

## Characterization of arsenic occurrence in the water and sediments of the Okavango Delta, NW Botswana

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### Abstract

Detailed chemical analyses were performed on surface water, groundwater and sediment samples collected from the Okavango Delta between February and November 2003 in order to examine the distribution and geochemistry of naturally occurring As in the area. Surface water in the Okavango Delta, which is neutral to slightly acidic and has high dissolved organic C (DOC), was found to be slightly enriched in As when compared to a global value for stream water. Of the 20 new borehole analyses from this project, six were found to have values exceeding 10 µg/L, the current World Health Organization provisional guideline value for As. The results from field speciation indicate that As(III) is slightly more predominant than As(V). There is a positive correlation between As and pH and between As and DOC in the groundwater samples. For the sediment samples, there is a positive correlation between As and Co, As and Fe, As and loss on ignition (LOI) and between As and the percent fines in the sample. Reductive dissolution of oxides and hydroxides in the sediments with organic C as an electron acceptor is the likely mechanism for the release of As from the sediments into the groundwater.

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### 1. Introduction

Botswana is a relatively large (582,000 km<sup>2</sup>) flat country with mean annual rainfall varying from 650 mm/a in the north to 250 mm/a in the south. Mean annual potential evapo-transpiration is estimated to be 4–5 times higher than the average mean annual rainfall (450 mm/a). Although

groundwater recharge rates are generally low in Botswana, averaging 2.7 mm/a (Snowy Mountains Engineering Corporation (SMEC), 1991), groundwater has been the main source of potable water supply. Figures released in the National Water Master Plan (SMEC, 1991), indicate that groundwater accounted for 67% of the 1990 total water consumption in Botswana.

The Okavango Delta (Fig. 1) is part of a large alluvial fan complex in north-western Botswana. Flow into the Delta system through the Okavango River, which originates in Angola, peaks after the

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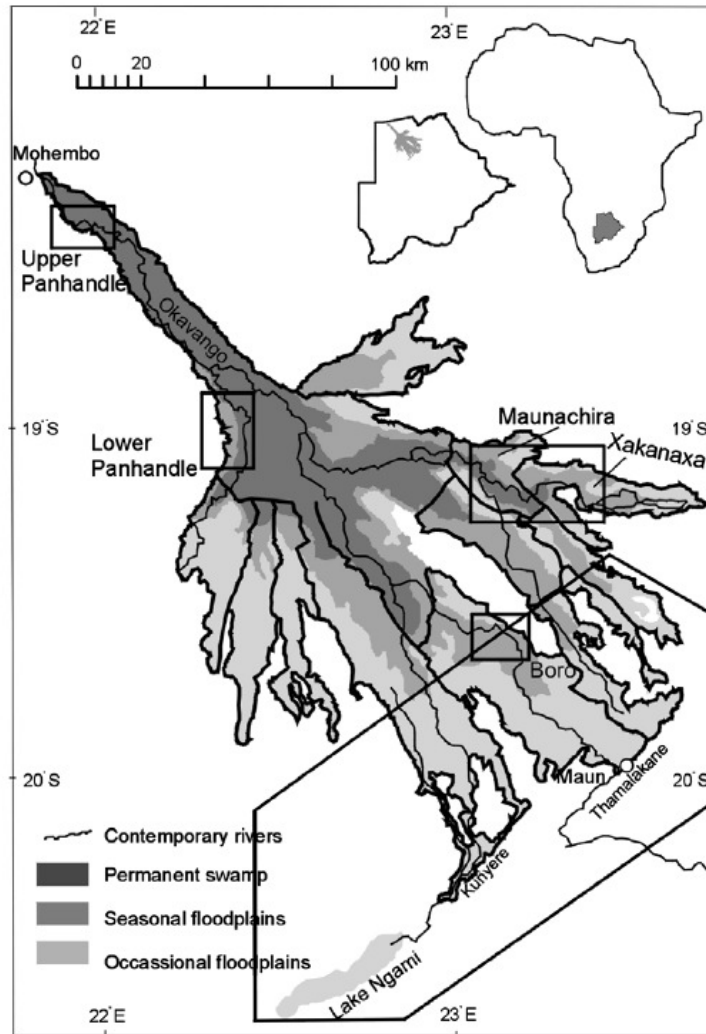


Fig. 1. Location map of the Okavango Delta in NW Botswana with rectangles showing the location of the surface water sampling.

rainy season in Angola. Only 2% of the mean annual inflow leaves the system as surface water outflow through the Thamalakane and Boteti Rivers.

Arsenic in the environment has received renewed attention due to the recent studies of elevated As in Bangladesh and neighbouring West Bengal, India (Chatterjee et al., 1995; Das et al., 1995; Nickson et al., 1998; BGS and DPHE, 2001; Welch and Stoltenwerk, 2003), and the debates in the USA over the maximum contaminant level (MCL) for drinking water (US EPA, 2001a; SenGupta and Greenleaf, 2002).

Evidence shows that high-As groundwater can be associated with reducing conditions prevalent in

alluvial and deltaic environments (Sengupta et al., 2004). Occurrence of As-enrichment in Bangladesh groundwater shows a close relationship with the geomorphological units. Quaternary sediments provide good aquifers in Bangladesh and As-enrichment is mainly restricted to the Holocene alluvial aquifers at shallow and intermediate depths (BGS and DPHE, 2001). The sediments are dominantly composed of sands, silts and clays (Ahmed et al., 2004).

The issue of potentially elevated As in groundwater of the Okavango Delta was first raised during the second phase of the Maun Groundwater Development Project (MGDP; Department of Water

Affairs (DWA, 2003). Based on those preliminary results, it was decided to conduct a more comprehensive study. The objectives of this paper are therefore: (1) to present new chemical analyses of groundwater and surface water samples and Quaternary sediments from the Okavango Delta; (2) to relate the presence of As in the Okavango Delta environment to the hydrogeological setting and geochemical processes; and (3) to determine the speciation of As in the ground water of the Okavango Delta.

## 2. Hydrological setting

In semi-arid NW Botswana, the development potential of the Okavango Delta has included plans for agriculture and water for mining, whilst more recent interest in development has focused on water supply for human consumption (UNDP, 1977; DWA, 2003). Maun, with a population of 49,822 in 2001 (Central Statistics Office, 2001), is the main source of demand for potable water in the region. The rapid growth of Maun, estimated at 3.5% per annum, has necessitated exploration of groundwater resources.

The construction of the Shashe wellfield (Fig. 2) began in 1986 and has subsequently been expanded by three groups of boreholes between the 1980s and 1990s. The main source of recharge to the lower Okavango Delta is river recharge with localised rainfall providing minimal groundwater recharge (DWA, 2003). With no river flow in the Shashe river since 1989 and increasing extraction, the Shashe wellfield was becoming stressed and the shallow unconfined aquifer dewatered. Recent groundwater development strategies (MGDP) have focussed on assessing the Matsibe, Xudum, Kunyere and Gomoti areas (Fig. 2; DWA, 2003).

The Okavango Basin is a Quaternary half-graben (McCarthy et al., 1993a; Modisi, 2000; Modisi et al., 2000) inferred to represent the south-western tip of the East African rift system (Kampunzu et al., 1998). It is located within the Kalahari Basin which is a shallow intracontinental basin covered by sand dunes, minor lacustrine deposits and precipitates (e.g., Thomas and Shaw, 2002).

The Okavango River enters the Middle Kalahari, through the Panhandle (Fig. 1) before extending into an inland alluvial fan. The Okavango River drains from the central highlands of Angola where annual inflow is estimated at  $1.01 \times 10^{10} \text{ m}^3$  measured at the apex of the Panhandle (McCarthy

et al., 2000). The Okavango Delta shows progressive degrees of inundation throughout its length and is subdivided into distinct geomorphic regions: (1) the Panhandle where the Okavango River is constrained; (2) the permanent swamps (3) the seasonal floodplains and (4) occasional (or intermittent) floodplains (Gumbrecht and McCarthy, 2003).

In the Upper Panhandle zone at Mohebo, the bulk of the water is confined to the main channel but immediately downstream, the Panhandle zone broadens and the main channel is flanked by areas of permanent swamp fringed by seasonal swamp. Guma Lagoon, located in the Lower Panhandle, is in the form of an old ox-bow. Narrow channels still link the lagoon to the main channel, however it now receives water primarily from the fringing swamp. In the Maunachira channel portion of the Okavango Delta, the water spreads out into numerous smaller channels with flow-through lagoons, such as Gadikwe and Xakanaxa. The Boro River on the south-western tip of Chief's Island is a major tributary of the Okavango Delta and has active flow throughout most of the year.

The sediments building up the Okavango Delta show a stratified profile and are predominantly sands with minor silts and clays that support both shallow unconfined and deeper semi-confined aquifers (DWA, 2003) which are hydraulically interconnected at a sub-regional scale. Groundwater recharge in the Okavango Delta is dominated by floodwater infiltration whilst the role of rainfall recharge is rather limited. Wolski and Savenije (2006), during 7 years of observation of floodplain and island groundwater, recognized only two diffuse recharge events. In contrast, the arrival of the annual flood to the distal parts of the Delta caused recharge of the shallow phreatic aquifer raising the groundwater table by 1–3 m within one month. The shallow groundwater was, however, subject to evaporation and transpiration after flood recession, and thus only some part of the infiltration flux contributed to the net annual groundwater recharge. Groundwater flows in the Okavango Delta are dominated by local flow systems developed between the floodplains where recharge occurs and islands, which are the discharge zones due to evapo-transpiration. The typical groundwater table gradients observed in these flow systems suggest that 95% of groundwater flow takes place in the top 60 m of the aquifer. The effect of groundwater discharge taking place through evapo-transpiration in islands, is that the island groundwater is enriched in

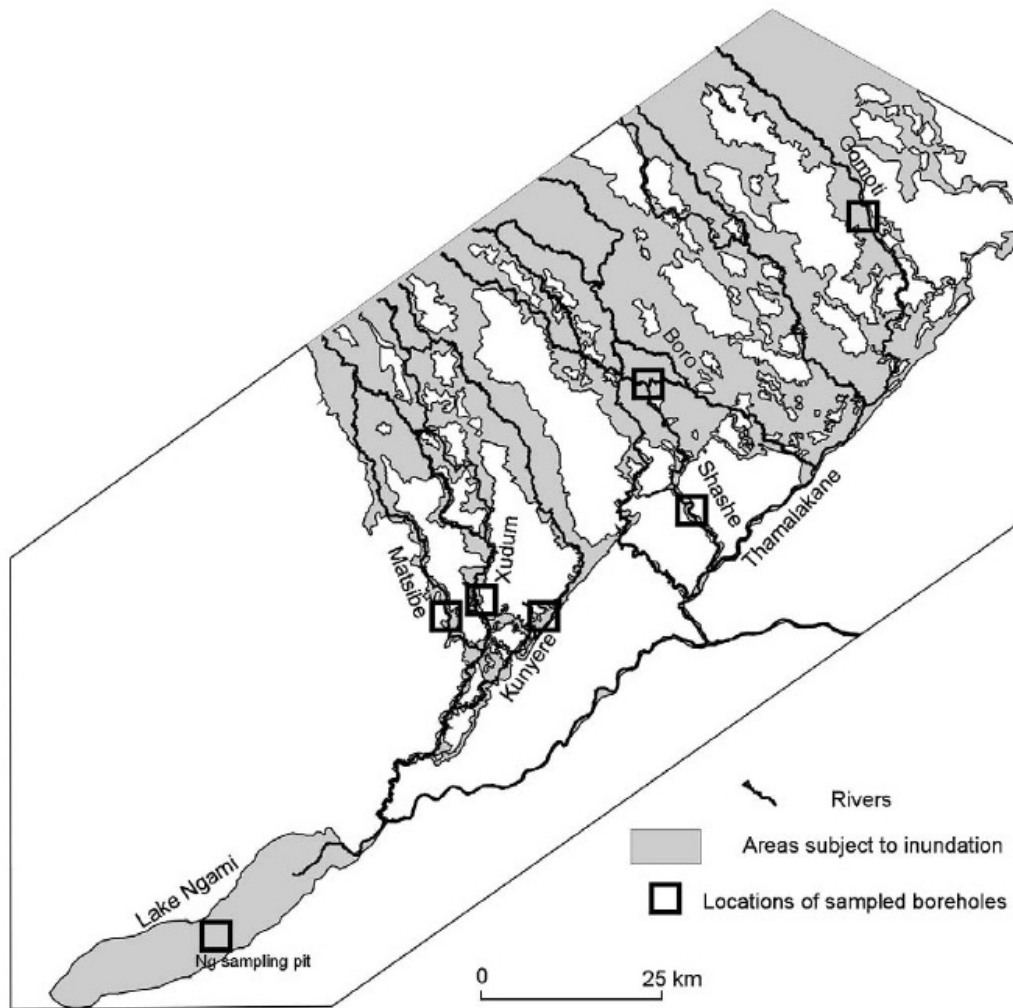


Fig. 2. Detailed map of the southern Okavango region showing the location of borehole sampling points.

dissolved salts (McCarthy and Ellery, 1994). That enrichment can reach the level where the shallow island groundwater is of much higher density than the underlying fresh groundwater, and can be subject to density-driven fingering and sinking to the bottom of the aquifer (Bauer et al., 2002).

### 3. Sampling and analytical procedures

#### 3.1. Water sampling

Nine surface water samples were collected in February 2003 as part of the Conservation International AquaRAP project (Huntsman-Mapila et al., 2005a). Surface water samples were collected from four different areas of the Okavango Delta: (1) the Upper Panhandle; (2) Guma Lagoon in the Lower Panhandle; (3) Maunachira Channel and eastern portion of

the Okavango Delta and (4) the Boro River on the south-western tip of Chief's Island (Fig. 1). Readings of pH, temperature, dissolved O<sub>2</sub> (DO) and electrical conductivity (EC) were taken in situ using a Hanna 991001 pH and temperature meter, a YSI 85 DO meter and a Hanna HI 9033 conductivity meter. Samples for metal analysis were filtered on site through a 0.45 μm cellulose nitrate filter and acidified to pH < 2 with HNO<sub>3</sub>. Samples for DOC were filtered through a Whatman glass fibre filter (GFF) and acidified with HNO<sub>3</sub>. Anion samples were not filtered. Alkalinity titrations were conducted on site. Field blanks were filtered and acidified like the samples each time the acid was renewed to ensure that there was no contamination from this process. All samples were stored in acid precleaned bottles (rinsed with 1% HNO<sub>3</sub> followed by thorough rinsing with water). Samples were stored in a cooler box

until transport to the laboratory where they were stored in a refrigerator until analysis.

Twenty groundwater samples were collected between June and November 2003 from boreholes in the occasional floodplain region of the Okavango Delta (Fig. 2). pH, temperature, DO, EC and alkalinity measurements were taken immediately. In the field, a 250 mL sample was filtered through a 0.45 µm filter and acidified with H<sub>2</sub>SO<sub>4</sub> to pH < 2. Field blanks were filtered and acidified as samples each time the acid was renewed to ensure that there was no contamination from this process. Anion samples were filtered but not acidified. All water samples were collected in HDPE bottles and refrigerated until analysis. Anion exchange columns for the speciation work were prepared in the laboratory using Bio-Rad econo-pac disposable 20 mL columns and the acetate form of Bio-Rad AG 1-X8 resin (100–200 mesh). Calibration of the packed columns using a spiked groundwater was conducted to ensure that the correct fractions were collected. 25 mL of the acidified filtered sample was passed through the column followed by 5 mL of 0.12 N HCl. This fraction was labelled as As(III). The column was then eluted with 20 mL of the same acid to collect the As(V) fraction. According to Ficklin (1990), the volume of eluent required for the removal of As(V) can be variable but elution has occurred when the dark yellow colour of the acetate form of the resin has been entirely converted to the light yellow chloride form.

### 3.2. Sediment sampling and pre-treatment

Sediments were collected at 1 m intervals as boreholes were being drilled in the occasional floodplains of the distal reaches of the Okavango Delta. A shallow core (1 m) was also collected using an Eijkelkamp Beeker sampler for undisturbed sediments from a shallow backwater lagoon near Xakanaxa Lagoon (Fig. 1). Samples from the core were collected from every cm and pore water was extracted, using a Buchner funnel, before storing the samples in plastic containers. An additional set of sediment samples was obtained from Lake Ngami, a drainage basin in the western portion of the Okavango Delta, where shallow (4 m) holes have been dug into the dry lake bed. All these sites are part of the Okavango Delta basin and receive water from the same source, the Okavango River.

Sediment samples were digested at 95 °C in 1:1 HNO<sub>3</sub> and 1:4 HCl according to US EPA method 200.9 (US EPA, 2001b). A total of 29 sediment sam-

ples were digested at the Harry Oppenheimer Okavango Research Centre (HOORC) Environmental Laboratory. In STSD-4, a stream sediment composite sample from the Geological Survey of Canada was digested in triplicate to determine recovery. Three sample blanks were also processed to ensure no contamination. A sample of the lubricant used during drilling was also digested to ensure that no contamination of the sediments had resulted from the drilling process. The samples from Lake Ngami (NG) for As analysis were digested in aqua regia at Chemex Laboratories in Canada.

### 3.3. Sample analysis

For the water samples, Ca, Mg, Fe and Mn were analysed by flame atomic absorption spectrophotometry (Varian Spectra 220). Sodium and K were analysed on a Sherwood 410 flame photometer. Anion samples were analysed on a Dionex DX-120 ion chromatograph. All analyses were conducted at the HOORC Environmental Laboratory. Water samples for As analysis were analysed on a Varian SpectrAA 220 atomic absorption spectrophotometer equipped with a Varian GTA 110 graphite furnace (GFA), a deuterium arc background correction system, and pyrolytically coated graphite tubes. A nickel nitrate solution in HNO<sub>3</sub> was used to stabilize the solution. The detection limit was 0.5 µg/L. An external reference was run every tenth sample. For surface water analyses, SLRS-4, a river water sample from the National Research Council of Canada was used (As concentration of 0.68 µg/L). Multiple injections were required for the surface water samples. For groundwater, two samples from the South African Bureau of Standards were used (As concentration of 15 and 33 µg/L) as external references. A blank and standard were run for re-sloping every 20 samples. An approximation of DOC was obtained by measuring the absorbance of the water samples at 280 nm using a Perkin Elmer Lambda 20 UV/Vis spectrophotometer (American Public Health Association (APHA) et al., 1995). Some organic compounds found in water such as lignin, tannin, humic substances and some aromatic compounds absorb ultraviolet radiation. The UV absorption is a useful approximate measure of DOC in the water of the Okavango Delta and absorbance at 280 nm in surface water has been found to give the best correlation ( $r = 0.97$ ; Mladenov et al., 2005) for water samples from the Okavango Delta.

For the analysis of As, Ni, Co and Fe in the sediments, samples were run using the GFA and STSD-4 was used as an external reference. For the samples from Lake Ngami (NG) analysed at Chemex, Co and Ni were determined by ICP-MS, As by GFA and Fe by ICP-AES. Loss on ignition (LOI) was determined on the sediments by combustion of the sample at 500 °C for 2 h. Particle size analysis of the sediments was conducted using sieving techniques (Smith and Atkinson, 1975).

## 4. Results

### 4.1. Water samples

For all geochemical data, the significance of correlation was tested at the 95% confidence level (Rollinson, 1993).

Results of the nine surface water analyses are presented in Table 1. Surface water samples in this study have low conductivity (35.9–75.2  $\mu\text{S}/\text{cm}$ ) and a near neutral to slightly acidic pH (5.53–7.24) typical of surface water from the Okavango Delta. DO values are variable (0.70–5.70 mg/L) as can be expected given the different habitats sampled (lagoons and channels). DOC in the surface water samples range between 5.5 and 16.6 mg/L. The predominant ions in the Okavango Delta water are Ca–HCO<sub>3</sub> and Ca–Na–HCO<sub>3</sub> as can be seen from the Piper Diagram (Fig. 3). Arsenic concentrations and precisions (% RSD) in these samples range from 1.1 to 3.1  $\mu\text{g}/\text{L}$  and 4.0% to 11.7%, with an average concentration and precision of 2.3  $\mu\text{g}/\text{L}$  and 6.2%, respectively. Recovery of As in the SLRS-4 reference material averaged 107%.

The results of the 20 borehole samples are presented in Table 1. In addition, results from five boreholes sampled during MGDGP have been included. Analysis of the MGDGP samples were conducted at CSIR in South Africa. Conductivity values of all samples range between 398 and 13 830  $\mu\text{S}/\text{cm}$  with an average of 1762  $\mu\text{S}/\text{cm}$ . pH values range between 6.29 and 8.60 with an average value of 7.72. DO values range between 0.81 and 4.91 with an average value of 2.06 mg/L. Sodium is the predominant cation in all groundwater samples with the exception of sample GOM16 where Ca is the predominant ion (Fig. 3). Thirteen of the boreholes have HCO<sub>3</sub> as the predominant anion (>50% meq) The remainder of the samples have variable proportions of HCO<sub>3</sub>, SO<sub>4</sub> and Cl. Sample SHM04 is a Na–SO<sub>4</sub>–Cl type water.

Arsenic values for all field blanks were less than the detection limit of 0.5  $\mu\text{g}/\text{L}$ . All As values for the groundwater samples have precisions (% RSD) of less than 10% with the exception of one sample (SHM04), which had a high background reading, with a RSD of 15%.

In samples from the 20 new boreholes, As in groundwater ranges between 3.2 to 116.6  $\mu\text{g}/\text{L}$ . Sample MAM06 with an As concentration of 97.5 was only available as an unfiltered sample. Of the 20 boreholes, six have As concentrations higher than 10  $\mu\text{g}/\text{L}$ , four have concentrations higher than 50  $\mu\text{g}/\text{L}$  and two are higher than 100  $\mu\text{g}/\text{L}$ .

For the speciation work, reliable results were only obtained for six of the boreholes tested due to difficulties experienced with calibration of the columns. Recovery of the As in the sample passed through the columns ranged between 86.7% and 136.3% with an average of 109.8%. Arsenic (III) in the samples ranged between 50.0% and 74.0% of the total As with an average of 57.1%. Arsenic (V) in the samples ranged between 29.3% and 79.0% of the total As, with an average value of 52.7%.

The data indicates a relationship between As and pH in groundwater where As concentrations greater than 50  $\mu\text{g}/\text{L}$  only occur in water at pH greater than 8.0 with the exception of sample 9904 with an As concentration of 90  $\mu\text{g}/\text{L}$  and a pH of 7.5 (Fig. 4a). This is also indicated by a significant positive correlation of As and pH in groundwater where  $r = 0.50$  ( $n = 25$ ) for groundwater. There is a relatively strong positive correlation between As and DOC (Fig. 4b) with  $r = 0.64$  ( $n = 9$ ) for surface water and  $r = 0.89$  ( $n = 19$ ) for groundwater. There is no significant correlation between As and EC (Fig. 4c) and no significant correlation between As and alkalinity (Fig. 4d), although pH and alkalinity are significantly correlated in groundwater. No correlation exists between Fe and As for surface water and groundwater samples.

### 4.2. Sediment samples

In this study, the fraction of sediments between 0.5 and 0.25 mm ( $2\Phi$ ) has been classified as medium grain sand whilst the fraction between 0.25a and 0.125 mm ( $3\Phi$ ) has been termed fine grain sand. The fraction that is <0.06 mm has been classified as “fines” (i.e., silts and clays). The samples analysed are predominantly fine to medium grain sands with the exception of KUM10-28 which has a high (>70%) amount of fines and the lake

Table 1  
Results from water quality analyses of Okavango Delta samples

Sample	UPO1 <sup>a</sup>	UP02 <sup>a</sup>	LP03 <sup>a</sup>	LP04 <sup>a</sup>	LP05 <sup>a</sup>	MC06 <sup>a</sup>	MC07 <sup>a</sup>	MC08 <sup>a</sup>	B09 <sup>a</sup>
Type <sup>b</sup>	sw	sw	sw	sw	sw	sw	sw	sw	sw
Date sampled	2-Feb-03	3-Feb-03	7-Feb-03	6-Feb-03	7-Feb-03	11-Feb-03	12-Feb-03	11-Feb-03	14-Feb-03
pH	6.55	6.45	6.5	6.62	5.53	7.09	7.2	7.24	6.67
Temperature °C	28.5	29.1	27.3	28.7	27.2	29	28.8	28.9	27.9
Conductivity (µS/cm)	35.9	39	63	49.2	51.2	51.5	51.7	49.7	75.2
DO (mg/L)	5.7	0.79	2.3	2.58	0.70	5.16	5.28	5.52	1.13
Location	U. Panhandle	U. Panhandle	L. Panhandle	L. Panhandle	L. Panhandle	Maunachira	Maunachira	Maunachira	Boro
Habitat	Lagoon	Channel	Channel	Lagoon	Channel	Channel	Lagoon	Lagoon	Channel
Ca (mg/L)	4.1	4.3	5.9	6.5	5.9	5.8	5.9	5.7	8.4
Mg	0.9	0.9	1.5	1.5	1.4	1.2	1.2	1.1	1.7
Na	2	2	3	2	3	3	3	3	5
K	2	2	3	1	3	2	2	2	3
Mn	<0.03	<0.03	<0.03	<0.03	0.03	<0.03	<0.03	<0.03	0.14
Fe	0.48	0.29	1.89	0.11	0.84	0.24	0.25	0.46	0.1
Alkalinity as CaCO <sub>3</sub>	25.6	26.8	27.7	35.4	31.7	34.2	34.2	31.7	50
Cl	0.28	0.48	0.88	0.24	0.24	0.61	0.29	0.35	0.31
SO <sub>4</sub>	0.23	0.20	0.26	0.07	0.25	0.30			0.09
NO <sub>3</sub>		0.45	0.20		0.03	0.60	0.04	0.01	0.84
DOC	5.6	5.5	14.1	6.5	7.7	8.4	11.1	16.6	13.3
As (µg/L)	2.1	2.1	2.5	1.1	2.1	2.6	2.7	2.6	3.1
% As (III)									
% As (V)									
% Recovery									
As (III)/As (v)									

<sup>a</sup> Field data from Huntsman-Mapila et al. (2005a).

<sup>b</sup> sw = surface water, gw = groundwater.

Table 1  
Results from water quality analyses of Okavango Delta samples continued

Sample	SHM01	SHM02	SHM03	SHM04	MAM05	MAM06	MAM07	KUM08	KUM09	KUM10	KUM11	KUM12	KUM13
Type <sup>a</sup>	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw
Date sampled	3-Oct-03	17-Oct-03	30-Nov-03	30-Nov-03	12-Aug-03	1-Jun-03	16-Jun-03	19-Aug-03	23-Jun-03	19-Jun-03	3-Jul-03	27-Jun-03	29-Aug-03
pH	7.94	8.45	7.18	7.22	7.5	8.19	8.46	8.06	8.07	8.53	8.21	7.64	7.04
Temperature °C	29.8	27.6	26.6	27	27.3	26.9	27.2	26.1	25.7	25.1	24.8	23.9	24.7
Cond. (µS/cm)	2230	1747	1740	3770	686	1046	948	810	13830	728	1296	471	1420
DO (mg/L)	4.91	2.38	2.45	2.95	1.85	1.96	2.01	2.85	2.14	1.95	1.54	1.88	1.84
Location	Shashe	Shashe	Shashe	Shashe	Xudum	Xudum	Xudum	Matsibe	Matsibe	Matsibe	Matsibe	Kunyere	Kunyere
Habitat													
Ca (mg/L)	36.6	5.0	22.0	84.5	0.6		3	1.4	83.6	2.3	1.6	16.3	56.6
Mg	12.1	0.8	8.0	63.3	0.1		0.3	0.3	75.8	0.2	0.2	5.7	19.1
Na	440	390	350	720	190		270	230	1680	220	310	100	230
K	14	15	14	20	4		9	9	48	5	6	10	30
Mn	<0.03	<0.03	<0.03	<0.03	<0.03		<0.03	<0.03	0.17	<0.03	<0.03	<0.03	0.27
Fe	0.14				0.94		1.16	1.1		1.49	0.38	0.7	
Alk	503	516	488	486	417		425	430	1026	437	543	264	632
Cl	380.1	158.4	224.5	419.0	1.2		11.0	4.5	1596.7	9.9	56.9	165.3	85.9
SO <sub>4</sub>	411.5	345.7	203.9	1162.0	4.6		38.0	24.2	666.4	31.3	138.8	318.8	59.4
NO <sub>3</sub>			0.4		0.5			0.2	4.7	0.3	0.1		
DOC	3.2	2.8	3.6	2.6	5.5		19.1	4.3	8.6	13.8	5.6	5.3	5.7
As (µg/L)	4.2	4.0	5.2	14.6	8.3	97.5 <sup>b</sup>	116.6	2.7	58.3	112.0	9.1	6.6	3.9
% As (III)			50.8	57.3									
%As (V)			44.1	29.3									
% Recovery			94.8	86.7									
As (III)/As (v)			1.2	1.95									

<sup>a</sup> sw = surface water, gw = groundwater.

<sup>b</sup> Result from unfiltered sample.



Table 1  
Results from water quality analyses of Okavango Delta samples continued

Sample	KUM14	KUM15	GOM16	GOM17	GOM18	GOM19	GOM20	9830 <sup>a</sup>	9834 <sup>a</sup>	9837 <sup>a</sup>	9904 <sup>a</sup>	9906 <sup>a</sup>
Type <sup>b</sup>	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw	gw
Date sampled	3-Jul-03	3-Sep-03	4-Nov-03	20-Nov-03	12-Nov-03	12-Nov-03	30-Nov-03	15-Feb-03	8/27/02	9/2/02	12/18/02	1/26/03
pH	7.44	6.78	6.29	7.49	7.39	7.51	6.81	8.1	8.5	8.6	7.5	8.1
Temperature °C	24.4	26.5	33.8	28.3	29.5	27.9	26.7	25.3	24.3	25.4	26.5	28
Conductivity (µS/cm)	725	809	398	1394	483	1025	542	1560	1457	1668	1340	1930
DO (mg/L)	2.43	1.67	0.81	1.22	1.02	1.59	1.57	1.7			1.25	3.5
Location	Kunyere	Kunyere	Gomoti	Gomoti	Gomoti	Gomoti	Gomoti	Boro	Boro	Boro	Gomoti	Gomoti
Habitat												
Ca (mg/L)	17.7	7.5	33.9	2.7	1.1	3.4	9.5	2.8	2.9	7.6	1.9	3.3
Mg	4.4	1.0	9.0	0.3	0.2	0.3	1.0	0.72	0.75	1.7	0.37	0.71
Na	150	170	28	300	170	260	130	454	392	390	370	404
K	10	10	8	3	2	10	12	6.6	14	9.3	5.2	4.2
Mn	<0.03	<0.03	0.19	<0.03	<0.03	<0.03	<0.03	<0.05	<0.05	<0.05	<0.05	<0.05
Fe	0.32	0.39	13.22		1.32	1.83		0.07	0.22	0.19	0.08	0.75
Alk	212	210	217	353	219	518	297	693	711	607	624	375
Cl	19.4	98.6	2.4	175.7	19.8	7.9	2.5	137.0	80.0	139.0	64.0	238.0
SO <sub>4</sub>	11.0	67.7	19.1	191.6	24.2	50.1	10.5	94.0	50.0	73.0	85.0	196.0
NO <sub>3</sub>	0.1	0.2	0.3	0.2	0.1	0.1	0.1	<0.1	<0.1	<0.1	<0.1	<0.1
DOC	3.5	3.6	10.4	3.1	4.4	4.6	2.2					
As (µg/L)	1.8	2.4	7.0	10.1	8.0	6.1	4.7	30	35.0	29.0	90	60
% As (III)			53.5	74.0	57.3	50.0						
% As (V)			56.7	48.3	79.0	58.6						
% Recovery			110.3	122.4	136.3	108.6						
As (III)/As (v)			0.94	1.53	0.73	0.85						

<sup>a</sup> Borehole data from MGD (DWA, 2003).

<sup>b</sup> sw = surface water, gw = groundwater.

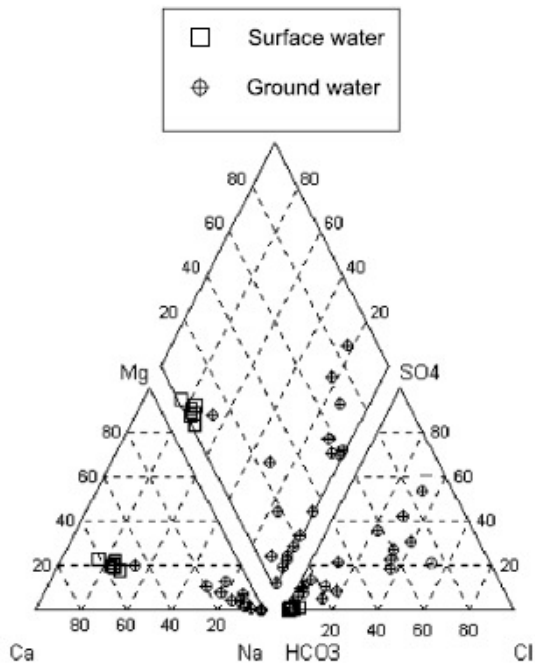


Fig. 3. Piper diagram for surface and ground water samples.

sediment samples from Lake Ngami, which also have a higher percentage of fines than the samples from the Okavango Delta proper. There was insuf-

ficient sample from the Xak core to conduct particle size analyses.

The sediment sample STSD-4 that was analysed in triplicate for As had an average recovery of 100.7%. The precision of the sediment sample data is <13% RSD for all samples with an average of 6.5%. Arsenic concentrations in the sediments analysed range between 0.2 and 7.0 mg/kg with an average value of 2.2 mg/kg (Table 2). Values are highest in the samples from KUM10, which also had elevated As in the water sample. A significant positive correlation exists between As and Co and between As and Fe (Fig. 5a and b) in the sediment samples (both  $r = 0.54$ ,  $n = 35$ ). However, the samples from the shallow core, Xak, do not plot with the other samples. With Ni, there is no clear relationship with As. There is a relatively strong correlation between As and the % fines in the samples ( $r = 0.64$ ,  $n = 25$ ) and between Fe and the % fines ( $r = 0.65$ ,  $n = 18$ ) and to a lesser extent between Co and the % fines ( $r = 0.30$ ,  $n = 25$ ; Fig. 5c–e).

A strong positive correlation exists between LOI and As (Fig. 5f) for the samples from KUM10 and MAM05 ( $r = 0.96$ ,  $n = 21$ ) and Xak, although plotting separately from the other data points has

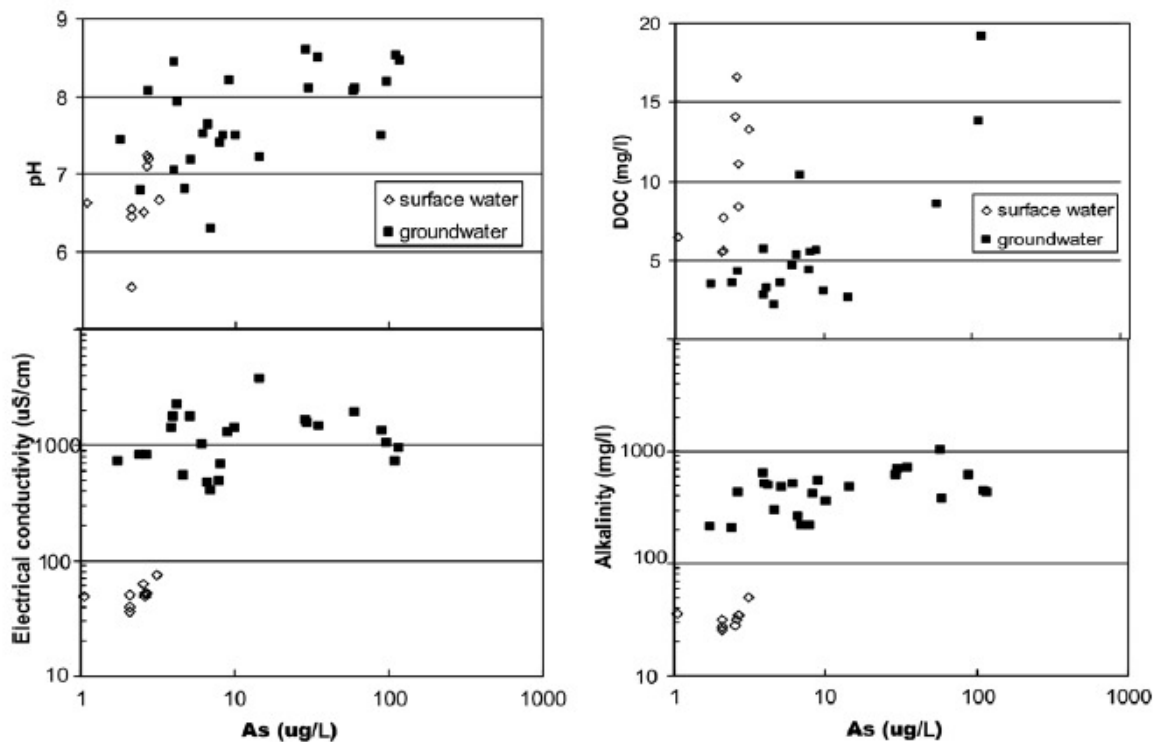


Fig. 4. Binary diagrams for Okavango Delta surface and groundwater compositions.

Table 2  
Results of sediment sample analysis of the Okavango Delta sediments

Sample	Sample depth (m)	As (mg/kg)	Co (mg/kg)	Ni (mg/kg)	Fe (%)	LOIorgC %	% medium sand	% fine sand	% fines
MAM05-2	2	2.6	8.2	39.5	0.40	4.9	27.4	12.3	4.6
MAM05-7	7	1.5	5.0	38.8	0.45	1.7	21.9	14.2	1.3
MAM05-13	13	2.6	7.1	92.4	0.53	4.5	23.3	8.2	9.9
MAM05-22	22	0.8	2.0	43.4	0.19	1.4	12.5	6.7	2.2
MAM05-33	33	1.2	5.8	42.8	0.26	1.5	41.0	20.6	6.6
MAM05-42	42	2.4	4.9	37.3	0.49	2.8	21.1	13.1	15.9
MAM05-63	63	0.4	3.9	50.4	0.24	0.1	51.1	38.1	0.2
KUM10-5	5	0.4	4.2	42.8	<0.05	0.3	51.6	29.3	0.2
KUM10-13	13	3.3	9.2	40.2	0.49	4.4	31.2	33.9	10.4
KUM10-17	17	3.7	7.6	40.0	0.57	5.2	21.6	20.1	32.3
KUM10-28	28	6.2	6.0	38.4	0.71	8.6	1.3	1.7	70.5
KUM10-29	29	3.6	5.8	32.1	0.44	5.0	17.6	5.7	16.1
KUM10-30	30	1.8	6.4	42.5	0.43	1.9	26.0	18.8	2.2
KUM10-32	32	2.3	6.6	44.4	0.39	2.0	24.3	14.9	4.9
KUM10-34	34	2.1	4.8	39.1	0.34	1.7	38.5	11.6	34.5
KUM10-47	47	1.4	5.6	36.1	0.44	2.3	30.7	10.8	10.7
KUM10-48	48	1.1	3.8	35.0	0.44	1.7	32.6	8.7	4.8
KUM10-49	49	0.9	2.6	32.6	0.37	1.3	15.0	21.1	10.5
KUM10-62	62	1.3	6.5	39.6	<0.05	1.7	51.9	15.0	3.7
KUM10-64	64	1.0	3.8	36.2	0.54	1.2	29.5	9.0	17.0
KUM10-65	65	0.9	2.8	62.3	0.42	1.3			
XAK-2	0.02	1.3	6.7	90.5	0.35	45.0			
XAK-7	0.07	1.2	6.7	83.5	0.72	30.0			
XAK-11	0.11	0.7	6.1	75.2	0.67	26.8			
XAK-18	0.18	0.9	3.2	64.3	0.53	14.2			
XAK-21	0.21	0.9	3.8	71.7	0.42	23.1			
XAK-24	0.24	0.8	1.8	80.6	0.52	21.8			
XAK-30	0.3	0.9	0.3	56.9	0.54	6.9			
XAK-49	0.49	0.2	0.4	58.4	0.35	0.5			
NG210 <sup>a</sup>	2.1	4	13.5	29	1.32	11.3	6.5	7.8	20.1
NG260	2.6	4	9.5	22	1.39	10.3	5.9	22.5	31.2
NG430	4.3	7	4.2	12	0.52	2.8	32.8	10.5	14.0
NG440	4.4	6	8.7	21	1.07	7.5	35.0	13.1	26.9
NG450	4.5	7	10.1	23	1.33	10.5			
NG460	4.6	2	11.7	25	1.44	10.8	28.0	25.8	27.9

<sup>a</sup> Co, Ni and Fe values for NG samples from Huntsman-Mapila et al. (2006).

$r = 0.78$  ( $n = 8$ ). There is no correlation between As and LOI for the NG samples.

## 5. Discussion

### 5.1. General hydrochemistry

The Okavango Delta surface water has a low conductivity with the major cation being Ca and the major anion being  $\text{HCO}_3$  (Hutton and Dincer, 1976; Sawula and Martins, 1991; Cronberg et al., 1995) with exceptions occurring in small saline pans on islands where the groundwater has come to the surface during the seasonal flood and experienced evaporative concentration. The surface water of the Delta has a typical tea colour from dissolved organic

compounds, in particular fulvic acids which are present in the water (Mladenov et al., 2005). The conductivity of the outflow of the Delta in Maun is typically double that of the inflow in the Panhandle due to evaporative concentration. This  $\text{HCO}_3$ -dominant fresh surface water dominates the ground-water recharge during the advance of the seasonal flood. McCarthy et al. (1998) plotted ground water chemistry of 251 boreholes and found that  $\text{HCO}_3$ -dominant water is confined to the area of the Delta proper whilst Cl-dominant water predominates in the region outside of the Delta. Three apparently isolated pockets of  $\text{SO}_4$ -dominant water occur in the western fringe of the Delta. These authors concluded that neither the Cl- nor the  $\text{SO}_4$ -dominant water could have originated from recharge from the Okavango Delta

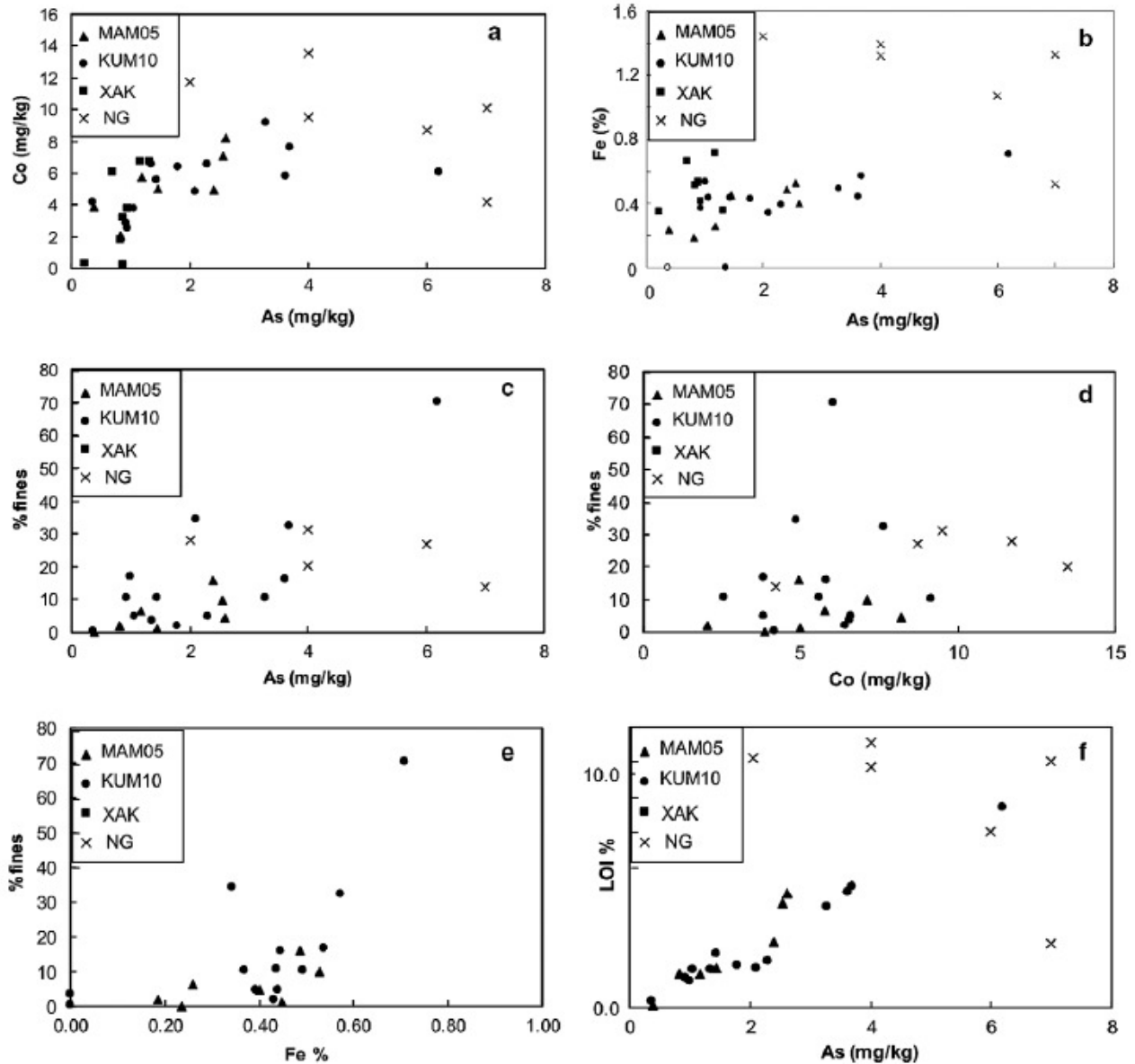


Fig. 5. Binary diagrams for the Okavango Delta sediment compositions.

surface water. Several studies have shown that lower salinity water frequently overlies water of higher salinity (Aquatec, 1982; BRGM, 1986; DWA, 2003; Bauer et al., 2002). In addition, salinity is spatially very variable, and can change by up to 3 orders of magnitude over a horizontal distance of a few tens of meters (McCarthy et al., 1993b).

### 5.2. Nature and source of surface water arsenic

The As concentrations in the surface water of the Okavango Delta exceed the global average value of

1.7  $\mu\text{g/L}$  for dissolved As in stream water (Martin and Whitfield, 1983) in all but one sample. The neutral to slightly acidic pH of the Delta water would suggest that the  $\text{H}_2\text{AsO}_4^-$  inorganic species predominates. The relatively high DOC of the surface water would also suggest the presence of the methylated form of As.

Dowling et al. (2002) suggest that dissolved As in the Bengal Basin is carried by the rivers and then removed by either flocculation of fine grained particles or bacterially reduced and precipitated to be stored in deltaic sediments. The suspended sediment

component in the Okavango Delta needs to be assessed. The particulate load in the Okavango Delta is however very low compared to the chemical precipitates. McCarthy et al. (2002) estimated that, under present conditions, 381,000 t of chemical sediment are accumulating on the fan each year compared to approximately 209,000 t of clastic load consisting of 170,000 t of bedload and 39,000 t of suspended load.

Slightly elevated As levels, compared to the global average, are occurring in the Okavango Delta surface water and possibly originate from the weathering of the bedrock in the region which comprises Proterozoic granitoids and mafic-ultramafic rocks (Huntsman-Mapila et al., 2005b). No As values are available for these source rocks. Koljonen (1992, quoted in Matschullat, 2000) reports that gabbros and basalts have an average concentration of 0.7 mg/kg and granites and granodiorites have an average As concentration of 3 mg/kg. Sulphide minerals in mineralised bedrock are vulnerable to attack in humid environments rich in  $O_2$  despite being relatively insoluble in pure water. The free  $O_2$  in the water causes the oxidation of pyrite and arsenopyrite and liberates  $H_2SO_4$ . Under these acid conditions, As is highly mobile.

The transport of As from source rocks in the Okavango Delta probably involves As being stored in various geochemical environments while in transit. This may include the short-lived Fe-monosulphide rich sediment which could accumulate in the system during wet periods.

### 5.3. Nature and source of sediment arsenic

Sediment extraction work in this study reveals As concentrations of 0.2–7 mg/kg. The Delta sediments are predominantly fine to medium grain sands and the average As concentration for sandstones is given as 0.5 mg/kg (Onishi, 1969). Sediment samples from the Ganges–Brahmaputra–Meghna (GBM) Delta range between 0.5 and 17.7 mg/kg (Ahmed et al., 2004). Sengupta et al. (2004), in their study on As in clay and sand sediments in the Bengal Delta, recorded 10 and 4 mg/kg, respectively.

The collection of the shallow core in Xakanaxa Lagoon was of interest as the core was taken in an active backwater swamp, where a vigorous release of gas was observed when the corer was placed in the organic rich sediments. This suggested a reducing environment perhaps comparable to the environment where the layers of the more fine grained/

organic rich sediments of the occasional floodplain were deposited in the past when this region was inundated with water (Huntsman-Mapila et al., 2006). It is thought that in the past, during periods of higher rainfall than today, water would have ponded up against the Thamalakane fault, resulting in the formation of what is referred to as Lake Thamalakane (Shaw, 1988).

It was hypothesized that the Xakanaxa lagoon site might represent a typical environment for active As deposition. In reducing environments, arsenite in the form of  $H_3AsO_3^0$  is present at pH values less than 9.2 (Sracek et al., 2004) and a pH of about 7.0 is the adsorption optimum for arsenite. In these conditions, As can diffuse to adsorption sites on  $Fe(OH)_3$  surfaces. In a reducing environment, the electron donor for the reduction of Fe(III) oxides and hydroxides can be organic matter in sediments.

Other potential adsorbents of As include Al oxides and hydroxides (Stollenwerk, 2003) with halloysite and chlorite found to adsorb As(V) to a much greater extent than kaolinite, illite and montmorillonite (Lin and Puls, 2000). XRD work on the Okavango sediments (Huntsman-Mapila et al., 2005b) has shown that kaolinite is the predominant clay mineral in the Okavango sediments present at no more than 3% for the samples analysed. It is therefore suggested that adsorption on Al oxides and hydroxides does not play a major role in fixing As in the sediments.

An earlier study of the sediments of the Okavango Delta revealed that in 90 samples analysed, the average S concentration was 0.04% with a range of <0.01–0.52%. The same samples had an average organic C content of 0.40% with a range of <0.01–1.64% (Huntsman-Mapila, unpublished data). This would suggest that organic C contributes to a much greater extent than S to the LOI of the sediments in the Delta. The strong positive correlation between As and LOI in this study suggests that the combustible elements of the sample play an important role in the adsorption of As in the Okavango Delta sediments.

### 5.4. Nature and source of groundwater arsenic

The occurrence of elevated As in the groundwater of the Okavango Delta is not attributed to any anthropogenic activities as minimal agricultural activities occur in the region. The current study suggests a natural source and the release in groundwater through natural processes. Oxidation of arsenopyrite due to a lowering of the groundwater table

was postulated as the dominant process for As mobilisation in the Ganges–Brahmaputra–Meghna (GBM) Delta (Bhattacharya et al., 1997; Nickson et al., 2000). However, recent studies (McArthur et al., 2001; Dowling et al., 2002; Anawar et al., 2003; Ahmed et al., 2004) have indicated that the reduction and dissolution of Fe oxy-hydroxide coating on sand grains is another likely release mechanism. This is coupled to the degradation of organic matter in sediments whereby organic matter facilitates the transformation of Fe(III) to Fe(II) and As(V) to As(III). Because dissolved As, Fe and  $\text{HCO}_3^-$  are positively correlated in some GBM ground waters and because organic C is abundant in the GBM sediments, reduction of Fe oxides and release of As may be coupled with organic C oxidation (Nickson et al., 2000).

Results from the MGDGP (DWA, 2003) indicate that there is a higher occurrence of elevated As in boreholes screened below 72.6 mbgl than boreholes screened between 27.7 and 50.3 mbgl suggesting that elevated As levels are predominantly occurring in the deeper aquifers. This would suggest that the oxidation of sulphide minerals due to lowering of the groundwater table is not a probable mechanism in this case.

Recent studies have shown that certain Fe reducing bacteria can use humic substances as electron acceptors and reduce amorphous Fe(III) oxide and crystalline Fe oxides producing siderite, in  $\text{HCO}_3^-$  buffered solutions (Little et al., 1997; Dong et al., 2000; Sengupta et al., 2004). There is a significant correlation between As and Fe in the Okavango sediments and between As and organic matter in both the sediments and groundwater. The oxidative degradation of DOC in the groundwater can produce high  $\text{HCO}_3^-$  and create a favourable environment for the reduction of Fe and As. This hypothesis would suggest that the mechanism for the mobilisation of As into the Okavango Delta groundwater is similar to the processes occurring in the GBM Delta.

However, no correlation exists between As and Fe in the Okavango Delta groundwater. Zheng et al. (2004) found similar results in Bangladesh, where not all As-rich waters had elevated dissolved Fe. They suggest a “re-oxidation scenario” whereby the introduction of  $\text{O}_2$  to  $\text{SO}_4$ -reducing ground water can induce the oxidation of Fe(II) and As(III). This re-oxidation can lead to As and  $\text{PO}_4$  co-precipitation or sorption onto newly formed Fe-oxyhydroxide. Phosphate analyses were not con-

ducted on the Okavango Delta samples so it is not possible to determine if a correlation exists between As and  $\text{PO}_4$ . This requires further investigation.

## 6. Conclusions

From this preliminary study of the occurrence of naturally occurring As in the sediments and water of the Okavango Delta, it has been found that:

- Surface water in the Okavango Delta is slightly enriched in As when compared to a global value for stream waters. The water is neutral to slightly acidic with a high DOC.
- Of the 20 new borehole analyses from this study, six were found to have values exceeding  $10 \mu\text{g/L}$ , the current provisional guideline WHO value for As (WHO, 2001).
- There is an indication that As(III) is slightly more predominant than As(V) in groundwater.
- Reductive dissolution of Fe oxides and hydroxides in the sediments using organic C as an electron acceptor is a likely mechanism for the release of As from the sediments into the groundwater. Further work is required in order to understand the mechanisms involved in the release of As into the ground water of the Okavango Delta.

The quantity of available water for supply to Maun was significantly increased through the drilling of the MGDGP boreholes. It is evident that to address water shortages and associated problems, significant emphasis has to be given to water quality. Non-selective continual incrementing of quantity could lead to increased deterioration of water quality in Maun’s water supply.

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