiterates that As is predominantly derived from surface sources related to anthropogen-

ic activity. Although at this site relative decrease in As in depth-wise direction is observed, the value of As=10.05 mg/kg suggests that this metal has leached to 2-m depth that is cause of concern from groundwater pollution point of view. Suyash and Pawar (2010) have already reported heavy metal pollution of groundwater from this area. Unlike PS-3, the industrial (PS-33) location (Fig. 5d) displays analogous trend with As=14.37 mg/kg in the surface layer that drops by about 62 % to attain value of 5.44 mg/kg at 1.25-m depth. In the mixed land use (agriculture + oil field) zone (Fig. 5b), the As concentration of 7.94 mg/kg in the surface sediments falls to 3.62 mg/kg at 1.80 m depth, while at PS-38 (Fig. 5e), the trend is reverse (from 10.07 to 14.46 mg/kg). To summarize, the depth integrated data suggests that sources of As are related to industrial and urban activities and that, at some places, the metal is migrated to shallower levels. All the profiles except PS-19 exceed values of permissible I_{geo} and standard values of shale and UCC.

Quality of sediments

Several indices have been proposed by many workers to evaluate the contamination levels of sediments (Muller 1969; Singh and Kumar 2006; Ansari et al.

Table 2 Arsenic concentrations (mg/kg) and indices in profile sediment samples

| Land use type	Depth (cm)	Sample no.	As	I_{geo}	Pi
Urban area	0	PS3A	34.27	4.78	28.56
(PS-3)	−15	PS3B	17.74	3.83	14.78
	−25	PS3C	15.11	3.60	12.59
	−35	PS3D	9.95	2.99	8.29
	−45	PS3E	13.14	3.39	10.95
	−55	PS3F	11.40	3.19	9.50
	−70	PS3G	10.76	3.11	8.97
	−130	PS3H	9.13	2.87	7.61
	−200	PS3I	10.05	3.01	8.37
		Min	9.13	2.87	7.61
		Max	34.27	4.78	28.56
		Avg.	14.62	3.42	12.18
Mixed land use	0	PS8A	7.94	2.67	6.62
(oil field+agriculture)	−55	PS8B	6.84	2.45	5.70
(PS-8)	−110	PS8C	8.27	2.73	6.89
	−150	PS8D	5.83	2.22	4.86
	−180	PS8E	3.62	1.54	3.02
		Min	3.62	1.54	3.02
		Max	8.27	2.73	6.89
		Avg.	6.50	2.32	5.42
Narmada Bank	−5	PS19A	0.5	−1.85	0.41
(agriculture)	−13	PS19B	3.50	1.49	2.92
(PS-19)	−25	PS19C	4.76	1.93	3.97
	−30	PS19D	2.32	0.89	1.93
	−32	PS19E	4.39	1.81	3.66
	−40	PS19F	4.81	1.94	4.01
	−70	PS19G	0.5	−1.85	0.41
	−100	PS19H	4.79	1.94	3.99
		Min	0.5	−1.85	0.41
		Max	4.81	1.94	4.01
		Avg.	3.08	1.50	2.57
AIA	−25	PS33A	14.37	3.52	11.98
(industrial area)	−45	PS33B	16.42	3.72	13.69
(PS-33)	−125	PS33C	5.44	2.12	4.54
		Min	5.44	2.12	4.54
		Max	16.42	3.72	13.69
		Avg.	12.08	3.12	10.07
PIA	−10	PS38A	10.07	3.01	8.39
(industrial area)	−50	PS38B	11.83	3.24	9.86
(PS-38)	−140	PS38C	0.5	−1.85	0.41
	−220	PS38D	14.46	3.53	12.05
		Min	0.5	−1.85	0.41
		Max	14.46	3.53	12.05
		Avg.	9.10	2.70	7.59
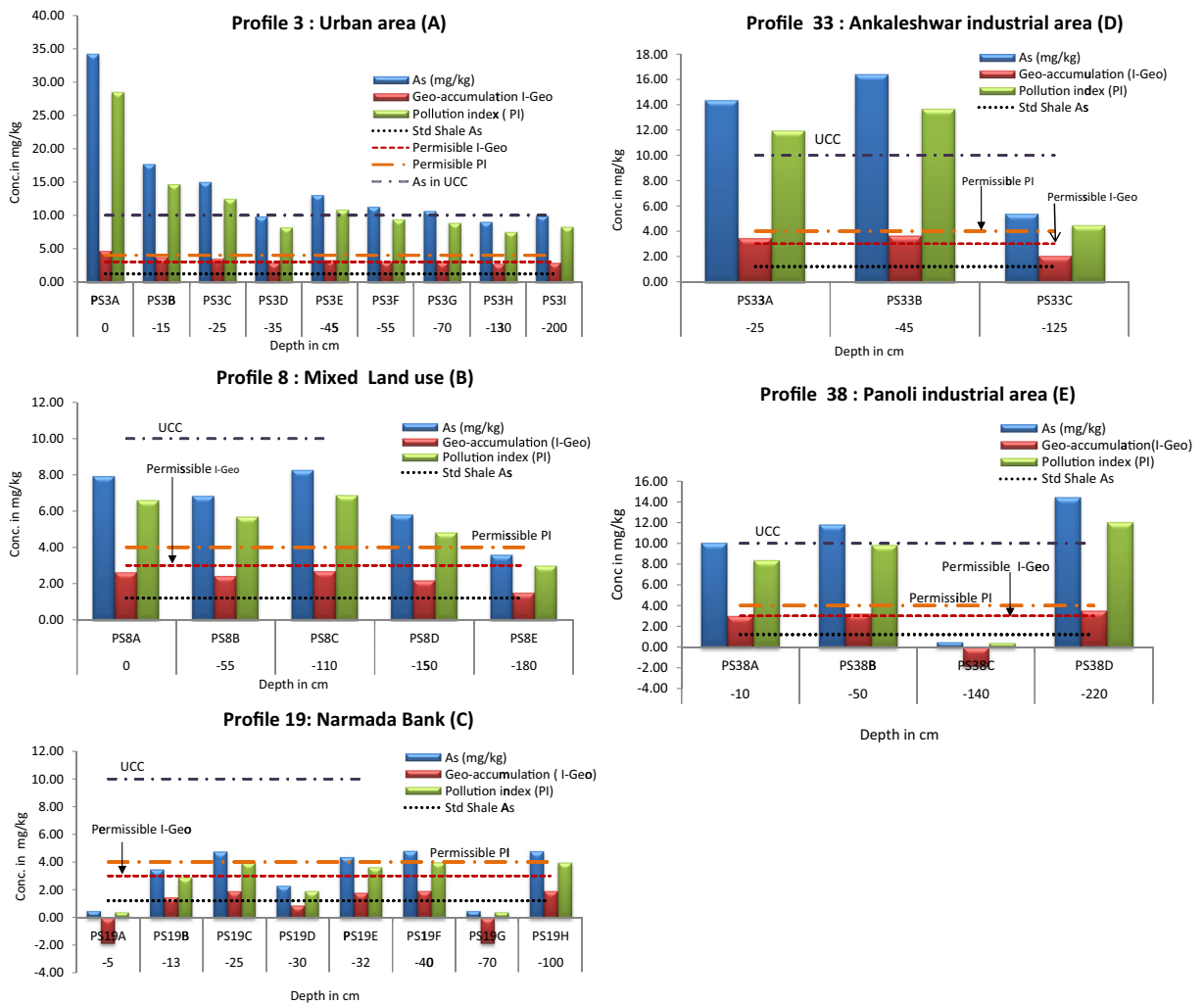
Fig. 5 Depth-wise variations in arsenic (mg/kg) form Ankaleshwar area, India

1998; Sutherland 2000; Jain et al. 2007; Upadhyay et al. 2007; Krishna and Govil 2007). Before determining the indices, the values of As for surface and profile sediments were normalized with UCC and Tertiary shale (Fig. 6) to get first-hand information about anomalous concentrations. It is interesting to note that almost all samples except S.No.S-18 display higher values of As than standard shale from the area. However, as compared to the UCC, "As anomaly" is clearly visible in the samples S-2, S-23, S-25, S-32, S-33, S-39, and S-41. These samples are predominantly from industrial and urban areas. This suggests that surface sources in the form of anthropogenic activity have contributed to arsenic in the sediments from the area.

Geo-accumulation index

To quantify the metal accumulation in the sediments, the geo-accumulation index proposed by Muller (1969) was used. As stated earlier, it is expressed as $I_{geo}=\log_2 C_n/1.5B_n$. In the study area, only four classes of I_{geo} are represented as the index values (min. −1.8 to max. 3.92, Av. = 2.46) are below 4 in the case of surface samples (Table 1). This indicates that sediment quality from the area belongs to unpolluted to highly polluted class. Interestingly, however, the profile samples bring out distinct pattern of variation in I_{geo} values based on land use pattern. The control profile from agricultural land (PS-19) shows minimum I_{geo} (av.=0.79) suggesting unpolluted to moderately polluted sediment class. On
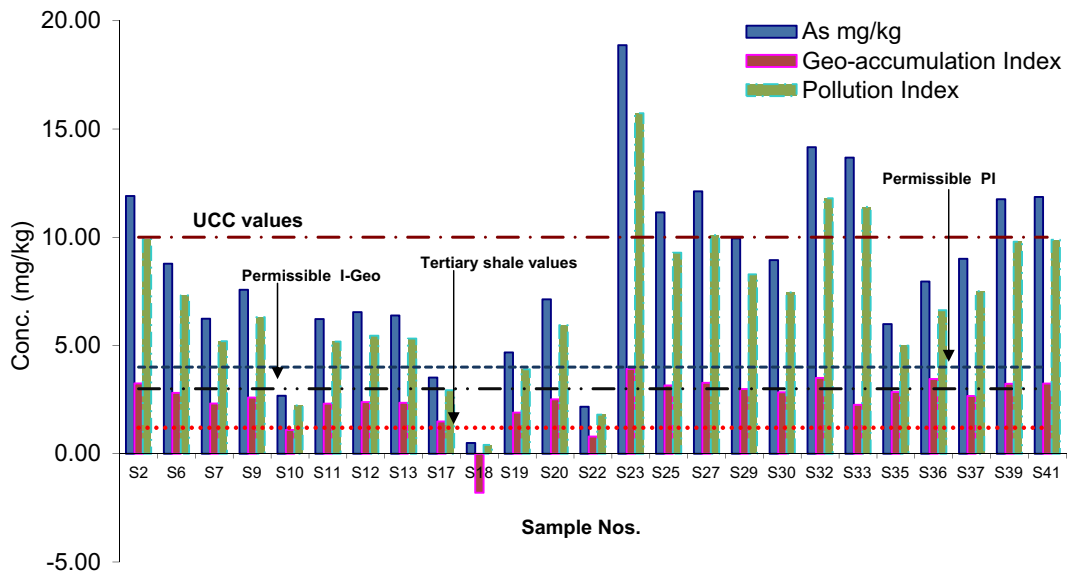
Fig. 6 Variations in Arsenic in surface samples along with Pi and I_{geo} indices

the other hand, the PS-3 from urban area depicts maximum I_{geo}=3.42 reflecting highly polluted category of sediments. Similarly, the PS-33 from industrial setup shows highly polluted sediment class (I_{geo}=3.12) followed by moderately to highly polluted type at PS-38 (I_{geo}=1.98) industrial locality. Further, it is to be noted that in the mixed land use zone (PS-8) with I_{geo}=2.32, the sediments are of moderately to highly polluted category. This implies that anthropogenic activities in the form of various land use practices have seriously influenced the sediment quality from the area. As a result of this, the arsenic values in the sediments from Narmada River flood plain have risen many fold and pose a serious threat to the environment in general and food chain in particular.

Pollution index

The value of arsenic and its comparison with background values of shale from the area were used in quick assessment of its enrichment. This index was used to understand the enrichment of As and its possible sources in the area. The index for surface samples ranges from 0.41 (S-18) to 15.72 (S-23 situated near brick kiln). The samples from industrial area (S-27, S-32, and S-33) illustrate higher values of Pi suggesting enrichment of arsenic in the sediments. The remaining samples with a little lower Pi numbers from urban and industrial segment show moderate enrichment of As in the sediments.

Similarly, the profile samples from different land use types display distinct difference in the values of Pi. This is to say that min. average value of Pi=2.66 is observed for PS-19 in the agricultural zone. In contrast, max. average value of Pi=12.18 is seen in the PS-3 from urban zone. This is followed by slightly lower Pi values in industrial (Pi=10.07 at PS-33 and 7.68 at PS-38) and mixed land use (Pi=5.42 at PS-8) areas. This reaffirms the fact that urban and industrial areas have contributed to increase in the values of arsenic in the area.

Conclusions

The data collected on levels of arsenic from Ankaleshwar industrial area reveals that the surface sediments in the area are contaminated and show relatively higher levels of As than standard values of shale. The high values of As from industrial area followed by urban and oil field regions demonstrate that varied anthropogenic activities have contributed significantly toward the arsenic content of the soil/sediment resources. This is evident from the high values of I_{geo} and pollution indices around industrial, urban, and oil field areas. Indices used as a tool for computing the degree of degradation of the sediment quality suggest that both the surface and profile samples have reached alarming levels in the industrial setup showing highly polluted sediment class followed by moderately to highly

polluted type in the mixed land use zone. The control profile from agricultural land suggests unpolluted to moderately polluted sediment class. In general, diverse precincts point to *minimal As enrichment* to *more significant As enrichment* reflecting impact of land use pattern on spatial distribution of arsenic. Although relative decrease in As depth-wise is observed, the metal has migrated up to a 2-m depth that is cause of concern from groundwater pollution point of view. Since the area is highly cultivated and is known for the intensive cultivation of crops of cotton, vegetables, and fruits, it is likely to lead to the bio-accumulation and bio-magnification of As and pose pathological hazards. This also has repercussion on the pollution of groundwater resources of the area that is used by the local populace for drinking, domestic, and irrigational needs. The study therefore calls for an urgent need for upgradation of the waste management practices and remedial methods as far as As pollution is concerned.

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