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Contribution of a medium-sized tropical river to the particulate heavy-metal load for the South Atlantic Ocean

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Abstract

The Paraíba do Sul River is a medium-sized river, 1145 km in length with a drainage basin of 55 400 km^2. The riverine fluxes of particulate metals (Cu, Cr, Zn, Mn and Fe) were investigated over 24 months. Particulate matter samples were monthly collected from April 1994 to March 1996. The first 12-month period presented lower rainfall than the second, although both periods presented average precipitation lower than the regional average. The particulate matter flux in the second period (2 042 080 t) was 250% higher than the first period (821 489 t). The same trend was observed for the associated metals, which presented higher fluxes in the second period. This study highlights the strong dependence of the transported mass on the rainfall, and consequently with the river water discharge. The Paraíba do Sul River presents a low contribution to the world oceans, although the local contribution could be considered relevant.

Keywords: Particulate matter; Metals; Tropical river; Annual load; South Atlantic Ocean

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1. Introduction

According to several authors, rivers are the main pathways for the transport of continental material to the world oceans. Riverine suspended particulate matter is represented by a wide combination of inorganic material (i.e. clay minerals and Fe and Mn oxyhydroxides) and organic matter (detritic or alive). These particles, due to their high surface area, and also to the carrier nature of oxides, are the main heavy metal carriers in fluvial systems (Jenne, 1968; Waren and Zimmerman, 1993).

Small and medium-sized rivers, although quite significant in terms of mass transfer, generally have not been considered in global mass transfer studies (Holeman, 1968; Meybeck, 1976; Martin and Meybeck, 1979; Milliman and Meade, 1983). The reliability of the global budget is further doubted, due to the lack of a proper database for the Third World rivers. A detailed monitoring of rivers on a local and regional scale will surely improve knowledge in terms of the global mass transfer (Jah et al., 1988).

In South America, very few studies have estimated the particulate matter loads and associated metals transported to the ocean. Most of them were performed in the northern region and studied the Amazon River basin input to the North Atlantic (Gibbs, 1967; Rodrigues et al., 1979; Boyle et al., 1982). Studies that deal with the riverine input to the South Atlantic, are very scarce, with only three rivers investigated: the São Francisco River (Bessa and Paredes, 1990) in the north-eastern Brazil; and the De la Plata (Urien, 1972) and Negro Rivers (Depetris, 1980) in the south of the continent.

In the present study, an attempt has been made to estimate the particulate matter and associated metals input from the Paraíba do Sul River, south-eastern Brazil, to the South Atlantic Ocean.

2. Sampling area

The Paraíba do Sul River (PSR) is a medium-sized river, 1145 km long, with a drainage basin of 55 400 km^2. It crosses three of the most important and developed states of Brazil (Minas Gerais, São Paulo and Rio de Janeiro). It is the main water supply for more than 11 000 000 people at Rio de Janeiro City, but it is also used as waste disposal for a very large number of industries along its course.

Previous work performed in the middle reaches of the PSR, as well as in some of its tributaries (Paraibuna, Pomba and Muriaé Rivers, Fig. 1), indicated that these rivers are contaminated by heavy metals. The main sources of these pollutants are industrial activity, urban wastes, particularly as major sources of Zn, Pb, Cu and Cr (Malm, 1986; Azcue, 1987; Torres, 1992), and gold-mining activities, a significant source of Hg to the PSR lower basin up to the late 1980s (Lacerda et al., 1993; Souza, 1994).

Although the above-mentioned authors showed that the middle PSR was contaminated by heavy metals, according to Carvalho et al. (1993), heavy metal concentrations found in continental shelf sediments in front of the river mouth showed little or no anthropogenic influence, and could be considered without metal contamination, with the exception of Hg (Lacerda, et al., 1993).

Carvalho et al. (1999), Molisani et al. (1999) and Salomão et al. (2001) have already described the sediment and particulate heavy-metal distribution, dynamic and mass balance along the lower drainage basin of the Paraíba do Sul River. These previous works concluded that this part of the system presented low or no heavy metal contamination. The observed absence of metal contamination is probably due to the dilution caused by the non-contaminated particulate matter that enters the river in the lower drainage basin (Carvalho et al., 1999).

3. Material and methods

Samples were monthly collected for 24 months, from April 1994 to March 1996. All water samples were collected with a polyethylene 'Van Dorn'-type sampler and stored in 10-l polyethylene flasks, which were kept in ice during transport to the laboratory. The samples were filtered in triplicate (Millipore cellulose acetate, 0.45 μm pore

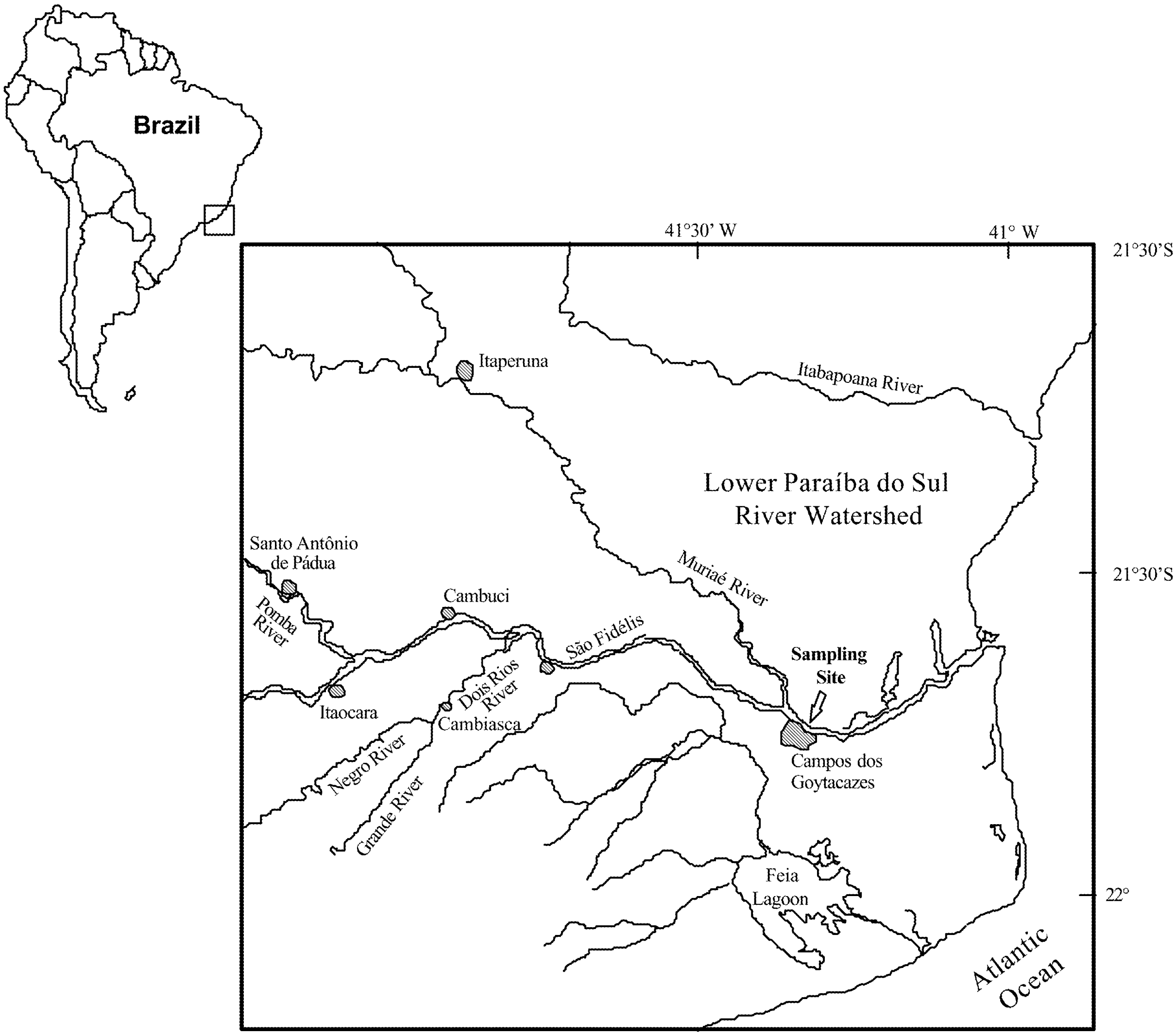

Fig. 1. Sampling site location.

diameter), and oven-dried (80°C for 48 h) for subsequent acid digestion and metal analysis. The filter digestion methodology followed a modified version of the procedure used by Watts and Smith (1994). The filters were placed in Teflon bombs with concentrated acids ($HCl + HNO_3 + HF$) in an oven (100°C for 24 h); after the digestion of the sample, the acid was evaporated to almost dryness, and redissolved in 0.5 M HCl (25 ml). The extracts were then analyzed by conventional flame atomic absorption spectrophotometry.

The coefficient of variation for triplicates of the same sample was generally lower than 20%; in cases of higher values, another replicate was analyzed in order to check the values obtained. Two standard reference materials were analyzed in order to estimate the accuracy of the method used (riverine sediment Standard Material 2704 and estuarine sediment Standard Material 1646a), both supplied by the National Institute of Standards and Technology (USA). The results of the heavy metal recovery from these standard materials are shown in Table 1.

River water-flow measurements were also per-

Table 1
Recovery of strongly bound metals from two standard materials

Standard material[a]	Recovery (%)				
	Cu	Cr	Zn	Mn	Fe
River sediment (NIST 2704)	103	90	91	87	86
Estuarine sediment (NIST 1646a)	95	85	105	91	90

[a] Standard material supplied by the National Institute of Standards and Technology (USA).

formed monthly using one or more coupled flow meters (General Oceanic model 2030). Riverine traversal sections were calculated, based on the bathometric measurements. After mapping the transversal section of the river, each section was divided in two to four sub-sections, where the current meters were installed. In each subsection, the current meters were positioned at different depths, starting at 1.5 m from the bottom, up to the surface, in order to minimize errors caused by differences in flow from the bottom to the surface (Kjerfve et al., 1981).

A mass balance estimation was performed in order to determine the volume of water and the mass of suspended particulate matter and heavy metals moved by the Paraíba do Sul river and its tributaries, following the method described by Kjerfve et al. (1981). The water discharge values measured were assumed to be constant between two sampling periods, and the total estimated flux for each year was obtained from the sum of that obtained for each month (Woodroffe, 1985).

Figueiredo (1999), working in the same area and at the same sampling sites, compared the monthly measured fluxes of diverse chemical species (COP, NOP, DIC, DOC, N-NO_3, N-NO_2, N-NH_4 and DON) and suspended particulate matter (SPM) at gauging station 7 (Campos) calculated by two methodologies. The first method assumed that the concentration of chemical species remains constant between two sampling periods (30 days), but used daily water discharge data calculated with rating curves from the National Water and Electric Energy Department (DNAEE, 1983). The second method assumed that both concentration and water discharge were constant between the two samplings. The results obtained showed a very low variation between both methodologies. The first method always presented higher fluxes than the second; the minimum flux difference was 5% (DIC) and the maximum difference was 15% (POC, PON, SPM). Although the maximum difference between the two methodologies was up to 15%, no statistically significant difference between daily discharges measured by rating curves from DNAEE and the methodology used in this study was evident ($P < 0.001$, Wilcoxon test, $n = 26$).

4. Results and discussion

The monthly suspended particulate matter concentrations, the associated heavy metal concentrations and the Paraíba do Sul river water discharge is presented in Table 2. The calculated particulate matter and associated metals loads transported by the Paraíba do Sul River during the sampling period are presented in Table 3.

The total suspended particulate matter load transported by the Paraíba do Sul River to the South Atlantic Ocean was 250% greater in the second period (from April 1995 to March 1996) than in the first period (from April 1994 to March 1995). This trend was also observed for all the metals studied, and is probably related to higher rainfall observed in the second period in the upper and/or middle PSR basin. However, no significant difference was observed between the rainfall averages of the two studied periods along the lower basin (first period, 733 mm; second period, 779 mm) that could justify such an increment. Both of the periods studied presented average rainfall lower than the regional average (890 mm/year). The main difference between both periods that affects the annual loads were the

Table 2
Monthly heavy metal concentration in suspended particulate matter, water discharge and total suspended particulate matter

	Cu ($\mu g\ g^{-1}$)	Cr ($\mu g\ g^{-1}$)	Zn ($\mu g\ g^{-1}$)	Mn ($\mu g\ g^{-1}$)	Fe ($\mu g\ g^{-1}$)	SPM ($mg\ l^{-1}$)	Discharge ($m^3\ s^{-1}$)
April 1994	44	92	483	2305	87 843	51	830
May 1994	40	53	274	2056	92 932	18	720
June 1994	52	56	196	2148	90 356	18	730
July 1994	74	73	156	2049	74 278	12	550
August 1994	90	88	271	1934	31 307	9	340
September 1994	46	44	296	3835	57 966	13	210
October 1994	79	79	257	2096	60 540	10	240
November 1994	125	49	210	1782	71 314	21	680
December 1994	125	79	453	1443	66 970	29	680
January 1995	134	52	353	1913	87 944	21	760
February 1995	134	47	156	1348	88 285	99	1800
March 1995	42	53	129	1559	55 998	21	640
April 1995	43	52	201	3734	73 229	20	510
May 1995	36	34	170	1331	55 460	17	400
June 1995	43	60	34	4324	47 271	6	300
July 1995	49	202	787	2987	77 121	5	270
August 1995	40	126	239	3324	58 593	13	110
September 1995	105	125	446	3698	64 644	17	220
October 1995	169	124	653	4072	70 694	22	330
November 1995	127	133	506	3778	71 623	17	390
December 1995	41	142	302	1887	80 250	13	540
January 1996	76	93	263	1705	74 505	193	2600
February 1996	43	46	135	948	62 770	108	900
March 1996	42	43	159	1002	64 511	92	1330
Average	75	81	297	2386	69 434	35	670
Standard deviation	41	42	178	1043	14 778	45	558

higher water fluxes and the higher particulate matter concentrations observed in January, February and March in the second period, where the SPM increased to 193, 108 and 92 mg l^{-1}, respectively. These higher SPM values observed are probably due to an increase in the river water velocity that is direct linked to the river transport capacity. The sources of these larger particles are probably from the river bottom sediments and/or particles derived from surface runoff. This observation can be confirmed from Fig. 2, which presents the maximum, minimum and average water discharge measured by DNAEE (1983) compared with the values measured for this study. Considering that all the transported load data in the present study are representative of low rainfall periods, it is possible to speculate that the estimated Paraíba do Sul River contribution to the South Atlantic Ocean is probably underestimated. More studies are needed in order to investigate periods of higher rainfall and water discharges.

The monthly metal flow was highly variable (Table 3). Generally, the highest loads were related to the highest water discharge and suspended particulate matter concentrations. This trend is probably due to the heavy metals associated with the suspended particulate matter. It is important to point out the great importance of one of the months for the total load transported in the year. In the second sampling period (1995/1996), February presented the highest water discharge, representing 34–66% of the total heavy metal loads. Extrapolating to the load in the rainy-season months, this contribution would rise to 82–93% of the annual load. This trend highlights the importance of the surface runoff in

Table 3
Monthly distribution of the heavy metals and suspended particulate matter (SPM) loads transported by the Paraíba do Sul River (t)

	Cu	Cr	Zn	Mn	Fe	SPM
April 1994	4.8	10.1	53.0	253	9638	113377
May 1994	1.4	1.8	9.5	71	3226	34712
June 1994	1.8	1.9	6.7	73	3077	35194
July 1994	1.3	1.3	2.7	36	1295	17432
August 1994	0.8	0.7	2.3	16	263	8413
September 1994	0.3	0.3	2.1	27	415	7154
October 1994	0.5	0.5	1.6	13	373	6168
November 1994	4.6	1.8	7.8	66	2640	37014
December 1994	6.7	4.2	24.2	77	3585	53526
January 1995	5.8	2.3	15.3	83	3811	43335
February 1995	57.4	20.1	67.0	578	37889	429166
March 1995	1.5	1.9	4.6	56	2016	35998
April 1995	1.1	1.4	5.3	99	1936	26438
May 1995	0.7	0.6	3.1	24	1010	18213
June 1995	0.2	0.3	0.2	20	221	4666
July 1995	0.2	0.7	2.8	11	279	3616
August 1995	0.2	0.5	0.9	13	226	3857
September 1995	1.0	1.2	4.4	37	643	9939
October 1995	3.2	2.4	12.6	78	1360	19239
November 1995	2.2	2.3	8.9	66	1255	17522
December 1995	0.8	2.7	5.8	36	1545	19249
January 1996	102.5	124.6	353.8	2296	100309	1346342
February 1996	10.6	11.3	32.8	231	15306	243845
March 1996	13.8	14.1	52.4	330	21234	329154
Total						
1994/1995	87	47	197	1350	68228	821489
1995/1996	137	162	483	3241	145323	2042080

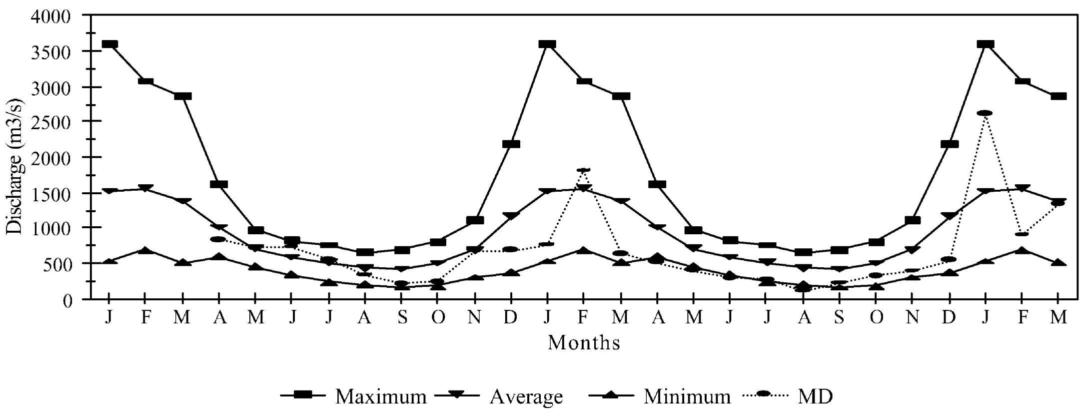

Fig. 2. Comparative graph of maximum, average and minimum monthly discharge measured by the National Water and Electric Energy Department (DNAEE) for Campos' station (based on measured discharges from 1934–1978) with the discharges measured during this study (MD).

Table 4
Percentile distribution of the heavy metals and suspended particulate matter loads transported by the Paraíba do Sul River during 1995/1996

Year 1995/1996	Cu (%)	Cr (%)	Zn (%)	Mn (%)	Fe (%)	SPM (%)
April[a]	5.5	21.5	26.9	18.7	14.1	13.8
May[b]	1.6	3.8	4.8	5.3	4.7	4.2
June[b]	2.1	4.0	3.4	5.4	4.5	4.3
July[b]	1.5	2.8	1.4	2.7	1.9	2.1
August[b]	0.9	1.5	1.2	1.2	0.4	1.0
September[b]	0.3	0.6	1.1	2.0	0.6	0.9
October[b]	0.6	1.1	0.8	1.0	0.5	0.8
November[a]	5.3	3.8	4.0	4.9	3.9	4.5
December[a]	7.7	8.9	12.3	5.7	5.3	6.5
January[a]	6.7	4.9	7.8	6.1	5.6	5.3
February[a]	66.0	42.8	34.0	42.8	55.5	52.2
March[a]	1.7	4.0	2.3	4.1	3.0	4.4
Rainy season[a]	92.9	85.9	87.3	82.3	87.4	86.7
Dry season[b]	7.1	14.1	12.7	17.7	12.6	13.3

[a] Rainy season.
[b] Dry season.

the transport of heavy metals, as well as studies on the seasonal variation in annual transported-load calculations.

The heavy metals and suspended matter loads showed a strong dependence on the rainfall, and consequently on the surface runoff. Although Salomão et al. (2001), studying the metal and SPM mass balance in this river, observed that the increase in load between two sampling points along the river in certain periods could not be explained by just the contribution of the tributaries. This fact probably indicates the importance of sediment resuspension and/or surface runoff input to the total load transported, mainly during months of high water discharge (Table 4).

Comparing the results obtained for particulate metal loads in the two periods analyzed with the calculations of Bewers and Yeats (1989) for fluvial heavy metals and suspended particulate matter input for the world oceans, it is possible to observe that the values found for PSR do not exceed 0.002% of the total metal transported load (Table 5) for all the metals studied.

Comparing the average particulate matter loads obtained in this study for the Paraíba do Sul River with some of the largest rivers of the planet, such as the Amazon, Ganges/Brahmaputra and Mississippi (Table 6), we can observe that this river is of very small importance. However, when we observe the calculated matter input for the South Atlantic Ocean carried out by Milliman and Meade (1983), where just the São Francisco and La Plata rivers were considered, we can observe that the Paraíba do Sul River transport is

Table 5
Comparison of the loads of heavy metals associated with the suspended particulate matter transported by the Paraíba do Sul River (PSR) with the global fluvial inputs (kg/year)

	SPM	Fe	Mn	Zn	Cr	Cu
Global input[a]	1.5×10^{12}	7.2×10^{11}	1.6×10^{10}	3.8×10^{9}	–	1.5×10^{9}
PSR 1994/1995	0.8×10^{9}	0.7×10^{8}	1.4×10^{6}	0.2×10^{6}	–	0.09×10^{6}
PSR 1995/1996	2.0×10^{9}	1.5×10^{8}	3.2×10^{6}	0.5×10^{6}	–	0.14×10^{6}
%	0.001	0.0002	0.0002	0.0001	–	0.0001

[a] Bewers and Yeats (1989).

Table 6
Average sediment input into the oceans for some of the world's largest rivers, comparison with Paraíba do Sul and São Francisco Rivers, and the estimate calculated for the South Atlantic Ocean (modified from Milliman and Meade, 1983)

River	Median SPM discharge (10^6 t/year)
Ganges/Brahmaputra	1670
Yellow River	1080
Amazonas	1100–1300
Mississippi	210
Huanghe	160
La Plata	92
Danube	67
Yukon	60
São Francisco	6
Paraíba do Sul	2
Estimate input for South Atlantic Ocean	98
Paraíba do Sul River	2

close to 2%. Considering the existence of some 50 other rivers of medium and small size in this area, we can speculate that the matter input for this part of the ocean is probably underestimated.

This study did not measure the flow in the Paraíba do Sul river mouth, or the role of this estuary in the retention of fluvial particles, and therefore the loads obtained cannot be considered as the heavy metal input to the South Atlantic Ocean. However, Carneiro et al. (1995), studying the inorganic nutrient dynamics in the Paraíba do Sul estuary, observed that only during the low water-discharge period did the estuary present intrusion of the saline edge. Based on this observation, we could consider that the Paraíba do Sul estuary probably functions as a channel linking the river to the adjacent shelf for most of the year. Considering that the rainy season represents more than 80% of the total load transported by the river (Table 4), we can speculate that the loads obtained for the Campos station are a good estimation of the particulate matter and heavy metal input for the adjacent continental shelf. This fact seems also seems to be corroborated by the study carried out on the continental shelf by Carvalho et al. (1993), which also demonstrated metal transport from the Paraíba do Sul River to the continental shelf.

Two other recent studies developed by Balzer et al. (1997) and Knoppers et al. (1997) on the continental platform adjacent to the outlet of the Paraíba do Sul River demonstrated high sedimentation rates for the area. According to Balzer (personal communication), 90% of the fluvial material is deposited directly at the shallow continental shelf, and later on is resuspended and transported to deeper zones.

5. Conclusions

Although this study was developed in the riverine portion of the Paraíba do Sul River, the measurements are probably very close to the real values. The study also highlights the strong dependence of the transported mass on the rainfall, and consequently on the river water discharge. The Paraíba do Sul River presents a low contribution to the world oceans, although the local contribution could be considered relevant.

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