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# SCREENING OF SEDIMENT POLLUTION IN TRIBUTARIES FROM THE SOUTHWESTERN COAST OF THE RIO DE LA PLATA ESTUARY

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**Abstract:** Sediment chemistry and textural properties of transported materials from different surface water basins of South America has recently started to be investigated in relation to provenance materials and pollution sources. The objective of the present study is to analyze and compare pollution burdens in bottom sediments from distal positions of three drainage basins running across urban and industrialized areas and to compare them with more preserved sectors (an upstream position and a water stream with low anthropic influence). The surface bodies of water cross wind and water-reworked substrate materials from the Pampean loess and discharge into the Río de la Plata. Sampling was done in distal positions of the Luján and Riachuelo rivers, Canal Oeste, and Juan Blanco creek, and in Las Flores creek, a tributary of the Luján River. Standardized methods for the determination of granulometric parameters, major matrix components, and organic and inorganic pollutants were employed. Assessment of similarities between rivers by Principal Component Analysis show that distal positions of the Luján and Juan Blanco rivers and the tributary group together, and that Riachuelo and Canal Oeste split from that group by the effect of the components 1 and 2. The last two bodies of water also split from each other mainly by effect of component 2. Variables contributing most to the separation of these two bodies of water between each other are mainly given by heavy metals and sulfide. A similar behavior is also shown by cluster analysis.

**Resumen:** Recientemente se han iniciado investigaciones sobre la química de sedimentos y las propiedades texturales de materiales transportados de diferentes cuencas hidrológicas de América del Sur, en relación con la proveniencia de los materiales y fuentes de contaminación. El objetivo del presente estudio es el de analizar y comparar cargas de contaminación de sedimentos de fondo extraídos en posiciones distales de tres cuencas que atraviesan zonas urbanizadas e industrializadas, y compararlos con sectores más preservados (un sector aguas arriba y un arroyo con baja influencia antrópica). Los cuerpos de agua superficial atraviesan sustratos compuestos por materiales retrabajados de origen eólico y fluvial pertenecientes al Loess Pampeano y descargan en el Río de la Plata. Los muestreos fueron realizados en posiciones distales del Río Luján, Riachuelo, Canal Oeste y Arroyo Juan Blanco, además del Arroyo Las Flores, tributario del Río Luján. Se emplearon métodos estandarizados para la determinación de parámetros granulométricos, componentes mayoritarios de la matriz, contaminantes orgánicos e inorgánicos. La evaluación de similitudes entre ríos por Análisis de Componentes Principales indica que las posiciones distales de los ríos Luján y Juan Blanco, además del tributario se agrupan entre sí, y que el Riachuelo y Canal Oeste se separan del grupo por efecto de las componentes 1 y 2. A su vez, estos dos últimos cuerpos de agua se separan entre si por efecto de la componente 2. Las variables que contribuyen en mayor medida a la separación de los mismos entre si, son principalmente los metales pesados y los sulfuros. El análisis de clusters muestra un comportamiento similar al mencionado.

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**Keywords**: bottom sediments, matrix composition, pollutants, principal component analysis.
**Palabras clave**: sedimentos de fondo, composición matriz, contaminantes, análisis de componentes principales.

## INTRODUCTION

The Río de la Plata is a complex water system that connects "del Plata Basin" with the Atlantic Ocean. Its southwestern coastal sector holds densely populated areas with tributaries running across them, which are also receptors of different discharges of pollutants (Ronco *et al*., 1995; Kreimer *et al*., 1996; Ronco *et al*., 2001, 2007). Streams flow from southwest to northeast discharging along the coast of the Río de la Plata Estuary. The region is mainly composed by Pleistocene silts and sands, with Holocene marine clays and silts predominating in the lower sectors of the coastal plain (Manassero *et al*., 2004; Dangavs, 2005). Human activity is the cause of serious pollution of surface waters (Cattogio, 1990; AA-AGOSBA-ILPLA-SHN, 1997; Salibián, 2006), sediments (Colombo *et al*., 1990; Kreimer *et al*., 1996; Ronco *et al*., 2007) and soils (Camilion *et al*., 2001; 2003) due to point and non-point industrial, agricultural and urban sources.

The sorption capacity and accumulation of pollutants in bottom sediments of different type of water environments has been studied and documented for several decades (Salomons and Förstner, 1984; Horowitz, 1985; Ferguson, 1991), although its knowledge within the area of study has only been recently reported, mainly in relation to metal burdens (Villar *et al*., 1999; Ronco *et al*., 2001; Camilión *et al*., 2003; Ronco *et al*., 2007).

This contribution provides new sedimentological data, major matrix components, and organic and inorganic compound loads in bottom sediments from four critical tributaries of the Río de la Plata coastal area, compared with streams under lower influence of human disturbance, within the frame of a broader study for the risk assessment of contaminated sediments in the region. Principal Components Analysis allowed the determination of the major contributions to pollution for the most polluted sampling sites.

## STUDY AREA AND METHODS

The Riachuelo is a distal river sector of the Matanza-Riachuelo Basin, an emblematic river that has been historically reported as one of the most polluted bodies of water of the region due to wastes from slaughterhouses, food and metal industries, tanning factories, oil and petrochemical factories, among others, and that has been of concern for almost two hundred years (Brailovsky and Foguelman, 1991). Although a few projects for its decontamination have been mentioned as early as 1822, its pollution burden continued to increase over the past decades. Sediment cores over one and a half meters long from previous studies have shown high contents of heavy metals, inorganic and organic pollutants within the assessed depth, in spite of material removal by dredging activities to maintain water depth for navigation at the mouth (Kreimer *et al.*, 1996). The Canal Oeste is another example of a seriously damaged body of water, associated to discharges of an oil refinery and several petrochemical plants of an important industrial complex with metal and hydrocarbon pollution burdens (Cattogio, 1990; Ronco *et al.*, 1995; Alzuet *et al*., 1996). Although Luján River has been also found severely contaminated by multiple sources of pollution in upstream sectors, downstream sectors were found to be more preserved (Di Marzio *et al.*, 2005; Salibián, 2006; Salibián and Ferrari, 2006). On the contrary, Juan Blanco and Las Flores creeks showed much lower anthropic influence (Feijoó *et al.*, 1999).

Sampling sites (coordinates given between parenthesis) were located in the lower sectors of the Luján (34º 26' 50,20" S; 58º 31' 17,46" W) and Riachuelo (34º 38' 12,23" S; 58º 21' 12,03" W) rivers, the Canal Oeste (34º 52' 22,59" S; 57º 54' 20,47" W), Juan Blanco (35º 8' 28,49" S; 57º 26' 27,92" W), and Las Flores (34º 27' 41, 04" S; 59º 1' 36,69" W) creeks, the latter being a tributary of the Luján river (Fig. 1). At least, 20 sub-samples from the upper layers of the bottom sediments from each sampling site were obtained with an Ekman grab. The sampling points were located as follows: Luján in its middle section, at 2 m depth; Las Flores at 0.15 m depth, Riachuelo on its left margin, in a sector where no dredging activity was detected, within 2 and 4 m depth; Canal Oeste in the middle sector at 0.5 m depth; Juan Blanco creek in left margin sector, where sedimentation was evident, and below 0.5 m depth. Analyses were performed on representative homogenized subsamples from each study site.

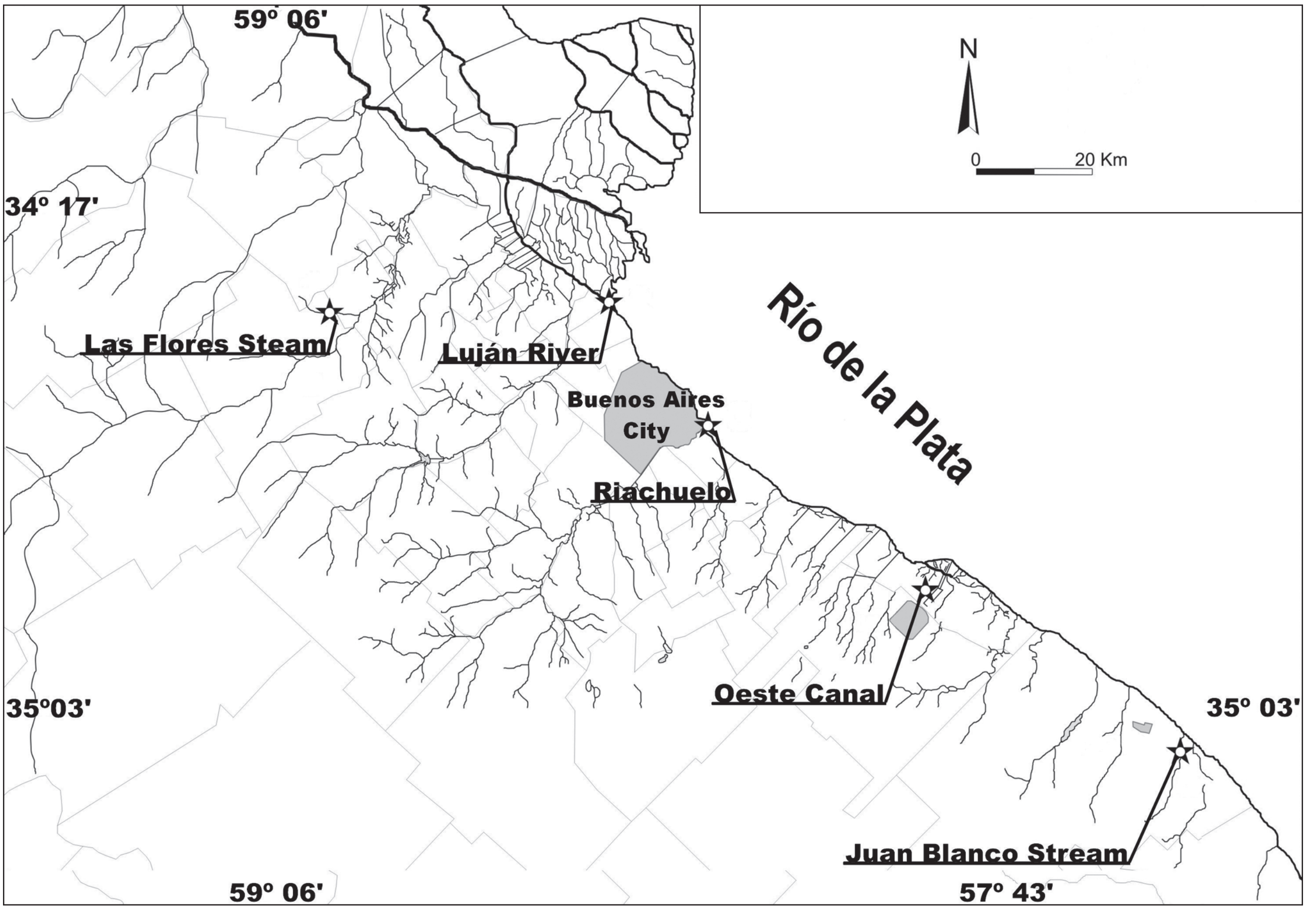


**Figure 1.** Study area and sampling point locations.
**Figura 1.** Área de estudio y localización de sitios de muestreo.

Sieving and settling velocity techniques, with previous cement removal (Day, 1965), was performed for grain size analysis. Loss on ignition (LOI) at 105 and 550°C was estimated by gravimetric methods. Analysis of total metal content following acid digestion of samples was done by flame atomic absorption spectrometry (direct air-acetylene flame method), except for Hg and As, that were analyzed, respectively, by cold-vapor and hydride generation atomic absorption spectrometry (USEPA, 1986; APHA, 1998). Ca and Mg were determined by titration (Method 3500, APHA, 1998) previous extraction in 1 N ammonium acetate followed by filtration; sulfide (Method 9030, USEPA, 1986); cyanide (Method 9010, USEPA, 1986); nitrate and phosphorus (Black, 1965); chloride (Method 4500, APHA, 1998) previous extraction in 0.1 N $K_2SO_4$; pesticides by gas chromatography electron capture detection, previous extraction and clean up (Methods 3550 and 3630, USEPA, 1986); hydrocarbons were analyzed by infrared spectrometry, previous extraction in carbon tetrachloride (Method 418.1, USEPA, 1983). Reagents: Traceable certified standards for the analysis of metals were from AccuStandard, Inc. (1000 mg/L standard stock solutions, traceable to National Institute of Standards and Technology, USA); and pesticides from SENASA (Argentinean National Service for Sanitary and Quality of Agriculture and Food). Chemicals for sample treatment or analysis of major matrix components were analytical grade. Qualities of solvents used were for the analysis of pesticides residues or certified for IR analysis. Sediment samples were classified according to Folk (1954). Statistical analysis of results of raw data was done by principal component analysis (PCA) and cluster analysis (Hair *et al.*, 1999).

## RESULTS

### Matrix composition

Results of grain size analysis, LOI and water proportion can be seen in Table 1. Samples fall into sandy silt and sandy mud classification. The LOI -550 and 900°C ranging from 2.8 through 21.5%, and 3.5 through 23.7%, respectively, indicates a variable content of organic and inorganic carbon. Both Canal Oeste and Riachuelo bottom sediments exhibited the highest values.

Data corresponding to chemical analysis of major and minor inorganic or organic constituents can be seen in Table 2. Mean major metal content values indicate that the order of abundance is the following: Fe> Ca> K> Mg> Mn> Na, with concentrations ranging in almost two orders of magnitude between most and least abundant. Highest concentrations were always detected in sediments from Canal Oeste. Although a higher variability regarding minor metal constituents can be observed, mean values (not including in the mean value estimation the high content of Cr from Riachuelo, two orders of magnitude higher than background levels) indicate the following trend: Zn> Pb> Cu> Ni≅ Cr≅ As> Cd> Hg. The highest concentrations were found in the sediments of Riachuelo and Canal Oeste.

Sulfides were not detected in the Luján, Las Flores and Juan Blanco bottom sediments, though Canal Oeste

| Sample | LOI (% dry weight) | | Water (%) | Particle size (% dry weight) | | | Sediment Classification (Folk, 1954) |
|---|---|---|---|---|---|---|---|
| | 550°C | 900°C | | Sand | Silt | Clay | |
| Luján | 2.8 | 3.5 | 38.7 | 15.8 | 61.5 | 21.9 | Sandy silt |
| Las Flores | 8.0 | 9.4 | 62.4 | 33.2 | 30.0 | 37.0 | Sandy mud |
| Riachuelo | 13.0 | 14.3 | 61.9 | 39.5 | 53.5 | 7.0 | Sandy silt |
| C. Oeste | 21.5 | 23.7 | 90.5 | 18.6 | 43.4 | 37.5 | Sandy mud |
| J.Blanco | 12.2 | 13.4 | 67.8 | 14.8 | 48.7 | 36.4 | Sandy mud |

**Table 1.** Loss on ignition, water proportion, grain size composition and sediment sample classification.
**Tabla 1.** Pérdida por ignición, proporción de agua, composición granulométrica y clasificación de muestras de sedimentos.

| Compound | Sediment sample source | | | | |
|---|---|---|---|---|---|
| (mg/kg)dw | Luján | Las Flores | Riachuelo | Canal Oeste | Juan Blanco |
| As | 3.4 | 5.2 | 15.1 | 18.2 | 9.0 |
| Cd | 0.5 | 0.3 | 2.4 | 3.2 | 1.0 |
| Cr | 20.3 | 15.5 | 1141.1 | <2.5 | <2.5 |
| Cu | 7.9 | 8.1 | 136.2 | 55.8 | 15.8 |
| Hg | <0.1 | <0.1 | 1.9 | 6.8 | 0.6 |
| Ni | 3.6 | 2.3 | 19.8 | 39.7 | 7.1 |
| Pb | 33.4 | 22.2 | 204.9 | 124.7 | 37.3 |
| Zn | 44.3 | 32.0 | 706.7 | 387.3 | 69.9 |
| Fe | 16653 | 14710 | 27585 | 34263 | 24301 |
| Mn | 1074 | 2132 | 1066 | 3079 | 1458 |
| Na | 418.3 | 883.7 | 912.1 | 3026.3 | 345.5 |
| K | 2327 | 2911 | 3067 | 5437 | 4557 |
| Ca | 4858 | 8501 | 8258 | 12994 | 6679 |
| Mg | 822.1 | 2720 | 1784 | 5972 | 1690 |
| Nitrate | 1.0 | 6.0 | 0.2 | 9.5 | 1.6 |
| Chloride | 95.8 | 46.5 | 19.6 | 1673 | 117.8 |
| Sulfate | 565 | 169 | 175 | 1177 | 461 |
| Sulfide | <2.0 | <2.0 | 2923 | 1343 | <2.0 |
| Cyanide | <1.0 | <1.0 | 15.9 | 33.1 | <1.0 |
| P total | 539.0 | 482.5 | 4430 | 5334 | 469.0 |
| Hydrocarbons | <200 | <200 | 286 | 22361 | <200 |

**Table 2.** Data from sediment chemical analysis of major components and minor matrix components and pollutant contents.
**Tabla 2.** Datos de análisis químicos de muestras de sedimentos de componentes mayoritarios, minoritarios y contaminantes.

| | F1 | F2 | F3 | F4 |
|---|---|---|---|---|
| As | 7.894 | 0.758 | 2.937 | 4.945 |
| Cd | 7.990 | 0.812 | 2.295 | 1.775 |
| Cr | 0.544 | 17.108 | 3.020 | 4.373 |
| Cu | 2.986 | 12.114 | 0.963 | 1.793 |
| Hg | 7.836 | 1.050 | 0.429 | 8.209 |
| Ni | 8.315 | 0.040 | 0.743 | 6.486 |
| Pb | 4.360 | 9.228 | 0.458 | 0.072 |
| Zn | 4.220 | 9.477 | 0.892 | 0.113 |
| Fe | 7.264 | 0.186 | 19.171 | 0.025 |
| Mn | 3.685 | 9.599 | 6.116 | 6.156 |
| Na | 7.009 | 2.588 | 3.716 | 5.445 |
| K | 4.365 | 3.523 | 32.441 | 22.396 |
| Ca | 7.059 | 1.788 | 6.055 | 11.890 |
| Mg | 6.120 | 4.856 | 3.084 | 1.818 |
| Nitrate | 2.908 | 10.444 | 13.922 | 2.595 |
| Chloride | 5.953 | 4.702 | 0.584 | 17.265 |
| Sulfide | 3.739 | 10.350 | 2.279 | 0.164 |
| Total P | 7.754 | 1.379 | 0.895 | 4.480 |

**Table 3.** Principal Component Analysis of sediment data. Percentage of variable contributions.
**Tabla 3.** Análisis de Componentes Principales de los datos. Contribución porcentual de variables.

| | F1 | F2 | F3 | F4 |
|---|---|---|---|---|
| Obs1 | 5.854 | -2.156 | 0.043 | -0.236 |
| Obs2 | -1.835 | -0.583 | 1.423 | 0.510 |
| Obs3 | -2.294 | -1.564 | -1.252 | 0.528 |
| Obs4 | -3.520 | -0.044 | 0.024 | -0.935 |
| Obs5 | 1.796 | 4.348 | -0.238 | 0.132 |

**Table 4.** Principal Component Analysis of sediment data. Observation coordinates.
**Tabla 4.** Análisis de Componentes Principales de los datos de sedimentos. Coordenadas de observación.

and Riachuelo reached concentrations much above the 1000 mg/kg dry weight, in agreement with evidence of anaerobic conditions in those bodies of water. Also, enrichment of total phosphorus and cyanide pollution were detected in these sediments. High concentrations of chloride, sulfate and nitrate were seen the Canal Oeste, in addition to high burdens of hydrocarbons.

The analyzed pesticides were: hexachlorocyclohexane (HCH), hexachlorobenzene (HCB), lindane, heptachlor, aldrin, chlorpyrifos, heptachlor epoxide, dieldrin, o,p´-DDT, p,p´-DDT, diazinon, methyl-parathion, malathion, D-allethrin, cypermethrin, tetramethrin, permethrin, endosulphan-1 and 2. Concentrations were mostly below the detection limits of the method (2 μg/kg) in all streams, except for the following (concentrations given in μg/kg of dry sediment): Luján River with 272.2 of cypermethrin, and 36.8 of chlorpiryfos; Riachuelo with 3.3 of Endosulphan-1; Canal Oeste showed 15.9 of p,p´-DDT and 6.7 of Cypermethrin; and Juan Blanco stream 2.7 of Aldrin plus chlorpiryfos.

### Statistical analysis

Principal component analysis shows a clear differentiation of the Riachuelo and Canal Oeste sediments with respect to the rest of the sampling places. The Tables 3 and 4 show the percent contribution of each variable to different components and the observed relative position with respect to components. Figure 2a indicates that the Luján and Juan Blanco Rivers and Las Flores creek are neither separated by the component 1 nor the 2; and that Canal Oeste and Riachuelo split from that group by the effect of the component 1 and 2; and also from each other principally by effect of the component 2. Variables contributing most to the separation of these two bodies of water from each other are given by the Cr, Cu, Zn and Pb, added to sulfide, nitrate and Mn in a positive way. On the other hand, the group of Las Flores, Luján and Juan Blanco split from Riachuelo and Canal Oeste by component 1, mainly determined by the majority of the variables. Therefore, the first three components do not exhibit large differences between them in relation to the data included in the PCA. A very similar behavior is also given by the cluster analysis (Fig. 3). The analysis of PCA did not include the variable pesticides and total hydrocarbons since the majority of samples were below detection limits, and therefore yielding a meaningless PCA.

## DISCUSSION

The Fe and Mn concentration levels in bottom sediments of the studied streams are within the range of those found in stream flowing into the lower Paraná Basin (Ronco *et al.*, 2007) and coastal plain of the Río de la Plata (Camilión *et al.*, 2003). The high concentrations of almost all the measured heavy metals reflect the presence of pollution sources in Riachuelo and Canal Oeste, with very high levels of chromium in the former and high mercury in the latter. This is consistent with tanning and metal industry-location in the former basin and oil or petrochemical plants in the latter. Previous reports of Cr levels in sediments from equivalent positions of Riachuelo were found to be within the same order of magnitude (Kreimer *et al.*, 1996), indicating that the historic levels of this metal pollution remain at present. Higher Cr concentration with respect to background levels in Luján River, are possibly related to discharges from tanning factories located in upstream positions of its basin (Salibián, 2006). Slightly higher values were also detected in sediments of the area by Cataldo *et al*. (2001). Concentrations of this metal in the other studied streams are within the range of those reported by other authors for

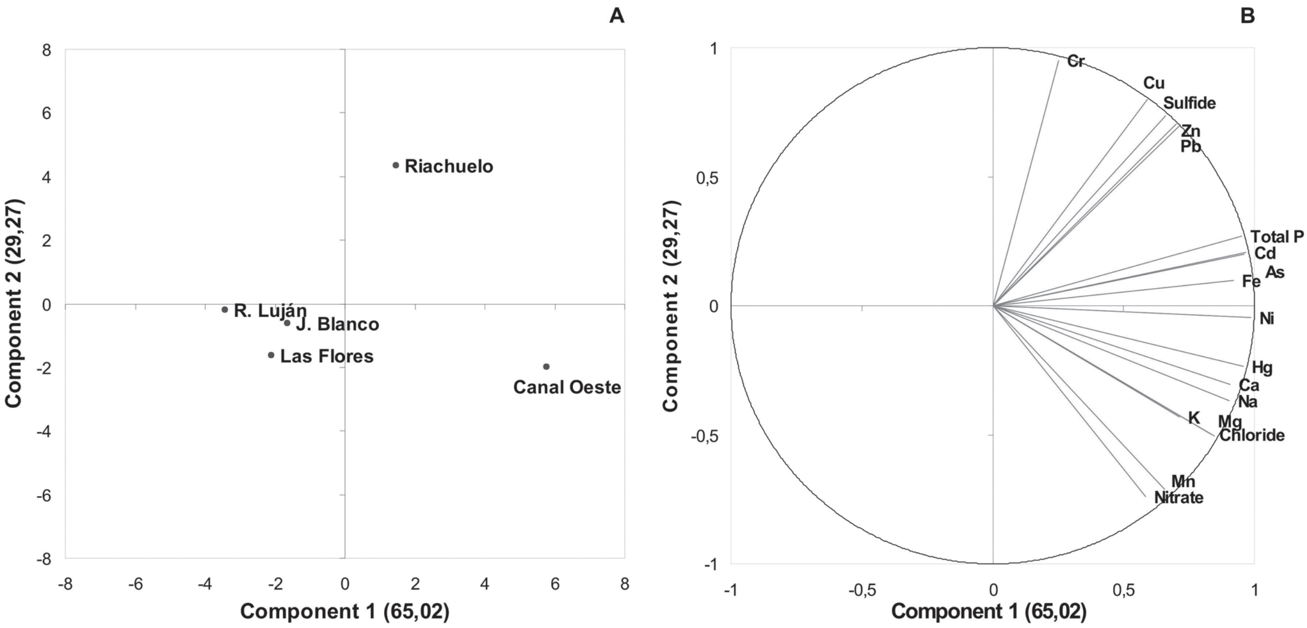


**Figure 2.** Two dimension plots from Principal Component Analysis, with two components explaining 94.3 % of the total variance. a) corresponds to sampling sites, b) corresponds to variable distribution with individual partial correlation.

**Figura 2.** Diagrama en dos dimensiones del Análisis de Componentes Principales con dos componentes que explican el 94.3% de la varianza total. a) corresponde a los sitios de muestreo, b) corresponde a la distribución de las variables y su correlación parcial.

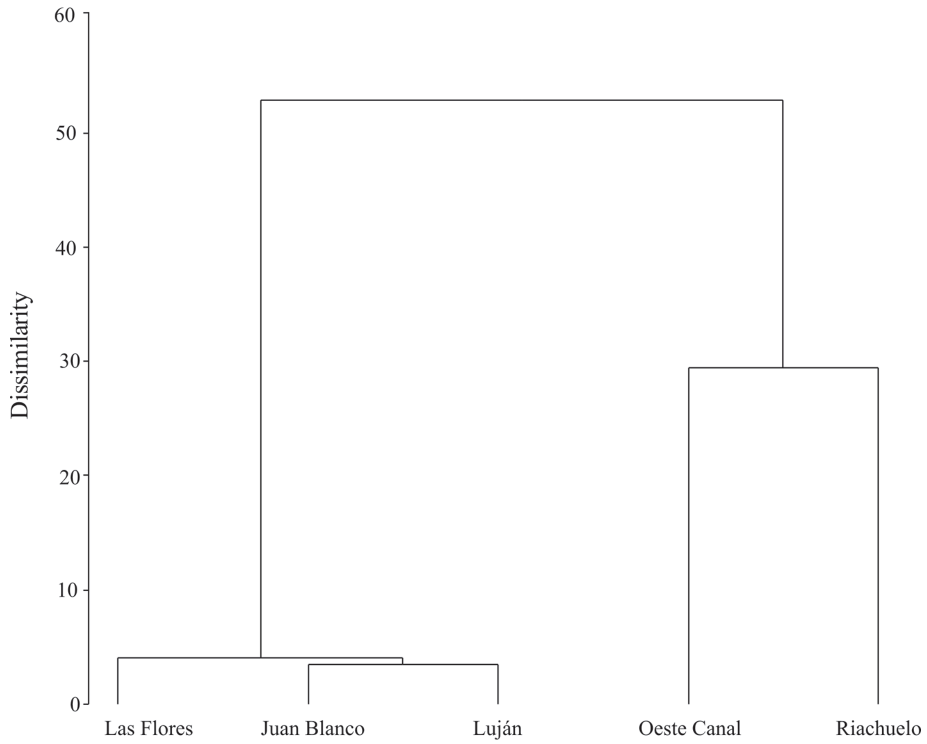


**Figure 3.** Cluster analysis using Euclidian distances and single linkage of the five sample data set using raw concentrations of chemical components. Data from pesticides and total hydrocarbons were not included.
**Figura 3.** Análisis de clusters usando distancias Euclidianas y distancias mínimas para el banco de datos correspondiente a las cinco muestras con contenidos no normalizados de concentraciones de compuestos químicos. No se incluyen datos de pesticidas e hidrocarburos totales.

the southern streams discharging in the Río de la Plata (Bilos *et al.*, 1998; Villar *et al.*, 1998; Ronco *et al.*, 2001) and right margin streams flowing into the lower Paraná Basin (Ronco *et al.*, 2007). Regarding the detected Hg levels, comparisons of reported concentrations from contaminated sectors of the del Gato creek reaching 0.88 mg/Kg (Ronco *et al.*, 2001), with those observed in Canal Oeste, one order of magnitude above, make the pollution burdens in this canal evident. Also, bottom sediments from Riachuelo double the reported levels of the del Gato creek. The concentration levels of Cd, Cu, Ni, Pb and Zn exhibit a similar trend; they are approximately one order of magnitude higher in Riachuelo and/or Canal Oeste, with respect to the other studied places, and comparable to those reported for polluted downstream sectors of the el Gato stream (Ronco *et al.*, 2007). Sediment features from Riachuelo and Canal Oeste exhibit dark coloration associated to metal sulfides. Comparing the measured concentrations of metals, phosphorus and sulfur levels with mean abundances and abundance interval values for sediments of the lithosphere according to Frink (1996), it can be observed that either one of them or both, Riachuelo and Canal Oeste, fall beyond the upper values of concentration for Cd, Hg, Pb, P and sulfur (as sulfide), being also Cr and Zn almost on the upper value ranges.

Bottom sediments from the Canal Oeste also show high levels of both hydrocarbons and organic matter. Several reports on hydrocarbon pollution and associated biological effects from sediments of this canal have been found (Cattogio, 1990; Ronco *et al*, 1995, 1996; Alzuet *et al.*, 1996), showing the persistence of this problem to the present. Although the concentration of total hydrocarbons were below the detection limit of the employed method (200 mg/kg) in the other studied sediments (except Riachuelo), previous reports on the mouth of the Luján River and the Paraná de las Palmas (Cataldo *et al.*, 2001) showed aliphatic hydrocarbon concentrations of 11 and 2 mg/kg, respectively (Cataldo *et al.*, 2001).

The highest concentrations of pesticides were detected in Luján River, due to intense agricultural activities in both margins. Concentration of chlorpyrifos and cypermethrin, the two insecticides of regional significance associated to biotech crops, are even higher than those reported for sediments from surface bodies of water of the core soybean production area of the Rolling Pampas (Marino and Ronco, 2005). Lower concentrations were also detected in Juan Blanco stream, probably related to rural areas in upstream positions. Relatively high levels of a chlorinated pesticide in Canal Oeste could be attributed to old industrial burdens from past production of this compound, not marketed for more than three decades. The presence of other pesticides in Canal Oeste could be explained by their domestic use.

## CONCLUSIONS

Results of the study show high burdens of different chemical loads in Riachuelo and Canal Oeste. Concentration levels of pollutants are within ranges expected to produce biological effects on aquatic biota. If we compare the concentrations of heavy metals with probable effects levels (PEL) provided by the Canadian Council of Ministers of the Environment (1999), with values (mg/kg) for As: 17; Cd: 3.5; Cr: 90; Cu: 197; Hg: 0.486; Ni: 35.9; Pb: 91.3; Zn: 315, we can observe that measured concentrations of almost all metals in Riachuelo and Canal Oeste sediments do not meet those levels. This is in agreement with previous reports on toxicity test assessments with different type of organisms inducing lethal, sub-lethal and mutagenic effects with samples of sediments of those sources. It should be pointed out that the Hg level detected in Juan Blanco creek sampled sediment is slightly above the PEL for this metal. This should be further investigated, the stream being with low evidence of pollution sources.

Although the Luján River shows evidence of depuration processes along its course, yielding to lower pollution levels in the mouth of the river, sediments in this last position exhibit the highest concentrations of pesticides found in the bodies of water analyzed in this study. Las Flores and Juan Blanco bottom sediments are less contaminated, making them good for potential use as reference control materials for toxicity assessment surveys in the region.

Evidence from previous reported data and results from the present study show concern for pollution levels in sediments of two of the studied bodies of water from the region, pointing to the need for mitigation interventions. The present study was done within the frame of a major project in relation to providing geochemical and ecotoxicological methods for environmental risk assessment of contaminated sediments in the region, aiming to provide management tools for decision makers.

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