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## The use of the $^{210}\text{Pb}$ method for preliminary evaluating the sedimentation rate in Amazon river mouth

SOPHIE ALTHAMMER<sup>1</sup>, ERICH FOBHAG<sup>1</sup>, HARALD MARTIN HOFFMANN<sup>1</sup>,  
 JOSÉ REINALDO CARDOSO NERY<sup>2</sup>, DANIEL MARCOS BONOTTO<sup>3</sup>

<sup>1</sup>Institute for Physical Chemistry and Radiochemistry, Mannheim University of Applied Sciences, Germany

<sup>2</sup>Universidade Federal do Amapá (UNIFAP), Brasil

<sup>3</sup>Departamento de Petrologia e Metalogenia, Universidade Estadual Paulista (UNESP), Brasil

**Abstract:** *The use of the  $^{210}\text{Pb}$  method for preliminary evaluating the sedimentation rate in Amazon river mouth.* This investigation reports the preliminary results for determining the sedimentation rate at the Amazonas River mouth, Brazil, by the  $^{210}\text{Pb}$  method. The CIC (constant initial concentration) of unsupported/excess  $^{210}\text{Pb}$  model was successfully applied to one sediments core. The measurements were made in a sediments core comprising ten samples 10 cm – thick each. The year of the sampling (2006) was used as reference for the establishment of the rate of sedimentation from the beginning of the top of the testimony. However, the atmospheric conditions and climate in Amapá State, Brazil, claimed for optimization of the results. Because the Channel North is opened into the Atlantic Ocean, it exhibits a huge water volume, causing a permanent motion of sediments and water. So, heavy waves are formed at the water surface, stirring the upper sand strata. Only at a depth of 40–50 cm the profile tends to be more homogeneous, where it is mostly composed by fine sand and very fine sand. Thus, it is reasonable utilize the CIC model from  $^{210}\text{Pb}$  data, which allows estimate a sedimentation rate corresponding to 0.61 g/cm<sup>2</sup>yr and a mean linear rate of 0.8 cm/yr. The  $^{238}\text{U}$  and  $^{210}\text{Pb}$  activities in the sediments allowed the reconstruction of the sedimentation processes over a period of about 130 years.

**Key words:** sedimentation rate; unsupported/excess  $^{210}\text{Pb}$  model;  $^{210}\text{Pb}$ -method; Amazonas River mouth.

## INTRODUCTION

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

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

Numerous studies have utilized  $^{210}\text{Pb}$  data as chronometer for sediment accumulation and mixing in lakes, estuarine, marsh, and coastal areas (see Appleby and Oldfield 1992, for a comprehensive

review), since they provide a reliable dating method over the last 100–150 years (Smith 2001). In general, there are difficulties in obtaining  $^{210}\text{Pb}$  concentration data that are above the detection limit in river sediments, a situation commonly verified in Brazil, where the  $^{210}\text{Pb}$ -derived chronology has been mainly developed at lakes and coastal areas (Godoy et al. 1998). Additionally, the radioactive disequilibrium in the U decay series generally caused by  $^{222}\text{Rn}$  loss from sediments has to be considered, since it would cause a depletion of supported  $^{210}\text{Pb}$ , leading to non-equilibrium conditions between  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  (Ravichandran et al. 1995). All these aspects were properly addressed in the present study realized at the Amazonas River mouth, Macapá city, Amapá State, Brazil.

## AREA OF INVESTIGATION

The Amazonas River starts flowing in the Andes Cordillera at Peru (5,600 m of altitude), intercepts a short part of south Colombia and flows through the northern portion of Brazil in the west-east direction until its discharge in the Atlantic Ocean between Amapá and Pará States. It is the largest in world either in extension or amount of water discharged ( $\sim 200,000 \text{ m}^3 \cdot \text{s}^{-1}$  during the rainy season) (Domínguez 2004). The Peruvian plains are reached after an extension of 1,900 km and altitude difference of 5,440 m from the beginning of the flow. However, the altitude difference is only 60 m from the Peruvian plains up to the Atlantic Ocean. The total number of tributaries corresponds to about 7,000 along its flow, where the principal in Brazil are: left

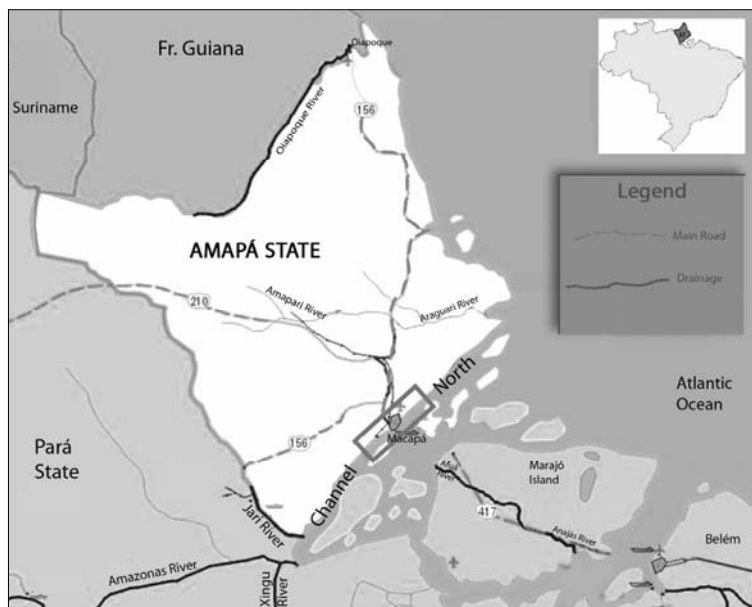


FIGURE 1. Location of Macapá city at Amapá State in Brazil and of the sampling point for the sediments core at the Amazonas River mouth

margin – Iça, Japurá, Negro, Trombetas, Paru and Jari; right margin – Javari, Jataí, Juruá, Tefé Madeira and Xingu. The discharge of Amazonas River is ~ 60 times higher than that of the Nile River, the second largest in world. The width of the Amazonas River is variable, ranging from 13 to 50 km in Manaus city (Amazonas State, Brazil), depending on the dry and rainy season. It is narrower (1,800 m) and deeper (50 m) at Óbidos city (Pará State in Brazil), where the discharge is  $200,000 \text{ m}^3 \cdot \text{s}^{-1}$ . The average water level high is 10 m, reaching 16 m in the rainy season. The amount of sediments transported into Atlantic Ocean is circa 800 billion tons per year (Brandini 2005).

The area of investigation is situated in the border of Amapá and Pará States, in the Amazon region at the northern part of Brazil (Fig. 1). It lies between the coordinates: latitude –  $1^{\circ}02' \text{ N}$  and  $0^{\circ}29' \text{ S}$ ; longitude –  $49^{\circ}21' \text{ W}$  and  $51^{\circ}11' \text{ W}$ . The region comprises the estuarine portion of Amazonas River, named Channel North, and includes Macapá city, the capital of Amapá State. The Channel North in the area exhibits a huge water volume that is discharged into Atlantic Ocean along 320 km of coastal line, reaching inclusive the Caribbean Sea (Brandini 2005).

The fieldwork was undertaken at Macapá city during the period of low discharge of Amazonas River, when sand beaches are available in the urban area of the municipality. A 1-1.1 m long PVC tube (7.5 cm in diameter) was driven into the sediments, each profile being cut in 10 cm thick slices that were transferred to polyethylene bags.

## ANALYTICAL METHODS

All bags containing the sediments were stored in iceboxes and transported to the laboratory. The water content of each section of sediments was determined after drying at  $60^{\circ}\text{C}$  for about 24 hours for organic matter preservation and minimal loss of volatile compounds. The dried sediments were classified in terms of color according to the Munsell (1975) chart, disaggregated with a porcelain mortar, homogenized, weighted, and separated into aliquots for radiochemical analysis. Aliquots for granulation analysis were pulverized using an agate mortar and pestle, sieved through a screen net defined by the Udden (1898) scale, and classified according to Wentworth (1922).

$^{210}\text{Pb}$  in the river sediments was determined by the quantification of its granddaughter  $^{210}\text{Po}$ . Homogeneous portions of dried samples from the core were used, with a 0.5 g aliquot of these powders being digested in a mixture of  $\text{HCl} + \text{HNO}_3$  at  $\sim 60^{\circ}\text{C}$ . The solution was brought to dryness, and the dry residue was dissolved with 8M HCl to a volume of 15 mL. A known amount (0.2 mL; activity = 40 dpm/mL) of  $^{209}\text{Po}$  spike was added at the beginning of each digestion to assess  $^{210}\text{Po}$  recovery. Then, 5 mL of 20% hydroxylamine hydrochloride and 2 mL of 25% sodium citrate solution were added, and the pH was adjusted to 2 with concentrated ammonia solution (Flynn, 1968). Polonium was plated onto a copper disc suspended in the solution placed on a hot plate magnetic stirrer heated to  $85\text{--}90^{\circ}\text{C}$ . Then, the disc was removed and counted by conventional alpha spectroscopy with surface barrier

detector. The system was calibrated in the 4.8–5.5 MeV region through a  $^{232}\text{U}$ – $^{228}\text{Th}$  standard radioactive source.

To obtain information about the parent-supported (*in-situ* produced)  $^{210}\text{Pb}$ , measurements of eU (=  $^{226}\text{Ra}$ ) content were performed on the same homogeneous portions of dried samples from each core used for  $^{210}\text{Po}$  analysis. Aliquots ranging from 50.9 to 64.2 g were submitted to  $\gamma$ -ray spectrometry through a 27×27 NaI(Tl) scintillation detector and a 2,048-channels multichannel analyzer provided by Ortec Ace 2K hardware controlled by MAESTRO software. The  $\gamma$ -spectrometer was calibrated for  $^{214}\text{Bi}$  (equivalent uranium, eU) readings through pitchblende standards from New Brunswick Laboratory, U.S. Department of Energy, Argonne, Illinois, USA.

## RESULTS AND DISCUSSION

The total  $^{210}\text{Pb}$  activity in the sediments profile ranged from 0.54 to 4.55 dpm/g and these values are lower than those obtained for eU, implying disequilibrium among  $^{210}\text{Pb}$  and its ancestors in the mass number  $^{238}\text{U}$  decay series.  $^{226}\text{Ra}$  reaches

$^{210}\text{Pb}$  after four  $\alpha$ -decays and two  $\beta$ -decays, producing the noble gas  $^{222}\text{Rn}$  in this decay succession. Since some of the produced  $^{222}\text{Rn}$  escapes from sediments to the surrounding water and air, only a fraction of Rn atoms formed in the solid phase will contribute to the generation of  $^{210}\text{Pb}$ . The  $^{222}\text{Rn}$ -loss was computed by the emanation coefficient (E) (Wanty et al. 1992):  $E = \frac{(^{222}\text{Rn})_{\text{fluid}}}{(^{222}\text{Rn})_{\text{fluid}} + (^{222}\text{Rn})_{\text{solid}}}$ . An average value of  $E = 0.84$  was obtained by Bonotto and Caprioglio (2002) on laboratory time-scale experiments conducted with sediments from Botucatu and Pirambóia formations whose grain size is practically similar to that of the sediments core in this study. Therefore, it was assumed that only 16% of the  $^{222}\text{Rn}$  generated by  $^{226}\text{Ra}$  decay contributed to the production of  $^{210}\text{Pb}$ . The supported  $^{210}\text{Pb}$ ,  $^{210}\text{Pb}_s$ , was calculated by the equation:  $^{210}\text{Pb}_s = 0.16 (\text{eU})$ . It ranged from 0.52 to 1.89 dpm/g. The excess  $^{210}\text{Pb}$  activity,  $^{210}\text{Pb}_{xs}$ , was calculated by the difference between total and  $^{238}\text{U}$  supported activities, i.e.,  $^{210}\text{Pb}_{xs} = ^{210}\text{Pb}_T - ^{210}\text{Pb}_s$ . It ranged from 0.01 to 2.89 dpm/g, as shown in Figure 2.

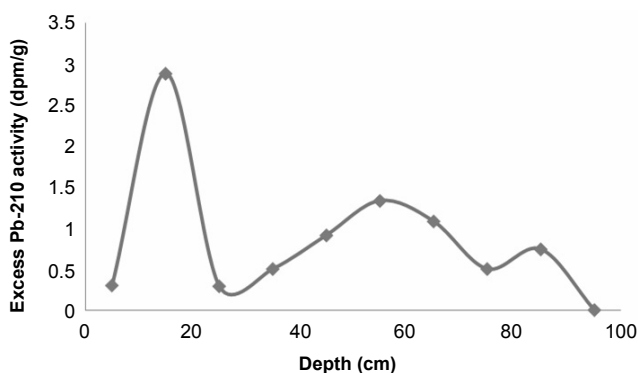


FIGURE 2. The excess  $^{210}\text{Pb}$  activity plotted against depth in the sediments profile sampled at the Amazonas River mouth

The sedimentation rate in this study was evaluated by the CIC (constant initial concentration) of unsupported/excess  $^{210}\text{Pb}$  model (Appleby and Oldfield 1978). It assumes the occurrence of sediments compaction and that the flux of  $^{210}\text{Pb}$  from the water to the sediments is constant or that the flux of the solids coming from the atmosphere and brought by rainfall is constant. This implies that the concentration of unsupported  $^{210}\text{Pb}$  in the initial sediments bed is constant. Another condition for the applicability of this model is that the sedimentation rate has to be constant during the investigated period of time, producing an exponential diminution of the  $^{210}\text{Pb}$  activity in accordance with the depth of the sediments column.

The excess  $^{210}\text{Pb}$  activity at any layer  $z$  of the sediment column,  $^{210}\text{Pb}_{\text{xs}(z)}$ , is (Baskaran and Naidu, 1995):  $^{210}\text{Pb}_{\text{xs}(z)} = ^{210}\text{Pb}_{\text{xs}(0)} e^{-\lambda_{210} t}$  where  $^{210}\text{Pb}_{\text{xs}(0)}$  represents the excess  $^{210}\text{Pb}$  activity at the sediment-water interface,  $\lambda_{210}$  is the  $^{210}\text{Pb}$  decay constant ( $0.0311 \text{ yr}^{-1}$ ), and  $t$  is the deposition time (age, in years). This equa-

tion can be simplified and rewritten as  $\ln ^{210}\text{Pb}_{\text{xs}(z)} - \ln ^{210}\text{Pb}_{\text{xs}(0)} = (-\lambda_{210} f^{-1}) w$ , where the cumulated dry mass per unit area ( $\text{g}\cdot\text{cm}^{-2}$ ),  $w$ , is related to the deposition time according to the expression  $t = w f^{-1}$  ( $f$  is the sediment mass flux in  $\text{g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$ ). When  $\ln ^{210}\text{Pb}_{\text{xs}(z)}$  is plotted against the cumulated dry weight per unit area,  $w$ , the resulting  $^{210}\text{Pb}$  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 and Naidu 1995). The relevant data for this study are given in Table 1.

The  $\ln (^{210}\text{Pb}_{\text{xs}})$  data obtained in this study were plotted against the cumulated dry mass per unit area in Figure 3. The straight line allows to calculate a sedimentation rate of  $2.19 \text{ g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$ . The deposition time (in years) has been calculated by dividing the cumulated dry mass per unit area by the sediment mass accumulation rate, and is reported in Table 1. The expected deposition year for each sediments layer was estimated considering the sampling year and the

TABLE 1. Deposition year of the sediments core sampled at the Amazon River mouth, Amapá State, Brazil

Depth range [cm]	Dry mass [g]	Mass accumulated/area [ $\text{g}\cdot\text{cm}^{-2}$ ]	Excess $^{210}\text{Pb}$ activity, $^{210}\text{Pb}_{\text{xs}}$ [dpm·g <sup>-1</sup> ]	$\ln (^{210}\text{Pb}_{\text{xs}})$ [dpm·g <sup>-1</sup> ]	Deposition time [years]	Deposition year
0–10	473.0	11.30	0.31	-1.17	5	2001
10–20	578.5	25.13	2.89	1.06	11	1995
20–30	556.0	38.41	0.30	-1.20	18	1988
30–40	552.5	51.61	0.51	-0.67	24	1982
40–50	582.0	65.52	0.92	-0.08	30	1976
50–60	564.0	79.00	1.34	0.29	36	1970
60–70	559.5	92.37	1.09	0.09	42	1964
70–80	585.0	106.34	0.51	-0.67	49	1957
80–90	505.0	118.41	0.75	-0.29	54	1952
90–100	526.5	130.99	0.01	-4.60	60	1946

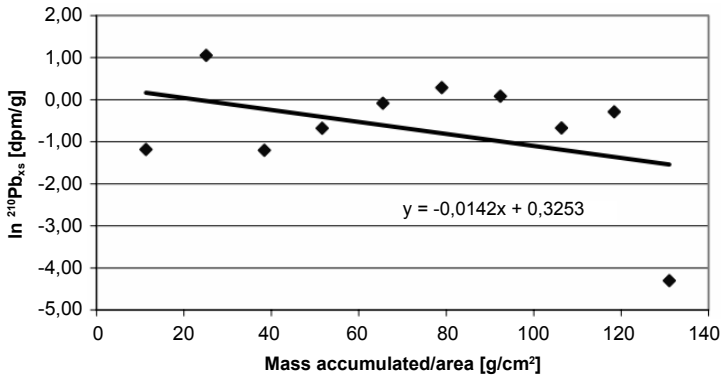


FIGURE 3. The excess <sup>210</sup>Pb activity plotted against the mass accumulated per area in the sediments profile sampled at the Amazonas River mouth

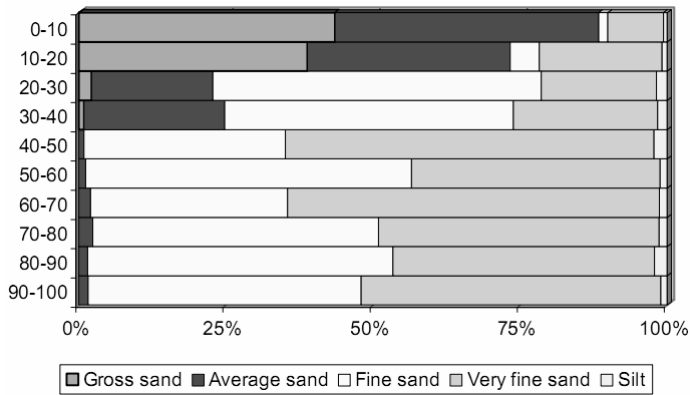


FIGURE 4. The dominant grain size of the sediments core sampled at the Amazonas River mouth

water-sediments interface at the uppermost layer as reference for establishing the chronology. The average linear sedimentation rate was evaluated through the division of the total thickness of the sediments column by the deposition time at the deepest layer, and corresponded to 1.7 cm·yr<sup>-1</sup>.

However, the atmospheric conditions and climate in the area studied request optimizations of the results. Because the Channel North is opened into the Atlantic Ocean, it exhibits a huge water volume, causing a permanent motion of

sediments and water. So, heavy waves are formed at the water surface what stirs the upper sand strata, and, consequently, the sand is very mixed and not suitable for chronological purposes. Figure 4 illustrates the dominant grain size of the sediments column, where it is possible to verify that only at a depth of 40–50 cm the profile tends to be more homogeneous, i.e. it is mostly composed by fine sand and very fine sand. Thus, the uppermost four sediments layers can be disregarded and the new data set for applying the CIC of excess <sup>210</sup>Pb model is reported

TABLE 2. Revised deposition year of the sediments core sampled at the Amazon River mouth, Amapá State, Brazil

Depth range [cm]	Dry mass [g]	Mass accumulated/area [ $\text{g}\cdot\text{cm}^{-2}$ ]	Excess $^{210}\text{Pb}$ activity, $^{210}\text{Pb}_{\text{xs}}$ [ $\text{dpm}\cdot\text{g}^{-1}$ ]	$\ln(^{210}\text{Pb}_{\text{xs}})$ [ $\text{dpm}\cdot\text{g}^{-1}$ ]	Deposition time [years]	Deposition year
40–50	582.0	13.91	0.92	-0.08	23	1983
50–60	564.0	27.38	1.34	0.29	45	1961
60–70	559.5	40.75	1.09	0.09	66	1940
70–80	585.0	54.73	0.51	-0.67	89	1917
80–90	505.0	66.80	0.75	-0.29	109	1897
90–100	526.5	79.38	0.01	-4.60	129	1877

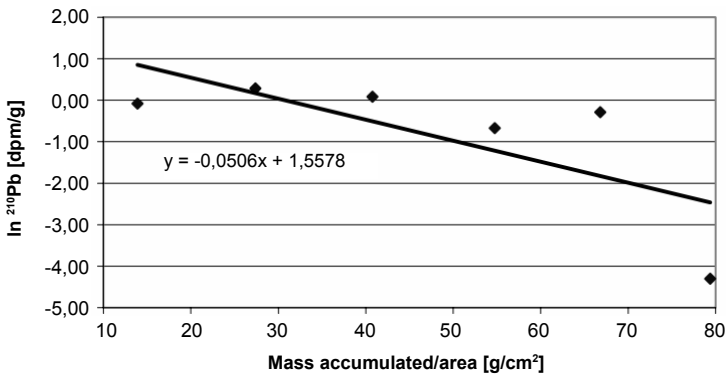


FIGURE 5. The excess  $^{210}\text{Pb}$  activity plotted against the mass accumulated per area in the sediments core sampled at the Amazonas River mouth without the four uppermost sediments layers

in Table 2 and plotted in Figure 5. The new straight line allows to calculate a sedimentation rate of  $0.61 \text{ g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$  and an average linear rate of  $0.8 \text{ cm}\cdot\text{yr}^{-1}$ . This result appears to be more realistic, tracing back the time to year 1877 that is within the time frame of the method, i.e. up to 150 years.

### CONCLUSION

Estuaries and the lands surrounding them are places of transition from land to sea, and are formed when freshwater from rivers and streams meet and mix with salt water from the ocean. Estuarine

environments are among the most productive on earth, creating more organic matter each year than comparably sized areas of forest, grassland, or agricultural land. Many different habitat types are found in and around estuaries, including shallow open waters, freshwater and salt marshes, swamps, sandy beaches, mud and sand flats, rocky shores, oyster reefs, mangrove forests, river deltas, tidal pools, and sea grasses. The productivity and variety of estuarine habitats foster an accentuated abundance and diversity of wildlife. Thousands of species of birds, mammals, fish, and other wildlife depend on estuarine habitats as places to live,

feed, and reproduce. And many marine organisms, including most commercially important species of fish, depend on estuaries at some point during their development. However, the formation of estuarine environments is greatly dependent of the sedimentation rates, and the results of this study indicated that the CIC (constant initial concentration) of unsupported/excess  $^{210}\text{Pb}$  model was successfully applied to one sediments core sampled at the Amazonas River mouth in Macapá city, the capital of Amapá State in Brazil. It was possible to estimate a sedimentation rate corresponding to  $0.61 \text{ g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$  and a mean linear rate of  $0.8 \text{ cm}\cdot\text{yr}^{-1}$ , and, thus, the  $^{238}\text{U}$  and  $^{210}\text{Pb}$  activities in the sediments allowed the reconstruction of the sedimentation processes in the area over a period of about 130 years that is within the time frame of the  $^{210}\text{Pb}$ -method.

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**Streszczenie:** Zastosowanie metody  $^{210}\text{Pb}$  do wstępnej oceny intensywności sedymentacji w ujściu rzeki Amazonki. W pracy przedstawiono wyniki badań przebiegu procesu sedymentacji w obszarze ujściowym rzeki Amazonki. Do oceny intensywności sedymentacji wykorzystano model CIC



(Constant Initial Concentration) aktywności  $^{210}\text{Pb}$ , którą badano w dziesięciu 10-cm warstwach osadu pobranego w 2006 roku. Zastosowana metoda pozwoliła na ocenę przebiegu procesu sedymentacji osadów w okresie 130 lat. Górne warstwy osadów o miąższości 40–50 cm posiadają zaburzoną strukturę i aktywność  $^{210}\text{Pb}$ , spowodowaną okresowym oddziaływaniem fal oceanicznych i wód słonych. Dolne warstwy osadu, w których dominują piaski bardzo drobne i drobne, posiadają homogeniczną strukturę oraz liniowo zmienną aktywność  $^{210}\text{Pb}$ . Stwierdzono, że średnia intensywność sedymentacji w analizowanym obszarze wynosi  $0,61 \text{ g}\cdot\text{cm}^{-2}\cdot\text{rok}^{-1}$ , co odpowiada warstwie o miąższości  $0,8 \text{ cm}\cdot\text{rok}^{-1}$ .

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Authors' addresses:

**Sophie Althammer, Erich Föbshag,  
Harald Martin Hoffmann**

Institute for Physical Chemistry and Radiochemistry,  
Mannheim University of Applied Sciences,  
Paul-Wittsack-Straße 10, 68163 Mannheim  
Germany

e-mail: [sophie.althammer@daimler.com](mailto:sophie.althammer@daimler.com)

[e.fosshag@hs-mannheim.de](mailto:e.fosshag@hs-mannheim.de)

[H.Hoffmann@hs-mannheim.de](mailto:H.Hoffmann@hs-mannheim.de)

**José Reinaldo Cardoso Nery**

Universidade Federal do Amapá (UNIFAP) Campus  
Marco Zero, Rodovia Juscelino Kubitschek,  
68902-280 Macapá, Amapá

Brasil

e-mail: [jrnery@unifap.br](mailto:jrnery@unifap.br)

**Daniel Marcos Bonotto**

Departamento de Petrologia e Metalogenia

Universidade Estadual Paulista (UNESP)

Câmpus de Rio Claro, Av. 24-A No 1515, C.P. 178

13506-900 Rio Claro, São Paulo

Brasil

e-mail: [dbonotto@rc.unesp.br](mailto:dbonotto@rc.unesp.br)