

MERCURY CONTENTS IN AQUATIC MACROPHYTES FROM TWO RESERVOIRS IN THE PARAÍBA DO SUL: GUANDÚ RIVER SYSTEM, SE BRAZIL

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ABSTRACT

This paper reports on a study to determine the Hg content in the five most abundant aquatic macrophyte species (*Elodea densa*, *Sagittaria montevidensis*, *Salvinia auriculata*, *Pistia stratiotes* and *Eichhornia crassipes*) in two artificial reservoirs flooded by water diverted from the Paraíba do Sul river, SE Brazil. The potential of these species for Hg accumulation and their role in Hg transport along the river system due to macrophyte management were evaluated. Mercury concentrations were higher in free-floating than in rooted species. Roots were also richer in Hg than were leaves. Dry weight Hg concentrations in leaves and roots from all species varied from 46-246 ng.g⁻¹ to 37-314 ng.g⁻¹, respectively. These values are higher than those reported for uncontaminated lakes in Brazil and in other tropical areas and similar to those reported for moderately contaminated sites. Mercury concentrations can be attributed to fluvial transport from the heavily industrialized Paraíba do Sul river basin. Intensive sampling of *Pistia stratiotes* from two sites in the Vigário reservoir was performed to evaluate the capacity of Hg incorporation in short periods of time. The results showed a significant negative correlation between Hg content and size class of individual plants, demonstrating the importance of juveniles, fast growing plants in absorbing Hg. The foremost impact related to Hg contents in the studied area concerns the periodic removal of macrophytes for reservoir management, followed by disposal in nearby areas. This results in the mobilization of 0.52 to 1.3 Kg of Hg per year, a significant fraction of the Hg burden present in reservoir waters. Disposal of such material may result in Hg leaching to river systems, affecting the Hg transfer throughout the basin.

Keywords: mercury, aquatic macrophytes, artificial reservoirs.

RESUMO

Concentração de mercúrio em macrófitas aquáticas em duas represas no sistema Paraíba do Sul-Rio Guandu, SE do Brasil

O presente estudo apresenta as concentrações de Hg em cinco espécies abundantes de macrófitas aquáticas (*Elodea densa*, *Sagittaria montevidensis*, *Salvinia auriculata*, *Pistia stratiotes* e *Eichhornia crassipes*) coletadas em duas represas que recebem águas da transposição do rio Paraíba do Sul, SE do Brasil. A acumulação de Hg nessas espécies e seu papel no transporte de Hg ao longo do sistema fluvial devido ao manejo das macrófitas são discutidos. As concentrações de Hg foram maiores nas macrófitas flutuantes que nas enraizadas. Em geral, as raízes apresentaram maiores concentrações de Hg que as folhas para todas as espécies. As concentrações de Hg variaram de acordo com as espécies entre 46-246 ng.g⁻¹ e 37-314 ng.g⁻¹,

respectivamente. Estas concentrações são maiores que aquelas relatadas para macrófitas coletadas em lagos não contaminados no Brasil e em outras regiões tropicais, e similares àquelas relatadas para áreas moderadamente contaminadas. As concentrações de Hg podem ser atribuídas ao transporte fluvial a partir da região industrializada do vale do rio Paraíba do Sul. Uma amostragem intensiva de *Pistia stratiotes* na represa do Vigário foi realizada para avaliar a capacidade de incorporação de Hg por esta macrófita. Os resultados mostraram uma correlação negativa, significativa, entre as concentrações de Hg e o tamanho dos indivíduos, demonstrando a importância de juvenis desta espécie na absorção de Hg. A retirada periódica de macrófitas da represa, seguida por sua disposição em áreas adjacentes, pode afetar a dinâmica do Hg. Os resultados mostram uma mobilização de 0,52 a 1,3 kg Hg por ano, uma fração significativa da carga total de Hg presente nas águas da represa. A disposição inadequada deste material poderá resultar em um aumento da mobilização de Hg na bacia.

Palavras-chave: mercúrio, macrófitas aquáticas, represas.

INTRODUCTION

Aquatic macrophytes accumulate Hg with high concentration factors relative to the Hg concentrations present in water, thus affecting the distribution of this metal among the different compartments of aquatic systems (Lacerda *et al.*, 1991; Dushenko *et al.*, 1995; Villar *et al.*, 1999). Also, macrophyte rhizospheres are important sites of Hg methylation in tropical freshwater ecosystems (Guimarães *et al.*, 1998; 2002), acting as a potential Hg source for herbivorous organisms and enhancing Hg transfer along food chains (Suarez *et al.*, 2001).

The extensive growth and large biomass of aquatic macrophytes observed in tropical reservoirs may influence the Hg balance and distribution among reservoir compartments and alter the system's ability to retain pollutants. Therefore, reservoir management strategies such as the periodic removal of macrophytes and their disposal in nearby areas must take into consideration their effects on Hg mobilization.

The Paraíba do Sul river (PSR) is a natural link between Rio de Janeiro and São Paulo states, SE Brazil, receiving effluents from the largest industrialized region of the country. In the late 1950s, water from the PSR was diverted to form artificial reservoirs to supply water to the metropolitan area of Rio de Janeiro city, resulting in an artificial waterway – the PSR-Guandú river system (Fig. 1). After treatment, part of this water is discharged back into an artificial canal that flows into Sepetiba Bay, about 100 Km from the PSR, an important fishery and tourism pole of southern Rio de Janeiro state. This has resulted in an increase in the freshwater flux to the bay from the original $20 \text{ m}^3 \cdot \text{s}^{-1}$ to $160 \text{ m}^3 \cdot \text{s}^{-1}$ (Molisani *et al.*, 2002). Marins *et al.* (1999) suggested that the PSR may also be an important source of pollutants flowing into Sepetiba Bay. A preliminary mass balance calculation showed that the Hg load brought into Sepetiba Bay by the PSR-Guandú river system represents about 30% of the total Hg dis-

charged into Sepetiba Bay (Marins *et al.*, 1999). Further studies on the Hg distribution along the entire length of the waterway systems confirmed this calculation (Molisani *et al.*, 2002). Two reservoirs, Santana and Vigário, control the system's water discharge and retain most of the suspended matter, and possibly associated pollutants, from the contaminated PSR. Both present extensive macrophyte growth due to partially eutrophicated waters. Therefore, macrophytes may play a significant role in the Hg dynamics through the PSR-Guandú system waterway.

In order to investigate the importance of aquatic macrophytes in the Hg dynamics of the PSR-Guandú system, we studied the distribution of Hg in the most abundant macrophyte species in the two reservoirs that control the water flux through the system.

MATERIAL AND METHODS

Study site

The two most important reservoirs in the Paraíba do Sul-Guandú river system were sampled. Santana reservoir has a watershed of 433 Km² and covers an area of 5.95 Km² with a mean depth of 3.3 meters. Vigário reservoir has a watershed of 30 Km² and an area of 3.8 Km² with a mean depth of 13.5 m. The widespread distribution of aquatic macrophytes is evident in the two reservoirs. Their spatial distribution is determined by environmental conditions, particularly morphometric variables such as reservoir width, depth and width/wetted perimeter ratio, current velocities and water chemistry (Gantes & Caro, 2001; Heegaard *et al.*, 2001). In the study area, *Elodea densa* Planchon and *Sagittaria montevidensis* Bogin are present only in Santana reservoir, which is less deep, with many sediment banks and is a suitable habitat for rooted macrophytes. The greater depth of Vigário reservoir and its steep banks provide habitat only for the free-floating

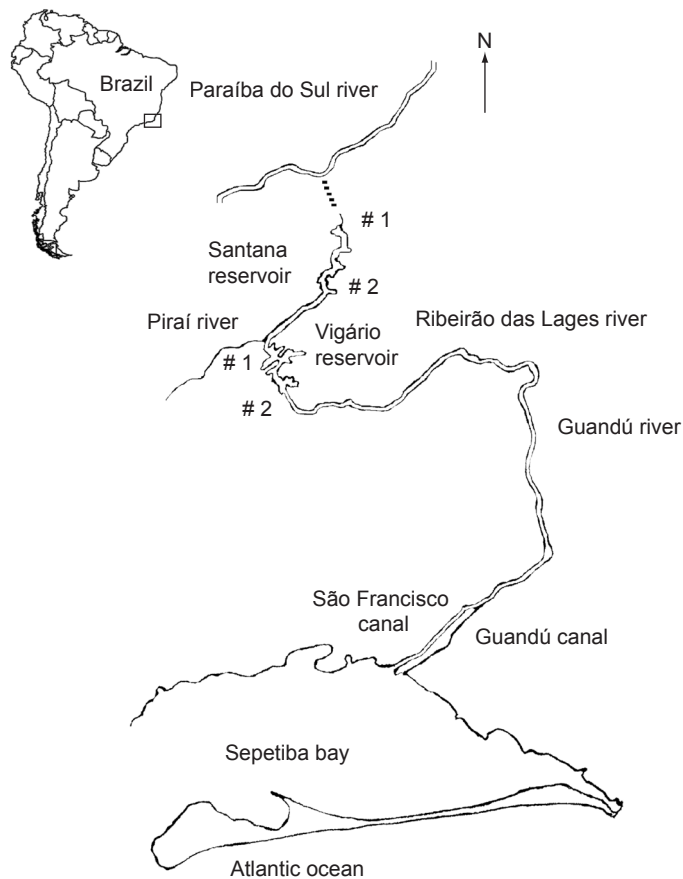


Fig. 1 — Scheme of the water diversion system of the Paraíba do Sul river to Sepetiba bay and sampling sites (scale 1: 400,000) (Molisani *et al.*, 2002).

species *Salvinia auriculata* Aublet, *Pistia stratiotes* Linnaeus and *Eichhornia crassipes* Solms-Laubach. Rooted macrophytes occur as belts along shallow shores, whereas floating macrophytes occur both as small mats floating on the reservoir surface and as continuous cross-sectional covers. In both reservoirs, macrophytes are artificially managed and periodically removed. Removed plants, however, are frequently dumped locally.

Sampling and chemical analyses

Two samplings were carried out in Santana reservoir and three in Vigário reservoir between March 2001 and February 2002. Samples of *E. densa* ($n = 8$) were collected in Santana reservoir at a depth of about 1 m at two sites (sites 1 and 2), while for *S. montevidensis* about 5 plants were sampled at one site (site 2) of the reservoir. In Vigário reservoir, 5 composite samples of *S. auriculata*, *P. stratiotes* and *E. crassipes* were collected from two sampling sites, in the middle (site 1) and lower (site 2) reservoir (Fig. 1). Each composite sample consisted of 10 to 20 individual plants. The plant samples were washed in

lake water to remove particles of sediment, put into plastic bags and taken to the laboratory. In February 2002, an intensive sampling of *P. stratiotes* was performed at the Vigário reservoir. A large number of plants ranging in size from juveniles to full adult individuals were sampled in order to estimate the importance of the artificial management of this species in removing Hg from the reservoir.

In the laboratory, the samples were washed and roots and leaves were separated and dried at 50 °C for 24 h. The Hg content was determined in duplicate after digestion of 1 g sub-samples in 20 mL of concentrated HNO_3 for 1 h at 70 °C in a “cold-finger” system. After cooling, 20 mL of HCl 50% was added and the mixture heated at 70 °C for one hour. The Hg concentration was determined by cold vapor atomic absorption spectrophotometry. The accuracy of Hg determination in plant tissues was assessed using standard reference material SRM 1515 (Apple Leaves), reaching acceptable (< 15% difference) results ($53 \pm 3 \text{ ng}\cdot\text{g}^{-1}$ and $44 \pm 4 \text{ ng}\cdot\text{g}^{-1}$; $n = 3$) for the measured and certified values, respectively.

Water and sediment samples were collected from

two sites in each reservoir in March 2001. All the sampling and analytical equipment was pre-cleaned according to acceptable protocols (Guentzel *et al.*, 1996). The water samples were stored in 0.5 L pre-cleaned PET bottles (Cope-land *et al.*, 1996). The sampling bottles were rinsed three times with local water before being filled, double bagged in acid-washed plastic bags and transported in an icebox to the laboratory. Bottom sediments were taken manually with a Teflon shovel and stored in plastic bags.

Total dissolved Hg concentration in reservoir water was determined in unfiltered, non-acidified, sub-samples (50 mL) after oxidation with 0.2 mL of a bromine mono-chloride solution (0.1 mL KBrO_3 1% m/v + 0.1 mL HCl 20% v/v) at room temperature (Lacerda & Gonçalves, 2001) through cold vapor atomic fluorescence in a Tekran Model 2500 AFS. The detection limit for the method is $0.2 \text{ ng}\cdot\text{L}^{-1}$.

The Hg concentrations in suspended particles (separated through filtering with Millipore $0.45 \mu\text{m}$ cellulose acetate filters) and bottom sediments were determined after extraction with 20 mL 3:HCl and 1: HNO_3 50% v/v for 1 h in 60°C using a "cold-finger system" (Lacerda & Gonçalves, 2001). The Hg in sediments was determined by cold vapor atomic absorption spectrophotometry following reduction with SnCl_2 , in a Bacharatt Model Mercury Analyzer. The accuracy of the Hg determination in sediment samples was assessed by simultaneous analysis of reference material (NIST 2704 Buffalo River), reaching $58 \pm 3 \text{ ng}\cdot\text{g}^{-1}$ ($n = 5$) compared with a certified value of $60 \text{ ng}\cdot\text{g}^{-1}$.

RESULTS AND DISCUSSION

Table 1 shows the Hg concentrations in leaves and roots of studied species sampled in Santana and Vigário reservoirs. Within the same plant species and particularly in free-floating macrophytes, the Hg concentration was generally higher in root than in leaf tissues, with the excep-

tion of *E. crassipes* from Vigário reservoir, which showed similar Hg concentrations in leaves and roots, and *Sagitaria montevidense* from Santana reservoir, which showed higher concentrations in leaves. The results agree, in general, with the reported pattern of Hg distribution in aquatic macrophytes (*e.g.* Aula *et al.*, 1994; 1995; Gupta & Chandra, 1998; Coquery & Welbourn, 1994), but the small number of samples and the relatively large variability within species hinder a better statistical analysis of these data.

The free-floating species, particularly the fast growing species *P. stratiotes* and *S. auriculata* from Vigário reservoir, displayed the highest Hg contents of all the species sampled, while the rooted emergent *S. montevidenses* from Santana reservoir presented the lowest Hg concentrations. Metal accumulation in aquatic macrophytes is known to vary among species; for example, emergent aquatic macrophytes usually accumulate lower amounts of metals than submerged or floating species (Albers & Camardese, 1993). Metal uptake by these plants is dependent on exposure pathways. While water is the main metal source for floating macrophytes radio-tracer studies have demonstrated that the underlying sediments is the principal pathway for metal uptake by rooted macrophytes (Jackson, 1998). Moreover, the proportion of Hg readily available for uptake by rooted aquatic macrophytes depends on sediment geochemistry rather than on total Hg concentration in sediments. Also, rooted macrophytes develop oxidized rhizospheres capable of immobilizing metals at the external surfaces of roots, acting as a barrier to metal uptake (Coquery & Welbourn, 1995; Campbell *et al.*, 1985).

The Hg content in the free-floating macrophytes *E. crassipes*, *P. stratiotes* and *S. auriculata* is related to the Hg concentrations in water, while the Hg content in the rooted emergent *S. montevidense* and the rooted submerged *E. densa* species is related to the concentrations and availability in bottom sediments. In addition to water and bottom sediments, suspended sediments may be

TABLE 1
Mean, standard deviation and range of Hg concentrations ($\text{ng}\cdot\text{g}^{-1}$) in aquatic macrophyte species from Santana and Vigário reservoirs, Rio de Janeiro State, SE Brazil.

Species	Type	Reservoir	Leaf	Root
<i>Elodea densa</i> (n = 4)	Submerged	Santana	82 ± 57 46 - 167	177 ± 119 106 - 314
<i>Sagitaria montevidense</i> (n = 4)	Emergent	Santana	103 ± 4 100 - 106	62 ± 35 37 - 87
<i>Pistia stratiotes</i> (n = 5)	Free-floating	Vigário	154 ± 9 146 - 167	215 ± 60 156 - 311
<i>Salvinia auriculata</i> (n = 5)	Free-floating	Vigário	139 ± 74 85 - 246	191 ± 75 88 - 265
<i>Eichhronia crassipes</i> (n = 5)	Free-floating	Vigário	125 ± 40 78 - 167	119 ± 75 101 - 136

considered a source of Hg for floating plants, since their roots act as traps for the suspended sediment flow and the particles adhering to roots are important sources of readily available Hg for plant uptake (Guimarães *et al.*, 1998). Moreover, the general trend of lower Hg contents in the rooted *S. montividenses* and *E. densa* species than in the free-floating *P. stratiotes*, *E. crassipes* and *S. auriculata* may be influenced by the lower Hg concentrations in both water and sediments in Santana reservoir than in Vigário reservoir (Table 2).

A review published by Outridge & Noller (1991) reported Hg concentration in freshwater plants from uncontaminated and contaminated environments to range from 20 to 19,000 ng.g⁻¹. These data demonstrate the considerable variations in Hg concentrations in aquatic macrophytes that settle in a wide diversity of aquatic environments. This wide range of concentrations is attributed to variables such as Hg partitioning in water and sediment, water physicochemistry, and plant physiology and genotypic features (Lacerda *et al.*, 1991; Outridge & Noller, 1991; Jackson, 1998; Maury-Brachet *et al.*, 1990). Therefore, meaningful comparisons with literature data should at least be restricted to the tropics, notwithstanding the paucity of data from that region. Compared to other reported Hg concentrations in tropical aquatic macrophytes, the concentrations found in leaves and roots of *E. crassipes* and *S. auriculata* in the reservoirs of the RPS-Guandú system are 2 to 3-fold higher than those reported for the same species in large artificial reservoirs in the Brazilian Amazon (Aula *et al.*, 1994) and from unpolluted Cuban waters (Gonzalez *et al.*, 1994), which varied from 30 to 75 ng.g⁻¹ and 60 to 120 ng.g⁻¹ for leaves and roots, respectively. The concentrations found in the other species (*P. stratiotes*, *S. montevidensis* and *E. densa*) are also higher than the average concentrations found in macrophytes from uncontaminated water bodies in Africa (Biney *et al.*, 1994) and other parts of the tropics. In contrast, data from lowly to moderately contaminated sites in African aquatic environments showed Hg values in *P. stratiotes* and *E. crassipes* similar to those found in our study area (Biney *et al.*, 1994). In several marginal lakes of the Madeira River in the western Amazon, which are affected by effluents from small-scale gold mining, Hg concentrations found in *E. crassipes* reached 1,000 ng.g⁻¹

(Martinelli *et al.*, 1988), *i.e.*, well above the maximum concentrations found in our site. Though based on scanty data, it seems clear that the macrophytes from the PSR-Guandú systems contain abnormal Hg concentrations, suggesting a major role in the transport of Hg through the system.

A sample of *S. auriculata* collected in a small lake located in the same drainage basin but outside the influence of the PSR-Guandú system showed a lower Hg concentration than those measured in the plants collected in Vigário reservoir (63 + 38 ng.g⁻¹ and 120 + 47 ng.g⁻¹ for leaves and roots, respectively). As a small artificial lake with no fluvial input, regional atmospheric deposition is most probably the only Hg source. Therefore, fluvial transport from the contaminated Paraíba do Sul river is responsible for the higher Hg concentrations found in macrophytes from the RPS-Guandú system's reservoirs.

The capacity of aquatic macrophytes to accumulate Hg can enable these plants to play at least three important roles in aquatic environments: as biomonitors of environmental Hg concentrations (Jackson *et al.*, 1991); as conveyors of Hg to food chains Simon & Boudou (2001); and as important sites for Hg methylation (Guimarães *et al.*, 1998). Regarding the capacity for responding to environmental concentrations, the relationship between metal concentrations in the environment (water and substrate) and in the macrophytes showed that, at the very least, *E. densa* and *P. stratiotes* can be considered good monitors of Hg contents, the former in bottom sediments and the latter in the water column of the reservoirs studied here.

In the study area, however, another important role of aquatic macrophytes concerns their periodic removal from reservoirs for their management and their disposal in surrounding areas, which may be enriched by Hg due to decomposition of plant material; this may significantly affect Hg transport through the RPS-Guandú system. Intensive sampling of *P. stratiotes* at two sites in Vigário reservoir was done in order to estimate the capacity of Hg incorporation in short periods of time by juveniles of this species, since macrophyte removal may occur as frequently as at monthly intervals. Fig. 2 presents the concentration of Hg in plants distributed according to size classes, which included juveniles and mature individuals. The results revealed a significant negative correlation

TABLE 2
Mean Hg concentration in water and sediment samples from the Santana and Vigário Reservoirs (n = 4).

Reservoir	Water ¹ (ng.L ⁻¹)	Suspended particles (ng.g ⁻¹)	Bottom sediments (ng.g ⁻¹)
Santana	0.05 - 0.10	320 - 550	15 - 22
	0.08 ± 0.03	390 ± 100	19 ± 3
Vigário	0.05 - 0.18	600 - 1,046	70 - 90
	0.1 ± 0.07	1,130 ± 430	80 ± 14

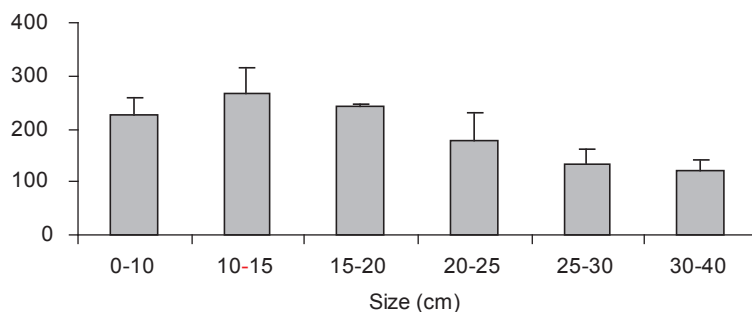


Fig. 2 — Mean Hg concentration (ng.g⁻¹) in different size classes of *Pistia stratiotes* collected from two sampling sites in the Vigário reservoir (n = 6 for each size class) (Molisani *et al.*, 2002).

($r = -0.88$; $p < 0.05$) between Hg concentrations and size. The higher Hg concentration in smaller individuals relative to adult plants demonstrates the importance of juveniles in absorbing Hg from the water, suggesting the biomonitoring value of juvenile individuals in monitoring studies of Hg contamination of these environments. On the other hand, the lower Hg content in adult plants suggests a dilution effect on Hg content as the plant's biomass increases. This finding confirms the potential importance of macrophyte removal in affecting the Hg dynamics in the system, since the plants may accumulate high Hg concentrations even within short periods.

To estimate the amount of Hg involved in the process, the amount of plants removed from the reservoir was monitored and analyzed between May 1997 and January 1998. During this period about 93,203 m³ of macrophytes were removed from both reservoirs and disposed in nearby areas.

To evaluate the potential Hg transfer from aquatic plants to disposal areas, an estimate was made based on the biomass removed in dry weight, representing about 20% of total biomass removed, an estimate of plant organic matter density (0.2-0.5 g.cm⁻³) and the mean Hg concentration in the leaves and roots of all aquatic macrophytes in the two reservoirs (0.14 µg.g⁻¹ dr. wt.). According to the above data, between 5 and 13 Kg of Hg per year is transferred to disposal areas through aquatic macrophytes removed from the reservoirs. Compared with the total amount of Hg transported annually by the systems (Molisani *et al.*, 2002), this may represent up to 10% of the total Hg transported. Once in soils, the Hg might be immobilized in soil organic matter, reach the pore water and return to the river systems or be reemitted into the environment through volatilization (Stein *et al.*, 1996; Lindberg & Meyers, 2001). Therefore, the potential importance of reservoir management strategies in the Hg dynamics of the RPS-Guandú system calls for more detailed studies on this subject.

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