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Trace-element behaviour in sediments of Ugandan part of Lake Victoria: results from sequential extraction and chemometrical evaluation

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Abstract

Lake Victoria is the second largest freshwater lake and the largest tropical lake in the world. The transboundary lake has the fastest growing population in its catchment, which can impact the water and sediment quality. To determine the extent of anthropogenic effects on sediment quality in the Ugandan part of Lake Victoria, the contents and binding behaviour of trace elements were analysed, as well as organic matter and phosphorus in different sediment layers of both deep and coastal sediments near the coastal cities of Entebbe, Kampala and Jinja. The data were assessed using the German LAWA criteria for trace-element pollution, the Geo-Index, Cluster- and Factor analyses. Mostly, no critical trace-element contamination in the sediments of the investigated area was observed. However, changes in element distributions caused by anthropogenic influences from around the lake were detected, like higher contents of Cu, Ti and V in near shore sediments with urban surrounding. Near Jinja, industrial wastewaters caused particularly elevated contents of Cu in the sediments (70–121 mg/kg, 3.5–6 times the geogenic background), exceeding the LAWA criteria and potentially harming the aquatic habitat. In addition, temporally growing organic matter contents in the lake sediments near the estuary of River Nzoia (from 4.2 to 17.6% in around 60 years) due to increased soil erosion in the river's catchment area and blooms of the water hyacinth became visible. This study demonstrates that the whole catchment area is responsible to ensure a healthy aquatic ecosystem in Lake Victoria.

Keywords Lake Victoria · Sediments · Trace elements and heavy metals · BCR extraction · $I_{\rm geo}$ and LAWA · Chemometrical judge- and assessment

Introduction

Trace-element contamination in lakes is a major concern especially in lakes still impacted by anthropogenic and population pressure. Despite being the largest freshwater lake,

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trace-element distribution in Lake Victoria sediments is not clearly understood due to limited studies mostly done on the Kenyan part of the Lake (Machiwa 2010; Outa et al. 2020). During the last 90 years, a strong population growth in the lake catchment area (growth rate of 3.5%/a) has led to a tenfold increase of the number of inhabitants in the catchment area which currently is approximately 40 million (Lake Victoria Basin Commission and GRID-Arendal 2017; Verschuren et al. 2002). The high population density around the lake (e.g. 549 inhabitants/km² in Kisumu County, Kenya (UNEP 2006) or 649 inhabitants/km² in Jinja District, Uganda (Uganda Bureau of Statistics 2016)) has an impact on the ecosystem's services of the lake. Besides a strong eutrophication of the lake, caused by a variety of multiple stressors (Hecky et al. 2010), an elevated input of trace elements is expected. Several factors contribute to that. First, growing cities and industries, together with immature wastewater treatment, led to an increased anthropogenic emission of trace elements and phosphorus (Oguttu et al. 2008).



Second, progressing deforestation for wood production and agriculture in the catchment area of Lake Victoria intensified soil erosion (de la Paix et al. 2013). The consequence is an increased transportation of trace elements and nutrients through the rivers into the lake. Furthermore, mining activities (gold, iron, rare earth metals) contribute to the enhanced input of trace elements into lake (Ngure et al. 2014).

Above a certain concentration, trace elements have a toxic effect on the living organisms of aquatic ecosystems (Wong et al. 2000). Furthermore, they cannot be degraded biologically or chemically. However, a deposition of trace elements in the sediments is possible, enabling their immobilisation (Förstner 1985). Under certain circumstances, trace elements might be remobilised when converted back to a more mobile form (Duruibe et al. 2007; Peng et al. 2009). This process is strongly dependent on chemical conditions in the water body and the binding form of the trace element in the sediment (Jenne and Luoma 1975). Trace elements that are adsorbed on clay minerals are remobilised after an increase of the salinity. When precipitated as carbonates, they redissolve after a decrease of pH. Trace elements adsorbed on Fe- and Mn-oxides and -hydroxides are remobilised under reducing conditions. Oxidising conditions trigger the remobilisation of trace elements precipitated as sulphide or being adsorbed on organic matter. Some binding forms just release the trace element under the influence of a strong acid, like aqua regia or perchloric acid, something that is unlikely under natural conditions. Being bound inside the crystal lattices of silicates, trace elements are solely remobilised by hydrofluoric acid (Rauret 1998). Clay-adsorbed, carbonate and Fe- and Mn-oxides and -hydroxide-adsorbed trace elements are considered to be bioavailable. The other binding forms thus are non-bioavailable (von Tümpling et al. 2013).

Thus, determining the trace elements binding form in addition to their total content is an easy way to enable risk evaluation of potential trace-element contamination. During the last decades, several analytical procedures have been developed to determine trace-element binding forms, so called sequential extraction procedures (Filgueiras et al. 2002). Throughout a sequential extraction, the sediment is extracted successively by different extraction agents, which specifically remobilise certain fractions. In case of the BCR sequential extraction procedure (developed by the Community Bureau of Reference, BCR), the sediments are subdivided into three fractions—namely the acid soluble, the reducible and the oxidizable fraction (Rauret et al. 2001).

The paper aims to provide a holistic view on the traceelement distribution on the Ugandan part of Lake Victoria. The focus of this paper is to investigate the influence of the distance to the shoreline as well as the sampling point's (SP) surroundings on trace element, phosphorus and organic matter distribution in sediments and the vertical distribution at each SP as well as the mobility of trace elements based on BCR extraction. This provides both insights into local differences and temporal developments of trace-element pollution into Lake Victoria. It is hypothesised that sediment trace-element contents decrease with increasing distance from the lake shoreline. It is also hypothesised that trace-element contents at shoreline depend on the land use type at the shoreline, e.g. differences between urban, rural or agricultural areas, respectively. Furthermore, it is expected that parts of the trace-element contents in the sediments are bioavailably bound.

Materials and methods

Study site

Lake Victoria, with a surface area of 68,870 km² is the second largest freshwater lake and the largest tropical lake in the world. The transboundary is located in East Africa and covers parts of Tanzania (49%), Uganda (45%) and Kenya (6%) with its catchment area extending over Burundi and Rwanda. The largest inflow, the River Kagera, arises at the Rwandan/Burundian border and enters Lake Victoria in Uganda. The only outflow of the lake is located at its northern shore near the city of Jinja, Uganda (Kayombo and Jorgensen 2005).

The sediment sampling took place in the Ugandan waters of Lake Victoria on seven different sampling points. Both, coastal (sampling points 1 and 4–7) as well as remote areas with a water depth of up to 68 m at sampling point (SP) 2 and SP 3 (Table 1; Fig. 1) were investigated.

Sampling

Sampling was carried out in May 2019. Undisturbed sediment cores were retrieved from each sampling point using a modified Kajak gravity corer (UWITEC, Austria) and plastic tubes of 60 cm length with an inner diameter of 9 cm. From each SP, the upper 15 cm of sediments was taken and subdivided into three subsamples with a thickness of 5 cm each. Only the subsamples 0-5 and 10-15 cm (X_00-05 and X 10–15; X: placeholder for SP; numbers represent the sediment layer in cm) were analysed, with the exception of subsample 7_05-10 near Jinja. In addition, three more sediment cores were retrieved from sampling points 1, 3 and 4 up to a depth of 38 cm, sliced in 1 cm layers for the first 6 steps and 2 cm layers for the following steps, from top to bottom. The lowermost 6 cm were used for analyses, since they deliver the furthest look back into the past. All samples were filled in plastic bags, carefully closed and cooled. After



Table 1 Coordinates, water depth and shore distance from the investigated sampling points

SP	Location	Coordinates	Water depth (m)	Distance from shore (m) ^a
1	Near estuaries of rivers Nzoia and Sio	00°09.613′ N, 033°54.670′ E	9.8	1.85 km
2	Deep waters	00°00.040′ S, 033°26.107′ E	40	20.55 km
3	Deep waters	00°30.438′ S, 033°15.599′ E	68	73.37 km
4	Murchison Bay	00°03.375′ N, 032°38.300′ E	20	3.65 km
5	Buvoma Channel	00°24.459′ N, 033°14.239′ E	15	4.50 km
6	Buvoma Channel	00°14.827′ N, 033°11.641′ E	25	3.75 km
7	Jinja	00°18.002′ N, 033°13.282′ E	25	0.98 km

^aGoogle, n.d.

Fig. 1 Map of the sampling points, located in the northeastern part of Lake Victoria (Google, n.d.)



bringing them to Germany, the samples were stored in a fridge at a temperature of 5 $^{\circ}$ C until further sample preparation and analysis.

Measurements

Sediment pH and redox potential was measured in the field immediately after slicing, using multi-parameter meter fitted with conventional pH and redox potential electrodes (WTW Multi3430, Germany), calibrated using standard pH and redox potential solutions. The results are listed in the supplementary information (Table S3). Loss on ignition (LOI) was determined by drying an aliquot of the sediment samples at 105 °C followed by combustion of the oven dried aliquot in a muffle oven at 550 °C for 2 h with pre- and post-muffling weighing of the samples. Since LOI can be used as a proxy for the amount of organic matter in the sediment, it is referred to as organic matter in the

following as well. Total phosphorus (TP) was analysed photometrically (Skalar, Netherlands) after combustion at 550 °C for 2 h and boiling in 1 N HCl for 15 min according to the German Norm EN ISO 6878:2004.

For the element analysis, an aliquot of the samples was air-dried for 48 h in a clean air (dust-free) laboratory before freeze-drying it for 72 h (CHRIST Delta 1-24 LSC, Germany). High-temperature drying would make the sediments unviable for the BCR extraction due to probable changes in the binding form of the elements.

The dry sediments were sieved with a sieving machine (Vibratory Sieve Shaker Analysette 3, Fritsch, Germany) and a 63- μ m mesh plastic sieve (Delrin®). The \leq 63- μ m sediment fraction was stored in sealable plastic containers at room temperature. 250 mg of each of the 18 sieved sediment samples were digested in quartz vessels with 6-mL 65% HNO₃ and 2-mL 37% HCl ("reverse aqua regia"; Suprapur® grade, Merck, Germany). The digestion was done in a pressure- and



temperature-controlled microwave (Discover® SP-D, CEM Corporation, USA) at the following settings: $T_{\rm Max}$ 170 °C, 18.5 min. Element determination within the final solution is called "pseudo-total concentration" since digestion by reverse aqua regia does not dissolve silicates. Furthermore, sequential extraction was conducted on every sample excluding sample 4_00–05 (due to small amount of sample), following the BCR protocol by Rauret et al. (2001) with a conclusive digestion determining the residual fraction.

All the reagents used for analysis were of analytical grade or higher. Element concentrations in the soil extracts and digested samples were measured by inductively coupled plasma mass spectrometry (ICP-MS; Agilent 8800 Triple-Quadrupole, Agilent Technologies[®], USA) following the Norm for application of ICP-MS EN ISO 17294-2:2004. All the quantified elements are listed in the following: As, Cd, Cr, Cu, Ni, Pb, Zn, Li, Be, Mg, Al, Ca, Ti, V, Mn, Fe, Co, Se, Rb, Sr, Mo, Sn and Ba.

As quality assurance measures, blanks (clean extraction agents) and certified reference materials (CRM; BCR 701 for BCR extraction and LGC6187 for digestion) were investigated at every step of the extraction process and at the digestion. In case of the sequential extraction, element recovery rates were between 87 and 121% and comparable to other values published in the literature (Margui et al. 2004; Tokahoglu and Kartal 2006). The results of the digestion of LGC6187 overall lay inside the 95% confidence interval of the certified values. A more detailed list with the results of the quality assurance samples and its description can be found in Tables S1 and S2 in the supplementary information.

Data analysis and evaluation

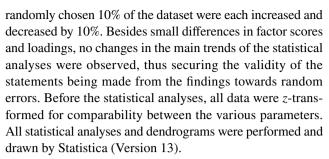
The results of the BCR extraction were transformed to "summed contents" (SC; Eq. 1) and "fraction percentages" (FP; Eq. 2). The former serves verifying the BCR results by comparing the summed contents with the pseudo-total contents and the latter was used to extract information about the binding behaviour of the studied elements:

$$SC_x = \sum_{n=1}^4 c_{n,x} \tag{1}$$

$$FP_{n,x} = \frac{c_{n,x}}{SC_x} \tag{2}$$

where SC is the summed content, FP is the fraction percentage, *n* is the *n*th fraction, and *x* is the element *x*.

To detect structures and correlations in the obtained dataset, cluster analyses were used, according to Ward's method with the squared Euclidean distance as distance measure. In addition, factor analyses were conducted (Varimax raw rotation). To verify the results of the statistical analyses,



The results of the digestion were evaluated using the Geoindex I_{geo} by Müller, which gives information about the local enrichment of an element in a certain sample (Müller 1979). Its local content is ranked depending on the geogenic background of the element. The higher the rank is, the higher the enrichment. In this study, the reference value (RV; reference for geogenic background) was set to its lowest element content out of all analysed samples. In addition, the criteria published by the German Working Group of the Federal States on Water Issues (LAWA) on heavy metal contamination of aquatic ecosystems helped evaluating the risks of aquatic ecosystem damage caused by some heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) (LAWA-working groups "Zielvorgaben" und "Qualitative Hydrologie von Fließgewässern" 1998). Besides, the metalloid arsenic was evaluated with values published by the ARGE ELBE (Krüger et al. 2000), due to its present potential of harming the aquatic ecosystem. In the following, the mentioned elements are referred to as "priority" trace elements and were looked at in more detail during evaluation.

Results and discussion

Trace-element and nutrient contents

The pseudo-total contents of the most relevant elements, having been declared as "priority" trace elements and exhibiting a risk to the aquatic ecosystem of Lake Victoria, of all samples are summarised in Table 2. In addition, 16 more elements were measured (see "Measurements"). The contents of the elements not listed in Table 2 are given in the supplementary information (Table S3). To evaluate the status of trace-element pollution in the Ugandan part of Lake Victoria, the presented results were compared with the results from most recently published studies of the other countries around the lake (see Table 2). The sediments from the Ugandan part of the lake contain less Zn and Cd compared to results from Kenyan and Tanzanian studies—maximum contents are below or close to the average of these studies. On the other hand, higher loads of Cu and Cr are noticeable in the investigated area. Since the element distribution differs between the sampling points, the differentiated element



Table 2 Range and mean of the "priority" trace element (mg/kg) in sediments from this study (Uganda) and the most recently published studies of the Tanzanian and Kenyan part of Lake Victoria (Machiwa 2010; Outa et al. 2020)

	Uganda (this study) $n = 18$			Tanzania (N	1achiwa 2	2010)	Kenya (Outa et al. 2020) n = 147		
	Range	Mean	Median	Range	Mean	Median	Range	Mean	Mediana
As	2.9–6.6	4.7	4.7	_	_	_	1.2-6.2	3.43	_
Cd	0.06-0.26	0.16	0.16	0.01-1.75	0.74	0.7	0.09-0.57	0.25	_
Cr	29-100	69	74	16.5-54.6	29.3	29	26.7-56.9	36.4	_
Cu	21-121	39	31	ND-89.7	24.2	18.9	23.7-60.1	37.7	_
Ni	19-56	29	27	_	_	_	15.9-29.0	19.4	_
Pb	10-25	17	17	ND-71.4	18.3	13.8	6.82-195	51.7	_
Zn	49–103	71	72	1-294	98.9	86.6	81.6–295	137	_

aNot indicated

Table 3 "Priority" trace-element contents (mg/kg) of all 18 samples with Geo-index classifications in brackets (class 0: 1-1.5×RV; class 1: 1.5-3×RV; class 2: 3-6×RV; italics: RV, bold: maximum value),

target trace-element contents by LAWA (aAs from Krüger et al. 2000) and information about LOI (%) and TP (g/kg)

Sample	As	Cd	Cr	Cu	Ni	Pb	Zn	LOI	TP
1_00-05	3,2 (0)	0,06 (0)	59 (1)	26 (0)	36 (1)	10 (0)	58 (0)	17.6	1
1_10-15	2,9 (0)	0,07(0)	70 (1)	28 (0)	39 (1)	11 (0)	71 (0)	15.9	0.4
1_00-36	4,5 (1)	0,12 (1)	95 (2)	40 (1)	56 (2)	14 (0)	103 (1)	4.2	0.1
2_00-05	6,6 (1)	0,18 (1)	46 (1)	32 (0)	26 (0)	18 (1)	81 (1)	37	2
2_10-15	5,7 (1)	0,22 (2)	64 (1)	32 (1)	29 (1)	18 (1)	89 (1)	36.3	2.7
3_00-05	3,7 (0)	0,25 (2)	39 (0)	21 (0)	24 (0)	14 (0)	77 (1)	29.7	1.9
3_10-15	4,2 (0)	0,19 (2)	34 (0)	23 (0)	22 (0)	14 (0)	73 (0)	27.2	1.3
3_24-30	3,0 (0)	0,26 (2)	29 (0)	21 (0)	19 (0)	12 (0)	79 (1)	22	3.8
4_00-05	6,5 (1)	0,15 (1)	75 (1)	24 (0)	21 (0)	21 (1)	75 (1)	42.1	2.6
4_10-15	5,0 (1)	0,13 (1)	75 (1)	26 (0)	21 (0)	16 (0)	59 (0)	42.2	1.4
4_32-38	4,1 (0)	0,14(1)	74 (1)	28 (0)	19 (0)	14 (0)	54 (0)	42.8	1.2
5_00-05	4,8 (1)	0,18 (1)	77 (1)	31 (0)	24 (0)	20 (1)	61 (0)	43.6	2.3
5_10-15	5,3 (1)	0,19 (2)	82 (1)	40 (1)	27 (0)	16 (1)	49 (0)	43.7	1.7
6_00-05	5,7 (1)	0,16 (1)	78 (1)	32 (0)	27 (0)	22 (1)	63 (0)	39.9	1.5
6_10-15	4,7 (1)	0,12 (1)	73 (1)	32 (0)	23 (0)	18 (1)	73 (0)	37.5	1.2
7_00-05	4,3 (0)	0,16 (1)	84 (1)	70 (2)	44 (1)	23 (1)	72 (0)	42.4	1.7
7_05-10	4,9 (1)	0,09 (0)	81 (1)	79 (2)	30 (1)	25 (1)	79 (1)	40.8	1.4
7_10-15	5,0 (1)	0,23 (2)	100 (2)	121 (2)	30 (1)	24 (1)	60 (0)	37.1	1.3
Target by	20a	1.2	200	60	50	100	200		
LAWA	20	1.4	200	00	50	100	200		
legend	Class 0	Class 1	Class 2			(I_{geo})			

contents of each of the studied samples are listed in Table 3. To facilitate the evaluation of the element contents, the $I_{\rm geo}$ has been added to Table 3. For reasons of clarity, the table was restricted to the "priority" trace elements. As mentioned before, a complete presentation of the results is given in Table S3.

It is clear, that SP 3 is the sampling point with lowest contents of trace elements (except for Cd). This might be due to the maximum distance between SP 3 and the shore. Since direct input of trace elements from the shoreline has far less influence on the sediment composition in the deep waters, the Cd enrichment should have been caused by the geogenic variance of Cd distribution rather than anthropogenic influences. Sediment contamination seems to increase with decreasing distance to the shore. Sampling point 7 near Jinja is the sampling point closest to the shore having the



strongest enrichment of "priority" trace elements. Besides an increased input of trace elements from general sources like urban runoff, this enrichment might originate from some industrial point sources. For example, the leather industry, situated in close distance to the lake, discharges considerable amounts of Cr-contaminated wastewater into the lake (approximately 2 t/a) (Oguttu et al. 2008). Until 1982, a Cu smelter was operated in Jinja (Kasese Municipality), causing a huge emission of Cu and other metals into the lake by directly discharging its effluents through the Masese wetland into Lake Victoria. 5940 ± 56.2 mg/kg of Cu was found in the soil of the Masese wetland near the shore of Lake Victoria (Nabulo et al. 2008). Moreover, a waste dump was abandoned on the ground of the Cu smelter after its closure (Ugandan Radio Network 2009; New Vision 2013), still causing Cu concentrations of 357 mg/L in its effluents in 2008 (Nabulo et al. 2008) and thus prolonging the negative consequences for the environment in the region.

The amount of organic matter depends on the location of the sampling points as well. Whereas in remote areas (far away from the shore) the deposition of organic matter is mostly restricted to aquatic plants and organisms, it is strongly influenced by the degradation of the thin humus rich layer in soils in nearshore areas, as exemplified by the difference between LOI of SP 3 (and SP 2) and sampling points 4–7 (Piccolo 1996).

Sampling point 1 has a remarkably different sediment composition in comparison to the other sampling points. It has a very low LOI and TP, thus contrasting the established relation between LOI and the distance between the SP and the shore. The reason for that is most likely found in the surrounding area of the SP, where the estuaries of the rivers Sio and Nzoia (approximately 12 km distance to the sampling point) are located. The deposited riverine sediments seem to influence the sediment composition of SP 1 strongly, containing eroded soils from the river's catchment area. With a discharge of 170 m³/s (more than 20 times higher than river Sio), river Nzoia has the highest discharge of all Kenyan rivers entering Lake Victoria and should be the main contributor to the LOI (Okungu and Opango 2002). Sample 1_30-36 has much less LOI and TP than the overlying samples, thus indicating a temporal change in the condition of the river's catchment area leading to an altered composition of the riverine sediments entering Lake Victoria. The trace-element contents are affected by that as well, as seen by decreasing contents of As, Cr, Cu, Ni and Zn. With a suitable sedimentation rate, the studied samples can be assigned to a certain space of time. Since dating was not part of this study, the sedimentation rate determined by Ramlal et al. (2003) for the Buvuma Channel (near sampling points 5 and 6) was applied as an approximation (0.67 cm/a). With that, the 5-cm-thick sediment layers are corresponding

to 7.5 years and the earliest point described by sample 1 10–15 is near the end of the twentieth century. Accordingly, the sample 1_30-36 dates to the 1960s. Since the 1960s, the catchment area of river Nzoia has undergone significant changes concerning its land use, pictured by a significant increase in agricultural areas (from 39.6% in 1973 to 64.3% in 2000-2001) and a decrease in forest areas, i.e. from 12.3% in 1973 to 7% in 2000-2001 (Githui et al. 2009). This resulted in enhanced runoff of rainwater and soil erosion, washing parts of the upper soil layer containing higher amounts of humus into the river, which explains the increase of LOI in the upper sediment layers of SP 1 (Githui et al. 2009). In addition, the population increase in the shore area of Lake Victoria, as for example in Budalangi was from 53,000 in 2001 (Onywere et al. 2007) to 66,500 in 2008 (Hongo et al. 2008), contributed to that as well. Formerly, the riverine sediments were most likely dominated by lithogenic ferralsols (Sombroek et al. 1980). The increased input of organic matter into the river might have diluted the lithogenic component, lowering the content of trace elements in the sediment. The decreased percentage of the lithogenic component is also indicated by shrinking Fe and Al contents in the samples of SP 1 (Table S3). It is noticeable, that both bulk elements do not decrease with the same ratio (-33% for Al; -14% for Fe), which might be due to their different sedimentation behaviour. Another source of organic matter could have been the bloom of the water hyacinth that happened around the end of the twentieth century in nearshore areas of Lake Victoria and effected the area around SP 1 as well (Wanda et al 2016). After the release of the Neochettina weevil in 2000-2001 (Ochiel and Njoka 2005), dead or bitten off plants could have sunk to the bottom and increased the organic carbon of the sediments. The lack of a major increase of LOI at the other sampling points might originate in municipal inputs of organic matter overshadowing the influence of the water hyacinth.

Regarding the other sampling points, no significant increase in trace-element contents is observable despite the long timescale displayed by the samples, apart from slightly elevated contents of Zn and Pb in sampling points 4 and 5. This indicates the low human impact on trace-element pollution from the shore areas into Lake Victoria during the last decades. At SP 7, the decreasing impact of the closed Cu smelter is demonstrated. Over time, the contaminated soils and the waste dump probably released most of the remaining Cu, reducing its input into Lake Victoria and causing smaller Cu contents in the upper sediment layers of the lake. However, as the LAWA criteria show, it still causes probable damage to the aquatic ecosystem exceeding the limit concentration for Cu contamination (60 mg/kg) in sample 7_00-05 (75.6 mg/kg). Apart from that, except for the Ni content in sample 1_30-36, the trace-element contamination of all



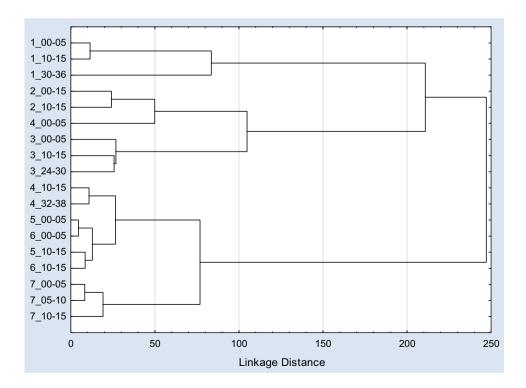
sediments in the investigated area seems to be of no concern. However, since the high Ni content in sample 1_30-36 seems to be of lithogenic origin, the environment might have been adapted to it.

Local differences in sediment composition

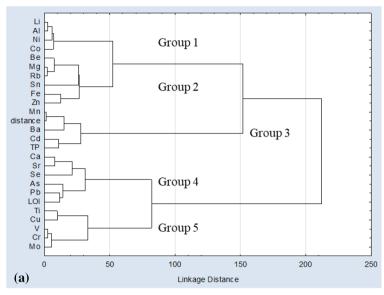
To study the element pattern of the sampling points in more detail, a cluster analysis was done with the pseudo-total contents of all 23 elements, LOI and TP, grouping together samples with similar sediment composition. Two main groups become apparent (sampling points 1–3 and sampling points 4–7, mismatch of sample 4_00–05), differing from the extent of urbanisation in their surroundings (Fig. 2). The SP 1 is located at a rural coastline and sampling points 2 and 3 are 20 respectively 70 km, far away from the nearest shore. SP 4 in front of the Murchison Bay near Kampala and Entebbe, sampling points 5 and 6 in the Buvuma channel and SP 7 in front of Jinja are altogether exposed to a much bigger anthropogenic influence, which is described in more detail in the following and most likely causes the observable split in the cluster. Examples of the influence of Jinja on the lake's sediment composition have been mentioned already in the last part. The influence of Kampala on the trace-element content of surrounding wetlands and the Murchison Bay by the uncontrolled effluent of industrial and municipal wastewaters was reported several times in the past as well (Nabulo et al. 2008; Mbabazi et al. 2010; Fuhrimann et al. 2015). An extension of that influence beyond the Murchison Bay seems plausible. Sampling points 5 and 6 could be influenced by the settlement of Kiyindi with its ferry services to Buvuma Island. The high distance between SP 1 and sampling points 2 and 3 highlights the special status of SP 1, which is caused by the river sediments from rivers Nzoia and Sio, entering the lake nearby and migrating towards SP 1.

To understand the backgrounds of these results, a closer look on the element distribution was assessed. Thus, patterns might be identified causing the apparent split of the sampling points. Figure 3a shows the dendrogram emerging from the cluster analysis of the element contents. Five groups of elements with differing patterns in their distribution among the 7 sampling points are evident. For each group, examples of those patterns are demonstrated in Fig. 3b-f. Group 1 (Li, Al, Ni, Co) demonstrates the mentioned influence of the estuaries of the rivers Nzoia and Sio on the sediment composition of SP 1, which is reflected by elevated contents of the associated elements in the sediments of SP 1 compared to the other sampling points. Another characteristic of the riverine influence on SP 1 are increasing contents of the associated elements in the deeper sediment layers. This phenomenon was traced back to a temporally increasing entry of organic matter probably resulting in a dilution of the trace elements in the upper sediment layer. The same course of element contents in SP 1 also applies to group 2 (Be, Mg, Rb, Fe, Zn, Sn). However, other than in group 1, related elements show an enrichment in sampling points 2 and 3 as well. Mwamburi (2003) studied the sediment composition of river Nzoia. A comparison with his results should enable a verification of our finding that SP 1 is influenced by river sediments from river Nzoia. It is noticeable, that especially

Fig. 2 Cluster analysis of the element composition using Ward's method with the squared Euclidean distance and *z*-transformed concentration values







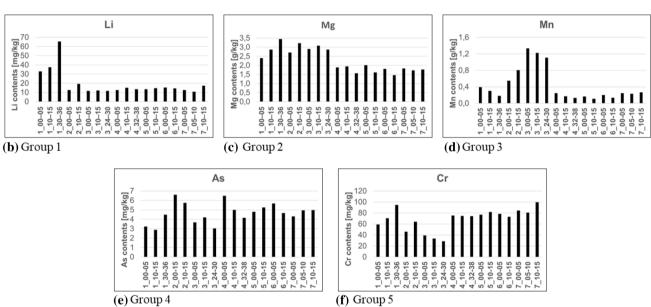


Fig. 3 a Cluster analysis of the element distribution patterns using Ward's method with the squared Euclidean distance and z-transformed concentration values; **b-f** Graphical representation of the characteristic element distribution patterns for each of the five groups

Al contents differ a lot from our results (8–12 times more Al in SP 1 (our study) than at the SP XIV near Nyandorera—SP closest to the lake (Mwamburi 2003)). This disagreement is most likely caused by the digestion method (30 min at 60 °C in aqua regia on a hotplate). Studies show that lower Al recoveries for hotplate digestions already become visible at hotplate conditions of 3 h at 110 °C (40% less in comparison to the applied microwave digestion) (Cheng and Ma 2001). This effect rather worsens with decreasing temperature and digestion time; therefore, comparisons are not possible for parameters analysed by different methods,

i.e. aluminium and trace elements. LOI seems comparable between SP 1 (our study) and SP XIV, though (4.2–10% (Mwamburi 2003) vs. 4.2–17.6% (this study)). Group 3 (Mn, distance, Ba, Cd, TP) describes the influence of the distance between the shore and the sampling point, containing elements with an enrichment visible in sampling points 2 and 3, with Mn correlating most with it. This is the result of the geochemical focussing of Mn, which shifts bioavailable Mn²⁺ from nearshore towards deeper areas (Schaller and Wehrli 1997). The effects of that are also visible in the Group speciation results (next section). Group 4 (Ca, Sr, Se,



As, Pb, LOI) combines the elements with seemingly no visible pattern in their distribution over the different sampling points. Group 5 (V, Cr, Mo, Cu, Ti) includes elements showing an enrichment in sampling points 4-7. The fact, that those elements mostly have an anthropogenic background (for example steel) matches the locality of sampling points 4-7 with their urban surroundings. The cluster analysis of the element patterns explains the apparent split of sampling points 1-3 and sampling points 4-7. In addition, it reinforces the hypothesis that the difference in land use and the extent of urbanisation in areas near the sampling points play an important role on the sediment quality. The results of the factor analysis given in Table 4 can be used to clearly contrast the sampling points located at the shoreline with sampling points 2 and 3, as stated by factor 1. The distance to the shore correlates positively with the element contents of Mn (group 3), whereas Cr, Mo and V contents (group 2) are correlating negatively with it. Factor 1 has a strong positive effect on sampling points 2 and 3 and a moderate to strong negative effect on sampling points 4-7 and the lowest sample of SP 1. Excluded from this is the sample 4 00–05, as already observed in the cluster analysis of the samples. Factor 2 mainly affects SP 1, correlating positively with Ni, Li, Al, Co, Be, Mg, and Rb contents (groups 1 and 2) and negatively with the LOI. It emphasises the impact of the rivers Sio and Nzoia on the sediment composition of SP1 and the change of the riverine sediment composition due to increasing degradation of the catchment areas, since the factor loadings for sample 1_30-36 is about three times higher than the ones of the other samples of SP 1. Factor 3 which describes only 15.6% of the variance with significant loadings of only As and Sr could not have been assigned to a specific cause.

Statistical analyses of speciation results (BCR procedure)

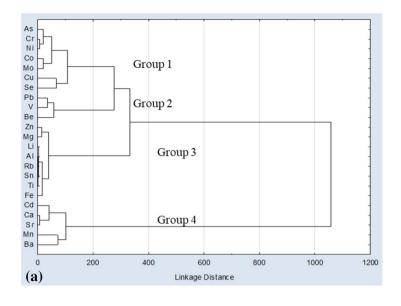
The FP and SC of each of the studied elements resulting from the BCR extraction is indicated in the supplementary information (Tables S5 and S6). When using the data obtained by the BCR extraction for cluster analysis, both dendrograms of summed contents and fraction percentages show a split of sampling points 1-3 and sampling points 4-7 (Fig. S1). The former indicates a good agreement between the results of the pseudo-total digestion and the sequential extraction. The latter might be caused by differences in the binding behaviour of the two sampling point groups. The binding behaviour of the elements was assessed by cluster analysis trying to find regional differences between the sampling points. Furthermore, the results for the "priority" trace elements (As, Cd, Cr, Cr, Ni, Pb, and Zn) will be compared with the literature (Bódog et al. 1997; Cao et al. 2015; Cappuyns et al. 2006; Cuong and Obbard 2006; Dundar et al. 2012; Gomez Ariza et al. 2000; Tokalioğlu et al. 2000; Zhigang 2008). Figure 4a shows the dendrogram resulting from the cluster analysis with the data containing the

Table 4 Factor loadings (a) and scores (b) resulting from the factor analysis using the normalised data containing pseudo-total element contents, TP, LOI and the closest distance between shore and the SP

(a)	F1	F2	F3	(b)	F1	F2	F3
Mn	0.92		'	1_00-05	0.22	0.68	- 1.25
Distance	0.91			1_10-15	-0.06	1.39	- 1.22
V	- 0.89			1_30-36	-0.68	3.15	-0.18
Cr	- 0.96			2_00-05	0.80	0.02	1.20
Mo	-0.85			2_10-15	0.70	0.77	1.74
Li		0.92		3_00-05	1.98	-0.11	0.15
Be		0.84		3_10-15	1.48	0.02	-0.05
Mg		0.71		3_24-30	2.00	-0.48	-0.71
Al		0.86		4_00-05	-0.17	-0.66	1.75
Co		0.79		4_10-15	-0.40	-0.62	-0.43
Ni		0.83		4_32-38	-0.40	-0.71	- 1.31
Rb		0.84		5_00-05	-0.38	-0.94	-0.17
LOI		-0.84		5_10-15	-0.74	-0.89	-0.94
As			0.70	6_00-05	-0.57	-0.54	0.16
Sr			0.81	6_10-15	-0.54	-0.68	-1.08
				7_00-05	- 1.01	-0.13	0.73
				7_05-10	-0.87	-0.28	0.74
				7_10-15	- 1.35	0.03	0.88

Only elements showing a factor loading of 0.7 or higher (absolute value) are listed, since no significant correlation occurs below that (F1: 29.5% of total variance; F2: 28.6% of total variance; F3: 15.6% of total variance). A complete indication of the results is listed in the supplementary information (Table S4)





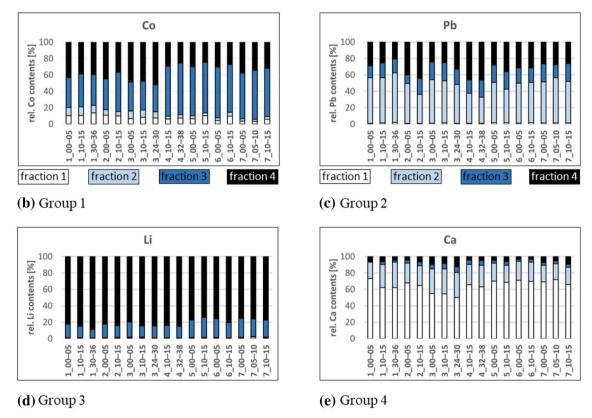


Fig. 4 a Cluster analysis of the element's binding behaviour using Ward's method with the squared Euclidean distance and z-transformed concentration values; **b**-**e** graphical representation of the characteristic binding behaviours for each of the four groups

fraction percentages. For each element group with similar binding behaviour (named BCR-group for clarity), an example is given in a chart next to the dendrogram (Fig. 4b–e). BCR-group 1 (As, Cr, Ni, Co, Mo, Cu, and Se) includes elements bound mainly in the third and fourth fraction in equal parts, with the exceptions of Cu (bound in the 2nd

fraction with 20–30%) and Se (share of 3rd fraction equals 90%). Comparing the results to the literature, As arsenic shows the expected behaviour, as it is usually found as an arsenate impurity in geogenic apatite (Moore and Ramamoorthy 1984) and precipitates as sulphide, incorporated with Fe (Chakraborti et al. 2001). Nickel has the typical



binding percentages for lake sediments. Copper and Cr exhibit a varying binding behaviour probably depending on the amount of organic matter present in the water. There seem to be a weak correlation between the FP of the third fraction and the LOI in the samples. The elements of BCRgroup 2 (Pb, V, Be) are all bioavailable with a percentage of ca. 50%. Whereas Pb and V are mainly bound to the second fraction, Be is as well found in the first fraction of the BCR extraction. No comparable results for the binding behaviour of Pb were found in the literature. Since all samples share the same characteristics for Pb, it can be assumed that regional specificities of (northeastern) Lake Victoria are the reason for that. BCR-group 3 (Zn, Mg, Zr, Li, Al, Rb, Sn, Ti, K, and Fe) consists of elements with low potential of damaging the aquatic ecosystem of Lake Victoria, since the percentage of the fourth fraction lies between 80 and 90%, except for Zn, Mg, K and Fe (share of 4th fraction is around 60%). Zn has a slightly elevated percentage of the first fraction in the samples of sampling points 4–7 which, according to literature, might imply an increased anthropogenic input of Zn into the lake. This is not reflected in the pseudo-total Zn contents of the sediments, rather exhibiting an enrichment of Zn in sampling points 1–3, though. However, it is possible, that higher anthropogenic inputs of Zn at sampling points 4–7 are just overlapped by the natural regiospecific enrichment of Zn at SP 1-3. BCR-group 4 (Cd, Ba, Mn, Ca, Sr) contains elements with a high bioavailable percentage (80%). For Mn, this partly applies, since the high bioavailability is only present in the samples of SP 2 and 3. For coastal sediments, the binding behaviour of Mn equals the third BCR-group. This is due to the geochemical focussing of Mn, which causes a migration of bioavailable Mn²⁺ into the deeper parts of the lake (Schaller and Wehrli 1997). The enrichment of Mn in SP 2 and 3 supports this hypothesis. Cadmium is known to be a bioavailable element. However, with no noteworthy pseudo-total Cd contents in the sediments, this fact is not alarming. As in the case of the pseudototal contents, the results of the factor analysis regarding the FP of the BCR extraction (Table 5) can be used to verify the cluster analysis. Both statistical methods are in good accordance with each other. BCR-factor 1 contains the elements of BCR-group 3, which show a clear preference of being bound in the fourth fraction of the BCR extraction. In addition, Cr and Ni changed sides from former BCR-group 1. BCRfactor 2 represents BCR-groups 1 (positive correlation) and 4 (negative correlation). Copper from BCR-group 1 could not have been clearly assigned to any factor, caused by its unique behaviour of being bound in the second fraction significantly. Nevertheless, it shows a tendency towards BCRfactor 2 as well (factor loading 0.65). The fact, that Cr and Ni are not found in BCR-factor 2 might be caused by their weaker preference to bind in the third fraction. BCR-group 4 represents the opposite of BCR-group 1, hardly being bound in the third fraction and exhibiting a significant bioavailability. Calcium and Mn of BCR-group 4 show no significant correlation with BCR-factor 2, but a tendency towards that is present as well (factor loading -0.68, respectively, -0.65). Lastly, BCR-factor 3 describes the elements of BCR-group 2, which prefer to bind in the second fraction. The low factor score for the first fraction matches Pb and V, but does not correspond to Be which shows a fraction percentage of the first fraction equal to that of the second fraction. That difference might have caused the lower factor loading of Be for BCR-factor 3 (0.71 compared to 0.8 (V) and 0.93 (Pb)).

Summarised discussion

Comparing the pseudo-total contents of trace elements in sediments of the northeastern part of Lake Victoria with other regions of the lake, lower levels of Zn and Cd and higher levels of Ni, Cu and Cr become apparent in the investigated area of this study. By the application of I_{geo} , it was possible to identify SP 3 as the least and SP 7 as the most contaminated sampling point, drawing a connection between the distance to the shore/urbanity and trace-element contamination of the sediments. At SP 7, two main sources for Cr and Cu pollution were found—the leather industry and a Cu smelter, which was closed in 1982. Untreated wastewaters from the Cu smelter were directly discharged into Masese Wetland and led to a massive contamination of its soils, which influences on the sediment composition of Lake Victoria near Jinja, being intensified by a waste dump having

Table 5 Factor loadings (a) and scores (b) resulting from the factor analysis using the normalised data containing the fraction percentages in each fraction of all of the studied elements

(a)	F1	F2	F3	(b)	F1	F2	F3
≥0.7	Cr, Ni, Zn, Li, Al, Mg, Ti, Fe, Rb, Sn	Co, Se, Mo, As	Pb, Be, V	Fraction 1 Fraction 2	-0.31 ± 0.2 -0.76 ± 0.16	-0.81 ± 0.31 -0.81 ± 0.25	-1.44 ± 0.32 1.25 ± 0.62
≤ -0.7		Cd, Ba, Sr		Fraction 3 Fraction 4	-0.62 ± 0.17 1.69 ± 0.26	1.57 ± 0.4 0.06 ± 0.58	0.1 ± 0.43 0.25 ± 0.43

Only elements showing a factor loading of 0.7 or higher (absolute value) are listed, since no significant correlation occurs below that (F1: 57.7% of total variance; F2: 20.9% of total variance; F3: 11.7% of total variance). The factor scores of the samples are similar within one fraction, so only the mean ±95% CI is indicated. Full indication of the individual samples and fractions as well as the factor loadings of each element are shown in the supplementary information (Table S7)



been left on the ground of the former Cu smelter, can still be seen today. As the Cu content exceeds the LAWA criteria, the waste dump causes potential Cu contamination to the aquatic ecosystem. Apart from that, according to LAWA, no major trace-element contamination becomes apparent in the sediments of northeastern Lake Victoria. However, some changes in the element distribution are recognisable that might be caused by anthropogenic influences. The element patterns suggests an enrichment of Cr, V and Mo (+Cu and Ti) in the coastal sediments near urban areas (sampling points 4-7). Addressed elements are altogether mainly distributed by anthropogenic emissions. SP 1 near the estuary of river Nzoia demonstrates the change of land use taking place in the catchment area of the lake and its influence on the lake's sediment composition. Increased agricultural area and decreased forest cover most likely led to enhanced soil erosion causing a higher input of organic matter into Lake Victoria. The bloom of the water hyacinth might have contributed to that as well.

Conclusion

Most trace-element loads are harmless and no strong increase in trace-element inputs over time was observed. However, anthropogenic induced changes in the sediment composition of coastal areas with urban influences were apparent as shown by elevated levels of trace elements with an anthropogenic background (Cr, V, Mo, Cu and Ti).

The strong influence of river Nzoia on the sediment composition of SP 1 demonstrates that the whole catchment area is responsible for ensuring a healthy aquatic ecosystem in Lake Victoria. The city of Jinja plays an important role since it is located near the outflow of Lake Victoria (River Nile) and influences the water quality even beyond the borders of the lake. High trace-element inputs into Lake Victoria might affect aquatic cohabitation of the River Nile. High inputs of Cu and Cr by industrial wastewaters have already been reported in the past. Since Cu and Cr contents are lower in the upper sediment layers, the situation has slightly been improved though.

Further studies should extend the investigated area. It would be interesting to see, if the connections between land use changes and the sediment composition found for River Nzoia also apply to River Kagera in the north-western Lake Victoria. In addition, a closer look should be taken closer to Kampala. There, the city's influence on the ecosystem might be more distinctive and better to investigate than at SP 4 of this study.

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Author contributions All the authors excluding Tallent Dadi and Nils Ribbe contributed to the study conception and design. Sampling, sediment preparation, data collection and analysis were performed mainly by Kenneth Arinaitwe, Tallent Dadi and Nils Ribbe. The judge- and assessment has been done by all the authors. The manuscript was written by Nils Ribbe and Wolf von Tümpling with support of the other authors in revision of the manuscript. All the authors read and approved the final manuscript.

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Availability of data and material All data are given in the supplementary material of this paper.

Code availability Not applicable.

Declarations

Conflict of interest Finally, the authors declare that they have no conflict of interest.

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