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Technical Article

The Impact of Phosphate Mine Tailings on the Bioaccumulation of Heavy Metals in Marine Fish and Crustaceans from the Coastal Zone of Togo

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Abstract. About 2.5 million t of sedimentary phosphorite mine tailings, highly enriched with Cd, Cr, Cu, Ni, Fe, F, and Zn, are dumped annually in the coastal waters of Togo without any pre-treatment, causing serious pollution problems in the region. We conducted bioaccumulation investigations on fish and crustaceans sampled from the polluted coastal zone. The highest concentrations of metals in fish and crustacean were found close to the tailings outfall and the values decreased further away from the source of pollution. Compared to the international reference norms for seafood given by the WHO, Cd is enriched 10 to 168 fold, Pb 20 to 107 fold, Cu up to 5 fold, Fe up to 15 fold, and F up to 3 fold.

Key words: Bioaccumulation; heavy metals; marine pollution; phosphorite mine tailings; Togo

Introduction

Industrial and urban activities in the coastal zone are the main sources of heavy metals in the marine environment. Although heavy metals are natural components of the marine environment, their enrichment by anthropogenic activities disturbs the marine ecosystem's equilibrium and can affect human health (Berrow 1991; Miguel 2001).

The marine sedimentary phosphate deposits of Hahotoé-Kpogamé (southern Togo), like those elsewhere in the world, are highly enriched with numerous heavy metals, such as Cd, Cr, Cu, Ni, V, Zn, Pb, U, Th, Mo, Ag, F, Y, and rare earths (Altschuler 1980; Gnandi 1998; Johnson 1987, Kunkel 1990; Piper 1991). Togo produces about 3.5 million t of industrial raw phosphate annually and is the 5th largest producer of phosphate in the world. The processing of the Haotoé-Kpogamé phosphate ore to commercial grade is done mechanically by wet sieving using sea water in a factory close to the Kpémé beach. Two types of mine wastes are produced: fine-grained muddy tailings and coarse-grained wastes. About 40% of the raw ore is rejected as tailings during the processing. The muddy tailings, about 2.5 million t of sludge, are annually dumped directly into the coastal waters of Togo without any pre-treatment (Figure 1). The main effluent outfall is by Goumou Kopé, but when there are mechanical problems, the IFG-Togo (International Fertilizer Group-Togo) effluent is sometimes discharged by Kpémé, near the factory. Coarse wastes are discharged on the beach by Agbodrafo, where they are often washed

Figure 1. The dumping of heavy metal rich phosphorite mine tailings into the sea near Gomou-Kope, Togo

out into the sea. Solid wastes are also discharged on soils around the facility and sometimes are used to repair the roads in the surrounding villages.

This disposal greatly decreases the clarity of the seawater and contaminates coastal sediments and waters with heavy metals enriched in phophorites. The impact of these activities on the pollution of coastal sediments and the bioavailability of heavy metals in phosphorites were already investigated by the author (Gnandi and Tobschall 1999a, b). The aim of the present work was to assess the impact of these mining activities on the bioaccumulation of Cd, Pb, Cu, Fe, and F in the tissues of marine organisms (fish, shrimp, and crabs) in the nearby coastal zone.

Methodology

Sampling of marine organisms (fish and crustaceans) was carried out by fishing along the coast at five different sites: Agbodrafo, Kpeme, Goumou-Kopé, Lomé, and Aného (Figure 2). We sampled species (Table 1) that are often fished and eaten by the local population. Species were regrouped according to their age, and 40 g of each group (sections of each fish without head) were weighed for analysis. For comparison, two fish species (*Sardinella aurita* and *Galeoides decadactylus*) were sampled at a site near Lome, 35 km from the phosphate pollution source. Samples were aspersed with 1 ml H_2SO_4, and progressively (50°C/h) ashed at 700 ± 50°C in a furnace for 6 hours. After cooling, the ash was crushed and then homogenised. Then, 2g of ash were digested in a combination of $HClO_4$ - HNO_3 acids on a sand bath at 200°C (Jorhem 1993). The resulting solution was diluted to 100 mL volume with distilled water and then filtered through Whatman filters.

Seawater was sampled at five sites (Agbodrafo, Kpémé, Goumou-Kopé, Aného and Lomé) (Figure 2) and, in addition, seawater decanted from the mining effluents was analysed. Water samples were decanted and acidified with supra-pure HNO_3. The pH, T°C, and electrical conductivity were measured in the field using a portable Fisher Scientific pH-meter model 63500 equipped with Quattrode. Cadmium, Pb, Cu, Zn and Fe were determined using a Perkin Elmer 2380 atomic absorption spectrometer equipped with a graphite furnace oven at the Office of Mine and Geology in Lome. Fluorine was determined using a Fisher Scientific Aqua Fast IV portable colorimeter and test kits especially designed for this purpose. The exactitude and the accuracy of the procedures were checked using certified organic matter standard samples MA-A-2 (fish) from the IAEA, which were analyzed using the same method as our fish samples. The errors in metal

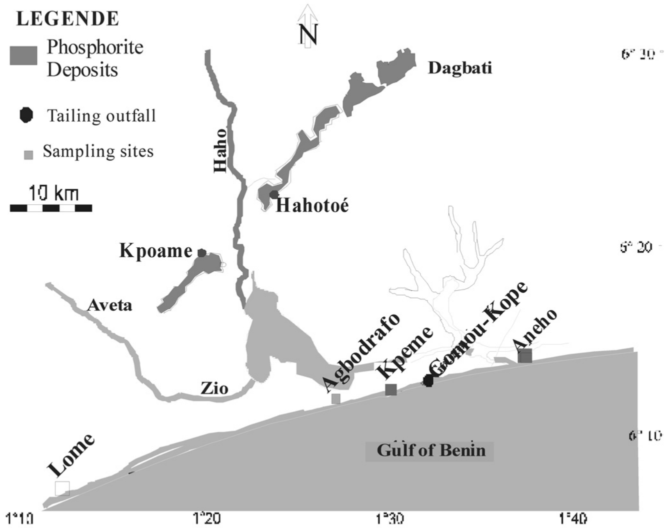

Figure 2. Location of the phosphorite deposits of Hahotoé-Kpogamé and the sampling sites

concentrations for the reference samples were 4% for F, 5% for Pb, 8% for Cd, 10% for Cu, 11% for Fe, and 14% for Zn.

Results

Chemical Characterization of the Hahotoé-Kpogamé Phosphorites

The raw ore was chemically characterised using phosphorite samples taken from the exploited Hahotoé-Kpogamé phosphatic layer. Table 2 shows that the phosphorites of Togo are naturally highly enriched with Cd, Cr, Cu, Ni, Zn, F, Fe, Mn, P, and Al (Gnandi and Tobschall 1999a). Toxic metals such as Cd have an enrichment factor of 18 compared to similar phosphatic deposits elsewhere in the world (Altschuler 1980; Wedepohl 1991). The heavy metal enrichment in phosphorite is caused by isomorphic substitutions for Ca^{2+} in the apatite structure and by adsorption onto iron

Table 1. The fish and crustacean species used for the analyses

Species	Name	Genus	Local name
	Fish		
Chloroscombrus chrysurus	*Sapater*	*Carangidae*	*Zozrui*
Sardinella aurita	*Allache*	*Clupeidae*	*Manvi*
Ilisha africana	*Alose razor*	*Clupeidae*	*Kaflan*
Galeoides decadactylus	*Small captain*	*Polynemidae*	*Tikoe*
Caranx latus	*Carangue*	*Carangidae*	*Glamata*
	Crustaceans		
Penaeus duorarum	*Rose shrimp*	*Penaedae*	*Bolou*
Callinectes pallidus	*Crab*	*Pordunidae*	*Aglan*
Cardisoma armatum	*Earth crab*	*Ocypodidae*	*Agoglan*

Table 2. Chemical composition of the raw phosphorite of Togo (major elements in %, trace elements in mg/kg) (Gnandi and Tobschall 1999a)

	Minimum	Maximum	Average
TiO_2	0.90	0.42	0.19
Al_2O_3	0.56	30.86	5.28
Fe_2O_3	0.002	16.04	3.87
MnO	0.09	0.09	0.02
MgO	6.89	0.57	0.23
CaO	0.19	48.32	34.12
Na_2O	0.04	0.52	0.28
K_2O	4.96	0.25	0.11
P_2O_5	0.02	40.89	28.37
F*	1.05	4.35	3.92
Cd	2	109	44
Cr	126	707	356
Cu	38	363	110
Ni	55	511	158
V	92	339	173
Zn	116	1869	465
Zr	19	93	46
Pb	13	160	67
Sr	207	18004	1429

by Kunkel 1990

and manganese oxides, clay, and calcite mineral surfaces (Jarvis and Al 1994; Nathan 1984; Prévôt 1990).

Chemical Characterization of the Coastal Zone Seawater

The pH of seawater, which is alkaline (8.28), becomes neutral (7.52 and 7.32) near the discharge point due to the low pH of the effluent water (6.5). The electrical conductivity (EC) also decreased, from 47,000 μS/cm for seawater far away from the tailings outfall to 4640 μS/cm for seawater near the discharge; the effluent water had an EC of 4.60 μS/cm (Table 3). The effluent water had high concentrations of Cd, Pb, Fe, F, Fe, Cu, and Zn compared to seawater. The concentrations of metals in seawater decrease as one moves away from the tailings outfall, which strongly suggest that the phosphorite wastewater is the main source of heavy metals in the studied coastal area. River input, urban effluents, atmospheric deposition, harbour industry, and other industrial sources also contribute to marine coastal pollution in the studied region.

Bioaccumulation of Heavy Metals in Fish

Table 4 lists the concentrations of heavy metals found in the studied marine species.

Cadmium (Cd): Cadmium is the second most toxic commonly found element after mercury (UNEP 1994). Compared to its concentrations in the earth's crust (0.13 mg/Kg, Wedepohl 1991), Cd is very abundant in the phosphorites of Togo, ranging between 2 and 109 mg/Kg (average 49 mg/kg). Cd is incorporated into the crystal lattices of the main phosphate mineral, apatite. Its bioavailability depends on the weathering state of the ore, which is moderate. The bioavailability test shows that high amounts of Cd are dissolved during the phosphorite/seawater contact. All studied species were strong contaminated with Cd; Cd concentrations ranged from 0.12 to 1.68 mg/kg, while the international reference norm given by the World Health Organization (WHO 1993) for seafood is 0.01 mg/kg. The level of contamination varies from one species to another; *Chloroscombrus chrysurus* appears to be a strong bioaccumulator for Cd. The Cd concentration in fish decreased further from the tailings outfall, showing that the effluents are the main cause of the strong bioaccumulation of this element in the studied species.

Lead (Pb): Classified as the third most toxic element commonly found in nature, lead is not very abundant in the phosphorites of Togo (average content, 67 mg/kg). Thus, Pb levels in the studied marine environment can

Table 3. Physical and chemical parameters of seawater in the study area

	Effluent Site	Agbodrafo Site 1	Kpémé Wharf Site 2	Goumou-Kopé Site 3	Aného Site 4	Lome
T°C	27.10	30	31.5	27	28.3	31.2
pH	6.5	7.52	7.32	8.28	8.21	8.30
conductivity (μS/cm)	42600	46400	46700	46800	47000	46 600
Cd (μg/L)	41	5.6	12	7	3.5	< 0.02
Pb (μg/L)	9	6.7	7	8.5	3.7	< 0.1
Fe (mg/L)	360.1	152	265	300	102	1
F (mg/L)	1.31	1.6	0.558	1.36	0.435	<0.2
Zn (mg/L)	0.982	3.11	0.417	0.678	0.791	<0.02
Cu (mg/L)	0.543	1.73	0.356	0.452	0.512	<0.05

Table 4. Heavy metal concentrations (in mg/Kg) in the tissues of fish species from the coastal zone of Togo contaminated by mine tailings

	Sampling sites					
	Agbodrafo	Kpeme	Goumou-Kope	Aneho	Lome	WHO
Species	**Concentrations of Cadmium (Cd) in mg/kg**					
Chloroscombrus chrysurus	0.25	0.36	1.68	0.12		
Sardinella aurita	0.24	0.24	1.00	0.12	< 0.020	0.01
Ilisha africana	0.38	0.38	0.46	0.13		
Galeoides decadactylus	0.25	0.46	1.06	0.12	< 0.020	
Caranx latus	0.28	0.37	0.58	0.12		
	Concentrations of Lead (Pb) in mg/kg					
Chloroscombrus chrysurus	10.37	8.10	5.99	8.19		
Sardinella aurita	5.99	8.03	6.01	6.14	< 0.100	0.1
Ilisha africana	8.34	6.26	6.31	8.40		
Galeoides decadactylus	8.23	6.27	8.49	8.04	< 0.100	
Caranx latus	8.25	6.21	6.25	8.27		
	Concentrations of Copper (Cu) in mg/kg					
Chloroscombrus chrysurus	1.82	2.26	1.8	0.97		
Sardinella aurita	2.58	2.18	2.45	0.98	< 0.05	1
Ilisha africana	2.86	2.16	2.20	1.06		
Galeoides decadactylus	3.86	2.41	3.86	1.98	< 0.05	
Caranx latus	4.75	2.41	4.10	2.25		
	Concentrations of Zinc (Zn) in mg/kg					
Chloroscombrus chrysurus	1.08	0.16	1.35	1.30		
Sardinella aurita	0.45	0.87	1.36	0.83	< 0.02	5
Ilisha africana	0.21	0.24	1.48	1.56		
Galeoides decadactylus	0.1	0.90	1.27	0.76	< 0.02	
Caranx latus	0.50	1.33	1.08	0.44		
	Concentrations of total Iron (Fe) in mg/kg					
Chloroscombrus chrysurus	0.68	1.36	1.81	0.83		
Sardinella aurita	1.37	2.33	1.97	0.68	1	0.2
Ilisha africana	0.62	2.22	2.99	0.31		
Galeoides decadactylus	0.23	1.20	1.44	0.78	1	
Caranx latus	0.74	1.89	2.87	0.20		
	Concentrations of Fluorine (F) in mg/kg					
Chloroscombrus chrysurus	2.61	1.08	2.30	1.12		
Sardinella aurita	1.93	2.58	3.92	0.19	0.2	1.5
Ilisha africana	1.11	0.26	0.89	0.63		
Galeoides decadactylus	0.68	1.44	1.37	0.62	0.2	
Caranx latus	1.54	0.93	4.56	0.18		

have other sources, e.g. river inputs, dumping of urban effluents, and atmospheric deposition. The use of leaded fuel in Togo and the very dense urban traffic in the maritime region of Togo can strongly effect the contamination of the Togo Sea. The Pb contents in the studied species ranged from 2 to 10.7 mg/kg, with factors of enrichment ranging from 20 to 107, compared to the WHO reference norms (0.1 mg/kg). Again, *C. chrysurus* is a strong bio-accumulator. Lead concentrations increase as one moves away from the tailings effluent, which is contrary to the distribution of Cd in the analysed organisms, suggesting that most of the lead pollution is not from the mine tailings.

Copper (Cu): Copper is an abundant element in the Togo phosphorites, with values ranging between 38 and 363 mg/kg (averaging 110 mg/kg). The Cu is associated with the clayey cement phase of the phosphorite. The Cu concentrations in the analysed tissues ranged from 0.97 to 4.75 mg/kg, compared to the WHO reference norms of 1 mg/kg, representing factors of enrichment of 1 to 4.75. The highest concentrations were found at Gomou-Kopé, Agbodrafo, and Kpémé and the concentrations decrease towards the east (Aneho). The bioaccumulation of Cu is highest in *Caranx latus*.

Zinc (Zn): Zn is also a very abundant element in Togo phosphorites, with concentrations ranging between 116 and 1869 mg/kg (averaging 465 mg/kg). It is also associated with the clayey matrix phase. The tissue concentrations ranged from 0.1 to 1.56 mg/kg, which are lower than the WHO international reference norms of 5 mg/kg. Nevertheless, the highest concentrations were measured in samples close to the Gomou-Kopé effluent outfall point. *Chloroscombrus chrysurus* shows the highest bioaccumulation for Zn.

Iron (Fe): Iron is a very abundant element in Togo phosphorites, ranging between 0.002 and 16.04% (as Fe_2O_3). In the analysed species, the concentrations ranged between 0.23 and 2.99 mg/kg (total Fe), compared to the WHO reference norms of 0.2 mg/kg; the enrichment factors ranged from 1-15. Generally, the highest concentrations were measured close to the discharges in Goumo-Kopé and in Kpémé and the concentrations decrease in areas far away from Kpéme and Gomou Kope.

Fluorine (F): Fluorine is also very abundant in Togo phosphorites, with contents ranging between 1.05 and 4.35%. Like Cd, F is incorporated in the apatite mineral phase, and its abundance confers the name of carbonate fluorapatite or francolite to the main mineral of the phosphorites of Togo. The concentrations in the analysed species ranged between 0.18 and 4.56, compared to the WHO reference norms of 1.5 mg/kg. Generally, the highest F concentrations were measured at Goumo-Kopé, Kpémé, and Agbodrafo; the values decrease towards the east, away from the pollution sources.

The Bioaccumulation of Cd and Pb in Crustaceans

Three shellfish species were analysed (Table 5). We found that the Pb accumulation was higher than that of Cd. The factors of enrichments varied from 14 to 24 for Cd, and from 84 to 104 for Pb, relative to the WHO reference norms. The high bioaccumulation of metals by these species can be related to their mode of feeding; they are in permanent contact with the sea-bed sediments, which are highly contaminated with tailings.

Table 5. Cd and Pb concentrations (mg/kg) in shrimp and crabs

	Penaeus duorarum	*Callinectes pallidus*	*Cardisoma armatum*
Cadmium	0.24	0.14	0.16
Lead	10.4	8.49	8.4

Discussion

Bioaccumulation in fish and crustacean was high for all of the analysed elements except Zn. The highest concentrations were found in areas near the discharge points (Agbodrafo, Kpeme, and Gomou-Kope) and concentrations were lower far away from the factory (Lome, Aneho) Metal contents in fish from a coastal area not polluted by phosphate tailings (Lome) are very low (Table 4), again suggesting that the mine waste is the main source of heavy metals for the marine pollution in Togo, though other potential metal sources include urban effluents, river input, atmospheric deposition, rural activities, coastal zone industrial activities, and harbour industry. Similar results were obtained when the coastal sediments of the same area were analysed; the concentrations of heavy metals increased (or decreased) away from the discharge point, according to their speciation (Gnandi and Tobschall 1999a). For example, the concentrations of Cd and F decreased away from the discharge point, because those elements are incorporated in the apatite structure, which settles rapidly because of the high density of apatite. Metals that are associated with the light clay minerals, such as Cu, Pb, Zn, and Fe, increased in concentration away from the discharge point because those minerals are light and are transported further before settling. Littoral drive plays an important role in sediment transport along the coast of Togo; the current is generally from the west to the east, but occasionally the direction can change from the east to the west.

Elements that are incorporated in the apatite structure are not easily bioavailable unless the ore is weathered. Earlier investigations have shown that the phosphorites of Togo are moderately weathered (Gnandi 1998; Kunkel 1990; Van Kauwenberg and McClellan 1990), which would make these elements partially bioavailable. Heavy metal bioavailability in a marine environment is also enhanced by their strong solubility in seawater and by the acidity of the mine effluents (Bourg 1983). Laboratory tests carried out by the author showed that in contact with seawater, heavy metals (Cd, Pb, Cu, Ni, Fe, Al, Mn, F) fixed by adsorption on sediment particles (manganese and iron oxides, clays, carbonates, organic matter, and phosphate minerals) are solubilised in high amounts in seawater (Gnandi and Tobscahall 1999b). The high concentrations of heavy metals found in the mine effluents attest to these results (Table 3).

The capacity of bioaccumulation of metals in such a polluted medium depends not only on the bioavailability of metals but also on the mode of feeding of the marine organisms (Giordano et al. 1991). Bioassimilation by marine organisms can occur through the food chain (bioaccumulation) or by direct assimilation (bioconcentration). Generally the level of bioaccumulation increases from the first to the last member in the food chain. The accumulation of heavy metal charged phosphate particles on the surfaces of the eaten chain members can significantly increase the degree of bioaccumulation by feeding, especially for species living in contact with the polluted sea bed sediments (shrimps, prawn, and some fish species). Thus, even though heavy metals may not be dissolved in the water, the presence of heavy metal enriched phosphate particles can lead to significant bioaccumulation by filter feeders (Table 5).

Conclusions and Recommendations

Our results show that the dumping of mining tailings into the Togo coastal waters seriously endangers the marine ecosystem and human health through its effects on the food chain. The mining company, IFG-Togo, considers the mine tailings as harmless and believes that they contribute to the development of the fishery in the region (Banou 2004). Our results indicate the opposite, and fishermen along the coast are claiming that the fish stocks are reduced since mining activities began. Dental fluorosis is endemic among the inhabitants of Kpeme, in particular among the children (Gnandi et al. in prep.). This illness is caused by the element fluorine, which is abundant in the Togo phosphorites. We recommend that the mining authorities should search for new methods to manage the mine wastes. We have demonstrated that treating the mine effluents by coagulation and flocculation greatly reduces their toxicity (Gnandi et al. 2005).

In research on marine pollution, Mythilus mussels are often used as suitable bio-indicators of the degree and extent of marine pollution (Kaimoussi and Al 2001). Future investigations will focus on this species.

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