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Historical economic and environmental policies influencing trace metal inputs in Montevideo Bay, Río de la Plata

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ABSTRACT

Montevideo Bay is located in the middle zone of the Rio de la Plata, and since the foundation of the city, several key economic and environmental policies affected the industry, and thus, metal inputs into this ecosystem. The aim of this study is to evaluate the sedimentary geochemical record of Montevideo Bay, in order to determine the historical inputs of anthropogenic metals to the system. In addition, environmental and economic policies of the country were taken into account to infer the relationship between them and the historic metal input. Concentrations of aluminum, chromium, copper, lead, scandium and zinc were analyzed and the EF and SPI indices were calculated. The analysis showed that since Montevideo foundation, metal concentrations increased in accordance with industry development, and the indices as well as the metal concentration represent a reliable footprint of the history of different economic and environmental policies influencing historical industrial activities.

1. Introduction

Estuaries are semi-enclosed coastal systems connected to the ocean and influenced by tides and the exchange of oceanic and freshwater from river discharges (McLusky & Elliot, 2004). In addition, these systems are subject to constant anthropogenic pressure, with population settlements on their coasts that grow faster than the world population of other eco-regions (Borja et al., 2012; Jordan, 2012). As a consequence, anthropogenic activities such as energy production, fisheries, and tourism among others, estuaries receive river inputs, urban and industrial effluents with sediments and pollutants that commonly lead to severe environmental degradation (McLusky & Elliot, 2004; Simpson et al., 2005; Borja et al., 2012; Jordan, 2012).

Sediments are the ultimate fate of most of the contaminants, thus becoming a source of pollution to the environment (Burton, 2002; Simpson et al., 2005). In particular, trace metals are significantly more abundant in the sediments since most of them are cations, and since sedimentary organic matter and clay exhibit a net negative charge, metals are adsorbed by the sediment (Horowitz 1985). There are several sources of these contaminants, such as mining/industrial activities, sewage sludge, pesticides, smelters and leaded gasoline and paints (Luoma & Rainbow, 2008; Wuana & Okieimen, 2011). In this sense, trace metals can be used as proxies for inferring human-induced change as an indicator of biological stress related to contaminants and ecosystem health (Du Laing, 2011; Birch, 2011), In contaminated sites, the most commonly found metals are cadmium (Cd), copper (Cu), chromium (Cr), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn). All above mentioned elements accumulate in sediments, are toxic and exert a potential effect on biological processes (Wuana & Okieimen, 2011; Luoma & Rainbow, 2008).

Montevideo Bay is a coastal system affected by several activities and industrial facilities such as the ANCAP-La Teja oil refinery, Batlle-UTE thermoelectric power plant and Montevideo harbor. Its main tributaries are Pantanoso, Miguelete and Seco streams (the latter currently flowing through an underground pipe). All streams are highly impacted by domestic and industrial effluents without treatment and solid waste disposal. Several studies classified this coastal system as highly degraded, especially the inner bay area with high levels of Cr, Pb and petroleum hydrocarbons in its sediments (Danulat et al., 2002; Gautreau, 2006; Muniz et al., 2002, 2004, 2006, 2015; Venturini et al., 2015). In addition, other historical studies reported high metal concentrations in sediment cores collected at the mouth of Miguelete and Pantanoso streams (Cranston & Kurucz, 2002; García-Rodríguez et al., 2010; Burone et al., 2011).

The aim of this study is to evaluate the geochemical record of the sedimentary column of Montevideo Bay, in order to determine the historic inputs of metals in the area and assess the environmental degradation evolved from such anthropogenic impacts. Consequently, concentrations of Al, Cr, Cu, Pb, Sc and Zn were analyzed. In addition, the Enrichment Factor (EF) (Szefer et al., 1998) and Sediment Pollution Index (SPI) (Singh et al., 2002) were calculated in order to establish the relationship between the environmental and economic policies of the country and the historical metal inputs.

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2. Material and methods

2.1. Study area

Montevideo Bay (34°52′18″ - 34°55′48″ S, 56°11′48″ - 56°14′42″ W) is located on the north coast of the Río de la Plata which is contained into the second largest river basin of South America (Fig. 1).

The first Spanish navigators considered Montevideo Bay as the natural harbor of the "New Continent". Despite this, due to the lack of metal wealth, the condition of city-harbor was explicitly denied when Montevideo was founded (between 1724 and 1726). After 1778, when free-trade was first decreed Montevideo harbor was recognized as such, and import/export of goods was only then allowed, although it had already been used by locals to market leather products clandestinely (Baracchini & Altezor, 2010). Thereafter, several key-events affected the dynamics and environmental conditions of Montevideo Bay (Table 1).

2.2. Collection of samples

In May 2010, a 149 cm long core (BAT 1) was collected with a 63 mm. internal diameter piston in the inner area of Montevideo Bay (near the Batlle thermoelectric power facility and Montevideo harbor) (Fig. 1). The core was longitudinally sectioned every 1 cm. and samples were preserved in the Oceanography and Marine Ecology Lab of Facultad de Ciencias.

2.3. Sedimentation rate

The sedimentation rate was determined based on unsupported ^{210}Pb activities using the CIC (Constant Initial Concentration) model (Appleby & Oldfield, 1978; Joshi and Shukla, 1991), and validated with ^{137}Cs data. In this sense, approximately 10 g of sediment were transferred into air-sealed cylindrical polyethylene containers for gamma counting in an EG&G ORTEC® low-background gamma spectrometer (hyperpure Ge, model GMX25190P).

The sedimentation rate was calculated using the following formula:

$$S = (\lambda \cdot D)/\mathrm{Ln}\left[\left(C_0{}^{210}\mathrm{Pb}\right)/\left(C^{210}\mathrm{Pb}\right)\right]$$

where S: sedimentation rate in cm·yr^{-1}; λ: radioactive decay constant of ^{210}Pb (0.31076); D: distance between the core-top and the measured interval (cm); $C_0^{210}Pb$: count of unsupported ^{210}Pb at the core-top; $C^{210}Pb$: unsupported ^{210}Pb at the measured stratum.

Table 1
Chronology of events that influenced the dynamics and environmental conditions of Montevideo Bay.
Sources: Nahum, 1999; Baracchini & Altezor (2010), Burone et al. (2011), Muniz et al., 2011.

1781	First dock construction in Montevideo Bay
1834–1860	Proposals for managing industrial settlement in Montevideo (harmful to public health)
1839–1851	Uruguayan civil war ("Guerra Grande"). The port infrastructure built in Montevideo Bay surpasses all the work done in the area since the city foundation.
1870–1914	Beginning of the second industrial revolution. Changes in energy sources (replacement of steam for electricity and petroleum products), and new materials with the use of different metals
1901–1909	Montevideo harbor construction
1917–1930	Industrial exchange. Salting industry is replaced by the meat-packing industry and leather tanneries emerge in Miguelete and Pantanoso basins.
1930–1936	Construction of La Teja oil refinery and thermoelectric power station "José Batlle y Ordóñez". Both industries located on Montevideo Bay margin.
1947–1953	*Neobatllismo*. Boom of the ISI model (Import Substitution Industrialization). State lead economic development through nationalization, subsidization for national firms, increased taxation, and highly protectionist trade policies. Little international trade.
1973	End of ISI model. Beginning of Uruguayan dictatorship.
1980	Controls for the industrial waste disposal to the environment
2000	Important decrease in the leather industry since 2000
2004	Law 17.775 – gasoline and paint lead free.

The quality control of the method was evaluated through the determination of the radionuclides of interest (^{210}Pb, ^{226}Ra and ^{137}Cs) in three certified reference materials: IAEA-326 (soil), IAEA-327 (soil) and IAEA-385 (marine sediment).

2.4. Metal determination

Analyses were performed at the Marine Inorganic Chemistry Laboratory of the Oceanographic Institute of São Paulo University (IO-USP). The concentration of Al, Cr, Cu, Pb, Sc and Zn was determined using the partial digestion method USEPA 3050B (USEPA 1996). The method consists of a strong acid digestion with HNO_3 at high temperature and the addition of H_20_2 for the organic matter elimination. Then, the solution was filtered and diluted to 50 ml to be analyzed with a Varian Vista MPXICP-OES. For the analysis of Al, samples were diluted 1 to 100. Method accuracy was determined by analyzing the certificated reference material SS-2 EnviroMAT. Values obtained for the reference

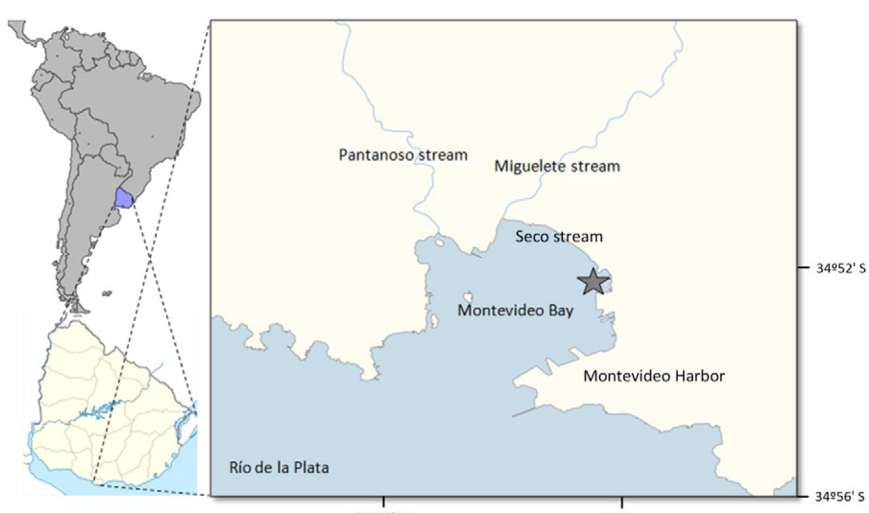

Fig. 1. Study area. The star indicates coring site in Montevideo Bay.

material were always satisfactory and lied within the range recommended by USEPA (1996) (Table 2).

2.5. Indices calculation

To establish the degree of sediment contamination throughout the core, both the Enrichment Factor (EF) (Szefer et al., 1998) and Sediment Pollution Index SPI (Singh et al., 2002) were calculated.

The EF provides information about sediment contamination for a specific element, and it was calculated following Szefer et al. (1998):

$$EF = \frac{\left[\frac{C_X}{C_N}\right]_{stratum}}{\left[\frac{C_X}{C_N}\right]_{base.}}$$

where C_X: X element concentration, C_N: normalizing element concentration

We used Sc as normalizing element, since it is an important constituent of fine grain-size fraction. It does reflect grain-size variability in the sediments and it is rarely added by anthropogenic sources (Loring, 1988; Loring & Rantala, 1992). In addition, background values correspond to the average element concentration of the three basal samples of the sediment core.

To assess levels of contamination considering the EF, we used the classification proposed by Sutherland (2000) (Table 3).

The SPI corresponds to a linear sum of the enrichment factor considering the relative toxicity of each metal, by assigning a differential weight; i.e., Cr and Zn (less toxic metals) = 1, Ni and Cu = 2, Pb = 5, and Cd (most toxic metal) = 300. In this paper we considered Cr, Cu, Pb and Zn using the formula proposed by Singh et al. (2002):

$$SPI = \frac{\sum (EF_m * W_m)}{\sum W_m}$$

where: EF: enrichment factor of metal m in the sample, W: toxicity weight of metal m

SPI is classified in five different classes according to the contamination level (Singh et al., 2002) (Table 4).

2.6. Statistical analysis

Normality of data was performed by running Shapiro-Wilk test. Significant differences for metal concentrations among the different lithological units were tested using the U-Mann-Whitney non-parametric test (Sokal & Rohlf, 2012), with a significance level of 0.05.

3. Results

According to CIC model (Fig. 2), the average sedimentation rate was estimated to be 0.53 ± 0.06 cm·yr^{-1}. Therefore, sediment core BAT1 encompasses the last 280 yr of Montevideo Bay's history.

According to the color and texture of the sediment, three lithological units were identified (Fig. 3). Unit I, from the base of the core to 40 cm. depth, with a dominance of gray silt-clay sediments. The layer 40–6 cm. (unit II) was dominated by darker gray silt sediment, and in unit III, from 6 cm. to the surface, a black mud was observed (Fig. 3).

The changes observed in color and texture between the lithological units I and II match with a significant increase in metal concentration ($p < 0.05$) for all analyzed metals. Moreover, in unit II maximum values of all metals were observed (Cr 280.52 mg·kg^{-1}, Cu 257.82 mg·kg^{-1}, Pb 157.55 mg·kg^{-1} and Zn 477.77 mg·kg^{-1}) at 24 cm depth which corresponds to 1967 CE (Fig. 3).

Table 2
Assigned and obtained values for the certified reference material SS-2 EnviroMAT.

	Reference (mg·kg^{-1})	Confidence (95%) (mg·kg^{-1})		Tolerance (mg·kg^{-1})		Obtained (mg·kg^{-1})
Al	13,265	12,114	14,416	9948	16,581	11,692
Cr	34	30	38	14	54	30
Cu	191	182	200	139	243	163
Pb	126	116	136	68	184	109
Sc	–	–	–	–	–	–
Zn	467	444	490	337	597	462

Table 3
Classification of EF proposed by Sutherland (2000) and used in the present study.

EF value	Classification
EF < 2	Minimal enrichment. Suggests null or minimal contamination.
2 < EF < 5	Moderate enrichment.
5 < EF < 20	Significant enrichment.
20 < EF < 40	Very high enrichment, indicting high level of contamination
EF > 40	Extremely high enrichment, indicating extreme contamination.

3.1. Indices

The maximum value of EF was registered for Cr and Cu at 24 cm. depth, which corresponds to 1967 CE (Cr 13.82 and Cu 13.07), and the maximum EF for Pb and Zn (Pb 57.94 and Zn 9.54) was observed at 20 cm depth which corresponds to 1974 CE (Fig. 4).

Considering the classification proposed by Sutherland (2000), an initial moderate sediment enrichment was observed for Cu, Cr and Zn between the 1930s and 1950s (2 < EF < 5), and after the 1950s a significant enrichment was recorded (5 < EF < 20) (Fig. 4). However, Pb EF showed a different trend. A moderate enrichment was observed from the late 18th Century to the late 19th Century, but in the early 20th Century a significant enrichment was measured. In early 1950s an increase in the EF was recorded, with values indicating high and extreme pollution levels (20 < EF < 40 and EF > 40), but then returning again to values of significant enrichment in the surface of the sediment core (Fig. 4).

Considering the SPI values, the results are essentially controlled by the Pb EF, due to the toxicity of this element for biota and human health compared to the other analyzed metals. The results of the SPI classification according to Singh et al. (2002) are shown in Table 5.

4. Discussion

Since the foundation of Montevideo specific historical well documented events and different economic policies were the driving forces on the urban and industrial development, thus influencing the inputs of metals into Montevideo Bay (Fig. 5).

Considering Montevideo's historical industry development, the enrichment of Pb (metal associated specifically with batteries, gasoline additives, paint, inks and pigments for leather and textiles (Sutherland, 2000; Wuana & Okieimen, 2011) showed an initial increase after 1760 CE (EF 3.82). Even though since 1778 Montevideo was only allowed to free trading, the harbor had previously been used by locals to market their industrial production (Baracchini & Altezor, 2010) thus explaining the first increase in Pb concentrations by local industry goods.

Table 4
SPI classes established by Singh et al. (2002) and used in the present study.

Class	SPI	Classification
SPI0	0–2	Natural sediments
SPI1	2–5	Low polluted sediments
SPI2	5–10	Moderately polluted sediments
SPI3	10–20	Highly polluted sediments
SPI4	>20	Dangerous sediments

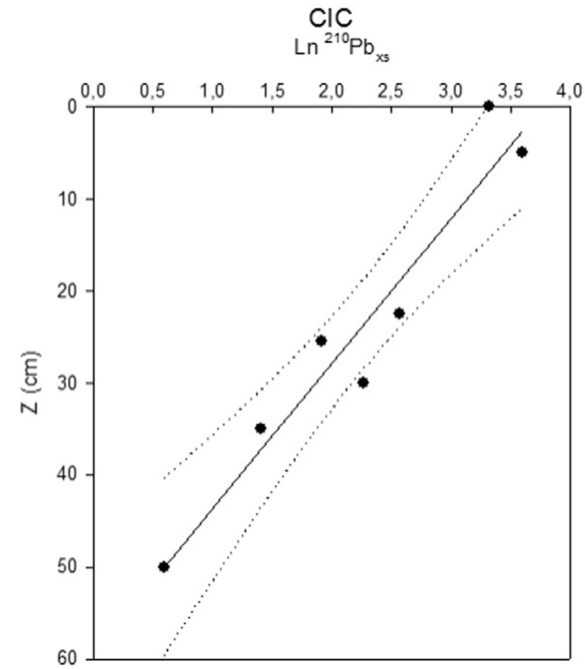

Fig. 2. Plot of Ln$^{210}Pb_{xs}$ activity vs. depth for BAT1 core. Solid line represents the regression line and dotted lines define the 95% confidence band.

The further increase in the Pb enrichment (EF 8.09) observed in the late 1880s. could be related to the second industrial revolution (1870–1914) that led to major technological innovations, with changes in the sources of energy (i.e., replacement of steam for electricity and petroleum). In addition, during the second industrial revolution the utilization of new materials using different metals, as well as more modern transport systems such as cars was documented by Bilbao & Lanza (2009). In 1909 the inauguration of Montevideo harbor was commemorated, which represented a major driving force for the development of the Uruguayan economy. However, due to high taxes and costs it did not fulfill functions as a passenger and cargo terminal until mid 1920s (Baracchini & Altezor, 2010), where a further increment of the SPI was observed (Fig. 5).

Between 1930 and 1959 Uruguay exhibited a period of strong economic growth, mainly driven by the development of the national industry. During this period, the Government promoted an economic model known as Industrialization Substitution Import (ISI) (1930–1973) to stimulate industrialized products with higher added value (eg.: fabrics instead of raw wool) (Nahum, 1999). The footprints of this industrial boom are expressed in core BAT1, as a further increase in the enrichment of Pb (EF 39.33), Cu (EF 7.35), Cr (12.17) and Zn (EF 9 31). All above mentioned metals are associated to the industrial activity; i.e., Cr: dyes, leather tanning, pesticides, textile processing, wood processing; Cu: fertilizers, electrical equipment, pesticides, pigments, paints, mining, wood treatment; Zn: fertilizers, electroplating, pesticides, paints, mining, inks, wood processing (Wuana & Okieimen, 2011; Sutherland, 2000).

While the decade of 1960 was characterized by a period of general stagnation of the Uruguayan economy, a continuous increase in the analyzed metals is observed until the early 1970s. This increase could be associated to changes in the traditional industries, where the activities of the most industrialized companies were encouraged. The consequence of this event led to the closure of large facilities and the opening of smaller factories (Camou & Maubrigades, 2006). On the other hand, non-traditional export industries responded favorably to the policies developed by the Government, and therefore, increases in food, drinks, tobacco and electric appliances were observed. In addition, paper, printing industries, rubber, petroleum and metallurgical industries also increased but to a lesser extent (BCU, 1975).

In the late 1960s, new laws and regulations about waste disposal to the environment (1968: Ordinance No. 13,982 industrial wastewater, 1979: Water Code Decree 253/79) were implemented. By 2001 the decrease in the number of industries caused a reduction in the input of pollutants to the watercourses (García-Rodríguez et al., 2010). Some companies implemented the environmental management system ISO 14000, thus improving their processes and control of their emissions

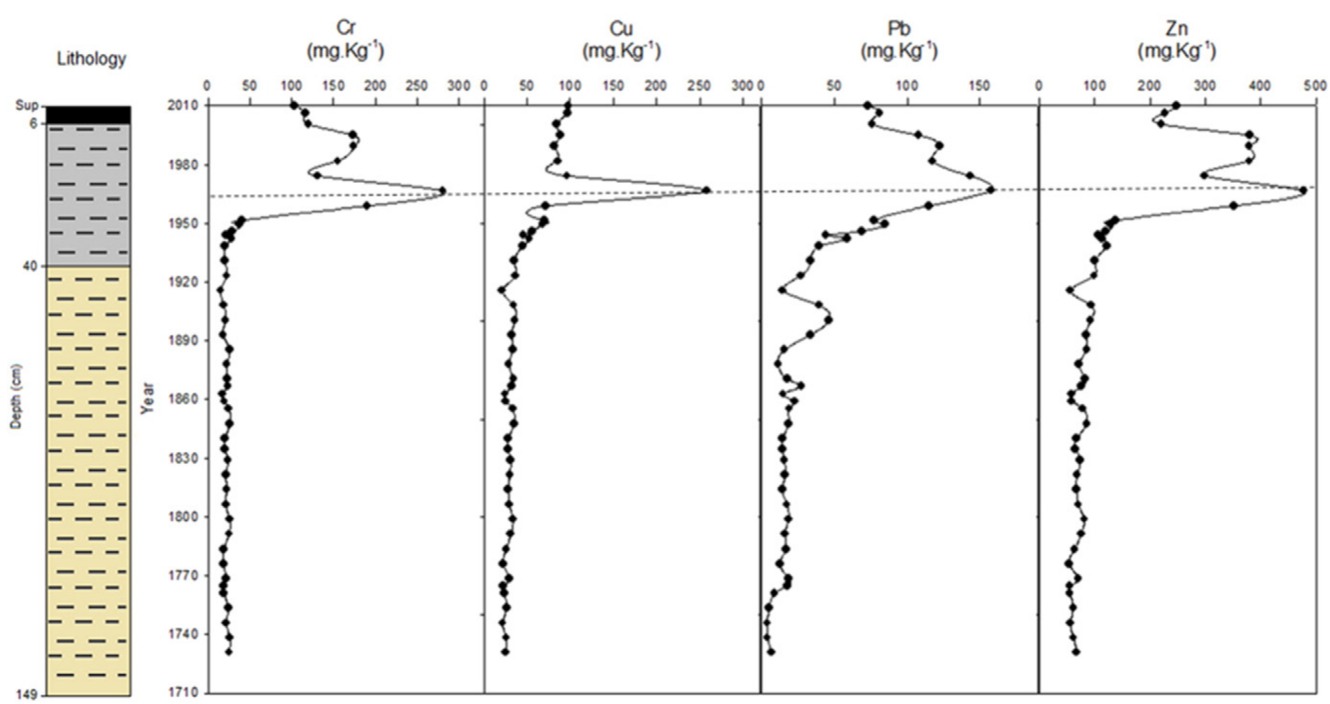

Fig. 3. Lithology and concentrations of Cr, Cu, Pb y Zn (mg·kg^{-1}) in sediment core BAT1. With a dotted line highlighting the 24th stratum, where the maximum metal concentrations are observed.

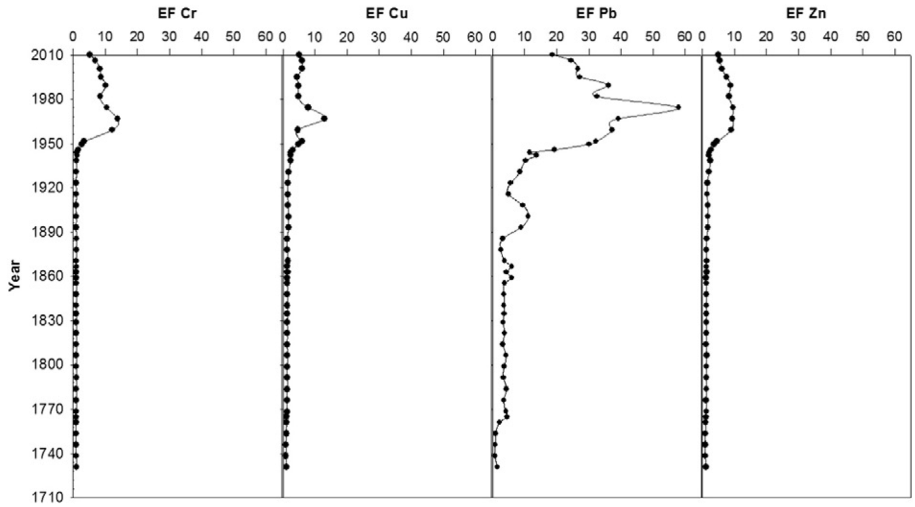

Fig. 4. Enrichment Factor (EF) for Cr, Cu, Pb and Zn.

and disposals into the environment (IMM 2001). For the period 2002–2008, further reductions were observed for the values measured in the effluents. Such a reduction is even more remarkable considering the fact that, in the same period, there was a significant increase in the number of active industries thus leading to a recovery of the industrial activity after the 2002 economic crisis (IMM, 2008). These results are mainly due to the Plan for the Reduction of Industrial Pollution proposed by the Municipality of Montevideo in 1997. In 1999, results of this Plan indicated a reduction of metal inputs and recovery of water quality in the Miguelete and Pantanoso streams, which are the main tributaries of Montevideo Bay (IMM, 2008). The observed decrease in the concentration of metals for the surface layers are in agreement with Muniz et al., (2015) and García-Rodríguez et al., (2010), because of the relocation of the leather tanneries and the use of Pb-free fuel.

Other studies, Burone et al., (2011) and García-Rodríguez et al., (2010) observed different metal content in sediment cores collected at the mouth of the Pantanoso stream (western internal zone of Montevideo Bay). Such studies, reported the highest Cr concentrations with maximum values close to 600 $mg \cdot kg^{-1}$ and an EF close to 20, both values are higher than the maximum registered in this study. However, for Cu, Zn and Pb concentrations we recorded higher EF values in the present study. Lower values of Cr registered in this study might be associated to the location of the coring station. Historical Cr inputs into Montevideo Bay came off mainly through Pantanoso stream due to the higher number of tannery facilities (Muniz et al., 2002), and core BAT1 was extracted near the mouth of Miguelete stream where no tannery facilities were constructed. In addition, the circulation of Montevideo Bay which influences sediment transportation, which is predominantly clockwise (Santoro et al., 2013), could drag the contributions away from the Miguelete stream, and to a lesser extent Pantanoso stream.

This relation between the economic policies and the metal input to Montevideo Bay as a final consequence can be analyzed considering the Driving Forces/Pressures/States/Impacts/Responses (DPSIR) Framework approach. This framework considers that social and economic developments are the driving forces (D) that exert pressure (P) on the environment, and consequently, does change the environmental quality. Finally, this leads to impacts (I) on human health/ecosystems that cause a response (R) from society to mitigate or reverse unacceptable conditions (Smeets & Weterings, 1999; Borja & Dauer, 2008). Therefore, this concept as an environmental indicator or proxy for the pressures or the evolution of the environment, play a key role for environmental management (Niemeijer & de Groot, 2008).

From this point of view, economic policies and social development are the driving forces that exert pressure on the environment of Montevideo Bay, leading to changes in environmental quality. In this sense, the SPI and the EF are consistent environmental indicators of the pressure and reliably reflect the state of the environment. In addition, environmental policies are the response of the society to mitigate unacceptable environmental conditions.

Table 5
Sediment classification for BAT1 core considering the SPI classes established by Singh et al. (2002).

Class	Classification	Years
SPI0	Natural	Until the second half of the XVIII century
SPI1	Slightly contaminated	Second half of the XVIII century until the end of XIX century
SPI2	Moderately contaminated	End of XIX century until the mid – twentieth century
SPI3	High contaminated	1945–1950
SPI4	Dangerous	50s until the late 80s
SPI3	High contaminated	Last 20 years

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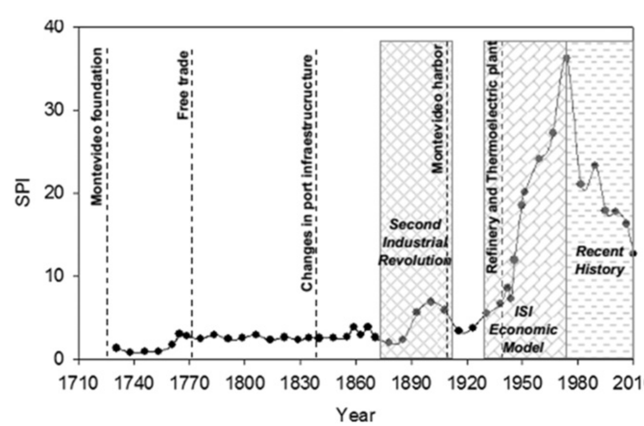

Fig. 5. Results of SPI for sediment core BAT1, highlighting events that influenced the dynamics and environmental conditions of Montevideo Bay.

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