



## Distribution of Arsenic (As) in Water, Sediment and Fish from a Shallow Tropical Reservoir (Aiba Reservoir, Iwo, Nigeria)

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**ABSTRACT:** The status of arsenic in Aiba Reservoir, Iwo, Nigeria was assessed to determine its levels and distribution in water, sediment and tissues of fish. Total arsenic was estimated by atomic absorption spectrophotometry. The mean levels detected for reservoir water ( $1.50 \pm 0.22$ ppb) and sediment ( $2.00 \pm 0.17$ ppb) were below the World Health Organization recommended limit of 0.01mg/L (10ppb) for drinking water. Arsenic in sediments significantly followed ( $r = 0.588$ ,  $p = 0.002$ ,  $n = 24$ ) the level of contamination of water. The distribution of arsenic in reservoir water shows significant spatial and temporal heterogeneity; while that for sediment shows temporal homogeneity. Mean As levels for fish kidney ( $15.72 \pm 4.14$ ppb) and liver ( $12.04 \pm 2.73$ ppb) were significantly higher than levels for fish gills ( $2.03 \pm 0.34$ ppb) and muscle ( $1.46 \pm 0.13$ ppb). The first and second Canonical Variate showed 49.82% and 34.75% between-species variation respectively. This report suggests that fish at the lower level of the food web have higher levels of As compared to those at a higher trophic status. The current low levels of arsenic in the abiotic component of the reservoir suggest that contamination is mainly from anthropogenic rather than from natural sources. This report suggests seasonal, municipal and agricultural sources of arsenic contamination of Aiba Reservoir. ©JASEM

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Arsenic is a ubiquitous metalloid and a notorious poison (Ravenscroft *et al.*, 2009) because it is injurious to human health. It has also been classified as a human carcinogen. Apart from natural sources, mining related activities are major sources of arsenic contamination of surface waters. Others are coal burning, industrial sources, municipal wastes, pesticides, chemical fertilizers, wood preserving arsenicals and traffic emissions (Eisler, 1988; Garelick *et al.*, 2008; Garba *et al.*, 2012a; Laniyan, 2013; Olmedo *et al.*, 2013; Luilo *et al.*, 2014). Trace metals are accumulated by aquatic organisms and persist in food webs (Oliveira Ribeiro *et al.*, 2005). Biota such as fish have been recommended as valuable biological indicators in aquatic environmental contamination and pollution assessments (Naigaga *et al.*, 2011; Rosso *et al.*, 2013; Ayeloja *et al.*, 2014) because they are large and easily identified (Kumolu-Johnson *et al.*, 2010); have longer life-span and high position in the aquatic food chain (Farkas *et al.*, 2001). Trace metals affect fish health and disrupts reproductive patterns in fish (Ebrahimi and Taherianfard, 2011).

Arsenic has no known essential role in living organisms; exhibit extreme toxicity even at very low (trace) levels and exposure and has been regarded as a threat to all forms of life especially human health (Eisler, 1985; Järup, 2003; Olmedo *et al.*, 2013). However, some authors have suggested that arsenic

might be an essential element for organisms at low concentrations (De Gieter *et al.*, 2002). Consumption of fish and drinking water are sources of human exposure to arsenic poisoning; while water is an important source of the more toxic inorganic arsenic, food fish is an important source of exposure to the less toxic organic arsenic (WHO, 2008). It is reported that about 90% to 95% of total arsenic in many organisms is in form of the inert, less toxic organic form (De Gieter *et al.*, 2002; Olmedo *et al.*, 2013; Medeiros *et al.*, 2014). Organic arsenic in humans is rapidly excreted; a strong correlation between total arsenic in the urine of pregnant Mediterranean women and total lean fish consumption has been reported (Fort *et al.*, 2014). Values greater than the provisional WHO permissible level of 10ppb arsenic in drinking water (WHO, 2008) have been reported in surface waters in parts of Biu Volcanic Province, North-Eastern Nigeria with inhabitants of the area showing obvious symptoms of arsenic poisoning (Usman and Lar, 2013); underground water from Kaduna State (Musa *et al.*, 2008; Garba *et al.*, 2012b); Ibadan, Oyo State (Egbinola and Amanambu, 2014); underground and surface waters from some major towns in Ogun State (Kayode *et al.*, 2011). However, arsenic values within WHO maximum permissible limit has been reported for the underground waters of Odede, Ogun State (Amori *et al.*, 2013). For food fish, greater than the WHO recommended limit has been reported from

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freshwaters in Ibadan, Oyo State (Tyokumbur and Okorie, 2014). Ravenscroft *et al.*, (2009) noted the paucity of information on natural contamination of arsenic in the inland waters of Africa and contradictory reports from Nigeria. This study will add to the little information on arsenic contamination of surface waters in southwest Nigeria.

This study, therefore, assesses the sources and levels of arsenic in water, sediment and tissues of different fish species inhabiting Aiba Reservoir, Iwo, Nigeria, in order to determine their distribution and probable indicator fish species.

## MATERIALS AND METHODS

A detailed account of the characteristics of Aiba Reservoir has been given by Atobatele and Ugwumba (2010). Location A is North-east of the reservoir, near source of inflow (upstream) with little human activity such as occasional washing/bathing (Figure 1); Location B is South-east and drains land used as forest reserve and for agricultural activities; Location C is North of the reservoir and drains an area with rapid residential and occupational encroachment; while Location D is South of the reservoir, close to outflow (downstream) with high human activity such as swimming, bathing, domestic washing and removal of fish intestines.

Water and sediment samples were collected from four locations (A, B, C and D) within the reservoir and at three different months (November, at the onset of the dry season; April, at the onset of the wet season; and June during the wet season). Prior to sampling, all sample bottles were washed with liquid detergents, rinsed with tap water, soaked in 10% HNO<sub>3</sub> for 48 hours, and thereafter rinsed with distilled water. The plastic bottles meant for sampling were further rinsed on site with surface waters prior to collection of reservoir water. Surface water samples were drawn into a 2 L plastic container. The water samples were transported to the laboratory within one hour of collection and kept in the refrigerator until ready for analyses.

Sediment samples were collected from each location using a Van-Veen grab sampler. The sediments samples were collected in aluminium foil paper, thoroughly mixed and stored in labelled black polythene bags for laboratory analysis. Sediment samples were air dried for about five days; ground using a silica pestle and mortar and sieved through a 2-mm mesh sieve.

Fish species were bought from fishermen at their landing sites and taken to the laboratory in a cool box. In the laboratory, each fish sample was dissected with stainless steel forceps, scalpels and scissors in a dissecting tray to bring out different tissues (gills, kidneys, liver and muscle). Fish tissues were then

dried in labelled pre-washed crucibles at 70°C for 24h in an oven. Each dry sample was then pulverised and homogenized with porcelain mortar and pestle and stored in labelled pre-treated specimen bottles. Each sample was transferred to pre-weighed crucibles and dried again in the oven until constant weight was obtained. One gram of the dry sample was digested with analytical grade nitric acid (HNO<sub>3</sub>) on a hot plate at 100°C to near dryness when a clear solution was obtained. The solution was then diluted up to 25ml with distilled water. Total arsenic estimation was made by the method proposed by Norris and Lake (1984). Blank experiments were run to check for background contaminants by the reagents and apparatus used. The values obtained from running blank experiments were subtracted from the analyte values as applicable. The Atomic Absorption Spectrophotometer (AAS) used was calibrated to evaluate the response of the analytical procedure with respect to known quantity of the standard of the metalloid so that the response to unknown quantities in the samples could be reliably estimated. Analysis of each sample was carried out in duplicate and results are expressed in parts per billion (ppb); picogram per litre for water and microgram per kilogram for dry weight of sediment and fish.

Pearson correlation coefficient was applied to explore the relationship between arsenic levels in reservoir water and sediment. One way Analysis of Variance was carried out to determine significant difference among fish species for all tissues combined and among tissues for all fish species combined. Duncan Multiple Range Test was adopted as the post-hoc test using SPSS software (SPSS 14.0 for Windows Evaluation Version, Release 14.0.0; 5 September, 2005). Graphical illustration of results was made using Microsoft Excel Software (2007). Canonical Variate Analysis plot was used to discriminate between fish species with 95% confidence regions about means using GenStat Release10.3 Discovery Edition.

## RESULTS AND DISCUSSION

There was a gradual and significant decrease ( $p < 0.05$ ) in mean water As levels from location A ( $2.17 \pm 0.48$ ppb) to location D ( $0.83 \pm 0.31$ ppb); the sediment from location B had a significantly higher mean As concentration ( $2.83 \pm 0.31$ ppb) compared to other locations (Figure 2). Although mean sediment values for all locations except location A were higher than mean water values, only locations B and D had significantly higher mean sediment concentrations ( $2.83 \pm 0.31$ ppb and  $1.83 \pm 0.31$ ppb respectively) compared to their respective mean water concentrations ( $1.83 \pm 0.31$ ppb and  $0.83 \pm 0.31$ ppb respectively). Arsenic levels less than 1ppb have been reported for uncontaminated freshwater ecosystems of sub-Saharan Africa, especially for Burkina Faso (Ouédraogo and Amyot, 2013); therefore, the mean

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water arsenic levels recorded for this study show contamination of the reservoir water. The distribution of arsenic in reservoir water shows significant spatial and temporal heterogeneity; while that for sediment shows spatial heterogeneity.

The mean water As concentration in the month of June ( $2.50 \pm 0.33$ ppb) was significantly higher ( $p < 0.05$ ) than for other sampled months (Figure 3). There was no significant difference in the mean sediment As concentration for the sampled months. However, a significantly higher mean sediment value compared to mean water value was recorded for the month of November ( $2.00 \pm 0.27$ ppb and  $1.00 \pm 0.27$ ppb respectively) and April ( $1.75 \pm 0.25$ ppb and  $1.00 \pm 0.27$ ppb respectively). Arsenic in sediments significantly followed ( $r = 0.588$ ,  $p = 0.002$ ,  $n = 24$ ) the level of contamination of water. The significant higher water levels of As in June during the wet season; the gradual but significant decrease downstream and the significant high sediment levels at location B suggests that run-off from catchment area during the wet season and agricultural activities are major sources of arsenic contamination of the reservoir. Agricultural source of freshwater contamination by arsenic compounds is supported by Garba *et al.*, (2012). The presence but low level arsenic concentration in commercially available inorganic fertilizers in Nigeria has been reported (Benson *et al.*, 2014). The results also suggest that arsenic sediments thereby increasing its levels as the reservoir water moves downstream.

A total of nine fish species (Table 1) were used in the analysis. All except *Channa obscura* are common in the daily catch of artisanal fishermen of Aiba Reservoir. The combined mean concentrations of As in fish gills ( $2.03 \pm 0.34$ ppb) and muscles ( $1.46 \pm 0.13$ ppb) were significantly lower than those recorded for fish kidneys ( $15.72 \pm 4.14$ ppb) and liver ( $12.04 \pm 2.73$ ppb) (Table 1). *Tilapia zillii* had the lowest mean gill As level ( $0.50 \pm 0.29$ ppb) while *Channa obscura* had the highest ( $6.50 \pm 2.22$ ppb). The muscles of *Marcusenius senegalensis* and *Oreochromis niloticus* had lower values of As ( $1.00 \pm 0.41$ ppb and  $1.05 \pm 0.39$ ppb respectively) compared to those of *Labeo senegalensis* and *C. obscura* ( $2.50 \pm 0.50$ ppb and  $2.50 \pm 0.29$ ppb respectively). The lowest mean kidney As value was recorded for *Chrysichthys nigrodigitatus* ( $1.08 \pm 0.79$ ppb) while the three cichlids studied each had greater than 20ppb mean As kidney levels. *C. nigrodigitatus* had the lowest mean liver value ( $2.13 \pm 1.09$ ppb) while *O. niloticus* ( $37.50 \pm 12.50$ ppb) and *L. senegalensis* ( $49.95 \pm 16.65$ ppb) had higher mean values. Freshwaters are major repositories of arsenic and fish are key constituents in the freshwater environment. The mean fish arsenic levels recorded for this study are lower than those reported for 19 fish species from Three Gorges Reservoir, China (Zhang *et al.*, 2007); and from edible tissues of 10 fish species from naturally

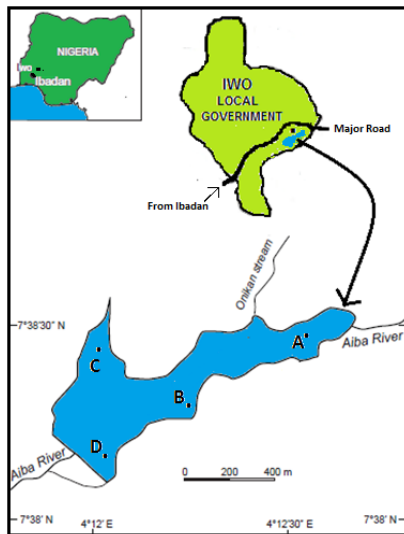
contaminated rivers in a central region of Argentina (Rosso *et al.*, 2013). The significantly higher mean arsenic levels in fish kidneys and liver suggests that they have relatively high potential for arsenic accumulation compared to gills and muscles. Muscle, the edible portion of all fish species, had the lowest mean As values in all cases studied, and this was closely followed by the mean levels in fish gills. *C. obscura* had significantly higher As value for gills compared to other fish species. This suggests that this bottom feeding fish accumulates As from the sediment through their gills; in contrast with *T. zillii* that forages in the open waters. The mean As concentration in the kidneys of cichlids (*T. zillii*, *S. galilaeus* and *O. niloticus*) were higher than those for *H. odoe*, *C. auratus*, *M. senegalensis* and *C. nigrodigitatus*. *L. senegalensis* and *C. obscura*, both bottom feeders, recorded moderate mean As kidney values.

This study revealed that both *O. niloticus* ( $21.05 \pm 7.71$ ppb) and *L. senegalensis* ( $17.24 \pm 8.11$ ppb) had significantly higher mean As levels compared with *C. nigrodigitatus*, *C. auratus*, *Hepsetus odoe* and *M. senegalensis* ( $1.55 \pm 0.33$ ppb,  $3.07 \pm 0.51$ ppb,  $3.44 \pm 0.76$ ppb and  $4.26 \pm 1.25$ ppb respectively). Canonical Variate Analysis shows that the first canonical variate accounted for 49.82% of the between species variation while canonical variate 2 accounted for 34.75% (Figure 4). *Tilapia zillii* and *Channa obscura* are distinctly and significantly different in organ arsenic level distribution from each other and from all other sampled species in the reservoir. The result also shows that the three cichlid species differ in arsenic level distribution while the two bagrid species have similar distribution with the piscivore *Hepsetus odoe*. This suggests that cichlids especially *O. niloticus* and *L. senegalensis*, a substrate feeder at the lower level of the food web have higher levels of As, while the bagrid fishes of the genus *Chrysichthys* and *H. odoe*, a piscivore with a higher trophic status, have relatively lower levels of As in their systems. This agrees with the report of De Gieter *et al.*, (2002) that interspecies differences in arsenic levels of fish in the North Sea is directly related to their food source and that arsenic does not seem to biomagnify along the food chain but is metabolized. This supports the evidence that arsenic may be nutritionally essential or beneficial (Eisler, 1988). Budiati (2010) opines that arsenic and its metabolite bioaccumulate in tissues of aquatic organisms. This is obvious for this study as fish tissue concentration, except for muscle, is higher than for the surrounding water. In terms of arsenic contamination of surface and underground waters, Africa is reported to be one of the two least impacted continents of the world (Ravenscroft *et al.*, 2009). The current low levels of arsenic in Aiba Reservoir water and sediment suggests that contamination is mainly from anthropogenic rather than from natural sources; however it has been reported that the effects

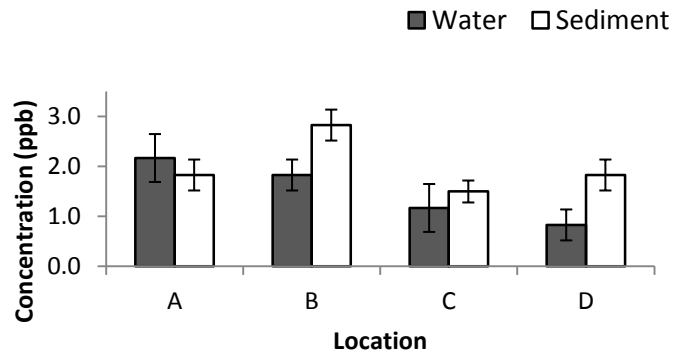
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of arsenic in food and water are both additive and cumulative (Ravenscroft *et al.*, 2009).

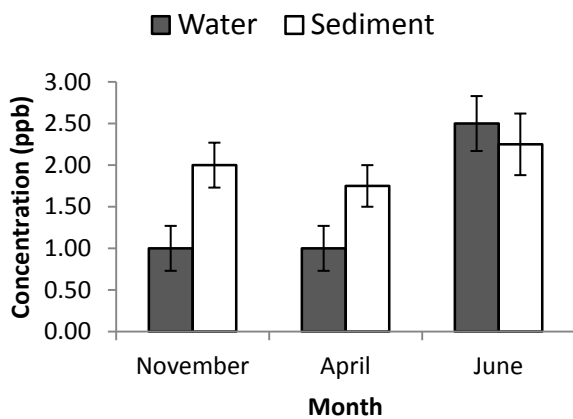
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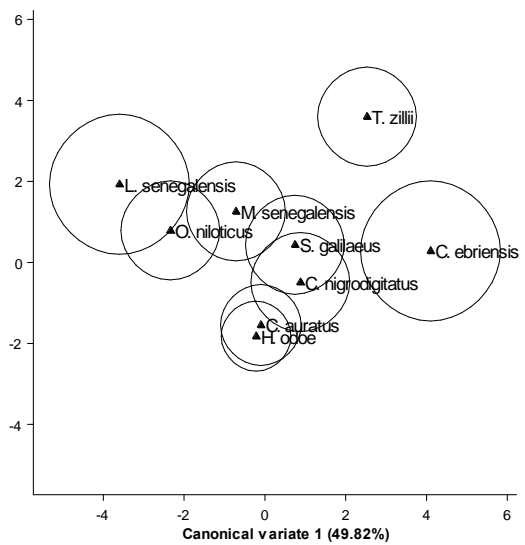
**Fig 1:** Aiba Reservoir Iwo showing locations for water and sediment samples.



**Fig 2:** Water and sediment mean ( $\pm$  standard error) arsenic concentrations from four locations in Aiba Reservoir, Iwo.



**Fig 3:** Water and sediment mean ( $\pm$  standard error) arsenic concentrations from Aiba Reservoir, Iwo in November, April and June.



**Fig 4:** Canonical variate analysis plot of arsenic levels in sampled organs of fish against sampled species with 95% confidence regions about means.

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**Table 1:** Mean and standard error arsenic (As) values in parts per billion (ppb) for different tissues of nine fish species from Aiba Reservoir. One-Way Analysis of Variance was carried out for total mean values with post-hoc test to determine significant differences in measured parameters.

S/no	Fish species	Gill Mean ± SE (ppb) (Range)	Kidney Mean ± SE (ppb) (Range)	Liver Mean ± SE (ppb) (Range)	Muscle Mean ± SE (ppb) (Range)	Total Mean ± SE (ppb) (Range)
1	<i>Marcusenius senegalensis</i>	1.01 ± 0.58	5.78 ± 1.93	10.00 ± 2.04	1.00 ± 0.41	4.26 ± 1.25 <sup>ct</sup> (0 - 15)
2	<i>Labeo senegalensis</i>	1.50 ± 0.50	15.00 ± 5.00	49.95 ± 16.65	2.50 ± 0.50	17.24 ± 8.11 <sup>ab</sup> (1 - 67)
3	<i>Hepsetus odoe</i>	2.00 ± 0.27	6.25 ± 1.25	4.94 ± 2.41	1.25 ± 0.25	3.44 ± 0.76 <sup>c</sup> (0 - 20)
4	<i>Chrysichthys auratus</i>	2.00 ± 0.45	6.00 ± 1.16	4.17 ± 0.87	1.17 ± 0.31	3.07 ± 0.51 <sup>c</sup> (0 - 8)
5	<i>Chrysichthys nigrodigitatus</i>	1.50 ± 0.29	1.08 ± 0.79	2.13 ± 1.09	1.50 ± 0.29	1.55 ± 0.33 <sup>c</sup> (0 - 5)
6	<i>Channa obscura</i>	6.50 ± 2.22	15.00 ± 5.00	ND	2.50 ± 0.29	6.60 ± 1.88 <sup>bc</sup> (2 - 20)
7	<i>Tilapia zillii</i>	0.50 ± 0.29	37.50 ± 12.50	6.66 ± 2.36	1.50 ± 0.29	7.83 ± 3.72 <sup>bc</sup> (0 - 50)
8	<i>Sarotherodon galilaeus</i>	1.50 ± 0.29	20.63 ± 11.01	11.13 ± 5.09	1.50 ± 0.65	8.69 ± 3.40 <sup>bc</sup> (0 - 50)
9	<i>Oreochromis niloticus</i>	1.50 ± 0.29	44.16 ± 23.62	37.50 ± 10.13	1.05 ± 0.39	21.05 ± 7.71 <sup>a</sup> (0 - 100)
	Total	2.03 ± 0.34 <sup>**</sup> (0 - 12)	15.72 ± 4.14 <sup>a</sup> (0 - 100)	12.04 ± 2.73 <sup>a</sup> (0 - 67)	1.46 ± 0.13 <sup>b</sup> (0 - 3)	7.15 ± 1.19 (0 - 100)

ND = Not Determined;

\* = mean values with different column superscripts are significantly different from each other;

\*\* = mean values with different row superscripts are significantly different from each other.

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