

Sedimentation rates in the Corumbataí River basin, Brazil, derived from ^{210}Pb measurements

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Abstract Activity profiles of excess ^{210}Pb measured in four sediment cores from the Corumbataí River basin, São Paulo State, Brazil, provided an opportunity to evaluate sedimentation rates that are helpful for defining appropriate management strategies for the hydrological resources in the basin. This is because Rio Claro city and other municipalities make extensive use of surface waters for drinking water supply. The radiochemical analysis of the sediment cores yielded apparent sediment mass accumulation rates of between 406 and 1014 $\text{mg cm}^{-2} \text{year}^{-1}$ for secondary drainage lines, whereas an intermediate value of 546 $\text{mg cm}^{-2} \text{year}^{-1}$ was found in the Corumbataí River, the main drainage system of the studied area. These values provided estimates of average linear sedimentation rates of between 3.1 and 16.2 mm year^{-1} that are compatible with field evidence, with the highest value corresponding with an area characterized by accumulation of sediment.

Key words Corumbataí River basin, Brazil; erosion; excess ^{210}Pb ; physical weathering; radiochemical analysis; sediment; sedimentation rate

INTRODUCTION

Pb-210 is an intermediary member of the natural mass number $(4n+2)$ ^{238}U decay series that terminates with stable ^{206}Pb , according to the sequence: ^{238}U (4.49 Ga, α) \rightarrow ^{234}Th (24.1 d, β^-) \rightarrow ^{234}Pa (1.18 min, β^-) \rightarrow ^{234}U (0.248 Ma, α) \rightarrow ^{230}Th (75.2 ka, α) \rightarrow ^{226}Ra (1622 a, α) \rightarrow ^{222}Rn (3.83 d, α) \rightarrow ^{218}Po (3.05 min, α) \rightarrow ^{214}Pb (26.8 min, β^-) \rightarrow ^{214}Bi (19.7 min, β^-) \rightarrow ^{214}Po (0.16 ms, α) \rightarrow ^{210}Pb (22.26 a, β^-) \rightarrow ^{210}Bi (5 d, β^-) \rightarrow ^{210}Po (138 d, α) \rightarrow ^{206}Pb .

Rn-222 emanating from land surfaces is responsible for ^{210}Pb present in the atmosphere, which is removed by precipitation. The atmospheric ^{210}Pb returning to the Earth's surface as fallout is commonly referred to as unsupported (excess) ^{210}Pb , whereas the ^{210}Pb resulting from the decay of ^{238}U within rocks, soils, minerals and sediments has been termed supported (produced *in situ*) ^{210}Pb (Baskaran & Naidu, 1995).

Numerous studies have utilized ^{210}Pb data as a chronometer for sediment accumulation and mixing in lakes, estuarine, marsh, and coastal areas (see Appleby & Oldfield, 1992, for a comprehensive review), since they provide a reliable dating method over the last 100–150 years. In general, there are difficulties on obtaining ^{210}Pb concentration data that are above the detection limit in river sediments, a situation commonly encountered in Brazil, where ^{210}Pb -derived chronologies have been mainly developed for lakes and coastal areas (Godoy *et al.*, 1998). Additionally, the radio

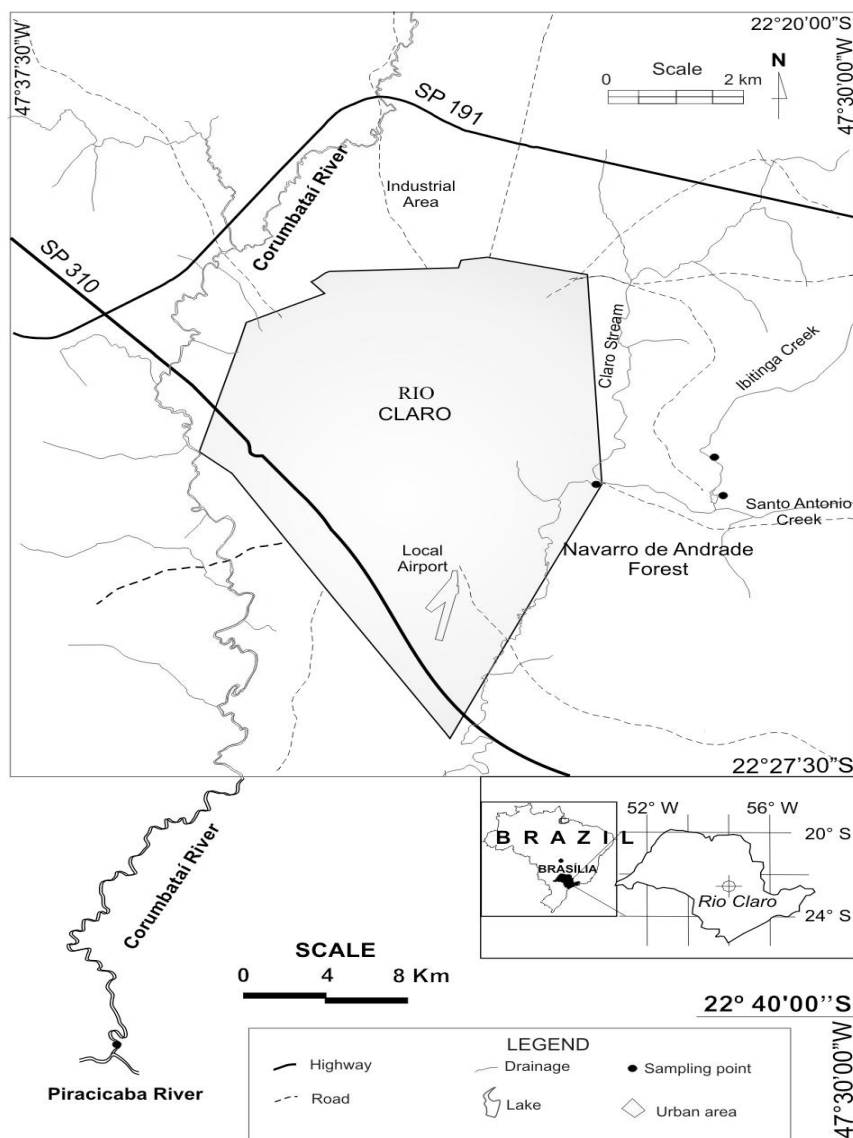


Fig. 1 Sketch map of the study area and location of the sampling sites.

active disequilibrium in the U decay series caused by ^{222}Rn loss from sediments has to be considered, since it will cause a depletion of supported ^{210}Pb , leading to non-equilibrium conditions between ^{226}Ra and ^{210}Pb (Ravichandran *et al.*, 1995). All these aspects were properly addressed in the present study undertaken in the Corumbataí River basin, an important sedimentary basin in São Paulo State, Brazil, occupied by municipalities that extensively utilize the waters of the Corumbataí River and its tributaries for drinking water supplies.

THE STUDY AREA

The Corumbataí River basin extends over an area of about 1581 km² in the middle-eastern part of São Paulo State (Fig. 1). It occurs as an eroded belt in a geomorphological province that delimits the northeastern edge of the basaltic flows in

the Paraná sedimentary basin and the crystalline plateau. Several stratigraphic units of the Paraná basin (Palaeozoic–Cenozoic) outcrop in it, and the main rock types are sandstones, conglomerates, diamictites, tillites, siltstones, mudstones, shales, limestones, basalts, diabases and related basic intrusives, and different types of Cenozoic sediments. About 65% of the Corumbataí River basin area is covered by yellow-red podzols and latosols (Köffler, 1993).

The Corumbataí River flows to the confluence with the Piracicaba River, where monthly measurements of the flow rate during the last 26 years, at Santa Terezinha (Piracicaba city) indicate that 37.4% of the observed values are between 10 and 20 m³ s⁻¹ (DAEE, 2002). Rio Claro city is the most important municipality in the basin, with about 170 000 inhabitants. The climate of the region is tropical, being characterized by wet summers (October–March) and dry winters (April–September) (Bonotto & Mancini, 1992). The area often has 55–65 days of rain per year, with more than 80% of the precipitation falling between October and March. The mean annual rainfall is ~1600 mm, with the flow rate in the Corumbataí River being directly linked to the rainfall over its catchment.

EXPERIMENTAL METHODS AND RESULTS

The sediment cores were collected in December 2001 and March 2002 from widely spaced locations in the Corumbataí River basin (Fig. 1) viz.: downstream from Rio Claro city, at the confluence of the Corumbataí River with the Piracicaba River (Santa Terezinha district, Piracicaba city) (low reach); from the Claro Stream, a tributary of the Corumbataí River; from Ibitinga Creek, a secondary tributary of the Claro Stream; from a small lake situated in the Navarro de Andrade forest, which receives water from Ibitinga Creek and discharges into Santo Antonio Creek, a tributary of the Claro Stream. The Navarro de Andrade forest is an environmentally protected area, dominated by eucalyptus trees that were extensively used for fencing rails in the past.

One core was collected from each sampling site using a Wildco Model 77263 hand core sediment sampler that contains a transparent acrylic 51 cm long (5 cm internal diameter) liner tube. No compaction of sediment and minimal perturbation of the more muddy superficial core top were noticed in the core tubes during coring. The cores were extruded in the laboratory with a polyethylene embolus, and cut into 2–6 cm sections with a porcelain spatula. The resulting sediment samples were dried, disaggregated with a porcelain mortar, homogenized, weighed, and separated into aliquots for radiochemical analysis. Aliquots for grain size analysis were pulverized using an agate pestle and mortar, sieved and classified according to Wentworth (1932). The sediment was relatively homogeneous, with dominance of fine (0.125–0.25 mm) to very fine (0.062–0.125 mm) sand throughout the profile.

The heavy metals, Cu, Zn and Pb, were analysed in one core by spectrophotometry after digestion of 2 g of powdered aliquot in a mixture of HCl + HNO₃ + HClO₄ contained in a Teflon beaker. Standard analytical techniques were used for treating the resulting solution, in order to provide the concentration data, with the different coloured complexes produced being read by a Hach DR/2000 spectrophotometer at the appropriate wavelengths (Hach, 1992). Organic matter content was also evaluated in the same core by spectrophotometry (Hach, 1992).

The ^{210}Pb in sediments was determined by its granddaughter ^{210}Po . Homogeneous portions of dried samples from each core were used, with a ~0.5 g aliquot being taken into solution in a HCl + HNO₃ + HF mixture inserted in a Parr type digestion bomb (24 h at 110°C). The solution was brought to dryness, and the dry residue dissolved with 8M HCl. A known amount (8 dpm) of ^{209}Po spike was added to assess ^{210}Po recovery. 5 ml of 20% hydroxylamine hydrochloride and 2 ml of 25% sodium citrate solution were added to the sample in a 50-ml Teflon beaker, and the pH was adjusted to 2 with NH₄OH (Flynn, 1968). Polonium was plated onto a Cu disc (1-inch diameter) suspended in the solution, heated to 85–90°C and stirred for 75–90 min. The disc was counted by α -spectroscopy with a 450 mm² area, 300 μm depletion depth, 26 keV resolution Si(Au) surface barrier detector coupled to a EG&G Ortec multichannel buffer. The MAESTRO software provided 1024 channels to plot the α -spectrum containing the ^{209}Po and ^{210}Po peaks, with the ^{210}Po activity in the sample being evaluated by isotope dilution. The total ^{210}Pb activity, $^{210}\text{Pb}_T$, was calculated from the ^{210}Po activity by considering the time Δt elapsed since the finishing of the plating procedure and the completion of α -counting, according to the equation:

$$^{210}\text{Pb}_T = ^{210}\text{Po} (e^{\lambda\Delta t}) \quad (1)$$

where λ is the ^{210}Po decay constant (0.005 day⁻¹).

The ^{238}U content of cores collected from Navarro de Andrade Lake and the Claro Stream was determined by α -spectrometry. An aliquot of about 0.5 g was dissolved in a mixture of HCl + HNO₃ + HF and placed in a Parr digestion bomb with a known amount (3.39 dpm) of ^{232}U - ^{228}Th spike. The dry residue was dissolved in 8M HCl, the U-isotopes were co-precipitated with Fe(OH)₃, the Fe³⁺ extracted with isopropyl ether, and the U separated from other interfering ions by anion exchange. The sorbed U was eluted with 0.1M HCl and electrodeposited on stainless-steel planchets that were placed in a counting chamber under 0.1 Torr vacuum for assay by α -spectrometry using a Si(Au) detector. ^{238}U activity was calculated by isotope dilution using the ^{232}U spike.

The ^{238}U content of cores collected from the Corumbataí River and Ibitinga creek (aliquots ranging from 54 to 125 g) was measured by γ -ray spectrometry using a 2" \times 2" NaI(Tl) scintillation detector and a 2048-channel multichannel analyser controlled by MAESTRO software. ^{137}Cs and ^{60}Co radioactive sources plus a pitchblende standard (1% uranium) were used to calibrate the system using the equation:

$$E = 2.15 \times 10^{-5} + 0.00169Ch \quad (2)$$

where E = energy in MeV and Ch = channel.

The γ -spectrometer was calibrated for ^{214}Bi (equivalent uranium, eU) readings using pitchblende standards from the New Brunswick Laboratory, US Department of Energy, Argonne, Illinois, USA, after waiting for ^{222}Rn to reach secular radioactive equilibrium with ^{226}Ra (at least 25 days). The calibration curve takes the form:

$$\log C_U = 1.057 \times \log I_U + 2.578 \quad (3)$$

where C_U is the eU concentration, in ppm or $\mu\text{g g}^{-1}$ and I_U = the effective intensity in cpm g^{-1} .

All radiochemical data are given in Table 1. Table 2 reports the results obtained for Pb, Cu, Zn, and organic matter analyses. The data for the more superficial and deeper layers of the Corumbataí River core were not used for sedimentation rate estimation, since a number of researchers (see Santschi *et al.*, 2001, and Baskaran &

Table 1 Radiochemical data for the sediments cores sampled from the Corumbataí River basin, São Paulo State, Brazil. analytical uncertainties of $\pm 10\%$ corresponding to 1σ (sd).

Depth range (cm)	Total dry weight (g)	Cumulated dry weight per area (g cm^{-2})	^{238}U content ($\mu\text{g g}^{-1}$)	^{238}U activity, ($^{238}\text{U}_s$) (dpm g^{-1})	^{210}Po activity, (dpm g^{-1})	Δt^a (day)	Total ^{210}Pb activity ($^{210}\text{Pb}_T$) (dpm g^{-1})	<i>In situ</i> ^{210}Pb activity ^b ($^{210}\text{Pb}_s$) (dpm g^{-1})	Excess ^{210}Pb activity ^c ($^{210}\text{Pb}_{xs}$) (dpm g^{-1})	ln ($^{210}\text{Pb}_{xs}$) (dpm g^{-1})	Dep. Time ^d (year)	Dep. year
Corumbataí River												
0–3.4	79.08	4.03	2.29	1.69	0.56	2.1	0.56	0.27	0.29	-1.24	7	1995
3.4–6.8	72.55	7.72	0.72	0.53	0.88	2.1	0.89	0.08	0.81	-0.21	14	1988
6.8–10.2	65.52	11.05	3.09	2.29	1.04	2.1	1.05	0.37	0.68	-0.38	20	1982
10.2–13.6	86.45	15.45	0.02	0.01	0.50	1.7	0.50	0.002	0.50	-0.69	28	1974
13.6–17.0	84.99	19.78	1.10	0.81	0.52	1.7	0.52	0.13	0.39	-0.94	36	1966
17.0–20.4	68.53	23.27	2.28	1.61	0.60	1.7	0.60	0.27	0.33	-1.11	43	1959
20.4–23.8	66.87	26.68	2.29	1.69	0.80	1.7	0.81	0.27	0.54	-0.62	49	1953
Navarro de Andrade Lake												
4.5–9.0	41.63	2.12	9.16	6.78	3.56	10	3.74	1.08	2.66	0.98	3	1998
9.0–13.5	47.40	4.54	5.19	3.84	2.84	10	2.99	0.61	2.38	0.87	6	1995
13.5–18.0	45.44	6.85	4.58	3.39	1.92	10	2.02	0.54	1.48	0.39	9	1992
18.0–22.5	53.14	9.55	4.27	3.16	1.96	10	2.06	0.51	1.55	0.44	13	1988
22.5–27.0	45.79	11.88	4.18	3.09	1.86	10	1.96	0.49	1.47	0.38	16	1985
27.0–31.5	48.40	14.34	7.52	5.56	1.94	10	2.04	0.89	1.15	0.14	19	1982
31.5–36.0	52.86	17.03	8.64	6.39	2.18	10	2.29	1.03	1.26	0.23	23	1978
36.0–40.5	37.08	18.91	9.16	6.78	2.34	10	2.46	1.08	1.38	0.32	25	1976
Ibitinga Creek												
4.2–8.4	124.64	6.35	0.13	0.10	0.34	10	0.36	0.02	0.34	-1.08	6	1995
8.4–12.6	104.05	11.67	0.17	0.12	0.52	10	0.55	0.02	0.53	-0.64	12	1989
12.6–16.8	85.90	16.04	0.24	0.18	0.34	3	0.35	0.03	0.32	-1.14	16	1985
16.8–21.0	101.34	21.20	0.20	0.15	0.36	10	0.38	0.02	0.36	-1.02	21	1980
21.0–25.2	97.92	26.19	0.35	0.26	0.24	3	0.25	0.04	0.21	-1.56	26	1975
25.2–29.4	102.36	31.40	0.60	0.45	0.32	10	0.34	0.07	0.27	-1.31	32	1969
29.4–33.6	111.04	37.05	0.24	0.18	0.18	10	0.19	0.03	0.16	-1.83	37	1964
33.6–37.8	110.82	42.69	0.42	0.31	0.18	10	0.19	0.05	0.14	-1.97	43	1958
Claro Stream												
1.0–2.0	32.88	1.67	0.93	0.69	1.14	10	1.20	0.11	1.09	0.09	4	1997
2.0–3.0	34.39	3.42	4.58	3.39	1.10	10	1.16	0.54	0.62	-0.48	8	1993
3.0–4.0	35.61	5.23	0.51	0.38	0.52	10	0.54	0.06	0.48	-0.73	13	1988
4.0–5.0	23.27	6.41	0.76	0.56	0.64	10	0.67	0.09	0.58	-0.55	16	1985
5.0–6.0	30.05	7.94	0.68	0.50	0.46	10	0.48	0.08	0.40	-0.91	20	1981
6.0–7.0	26.86	9.31	2.70	2.00	0.76	10	0.80	0.32	0.48	-0.73	23	1978
7.0–8.0	30.72	10.87	1.19	0.88	0.64	10	0.67	0.14	0.53	-0.64	27	1974
8.0–9.0	21.15	11.95	1.01	0.75	0.44	10	0.46	0.12	0.34	-1.07	29	1972

^a Time range between ^{210}Po plating and ending of alpha counting;

^b $^{210}\text{Pb}_s = 0.16 (^{238}\text{U})_s$;

^c $^{210}\text{Pb}_{xs} = ^{210}\text{Pb}_T - ^{210}\text{Pb}_s$;

^d Deposition time = cumulated dry weight/area ÷ sediment mass flux.

Naidu, 1995, among others) have questioned the validity of the assumption of steady-state accumulation, because mechanical mixing and diffusion of ^{210}Pb can occur in the first and last few cm of the sediment core and bioturbation can also affect the uppermost layer of the sediment, so that ^{210}Po may not have grown completely into equilibrium with ^{210}Pb .

Table 2 Chemical analyses of the sediment core from the Corumbataí River.

Depth range (cm)	Organic matter (%)	Cu ($\mu\text{g g}^{-1}$)	Zn ($\mu\text{g g}^{-1}$)	Pb ($\mu\text{g g}^{-1}$)
0–3.4	2.08	496	40	45.2
3.4–6.8	2.28	560	35	45.6
6.8–10.2	2.66	688	40	40.4
10.2–13.6	1.84	480	80	39.2
13.6–17.0	2.28	540	<0.1	50.4
17.0–20.4	3.17	360	160	45.2
20.4–23.8	4.71	162	35	56.4

The total ^{210}Pb activity in Table 1 is generally lower than the ^{238}U activity ($^{238}\text{U}_s$) that supplies the parent-supported (*in situ* produced) ^{210}Pb . This implies a radioactive non-equilibrium among ^{210}Pb and its ancestors in the ^{238}U decay series, that is caused by ^{222}Rn escaping from sediments to the surrounding fluid phase, such as water and air, i.e. only a fraction of Rn atoms formed in the solid phase will contribute to the generation of ^{210}Pb . The ^{222}Rn -loss may be computed from the emanation coefficient or emanating efficiency (E) expressed by Wanty *et al.* (1992) as:

$$E = (^{222}\text{Rn})_{\text{fluid}} / (^{222}\text{Rn})_{\text{fluid}} + (^{222}\text{Rn})_{\text{solid}} \quad (4)$$

An average value of $E = 0.84$ was obtained by Bonotto & Caprioglio (2002) on laboratory time-scale experiments conducted with sediments from the Botucatu and Pirambóia formations, whose grain size composition is very similar to that of the sediment cores analysed here. Therefore, it is possible to assume that only 16% of the ^{222}Rn generated by ^{226}Ra decay contributes to the production of ^{210}Pb , i.e. the supported ^{210}Pb , $^{210}\text{Pb}_s$, may be calculated as:

$$^{210}\text{Pb}_s = 0.16 (^{238}\text{U})_s \quad (5)$$

The excess ^{210}Pb activity, $^{210}\text{Pb}_{xs}$, is calculated by:

$$^{210}\text{Pb}_{xs} = ^{210}\text{Pb}_T - ^{210}\text{Pb}_s \quad (6)$$

DISCUSSION AND CONCLUSIONS

The sedimentation rates estimated in this study were obtained using the CRS (constant rate of supply of unsupported/excess ^{210}Pb) model of Appleby & Oldfield (1978). This assumes a constant rate of supply of unsupported/excess ^{210}Pb , a variable sedimentation and sediment compaction rate, as well a constant flux of unsupported ^{210}Pb to the sediment/water interface. The excess ^{210}Pb activity in any layer z of the sediment column, $^{210}\text{Pb}_{xs(z)}$, is (Baskaran & Naidu, 1995):

$$^{210}\text{Pb}_{xs(z)} = ^{210}\text{Pb}_{xs(0)} e^{-\lambda_{210}t} \quad (7)$$

where $^{210}\text{Pb}_{xs(0)}$ represents the excess ^{210}Pb activity at the sediment-water interface, λ_{210} is the ^{210}Pb decay constant (0.0311 year^{-1}), and t is the deposition time (age, in years). This equation can be simplified and rewritten as:

$$\ln ^{210}\text{Pb}_{xs(z)} - \ln ^{210}\text{Pb}_{xs(0)} = (-\lambda_{210}f^1)w \quad (8)$$

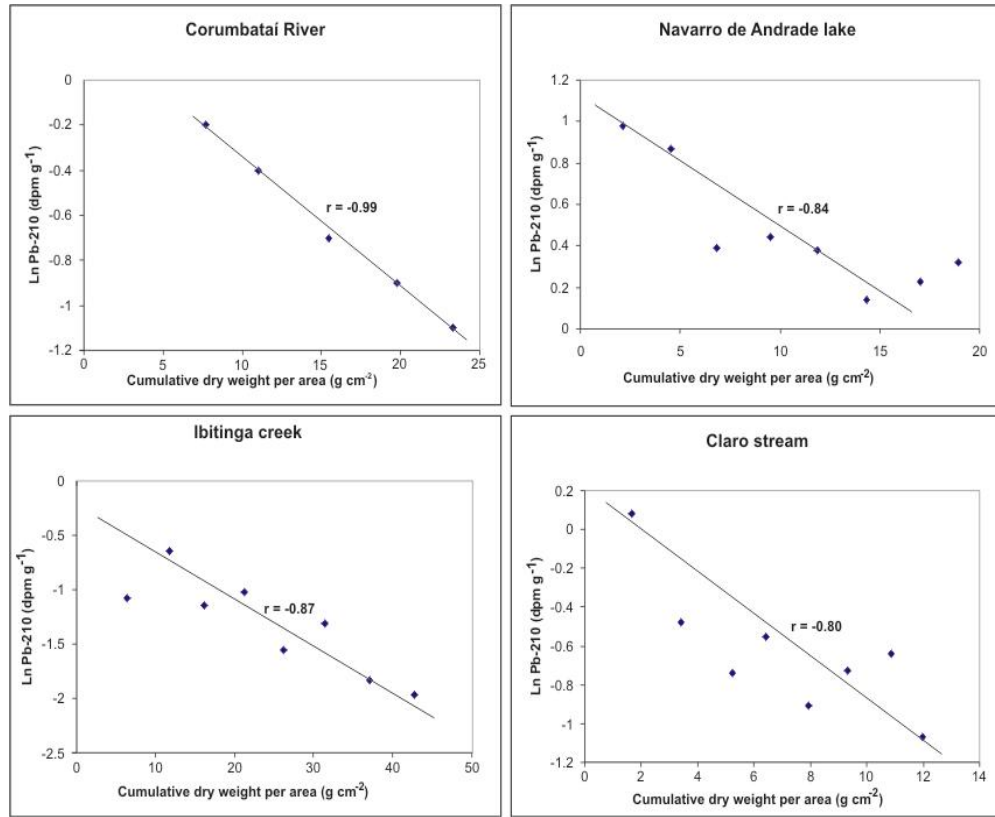


Fig. 2 Excess ^{210}Pb vs cumulative dry mass relationship for sediment cores collected from the Corumbataí River basin, São Paulo State, Brazil.

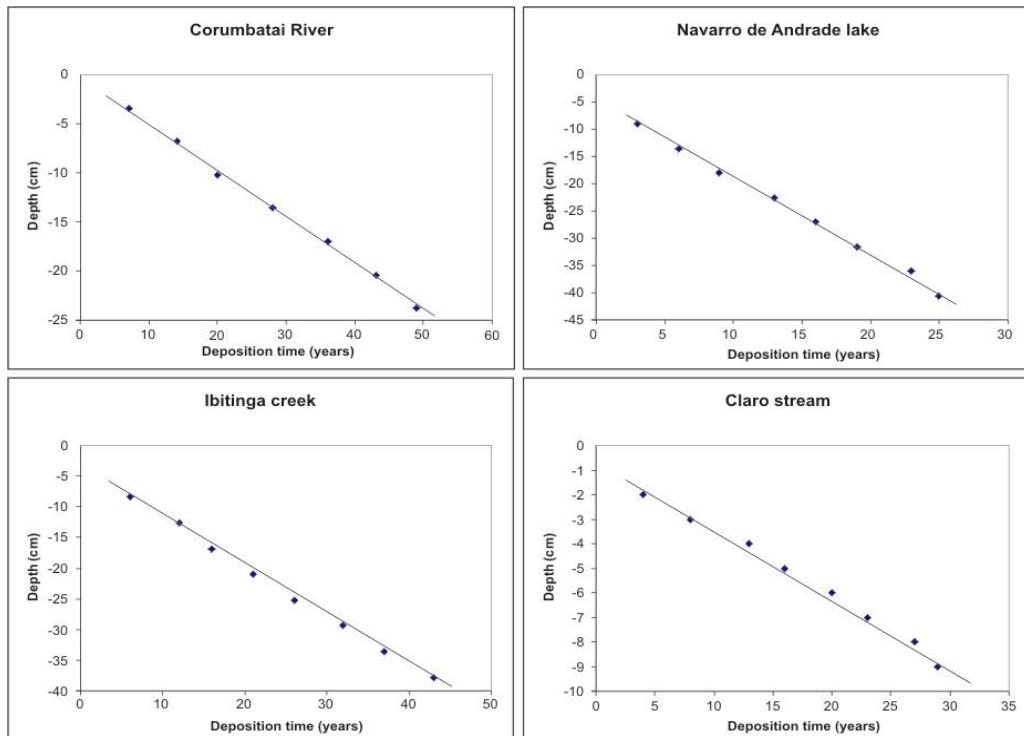


Fig. 3 Depth vs deposition time relationship for sediment cores collected from the Corumbataí River basin, São Paulo State, Brazil.

where the cumulated dry weight per unit area (g cm^{-2}), w , is related to the deposition time according to the expression

$$t = w f^{-1} \quad (9)$$

where f is the sediment mass flux in $\text{g cm}^{-2} \text{ year}^{-1}$. When $\ln^{210}\text{Pb}_{xs(z)}$ is plotted against the cumulated dry weight per unit area, w , the resulting ^{210}Pb profile will be linear, with slope $-\lambda_{210} f^{-1}$. The sediment mass flux, f , may then be determined from the mean slope of the profile, using the least-squares fit procedure (Baskaran & Naidu, 1995).

The $\ln(^{210}\text{Pb}_{xs})$ data in Table 1 are plotted against the cumulated dry weight per unit area in Fig. 2. The following mass accumulation rates are obtained: $546 \text{ mg cm}^{-2} \text{ year}^{-1}$ (Corumbataí River), $752 \text{ mg cm}^{-2} \text{ year}^{-1}$ (Navarro de Andrade Lake), $1014 \text{ mg cm}^{-2} \text{ year}^{-1}$ (Ibitinga Creek), and $406 \text{ mg cm}^{-2} \text{ year}^{-1}$ (Claro Stream). The deposition time (in years) is calculated by dividing the cumulative dry weight per unit area by the sediment mass accumulation rate, and is plotted against depth in Fig. 3. The expected deposition year for each sediment layer is also shown in Table 1, which was estimated by considering the sampling year and the water-sediment interface at the uppermost layer as a reference for establishing the chronology. The average linear sedimentation rate (in mm year^{-1}) is evaluated by division of the total thickness of the sediment column by the deposition time for the deepest layer, and corresponds to 4.8 mm year^{-1} (Corumbataí River), $16.2 \text{ mm year}^{-1}$ (Navarro de Andrade Lake), 8.8 mm year^{-1} (Ibitinga creek), and 3.1 mm year^{-1} (Claro Stream). They are compatible with values estimated for other sites, for instance, 2.9 and 6.5 mm year^{-1} , respectively, in Galveston Bay and the Mississippi River Delta, USA (Santschi *et al.*, 2001). They also agree with field evidence indicating that the highest value found in Navarro de Andrade Lake is due to the fact that it is a flatter area where the water velocity is negligible, conditions that are highly favourable to the accumulation of sediment.

Beyond modifying the bulk density and porosity of soils and sediments, organic matter also influences their specific surface area, by increasing it. Consequently, other parameters are also significantly affected, for instance, the cation exchange capacity and heavy metal adsorption. The data in Table 2 indicate that a significant direct correlation exists between Pb and organic matter ($r = 0.74$), but an inverse relationship exists between Cu and organic matter ($r = -0.80$). Pb and Cu concentrations in the sediments exceeded the maximum allowable values established by USEPA (2002), corresponding to 30 and $18.7 \mu\text{g g}^{-1}$, respectively. These preliminary results emphasize the need to undertake more detailed investigations focusing on a broad geochemical survey of the bottom sediments in the study area. Further investigations could usefully address the reasons for the differential distribution of the elements/compounds analysed, which may have implications for the generation of information to support the appropriate management of the hydrological resources in the basin.

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