Advantages of combining <sup>210</sup>Pb and geochemical signature determinations in sediment record studies: application to coral reef lagoon environments

Jean-Michel Fernandez Benjamin Moreton Renaud Fichez Ludovic Breau Olivier Magand Christian Badie

# Introduction

During the past 150 years, terrestrial and coastal environments have been strongly modified by human activities. In the tropics, population growth and economic development imposed serious constraints on lagoon ecosystems (Hatcher *et al.*, 1989). Deforestation and mining, which are primarily responsible for hyper-sedimentation and metar pollution, are two of the major causes of disturbance in coral-lagoon environments (Carey, 1981; Naidu and Morrisson, 1994; Zan, 1994). Traces of these upheavals may be preserved in the sediment layers that gradually build up a memory of the various successive events affecting the environment. Interpretation of these "sedimentary records" is mostly based on the dating of deposits, using natural timers such as radionuclides to establish a geochronology of sediment deposition. Most of the studies dealing with recent sedimentary records of environmental changes have been based on <sup>210</sup>Pb determinations, the decrease in unsupported <sup>210</sup>Pb permitting age determinations back to about 100 years (Faure, 1986). However, deciphering both the geochemical and sedimentological data collected from these layers is not always straightforward, with results often leading to misinterpretations due to a lack of converging information.

Four cores have been selected here to demonstrate that in most cases a combination of excess <sup>210</sup>Pb measurements, geochemical and sedimentary data is necessary to reach a solid interpretation of recorded environmental changes. This paper shows how excess <sup>210</sup>Pb values may be used to distinguish between real human effects and sampling errors or natural processes.

## Materials and methods

Sediment cores were collected in the coral reef lagoons of New Caledonia and Fiji in the vicinity of Noumea and Suva towns (Table 1). Two cores were sampled in the New Caledonia Lagoon (Figure 1). The first core was taken adjacent to Noumea in Sainte Marie Bay (N12) and the second (M15) was extracted from the Dumbea sub-marine valley formed during the last glaciation, 20 000 years ago, when the sea level was roughly 120 m below present sea level. Sainte Marie Bay is a fairly enclosed system with two narrow passes connecting it to Boulari Bay in the northeast part, and the middle lagoon in the south part. The N12 core was extracted in an area where evidence of fine material deposition has been demonstrated (Dugas & Debenay, 1979, Chevillon unpublished results). Core M15 was extracted at the base of the northern slope of the submarine valley, close to the Dumbea pass.

In the lagoon of Fiji, two other cores were collected, in Suva Harbour (S14) and in Lauthala Bay (S31) (Figure 2). Suva Harbour is known to have experienced a large increase in industrial and urban developments in the coastal zone over the past 30-40 years. Lauthala Bay, where the other core was sampled, is a coral reef lagoon acting as a major recipient of the 2900 km<sup>2</sup> wide catchment area of the Rewa river.

Core	Geographical Coordinates	Depth (m)
N12	22° 17.66 E – 166° 27.74 S	13.5
M15	22° 18.25 E – 166° 15.69 S	45
S14	18° 07.34 E - 178° 24.78 S	18
S31	18° 08.89 E - 178° 28.66 S	11
		1

Table 1

Geographical coordinates and collecting depths.



#### Figure 1

Sampling locations of sediment cores collected in the Noumea Lagoon, New Caledonia.

The sediment cores were sampled using a specially devised PVC corer operated by SCUBA divers (Harris *et al.*, 2001). The corer consisted of a 1.2 metre long PVC tube, 25 cm in diameter, which had been cut in half from top to bottom. The two halves were clamped together during coring and transportation to sustain the core until sampling. The corer was forced down into the sediment by hammering on a cap placed on the top of the corer to about half a meter deep. The sediment surrounding it was then pumped away until the bottom could be sealed with a second cap. The corer was then removed and kept vertical.

Once onboard the core was allowed to settle vertically, the top was then removed and the overlying water carefully pumped out. The first fluid layers were sampled using a spatula and the core was then laid horizontally and 2 cm slices taken for geochemical and sedimenta-logical analysis. A sub-sample of each slice was used for <sup>210</sup>Pb measurements.

Finally, to obtain water content additional 10 ml sub-samples were taken and weighed before and after drying, in an oven at 110°C until constant weight. Results are expressed as a percentage of the initial sediment dry weight. This was subsequently used to calculate sediment accumulation in g.cm<sup>-2</sup>.

The concentrations of three transition metals (Fe, Mn and Ni) were determined using a sequential extraction scheme (Tessier, 1979) on the pelitic ( $<40\mu$ m) fraction of the sample. The residual phase, primarily composed of terrigeneous material, was extracted with an acidic mixture (HCI/HNO<sub>3</sub>/HF) in a high pressure vessels made of TFM (Anton Paar, MF100) and subject to micro-wave exposure in an Anton Paar Multiwave. Metals were analysed by ICP-OES (Perkin-Elmer, 3300DV model). Results are expressed in *absolute concentration* (mg.g<sup>-1</sup> extracted from the residual phase per mass unit of pelitic fraction) and *relative concentrations* (mg.g<sup>-1</sup> extracted from the residual phase).

Geochronology was defined using <sup>210</sup>Pb activity determined indirectly by measuring alpha emission from its granddaughter isotope <sup>210</sup>Po. Each sample was spiked with <sup>208</sup>Po in order to appraise possible losses incurred during application of the digestion protocol. The <sup>210</sup>Po measurement was performed in a NUMELEC gridded-chamber (NU 114B model) by alpha counting.



Sampling locations of sediment cores collected in the Suva Lagoon, Fiji.

Direct <sup>210</sup>Pb determination using gamma spectrometry at the 46.54 keV, was also conducted on the two Fijian cores. These measurements were carried out on an ORTEC X beryllium window Diode with a relative efficiency of 80%. The unsupported <sup>210</sup>Pb results were subsequently plotted for the 4 cores.

#### Results and discussion

The Lauthala Bay S31 core (Figure 3) core is one example where the data shows linear concentration profiles of Mn and Ni, in both relative and absolute values, indicating a regular supply in particulate matter (Schneider *et al.*, 1995). The <sup>210</sup>Pb measurements demonstrated the existence of a limited bioturbation layer extending down to 10 cm (5 g.cm<sup>-2</sup>) as most commonly reported world-wide (Boudreau, 1998). Below 10 cm (5 g.cm<sup>-2</sup>), a very good fit can be observed between the data and a "log (excess <sup>210</sup>Pb) vs. sediment





accumulation" regression giving an estimated accumulation rate of 0.39 g.cm<sup>-2</sup>.yr<sup>-1</sup>. In this very unambiguous case the excess <sup>210</sup>Pb and geochemical variables profiles converged to indicate that no changes in sediment deposition regime occurred during the last century and that sediment dating could be inferred from excess <sup>210</sup>Pb. This conclusion is consistent with other results demonstrating that the Rewa River basin experienced very little change during the recent past (Shorten, 1993).

In core S14 (Figure 4) a constant reduction in Ni concentrations down the core can be observed, in both relative and absolute values, together with a continuous increase in Mn between 0 and 33cm depth (0 and 17.5 g.cm<sup>-2</sup>). Below 33 cm (17.5 g.cm<sup>-2</sup>) a sharp increase in Ni and decrease in Mn was recorded in the deeper layers. The highest Ni and lowest Mn values measured in these deeper layers were equivalent to those measured in the two first centimetres of the core. This global trend was similarly marked when con-



Suva Harbour, Fiii.

sidering other transition metals profiles, leading to the hypothesis that major changes in particle supply occurred in the past over a short time period.

The excess <sup>210</sup>Pb profiles indicated that the main bioturbation layer did not extend beyond 6-8 cm (3-4 g.cm<sup>-2</sup>). Excess <sup>210</sup>Pb activity exponentially decreased with depth down to 35 cm (20 g.cm<sup>-2</sup>), where a strong anomaly was observed. At this depth, a strong increase in excess <sup>210</sup>Pb was recorded with values of a similar magnitude to those recorded in the top layers. This profile clearly indicated that the sediments in core S14 were not chronologically deposited and that the observed change at 35 cm (20 g.cm<sup>-2</sup>), was most likely a result of an artificial disturbance of the sedimentary column. This interpretation was further sustained by the documented absence of drastic changes in sediment supply in the Suva Harbour during the early Holocene (Shorten, 1993).

Sample preparation for <sup>210</sup>Po alpha counting was the first potential source of error to be considered. Additional direct gamma spec-

trometry <sup>210</sup>Pb provided results consistent with alpha determination therefore ruling out analytical failure. Furthermore, <sup>234</sup>Th and <sup>137</sup>Cs activity profiles were also strongly consistent with the distribution pattern of <sup>210</sup>Pb. An increase in water content was also observed in the last 15 cm together with a change in mud colour from black to brown. These discrepancies reinforced the hypothesis of nonchronological sediment deposition.

Two hypotheses were suggested to explain the profiles. The first one, was based on the consideration that the sampling site was located next to the commercial port of Suva where ships are often anchored before berthing. The traction pull of an anchor may have led to the displacement of a sediment layer and its subsequent deposition on top of adjacent unaltered sediments, therefore resulting in the occurrence of two similar sedimentary interfaces in a single vertical sediment column. Even though this occurrence cannot be fully dismissed it seems unlikely that the displacement of such a sediment layer would occur without significant reworking. On the contrary the <sup>210</sup>Pb and the metal profiles show a superposition of two well-stratified layers. The second hypothesis arosed from the fact that divers had experienced some problems while removing the core from the sediment bed. During this process the core, once extracted, was momentarily settled on the bottom before bringing it back to the surface and during this period it is possible that the bottom stopper of the corer was not in place. In such a situation and considering that the upper layers of the sediment were mainly composed of fluid mud, the corer by its own weight might have sampled again a 15 cm thick section of surface sediment. This scenario, resulting in the occurrence of two similar overlaying sediment sections, appears the most likely explanation to account for the almost perfectly mirrored distribution of all sediment parameters studied. Whatever the interpretation, the bottom layer was dismissed as an artefact and the 0 to 35 cm depth (0 to 20 g.cm<sup>-2</sup>) layer being solely useable to study sediment geochronology. In this layer the maximum accumulation rate calculated from the slope of the regression fit was 0.17 g.cm<sup>-2</sup>.yr<sup>1</sup>.

The M15 core produced specific profiles (Figure 5); the pelitic fraction together with absolute and relative Mn and Ni concentrations core showed a relatively constant pattern between 25 and 42 cm depth (14 to 24 g.cm<sup>-2</sup>). This trend demonstrates the sediments to be



Dumbea canyon, New Caledonia.

fairly homogeneous over more than 18 cm section of the core. The unsupported <sup>210</sup>Pb profile display a very regular and slow decrease in activity with depth, down to 43 cm (24 g.cm<sup>-2</sup>), followed by a rapid decrease below. In the upper layer, strong bioturbation is not evidenced as no homogeneous layer is observed.

A first analysis of the <sup>210</sup>Pb profile suggests a strong change in sediment deposition rate occurred at 43 cm depth. Considering this scenario, the corresponding sediment accumulation rates would have increase sharply from 0.11 g.cm<sup>-2</sup>.yr<sup>-1</sup> below 43 cm depth to 0.56 g.cm<sup>-2</sup>.yr<sup>-1</sup> in the upper layer and the age of change could be dated at around the year 1952.

A second interpretation of this data might be proposed based on the location of the sampling site. The core was sampled at 55 m depth in a submarine canyon that is delimited by steep slopes extending through a lagoon area with an average depth of 22 m. The canyon, formed by river erosion during the last low sea-level period, is now progressively filling up with sediments imported from the adjacent

and shallower lagoon bottoms. Under such conditions sediment slumps are a very likely occurrence and the presence of a very homogeneous 18 cm thick sediment layer in the middle of the core could be attributed to such an event. Similar homogeneous geochemical signatures have been documented in deposits resulting from sediment slides (Monaco *et al.*, 1982).

Interpretation of this core therefore calls for additional information which could be obtained from the study of grain size distribution throughout the core as slumps are known to cause gravimetric grain size sorting (Mear, 1984).

From the bottom to the top of the core, the pelitic fraction and the absolute Fe concentrations of the sieved sample increase in parallel with a corresponding decrease in carbonates. The Ni concentration profile clearly identifies a drastic change in the amount of the terrigeneous inputs, with a sharp increase in both absolute and relative concentrations between 12 and 11 g.cm<sup>-2</sup> corresponding to 31 and 30 cm depth, respectively.

The beginning of the <sup>210</sup>Pb profile of the N12 core indicates a large layer of almost invariable activities between 0 and 20 cm (0 to 7 g.cm<sup>-2</sup>) reflecting intense bioturbation processes and/or large accumulation rates (Figure 6). The magnitude of this phenomena makes any interpretation difficult, particularly when considering the unsupported <sup>210</sup>Pb data alone. Below 20 cm (7 g.cm<sup>-2</sup>), excess <sup>210</sup>Pb activity decreases roughly exponentially but using a single linear fit for the whole data set resulted in a poor regression fit. When comparing <sup>210</sup>Pb and metal data it appears possible to propose an environmental change scenario compatible with the <sup>210</sup>Pb profile. There is a clear shift in sediment composition and especially in Ni concentrations at 30 cm depth (11 g.cm<sup>-2</sup>) which is very likely to be associated with change in deposition regime. Hence, the linear fit calculated for the <sup>210</sup>Pb data below 30 cm (11 g.cm<sup>-2</sup>) was not extended to the above layer and another fit was drawn between 30 and 20 cm (11 and 7 g.cm<sup>-2</sup>). Two regression lines were calculated for the <sup>210</sup>Pb distribution yielding different sediment accumulation rates and allowing this major environmental change to be dated at 1952 +/- 5 years.





Despite large uncertainties regarding the calculation, the proposed date of sedimentation change corresponded with the beginning of large scale and extensive open-cast mining activities in the Southern part of New Caledonia (Bird et al., 1984; Mermoud, 1994). Those past activities were responsible for the long term erosion of soils with the lagoon acting as a final reservoir of all eroded land material. The influence of the mining exploitation has also been clearly recorded in another core taken from the Dumbea Bay (Ambatsian, 1997), a Bay where sedimentation mechanisms have been widely studied in the past (Launay, 1972). The values from both Bays present converging results regarding the decrease in excess <sup>210</sup>Pb with sediment depth. Unlike Dumbéa Bay, Sainte-Marie Bay where our N12 core was sampled is not directly under the influence of river inputs, the nearest estuary being 11 km away, indicating that additional terrigeneous inputs related to poor mining practice affected the coastal zone on a large scale.

### Conclusion

Most of the sediment cores we have been dealing with are not as easily interpreted as core S31. This relatively rare case seems to present the ideal conditions regarding particle supply and biological mixing to obtain reliable estimate of accumulation rates (consistent and constant particulate matter inputs and a reduced bioturbation layer) and therefore of dating. In sedimentary deposits subject to various sources of perturbation including the impact of the human activities, the combination of excess <sup>210</sup>Pb with geochemical data can provide necessary additional information to allow for a proper interpretation of sediment deposition processes.

Aknowledgements

This work was part of a research project financially supported by IRD and the French "Programme National Environnement Côtier" (PNEC).

#### Bibliography

AMBATSIAN P., FERNEX F., BERNAT M., PARRON C., LECOLLE J., 1997 – High metal inputs to closed seas: the New Caledonian Lagoon. Journal of Geochemical Exploration., 59: 59-74.

BOUDREAU B.P., 1998 – Mean mixed depth of sediments: The wherefore and the why. *Limnol. Oceanogr.*, 43: 524-526.

BIRD E. C., DUBOIS J-P., ILTIS J. A., 1984 – The impacts of opencast mining on the rivers and coasts of New Caledonia. The United Nations University, NRTS-25 / UNUP-505, ISBN 92-808-0505-5, 53 p.

CAREY J., 1981 – Nickel minning and raffinery wastes in Coral Sea environs. *In: Proceed. 4<sup>th</sup> Intern. Coral Reef Symp., Manila*, 1: 137-146.

DUGAS F., DEBENAY J.-P., 1979 – Carte sédimentologique et carte annexe au 1/50 000° de Nouvelle-Calédonie. Orstom. Bondy.

FAURE G., 1986 – Principles of isotope geology. Wiley & Sons, New York, 374 p. HARRIS P., FICHEZ R., FERNANDEZ J. M., GOLTERMAN H.L., BADIE C., 2001 – Phosphorus enrichment in the Papeete Lagoon (Tahiti, French Poynesia): using sediment geochronology to reconstruct the evolution of phosphorus inputs during the past century. *Oceanologica Acta*, 24: 1-10.

HATCHER B. G., JOHANNES R. E., ROBERTSON A. I., 1989 – Review of research relevant to the conservation of shallow tropical marine ecosystems. *Oceanogr. Mar. Biol. Annu. Rev.*, 27: 337-414.

LAUNAY J., 1972 – La sédimentation en baie de Dumbéa (Côte ouest - Nouvelle-Caledonie), *Cah. Orstom, ér. Géol.*, IV (1): 25-51.

MEAR Y., 1984 -Séquences et unités sédimentaires du glacis rhodanien (Méditerranée occidentale). Thesis., University of Perpignan, 214 p.

MONACO A., ALOISI J. C., BOUYÉ C., GOT H., MEAR Y., BELLAICHE G., DROZ L., MIRABILE L., MATTIELO L., MALDONADO A., LE CALVEZ Y., CHASSEFIÈRE B., NELSON H., 1982 – Essai de reconstitution des mécanismes d'alimentation des éventails sédimentaires profonds de l'Ebre et du Rhône. *Bulletin de l'Institut de Géologie du Bassin d'Aquitaine*, 31 : 99-109. NAIDU S. D., MORRISON R. J., 1994 – Contamination of Suva Habour, Fiji. *Mar. Poll. Bull Special Issue.*, 29: 52-61.

MERMOUD Y., 1994 – Processus de dégradation du milieu naturel dans le bassin versant de la Rivière des Pirogues. Mémoire de Licence, Université française du Pacifique, centre de Nouméa.

SCHNEIDER W., SCHMELZER I., WURTZ J., 1995 – Sedimentoogical interplay of siliciclastic Rewa river input and organic carbonate production of the Suva barrier reef, Laucala Bay, Fiji. Technical Report, Marine Studies 95/4, The University of the South Pacific eds, 12 p.

SHORTEN G. G., 1993 – Stratigraphy, sedimentology and engineering aspects of Holocene organo-calcareous silts, Suva Harbour, Fiji. *Marine Geology*, 110: 275-302.

TESSIER, A., CAMPBELL P. G. C., BISSON M., 1979 – Sequential procedure for the speciation of particulate trace metals. *Analitycal Chemistry.*, 51: 844-851.

ZAN L. P., 1994 – The status of coral reefs in south western Pacific islands. *Mar. Poll. Bull Special Issue.*, 29: 52-61.