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Speciation of heavy metals in water from the Uganda side of Lake Victoria

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Different forms of copper Cu, zinc Zn, lead Pb and cadmium Cd in water from the Uganda side of Lake Victoria (25°C, pH 6.75–7.18), the second largest inland freshwater lake in the world, have been studied using ion-exchange, dialysis and atomic absorption spectrophotometry. The results indicate that heavy metals Cu, Zn, Pb and Cd are present mainly in the cationic form (80–83%). Small quantities of anionic (13–22%), non-ionic, dialyzable (4–8%), and non-ionic, non-dialyzable (< 1.3–4.4%) forms were also detected for all metals except Cd. The corresponding concentrations lay in the ranges: cationic, 0.06–0.99; anionic, < 0.001–0.25; non-ionic, dialyzable, < 0.001–0.08; non-ionic, non-dialyzable, < 0.001–0.06 $\mu\text{g ml}^{-1}$. The existence of the metals in non-ionic and non-dialyzable forms is attributable to metal associations with high relative molecular mass (RMM) organic matters.

Keywords: Heavy metals; Speciation; Dialysis; Ion-exchange; Lake Victoria; Atomic absorption spectrophotometry

1. Introduction

With ever-increasing population, migration to urban centres and increased industrialisation, the environment, in particular the water systems [1,2], have been considerably polluted. Many streams and rivers that are an important means of livelihood for numerous communities have been converted into slow-moving, polluted drains. The root cause is man's tendency to dispose of untreated waste directly into the catchment drainage system instead of removing it at the source. In polluted waters, the toxicity of heavy metals both to man and aquatic life depends on their physico-chemical forms. The determination of such forms, usually called speciation, is necessary for a better understanding of the level of toxicity, bioavailability, bioaccumulation and transport of a particular element in a given environmental medium.

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Lake Victoria in East Africa (figure 1) is the second largest inland freshwater lake in the world. The lake is the main source of domestic and industrial water for the surrounding towns and cities such as Jinja and Kampala. The lake receives a wide variety of pollutants, from industrial effluent, untreated sewage and run-offs from surrounding cities and townships into drainage systems that empty into it. Heavy metals found in such effluent may accumulate in aquatic life and reach toxic levels in man via food chains [3,4]. The metals are extremely resistant to changes that would otherwise lead to their partial or complete elimination from the environment. They undergo biogeochemical cycles, their potential toxicity being largely controlled by their physico-chemical modifications. They have a tendency to accumulate in vital human organs such as the brain, kidney, liver, intestinal tract and lungs. They may cause structural damage and/or cell malfunctioning, owing to their interaction with nucleic acids. Heavy metal toxicity may also adversely affect the genetic carrier code via nucleic acid-chelate formation [5].

It is well known that the biological effects of a number of heavy metals depend on the speciation of the metals in a given aqueous environment [3]. Speciation analyses are of great importance for a better understanding of the fate of the elements in aqueous media [6–10]. In addition, speciation data are very helpful in the formulation of water-quality criteria [11]. There are a number of speciation laboratory techniques for heavy metals in natural waters. These include electro-analysis [11], ion-exchange [12–15], dialysis [15–17], solvent extraction [18] and atomic absorption spectrophotometry [15,19]. A combination of ion-exchange, dialysis and atomic absorption spectrophotometric techniques has been applied to the speciation of trace metals in river waters [15].

The aim of the present study therefore was to establish the distribution patterns of Cu, Zn, Pb and Cd in the waters of Lake Victoria at Jinja (latitude 00' 25" N, longitude 35' 12" E), a major industrial area in Uganda, located at the source of the River Nile.

2. Materials and methods

2.1. Water sampling

Water samples were collected within 1 metre of the surface at 12 different sites (W1–W12), separated from each other by at least 800 metres, from Lake Victoria at Jinja (figure 1), using 20-litre plastic containers. The containers were cleaned and rinsed several times with the lake

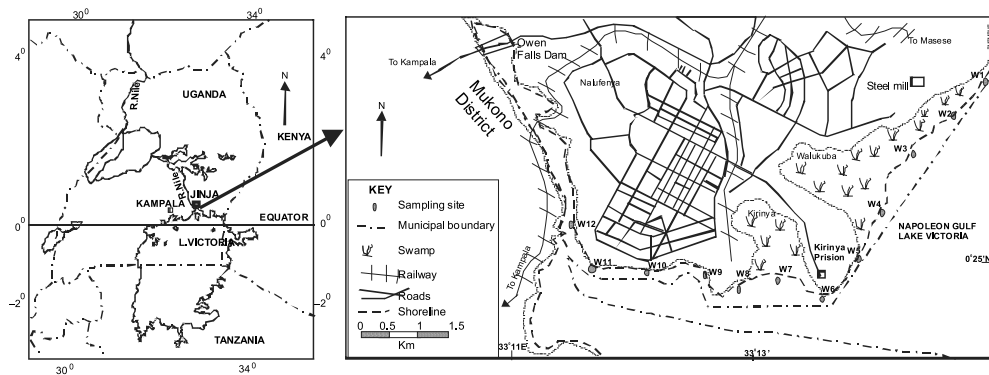


Figure 1. Schematic map of Jinja Municipality showing studied and sampling area.

water before use. The water was filtered within a few hours of sampling, transferred to 5-litre polythene containers and stored at room temperature (25°C) until analysis.

2.2. Analytical procedures

At the time of analysis, 1 litre of the water samples from each site was evaporated to dryness. To the residue, the triple acid system, *viz.* concentrated nitric acid (10 ml), perchloric acid (2 ml) and hydrofluoric acid (4 ml), was added. Then this was reheated to dryness. The final residue was reconstituted in 2 ml of 2 M hydrochloric acid, transferred to a 25 ml volumetric flask and made up to the mark with distilled water. The solution was then analysed using an atomic absorption spectrophotometer (Perkin-Elmer, Model 2380) using the Paul and Pillai method [15]. This gave the total metal concentration in the sample.

To obtain the concentration of all the cationic species, 2 litres of the water sample were passed through a cation-exchange resin [Amberlite CG-120 (Na), chromatographic grade, type 1, 100–200 mesh, conditioned with 1 M HCl]. The eluate was processed as before and analysed using the atomic absorption spectrophotometer (AAS). This gave the concentration of all the anionic and non-ionic species. The concentration of the cationic species was obtained as the difference between the total metal concentration and the concentration for both the anionic and non-ionic species.

The concentration of the anionic species was obtained by passing 2 litres of the freshwater sample through an anion-exchange resin [Amberlite IRA-400 (Cl), standard grade, conditioned with 1 M NaOH]. The resulting eluate was treated and analysed as before. This gave the total concentration of all the cationic and non-ionic modifications. The difference between these results and those of the total metal concentration gave the concentration of the anionic form.

The resins were periodically regenerated. The concentration of the total non-ionic, or neutral, species was obtained by successively passing 4 litres of the water sample through the cation resin and the anion resin, followed by digestion and analysis of the eluate.

The concentration of the non-ionic, non-dialysable forms was obtained as follows. Dialysis was performed on 4 litres of the eluate (obtained after successive cation and anion exchange) by immersing the dialyser tubes (Arthur Thomas and Co; 5 cm, dia., 4.8 nm pore size), containing the test water, into de-ionised water which was periodically changed for 24 hours. The final dialysed solution was processed as before, and analysed using the AAS.

3. Results and discussion

The pH of the samples at 25°C was in the range 7.0 ± 0.2 , indicative of the relative neutrality of the lake waters. Solubilisation of heavy metals was detected in the water as cationic, anionic, non-ionic (dialysable) and non-ionic (non-dialysable) modifications. Distribution among the different forms or modifications varied widely according to the nature of the metal. Table 1a shows the total sum of heavy metal composition of the filtered water samples as analysed through the digestion procedure. The total concentration, in $\mu\text{g ml}^{-1}$, of the metals in all the samples decreased in the order: Zn > Pb > Cu >> Cd. The results obtained for the metals after cationic and anionic exchange are shown in tables 1b and 1c, respectively, while table 1d displays the data after successive cation and anion exchange. Table 1e shows the results obtained for non-ionic, non-dialysable forms. The fairly good agreement (table 2)

Table 1. Concentrations of heavy metals obtained as in Procedures (a) to (e)

Site	Concentration ($\mu\text{g ml}^{-1}$)				Concentration ($\mu\text{g ml}^{-1}$)				Concentration ($\mu\text{g ml}^{-1}$)			
	Cu	Zn	Pb	Cd	Cu	Zn	Pb	Cd	Cu	Zn	Pb	Cd
	(a) Total in filtered water sample											
W1	0.99±0.01	1.23±0.06	1.00±0.01	0.090±0.001	0.88±0.03	1.08±0.07	0.75±0.03	< 0.001	0.04	0.06	0.01	< 0.001
W2	0.97±0.03	1.22±0.05	1.00±0.02	0.081±0.002	0.86±0.01	1.05±0.06	0.70±0.01	< 0.001	0.04	0.04	0.02	< 0.001
W3	0.99±0.01	1.23±0.06	0.90±0.02	0.088±0.001	0.87±0.02	1.03±0.06	0.78±0.02	< 0.001	0.01	0.05	0.02	< 0.001
W4	0.98±0.01	1.20±0.07	0.90±0.03	0.072±0.002	0.86±0.03	1.07±0.05	0.70±0.04	< 0.001	0.03	0.05	0.01	< 0.001
W5	0.98±0.01	1.21±0.06	0.80±0.01	0.077±0.003	0.88±0.04	1.04±0.07	0.76±0.01	< 0.001	0.04	0.06	0.01	< 0.001
W6	0.96±0.04	1.20±0.07	0.90±0.02	0.091±0.001	0.87±0.01	1.06±0.05	0.77±0.04	< 0.001	0.03	0.04	0.02	< 0.001
W7	0.98±0.02	1.22±0.05	1.00±0.02	0.072±0.001	0.88±0.01	1.05±0.07	0.72±0.02	< 0.001	0.02	0.06	0.03	< 0.001
W8	0.99±0.01	1.23±0.04	1.00±0.01	0.081±0.002	0.85±0.03	1.06±0.02	0.73±0.04	< 0.001	0.01	0.05	0.02	< 0.001
W9	0.97±0.02	1.21±0.03	1.00±0.02	0.082±0.001	0.88±0.02	1.05±0.05	0.75±0.04	< 0.001	0.03	0.06	0.03	< 0.001
W10	0.98±0.02	1.22±0.03	0.90±0.04	0.080±0.003	0.86±0.03	1.04±0.07	0.74±0.05	< 0.001	0.04	0.04	0.02	< 0.001
W11	0.98±0.02	1.22±0.04	1.00±0.03	0.071±0.001	0.87±0.02	1.08±0.04	0.72±0.06	< 0.001	0.03	0.07	0.01	< 0.001
W12	0.97±0.03	1.20±0.05	0.90±0.02	0.068±0.002	0.88±0.01	1.02±0.05	0.69±0.01	< 0.001	0.02	0.05	0.02	< 0.001
Average	0.978 ± 0.019	1.216 ± 0.051	0.942 ± 0.021	0.079 ± 0.002	0.799 ± 0.042	1.053 ± 0.009	0.734 ± 0.016	< 0.001	0.028 ± 0.006	0.053 ± 0.005	0.018 ± 0.004	< 0.001
	(b) In eluate after cation exchange											
W1	0.21±0.02	0.27±0.04	0.14±0.02	0.021±0.002	0.09	0.10	0.10	< 0.001				
W2	0.20±0.03	0.27±0.03	0.15±0.01	0.009±0.004	0.09	0.12	0.10	< 0.001				
W3	0.19±0.03	0.19±0.03	0.17±0.01	0.022±0.003	0.08	0.09	0.09	< 0.001				
W4	0.19±0.03	0.25±0.02	0.16±0.02	0.022±0.002	0.10	0.09	0.10	< 0.001				
W5	0.19±0.01	0.23±0.04	0.15±0.01	0.011±0.003	0.10	0.10	0.09	< 0.001				
W6	0.20±0.01	0.25±0.03	0.16±0.02	0.009±0.002	0.09	0.12	0.10	< 0.001				
W7	0.21±0.02	0.22±0.05	0.17±0.01	0.020±0.003	0.09	0.11	0.09	< 0.001				
W8	0.20±0.02	0.20±0.01	0.17±0.02	0.011±0.002	0.10	0.10	0.09	< 0.001				
W9	0.19±0.01	0.21±0.02	0.16±0.03	0.008±0.002	0.08	0.09	0.10	< 0.001				
W10	0.19±0.02	0.28±0.01	0.17±0.01	0.021±0.002	0.08	0.08	0.10	< 0.001				
W11	0.20±0.03	0.26±0.02	0.18±0.02	0.012±0.003	0.09	0.09	0.10	< 0.001				
W12	0.21±0.04	0.28±0.02	0.13±0.03	0.007±0.002	0.10	0.10	0.09	< 0.001				
Average	0.198±0.004	0.248±0.016	0.159±0.006	0.014±0.003	0.091±0.004	0.099±0.007	0.096±0.003	< 0.001				
	(c) In eluate after anion exchange											
W1	0.21±0.02	0.27±0.04	0.14±0.02	0.021±0.002	0.09	0.10	0.10	< 0.001				
W2	0.20±0.03	0.27±0.03	0.15±0.01	0.009±0.004	0.09	0.12	0.10	< 0.001				
W3	0.19±0.03	0.19±0.03	0.17±0.01	0.022±0.003	0.08	0.09	0.09	< 0.001				
W4	0.19±0.03	0.25±0.02	0.16±0.02	0.022±0.002	0.10	0.09	0.10	< 0.001				
W5	0.19±0.01	0.23±0.04	0.15±0.01	0.011±0.003	0.10	0.10	0.09	< 0.001				
W6	0.20±0.01	0.25±0.03	0.16±0.02	0.009±0.002	0.09	0.12	0.10	< 0.001				
W7	0.21±0.02	0.22±0.05	0.17±0.01	0.020±0.003	0.09	0.11	0.09	< 0.001				
W8	0.20±0.02	0.20±0.01	0.17±0.02	0.011±0.002	0.10	0.10	0.09	< 0.001				
W9	0.19±0.01	0.21±0.02	0.16±0.03	0.008±0.002	0.08	0.09	0.10	< 0.001				
W10	0.19±0.02	0.28±0.01	0.17±0.01	0.021±0.002	0.08	0.08	0.10	< 0.001				
W11	0.20±0.03	0.26±0.02	0.18±0.02	0.012±0.003	0.09	0.09	0.10	< 0.001				
W12	0.21±0.04	0.28±0.02	0.13±0.03	0.007±0.002	0.10	0.10	0.09	< 0.001				
Average	0.198±0.004	0.248±0.016	0.159±0.006	0.014±0.003	0.091±0.004	0.099±0.007	0.096±0.003	< 0.001				
	(d) In eluate after successive cation and anion exchange											
W1	0.21±0.02	0.27±0.04	0.14±0.02	0.021±0.002	0.09	0.10	0.10	< 0.001				
W2	0.20±0.03	0.27±0.03	0.15±0.01	0.009±0.004	0.09	0.12	0.10	< 0.001				
W3	0.19±0.03	0.19±0.03	0.17±0.01	0.022±0.003	0.08	0.09	0.09	< 0.001				
W4	0.19±0.03	0.25±0.02	0.16±0.02	0.022±0.002	0.10	0.09	0.10	< 0.001				
W5	0.19±0.01	0.23±0.04	0.15±0.01	0.011±0.003	0.10	0.10	0.09	< 0.001				
W6	0.20±0.01	0.25±0.03	0.16±0.02	0.009±0.002	0.09	0.12	0.10	< 0.001				
W7	0.21±0.02	0.22±0.05	0.17±0.01	0.020±0.003	0.09	0.11	0.09	< 0.001				
W8	0.20±0.02	0.20±0.01	0.17±0.02	0.011±0.002	0.10	0.10	0.09	< 0.001				
W9	0.19±0.01	0.21±0.02	0.16±0.03	0.008±0.002	0.08	0.09	0.10	< 0.001				
W10	0.19±0.02	0.28±0.01	0.17±0.01	0.021±0.002	0.08	0.08	0.10	< 0.001				
W11	0.20±0.03	0.26±0.02	0.18±0.02	0.012±0.003	0.09	0.09	0.10	< 0.001				
W12	0.21±0.04	0.28±0.02	0.13±0.03	0.007±0.002	0.10	0.10	0.09	< 0.001				
Average	0.198±0.004	0.248±0.016	0.159±0.006	0.014±0.003	0.091±0.004	0.099±0.007	0.096±0.003	< 0.001				
	(e) In water sample, in non-ionic non-dialyzable form											
W1	0.99±0.01	1.23±0.06	1.00±0.01	0.090±0.001	0.88±0.03	1.08±0.07	0.75±0.03	< 0.001	0.04	0.06	0.01	< 0.001
W2	0.97±0.03	1.22±0.05	1.00±0.02	0.081±0.002	0.86±0.01	1.05±0.06	0.70±0.01	< 0.001	0.04	0.04	0.02	< 0.001
W3	0.99±0.01	1.23±0.06	0.90±0.02	0.088±0.001	0.87±0.02	1.03±0.06	0.78±0.02	< 0.001	0.01	0.05	0.02	< 0.001
W4	0.98±0.01	1.20±0.07	0.90±0.03	0.072±0.002	0.86±0.03	1.07±0.05	0.70±0.04	< 0.001	0.03	0.05	0.01	< 0.001
W5	0.98±0.01	1.21±0.06	0.80±0.01	0.077±0.003	0.88±0.04	1.04±0.07	0.76±0.01	< 0.001	0.04	0.06	0.01	< 0.001
W6	0.96±0.04	1.20±0.07	0.90±0.02	0.091±0.001	0.87±0.01	1.06±0.05	0.77±0.04	< 0.001	0.03	0.04	0.02	< 0.001
W7	0.98±0.02	1.22±0.05	1.00±0.02	0.072±0.001	0.88±0.01	1.05±0.07	0.72±0.02	< 0.001	0.02	0.06	0.03	< 0.001
W8	0.99±0.01	1.23±0.04	1.00±0.01	0.081±0.002	0.85±0.03	1.06±0.02	0.73±0.04	< 0.001	0.01	0.05	0.02	< 0.001
W9	0.97±0.02	1.21±0.03	1.00±0.02	0.082±0.001	0.88±0.02	1.05±0.05	0.75±0.04	< 0.001	0.03	0.06	0.03	< 0.001
W10	0.98±0.02	1.22±0.03	0.90±0.04	0.080±0.003	0.86±0.03	1.04±0.07	0.74±0.05	< 0.001	0.04	0.04	0.02	< 0.001
W11	0.98±0.02	1.22±0.04	1.00±0.03	0.071±0.001	0.87±0.02	1.08±0.04	0.72±0.06	< 0.001	0.03	0.07	0.01	< 0.001
W12	0.97±0.03	1.20±0.05	0.90±0.02	0.068±0.002	0.88±0.01	1.02±0.05	0.69±0.01	< 0.001	0.02	0.05	0.02	< 0.001

Table 2. Amounts of heavy metals in the filtered water samples

Modification	Concentration ($\mu\text{g ml}^{-1}$)			
	(a)	(b)	(c)	(d)
	Cu	Zn	Pb	Cd
Total metal	0.978 \pm 0.019	1.216 \pm 0.051	0.942 \pm 0.021	0.079 \pm 0.002
Cationic	0.780 \pm 0.046	0.968 \pm 0.016	0.783 \pm 0.054	0.065 \pm 0.007
Anionic	0.179 \pm 0.008	0.163 \pm 0.023	0.208 \pm 0.044	< 0.001
Non-ionic, dialyzable	0.063 \pm 0.005	0.046 \pm 0.006	0.078 \pm 0.004	< 0.001
Non-ionic, non-dialyzable	0.028 \pm 0.006	0.053 \pm 0.005	0.018 \pm 0.004	< 0.001

between the total heavy metal value of 3.2 ± 0.1 from Procedure (a) and that of $3.4 \pm 0.2 \mu\text{g ml}^{-1}$ from Procedures (b)–(e) suggests that the methods used were reliable.

Copper in samples (table 2a) passed through a cation-exchange resin gave concentrations of cationic form ranging between 0.73–0.83; anionic, 0.17–0.19; non-ionic (dialysable), 0.06–0.07; and non-ionic, non-dialysable, 0.02–0.03 $\mu\text{g ml}^{-1}$. Copper in cationic form was more or less evenly distributed among all the sites in the total metal concentration (0.96–1.0 $\mu\text{g ml}^{-1}$). The total zinc concentration (table 2b) in the filtered water was found to be the highest (1.17–1.27 $\mu\text{g ml}^{-1}$) in all the samples, followed by that in cationic form (0.95–0.99 $\mu\text{g ml}^{-1}$). The zinc concentration in anionic form ranged between 0.14 and 0.18; non-ionic (dialysable), 0.04–0.06; and non-ionic (non-dialysable) 0.05–0.06 $\mu\text{g ml}^{-1}$. The lead concentrations in the samples decreased in the order: total (0.92–0.96); cationic (0.73–0.84); anionic (0.17–0.25 $\mu\text{g ml}^{-1}$). The lead concentrations in non-ionic (dialysable), and non-ionic (non-dialysable) forms were found to be comparatively low (table 2c). The total concentration of cadmium in the water samples ranged between 0.077 and 0.081, while cadmium in cationic form lay in the range 0.058–0.072 $\mu\text{g ml}^{-1}$. The study showed that cadmium concentrations were very low (table 2d) in anionic, non-ionic (dialysable), and non-ionic (non-dialysable) forms.

Copper, zinc, lead and cadmium were predominantly present in the cationic form in roughly similar amounts ($81 \pm 2\%$) (table 2). With the exception of cadmium, the existence of the other metals in non-ionic (dialysable), and non-ionic (non-dialysable) modifications could be attributed to associations with high RMM organic matter. The non-detection of cadmium in anionic, non-ionic (dialysable), and non-ionic (non-dialysable) forms may be accounted for by the view that the levels of cadmium in the cationic form (table 2d), though low, contributed 82.3% of the total, leaving other modifications of the element practically undetectable by the techniques at our disposal.

The WHO maximum permissible levels of Cu, Pb, Zn and Cd in drinking water are: Cu = 2; Pb = 0.01; Zn = 3; Cd = 0.003 $\mu\text{g ml}^{-1}$ [20]. The corresponding limits for the heavy metals, according to the Uganda environment management statutes [21], are, respectively, Cu, 1; Pb, 0.1; Zn, 5; Cd, 0.1 $\mu\text{g ml}^{-1}$. Our results show that whereas Cu (0.978) and Zn (1.216 $\mu\text{g ml}^{-1}$) levels (table 2), although undesirable, are still within the permissible limits, those of Pb (0.942) and Cd (0.079 $\mu\text{g ml}^{-1}$) are in the higher range. The relatively high level of cadmium in the Jinja waters may be attributable to the activities of steel-rolling mills in the town, which process even scrap metal and run their untreated effluent directly into the lake. Cadmium has been blamed for large-scale poisoning incidents [22]. The elevated level of lead is probably explained by the established habit of car-washing and emptying of dead lead-acid accumulators

directly on the banks of the lake. The presence of a copper-smelting plant at Jinja for the copper ore from the Kilembe mines in Western Uganda in the 1960s may be blamed for the relatively significant levels of copper in the surrounding lake waters. The plant was located at Jinja owing to its closeness to Uganda's chief source of hydroelectric power, the Owen Falls Dam.

Under Ugandan law [23], the Uganda National Environment Management Authority (NEMA) requires, under Statutory Instrument (S.I.) No. 153-4 [24], that (1) 'Every industry or establishment shall install at its premises anti-pollution equipment for the treatment of effluent chemical discharge emanating from the industry or establishment', and that (2) 'Anti-pollution equipment installed, under Regulation 1 shall be based on the best practicable means, environmentally sound practice or other guidelines as the Executive Director may determine.' Under S.I.153-6 [25] of the same law, 'Any person who contravenes these Regulations commits an offence and is liable, on conviction, to imprisonment for a term not exceeding eighteen months or to a fine not less than one hundred and eighty thousand shillings and not more than eighteen million shillings or both' (US\$1 = US\$2000). In spite of this, however, only the Nile Breweries Industry at Jinja seems to be taking serious steps towards pre-treatment of effluent before discharge into the lake.

The forms in which heavy metals exist in a given environment are of key importance in understanding their uptake and release, as well as their toxicological properties. Most studies of the toxicity of heavy metals in fish have indicated that the free metal-ion (hydrated) is the most toxic form [26]. In our findings, it is indicated that the predominant form of the heavy metals is the cationic species (80–83%). This percentage value may be a good indicator of the bioavailability of these metals in the lake. Because surrounding townships, cities and inland ports (figure 1) in East Africa (e.g. Kampala in Uganda, Kisumu in Kenya, Mwanza, Bukoba, Musoma and Bukakata, in Tanzania) draw their fresh water from Lake Victoria for both domestic and industrial use, our findings indicate a slight but significant increase in the levels of the heavy metals in the waters of this famous freshwater body. Considering that no such metals were detected in the bottled mineral water commonly consumed in Kampala City, there is therefore a need for the relevant urban authorities in central East Africa to apply measures for water pollution control, particularly from industries. Informed urban dwellers may be well advised to drink only mineral water and use it only for their cooking. Rejecting the lake because of its pollution could then be publicised to expedite the process of environmental improvement.

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