



# Metal geochemistry in suspended and bed sediments in the eutrophic lowland Salado River basin (Buenos Aires, Argentina)

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## Abstract

The Salado River is a lowland river that drains a vast and productive region of central Argentina into the Río de la Plata estuary. To evaluate water quality and the geochemistry of major and trace metals (Fe, Mn, Zn, Cu, Pb, Cr and Ni), water and sediments samples were collected along 500 km of the Salado River basin ( $n=21$ ). Waters were highly alkaline (pH:  $8.8 \pm 0.4$ ), saline ( $5.2 \pm 1.8$  mS/cm), turbid ( $467 \pm 237$  NTU) and eutrophic (chlorophyll *a*:  $187 \pm 119$  µg/l) with a decreasing gradient towards the river mouth reflecting the contribution of alkaline ground waters and eutrophication at the river head. Suspended particles (70–595 mg/l) were enriched in total organic carbon, nitrogen (5–7 times) and metals (1–4 times) relative to bed sediments, but metal concentrations were 2–10 times below the world river's average. Consistently, metal concentrations in bed sediments were lower than sediment quality guidelines with an increasing downriver trend following the enrichment of clays. The enrichment factors (EF) and geoaccumulation index (I<sub>geo</sub>), calculated with local background metal levels from the deepest layers of a dated sediment core, indicate a general prevalence of natural metal sources (EF < 1.5) with unpolluted sediments (I<sub>geo</sub> ≤ 0), excepted for the Chivilcoy stream, which showed a significant enrichment of Zn, Cu and Pb (EF = 2–12; 0 ≤ I<sub>geo</sub> ≤ 1) constituting a clear pollution hot spot in the basin.

**Keywords** Metals · Suspended particulate matter · Bed sediments · Salado River

## Introduction

The Salado River has a ~600-km-long lowland basin (slope ~0.1 m/km) that drains a vast (150,000 km<sup>2</sup>) and productive region of the Buenos Aires province into the Samborombón Bay (a protected RAMSAR site) in the outer

Río de la Plata estuary (Fig. 1). The Salado River basin is sparsely populated with small to medium urban centers (< 100,000 inhabitants) and has a heterogeneous land use. The main activity gradually shifts from intensive agriculture in the headwaters to extensive livestock in the lower basin (Gabellone et al. 2013). Furthermore, the basin has several small industrial settlements and it is also used for leisure activities, mainly recreational fishing. The most important urban–industrial complexes in the basin are Junín and Chivilcoy, the later was the first in the region, operating since 1969, basically with food processing, metallurgy and the production of plastics and textiles.

Salado River waters stand out by their high salinity, contributed by the drainage of aquifers enriched in sodium chloride, and a highly variable hydrological regime (< 100–1500 m<sup>3</sup>/s; Gabellone et al. 2005, 2008). The basin has two main tributaries, the Saladillo-Vallimanca and Las Flores streams, and many minor streams as well as permanent and temporary lakes (Fig. 1). To prevent recurrent floods, considerable hydraulic modifications were carried out all over the basin during the last century, including dredging and construction

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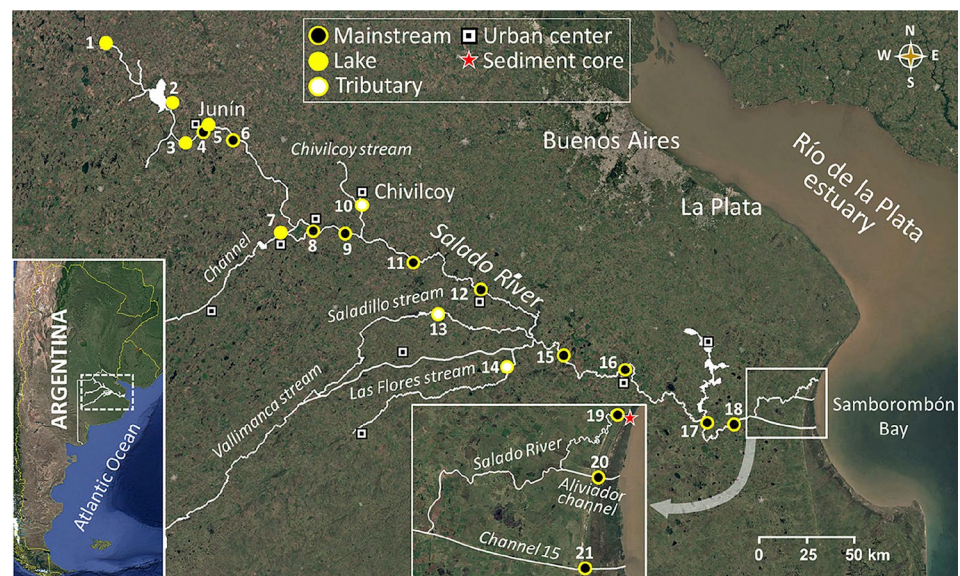
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**Fig. 1** Location of the Salado River basin and the sampling sites



of artificial channels and control gates (Conzonno et al. 2001; Bazurri et al. 2018).

The basic hydrochemistry and plankton dynamics of the Salado River have been extensively studied (Gabellone et al. 2005, 2008; Bazurri et al. 2018 and references therein), but despite the economic importance of the basin, no comprehensive study on the sources and distribution of pollutants has been yet conducted. To fill this gap, in this study, we analyze the variation of water quality parameters and the spatial distribution of major and trace metals in suspended and bed sediments along the Salado River basin.

## Materials and methods

### Sampling

Sampling was performed between August and September 2018 using a pneumatic boat at 21 sampling stations from the river head to the mouth including the main channel ( $n=13$ ), associated lakes ( $n=5$ ) and tributary streams ( $n=3$ ; Fig. 1). Sampling was performed in three consecutive field trips under low/normal flow conditions (discharge ranged between 79 and 90 m<sup>3</sup>/s (August–September; bdhi.hidrico-sargentina.gob.ar).

A flowchart of the general methodology is shown in S1 (Supplementary material). Water samples were collected manually 10 cm below the surface using 1-L amber glass bottles and bed sediments were sampled with a “Van Veen” stainless steel grab (Hydro-Bios), mixing three grab subsamples at each site. Water quality parameters (conductivity, pH, dissolved oxygen and turbidity) were measured in situ with a multi-parameter probe (Horiba U-52) calibrated previously at the laboratory with reference solutions.

### Chemical analyses

Suspended particulate matter (SPM) was separated by vacuum filtration of waters through 0.45 μm cellulose acetate filters. Grain size analysis of SPM and bed sediments was carried out by laser diffraction (CILAS 990L) using sodium hexametaphosphate as dispersing agent. Total organic carbon (TOC) and total nitrogen (TN) content were determined by catalytic high temperature combustion of SPM and bed sediments (Thermo Finnigan, CE Flash EA 1112 elemental analyser). The accuracy of TOC and TN analyses, validated using two certified reference materials (sediment B2150 and soil B2152, Elemental MicroAnalysis), ranged from  $97 \pm 3.6$  to  $98 \pm 8.4\%$  for TOC and  $92 \pm 17$  to  $97 \pm 21\%$  for TN, respectively. Chlorophyll *a* (Chl *a*) corrected for phaeopigments was determined spectrophotometrically (Hitachi 2001) on glass microfiber filters (Whatman GF/C) extracted with 90% acetone (Strickland and Parsons 1972).

For metal analyses (Fe, Mn, Zn, Cu, Pb, Cr and Ni), filters with SPM and homogenized sediments (1.0 g approx.) were digested with aqua regia (3:1 v/v, HCl to HNO<sub>3</sub>) and H<sub>2</sub>O<sub>2</sub> at 100 °C (Hseu et al. 2002). Metal concentrations were determined by flame atomic absorption spectrophotometry (Thermo Elemental Solaar M5) except for Cu, Pb, Cr and Ni in SPM samples which were measured by graphite furnace (GF95). The quality control and quality assurance scheme included the analysis of blanks, duplicate samples and standard reference material. Filter blanks ( $n=3$ ) and procedural blanks ( $n=5$ ) were negligible. The precision of trace metal determinations assessed by the relative standard deviation (RSD) of triplicate samples was below 10%. The accuracy was assessed by triplicate

analysis of certified freshwater sediment (CRM016-50G). The average recoveries for each metal ranged from 77 to 109% (Fe = 92 ± 10%; Mn = 80 ± 9.5%; Zn = 86 ± 2.9%; Cu = 93 ± 2.2%; Cr = 77 ± 4.4%; Ni = 91 ± 1.8%; Pb = 109 ± 13%).

**Enrichment factor and Geoaccumulation index**

To normalize trace metal concentrations of bed sediments and to identify the potential anthropogenic influence (Devesa-Rey et al. 2009), enrichment factors (EF) relative to local background values taking Fe as reference element was applied as:  $EF = (\text{metal}/\text{Fe})_{\text{sample}} / (\text{metal}/\text{Fe})_{\text{background}}$ . EF values around 1.0 suggest that the element comes from geogenic sources, whereas an EF much higher than 1.0 suggests anthropogenic sources. In addition, to further evaluate the extent of pollution in the river, the geoaccumulation index (Igeo; Müller 1969) was applied as:  $I_{\text{geo}} = \log_2 (C_n / 1.5 B_n)$ ; where  $C_n$  is the measured concentration and  $B_n$  is the geochemical background multiplied by 1.5 due to variability related to lithogenic effects. According to Müller (1969), metal pollution was categorized into seven classes ranging from  $I_{\text{geo}} \leq 0$  (unpolluted) to  $I_{\text{geo}} > 5$  (extremely polluted). The local background trace metal concentrations were obtained from the deepest sections of a dated sediment core (80–115 cm) collected in the Salado River mouth (Schuerch et al. 2016; Tatone et al. 2020). According to the average sedimentation rate calculated for this area (2.62 cm/year), these depths correspond to 1970–1982.

**Results and discussion**

**Water quality parameters**

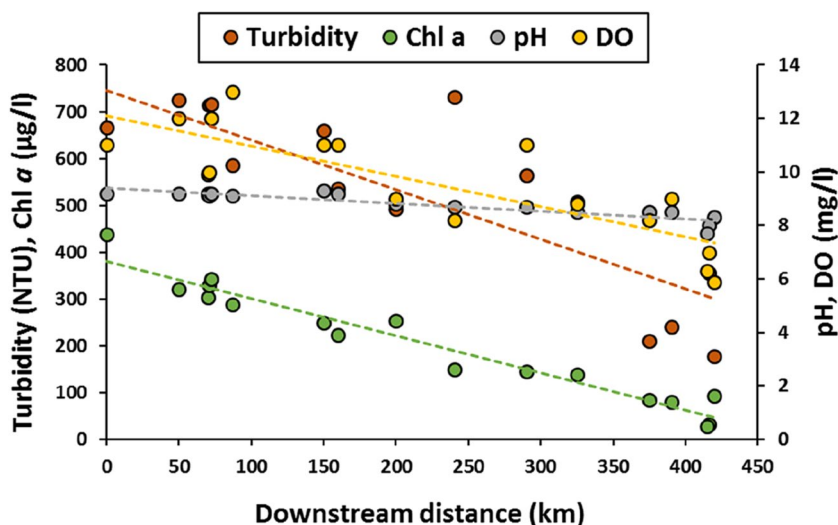
In general, sampling stations were very shallow (< 1.5 m), with highly alkaline (pH: 8.8 ± 0.4), saline (5.2 ± 1.8 mS/cm), turbid (467 ± 237 NTU), eutrophic (Chl *a*: 187 ± 119 µg/l) and relatively well-oxygenated waters (DO: 9.8 ± 2.1 mg/l). Chl *a* concentrations were very high and variable (20–439 µg/l; RSD 64%), in agreement with previously published values (5.0–416 µg/l, Gabellone et al. 2005; Bazurri et al. 2018) corresponding to eutrophic to hypereutrophic waters (Quirós and Drago 1999; Wetzel 2001). Chl *a* was significantly higher in lakes than in the main course and tributary streams ( $p < 0.05$ ) with the lowest value at Chivilcoy stream (20 µg/l).

In the mainstream, pH, dissolved oxygen, Chl *a* and turbidity displayed a significant ( $p < 0.001$ ) decreasing trend towards the river mouth (Fig. 2), reflecting the contribution of alkaline ground waters and nutrients from agricultural production in the headwaters (Gabellone et al. 2013) and the progressive sedimentation of particles along the river.

The high conductivity of Salado River waters (2.8–11 mS/cm) showed no clear spatial trend. The maximum value corresponds to the shallow Bragado lake which receives the contribution of salt-laden groundwaters from the NW endorheic region through artificial drainage channels constructed in the last decades (Bazurri et al. 2018).

SPM concentrations determined gravimetrically (70–595 mg/l) confirmed turbidity values obtained in situ (46–784 NTU) with a significant linear regression ( $R^2 = 0.93$ ; slope = 1.5,  $p < 0.0001$ ). The Chivilcoy and Las Flores streams showed the lowest turbidity values (46 and 63 NTU, respectively) whereas the Salado River mouth (site 19)

**Fig. 2** Spatial variation of water quality parameters along the Salado River mainstream





presented an atypically high turbidity (784 NTU), possibly due to resuspension of bed material in this extremely shallow site with restricted water flow, since most of the Salado River water (80%) is diverted to channel 15 and, to a lesser extent, Aliviador channel (Carol et al. 2014; Fig. 1).

To estimate the average suspended sediment load of the Salado River to the Samborombón Bay, the mean annual water discharge was taken from site 16, downstream the main tributaries (187 m<sup>3</sup>/s; bdhi.hidricosargentina.gob.ar). Considering the joint discharge of channels 15 and Aliviador, that make up 80% of total river discharge (Gabelleone et al. 2013), and their respective SPM concentrations (159–193 mg/l), the mean annual solid discharge would amount to ~0.8 × 10<sup>6</sup> t/year, representing a significant source of particulate matter to the Bay.

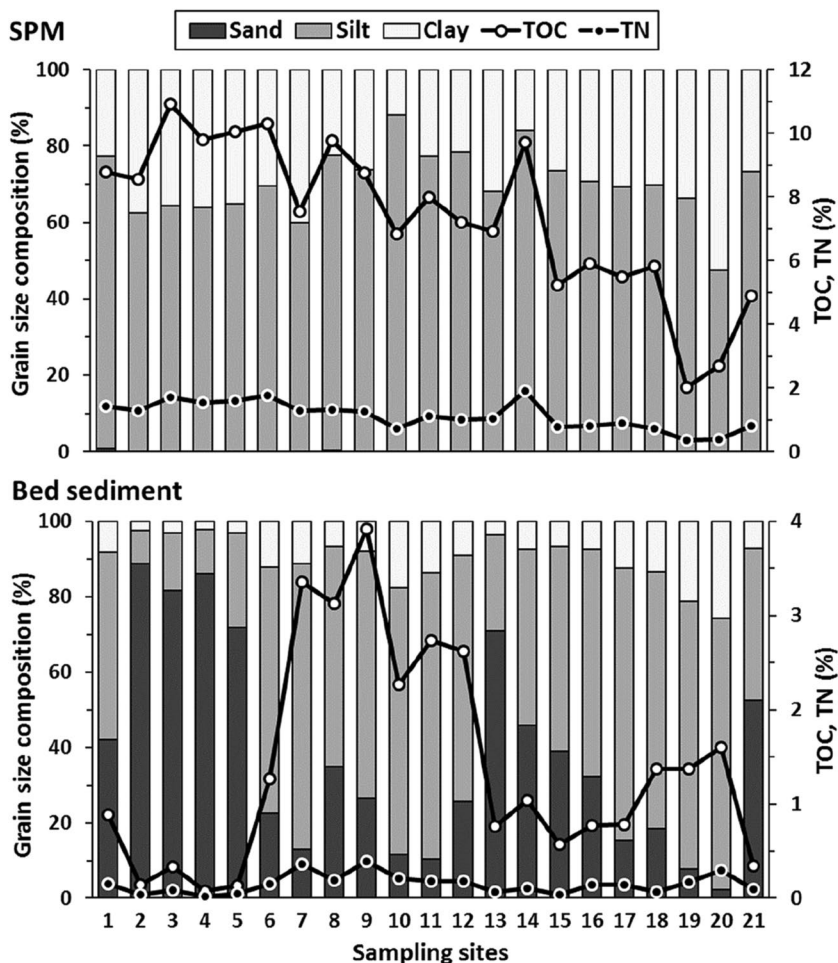
### Grain size composition, TOC and TN contents of suspended and bed sediments

Figure 3 presents the particle size distribution, TOC and TN contents of suspended and bed sediments. SPM shows a relatively homogeneous textural composition composed

mainly by particles with a diameter lower than 31 μm (mode 11 ± 3.5 μm) dominated by silts (70 ± 8.8%) and clays (30 ± 8.9%), with a significant reduction of the mode towards the river mouth (from > 13 to ~ 10 μm; *p* < 0.05) due to progressive deposition of coarser fractions during downstream transport (Walling and Moorehead 1989). As expected, bed sediments have a coarser textural composition (mode 72 ± 26 μm), with predominance of silts (53 ± 22%) and sands (36 ± 28%), especially in the upper Salado basin (stations 1–5, sands: 42–89%) and lower clay contents (10 ± 7.2%) that tends to increase downstream (*p* < 0.05), supporting the progressive gravitational settling deduced for SPM.

The TOC and TN content of SPM were relatively high (TOC: 7.4 ± 2.5%; RSD=33%; TN: 1.1 ± 0.4%, RSD=39%), with a decreasing trend towards the river mouth (*p* < 0.001; Fig. 3). Interestingly, TOC shows a very significant positive correlation with Chl *a* (*r* = 0.88; *p* < 0.0001; Fig. 5) suggesting that most of the suspended organic carbon comes from primary production in the river. This is supported by the low and homogeneous elemental ratio of TOC to TN (C/N: 7.7 ± 0.8; RSD = 11%), which is widely used as a proxy to

Fig. 3 Grain size composition, TOC and TN in suspended and bed sediments



elucidate the source of organic matter in aquatic environments (Thornton and McManus 1994), and confirms a prevailing phytoplanktonic TOC source (i.e., C/N: 6–8; Wu et al. 2007).

TOC and TN contents of bed sediments were significantly lower and more variable than in SPM (TOC:  $1.4 \pm 1.2\%$ , RSD = 82%; TN:  $0.15 \pm 0.10\%$ , RSD = 68%), reaching maximum values in the central sector of the basin (TOC: 2.3–3.9%; TN: 0.2–0.4 at stations 7–12). The C/N ratio for bed sediments was also variable ( $11 \pm 5.9$ ; RSD = 56%), with maximum values at the central stations (C/N 11–19)

suggesting a significant contribution of allochthonous organic matter.

In contrast with the general SPM pattern of the basin, the high C/N ratio (10.9) and low Chl *a* (20 µg/l) registered in Chivilcoy stream suggest allochthonous inputs of organic matter, confirming the abnormal character of this tributary. In fact, the high concentrations of coprostanol (fecal biomarker produced by microbial cholesterol reduction in human gut; Bull et al. 2002) registered in suspended and bed sediments of this stream (1349 and 29 µg/g, respectively; Heguilor et al. 2019) confirms fresh sewage inputs from the

**Table 1** Metal concentrations (µg/g, Fe: mg/g) in suspended (SPM) and bed sediments

Stations		DWD (km)	SPM							Bed sediments						
			Fe	Mn	Zn	Cu	Pb	Cr	Ni	Fe	Mn	Zn	Cu	Pb	Cr	Ni
<b>Lakes</b>																
1-	El Chañar	0	21	1063	71	15	29	13	8.6	9.5	393	31	8.4	3.9	4.3	2.6
2-	Mar Chiquita	50	17	946	62	10	22	12	7.6	6.8	125	15	3.4	2.1	3.1	1.8
3-	Gómez	70	17	873	64	10	23	11	7.3	12	178	22	6.2	3.6	4.2	2.7
5-	Carpincho	72	18	963	65	10	24	11	7.7	15	231	33	8.7	6.3	5.8	3.4
7-	Bragado	140	14	1153	33	9.4	25	9.2	5.9	16	909	42	12	5.4	6.1	4.8
<b>Tributary streams</b>																
10-	Chivilcoy	165	18	1503	102	26	47	18	6.3	9.1	842	190	64	29	13	3.7
13-	Saladillo	230	18	1099	40	11	15	11	6.6	11	270	28	7.0	4.6	4.1	2.9
14-	Las Flores	280	11	1704	15	8.5	10	7.6	3.8	17	386	31	11	4.8	5.8	4.6
<b>Main channel</b>																
4-	Junín	71	18	953	45	11	21	11	7.2	12	135	26	8.9	4.0	3.9	3.4
6-	RN7 O'Higgins	87	17	876	55	11	24	12	7.2	12	376	42	9.8	6.0	5.0	3.1
8-	RN5 Alberti	150	15	770	48	10	22	10	6.5	8.9	321	33	9.1	4.8	4.4	3.1
9-	RP51 Chivilcoy	160	15	904	46	10	22	10	6.0	10	423	37	9.7	5.2	4.7	3.8
11-	RP30 N. Riestra	200	16	860	48	10	22	10	6.2	16	497	47	12	6.6	6.4	5.2
12-	RN205 R. Pérez	240	15	787	47	10	20	11	6.1	13	391	38	10	6.1	5.6	4.1
15-	RN3 Monte	290	21	926	47	13	20	15	7.8	18	297	33	12	5.5	5.7	5.0
16-	General Belgrano	325	21	1057	49	12	20	14	7.9	18	305	34	12	5.7	5.4	5.2
17-	RP57 Lezama	375	21	1161	37	12	18	13	7.5	22	393	44	15	6.3	6.0	6.3
18-	RP2 Guerrero	390	21	1160	40	12	22	12	7.7	22	472	46	14	8.3	6.4	6.2
19-	Salado mouth	415	38	1756	57	18	38	34	15	32	734	63	20	14	13	13
20-	Aliviador channel	416	36	1427	57	16	34	33	15	36	830	72	22	15	14	14
21-	Channel 15	420	23	1424	42	15	21	16	8.6	18	417	31	7.6	5.6	5.1	4.3
<b>Basin grand mean</b>			20	1112	51	12	24	14	7.8	16	425	45	14	7.3	6.2	4.9
<b>SD</b>			6.5	290	17	3.8	7.8	6.9	2.7	7.3	226	36	12	5.9	2.9	3.1
<i>World Rivers SPM<sup>a</sup></i>			58	1679	208	76	61	130	75							
<i>Samborombón Bay<sup>b</sup></i>			48	1003	109	87	ND	35	76	33	587	81	23	15	21	15
<i>SQGs<sup>c</sup></i>																
<i>ISQG</i>										<i>n/a</i>	<i>n/a</i>	123	35.7	35	37.3	<i>n/a</i>
<i>PEL</i>										<i>n/a</i>	<i>n/a</i>	315	197	91.3	90	<i>n/a</i>

DWD downstream distance (km)

<sup>a</sup>Viers et al. (2009)

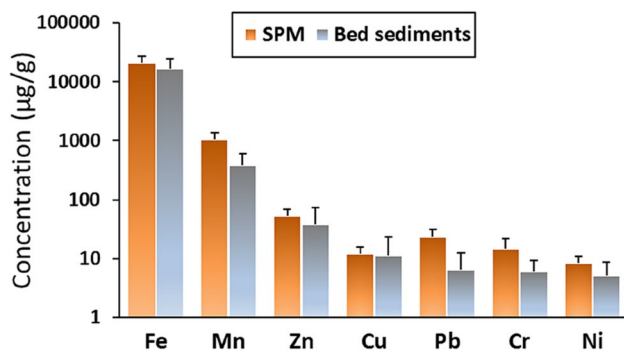
<sup>b</sup>Tatone et al. (2015)

<sup>c</sup>CCME (2001)

city of Chivilcoy, despite the fact that the city has an operational wastewater treatment plant.

### Metals in suspended and bed sediments

Table 1 presents metal concentrations in SPM and bed sediments. As expected, SPM values are consistently higher (1–4 times) than those of bed sediments, reflecting the finer nature of the material transported in suspension with respect to that deposited in the bottom (Fig. 4). The average SPM/bed sediment metal ratio decreased in the following order: Pb (4.3) > Mn (3.3) > Cr (2.5) > Ni (1.9) > Zn (1.6) > Fe (1.4) > Cu (1.2). However, metal concentrations in SPM were relatively homogeneous (RSD = 18–50%) and 1.5–9.6 times lower than the global average concentrations of riverine suspended sediments reported by Viers et al. (2009)

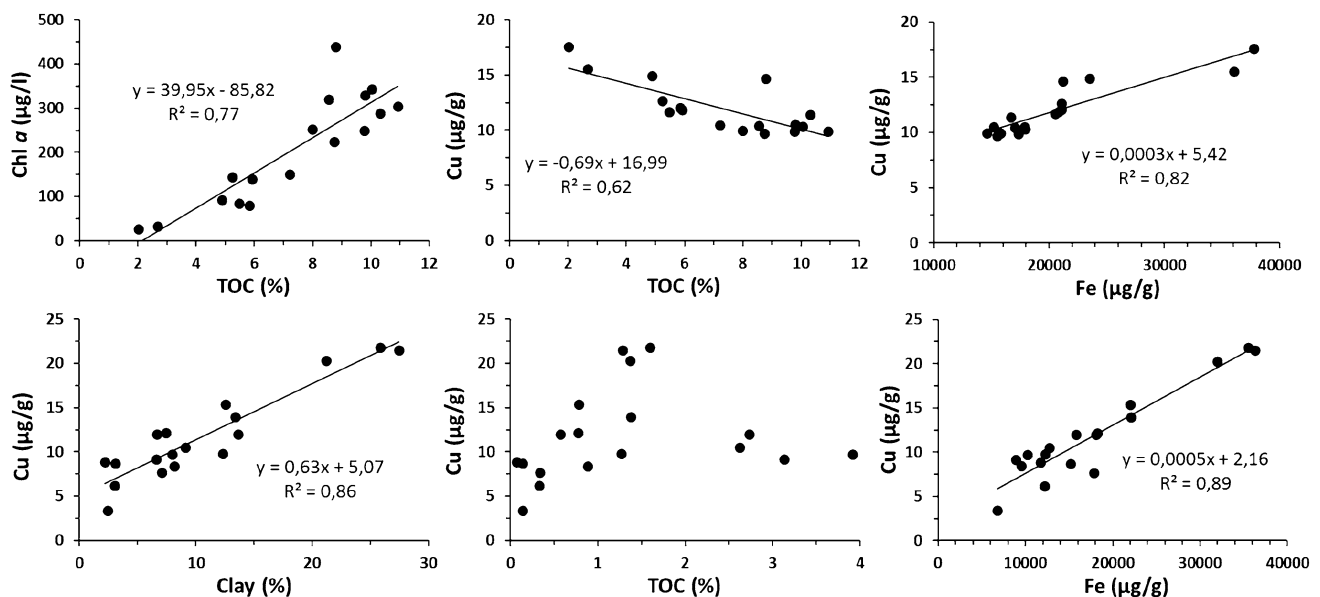


**Fig. 4** Comparison between metal contents in SPM and bed sediments. Error bars indicate the standard deviation

(Table 1). The exception was the Chivilcoy stream, where Zn, Cu and Pb concentrations (102, 26 and 47 µg/g, respectively) doubled the average values of the basin (Table 1). Metal concentrations in bed sediment were more variable (RSD = 37–67%) and showed a general increasing trend towards the river embouchure following the enrichment of clays. Consistent with SPM data, bed sediments of the Chivilcoy stream showed maximum concentrations of Zn, Cu and Pb (190, 64 and 29 µg/g, respectively). Metal concentrations in suspended and bed sediments of the Salado River were generally lower (2–10 times) than those reported previously by Tatone et al. (2015) in the Samborombón Bay (Table 1), confirming that Salado River is not a relevant source of metals to the Bay.

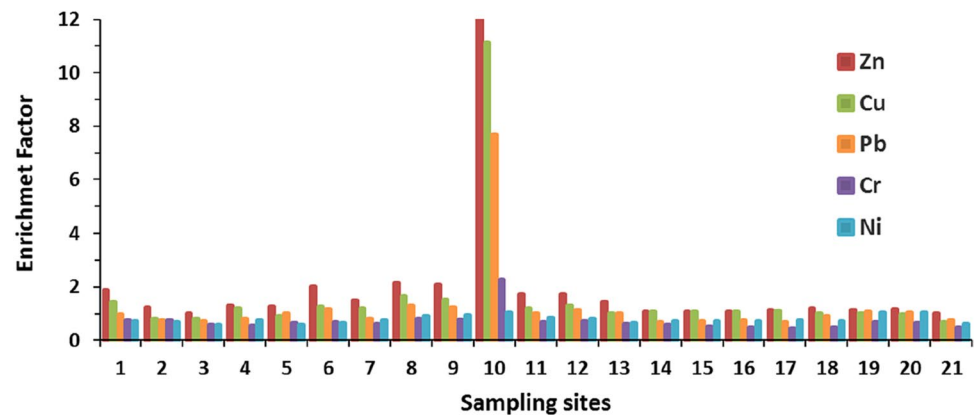
Spearman's correlation analysis revealed that, excepted Zn and Pb, suspended metals from Salado River display a significant positive correlation among them ( $r = 0.80$ – $0.94$ ) and a negative correlation with TOC contents of SPM ( $r = -0.58$ – $-0.76$ ; Fig. 5), suggesting a prevailing association of metals with the lithogenic matrix rather to autochthonous organic matter derived from phytoplankton. For bed sediments, all metals show a strong correlation among them ( $r = 0.82$ – $0.99$ ) and with the clay content ( $r = 0.89$ – $0.97$ ) indicating a similar distribution of metals, strongly influenced by the distribution of clays. Given the homogeneity of these patterns, only selected relationships are shown in Fig. 5.

To further evaluate the potential ecological risks, metal concentrations in bed sediments were compared with the Canadian sediment quality guidelines for the protection of aquatic life (CCME 2001). Metal concentrations in the Salado River and its main tributaries were generally well



**Fig. 5** Relationships of physicochemical parameters and selected metals for SPM (above) and bed sediments (below)

**Fig. 6** Enrichment factors of trace metals in bed sediments relative to local background values



below ISQG values (Table 1), suggesting no risks for adverse biological effects. As expected, the sole exception is the Chivilcoy stream whose Zn and Cu concentrations range between ISQG and PEL, indicating that occasional adverse effects may occur for aquatic biota.

### Enrichment factor and Geoaccumulation index

Most enrichment factors (EF) for Zn, Cu, Pb, Ni and Cr in the mainstream are lower than 1.5 ( $1.4 \pm 0.4$ ;  $1.1 \pm 0.3$ ;  $0.9 \pm 0.2$ ;  $0.8 \pm 0.1$  and  $0.6 \pm 0.1$ , respectively), confirming prevailing natural sources. In contrast to the previously discussed general increasing trend of metal concentrations towards the river mouth, EF values showed a slight increase in the middle sector relative to the rest of the basin, consistent with the TOC pattern, supporting some degree of anthropogenic influence (Fig. 6). This was particularly noticeably in the Chivilcoy stream whose EFs show high values for most metals (especially for Pb, Cu and Zn;  $EF = 8-12$ ), reflecting enrichment relative to background concentrations. Thus both the abnormal organic values and metal concentrations at this site indicate that mixed sewage and industrial anthropogenic sources contribute to this polluted tributary.

The geoaccumulation index (Igeo), calculated with local background metal levels, was in general low for all trace metals ( $I_{geo} \leq 0$ ), supporting the prevalence of unpolluted sediments in the Salado River and its main tributaries. Also, in agreement with the other indexes, the Igeo indicate a higher degree of pollution ( $0 \leq I_{geo} \leq 1$ ) for Zn, Cu and Pb in the Chivilcoy stream.

### Conclusions

This study presents the first comprehensive evaluation of water quality and metal geochemistry in suspended and bed sediments of the Salado River, the main river basin of the Buenos Aires province contributing to the outer Río de la Plata estuary. Water quality parameters displayed a

decreasing gradient towards the river mouth reflecting the change in land use and progressive particle sedimentation along the river. Due to smaller grain-size distribution, SPM was enriched in total organic carbon, nitrogen and metals, but with metal concentrations almost an order of magnitude lower than the global river average, reflecting reduced anthropogenic impact and the predominance of phytoplanktonic material in the SPM. Consistently, bed sediments also show low metal concentrations compared to background concentrations with a spatial distribution controlled by the clay content which increase downstream. None of the two main tributaries, the Saladillo-Vallimanca and Las Flores streams, contribute significant metal load to the Salado River. The Chivilcoy stream is the sole exception in the basin; organic and metal data reflect an abnormal situation with a significant enrichment of metals indicating that mixed sewage and industrial anthropogenic sources contribute to this polluted tributary. Continuous monitoring programs would be useful to assess the impact of organic and metal discharges under the highly variable hydrologic conditions of the Salado River.

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s12665-021-09624-4>.

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