MERCURY DISPERSAL IN WATER, SEDIMENTS AND AQUATIC BIOTA OF A GOLD MINING TAILING DEPOSIT DRAINAGE IN POCONE, BRAZIL

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Abstract. In the Pocone district, Brazil, Hg distribution was studied in a small watershed which drains tailings from a 10 yr old gold mining operation. Heavy regional rains are responsible for continuous weathering, thereby making it possible to transport Hg into the ecologically important Pantanal area. Mercury concentrations in creek sediments range from $<$ 0.02 to 0.18 mg. kg⁻¹. The highest concentrations occur close to the tailings deposit. Mercury concentrations in the water were always below the detection limit of the analytical method used $(<0.04 \mu g$. L⁻¹). Suspended matter samples collected before, during and after a storm, showed a Hg peak value of 0.61 mg kg^{-1} about 30 min after the event. Dissolved Hg concentration still fell below the detection limit. Among the biota, molluscs accumulated moderately high concentrations of Hg, while macrophytes and fish did not. Mercury concentrations in molluscs were dependent on size, with larger animals presenting higher Hg concentration. We conclude that Hg present in the tailings shows low mobility and that its eventual transport into the drainage system is dependent on the erosion of fine material from the wastes during rains, resulting in a restricted contamination of the area and low Hg concentration in the biota.

1. Introduction

Mercury is currently used in gold mining throughout the Brazilian North and Nortwestern regions, including the Amazon forest, the western Brazilian Savanna ('Cerrado' formation) and the northern swamps of the 'Pantanal" formation. According to recent estimates 90 to 120 t Hg are released annually in these regions (Pfeiffer and Lacerda, 1988).

A major concern of local environmental authorities is the potential for Hg contamination in complex ecosystems of great ecological and economic importance to the area. The fate of Hg is virtually unknown in these environments.

The Pocone mining region is located at the northern reach of the Pantanal, where typical savanna ('Cerrado') mixes with oxbow lakes, flood plains, bays, and lakes; it is therefore a region of major ecological and conservation concern.

During the last 10 yr gold mining has released 15 to 20 t Hg into the local environment. Mercury enters the environment directly associated with mining wastes and via atmospheric deposition of Hg emitted by roasting of the Au-Hg amalgam. The mining sites are dispersed throughout the area leaving the landscape dotted with tailings deposits. High Hg concentrations, up to 5 mg kg^{-1} , are found associated with these wastes. Tailings deposits are currently subject to leaching and weathering by local drainage and rains, comprising a potential, long term source of regional Hg contamination. Up to the present however the behavior of Hg in such deposits and its potential mobilization has not been investigated.

This study is part of a long term project of the Mineral Technology Center (CETEM) to investigate the fate of Hg in the region. We report here on Hg distribution and mobilization in water, sediments and aquatic biota (macrophyte, mollusc and fish) of a typical tailings area, the 'Tanque dos Padres' deposit and its drainage, in order to evaluate its potential availability for contamination in the local ecosystems.

2. Study area

The 'Tanque dos Padres' is a typical tailing deposit similar to others present in the region, located in the Pocone Municipality, (longitude 50 °C 07' 32" W and latitude 16 °C 15' 43" S), approximately 100 km from Cuiabá, capital of the Mato Grosso State (Figure 1). Tailings amount 300000 m^{-3} of exposed soils, including an artifical lake of *ca* 2 ha and a drainage basin of *ca* 500 ha (Figure 2).

Mercury is present mostly as Hg^0 randomly distributed in the tailings with concentrations between 0.04 and 0.23 mg kg^{-1} , amounting a total Hg content of 40 to 90 kg (CETEM, 1989). In small areas $(1 \text{ to } 4 \text{ m}^{-2})$ spread throughout the deposit, where old ore concentrating sites exist, Hg concentration can reach up to 5 mg kg^{-1} dw of mine waste. In these areas Hg drops can be easily seen in concentrates of the tailing material (CETEM, 1989).

The tailings are drained by a small creek with water flow varing from 16 to 35 L s⁻¹ depending on the season. During heavy rains, fine clay particles are transported form the tailings, reaching values as high as 70 mg L^{-1} . During most of the year however suspended matter load is only 5 mg L^{-1} . Details on the gold mining procedure and on Hg distribution in the tailings deposit itself are published elsewhere (CETEM, 1989).

The local aquatic environment is characterized by small creeks draining flood plains, dotted with small permanent and seasonal lakes, which play an important role in retaining sediments eroded from tailings. These water bodies are dominated by rooted and floating macrophytes. One of the dominant species is the rooted macrophyte *Pontederia lanceolata* Nutt.. The freshwater mollusc, *Ampullarius* sp. is the most frequent benthic animal in the entire drainage. These two organisms are important food items to most birds and fish in the area.

3. Material and methods

Water, sediment, macrophyte, molluscs and fish samples were collected along the drainage of the 'Tanque dos Padres' tailings deposit and the upper reaches of the Pantanal, potentially affected by this drainage (Figure 1).

Water samples were collected in 500 ml polyethylene bottles previously washed

Fig. 1. Location of the study site and sampling points at the 'Tanque dos Padres' drainage. Sediment and water samples were collected in all points with the exception of BG-10. Notation for biological samples are: (M) the macrophyte *Pontederia lanceolata;* (A) the mollusc *Ampularius* sp.; and (F) fish.

in 1N HCl. Water samples were preserved in the field with 2 ml of $K_2Cr_2O_7 0.01\%$ and 2 ml of concentrated $HNO₃$. These samples included dissolved and particulate Hg. Surface sediments (0 to 5 cm) were collected by inserting small plastic cores in bottom sediments of creeks and small lakes. Samples were frozen in the field for transport. In the laboratory, samples were sieved (\leq 74 μ m) and dried at 50 °C to constant weight. Sub-samples of 2.0 g dw were digested with 20 mL of 'aquaregia' for 5 min at 60 °C in closed system. Details of sample preservation and treatment are the same as in Hinton *et al.* (1987), with small modifications as proposed by Malm *et al.* (1989).

At the main effluent from the tailings deposit (Station MA-08; Figure 1) water samples were collected for 48 hr during a rain storm. From these samples, suspended

material was separated by passage through $0.45 \mu m$ Millipore filters, and both dissolved and particulate Hg were analyzed according to the procedures described above.

Macrophytes *(Pontederia lanceolata* Nutt.) and molluscs *(Ampullarius* sp.) were collected by hand along the entire drainage. Care was taken to collect molluscs of the same size, since size is an important determinant of Hg concentration in these animals (Mohlenberg and Riisgard, 1988). In the lake inside the tailings deposit, 60 individuals of different size were collected and pooled in 5 size classes to study Hg intraspecies variability. Fish, however, were only collected in the Bento Gomes River using hand nets, since most species do not occur along the small creeks of the drainage (Figure 1). Twelve individuals of the top predator *Serrasaulus natterii* ('piranha') of different size were also collected to study Hg intraspecific variability.

All biological samples were fully washed in local water, packed in plastic bags and frozen in the field for transport. In the laboratory, macrophytes were oven dried to constant weight (50 °C), while muscle tissues of fish and soft tissues of molluscs were digested without previous drying. Animal and plant samples were digested at 60 °C with concentrated H_2S_4O and H_2O_2 . After cooling, the samples were further oxidized with 5% KMnO₄ and 5% K₂S₂O₈ for 24 hr according to Agemian and Chau (1978).

All Hg analysis were done by cold vapor atomic absorption spectrophotometry using a Varian Techtron AA-1475 spectrophotometer and a Varian VGA-76 vapor generator. Reproducibility and accuracy were determined by means of triplicate analyses and analyses of certified reference standards (Malm *et al.,* 1989). The detection limit of the method used was determined according to Slavin *et al.* (1972) and Hinton *et al.* (1987), reaching 0.04 μ g L⁻¹ for water; 0.02 mg kg⁻¹ for sediments; 0.04 mg kg⁻¹ ww for fish and mollusc and 0.04 mg kg⁻¹ dw for plants.

In all sampling stations, water pH, Eh, electrical conductivity and temperature were measured using portable electrodes.

4. Results and Discussion

Major physico-chemical parameters and Hg concentrations in water and sediments are presented in Table I. The water of the entire drainage is slightly acidic (pH $= 6.5$ to 6.9); oxidized (Eh = +136 to +283 mV); warm (27 to 31 °C) and low to medium in conductivity (55 to 159 μ S cm⁻¹). These values are typical for most water bodies in the region (Silva and Pinto-Silva, 1989). Mercury concentrations in water were always below the detection limit of the method used $(< 0.04 \mu g$ L^{-1}), regardless of the distance relative to the tailing deposit. Under the physicochemical conditions prevailing in the drainage, Hg presents very low solubility (Lindqvist *et aL,* 1984; Nriagu, 1979), therefore the low dissolved Hg concentrations were expected. In sediments however, a clear gradient of Hg concentration occurred, with higher Hg concentration (0.18 mg kg^{-1}) close to tailing deposit, decreasing

sediments (mean of 3 sub-samples) of the 'Tanque dos Padros' drainage Station T pH Eh Cond. Hg water Hg sediment

^oC mV uS cm⁻¹ ug L⁻¹ mg kg⁻¹ dw mV μ S cm⁻¹ μ g L⁻¹ mg kg⁻¹ dw

MA-08 30 6.7 136 95 <0.04 0.18 FL-05 30 6.9 275 112 < 0.04 0.06 VA-04 31 6.7 243 119 <0.04 0.12 FA-03 28 6.5 255 84 ≤ 0.04 0.11 OL-02 28 6.6 258 90 <0.04 0.08 $CA-01$ 28 6.8 263 55 <0.04 <0.02 PE-07 27 6.8 283 159 <0.04 <0.02 DO-06 29 6.8 267 135 <0.04 0.04 PT-11 28 6.5 266 102 < 0.04 < 0.02

TABLE I

Sediment Hg concentrations in the Tanque dos Padres drainage are lower than those considered as the mean Hg concentrations in surface lake sediments of temperate region, namely ca. 0.33 mg kg^{-1} (Andren and Nriagu, 1979), where these water bodies are exposed to elevated Hg atmospheric inputs from anthropogenic sources (Nriagu, 1979). The concentrations however are slightly higher than background values found in temperate areas of ca. $0.10 \text{ mg} \text{ kg}^{-1}$ (Evans, 1986). Although much lower values were found in sediments far from the tailings deposit (Station PT-11). Therefore, comparisons with published data from north temperate, regions, characterized by strong industrial development, and frequently with mineralization of Hg-ore deposits are very difficult.

A comparison of Hg concentrations in water and sediments found in this study and those reported for other gold mining areas in Brazil, where Hg is used in gold recovery is presented in Table II. Mercury concentrations in water and sediments

Mercury concentration in water and sediments from the 'Tanque dos Padres' tailing area compared with other contaminated gold mining areas in Brazil

TABLE II

TABLE III

			storm collected at the main effluent creek (Station MA–08) of the 'Tanque dos Padres' tailing deposit. Mean values of three determinations					
Time	flow $L s^{-1}$	т °C	ph	Eh mV	Cond. μ S cm ⁻¹	TSS $mg L^{-1}$	Hg dis. ppb	Hg partic. $mg \, kg^{-1} \, dw$
-24 hr	16.8	32	6.8	280	72	6.0	< 0.04	
-30 min	16.7	32	6.9	260	71	6.0	< 0.04	< 0.02
$+10$ min	22.5	32	5.2	430	71	8.0	< 0.04	0.17
$+30$ min	23.0	31	5.1	420	72	66.0	< 0.04	0.61
$+4hr$	20.1	28	7.5	90	71	9.0	< 0.04	
$+24$ hr	18.0	33	7.0	230	71	6.0	< 0.04	< 0.02

Major physico-chemical parameters and Hg concentration in water and suspended matter during a

of the 'Tanque dos Padres' tailing area are considerably lower than in other contaminated areas, in particular those in the Amazon Region. In areas such as tha Mutum Parana and Madeira rivers, a total of 25 t Hg are released annually (Lacerda *et al.,* 1989), which is higher than the total amount of Hg released in the Pocone region during the last 10 yr. However, Hg concentrations in the area are slightly higher than in non-contaminated Amazon rivers.

Tabel III presents Hg concentrations in water and suspended particles, as well as major physico-chemical parameters in water draining the tailing deposit during a storm event. Major changes occurred in pH, Eh and total suspended solids (TSS) and Hg concentration in suspended particles. The water becomes progressively more acidic and oxidized, and suspended particles reach values 10 times higher than during dry conditions. Hg concentrations in suspended particles increases from < 0.02 to 0.61 mg kg⁻¹, although no modification could be found in Hg dissolved concentrations. These results show the capacity of rain to erode fine particles, enriched with Hg, from the tailings, suggesting that erosion, followed by Hg transport associated with suspended particles is the main pathway of Hg contamination of the drainage system. However as suggested by the gradient found in Hg concentrations in sediments, these Hg-rich suspended particles settle and are deposited near the tailings, restricting the zone of Hg contamination.

Siegel *et al.,* (1985) and Mudroch and Clair (1986) found similar Hg distribution patterns in waters draining tailing deposits in Canada. They have also suggested that contamination of sediments was due to transport and deposition of suspended particles brought into the drainage by erosion of mining wastes, and that these areas are intensively polluted locally but the polluting species are not highly mobile (Siegel *et al.,* 1985). Therefore, it seems that the results found in this study represents a typical distribution pattern of Hg in areas draining tailing deposits, that Hg presents very low mobility in the area and that most of Hg transport is associated with suspended particles. Thus, controlling waste erosion and suspended particle transport should be quite efficient in preventing Hg contamination in areas surrounding tailing deposits.

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TABLE IV

Mercury concentrations in macrophytes *Pontederia lanceolata* Nutt. callected in the 'Tanque dos Padres' drainage. Mean values of 5 samples in mg $kg⁻¹$ dw. Sediment Hg concentrations are mean values in mg $kg^{-1}dw$

MACROPHYTES

Table IV shows Hg concentration in macrophytes, P. *lanceolata,* along the drainage of the 'Tanque dos Padres' tailing deposit. The results showed low concentrations of Hg throughout the drainage, even in sampling points close to the tailings. Also, no relationship between Hg concentrations in sediments and macrophytes was found.

Macrophytes absorb Hg as the ion Hg^{2} , and present concentration factors (plant-Hg/soil-Hg) ranging from 0.1 to 10.0 depending on environmental Hg levels. In general higher concentration factors are found when low environmental Hg concentrations are present (Siegel *et al.,* 1987). Here we also found higher leaf/sediment concentration factors at low sediment Hg, however the high variability of the data hampers firm conclusions.

The low concentrations found even in sites closest to the tailings, suggest that Fig is exported strongly bound to suspended particles which would keep Hg unavailable to macrophytes. Similar results have been found in rooted macrophytes occurring in Hg-rich tailings elsewhere (Lane *et al.,* 1988).

Few data exists in literature on Hg content of macrophytes growing in areas affected by tailings associated with gold mining. Martinelli *et al.* (1988) reported Hg content in three species of Amazonian macrophytes from the Madeira River, a highly contaminated watershed in the Brazilian Amazon (Pfeiffer *et al.,* 1989a). They found similar results for rooted macrophytes to those reported in our study. However, concentrations up to 1.04 μ g g⁻¹ dw were found in floating plants probably as a result of incorporation of Hg present in Hg-enriched suspended material. Aquatic macrophytes growing on mine tailings in Canada, show Hg shoot concentrations ranging form 0.18 to 0.55 μ g g⁻¹. However root Hg concentrations reached much higher values (up to 6.1 μ g g⁻¹) (Lane *et al.*, 1988). Therefore, although the Hg content measured in the macrophytes from Pocone region was comparatively low, it may represent an underestimation of macrophyte contamination, since neither roots or floating species have been sampled in this study.

MOLLUSCS

Mercury concentrations in the mollusc *Ampularius* sp. are presented in Table V. Concentrations ranged from ≤ 0.04 to 0.93 mg kg⁻¹, depending on sample location. Samples collected close to the tailings presented much higher concentrations, decreasing sharply as the distance from the railings increases, similar to Hg distributions in bottom sediments. Proportional response to environmental Hg levels has been reported for many mollusc species and is one of the main reazons for using these animals as biological monitors for heavy metal contamination (Mohlenberg and Riisgard, 1988).

Table VI shows Hg distribution in *Ampullarius* sp. collected in the most contaminated site in the drainage. Mercury concentrations ranged from 0.43 ppm in the smallest individuals to 0.95 mg kg⁻¹ in the larger individuals. There is a significant correlationship between Hg concentration and shell size $(r=0.938,$ $p<0.05$), indicating that age is also an important factor determining Hg concentrations in this species. Relationship between Hg concentration and animal age has been previously reported (Davies *et al.,* 1979; Riisgard and Fame, 1988; Mohlenberg and Riisgard, 1988). These authors suggest that a probable cause for the phenomenon is the progressive accumulation of organic Hg, mostly alkyl-Hg

TABLE V

Mercury concentrations in the mollusc *(Ampullarius* sp.) from the 'Tanque dos Padres' drainage. Mean values of 5 samples in mg kg^{-1} ww

Station	Hg concentration	
MA-08	0.93 ± 0.09	
FA-03	0.10 ± 0.01	
VG-04	0.11 ± 0.02	
OL-02	0.08 ± 0.01	
PT-11	0.04 ± 0.01	
BG–10	< 0.04	

TABLE VI

Mercury concentrations in the mollusc *Ampullarius* sp. of different size classes. Mean values of three samples in mg kg^{-1} ww

	Shell size (cm) Hg concentration
-25	$0.43 + 0.07$
$2.5 - 3.0$	$0.70 + 0.03$
$3.0 - 3.5$	$0.73 + 0.29$
$3.5 - 4.0$	0.65 ± 0.30
$4.0 - 4.5$	$0.95 + 0.07$

compounds, in older individuals since in some species alkyl-Hg presents much longer residence times than inorganic Hg (Flower *et al.,* 1978; Cunningham and Tripp, 1975; Riisgard and Famme, 1986). Therefore, our results may suggest that Hg methylation is currently occurring in the area closest to the tailings, since Hg concentrations in both water and sediments are in general very low, even in the most contaminated sites of the drainage.

FISH

Table VII presents Hg concentration in fish species collected in the Bento Gomes River. The results show very low concentrations of Hg in all fish species sampled, ranging from ≤ 0.04 to 0.16 mg kg⁻¹. In general, carnivorous species contained more Hg than species at other trophic levels. However, the analysis of the 12 samples of *S. natterii* ('piranha'), a top carnivore, showed no correlation between fish size and Hg concentration. The low Hg concentrations found probably reflect the characteristics of the sampling site. The Bento Gomes river has a water flow of ca. 140000 L s⁻¹ which is at least 1500 times higher than the mean water flow of the Tanque dos Padres drainage which varies from 17 to 65 L s^{-1} , therefore, representing a huge dilution capacity for any Hg present in the drainage. Also, Hg is exported from the wastes mostly associated with suspended particles which are efficiently trapped by the various flood plains, swamps and lakes present in the drainage (CETEM, 1989; Lacerda *et al.,* 1990). Therefore, low Hg concentrations in these fish samples were expected.

Contamination of aquatic organisms, in particular of fish, by Hg released in the environment by the gold mining activity has been reported in various Amazonian rivers. In the Madeira River, the larger tributary of the Amazon River, Pfeiffer *et al.* (1989a) reported Hg concentrations up to 2.7 mg kg^{-1} ww in carnivorous fish. In the Carajás Mining District in Northern Pará State, Hg concentrations in carnivorous fish ranged from 1.01 to 2.19 mg kg^{-1} (Fernandes *et al.,* 1990). These high concentrations reflect in part the large Hg load in the environment

TABLE VII

Mercury concentrations in fish from the Bento Gomes River. Mean values of three samples, *S. natterii* was 12 samples, in mg kg⁻¹ ww

Fish species	Food habit	Hg concentration	
Panaque sp.	iliofagus	< 0.04	
Metynnis hypsauchen	herbivorous	< 0.04	
Potamotrygon motoro	carnivorous	< 0.04	
Prochilodus nigricans	iliophagus	< 0.04	
Leporinus friderici	herbivorous	< 0.04	
Serrasalmus eigenmanni	carnivorous	0.04 ± 0.06	
Serrasalmus rhombeus	carnivorous	0.06 ± 0.01	
Serrasalmus nattereri	carnivorous	0.06 ± 0.01	
Hemisorubim platyrhynchus	carnivorous	0.08 ± 0.02	
Cichlasoma spectabile	carnivorous	0.16 ± 0.03	

due to gold mining activities. However, they also are a result of the major environmental conditions of most Amazonian aquatic ecosystems, which favor high rates of Hg methylation, and therefore accumulation in hight trophic level fish species (Lacerda *et al.,* 1989; 1990).

Mercury concentrations found in fishes of the 'Tanque dos Padres' drainage are in general much lower than those found for Amazonian areas, where carnivorous fish carry Hg contents form 1 to 2 orders of magnitude higher than those found in the Pocone region.

In conclusion, Hg contamination of the local biota reflects the low Hg mobility from the tailing deposits in the Pocone region, which results in small dispersal along the drainage. Therefore, Hg contamination of the local biota seems to be restricted. Notwithstanding that these species are not in the diet of the local human population, their contamination can represent a potential threat to the local ecosystems, as they are, due to their abundance, important components of the local food webs.

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