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Assessing the effect of natural variability and human impacts on the environmental quality of a coastal metropolitan area (Montevideo Bay, Uruguay)

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Abstract. Montevideo Bay is an extremely polluted coastal system because of multiple sources of contamination from industrial manufacturing, urban activities, energy production, harbor operations and population increase. Because rehabilitation measures will be implemented, this research project was undertaken to assess the contamination of the system. A sediment core that spanned to 3000 yr BP was taken to reconstruct the Holocene natural changes in salinity and trophic state, and also the effect of human activities on the environmental quality of the system. Before the human impact, the system was controlled by a sea level decrease, during which an increase in trophic state was observed. After the human impact, an intensification of the eutrophication process was observed, mainly due to the industrial activities linked to lack of sanitation. In addition, sharp increases in heavy metal concentration were recorded after 1917, when the leather tanning activities together with the construction and operation of the oil refinery and the thermoelectric generation plant were first documented. Thus, the data presented in this paper provide important chronological evidence of human impacts and environmental degradation, which at the minimum can be used to advise the managers and government agencies about the consequences of bad environmental practices on coastal aquatic systems.

Key words: heavy metal contamination, diatoms, eutrophication, coastal paloceanography

Resumen. Evaluación del efecto de la variabilidad natural e impactos humanos en la calidad ambiental de un área costera metropolitana (Bahía de Montevideo, Uruguay). La Bahía de Montevideo es un sistema extremadamente degradado como consecuencia de múltiples fuentes de contaminación de la industria manufacturera, actividades urbanas, generación de energía, operaciones portuarias e incremento poblacional. Ya que se implementarán medidas de rehabilitación, llevamos a cabo este trabajo de investigación para evaluar el estado de contaminación de la Bahía. Se tomó un testigo de sedimento y se reconstruyeron los cambios naturales de salinidad y estado trófico durante parte del Holoceno. También se infirió el efecto de las actividades humanas sobre la calidad ambiental del sistema. Antes del impacto humano el sistema estaba controlado por un descenso del nivel del mar. Luego del impacto humano se observó una intensificación del proceso de eutrofización principalmente por las actividades de la industria manufacturera junto a la ausencia de saneamiento. Marcados incrementos de la concentración de metales pesados fueron observados a partir de 1917, cuando se documentaron las primeras actividades de curtido de cuero, operación de la refinería y generación de energía termoeléctrica. En este sentido, los datos aquí presentados son una evidencia cronológica del efecto de los impactos humanos sobre la calidad ambiental, la cual al menos puede ser utilizada para aconsejar a los gestores ambientales y empleados gubernamentales acerca de las consecuencias de malas prácticas de manejo y desarrollo sobre los sistemas acuáticos costeros.

Palabras clave: contaminación, metales pesados, diatomeas, eutrofización, Paleoceanografía costera

Introduction

Monitoring information on the effect of human impacts on coastal systems is essential in view of sustainable management practices (Sanchez-Cabeza & Druffel 2009). Such information should include direct observations, which encompass time-scales of years and decades, but also long-term proxy data from paleoenvironmental reconstructions. Especially in those regions that have experienced sharp historical population increases, an understanding of historic conditions and relationships among stressors and ecosystem responses is necessary.

In the case of Montevideo Bay, an extremely degraded system because of industrial, economic and harbor activities, reliable direct data on the degree of contamination are only available since 1993 (Moyano *et al.* 1993). Later on, Danulat *et al.* (2002), Muniz *et al.* (2002, 2004a, b), Venturini *et al.* (2004), Burone *et al.* (2006), Brugnoli *et al.* (2007), undertook further assessments and also concluded that the bay is extremely degraded. Long-term data were first generated by Cranston & Kurucz (2002) who took cores in Montevideo Bay. They calculated sedimentation rates of 3 and 2 mm yr^{-1} in the inner and outer bay respectively, and detected an increase in zinc and chromium values in the last 150 yr. In addition, very elevated concentrations of these heavy metals were only observed within the top 20 cm of the sediment, representing the last 70 years, when industrial activities were intensified. However, they only briefly attributed contamination to the leather tanning industry, but failed to identify other sources of contamination and the relations between such sources and the several historical industrial activities, population growth and harbor development.

In this paper, a new sediment core was taken to analyze chromium, zinc, lead, copper, organic matter content and diatoms as proxies. An historical population increase was documented together with the major milestones in industrial activities and harbor development, and their relation to historical contamination trends. In addition, diatom and organic matter data were used to infer the Holocene changes in both trophic state and paleosalinity that were ultimately related to sea level variation for the past 3000 yr. The data presented here provide a good example of the increasing importance of using paleoenvironmental techniques in contamination assessments to improve our skills at environmental management.

Material and methods

Description of the study area

The Montevideo coastal zone (34°50'-34°56' S and 56°05'-56°25'W) is located in the middle of the coast of Río de la Plata, which is contained in the second largest basin in South America. The Río de la Plata is a coastal plain tidal river with a semi-enclosed shelf sea at the mouth.

Montevideo Bay is approximately 10 km^2 (Fig 1) with a mean depth of 5 m. Water circulation is mainly clock-wise (Moresco & Dol 1996). The bay contains one oil refinery, one thermoelectric facility, the harbor, and the city itself. Only part of the city of Montevideo has a sanitation system (Fig. 1). Three streams flow into the bay, Miguelete, Pantanoso and Seco; the latter entering through an artificial pipe-line. These streams carry wastes from many different industries, urban centers and from a great number of clandestine sewage pipes, where there is no sanitation system.

To the east of the Montevideo coastal zone, in the region of punta Carretas (Fig. 1), lies the most important sewage pipe of Uruguay. The authorities of Montevideo are planning the construction of a similar sewage pipe in the Punta Yeguas zone (Fig. 1) that will concentrate the sewage of both Pantanoso and Miguelete streams, which are at present discharging directly into Montevideo Bay.

The modern sediments are mostly clay and silt (Urien *et al.*, 1980). According to Muniz *et al.* (2002) the inner part of Montevideo Bay has greater sediment heterogeneity, higher organic load and lower oxygen content in bottom sediments than those of the outermost part of the bay and the adjacent coastal zone (*i.e.*, Punta Carretas and Punta Yeguas). This inner part of Montevideo Bay is grossly polluted by Cr, Pb and petroleum hydrocarbons, while the other zones show moderate pollution. Danulat *et al.* (2002) defined the Montevideo Harbor as a hyper-eutrophic system, which receives considerable nutrients and organic loads. Gómez-Erache *et al.* (2001) reported that water quality of Montevideo Bay is highly deteriorated because of several point and non-point sources, whereas harbor activities introduce great amounts of heavy metals and nutrients leading to a high biological oxygen demand.

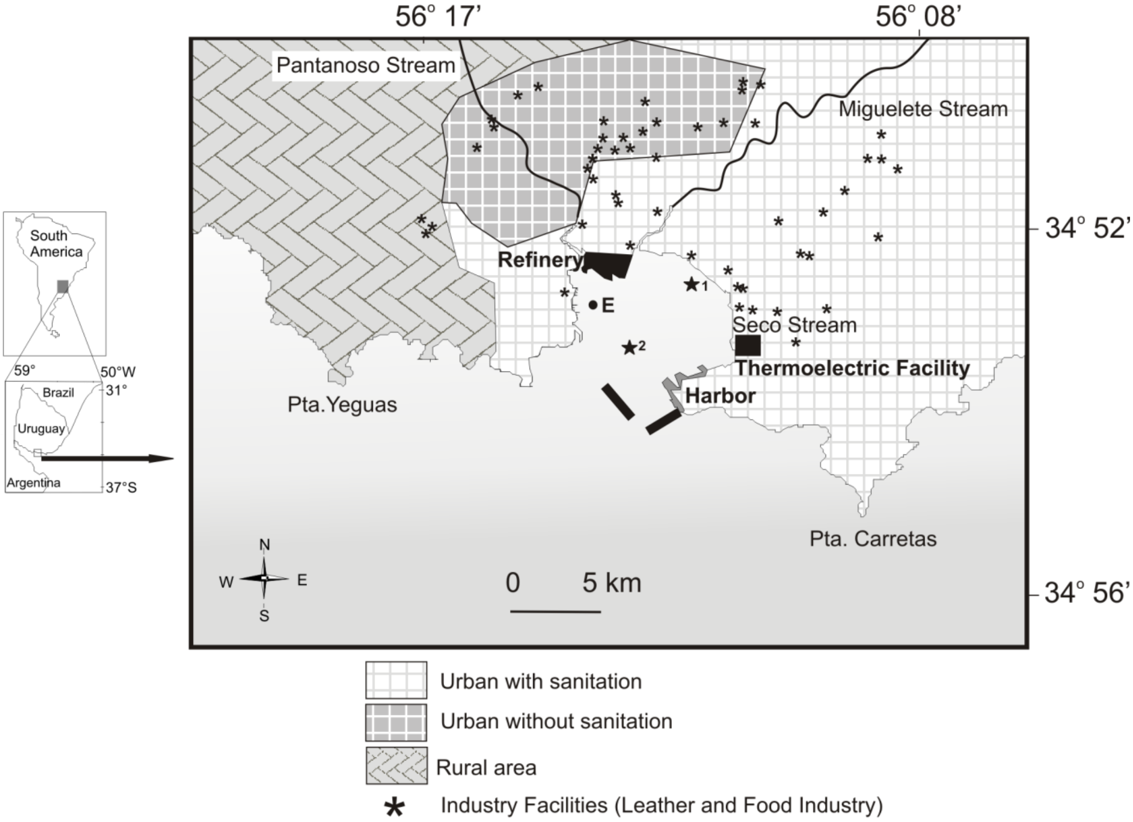

Figure 1. The study area. Solid dot indicates coring station E. Stars indicate coring stations of Cranston & Kurucz (2002).

On the history of the study area

Montevideo was founded in 1725 by Spanish settlers for war purposes against Portugal. The city was built on the bay (Fig. 1). The first dock in Montevideo Bay was built in 1796 (Gautreau 2006), but only in 1901-1909 was the modern harbor developed. During this stage, a population increase from ca. 50,000 to 300,000 inhabitants was documented (Fig. 2). After the harbor was built, there was a sharp development of both leather and food industry facilities between 1917 and 1930. Between 1923 and 1939 the coastal road was placed. Concomitantly, in 1930-1937 the oil refinery and the thermoelectric facility were also built on the coast of the bay. Because of the urban development, in 1963 population had increased to 1.2 million (Fig. 2). After 1977, the harbor has been progressively expanded and enlarged as several docks, container places and related facilities and logistics were erected.

Field and laboratory methods

A 200-cm-long core (Core E1, 34° 53' 00''S, 56° 14' 23''E) was taken with a 63 mm internal diameter piston corer in the inner zone of Montevideo Bay (Fig. 1). This region of the bay is relatively undisturbed because no dredging activities to allow navigation are undertaken. After retrieval, the core was immediately sealed and brought to the laboratory for lithological, chemical and biological analyses. The lithological subdivisions were based on the visual and textural appearance of the sediment.

The core was dated following the conventional ^{14}C method on entire/articulated shells of *Erodona mactroides* Daudin 1801. The sample was treated with dilute HCl to remove carbonates. Bulk organic material was converted to benzene and its ^{14}C activity was measured with a Packard Tri-Carb 2560 TR/XL liquid scintillation spectrometer. Age is expressed in uncalibrated conventional ^{14}C yr BP, corrected for isotopic fractionation by normalizing $\delta^{13}C$ values to –25 ‰. Quoted error ($\pm 1\sigma$) includes uncertainties in counting statistics. The radiocarbon date was calibrated using the computer program Radiocarbon Calibration Program Rev. 3.0, University of Washington, Quaternary Isotope Laboratory (Stuiver & Reimer, 1993). Bracco *et al.* (1999) estimated that the error caused by the reservoir effect for estimations on coastal biogenic material, is small in our study area. They concluded that the magnitude of the error caused by the reservoir effect is not higher than the inherent error of the dating technique. In addition, Angulo *et al.* (2005) estimated a low regional marine reservoir correction of 33 ± 24 ^{14}C yr for southern Brazil (states of Santa Catarina and Paraná) and an estimate of 8 ± 17 ^{14}C yr for the region between Rio de Janeiro and Santa Catarina.

Samples for diatom counting and identification were treated with 2N $Na_4P_2O_7$ to deflocculate the sediment and remove clay. Then, 15 ml of 35 % HCl were added and allowed to stand for 24 hours to eliminate carbonates followed by rinsing five times with distilled water. Next, 10 ml of 30 % H_2O_2 were added to eliminate organic matter, and then the samples were boiled for four hours and rinsed five times with distilled water. Permanent slides were mounted in Naphrax® for counting and identification. A minimum of 300 valves was counted at 1000 x magnification in each sample. Species were identified according to Frenguelli (1941, 1945), Metzeltin & García-Rodríguez (2003), Metzeltin *et al.* (2005), Müller-Melchers (1953, 1959), Witkowski *et al.* (2000). Chrysophyte cysts were also counted and the results expressed as the percentage ratio of their abundance to the total number of diatoms valves and chrysophyte cysts (Smol 1985). In addition a dictyochophycean species was also counted and identified.

Samples of ~1.5-2.0 g were taken every 1 cm for organic matter determination by weight loss on ignition at 550°C for 4 hours following Heiri *et al.* (2001).

Trace metal (Cr, Zn, Pb and Cu) samples were also analyzed every 1 cm within the upper 40 cm; for deeper sections only selected intervals were measured. The samples were dried at 85 °C to constant weight prior to homogenization in an agate mortar and pestle. In order to avoid interference of organic matter in the measurements and to convert the metals to their free form by method No. 3051 of the EPA (Anonymous, 1990), duplicates of subsamples (0.5–1 g) were mixed with 10 ml concentrated nitric acid and digested by microwave (CEM, MDS 2100) in a closed fluorocarbon vessel. Quantification was by ASS (Shimadzu AA-680) with graphite furnace atomization (Shimadzu GFA-4B). Quality control included procedural blanks, measurement of standards obtained from the National Institute of Standards and Technology, and spiked samples. Results presented in g^{-1} dry sediment for Zn, Cu, Cr and Pb correspond to mean values of duplicate analyses.

Cluster analyses were performed to identify Diatom Association Zones (DAZ) using the program PAST version 1.81, which is available on the internet for free (http://folk.uio.no/ohammer/past). DAZ were determined by cluster analysis using the Morisita method as advised by Hammer *et al.* (2008) for analyzing similarity of relative abundance species data.

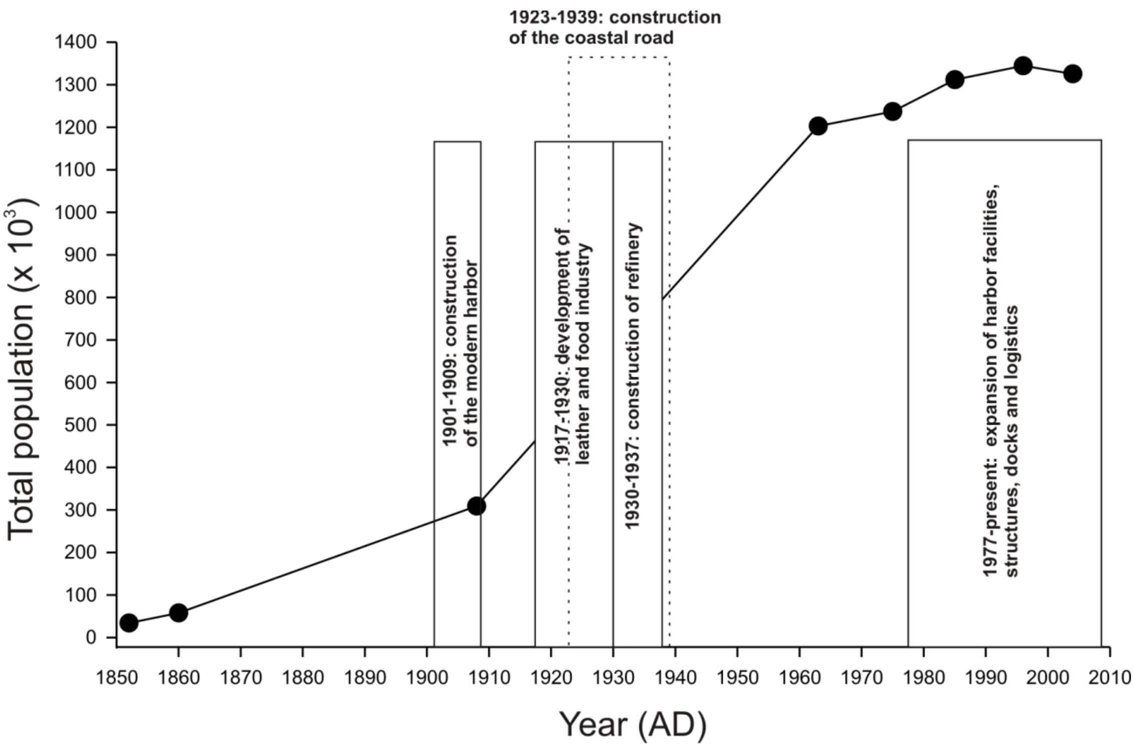

Figure 2. Population increase and chronology of most important human impacts in the study area. Total population data obtained from the National Institute of Statistics.

Results

Lithology and sediment age

Eight lithological units were identified according to changes in sediment color, sediment texture and conservation of biogenic material (Fig. 3). The basal unit of core E1 consisted of sandy sediments and exhibited entire/articulated biogenic remains of *Erodona mactroides* and *Heleobia australis* (D'orbigny, 1835).The section 200-195 cm (lab number URU 0507) was dated at 2900 ± 100 yr BP (Fig. 3). The conventional age was also calibrated for the 2 sigma range and yielded an age between 1386–826 BC. Unit VII was dominated by silty grey sediments with broken shells of *E. mactroides*. However, two black layers were observed at 163 and 154 cm depth. Units VI and IV were dominated by grey sandy sediments. Unit V and III were similar to unit VII in both sediment color and composition; however, at 77 cm depth a black layer was detected. Unit II consisted of sandy sediments with entire shells of *E. mactroides*, while unit I consisted of silty grey sediments with entire shells of both *E. mactroides* and *Tagelus plebeius* (Lightfoot, 1786).

Organic matter and heavy metals

The lowest organic matter values of the core (Fig. 3) were observed in those lithological units dominated by sandy sediments (*i.e.*, units VIII, VI, IV and II). Organic matter values were equal to or lower than 2%. Those lithological units dominated by silty sediments, exhibited higher values (usually between 4 and 8%).

Heavy metals showed a trend similar to that observed for organic matter (Fig. 3), as the lowest concentrations were also recorded in those lithological units dominated by sandy sediments (i.e. VIII, VI, IV and II). From 37 cm to the sediment surface, all heavy metals exhibited a sharp and steady increase in concentration, and the maximum values were observed 5 cm below sediment surface. The relative abundance of the heavy metals was Cr > Zn > Cu > Pb.

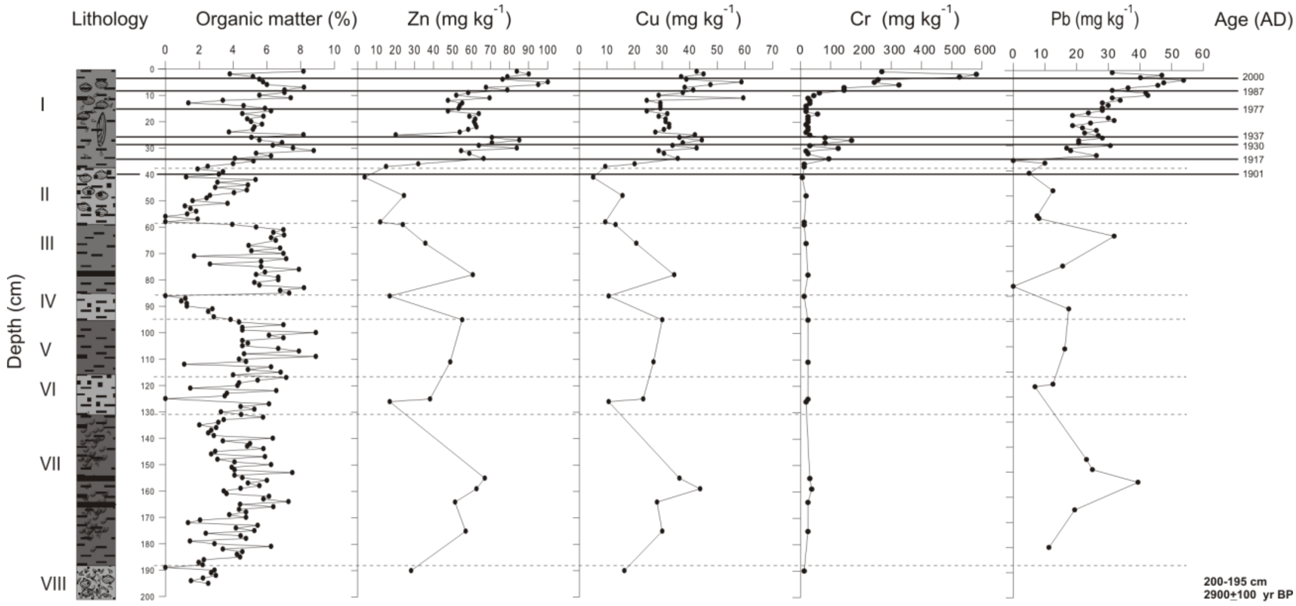

Figure 3. Lithology and vertical distribution of organic matter, zinc, copper, chromium and lead of core E1. Sediment age is plotted to the right of the plot.

Diatoms, dictyochophyceans and chrysophyte cysts

Ninety seven diatom species were identified. The most common taxa (*i.e.* > 2% in at least three sediment intervals, Kart & Smol 2000) are shown in Figure 4. Cluster analysis allowed us to identify six DAZ. Co-dominance of freshwater and marine/brackish species was observed. The most common freshwater taxa were *Aulacoseira granulate* (Ehrenberg) Simonsen , *A. granulata var. angustissima* (O. Müller) Simonsen , *A. italica* (Ehrenberg) Simonsen and *A. muzzanensis* (Meister) Krammer (Fig. 4). These species together accounted for minimum abundances of 10% and maxima of 60%. The most abundant marine brackish species were *Coscinodiscus radiatus*, *Hyalodiscus subtilis* and *Paralia sulcata* (Ehrenberg) Cleve (var. *coronata and radiata)*, although in DAZ II *Pseudopodosira echinus* Frenguelli (Metzeltin, Lange-Bertalot, García-Rodríguez) showed abundance values close to 20%. Cyst to diatom ratios showed the highest values in those DAZ dominated by freshwater taxa. Minimum values were detected where marine/brackish taxa were most abundant. In these sections, *Dictyocha fibula* Ehrenberg showed the highest frequencies. However, in DAZ V freshwater and marine/brackish taxa exhibited similar abundances (*i.e.* they accounted together for ca. 40%), and high cyst to diatom ratios were registered. The upper section of the core was characterized by the increase in *Biddulphia* sp., *Actinocyclus curvatulus* Janisch and *A. gallicus* Meister, although the most common historical freshwater and marine/brackish species still persisted.

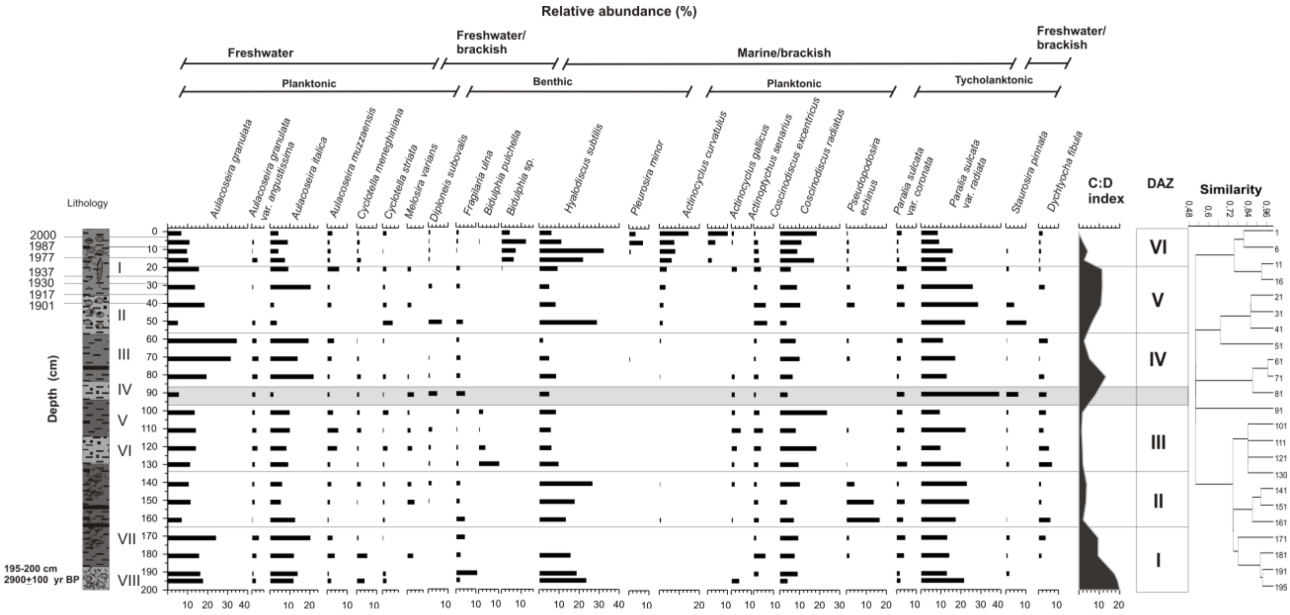

Figure 4. Vertical distribution of most abundant diatom species. DAZ = Diatom Association Zones. Cluster diagram following the method of Morisita is depicted to the right of the plot. Sediment age is plotted to the left of the plot. C : D index is also shown in the vertical profile.

Discussion

According to the regional models of relative sea level change, after 3000 yr BP there was a regression (Isla 1998, Cavallotto *et al.* 2004, Angulo *et al.* 2006, Bracco *et al.* 2008). The radiocarbon date of the basal zone of the core indicates that this unit was deposited when sea level was +2 m higher than that of the present. Therefore the coast of Montevideo was subject to extreme sediment resuspension processes. For this reason sediment hiatus(es) or lithological displacements were likely to occur. However, absolute resuspension and complete sediment mixture should also have been possible, because in those places of the bay where dredging activities are most intense, sediment cores exhibited sandy sediments at the bottom (i.e. 2.5 to 2 m below sediment surface) with a clay layer on the top (García-Rodríguez unpublished). In the case of our core, eight lithological units, with alternation of sand and silt layers were identified. Therefore, although our sediment core does not probably represent a continuous record, it is still possible to infer some Holocene paleoenvironmental changes. In this sense, the occurrence of the prehistoric sandy layers mentioned above is thought to be a consequence of tempestites, which could be attributed to either strong winds (i.e., hurricanes) or heavy rains in the hinterland.

The organic matter values showed an increasing trend from the basal section to 100 cm, thus indicating a eutrophication trend. However, between 130 and 120 cm a decrease in the organic load associated with a sandy layer was observed. The deposition of such a sandy layer might be associated to an increased marine influence as a consequence of either strong winds or ocean storms (De la Vega Leinert *et al*. 2000). This eutrophication trend matches with the Holocene sea level decrease. A similar relation between a sea level regression, and therefore paleosalinity decrease, and trophic state has been inferred for coastal lagoons of SE Uruguay (García-Rodríguez *et al.* 2004a, b). Similar trophic state changes in relation to sea level and paleosalinity have been also observed in South Africa (Gordon *et al*. 2008) and Australia (Saunders & Taffs 2009). Such an increase in trophic state is also supported by the decrease in cyst : diatom ratios (Smol 1985). This interpretation is based on the observation that in oligotrophic systems, cyst to diatom ratios often display relatively high values, whereas in meso–eutrophic systems the ratios are often lower.

Between 95 and 85 cm depth another sandy layer was deposited. Here, very low organic matter and heavy metal values were detected, which together with dominance of marine/brackish over freshwater diatom species suggest an increase in salinity levels. As observed elsewhere this might indicate a strong oceanic influence because of either strong winds from the SE or ocean storms (De la Vega Leinert *et al*. 2000). Above 85 cm depth, eutrophication probably intensified as inferred from high organic matter levels. In addition, increases in heavy metals associated with silt sediments and dominance of freshwater diatoms might indicate a continental influence. Therefore, the alternation between marine and freshwater conditions might have controlled the trophic state, salinity and reference levels of heavy metals (García-Rodríguez *et al*. 2004a, b).

At 34 cm depth, a major environmental change likely took place. The sharp and steady increases in all heavy metals until reaching values higher than those of downcore reference concentrations (for sand and silt), highlight the beginning of the human impacts and modification of the natural conditions. The first historical strong impact was the construction of the modern harbor between 1901 and 1909 (Fig. 2), thus leading to a more diminished marine influence because of the dock construction. Between 1917 and 1937 the strongest human industrial impacts, population increase and urban development were documented. In particular, the leather tanning industry mainly located in the Pantanoso Stream basin, the thermoelectric facility and the oil refinery were probably the most important heavy metal contamination sources. Therefore the 34 cm layer, which corresponds to the dramatic heavy metal increase, appears to correspond to 1917 AD. Assuming a constant sedimentation rate, mean annual sediment deposition is 3.74 mm yr^{-1}. Therefore the 40 cm layer should correspond to beginning of the construction of the harbor in 1901. Our calculation of sedimentation rate is in close agreement with that of Cranston & Kurucz (2002) who estimated a sedimentation rate of 3 mm yr^{-1} for the inner bay and 2 mm yr^{-1} for the outer bay (see Fig. 1). They also identified the leather tanning activities as the major input of heavy metals into the bay. According to our data, both the leather industry and the refinery are identified as the major heavy metal contamination sources to this system. The principal heavy metal of the effluents of the tanner industry is chromium (Muniz *et al*. 2004b). It is also important to mention that the region of the city where most leather tanning facilities are located does not have a sanitation system (Pantanoso stream region). According to Muniz *et al.* (2004b), the Pantanoso basin by itself received in 2000 an estimated annual load of 160 metric tons of Cr from untreated wastewaters of tanneries. Additional unpublished information stated that the leather industry has had an important decrease since 1993 and actually less than 10% of the past load was discharged directly to the Pantanoso stream (www.imm.gub.uy). On the other hand, as not all refineries have the same processes, different effluents will have different chemical composition depending on the type of treatment they receive. As a general rule, petroleum refinery wastewaters consists of many different chemicals that include oil and greases, phenols (creosols and xylenols), sulphides, ammonia, suspended solids, cyanides, nitrogen compounds and heavy metals, such as chromium, iron, nickel, copper, molybdenum, selenium, vanadium and zinc (Concawe 2004). The steady Pb increase registered in the sediment core could be also related to the continuous increase of fuel, due to the transportation and development of the city. Thus, population growth, unsustainable urban development, lack of sanitation and industrial waste were identified as the major causes of the environmental degradation of the bay.

After the construction of the harbor, in addition to heavy metal increases, a steady increase in organic load was registered. Such a eutrophication process is thought to be a consequence of the increased urban development without a sanitation system. In this sense, it is important to highlight that most of the facilities of the food industry are located in the area of the city where a sanitation system is lacking. Therefore, the effluents of such facilities that are deposited in the bay might be the cause of the increase in trophic state to hypertrophic levels, as several authors have classified the bay as hypertrophic (Nagy *et al*. 1987, Gómez-Erache *et al.* 2001, Danulat *et al*. 2002, Centurión *et al*. 2007).

Although our paleoenvironmental data cannot identify all the probable causes for environmental changes because of the inherently strong physical conditions influencing coastal systems, we observed shifts in heavy metals, organic matter and diatoms that appear to be the consequence of natural changes and human impacts. Thus, two main stages were identified. Before the human impacts, the system exhibited estuarine conditions as highlighted by the occurrence of both freshwater and marine/brackish taxa. After the construction of the harbor, population increase, beginning of industrial activities, and urban development led to the contamination and eutrophication. Therefore, the paleoenvironmental reconstruction presented here is indeed very important to show managers how to learn from historical bad practices and to start promoting environmentally friendly and sustainable policies in the future.

Acknowledgements

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