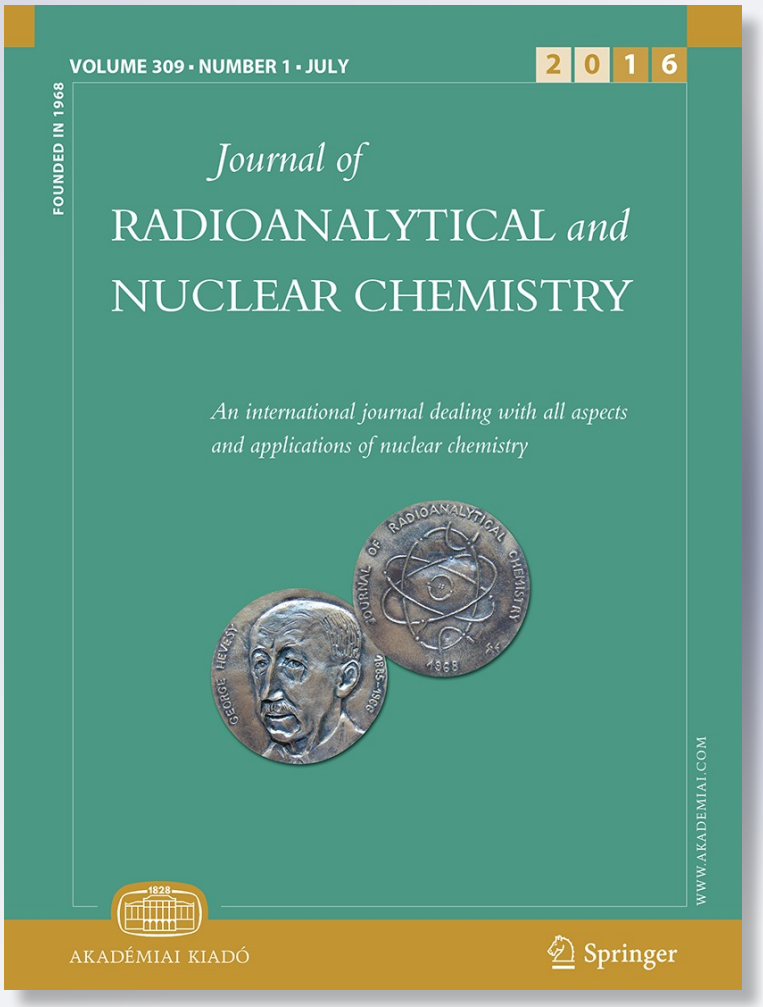
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Najat K. Mohamed, A. V. R. Ntarisa, I. N. Makundi & J. Kučera

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Impact of North Mara gold mine on the element contents in fish from the river Mara, Tanzania

Najat K. Mohamed[1] · A. V. R. Ntarisa[1] · I. N. Makundi[1] · J. Kučera[2]

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Abstract The aim of the study was to determine element concentrations in lungfish and catfish from the river Mara with INAA and EDXF to assess the health impact of the North Mara gold mine (NMGM) in Tanzania. Twenty samples of each species were collected from two sites downstream and upstream along the river Mara in the vicinity of NMGM. The sampling sites were 70 km apart. Significantly higher concentrations ($p \leq 0.05$) of Cr, Ni, Cu, and Se in one of the species taken downstream than in those taken upstream might indicate contamination of the river Mara caused by the mining activities.

Keywords Gold mining · Contamination · Lungfish · Catfish · INAA · EDXRF

Introduction

Tanzania introduced large-scale mining in 1998, which has been growing very rapidly after the economy reforms of the 1990s. There are three gold fields in Tanzania, the most prominent being the Lake Victoria Gold Field (LVGF) that spreads between Tabora, Shinyanga, Simiyu, Geita, Mwanza, Kagera and Mara regions. This gold field produces about 90 % of all gold that is mined in Tanzania. The NMGM, located near the river Mara, is a part of LVGF [1, 2].

The NMGM is located in Tarime District in the north-western part of Tanzania. This mine has three open pits, two of them located along the river Tighite that is the biggest tributary of the river Mara in the Tarime district. In 2009 a spill from a tailing dam and leachate was observed around NMGM. A small-scale study, performed after the event, detected high levels of several metals in this river above the permissible levels set by the World Health Organization (WHO). Since the river Tighite is a tributary of the river Mara, the metal levels were assumed to be high in this river as well. The river Mara is home to several species of fish, of which catfish (*Clarias mossambicus*) and lungfish (*Protopterus aethiopicus*) are the ones most consumed by the inhabitants.

Several studies conducted in different places in the world have also shown that metal contamination in fish is associated with mining activities [3, 4]. The fish in these studies were likewise collected from waters in the neighbourhood of metal mines. Hence, there is a compelling reason to measure metal concentrations in fish taken from the river Mara in the proximity of the biggest gold mine in Tanzania. Fish from the river Mara are consumed by approximately 300,000 people living near the river. If contaminated by extraneous and/or toxic metals, the fish will pose a health hazard to a large population. This study aims at determining metal concentrations in two most commonly eaten fish species (catfish and lungfish) from the river Mara near NMGM.

Experimental

Sampling and sample preparation

NMGM, operated by the African Barrick Gold Company is located in the North Western part of Tanzania in the

✉ J. Kučera
kucera@ujf.cas.cz

[1] University of Dar es Salaam, P.O. Box 35091, Dar es Salaam, Tanzania

[2] Nuclear Physics Institute CAS, 250 68 Husinec-Řež 130, Czech Republic

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Tarime district of the Mara region. The mine is 43 km distant from Tarime town, about 100 km east of Lake Victoria. There are three open-cast mines in the area, namely Nyabirama, Nyabigena and Gokona [5]. Both Nyabigena and Gokona mines are located along the river Tighite, which is the biggest tributary to the river Mara in the Tarime district. This river conflows with the river Mara near Matongo village, at about 5 and 3 km from the Matongo centre and the Nyabirama pit, respectively. In this study, samples of catfish and lungfish from downstream and upstream of the river Mara were collected at Wegita and Mrito villages, respectively. Wegita is about 35 km from Matongo village downstream whereas Mrito is about 30 km upstream from Matongo village. All fish samples were collected directly from the sites along the river Mara before being delivered to the market. Downstream samples were collected at the Wegita village whilst the upstream samples were collected at the Mrito village (Fig. 1). Twenty samples of each fish species were collected to make a total of forty.

The collected fish samples were washed with distilled water to remove external dirt. The flesh of the fish was cut, placed in jars and kept in a cooler box. The samples were then transferred to the University of Dar es Salaam, to be oven dried at 45–50 °C for 48 h. The dried samples were then ground with an electric grinder and sieved to obtain fine powder. This powder was stored in polyethylene (PE) bags and subsequently transferred to the Nuclear Physics Institute of the Czech Academy of Sciences (NPI) and to the Tanzania Atomic Energy Commission (TAEC) in Arusha for further preparation and analysis.

At NPI, the samples were additionally freeze dried, homogenized by cryogenic grinding and deep-frozen prior to analysis. Sample aliquots of approximately 100 mg and 200 mg for short-time and long-time irradiation, respectively, were sealed into acid-cleaned (PE) disk-shaped capsules.

At TAEC, about 14.7 g of fish powder from each sample were pelletized in a hydraulic press using a pressure of 20 tons to produce intermediate thickness pellets with an outer diameter of 32 mm. Since fish muscle powder has good binding properties, there was no need to use a binder for these samples.

Analysis

The samples were subjected to instrumental neutron activation analysis (INAA) performed at NPI and energy

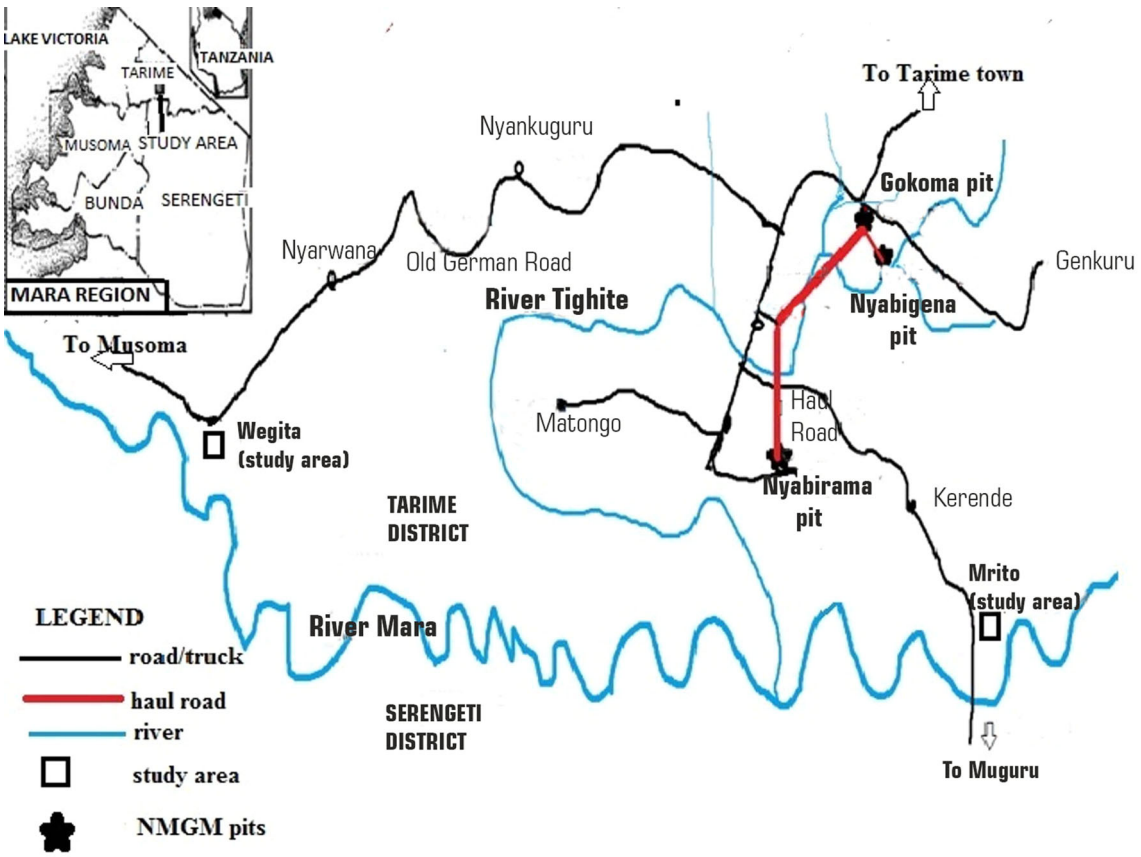


Fig. 1 Location of sampling sites Wegita (*downstream*) and Mrito (*upstream*) along the river Mara in the vicinity of the North Mara gold mine

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dispersive X-ray fluorescence (EDXRF) analysis performed at TAEC. Irradiations for INAA were carried-out in the nuclear reactor LVR-15 of the Research Centre Řež, Ltd., operated at 9 MW. Short- and long-time irradiations (30 s and 3 h, respectively), were carried out at neutron fluence rates of 3×10^{13}, 1×10^{13}, and 8×10^{13} cm^2 s^{-1} for thermal, epithermal, and fast neutrons, respectively. Nuclear reactions, radionuclides employed and their parameters have already been reported elsewhere [6]. For a relative standardization, multielement standards, containing known amounts of elements to be determined, were simultaneously irradiated with the fish samples. Counting of the irradiated samples and standards was performed under conditions given in Table 1, where t_i is irradiation time, t_d is decay time, t_c is counting time, and counting geometry is the distance of the sample from the cap of the detector. The samples and multielement standards were counted in the same geometry.

The detectors were coupled to the Canberra Genie 2000 computer-controlled gamma spectrometer via a chain of linear electronics, which contained a loss-free counting module (LFC Canberra 599, dual mode) to correct for the pile-up effect and dynamic changes of dead time. The Canberra Genie 2000 software was used for spectrum analysis. For quality control purposes, NIST SRM 1577b bovine liver was assayed, which was prepared for analysis in the same way as the samples. Irradiations and measurements for INAA were carried out within the CANAM infrastructure of NPI supported by MEYS project No. LM2011019.

The EDXRF spectrometer used in this study was a Spectro–Xepos™ system operated by X-lab Pro™ software. In this model, X-rays are generated by the X-ray tube and the excitation of elements in the sample was carried out using three different secondary targets depending on the atomic number of the investigated element. Each pellet with a mass of approximately 14.7 g was irradiated for 15 min to determine the elemental composition of the samples from spectra of the samples using the X-lab pro™ software. The tube was operated at a maximum power of 50 W and a maximum voltage of 50 kV. Element concentrations of the samples were also calculated with the X-lab Pro™ software with Turboquant (Tq 9232) algorithm to allow matrix effect correction [7]. The software corrected for the matrix effects (M_i) and the interference effects (K_i) based on the fundamental parameter methodology. This software package also corrected for the background effect on a spectral line intensity (I_i), given as counts per second (cps). After making the corrections, the intensity was converted into concentration of an element using Eq. (1) [8]

$$C_i = K_i I_i M_a \quad (1)$$

where C_i is the concentration of a given element i, M_α is the correction factor for matrix effects, K_i is the proportionality constant, I_i is the intensity of the fluorescent radiation from the element i.

Table 1 Irradiation and counting conditions for INAA

t_i	t_d	t_c	Counting geometry (cm)	Detector[a]
30 s	10 min	10 min	15	HPGe-1
3 h	4–5 days	1 h	20	HPGe-2
	1 month	3 h	1	HPGe-2

[a] *HPGe-1* coaxial HPGe detector with 23 % relative efficiency, resolution FWHM 1.80 keV for the 1332.5 keV photons of ^{60}Co

[a] *HPGe-2* coaxial HPGe detector with 53 % relative efficiency, resolution FWHM 1.80 keV for the 1332.5 keV photons of ^{60}Co

Results and discussion

Quality control analyses

INAA results of quality control analysis are shown in Table 2 both for elements, whose values were above and below limit of detection (LOD). Our values agree with the NIST certified values within uncertainty margins or they are close to the NIST noncertified values, thus proving the accuracy of our results.

Table 2 INAA results for NIST SRM 1577b Bovine Liver

Element, unit	This work[a]	NIST value [9][b]
Na, (%)	0.237 ± 0.006	0.242 ± 0.006
Cl, (%)	0.259 ± 0.014	0.278 ± 0.006
K, (%)	0.963 ± 0.040	0.994 ± 0.002
Ca, (mg kg^{-1})	<166	116 ± 4
Sc, (mg kg^{-1})	40.4 ± 1.2	(41)
Cr, (mg kg^{-1})	<0.17	–
Mn, (mg kg^{-1})	10.3 ± 0.4	10.5 ± 1.7
Fe, (mg kg^{-1})	185 ± 10	184 ± 15
Co, (mg kg^{-1})	0.24 ± 0.02	(0.25)
Zn, (mg kg^{-1})	120 ± 4	127 ± 16
Se, (mg kg^{-1})	0.64 ± 0.10	0.73 ± 0.06
As, (mg kg^{-1})	<0.3	(0.05)
Br, (mg kg^{-1})	9.54 ± 0.04	(9.7)
Rb, (mg kg^{-1})	12.3 ± 0.6	13.7 ± 1.1
Sr, (mg kg^{-1})	<6	–
Cd, (mg kg^{-1})	<8	0.50 ± 0.03

[a] Value ± expanded uncertainty (coverage factor $k = 2$)

[b] Certified values are given with uncertainties, noncertified values are in parenthesis

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For EDXRF measurements, reference material IAEA-359 Trace and Minor Elements in Cabbage [10] was used for quality control purposes. Table 3 shows that our results agree well with the IAEA values, where available.

Results for fish samples

Mean values and dispersion of element contents obtained with EDXRF and INAA are given in Tables 4 and 5 for lungfish and catfish, respectively. All results are reported on a dry mass basis. For the data sets ($n = 10$ for each category) with a normal distribution, we calculated the arithmetic mean, x, and standard deviation, sd. For those data sets deviating from a normal distribution we calculated the median,$\tilde{x}$, and median absolute deviation, MAD. Differences among the element mean values in fish samples from the upstream (presumably clean water) and the downstream (presumably polluted water) localities of the river Mara were tested using parametric t test or non-parametric Kolmogorov–Smirnov test for normally or non-normally distributed data sets, respectively, at $p = 0.05$ significance level. Results for 16 elements were obtained in both fish species by both analytical methods, namely for the following elements Na, Cl, K, Ca, Sc, Cr, Mn, Fe, Co, Ni, Cu, Zn, Se, Br, Rb and Sr. Other elements, including some toxic ones (As, Cd, Hg, etc.), could not be determined by INAA because of the high contents of the elements Na, P, Cl and K, which generated high activities of their respective radionuclides ^{24}Na, ^{32}P, ^{38}Cl and ^{42}K on neutron irradiation. Thus LOD of other elements were hampered. No attempt was made to determine Hg because of possible

Table 3 EDXRF results (mg kg^{-1}) in IAEA-359 Cabbage material

Element	This work[a]	IAEA value [10][b]
Ni	1.02 ± 0.10	1.05 ± 0.05[c]
Cu	5.18 ± 0.65	5.67 ± 0.18[d]
Hg	<0.5	0.013 ± 0.00015[c]
Pb	<0.5	–

[a] Value ± expanded uncertainty (coverage factor $k = 2$)

[b] Arithmetic mean ± 95 % confidence interval

[c] Information value

[d] Recommended value

Table 4 Element values determined in lungfish from the Mrito (upstream) and Wegita (downstream) villages on the river Mara

Element, unit	Method	Presumably clean area (upstream)		Possibly polluted area (downstream)		p value[a]	
		$x \pm sd$[b]	$\tilde{x} \pm MAD$[c]	$x \pm sd$[b]	$\tilde{x} \pm MAD$[c]	t-test	K–S[d]
Na, (%)	INAA	2.14 ± 1.24		1.81 ± 0.99		>0.05	
Cl, (%)	INAA	3.03 ± 1.80		2.86 ± 1.75		>0.05	
K, (%)	INAA	1.11 ± 0.15		1.10 ± 0.18		>0.05	
Ca, (%)	INAA		0.94 ± 0.12	2.525 ± 1.44			<0.05
Sc, (mg kg^{-1})	INAA	0.008 ± 0.001			0.006 ± 0.001		>0.05
Cr, (mg kg^{-1})	INAA	0.38 ± 0.03			1.05 ± 0.73		<0.05
Mn, (mg kg^{-1})	INAA	7.52 ± 2.03		8.96 ± 3.66		>0.05	
Fe, (mg kg^{-1})	INAA	55.5 ± 10.0		66.3 ± 21.8		>0.05	
Co, (mg kg^{-1})	INAA	0.051 ± 0.014		0.073 ± 0.036		>0.05	
Ni, (mg kg^{-1})	EDXRF	1.23 ± 0.18		2.88 ± 0.35		<0.05	
Cu, (mg kg^{-1})	EDXRF	168 ± 80		587 ± 99		<0.05	
Zn, (mg kg^{-1})	INAA	45.9 ± 6.0			49.4 ± 1.5		>0.05
As, (mg kg^{-1})	INAA	<0.6		<0.6			
Se, (mg kg^{-1})	INAA	0.58 ± 0.14		0.59 ± 0.13		>0.05	
Br, (mg kg^{-1})	INAA	18.7 ± 6.2		17.2 ± 3.2		>0.05	
Rb, (mg kg^{-1})	INAA	7.85 ± 1.47		6.84 ± 1.34		>0.05	
Sr, (mg kg^{-1})	INAA		16.2 ± 0.4		37.1 ± 13.0		<0.05
Cd, (mg kg^{-1})	INAA	<7		<7			
Hg, (mg kg^{-1})	EDXRF	<0.5		<0.5			
Pb, (mg kg^{-1})	EDXRF	<0.5		<0.5			

[a] Values ≤0.05 indicate significant difference of the mean values

[b] Arithmetic mean ± SD

[c] Median ± median absolute deviation

[d] Two-sample Kolmogorov–Smirnov test

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Table 5 Element values determined in catfish from the Mrito (upstream) and Wegita (downstream) villages on the river Mara

Element, unit	Method	Presumably clean area (upstream)		Possibly polluted area (downstream)		p value[a]	
		$x \pm sd$[b]	$\tilde{x} \pm MAD$[c]	$x \pm sd$[b]	$\tilde{x} \pm MAD$[c]	t-test	K–S[d]
Na, (%)	INAA		1.30 ± 0.14	1.89 ± 0.33			<0.05
Cl, (%)	INAA		1.97 ± 0.24	2.99 ± 0.54			<0.05
K, (%)	INAA	1.22 ± 0.13		1.25 ± 0.09		>0.05	
Ca, (%)	INAA	1.26 ± 0.19		1.24 ± 0.12		>0.05	
Sc, (mg kg^{-1})	INAA	0.005 ± 0.002		0.004 ± 0.001		>0.05	
Cr, (mg kg^{-1})	INAA		0.24 ± 0.05	0.30 ± 0.07			>0.05
Mn, (mg kg^{-1})	INAA	10.72 ± 2.29		14.27 ± 5.62		>0.05	
Fe, (mg kg^{-1})	INAA	39.13 ± 8.06		53.28 ± 6.56		>0.05	
Co, (mg kg^{-1})	INAA	0.031 ± 0.015			0.033 ± 0.006		>0.05
Ni, (mg kg^{-1})	EDXRF	1.07 ± 0.18		1.45 ± 0.40		>0.05	
Cu, (mg kg^{-1})	EDXRF	70 ± 4		205 ± 94		<0.05	
Zn, (mg kg^{-1})	INAA	26.1 ± 3.0		28.3 ± 2.1		>0.05	
As, (mg kg^{-1})	INAA	<0.6		<0.6			
Se, (mg kg^{-1})	INAA	0.80 ± 0.32		1.16 ± 0.31		<0.05	
Br, (mg kg^{-1})	INAA	17.2 ± 2.7		17.6 ± 2.7		>0.05	
Rb, (mg kg^{-1})	INAA	9.29 ± 1.06			8.94 ± 0.80		>0.05
Sr, (mg kg^{-1})	INAA	28.0 ± 5.1		31.3 ± 4.5		>0.05	
Cd, (mg kg^{-1})	INAA	<7		<7			
Hg, (mg kg^{-1})	EDXRF	<0.5		<0.5			
Pb, (mg kg^{-1})	EDXRF	<0.5		<0.5			

[a] Values ≤0.05 indicate significant difference of the mean values

[b] Arithmetic mean ± SD

[c] Median ± median absolute deviation

[d] Two-sample Kolmogorov–Smirnov test

volatility losses from the PE capsules during irradiation in the reactor. Levels of the above toxic elements were also below LOD of EDXRF.

It follows from Table 4 that the mean concentrations of Ca, Cr, Ni, Cu, and Sr were significantly higher in the samples from the possibly polluted area than those from the presumably clean area. The mean concentrations of other elements in the samples from both areas did not significantly differ, namely as a result of a large variations in the individual data sets.

Table 5 shows somewhat different patterns of element contents for catfish. In this particular case, other elements, namely Na, Cl and Se, and again Cu, as in lungfish, were significantly elevated in the possibly polluted area. These differences may result from a different element uptake by the individual fish species. Here again a large variation in the individual data sets was observed. Two reasons may be considered for the large variations of element contents in both fish species. First is a biological variation (age, sex, etc.), second is the factor of a free migration of fish along the river over a distance of 70 km (which may be considered as part of the first reason), which separates both sampling localities. The free migration of fish along river over the stated distance should not be surprising, because some fish species, e.g., salmon, are known to travel to egg-laying areas over thousands of kilometres.

Environmental contamination caused by the past or the present mining and/or ore processing activities has recently been addressed by a number of authors [2–4, 11–15]. High concentrations of Cr, Fe, Mn, Ni and Cu in various compartments of the environment have been associated with mining activities [2–4, 11, 12]. For instance, Almas and Manoko [2] reported elevated concentrations of Co, Cr, Cu, Ni, Pb and Zn in samples of soil, sediments and water in the vicinity of the Geita Gold Mine (GGM) and NMGM in Northwest Tanzania. A similar observation as found in this work has been reported by Mnali [12]. It has been reported that fish may accumulate the elements Hg, Pb, Cu, Zn, Mn and Fe, whose concentrations were elevated as a result of gold mining in the river Offin, Ghana [13]. Elemental analysis of fish tissues (muscle and viscus) has also been considered useful for environmental monitoring in a Brazilian study [16].

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Table 6 Maximum values and/or literature values of element contents in fish

Element, unit	Literature values[a]
Na, (%)	0.8 [20]
Cl, (%)	0.6 [20]
K, (%)	1.5 [20]
Ca, (%)	0.076–2.0 [20]
Sc, (mg kg^{-1})	–
Cr, (mg kg^{-1})	4 [17], 0.03–2 [20]
Mn, (mg kg^{-1})	0.3–5 [20]
Fe, (mg kg^{-1})	9–88 [20]
Co, (mg kg^{-1})	0.006–0.05 [20]
Ni, (mg kg^{-1})	0.1–4.2 [20]
Cu, (mg kg^{-1})	40–120 [17], 0.7–15 [20]
Zn, (mg kg^{-1})	160–600 [17], 9–82 [20]
As, (mg kg^{-1})	0.4–20 [17], 0.2–10 [20]
Se, (mg kg^{-1})	1.2–4 [17], 0.17 [20]
Br, (mg kg^{-1})	400 [20]
Rb, (mg kg^{-1})	0.8–5.6 [20]
Sr, (mg kg^{-1})	0.2–20 [20]
Cd, (mg kg^{-1})	0.2 [19]
Pb, (mg kg^{-1})	1.2 [19]
Hg, (mg kg^{-1})	2.0 [19]

[a] Values originally given on fresh mass basis [17–19] were recalculated to dry mass basis using multiplication by a factor of 4 as recommended by Bowen [20]

Results of this work suggest that the elevated concentrations of Cr, Ni, Cu in lungfish and Cu and Se in catfish from downstream of the river Mara are likely caused by contamination originating from the mining activities of NMGM. Apparently, lungfish is a better biomonitor of environmental pollution than catfish. So far, we have no explanation for higher concentrations of Ca and Sr in lungfish, and Na and Cl in catfish from the polluted area, because these elements, except for Sr, are macroconstituents of the fish meat.

We have also attempted to evaluate the results obtained in terms of health risk assessment, because excessive contents of extraneous elements may lead to serious health problems to consumers. For this purpose, Table 6 lists legal limits for hazardous substances in fish and fishery products found in the literature [17–19]. The legal limits set in various countries have been compiled by the Food and Agriculture Organization (FAO) of the United Nation in 1983 [17]. Later, maximum limits (ML) of food additives and contaminants have been critically evaluated by FAO and WHO in their Codex Alimentarius Commission [18], and later adopted by national and/or international legislation bodies, such as the European Union [19]. For elemental environmental contaminants in fish, the ML are presently given only for the toxic elements Cd, Pb, Hg (and methylmercury) [19]. For guidance on usual levels of other elements, we also present in Table 6 values for marine fish compiled by Bowen [20].

It follows from a comparison of the mean element values determined in this work (the mean or median, whatever appropriate—cf. Tables 4 and 5) exceed the Bowen's values [20] for Mn in the in fish samples from all localities, for Rb and Sr in most sampling sites, and the values for Cu from most localities exceed the FAO values [17]. Sufficiently low LOD values for the toxic elements As, Pb and Hg document that the legal limits [17] or the ML values [19] are not exceeded. No such inference can be made for Cd, because much higher LOD than ML was obtained by INAA, i.e., it cannot be concluded whether consumers of fish from downstream of the river Mara may be exposed to an excessively high Cd concentration. More specific information on the concentrations of the elements As, Cd, Pb and Hg could be obtained using, e.g., neutron activation analysis with radiochemical neutron activation analysis (except for Pb), atomic absorption spectrometry or inductively coupled plasma mass spectrometry. However, these techniques, which provide lower LOD values for the above elements than those of INAA and/or EDXRF were not available for this study. Concentrations of other elements slightly deviating from values given in the literature [20] do not seem to present a serious health risk to their consumers, even if significantly higher in fish from downstream of the river Mara (Ca, Ni and Sr in lungfish, Na, Cl in catfish).

Conclusions

Sixteen elements, namely Na, Cl, K, Ca, Sc, Cr, Mn, Fe, Co, Ni, Cu, Zn, Se, Br, Sr and Rb, were detected in concentration above the LOD in samples of both fish species. The concentrations of the toxic elements As, Cd, Hg and Pb were found to be below LOD of the analytical techniques used in this study. Concentrations of more than 50 % of elements were higher in both lungfish and catfish caught downstream than those caught upstream of the river Mara. Out of these, the concentrations of Ca, Cr, Ni, and Cu in were significantly higher ($p \leq 0.05$) in lungfish, whereas the significantly higher concentrations in catfish were found for Na, Cl and Cu. This might indicate contamination of the downstream river due to the mining activities of NMGM. Hence, lungfish appears to be a better indicator of elemental contamination than catfish. It may be concluded that the contamination observed presents a moderate health risk to the concerned population in the river Mara basin, mainly because of the excessively high concentrations of Mn and Cu, and long-lasting contamination of the river since 2009.

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