



Mercury accumulation in sediment cores and along food chains in two regions of the Brazilian Pantanal

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Abstract

The Pantanal is a 140,000 km² floodplain wetland stretching across western Brazil and parts of Bolivia and Paraguay. Gold mining with mercury (Hg) amalgamation has thrived since 1980 along its northern rim. We quantified Hg accumulation in sediment cores (N = 5) and food chains in this general region of the northern Pantanal and in a reference region, 200 km deeper into the wetland (Acurizal). Cores were dated with ²¹⁰Pb and ¹³⁷Cs using direct gamma-assay. Total Hg was analyzed by cold-vapor atomic fluorescence using a gold-mesh pre-concentration trap. Average pre-1940 Hg accumulation in cores was not significantly different (N = 5, $p = 0.14$) between both regions and comparable with rates calculated for global reference sites. Post gold-rush Hg (post-1980) deposition averaged $55 \pm 11.3 \mu\text{g m}^{-2} \text{yr}^{-1}$ in the northern impacted region and was more than 1.5 times higher than the post-1980 rate in Acurizal, implying a regional Hg effect of gold mining. Post-1980 Hg accumulation in Acurizal, in turn, was 2.1 times the rate reported for a global reference during that time period, suggesting an additional basin-wide effect over such reference sites. By combining our core data with assessments of the size of the impacted area and the amount of Hg released to the region since 1980, we estimated that only 2–8% of this Hg was recovered as a sedimentary signal. The remainder of the Hg was lost to the atmosphere, downstream areas, or stored in biota. Hg concentrations in surface sediments in the northern Pantanal ($45.5 \pm 5.5 \text{ ng g}_{\text{dry}}^{-1}$) were significantly higher than those in our reference region ($29.1 \pm 0.7 \text{ ng g}_{\text{dry}}^{-1}$). Hg levels in primary producers were also elevated in the northern Pantanal. *Eichhornia crassipes* roots contained 2.7–3.0 times more mercury than shoots in both regions and *Salvinia auriculata*, suggested as a biological monitor for Hg pollution, contained almost four times more mercury in the northern Pantanal ($90.7 \pm 9.1 \text{ ng g}_{\text{dry}}^{-1}$) than in Acurizal ($24.5 \pm 3.3 \text{ ng g}_{\text{dry}}^{-1}$). Plant grazers and scavengers, such as apple snails (*Pomacea* sp.) and adult water beetles (Fam. Hydrophilidae), were low in Hg, confirming previous data showing that the channeling of mercury from lower to higher trophic levels along herbivorous links was inefficient compared to transfer along carnivorous links. Collections of 12–16 individuals of four species of Characidae (*Aphyocharax* sp., *Tetragonopterus* sp., *Serrasalmus spiroleura* and *Pygocentris nattereri*) in both regions showed elevated Hg body burdens in both piranhas *S. spiroleura* and *P. nattereri* from the northern Pantanal (149.9 ± 84.2 and $302.2 \pm 159.1 \text{ ng g}_{\text{dry}}^{-1}$, respectively). Fish length for each species was not statistically different between regions. *P. nattereri* length correlated significantly ($p < 0.001$) with Hg content in both regions, but the slope of the regression in the northern Pantanal was 2.6 times the slope for the Acurizal collection, indicating an elevated rate of biomagnification in the Hg-impacted region. Signals of Hg use in mining can be quantified in sediment core chronologies and biological tissues, although species at different trophic levels show dissimilar impacts. Mechanisms involved in Hg magnification along food chains deserve more attention, particularly in tropical regions where the threat of chronic exposure to this neurotoxin may have the greatest implications for biodiversity.

Introduction

Mercury is a potent neurotoxin that readily bioconcentrates and biomagnifies. In the United States, sixty percent of the fish consumption advisories issued are for mercury contamination (USEPA, 1997). Globally, the major source of mercury to freshwater systems is atmospheric deposition from natural and anthropogenic sources (Swain et al. 1992) but discharges from industry, mining activities and direct watershed runoff may increase regional loadings (Gill and Bruland 1990; Lacerda et al., 1991a).

The vast majority of ecological studies on patterns of mercury accumulation are from the temperate zone. Mercury concentrations in sediment and biological samples have been documented in only a few tropical aquatic ecosystems (Cuvin-Aralar, 1990; Lacerda, 1991b; Filho and Maddock, 1997; Olivero and Solano, 1998; Guimaraes et al., 1998; Mauro et al., 1999) and long-term trends in mercury accumulation in the tropics are essentially unknown. In the tropical environment, with potentially high rates of methylation (Mauro et al., 1999) and poorly-controlled mercury use and discharges, the threat of chronic mercury toxicity for longer-lived organisms at higher trophic levels may be particularly severe. In addition, the implications of chronic exposure to mercury on biocomplexity in the tropics have not been evaluated.

We selected the Pantanal, one of the world's largest tropical wetland systems, to measure long-term trends in mercury accumulation and to quantify patterns of mercury transfer in food webs. Covering an area of some 140,000 km² in the upstream basin of the Paraguay River, the Pantanal stretches across western Brazil and parts of Bolivia and Paraguay. Plant and animal life in this savanna type wetland are strongly influenced by distinct seasonal flooding with maximum water level fluctuations of five meters between the dry and rainy seasons. Periods of severe flooding are followed by extreme droughts with only a small portion of the wetland remaining inundated year-round. The Pantanal is famous for the abundance and diversity of its fauna. Hyacinth macaws, jaguars, giant river otters and Victoria lilies prosper in this floodplain.

Deforestation, expanding agriculture, illegal hunting and fishing, drainage activities associated with a planned waterway project and mercury contamination associated with gold mining threaten the Pantanal's remarkable diversity (Alho et al., 1988). After an initial gold rush several centuries ago, gold mining returned to the northern edge of the Pantanal near Poconé in

the early 1980s. The Rio Bento Gomes, in the Poconé region, receives drainage from mining and ore dressing sites (Filho and Maddock, 1997) and an estimated 15–20 tons of mercury was released into the local environment between 1980 and 1990 (Lacerda et al., 1991a). In all of Brazil, gold refining utilizes more mercury (168 tons year⁻¹) than the industrial sector (105 tons year⁻¹) (Ferreira and Appel, 1992). These mines in the northern Pantanal region have remained active in the 1990s.

Trends in mercury deposition over decades to centuries have been successfully inferred from natural archives such as sediment cores (summarized in EPRI, 1996). Mercury accumulation rates over time can now be calculated from sediment cores analyzed stratigraphically for mercury and dated by ²¹⁰Pb and other radionuclides such as ¹³⁷Cs. As a measure of environmental input, results presented as such rates of accumulation are superior to those presented in units of concentration. The latter, expressed as a relative measure of sediment composition (mg g⁻¹), is the conventional expression of sediment stratigraphy. Such data, however, are vulnerable to variation in sedimentation of other components. These variations may result in the dilution of the target analyte and underestimation of its environmental input. Accumulation is normalized over time, thus avoiding the problem of covariance among different sedimentary components.

Food web transfer is important for the accumulation of mercury and the highest levels occur in the longer-lived species in upper trophic levels (Cabana et al., 1994). The majority of mercury invades aquatic systems in inorganic form. Methyl-mercury, however, is the most toxic and bioaccumulated form of the different mercury species and predators, such as some fish, contain primarily methyl-mercury. In spite of the acute and chronic impact of mercury contamination, we have an incomplete understanding of the transfer of mercury between trophic levels (Hill et al., 1996), particularly in tropical regions with less information on the ecology and natural history of species (Cleckner et al., 1998; Por, 1995). Quantifying mercury dynamics in lower and intermediate trophic levels seems particularly important to clarify the efficiency of mercury biomagnification.

In this work, we compare records of mercury accumulation in sediment cores and mercury dynamics in food webs in the region of gold mining in the northern Pantanal to a reference area 200 km deeper into this floodplain. This is the first study to report detailed radionuclide-dated mercury accumulation profiles for

the Pantanal and to document mercury levels in a variety of organisms linked in *in situ* food webs. We test the hypothesis that mercury accumulation rates are elevated in northern Pantanal sediments deposited since the return of gold mining to that region in 1980. In addition, we predict that tissue concentrations in biota will be higher in the northern Pantanal than levels in the same taxa at the reference sites.

Methods

Site description

We sampled in three sites in the region of gold mining and in two reference sites deeper into the floodplain (Figure 1) and selected wetland locations with minimum disturbance and with maximum water depth. Sites within 25 km of the Poconé mining region (northern Pantanal) included a marsh near Santo Antonio do Leverger (15°51'30"S / 56°05'30"W), a floodplain depression adjacent to the Rio Bento Gomes (16°19'00"S / 56°32'00"W) and a shallow lake (Baia Piuval, Fazenda Ipiranga) on the Rio Bento Gomes (16°22'30"S / 56°34'00"W) approximately 20 km south of Poconé. The reference marshes were located about 200 km into the floodplain near Acurizal (Acurizal 1: 17°50'11"S / 57°32'21"W; Acurizal 2: 17°49'59"S / 57°32'54"W), in an area owned and managed by the Ecotropica Foundation, Cuiaba, Brazil. Water depth at the time of sampling varied from 36 cm (Sto. Antonio and Bento Gomes) to 125 cm (Acurizal 2). Vegetation at all sites was dominated by floating aquatic species (including *Eichhornia azurea*, *E. crassipes*, *Salvinia auriculata*, *Azolla* sp. and *Pistia stratioides*), common in neotropical systems with highly-fluctuating water levels.

Sample collection

At each site, we took a sediment core using a thin-walled polycarbonate tube (i.d. 7.5 cm) and carefully selected biological samples to represent different trophic levels along a food chain. We focused on selecting the same species, number of individuals and size class in each sample location. Observations of the cores were made in the field to detect changes in sediment color or texture. Cores were carefully extruded *in situ* in 1 or 2 cm sections, stored in plastic bags and frozen. Plants and fish were identified to genus or species and frozen. All samples were handled using clean field techniques. *Aphyocharax* sp. was

analyzed whole. The larger fish species (*Tetragonopterus* sp., *Serrasalmus spiroleura* and *Pygocentris nattereri*) were analyzed as filets. We compared whole body mercury concentrations directly to filet mercury concentrations following Goldstein et al. (1996) who demonstrated a consistent 1 to 1 ratio between whole body and filet mercury content in Minnesota fish species. Standard lengths (i.e., length from nose to tail base) were not significantly different between sites for the fish *Aphyocharax* sp., *Tetragonopterus* sp., *Serrasalmus spiroleura* and *Pygocentris nattereri*.

Dating of sediment cores

Radioactive lead (^{210}Pb) is used widely to determine pollution chronologies in aquatic sediments. ^{210}Pb is part of the uranium-238 (^{238}U) decay series where it follows radon-222 (^{222}Rn), an inert gas that emanates from the earth's crust into the atmosphere. Because of the long half-life of ^{238}U (4.51×10^9 yr) and its widespread occurrence in the earth's crust, ^{222}Rn emanation is assumed to occur at a constant rate over time. ^{222}Rn decays in the atmosphere through its short-lived daughters to ^{210}Pb , which is deposited on land and water by both wet precipitation and dry fallout processes. Once deposited in aquatic systems, ^{210}Pb is buried by subsequent sedimentation. The activity of ^{210}Pb in each sediment layer declines with its age due to radioactive decay. Because of the relatively short half-life (22.3 yrs.) of ^{210}Pb , this technique is restricted to sediments deposited within the last 100 years (Gottgens et al., 1999). The ^{210}Pb activity in older sediment layers results from and is maintained ('supported') by continued decay of parent radionuclides contained in the soil. Subtracting the level of supported ^{210}Pb from the total ^{210}Pb activity yields the 'unsupported' activity upon which age-depth relationships are based.

We used direct low-background, high-purity germanium γ -counting (14.5 \times 40.0 mm well detector) following techniques described earlier (Gäggeler et al., 1976; Appleby et al., 1986) and summarized in EPRI (1996). The detector operates at a positive bias of 1800 Volts and at a temperature near that of liquid nitrogen provided by a high vacuum cryostat-dewar system. Counts for regions of interest are obtained with a 4096-channel analyzer calibrated at 0.186 keV/channel. γ -Counting allows simultaneous assay for supported and unsupported ^{210}Pb , as well as other radionuclides (such as ^{137}Cs), which may serve as ancillary age-markers. Additional specifics on the detector system, such as background suppression,

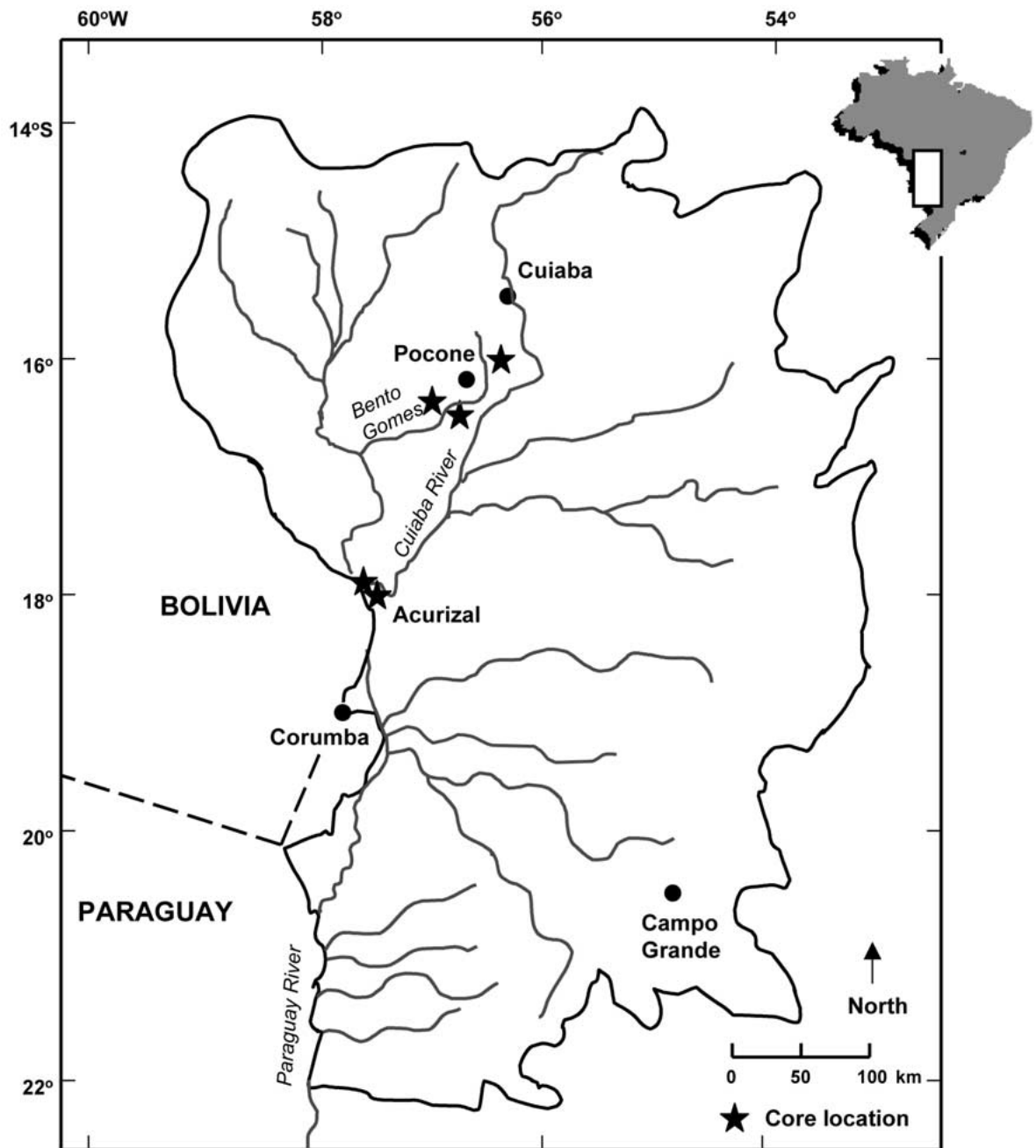


Figure 1. Map of the Pantanal with the five core locations indicated.

energy calibration and the computation of counting efficiencies may be found in Gottgens et al. (1999).

The samples were dried at 104 °C for 24 hours, pulverized and placed in low-density polypropylene vials. They were subsequently sealed with plastic cement and stored for a minimum of 21 days to equilibrate radon (^{222}Rn) with radium (^{226}Ra). Counting times per sample varied and were as long as 72 hours depending on the weight of the sample; small samples needed longer counting times to reduce error. Sample spectra were analyzed following Gottgens et al. (1999). Calculation of ^{210}Pb dates followed the Constant Rate of Supply model (Appleby and Oldfield, 1978).

^{137}Cs activities served as independent marker horizon in the sedimentary profile. The first occurrence of this man-made nuclide in the profile generally coincides with the onset of widespread atmospheric testing of nuclear weapons (i.e. 1954). 1959–1960 and 1963–1964 are reported as periods of maximum fallout in the northern hemisphere (Ritchie et al., 1973; Joshi, 1989) and additional support for ^{210}Pb -derived age/depth relationships may come from matching peak- ^{137}Cs activities in the profile with these time periods.

Mercury analysis

Mercury was analyzed using cold-vapor atomic fluorescence spectroscopy (CVAFS) at 253.7 nm (USEPA 1995) on a Tekran[®] Detector (model 2500). Sediment and plant material were air-dried and pulverized. Fish and other tissues were first analyzed wet and later converted to a dry-weight basis by analyzing separate tissue samples for moisture content. Results were expressed both on a wet- and dry-weight basis. Samples were digested with mixed warm acids for 2 hours. Following reduction with SnCl_2 , Hg^0 was purged with ultra high-purity argon gas (125 ml min^{-1} flow rate), scrubbed through a soda-lime trap, adsorbed onto a pure gold-mesh trap and thermally desorbed using a heated Nichrome coil. Determination of total mercury concentrations in samples, standards, spikes, replicates and certified reference materials used peak height on a Hewlett-Packard[®] Integrator (model 395). Detection limit, computed as twice the standard deviation of five method blanks, was $<1 \text{ ng g}^{-1}$ after correction for moisture content.

Results and discussion

Sedimentary records of mercury deposition

Unsupported ^{210}Pb activities decreased with sediment depth and approached background levels of supported ^{210}Pb in three of the five cores (Figure 2). Gamma-assay demonstrated that the lengths of both cores recovered from Acurizal were insufficient to reach background levels of ^{210}Pb . Therefore, unsupported ^{210}Pb activities had to be estimated for the lower 3 cm (Acurizal 1) and 4 cm (Acurizal 2). Sensitivity analysis revealed that halving and doubling of unsupported ^{210}Pb activities in those lower sections resulted for core Acurizal 1 in a mean age range of ± 0.5 years sediments dated to 1980, ± 1.3 years for 1960 deposits and ± 4.7 years for 70-year old deposits. The same analysis for core Acurizal 2 produced mean age ranges of ± 0.8 years, ± 2.9 years and ± 9.3 years for 1980, 1960 and 1930 deposits, respectively. We concluded that the dated profiles for both cores were robust enough to calculate average mercury accumulation rates for 20-year intervals. We opted for a coarse dating resolution separating post-1980 deposits from 1960–1980, 1940–1960, and pre-1940 sediments. Large dating uncertainty in the bottom of the cores made ^{210}Pb dates unreliable for sediments older than 80 years.

We used the Constant Rate of Supply (CRS) dating model, which assumes a constant flux of ^{210}Pb to the sediments regardless of variations in the rate of sediment accumulation. This model is commonly applied in studies that reconstruct pollution histories, particularly when sedimentation rates have varied over time. Its use in our work was supported by three conditions (cf. Appleby and Oldfield, 1983). First, the unsupported ^{210}Pb profiles were non-linear and non-monotonic indicating varying sedimentation rates (Figure 2). Second, cumulative ^{210}Pb residuals were similar within each of our two regions (Table 1) despite the differing sediment accumulation rates. Finally, the cumulative ^{210}Pb residuals in our five cores ranged from 20.5 to 29.0 pCi cm^{-2} , corresponding to ^{210}Pb fallout rates of 0.64 to 0.90 $\text{pCi cm}^{-2}\text{yr}^{-1}$. These rates were well within the range of fallout rates of 0.19 to 0.93 $\text{pCi cm}^{-2}\text{yr}^{-1}$, reported for a large number of geographically-distinct sites (Appleby and Oldfield, 1983).

^{137}Cs profiles were not as striking as those found in cores from the northern hemisphere (Figure 2). The onset of ^{137}Cs activity ($>$ our detection limit of 0.30

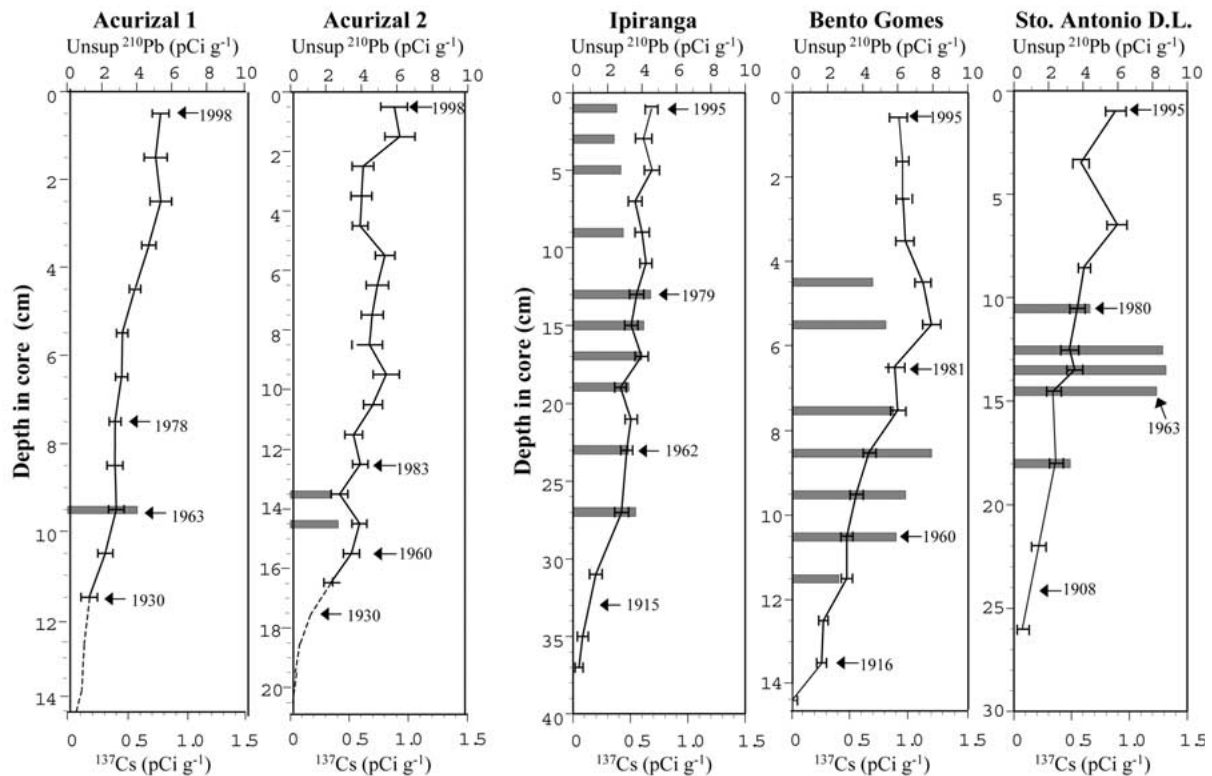


Figure 2. Unsupported ^{210}Pb (lines) and ^{137}Cs (bars) activities for two regions of the Pantanal – Acurizal (Acurizal 1 and 2) and the northern Pantanal (Ipiranga, Bento Gomes and Sto. Antonio D.L.). Selected ^{210}Pb -derived sediment ages are indicated. Counting errors are 1SD. ^{137}Cs activity was measured concurrently with ^{210}Pb but only values exceeding instrument detection (0.30 pCi g^{-1}) are given. Dotted lines in Acurizal 1 and Acurizal 2 cores represent down-core estimated values (see text for details).

pCi g^{-1}) corresponded with ^{210}Pb derived date of 1952 ± 1.8 (SD) in the northern Pantanal cores. Detectable ^{137}Cs occurred in deposits laid down since 1967 ± 5.7 SD in the Acurizal cores. Krishnaswami and Lal (1978) suggested that inter-hemispheric mixing rates may be slower than intra-hemispheric rates and demonstrated this effect with strontium. Atmospheric bomb testing, the source for ^{137}Cs , was conducted primarily in the northern hemisphere. Thus, it may be expected to find weak or delayed ^{137}Cs signals in southern hemisphere cores.

Average pre-1940 mercury accumulation in cores from Acurizal and the northern Pantanal were not significantly different (14 ± 3.3 and $18 \pm 4.2 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$, respectively, $p = 0.14$) (Figure 3). They were comparable to the rates of $12.3 \pm 5.0 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$ calculated for the 1900–1940 time period in cores from southeastern Alaska as a global reference (Engstrom and Swain, 1997). Mercury accumulation rates during 1940–1960 and 1960–1980 were also not significantly different between our two regions ($p =$

0.46 and $p = 0.17$, respectively). After the onset of gold mining in the early 1980s, however, northern Pantanal mercury deposition averaged $55 \pm 11.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$, which was more than 1.5 times higher than accumulation rates during the same time period in Acurizal sites (Figure 3). This difference between the two regions was statistically significant at a p-value of 0.072. We consider p-values less than 0.10 realistic in assigning statistical significance in coring studies where the number of samples (e.g., cores) is characteristically low. Modern mercury fluxes in the northern Pantanal were higher than those from systems in north temperate regions and comparable to rates found in subtropical South Florida (Table 2). Pre-1940 mercury accumulation in both Pantanal regions was higher than pre-industrial rates in the northern hemisphere, although flux ratios were intermediate averaging 3.2 ± 1.6 in the northern Pantanal and 2.7 ± 0.9 in Acurizal (Table 2).

Elevated accumulation rates of mercury in post-1980 deposits in the northern Pantanal (relative to

Table 1. Summary of radionuclide data for cores from two regions of the Brazilian Pantanal. Cores from the northern Pantanal are Bento Gomes, Santo Antonio D.L. and Ipiranga.

Coring site	Cumulative residual ^{210}Pb (pCi cm^{-2})	Calculated ^{210}Pb flux ($\text{pCi cm}^{-2}\text{yr}^{-1}$)	^{210}Pb derived age for ^{137}Cs onset*	^{210}Pb derived age for ^{137}Cs peak
Bento Gomes	24.1	0.75	1951	1973
Sto. Antonio D.L.	29.0	0.90	1954	1970
Ipiranga	22.9	0.71	1954	1979
Acurizal 1	20.7	0.65	1963	1963
Acurizal 2	20.5	0.64	1971	1971

* Onset defined as oldest section with ^{137}Cs activity exceeding instrument detection (0.3 pCi g^{-1}).

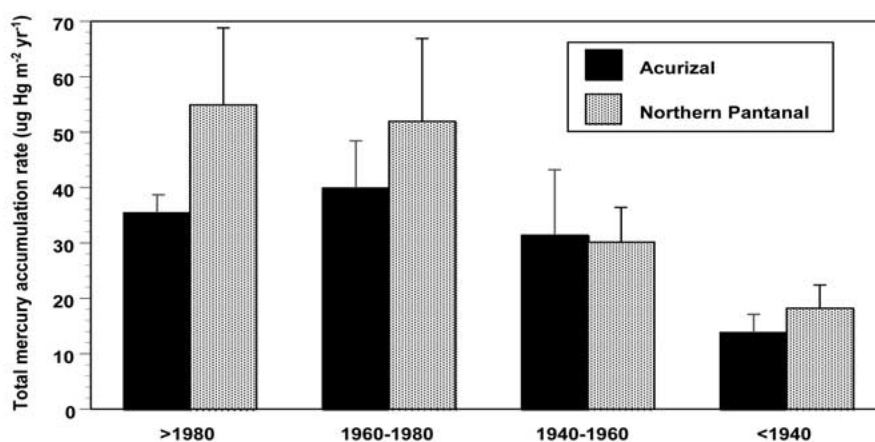


Figure 3. Average total mercury accumulation rates for selected time intervals for two regions of the Pantanal. Rates for the northern Pantanal are averaged for three cores. Acurizal rates represent average of two cores. Error bars represent +1 SD.

Table 2. Mean (and range) of modern and pre-industrial total mercury accumulation rates and ratios for selected regions arranged by descending Hg flux ratios. Ratios are computed by averaging all individual cores from each region. Modern and pre-industrial mercury accumulation rates generally apply to post-1980 and pre-1850 time periods, respectively, unless otherwise noted.

Location	Mercury flux ratio (modern/pre-industrial)	Number of core sites	Modern mercury flux ($\text{ug m}^{-2} \text{yr}^{-1}$)	Pre-industrial mercury flux ($\text{ug m}^{-2} \text{yr}^{-1}$)	Reference
South Florida	4.9 (1.6–19.1)	18	53 (23–141)	11** (8–14)	Rood et al. (1995)
Finland	4.4 (1.2–8.9)	9	26.5 (2.3–49.5)	7.8 (1.9–27.0)	Verta et al. (1989)
Minnesota-Wisconsin	3.7 (3.2–4.9)	7	23 (16–32)	6.4 (4.5–9.0)	Engstrom et al. (1994)
North Pantanal (Brazil)	3.2 (2.5–5.1)	3	54.9 (45.6–70.8)	18.2* (14.0–22.3)	This study
Sweden	3.0 (1.2–7.5)	11	12.5 (6.7–20.7)	5.8 (1.9–17.2)	Johansson (1985)
West Canada	2.7 (1.1–7.1)	9	12.1 (3.8–28.4)	5.6 (0.7–14.5)	Lockhart et al. (1995)
Acurizal (Brazil)	2.7 (2.0–3.3)	2	35.4 (33.1–37.7)	13.8* (11.5–16.2)	This study
South Ontario	2.2 (1.5–3.2)	5	22 (10–44)	10** (4.8–13.6)	Johnson et al., 1986
Southeastern Alaska	2.0 (1.8–2.1)	3	16.5 (11.5–26.0)	8.3 (5.5–12.7)	Engstrom and Swain (1997)

** Pre-1900 rates; * Pre-1940 rates.

rates for the same time interval at our reference sites) suggested a regional effect of gold mining. Furthermore, increased rates in both our Pantanal regions over global reference sites, such as southeastern Alaska, pointed to a basin-wide effect of mercury emissions. Cores from both regions in the Pantanal showed comparable mercury chronologies until the onset of mining. At that time, a significant regional signal was added to the northern cores. We estimated the magnitude of that regional signal at $19.5 \mu\text{g Hg m}^{-2} \text{ yr}^{-1}$, i.e., the difference between average post-1980 rates for both regions. This amounted to more than 70 kg yr^{-1} of mercury, if we estimated the size of the mercury impacted region at roughly $3,600 \text{ km}^2$ ($60 \times 60 \text{ km}$). Lacerda et al. (1991a,b) estimated that 15,000–20,000 kg of mercury were released into the local environment between 1980 and 1990. Using this average of $1,750 \text{ kg yr}^{-1}$ of mercury, our regional sedimentary signal only accounted for 4% of the mercury lost to the environment with the remainder lost to the atmosphere and downstream areas, or temporarily incorporated in biota. Halving ($1,800 \text{ km}^2$) and doubling ($7,200 \text{ km}^2$) of our estimate of the impacted region yielded similarly-low estimates of the regional sedimentary signal of 2 and 8% of the total released mercury. These may be realistic estimates, although our data are based on only five cores and a very rough estimate of the size of the impacted region and the amount of released mercury.

Mercury levels in surface sediments and biota

Surface sediments were defined as the top 5 cm of deposits in the sediment cores. Total mercury concentrations in these sediments averaged $45.5 \pm 5.5 \text{ ng Hg g}_{\text{dry}}^{-1}$ in the northern Pantanal (Table 3). These levels were lower than the $62\text{--}80 \text{ ng Hg g}_{\text{dry}}^{-1}$ found by Lacerda et al. (1991b) closer to gold mines near Poconé or the $71\text{--}116 \text{ ng Hg g}_{\text{dry}}^{-1}$ found at Baía Piuvai by Guimaraes et al. (1998). Surface sediments at Acurizal averaged $29.1 \pm 0.7 \text{ ng Hg g}_{\text{dry}}^{-1}$ and were significantly lower ($p < 0.05$) than those in the northern Pantanal. Although our impacted northern region was significantly higher in mercury in surface sediments than Acurizal, both our regions were in the low range of mercury concentrations documented for surface sediments in northern hemisphere wetland systems, including Lake Erie wetlands ($64.3 \text{ ng Hg g}_{\text{dry}}^{-1}$, $N = 5$; Cole 1997), the Okefenokee Swamp ($68.3 \text{ ng Hg g}_{\text{dry}}^{-1}$, $N = 3$; Leady, unpublished data)

and the Everglades ($120 \text{ ng Hg g}_{\text{dry}}^{-1}$, $N = 45$; Rood et al., 1995).

Mercury levels in biological samples from both regions in the Pantanal are given in Table 3. We report both wet and dry weight concentrations for animal tissues. Mercury levels per gram wet weight are commonly documented in the literature and our wet weights provide a measure for comparison. Mercury concentrations per dry gram, on the other hand, are a more appropriate measure when comparing trace levels in biological tissues with different moisture content along food chains.

Periphyton are major primary producers in freshwater systems and form an important trophic link between producers and herbivorous grazers. In addition, periphytic communities play a significant role in the methylation of mercury, as demonstrated by Cleckner et al. (1999) in the Florida Everglades. In other words, periphyton contribute to the conversion of inorganic mercury to the more toxic and bioaccumulated organic forms. Total mercury concentrations in periphyton were not significantly different between our two regions and levels were comparable to periphyton in contaminated sites in Tennessee ($5\text{--}50 \text{ ng Hg g}_{\text{dry}}^{-1}$) (Hill et al., 1996).

Mercury levels in *Salvinia auriculata* were significantly different between the two regions ($p = 0.006$). This floating fern contained almost 4 times more mercury in the northern Pantanal than in Acurizal (Table 3). The *Salvinia* samples probably represent the best population estimate of any of our biological samples. They were analyzed as a series of subsamples of a homogenate of a large number of whole plants from each region. Aula et al. (1995) proposed *S. auriculata* as a biological monitor for mercury pollution in their work in the Tucuruí reservoir in the Amazon basin. They found levels of $25\text{--}225 \text{ ng Hg g}_{\text{dry}}^{-1}$ in *S. auriculata* depending on season and exposure of leaves to air or water. A Russian reservoir study found the highest levels of mercury among the macrophytes in the related species *Salvinia natans* (Kipriyanova, 1997). High levels of mercury in *Salvinia* spp. may be related to the high surface area to volume ratio of this primitive plant, which maximizes exposure to mercury in the environment. Data summarized by Moore et al. (1995) on a collection of wetland plants from Canada show the same trend with highest mercury levels occurring in small plants with low vascular complexity and high surface to volume ratios.

Eichhornia crassipes can readily absorb and retain mercury (Lenka et al., 1990). Roots in the north-

Table 3. Comparison of average mercury content (ng g^{-1}) of biological tissues and sediment samples from two regions in the Pantanal. Left columns contain wet weight values; right columns contain dry weight levels after hygroscopic correction. Values in parentheses represent one standard deviation.

	Wet weight				Dry weight			
	N. Pantanal		Acurizal		N. Pantanal		Acurizal	
<i>Pygocentris nattereri</i>	67.7	(31.6)	37.0	(26.6)	302.2	(159.1)	172.3	(114.7)
<i>Serrasalmus spiroleura</i>	44.8	(24.5)	33.4	(18.0)	149.9	(84.2)	111.6	(60.0)
<i>Tetragonopterus</i> sp.	7.0	(3.1)	20.9	(3.7)	26.8	(6.4)	69.7	(12.3)
<i>Aphyocharax</i> sp.	13.4	(3.1)	14.7	(9.3)	56.0	(12.8)	61.8	(39.1)
<i>Pomacea</i> sp.	0.5	(0.3)	0.4	(0.2)	6.2	(2.1)	4.5	(2.7)
Hydrophilidae	0.7	(0.6)	1.7	(1.1)	5.5	(4.7)	9.2	(6.1)
<i>Salvinia auriculata</i>					90.7	(9.1)	24.5	(3.3)
<i>Eichhornia crassipes</i> - roots					102.0	(33.2)	64.1	(29.6)
<i>Eichhornia crassipes</i> -shoots					34.0	(9.8)	23.5	(4.5)
Periphyton					41.6	(1.7)	46.6	(5.7)
Surface sediment (top 5cm)					45.5	(5.5)	29.1	(0.7)

ern Pantanal averaged $102.0 \pm 33.2 \text{ ng Hg g}_{\text{dry}}^{-1}$ (Table 3), 1.6 times higher than Acurizal but not significantly different between both regions ($N = 8$, $p = 0.07$). Shoots in the northern Pantanal averaged $34.0 \pm 9.8 \text{ ng Hg g}_{\text{dry}}^{-1}$ (Table 3), 1.4 times higher than Acurizal but again not significantly different ($N = 8$, $p = 0.06$). Olivero and Solano (1998) found an average of $225 \pm 21 \text{ ng Hg g}_{\text{dry}}^{-1}$ in roots of *Eichhornia crassipes* from a gold mining area in Columbia. Our stem and leaf mercury levels fell in the low end of the range of 30–140 $\text{ng Hg g}_{\text{dry}}^{-1}$ reported for the emergent macrophyte *Pontederia lanceolata* in the northern Pantanal (Lacerda et al., 1991b). Shoots and leaves of aquatic vegetation in uncontaminated areas in north temperate zones contained less than $100 \text{ ng Hg g}_{\text{dry}}^{-1}$ and less usually less than $10 \text{ ng Hg g}_{\text{dry}}^{-1}$ (Cocking et al., 1991; Siegel et al., 1985). Mercury levels in water hyacinth leaves from our sites in the Pantanal fell within this range of contamination.

At both sites, mercury concentrations in *Eichhornia* roots were 2.7–3.0 times those in shoots. This matched our previous findings in the Okefenokee Swamp, where we found a trend for mercury to increase in concentration from leaves to stem to roots. For example, *Xyris smalliana* had root concentrations of $91.8 \text{ ng Hg g}_{\text{dry}}^{-1}$ and shoot concentrations of $26.0 \text{ ng Hg g}_{\text{dry}}^{-1}$ (unpublished data). Kipriyanova (1997) found a similar trend in a Russian reservoir with concentrations of almost all trace metals, including mercury, higher in roots and root stock than in macrophyte tissue above the sediment surface. Aula

et al. (1995) also found submerged roots and leaves of *Salvinia auriculata* to be significantly higher in mercury than floating leaves. Factors that may contribute to such concentration differences among plant tissues include the increased levels of mercury in sediments (relative to water and air) and the possibility that aerial plant parts may rid themselves of mercury as vapor by reducing ionic Hg to elemental Hg (Siegel et al., 1987). In addition, plant roots are generally older than stems and leaves and have had more time to accumulate mercury.

Aquatic insects represent a major component of the diet of many fish species. As such, they constitute an important pathway for the channeling of mercury along a food chain (Hall et al., 1997). Adult water scavenger beetles (Order Coleoptera, Family Hydrophilidae) were collected on macrophytes from both study regions. As adults, these beetles are scavengers on dead vegetation. Their mercury concentrations were among the lowest in this study and were not significantly different between our regions (Table 3). Mercury levels in apple snails (*Pomacea bridgesi*, formerly *Ampullarius australis*) were also low in our samples ($N = 10$, $0.4\text{--}0.5 \text{ ng Hg g}_{\text{wet}}^{-1}$) and not significantly different between regions (Table 3). *Pomacea* spp. are primary consumers feeding on periphyton and aquatic macrophytes. The snails in our collection averaged $11.5 \pm 1.7 \text{ mm}$ in diameter and were not different in size between our two regions ($p = 0.46$). Lacerda et al. (1991a) reported mercury levels in *Pomacea bridgesi* below their detection limit (e.g., 40 ng

Hg $\text{g}_{\text{wet}}^{-1}$) at a site approximately 15 km downstream from a gold mine in Poconé. Their snails ranged in size and were larger than our individuals making a direct comparison between the two collections problematic. In fact, they noted a significant positive relationship between shell size and mercury content in their snail collection (Lacerda et al., 1991a). Eisenmann et al. (1997) found an average of 63 ng Hg $\text{g}_{\text{wet}}^{-1}$ in south Florida apple snails (*Pomacea paludosa*) with a mean shell diameter of 36 mm. Again, these larger and likely older snails may not be directly comparable to our smaller individuals. Both the beetles and the snails consume predominantly plant matter and it is interesting to note that mercury levels in these primary consumers in our samples are considerably lower than in their potential food source (e.g., periphyton and macrophyte tissue). The channeling of mercury from lower to higher trophic levels along herbivorous links in food chains may be relatively inefficient compared to transfer along carnivorous links, a phenomenon previously noted in Lake Erie wetlands (Cole, 1997) and in the Florida Everglades (Roelke et al., 1991). Both the high water content of plant material consumed by herbivores, which in effect dilutes the mercury intake and the low percentage of organic mercury in plant material (Moore et al., 1995) may play a role in this inefficient transfer.

In both our regions, we made collections of four different species from the Characidae, a large family of about 300 species found mostly in South America. *Aphyocharax* sp. is a small planktivorous characid fish with a standard length of 2.5 ± 0.1 cm in our samples. This length was not different between our two sampling regions ($N = 12$, $p = 0.33$). Its mercury content was similar between the northern Pantanal and Acurizal ($N = 12$, $p = 0.37$). *Tetragonopterus* sp. consumes a variety of small aquatic invertebrates, as well as small fish. Contrary to our prediction, mercury levels in *Tetragonopterus* were significantly higher ($p = 0.00003$) in our reference region than in the northern Pantanal. Fish length was similar between both regions (standard length 7.8 ± 0.4 cm, $N = 12$, $p = 0.14$) and we have no basis to believe that we collected a different species of *Tetragonopterus* from each region.

Serrasalmus spiroleura, a predatory piranha, was also sampled at both regions. Although this piranha has a reputation of fin-biting, its diet is more varied and feeding strategy more opportunistic than our other large predator *Pygocentris nattereri* (Sazima and Machado, 1990). Mercury content of 12 filets

(Table 3) and standard fish lengths (10.6 ± 2.1 cm) were not significantly different between regions ($p = 0.19$, $p = 0.43$, respectively). *P. nattereri* was the largest predatory piranha sampled (standard length 13.7 ± 2.4 cm). The mercury content of filets was significantly higher in the northern Pantanal region ($N = 16$, $p = 0.04$). The standard length of our *P. nattereri* collections was not significantly different between regions ($N = 16$, $p = 0.37$). Our results compare well with Lacerda et al. (1991a) who reported 60 ± 10 ng Hg $\text{g}_{\text{wet}}^{-1}$ in this species in the Bento Gomes river some 20 km downstream from gold mines in Poconé. Lacerda et al. (1994) found 300 ± 360 ng Hg $\text{g}_{\text{wet}}^{-1}$ in this species in the Carajas mining region in the southeastern Amazon, i.e., 4.4 times the amount in our *Pygocentris* from the northern Pantanal.

We correlated standard fish length with mercury levels for *P. nattereri* from the two regions (Figure 4). Both correlations were significant (longer fish contain more mercury), but the slope of the regression line for the northern Pantanal was 2.6 times the slope for the Acurizal collection. This difference in slope was statistically significant ($p = 0.05$). It indicated that, as these piranhas get larger, *P. nattereri* gained 2.6 times more mercury per cm of growth near the mining region than in Acurizal. This implied an elevated rate of bio-magnification in the northern Pantanal compared with our reference region. Interestingly, we found nearly the same ratio for *S. spiroleura*, although the correlation between fish length and mercury content was not significant at 0.05 level for our Acurizal collection. Unfortunately, these piranhas were very difficult to age and an alternative explanation for these results may be that *P. nattereri* in the northern Pantanal grew more slowly than near Acurizal. Although the age/size relationship is complicated, in general larger fish of the same species are older and have had a longer time to accumulate mercury.

In summary, we documented for the first time radionuclide-dated mercury accumulation profiles from a mercury-impacted and a reference region in the Pantanal. We uncovered a regional effect of mercury released from gold mining in cores from the northern Pantanal. We also showed a basin-wide effect of elevated mercury accumulation in all our cores relative to global reference sites. In addition, we documented mercury levels in a variety of organisms linked in *in situ* food webs and we focused on comparing the same taxa, number of individuals and size classes between both regions. Plant tissues contained more mercury in the impacted northern Pantanal than in our refer-

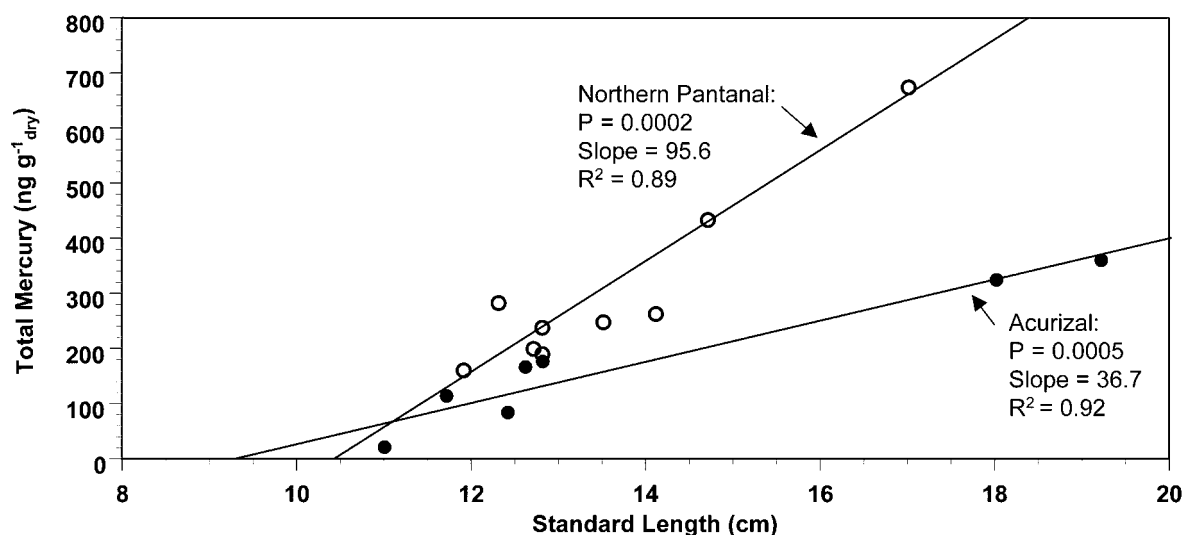


Figure 4. Regressions of standard length (cm) and mercury content ($\text{ng g}^{-1} \text{dry}$) of filets of *Pygocentris nattereri* from two regions of the Pantanal: Acurizal (closed circles) and the northern Pantanal (open circles).

ence region. Herbivores contained little mercury in both regions, while larger carnivorous piranha species showed elevated mercury body burdens in the northern Pantanal. This confirmed previous work which showed that the channeling of mercury from lower to higher trophic levels along herbivorous links is inefficient compared to transfer along carnivorous links. Our data also suggested that the rate of mercury magnification in upper trophic-level carnivorous fish was significantly higher in the northern Pantanal than in the reference region. Magnification of mercury along food chains and its chronic and toxic impact on biota may threaten biodiversity and human health. Understanding mercury dynamics in the Pantanal, with its remarkable biodiversity and with a human population that traditionally relies on a high per capita fish consumption, is therefore particularly important. Our work intended to contribute to this understanding.

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