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Vertical distribution and inventories of $^{239+240}$Pu and mercury in Sagua la Grande estuary, Cuba

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ABSTRACT

The vertical activity distribution and inventories of $^{239+240}$Pu profile and Hg were determined in Sagua la Grande estuary, Cuba. The shape of the $^{239+240}$Pu profile in the core column resembled very closely the history of atmospheric nuclear weapons' testing, and the maximum deposition in 1963 was recorded in the sediment core history. The $^{239+240}$Pu activity concentrations in the surface layer sediments varied from 0.163 to 0.611 mBq g^{-1}. The inventory of $^{239+240}$Pu was 42 ± 5.6 Bq m^{-2}, a value close to that expected from direct global fallout. Using the $^{239+240}$Pu as a chronomarker the mass sedimentation rate in the area for the last 60 years was calculated, reaching values of 0.173 g cm^{-2} y^{-1}. The mercury profile reflects the history of anthropogenic pollution in the estuary and perfectly describes the operation of the mercury-cell chlor-alkali plant, for production of NaOH, which began operations in 1980. The inventory of Hg was 2.42 ± 0.19 μg cm^{-2}. These results contribute to the scarce regional database for pollutants and anthropogenic radionuclides in the Caribbean marine environment, particularly in relation to $^{239+240}$Pu.

1. Introduction

In Cuba, several studies have been carried out to establish levels of artificial and natural radioactivity (^{137}Cs, ^{90}Sr, ^{210}Po, ^{210}Pb, ^{40}K) in the marine environment (Hernández et al., 1998; Alonso-Hernández et al., 2006) and their effects on the population's health (Alonso-Hernández et al., 2002), but levels and inventories of $^{239+240}$Pu are not known. Studies on the distribution and inventories of fallout Pu are important for source characterization of possible future non-fallout Pu contamination in aquatic environments and useful for dating recent sediments to understand better the pollution history of environmental contaminants.

Plutonium was introduced into the environment mainly through fallout from atmospheric nuclear weapons tests. Approximately 6 t of ^{239}Pu were released as a result of 541 atmospheric weapon tests (UNSCEAR, 2000). Isotopes of plutonium are used as a geochemical tracer to study sediment mixing rates (Cochran et al., 1995; Roberts et al., 1997), to evaluate scavenging processes in the water column (Buesseler, 1997; Gustafsson et al., 1998; Yamada et al., 2006), and to study the recent chronology of sediments (Santschi et al., 1980, 1984, 1999). Some advantages have been defined over other geochronograhy proxies, like ^{137}Cs. Plutonium has less mobility and can be used as a more stable chronographic tracer. Pu radioisotopes also have greater measurement sensitivity using alpha spectrometry and mass spectrometric techniques compared to ^{137}Cs measurements by gamma spectrometry. For estuarine and near-shore sediments Pu sometimes has another significant advantage over ^{137}Cs due to its enrichment in bottom sediment relative to ^{137}Cs, resulting from the more efficient scavenging of dissolved Pu in seawater by sediment particles. Further, with a half-life of 30.01 years, more than 60% of the ^{137}Cs inventory has already decayed away since the nuclear-testing era. This has significantly reduced the sensitivity of ^{137}Cs as a tracer of sediment transport. In contrast, only 0.2% of the plutonium atoms have decayed, owing to the much longer half-lives of ^{239}Pu (24,110 years) and ^{240}Pu (6561 years).

In recent years a large environmental programme, supported by the Technical Cooperation Department of the International Atomic Energy Agency (IAEA-TC), started in Cuba. The aims of the programme are to define levels, distribution and effects of environmental radioactivity in marine ecosystems as well as to use environmental radiotracers for understanding sedimentation regimens and pollution trends. As part of this programme, four areas in Cuba were selected as case studies: Havana Bay (highly

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polluted for petroleum and its derived), Cienfuegos Bay (where arsenic spills took place and a petrochemical plant is in development), Batabanó Gulf (main area for lobster fisheries in the country) and Sagua la Grande estuary (high mercury contamination). The principal results of this programme have been published in the past few years (Alonso-Hernández et al., 2005, 2006; Díaz-Asencio et al., 2009, 2011; Tolosa et al., 2009, 2010; Alonso-Hernandez et al., 2011).

Recently, Diaz-Asencio et al., (2009) published a study of sedimentation processes and temporal evolution of Pb and Hg fluxes in Sagua la Grande estuary for the past 100 years, deduced from ^{210}Pb and ^{137}Cs chronology. The historical sedimentary record showed a strong enrichment of mercury concentrations since 1980, caused by the incomplete treatment of industrial wastes from a chlor-alkali plant with mercury-cell technology in the Sagua la Grande river basin.

In this work we present the vertical distribution and inventories of $^{239+240}$Pu and mercury in the north central part of Cuba, reconstructed using a sediment core from Sagua la Grande estuary. The sedimentation rates are calculated using a sediment model age based on peak concentration and $^{239+240}$Pu fallout history. This will help in understanding the behaviour of mercury over time through the investigation of vertical concentration profiles in sediment.

2. Materials and methods

2.1. Study area

The basin of the Sagua la Grande river (Fig. 1) has a catchment of 2188 km^2, corresponding to 21% of Villa Clara Province, including fully or partially the municipalities of Santa Clara, Sagua la Grande, Santo Domingo, Manicaragua, Quemado de Güines, Cifuentes and Ranchuelo, with numerous industrial parks and urban establishments (approximately 390,000 inhabitants). The river discharges into Santa Clara Bay, an estuary with a mean depth of 6 m. This estuary is surrounded by mangrove forests, has many small islands and is the natural environment for many fish and mollusc species. This ecosystem is a natural boundary for protecting part of the coralline barriers located outside the Sabana–Camaguey archipelago. In addition, a small fishing town is located west of the river's mouth.

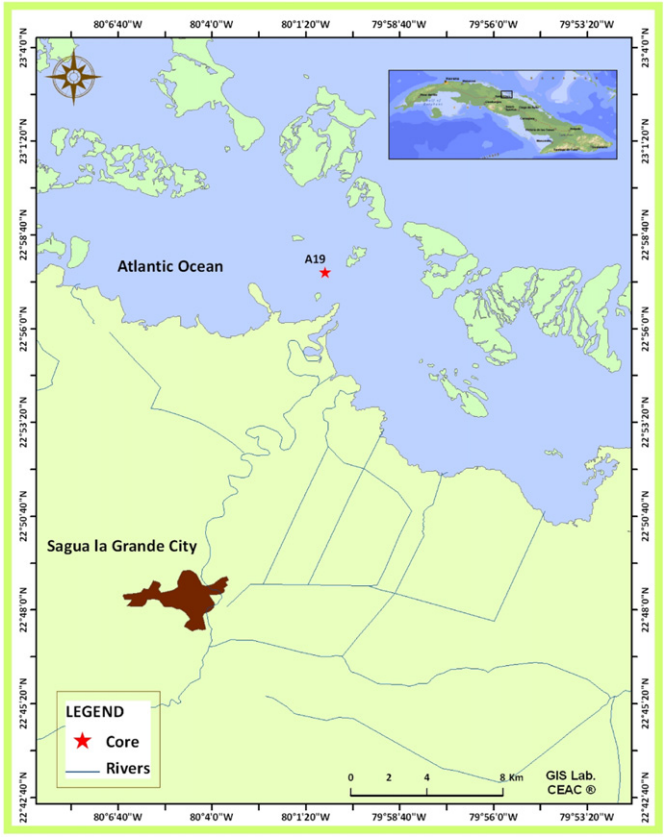

Fig. 1. Map of Sagua la Grande region.

2.2. Sampling

The sediment core A19 (12 cm inner diameter, 1 m length) was collected from Sagua la Grande estuary (22°57.36′ N 80°00.47′ W) at 6 m depth in September 2005, by a scuba diver gently inserting a plastic corer into the sediment in order to ensure minimal disturbance of the sediment–water interface. The sediment core was vertically extruded and sliced at 1.5 cm intervals. Each section was freeze-dried, homogenized and stored in the laboratory for subsequent analysis. The fraction of sand (>63 μm) and mud (clay and silt) (<63 μm) in each sample was estimated via grain size analysis using the gravimetric method (Loring and Rantala, 1992) after wet sieving through a 2 mm stainless steel sieve.

2.3. Quantification of $^{239+240}$Pu

Aliquots of 10 g were used to analyse $^{239+240}$Pu. Briefly, plutonium was extracted from the sediment using hot 8 M HNO_3 (3×) and the sediment residue discarded. Then, it was purified from chemical and spectrometric interferents by employing ionic resins AG 1 × 8 (20–50 mesh; 8 M HNO_3 medium) and AG 1 × 8 (50–100 mesh; 12 M HCl medium). Finally, plutonium was electroplated onto stainless steel discs and quantified by high-resolution alpha spectrometry. The average chemical recovery was calculated by using ^{242}Pu as an internal tracer.

Quality control was achieved by analysing certified samples (IAEA-385, IAEA-384), by checking the tracer activity and by controlling the counting instrumentation (background levels, electronical adjustment, calibration, etc.).

The $^{239+240}$Pu inventory, defined as the concentration of activity per surface unit (Bq m^{-2}), was calculated from the dry bulk density of the sediment samples and the $^{239+240}$Pu activities using the following equation:

$$I = \sum_{i=1}^{n} (A_i * D_{mi})$$

Where I is the $^{239+240}$Pu inventory in the sediment core (Bq m^{-2}), A_i is the $^{239+240}$Pu activity of the mass depth interval (Bq g^{-1}), and D_{mi} is the mass depth interval (g m^{-2}). Where depth intervals have not been measured, A_i was estimated by interpolation.

2.4. Mercury

Samples were dried in a laboratory oven at 40 °C for 48 h, ground using plastic mortars, and sieved through a 63 μm plastic mesh. Sample aliquots were weighed (20–50 mg, Gibertini scale with error of 0.001 mg) and placed in pre-cleaned combustion containers.

Total mercury concentrations were determined using an Advanced Mercury Analyser (LECO AMA-254) with a detection limit of 0.01 ng Hg. The samples were dried at 120 °C for 70 s and thermally decomposed at between 550 and 750 °C for 200 s under an oxygen carrier gas. Decomposition products were carried by carrier flow to an Au-amalgamator. Selectively trapped mercury

was released from the amalgamator by a brief heat-up and finally quantified (measuring cycle, 45 s) as Hg^0 by cold vapour AAS at 253.65 nm (EPA method 7473; US EPA, 1998). Quality control was assessed by analysing duplicate sediment samples and Standard Reference Materials (PACS-1, BEST-1, SRM 1645 and SRM 1646).

3. Results and discussion

The distribution of sediment particle sizes, $^{239+240}$Pu activities and Hg levels obtained in the sediment core A19 are reported in Table 1. The textural composition of the sediments in the core was classified as clay and silt clay. The total mud material (<63 μm) was 70–90%. Changes in the textural composition of the sediments are observed along the sediment core (Fig. 2). An increment of finer particles was obtained from the 35 cm layer towards the surface suggesting, since that layer was deposited, the existence of a system with smaller energy or changes in land use. These changes may be associated with the construction of the "Rio Nuevo" canal or the "Alacranes" dam, in 1953 and 1972, respectively. Both constructions substantially reduced freshwater flows to the estuary.

3.1. Vertical distribution of $^{239+240}$Pu and Hg

The $^{239+240}$Pu activities in the sediment core ranged between 0.16 and 0.61 mBq g^{-1} and displayed a considerable degree of variability with depth (Fig. 3a). The $^{239+240}$Pu activity in the surface sediment was 0.163 ± 0.065 mBq g^{-1}. The maximum value (0.609 ± 0.082 mBq g^{-1}) was recorded at 17 cm depth. From the 21.5 cm layer to the core bottom, $^{239+240}$Pu was not detected. Fig. 3b shows the concentrations per unit area and mass depth. In Table 2, the $^{239+240}$Pu concentrations obtained in this study are compared with $^{239+240}$Pu concentrations in sediments from other regions. The values of $^{239+240}$Pu quantified in the sediments from Sagua la Grande estuary showed similar levels those reported in the literature. Data on regional distributions of $^{239+240}$Pu in Caribbean Sea samples are scarce, however the values obtained are similar to those reported (0.01–1.09 mBq g^{-1}) by Yeager et al. (2004) in the Gulf of Mexico.

The history of atmospheric nuclear weapons' testing, and hence fallout plutonium deposition, can be broadly divided into two phases: the pre and post-moratorium weapons testing years (Buesseler, 1997). During the pre-moratorium years of 1952–1958,

Table 1
Distribution of sediment particle sizes $^{239+240}$Pu activities and Hg levels obtained in the sediment core A19, Sagua la Grande estuary.

Depth cm	Depth g cm^{-2}	Sand %	Silt %	^{239,240}Pu mBq g^{-1}	^{239,240}Pu mBq cm^{-2}	Hg μg g^{-1}
2	0.64	16.94	68.13	0.163 ± 0.102	0.105 ± 0.065	0.301 ± 0.014
3.5	1.13	11.25	69.62	0.43 ± 0.068	0.211 ± 0.034	0.305 ± 0.014
5	1.63	19.82	61.66	0.347 ± 0.095	0.174 ± 0.047	0.312 ± 0.014
6.5	2.37	17.19	66	0.486 ± 0.092	0.287 ± 0.054	0.260 ± 0.012
8	3.03	16.02	66.9	0.362 ± 0.028	0.241 ± 0.019	0.262 ± 0.012
9.5	3.77	20.7	61.51	0.412 ± 0.064	0.304 ± 0.047	0.270 ± 0.012
11	4.51	24.36	58.34	0.522 ± 0.094	0.386 ± 0.07	0.242 ± 0.011
12.5	5.28	15.68	65.15	0.611 ± 0.096	0.422 ± 0.066	0.139 ± 0.006
14	6.05	22.14	59.13	0.548 ± 0.072	0.418 ± 0.055	0.095 ± 0.004
15.5	6.93	16.91	63.28	0.425 ± 0.068	0.39 ± 0.062	0.113 ± 0.005
17	7.8	13.99	65.6	0.609 ± 0.082	0.529 ± 0.071	0.098 ± 0.004
18.5	8.63	12.69	65.33	0.286 ± 0.085	0.238 ± 0.07	0.105 ± 0.005
20	9.46	14.81	66.7	0.288 ± 0.12	0.241 ± 0.1	0.102 ± 0.005
21.5	10.35	16.55	62.41	0.206 ± 0.045	0.183 ± 0.04	0.083 ± 0.004
23	11.33	15.48	61.96	n.d.	n.d.	0.088 ± 0.004
24.5	12.24	19.05	59.86			0.080 ± 0.004
26	13.22	16.47	63.65			0.072 ± 0.003
27.5	14.37	15.61	61.77			0.060 ± 0.003
29	15.38	14.98	63.1			0.050 ± 0.002
30.5	16.28	15.56	62.31			0.052 ± 0.002
32	17.34	17.41	61.25			0.041 ± 0.002
33.5	18.25	28.21	54.71			0.048 ± 0.002
35	19.27	31.99	51.37			0.044 ± 0.002
36.5	20.36	26.43	54.21			0.039 ± 0.002
38	21.31	26.48	52.78			0.034 ± 0.002
39.5	22.35	24.36	55.61			0.047 ± 0.002
41	23.29	24.18	55			0.036 ± 0.002
42.5	24.24	21.64	56.86			0.036 ± 0.002
44	25.13	27.66	53.35			0.037 ± 0.002
45.5	25.93	32.87	49.91			0.037 ± 0.002
47	27.01	32.85	49.12			0.035 ± 0.002
48.5	27.64	29.05	50.62			0.032 ± 0.002
50	28.64	18.66	58.92			0.034 ± 0.002
51.5	29.7	23.35	55.52			0.037 ± 0.002
53	30.96	32.21	50.67			0.038 ± 0.002
54.5	32.09	26.88	53.18			0.033 ± 0.002
56	33.43	30.84	50.4			0.033 ± 0.002
57.5	34.06	36.12	45.63			0.032 ± 0.002
59	35.16	33.28	46.59			0.034 ± 0.002
62	35.97	32.16	49.03			0.036 ± 0.002
63.5	36.73	30.88	46.67			0.037 ± 0.002
65	37.56	20.99	52.29			0.033 ± 0.002
66.5	38.4	27.23	48.23			0.032 ± 0.002
68	39.31	17.96	52.66			0.031 ± 0.002
69.5	39.96	16.7	53.11			0.032 ± 0.002

n.d. Not detectable.

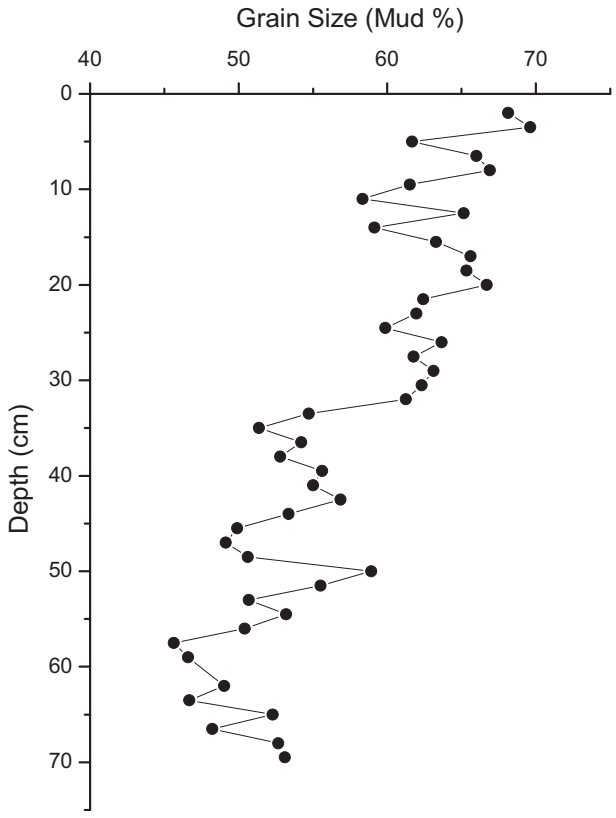

Fig. 2. Depth distribution of grain size composition in the A19 core.

the high-yield thermonuclear tests conducted by the United States dominated atmospheric pollution. Between November 1958 and September 1961, the testing moratorium between the USA and the former Soviet Union temporarily ended all new fallout inputs. The peak of global fallout deposition was in 1963 due to a series of large-scale atmospheric nuclear tests conducted by the former Soviet Union in 1961–1962. In 1963, the Limited Test Ban Treaty between the USA and the former Soviet Union put an end to the major atmospheric nuclear-testing programmes; since then, smaller amounts of nuclear debris have been introduced into the atmosphere by French and Chinese activities (UNSCEAR, 1982).

Assuming that the maximum $^{239+240}$Pu observed in Fig. 3b corresponds to 1963, then an average mass sedimentation rate of 0.173 g cm^{-2} y^{-1} was obtained for the period 1963–2005. This value agrees with the mass accumulation rate (0.177 g cm^{-2} y^{-1}) in the area reported by Díaz-Asencio et al. (2009), derived from the ^{210}Pb and ^{137}Cs chronology. These results confirm the possibility of using $^{239+240}$Pu as an independent chronostratigraphic marker for the study of sedimentation processes in Cuba and the Caribbean Sea for the last 60 years.

The inventory of $^{239+240}$Pu deposition in the core A19 is 42.0 ± 5.6 Bq m^{-2}. The inventory obtained is lower than those reported for other places (see Table 2). However, this value is in good agreement with the integrated atmospheric fallout of 36 Bq m^{-2} for 20–30 °N published by UNSCEAR (2000), and confirms that the area was affected only by global radioactivity deposition. The slight difference observed could be attributed to the lateral transport of sediments, effects commonly evidenced in Cuban estuaries (Alonso-Hernández et al., 2006).

It is well established that particle size distribution in sediment has a marked effect on the concentration of pollutants per unit mass, due to the larger surface area to mass ratio of fine grained sediments. Thus, significant variation in particle size distribution with depth in the sediment cores could lead to features in the concentration profile that do not represent real changes in contamination levels. In order to establish background contaminant levels and identify potential outliers, normalization is required to mitigate differences in grain size distribution among sediment samples. Fig. 4 presents the Hg profile concentration normalized to <63 grain-size fractions. The Hg profile (Fig. 4) shows concentrations below 0.05 mg kg^{-1} from the 30 cm layer to the core bottom. Between the 30 and the 13 cm layers, Hg levels progressively

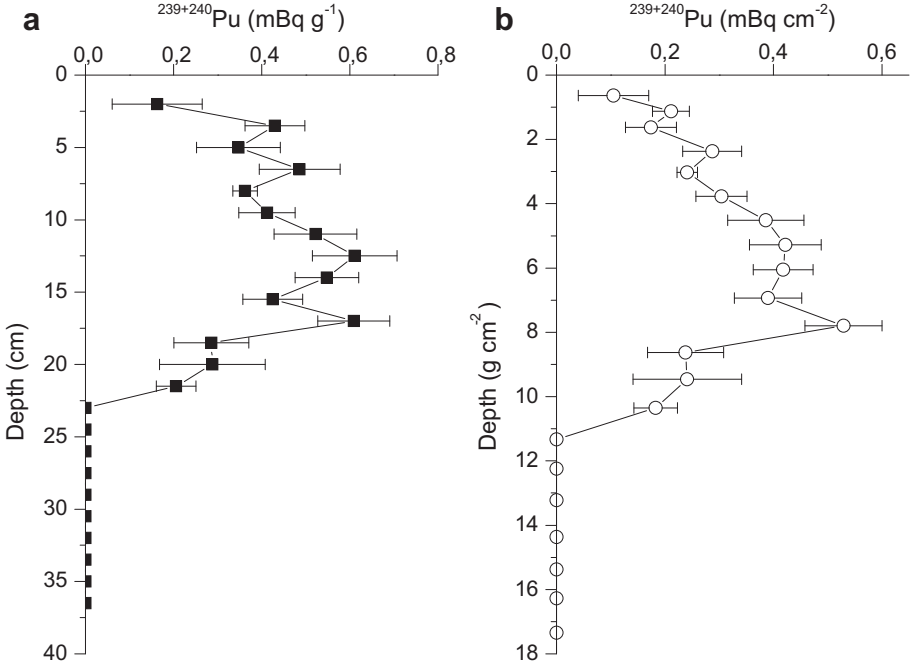

Fig. 3. Vertical profiles of $^{239+240}$Pu in sediment core A19: a) Concentrations in mBq g^{-1} vs cm depth, b) Concentrations in mBq cm^{-2} vs g cm^{-2} depth.

Table 2
Comparison of Pu concentrations and inventories with other sites.

Location	$^{239+240}$Pu (Bq kg^{-1})	Inventory $^{239+240}$Pu (Bq m^{-2})	Reference
Sagua la Grande Estuary, Cuba	0.03–0.609	42.0 ± 5.6	This work
Northern Gulf of Mexico	0.01–1.09	22–68.3	(Yeager et al., 2004)
Lake Tamogi, Japan		111–230	(Ueda et al., 2009)
Spain	0.04–0.57	31–101	(Gasco et al., 2006)
Gdansk, Baltic Sea	0.93–6.50	95–210	(Skwarzec et al., 2003)
Brazil	0.018–1.17		(Figueira et al., 2006)
Yangtze River estuary	0.142–0.716	407	(Pan et al., 2011)
30–40°		42	(UNSCEAR, 2000)

increased. An analogous trend has been observed in Cienfuegos Bay (Alonso-Hernández et al., 2005) and may reflect the cumulative development of the zone. In fact, industrial and economic expansion in Sagua la Grande appears to be continuous from 1900.

A substantial increase in mercury concentrations was observed between 10 cm depth and the surface, reaching levels of 0.3 mg kg^{-1}, indicating a considerable anthropogenic input. This important increase is clearly related to the operation of the mercury-cell chlor-alkali plant for the production of NaOH, which began operation in 1980. These values are comparable with those reported by other authors in sites where a Chlor-Alkali plant is present: 0.3–14 mg kg^{-1} in Estarreja Portugal (Pereira et al., 1998); 0.09–10 mg kg^{-1} in Cartagena Bay, Colombia (Alonso et al., 2000); and 0.1–3.22 mg kg^{-1} in Guanabara Bay, Brazil (Covelli et al., 2012). The inventory of Hg in the core A19 is 0.243 mg m^{-2}. A similar trend and inventory values (0.448 mg m^{-2}) were reported for the area by Díaz-Asencio et al. (2009).

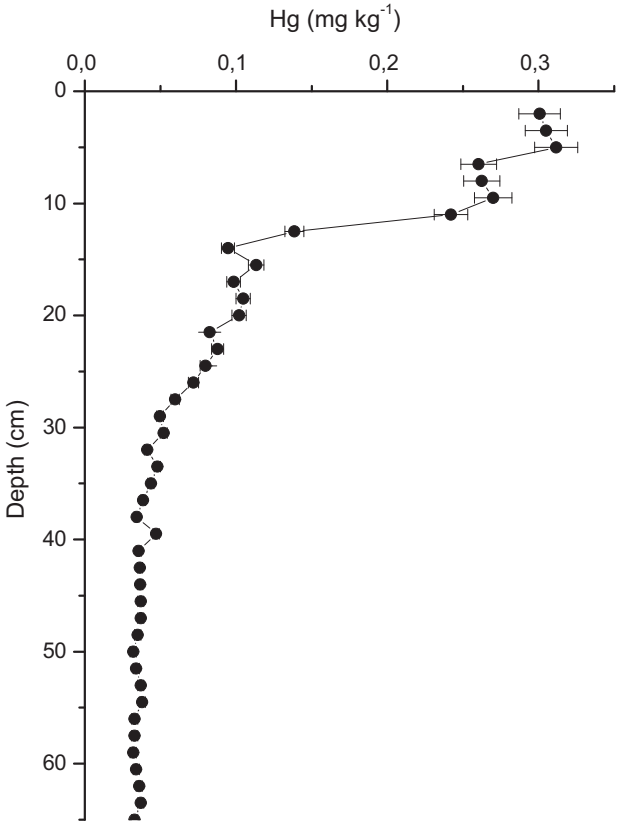

Fig. 4. Hg profile concentration normalizated to <63 grain-size fractions.

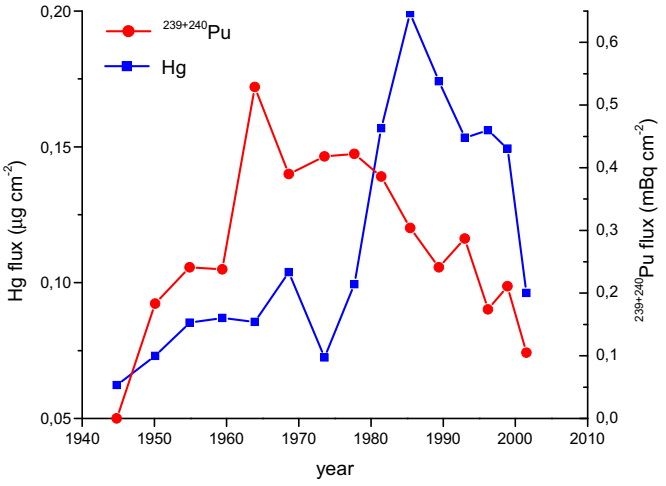

Fig. 5. Mercury and $^{239+240}$Pu fluxes in Sagua la Grande estuary.

3.2. *Hg and $^{239+240}$Pu fluxes*

Taking into account the calculated sediment accumulation rate, $^{239+240}$Pu and Hg fluxes to sediments were estimated (Fig. 5) and showed an overall behaviour similar to the one described above.

The observed $^{239+240}$Pu fluxes to sediments reflect the history of atmospheric nuclear weapons' testing in the area. In fact, the onset of $^{239+240}$Pu appeared in the early 50s with a maximum in 1963 followed by a subsequent decrease.

Furthermore, the Hg fluxes also clearly reflect the history of the chlor-alkali plant in Sagua la Grande and showed a trend similar to that reported for this region by Diaz-Asencio et al. (2009). From 1900 until the end of the decade of the 70s, a constant increment in the Hg fluxes is observed. This tendency is associated with the economic and demographic development of the area. The highest Hg flux is dated at 1990 ± 2, corresponding to the maximum production of NaOH by industry, while a reduction in Hg fluxes was observed after that date. In fact, in 1990 a waste treatment plant was put into operation in the chlor-alkali plant, thus limiting the release of mercury into the estuary and helping to recover the sediments quality (Balogh and Nollet, 2008; Cyr et al., 2002; Wan Ngah and Hanafiah, 2008). This is clearly reflected in the reduction of mercury flux to the sediments, since 1990, that reached values of 0.10 µg cm^{-2} y^{-1} in most recent years.

Both $^{239+240}$Pu and mercury can be used as markers for future studies in the Sagua la Grande estuary. The applications of $^{239+240}$Pu can be wider as this radio-tracer can be employed, as an independent marker, for dating sediments in Cuban and the wider Caribbean coastal areas.

4. Summary and conclusions

- Depth profiles of $^{239+240}$Puin sediment core samples collected at the delta of the Sagua la Grande estuary successfully identified the main atmospheric nuclear weapons tests carried out in the past.
- The inventory of $^{239+240}$Pu in the A19 core is 42 ± 5.6 Bq m^{-2}, a value close to that expected from direct global fallout.

- These results contribute to the scarce regional database for anthropogenic radionuclides in the Caribbean marine environment, particularly $^{239+240}$Pu.
- The mercury profile reflects the history of anthropogenic pollution in the estuary and describes excellently the operation of the mercury-cell chlor-alkali plant for the production of NaOH, which began operation in 1980.
- $^{239+240}$Pu and mercury are confirmed to be excellent chronomarkers for the study of sedimentation processes in the marine environment.

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