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# Heavy metals and trace elements in muscle of silverside (*Odontesthes bonariensis*) and water from different environments (Argentina): aquatic pollution and consumption effect approach

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## HIGHLIGHTS

- High concentrations of As, Pb, Cd, Hg and Zn were found in the water bodies studied.
- High concentrations of As, Hg and Pb were found in silverside muscle.
- As, Hg and Cd in water were above the recommended levels for the Biota Protection.
- A fraction of the population may consume fish contaminated with Hg and Pb.

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## ABSTRACT

The concentrations of As, Ag, B, Ba, Bi, Ca, Cd, Co, Cr, Fe, Ga, Hg, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sb, Se, Sn, Sr, Te, Ti, U, V and Zn were determined in water and silverside (*Odontesthes bonariensis*) muscle samples from four important commercial fishing sites (Argentina) by ICPMS. Trace element concentrations in water with well-documented human health effects were above the recommended maximum levels established by Argentinean and international guidelines for the aquatic biota protection in three sampling sites (e.g. As: 28.4–367 μg $L^{-1}$; Cd: 0.17–1.05 μg $L^{-1}$; Hg: 0.07–0.63 μg $L^{-1}$; Zn: 71.3–90.0 μg $L^{-1}$). High concentrations of As, Hg and Pb (0.03–0.76; 0.03–0.42 and 0.04–0.19 mg $kg^{-1}$ wet weight, respectively) were found in silverside muscle. Fishing communities associated with contaminated environments are likely to have higher consumption rates and are thus more likely to be exposed to higher concentrations of the toxic elements (As, Hg and Pb).

## 1. Introduction

Commercial fish species inhabiting polluted water bodies worldwide are exposed to a wide range of contaminants that may accumulate in tissues and reach harmful levels (e.g. Yousuf et al., 2000; Mol, 2011). In particular, the accumulation of heavy metals (some of which are highly toxic) and arsenic have been shown to depend on different factors such as the target tissue, fish species and water quality (Carriquiriborde and Ronco, 2008; Visnjic-Jeftic et al., 2010).

The trace elements found in the surface water of the South American Chaco–Pampean Plain are of natural origin (e.g. As, V, Mo and U) and anthropic origin (e.g. Pb, Hg, Cr) (Schenone et al., 2007; Rosso et al., 2013; Schenone et al., 2014). Heavy metal pollution in surface water bodies (e.g. lagoons, lakes and rivers) from this region may be mainly caused by increasing agricultural and industrial activities along with urban expansion. In this context, it becomes necessary to assess the heavy metal contents in commercial fish species and the resulting potential effects for consumers.

The silverside (*Odontesthes bonariensis*) is a native species of Argentina, Uruguay, Brazil and Chile and introduced in Europe and Asia (Chiba et al., 1989; Gandolfi et al., 1991; Brian and Dyer, 2006). In

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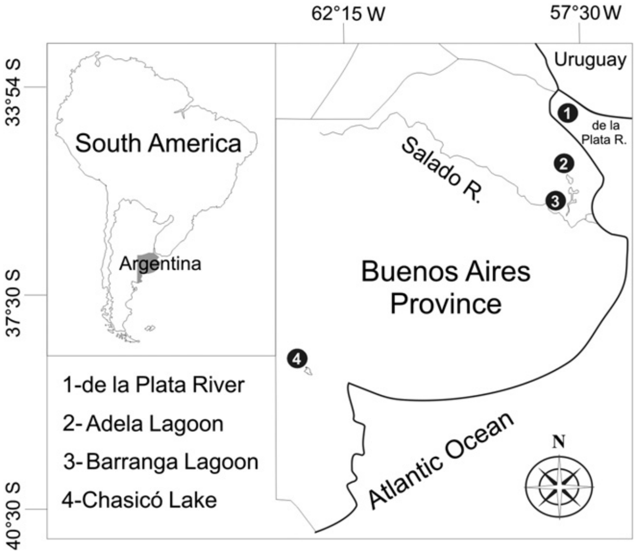

**Fig. 1.** Sampling sites of the silverside, *Odontesthes bonariensis*, Argentina.

Argentina it can be found in the lower section of the Plata Basin and in lentic inland water bodies (e.g. ponds and lagoons) (Brian and Dyer, 2006; Avigliano and Volpedo, 2013) from the Pampean plain (Fig. 1). The commercial and sport fishing of the silverside make it an economically important species (López et al., 2001). In particular, it represents the second most important fishery resource for Argentina and Uruguay with a large proportion of the production being marketed for consumption locally and exported to Europe (Italy, Netherlands and Ukraine), Russia and the United States of America (Minagri, 2014). Nowadays, an increasing number of consumers, policymakers, and managers require information on contaminants in biota to assess the health and well-being of organisms and consumers (Burger et al., 2007). Based on the above considerations, the objective of the present study was to determine the content of trace elements, including toxic metals and arsenic, in muscle of silversides from different aquatic environments, analyzing the relation to human exposure and to present a baseline for this important region.

## 2. Materials and methods

### 2.1. Study area

The region under study (Fig. 1) includes different environments where the silverside is one of the most important commercial and recreational fish species. The de la Plata River (35° 3'5.09"S-57° 2'47.79"W) is a 300-km wide estuary that discharges into the Atlantic Ocean (Guerrero et al., 2010). The salinity ranges from 0.04 to 25 g L$^{-1}$ (north–south), and the maximum depth ranges from 5 to 25 m (Guerrero et al., 2010). The Barrancas (35°52'35.29"S-58°3'19.88"W) and Adela (35°40'51.47"S-58°0'4.79"W) Lagoons (Pampean plain, Buenos Aires, Argentina) are shallow alkaline ecosystems with circulation pattern characteristics of polymictic lakes due to the nearly continuous vertical mixing that promotes a high concentration of suspended particulate matter and low transparency (Miretzky and Fernández Cirelli, 2014). It has a maximum depth of 2 m and salinity of less than 1 g L$^{-1}$. The Chasicó Lake (38°36'39.41"S-63°4'48.68"W) is located in a transition zone between the Pampean and Patagonian regions. It has a maximum depth of 16 m and salinity above 27 g L$^{-1}$ (Kopprio et al., 2010).

### 2.2. Samples collection and preparation

Water and fish samples were collected during August 2011 according to standard procedures (Fig. 1). Surface water samples for elements determination were collected manually at 0.5 m depth with 0.5 L polyethylene-tereftalate bottles. Samples were taken by triplicate and acidified to 0.2% (v/v) ($pH < 2$) with nitric acid (Merck® Pro Analysis) in darkness at 4 °C and immediately transported to the laboratory. Subsequently, these were vacuum filtered through acid-treated Whatman GF/F glass fiber filters (0.45 mm). These samples were stored in darkness at 4 °C up to the analytical treatment. Collection, preservation, preparation, pretreatment and analyses of water samples were conducted according to APHA (1995).

Adult fish were collected overnight using a 3-layer 3 × 3 cm mesh net of 10 m length. Once the nets were recovered, fish were killed by percussive stunning (Van de Vis et al., 2003) and transported on ice (in individual Ziploc® bags) to the laboratory. Fish were randomly selected for analysis considering commercial consumption sizes. Fish sizes are shown in Table S1 (Supplementary material). The axial muscle, below the pectoral fin (±6 g), of each specimen was dissected with a decontaminated ceramic knife. All laboratory tools were soaked in 10% $HNO_3$ for 48 h, rinsed five times with distilled water, and then five times with ultra pure Milli-Q water (Turkmen and Ciminli, 2007). After dissection, the muscle tissue was weighed using an electronic balance (±0.1 g) and freeze-dried.

### 2.3. Determination of element concentrations by ICP-MS

Silverside muscle and water samples were submitted to the Analytical Chemistry Laboratory within the Institute of Chemistry, Karl-Franzens-University, Graz, Austria, for chemical analysis. Element concentrations (As, Ag, B, Ba, Bi, Ca, Cd, Co, Cr, Fe, Ga, Hg, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sb, Se, Sn, Sr, Te, Ti, U, V and Zn) were determined by inductively coupled plasma mass spectrometer (ICP-MS), using an Agilent 7500 (Agilent, Waldbronn, Germany) equipped with a Micro Mist nebulizer (Glass Expansion, Melbourne, Australia) and a Scott double-pass spray chamber.

Water samples were analyzed with no previous digestion.

The freeze-dried muscle samples were pulverized in a coffee-mill, and digested in a microwave digestion system (Milestone ultraCLAVE III, EMLS, Leutkirch, Germany), using 5 ml of concentrated nitric acid (Merck, Darmstadt, Germany), before being analyzed by ICP-MS. Fish muscle contents were measured for dry tissue. However, in order to compare these values with the recommended and suggested guidelines, they were corrected to wet weight with a conversion factor of 0.58%.

### 2.4. Quality assurance and quality control

The water samples were analyzed by triplicate. A water standard reference material (SRM 1643e; National Institute of Standards and Technology, NIST, USA) was analyzed to support quality assurance and control (QA/QC) of water sample measurements.

Furthermore, each sample of dried muscle was divided into three parts. These were digested separately and analyzed in triplicate by ICP-MS. Muscle standard reference material (DORM-2; National Research Council of Canada) was digested in quadruplicate and analyzed to support the QA/QC of fish muscle measurements.

Replicate analysis of these reference materials showed good accuracy, with recovery ranging from 82% to 124% and 81% to 112% for water and muscle, respectively (Table S2, Supplementary material).

A blank was run to correct the all measurements (for water and muscle). The water used throughout the present study was obtained from a Milli-Q Academic water purification system (Millipore GmbH, Vienna, Austria) with a resistivity of 18.2 MOhm*cm. Pro-analysis reagents were used throughout the study (Scheer et al., 2012). For both water and fish muscle, for every nine samples, a procedure blank and spike sample, involving all reagents, was run to check for interference and cross-contamination. Triplicate analyses of blanks, spike sample and reference materials differed from each other within an acceptable range of ±25%.

The sample results were reviewed and evaluated in relation to the quality-assurance/quality-control samples worked up at the same time. Reported results have been corrected for the blanks.

The detection limits for water samples (LOD) in μg L$^{-1}$ based on three times the standard deviation of the blank signal was 0.01 for U; 0.002 for Bi and Li; 0.004 for Sb and Ti; 0.005 for Sn; 0.01 for Ag, Be and Mo; 0.02 for B, Cd, Co, Pb and V; 0.03 for As, Ba, Ga, Sr and Rb; 0.05 for Te; 0.07 for Cr and Hg; 0.12 for Ne; 0.2 for Ni; 0.3 for Fe, Mg and Se; 0.5 for Zn; for 3 Ca and Na;10 for K. The quantification limits (LOQ) in μg L$^{-1}$ based on the lowest calibration point was 0.003 for U; 0.01 for Sb, Ti; for 0.04 Mo; 0.06 for Pb and V; 0.09 for Rb, Sn and Sr; 0.4 for Mn and Te; 0.5 for Ni; 0.8 Se; 1 for Mg and Zn; 8 for Na. The LOD in μg Kg$^{-1}$ for muscle samples was 0.2 for U; 0.5 for Bi and Li; 0.8 for Sb and Ti; 3 for Be, Ag, Mo; 4 for Cd, V and Pb; 5 For Co; 8 for As, Ba, Sn, Rb and Sr; 14 for Cr, Ga and Hg; 24 for Mn and Te; 40 for B and Ni; 60 for Fe, Se; 90 for Mg and Zn; 650 for Ca and Na; 2000 for K. The LOQ in μg Kg$^{-1}$ for muscle samples was 1 for U; 2 for Bi, Li, Sb and Ti; 4 for Ag and Be; 8 for Mo; 15 for Cd, Co, Pb and V; 18 for Sn and Rb; 24 for As, Ba and Sr; 40 for Ga Cr, and Hg; 70 for Mn and Te; 108 for Ni; 150 for B and Fe; 170 for Se; 250 for Mg and Zn; 2000 for Ca and Na; 6300 for K.

### *2.4.1. Statistical analysis*

The Kruskal–Wallis non-parametric, one-way analysis of variance was used to compare the fish size among sampling sites. Furthermore, the Kruskal–Wallis test was used to compare the concentration of each element between sites. Differences were considered to be statistically significant at $p < 0.05$. The statistical analyses were performed using the InfoStat® software. All values were expressed as mean ± standard deviation.

**Table 1**
Concentration (means ± SD in μg L$^{-1}$ and mg L$^{-1*}$) of trace elements in water.

| | De La Plata River | Adela Lagoon | Barrancas Lagoon | Chasicó Lake |
|---|---|---|---|---|
| Ag | 0.10 ± 0.00$^{a}$ | 0.08 ± 0.02$^{ab}$ | 0.12 ± 0.01$^{ab}$ | 1.17 ± 0.07$^{b}$ |
| As | 3.90 ± 0.40$^{a}$ | 28.4 ± 1.10$^{ab}$ | 42.9 ± 1.70$^{ab}$ | 367 ± 2.02$^{b}$ |
| B | 123 ± 11.0$^{a}$ | 364 ± 57.0$^{b}$ | 475 ± 12.0$^{b}$ | 8425 ± 79.0$^{c}$ |
| Ba | 51.4 ± 0.52$^{a}$ | 67.6 ± 0.90$^{b}$ | 58.2 ± 1.30$^{b}$ | 118 ± 2.00$^{c}$ |
| Bi | 0.02 ± 0.00$^{a}$ | 0.02 ± 0.01$^{a}$ | 0.07 ± 0.04$^{a}$ | 0.03 ± 0.03$^{a}$ |
| Ca* | 9.93 ± 0.18$^{a}$ | 33.4 ± 0.50$^{b}$ | 25.9 ± 0.50$^{b}$ | 65.1 ± 1.20$^{c}$ |
| Cd | 0.32 ± 0.00$^{bc}$ | 0.17 ± 0.02$^{a}$ | 0.21 ± 0.01$^{a}$ | 1.05 ± 0.07$^{c}$ |
| Co | 0.43 ± 0.01$^{a}$ | 0.47 ± 0.12$^{a}$ | 0.91 ± 0.10$^{a}$ | 0.47 ± 0.40$^{a}$ |
| Cr | 1.17 ± 0.21$^{a}$ | 1.51 ± 0.76$^{a}$ | 2.34 ± 1.12$^{a}$ | 6.70 ± 0.75$^{a}$ |
| Fe | 681 ± 14,0$^{a}$ | 57.8 ± 2.20$^{ab}$ | 79.4 ± 9.40$^{ab}$ | 50.7 ± 22.5$^{b}$ |
| Ga | 0.08 ± 0.02$^{a}$ | ND | 0.04 ± 0.02$^{a}$ | 0.10 ± 0.00$^{a}$ |
| Hg | 0.07 ± 0.01$^{a}$ | 0.13 ± 0.03$^{b}$ | 0.29 ± 0.09$^{b}$ | 0.63 ± 0.24$^{c}$ |
| K* | 5.71 ± 0.28$^{a}$ | 25.5 ± 0.70$^{b}$ | 37.1 ± 1.01$^{b}$ | 241 ± 3.01$^{c}$ |
| Li | 2.92 ± 0.10$^{a}$ | 20.8 ± ± 0.30$^{b}$ | 22.7 ± 0.50$^{b}$ | 69.2 ± 0.60$^{c}$ |
| Mg* | 8.62 ± 1.16$^{a}$ | 29.2 ± 2.30$^{b}$ | 50.7 ± 8.60$^{b}$ | 815 ± 23.0$^{c}$ |
| Mn | 25.6 ± 0.50$^{c}$ | 3.42 ± 0.17$^{a}$ | 17.9 ± 0.30$^{bc}$ | 12.2 ± 2.50$^{b}$ |
| Mo | 1.87 ± 0.29$^{a}$ | 30.2 ± 0.20$^{b}$ | 33.3 ± 0.70$^{b}$ | 92.7 ± 1.10$^{c}$ |
| Na* | 104 ± 2.00$^{a}$ | 443 ± 32.0$^{b}$ | 764 ± 8.00$^{b}$ | ND |
| Ni | 3.97 ± 0.74$^{a}$ | 3.54 ± 0.61$^{a}$ | 6.05 ± 0.47$^{b}$ | 4.49 ± 0.31$^{a}$ |
| Pb | 2.48 ± 0.04$^{a}$ | 1.40 ± 0.10$^{ab}$ | 1.47 ± 0.11$^{ab}$ | 0.91 ± 0.17$^{b}$ |
| Rb | 4.13 ± 0.11$^{a}$ | 6.59 ± 0.03$^{ab}$ | 8.40 ± 0.02$^{ab}$ | 61.7 ± 0.90$^{c}$ |
| Sb | 0.57 ± 0.02$^{ab}$ | 0.47 ± 0.01$^{a}$ | 0.55 ± 0.02$^{ab}$ | 1.26 ± 0.08$^{c}$ |
| Se | 1.40 ± 0.35$^{a}$ | 4.70 ± 1.17$^{ab}$ | 5.47 ± 0.27$^{ab}$ | 75.9 ± 2.70$^{b}$ |
| Sn | 0.01 ± 0.00$^{a}$ | 0.05 ± 0.01$^{a}$ | 0.12 ± 0.03$^{a}$ | ND |
| Sr | 98.8 ± 2.90$^{a}$ | 403 ± 4.01$^{b}$ | 426 ± 4 .01$^{b}$ | 4561 ± 46.0$^{c}$ |
| Te | ND | ND | ND | 0.50 ± 0.15 |
| Ti | ND | ND | ND | ND |
| U | 0.81 ± 0.08$^{a}$ | 14.1 ± 0.20$^{b}$ | 17.9 ± 0.30$^{b}$ | 30.6 ± 0.50$^{c}$ |
| V | 6.22 ± 0.32$^{a}$ | 33.4 ± 0.60$^{b}$ | 41.9 ± 1.20$^{b}$ | 290 ± 2.00$^{c}$ |
| Zn | 81.2 ± 13.0$^{a}$ | 59.4 ± 12.1$^{a}$ | 71.0 ± 51.0$^{a}$ | 90.0 ± 0.70$^{a}$ |

a, b, c: Different letters indicate statistical significant differences ($p < 0.05$); ND: not detected.

## 3. Results and discussion

No significant differences in fish size between sampling sites were found ($p > 0.07$). In environmental studies As, Co, Zn, Cr, Mn, Fe, Pb, Cd, Ni, Se, Hg and Ag are the most studied and significant elements regarding environmental impact and human health (Copat et al., 2012; Schenone et al., 2014). Less studied elements in fish tissues like Li, B, Al, V, Ga, Rb, Sr, Mo, Sn, Sb, Te, Ba, Ti, Bi, and U (e.g. Eisler, 1994; Hothem et al., 2007; Schenone et al., 2014) are highly useful to understand the anthropic effects on wild fish from an environmental point of view.

### *3.1. Aquatic pollution*

High concentrations of trace elements related to adverse health effects such as As, Pb, Cd, Hg and Zn were found in the water bodies studied (Table 1).

The As concentration was significantly higher in water at Chasicó Lake, while Pb was higher in water from de la Plata River. Based on the Argentinean National Guidelines for the Aquatic Biota Protection (NGABP), the As concentration in water from the Chasicó Lake and Adela and Barrancas Lagoons, along with the Pb concentration in water from de la Plata River, were above the recommended levels (15 μg L$^{-1}$ and 1.6 μg L$^{-1}$, respectively). Based on the Canadian Guidelines for the Aquatic Biota Protection, the As concentration was above the recommended maximum level (5 μg L$^{-1}$) in all sampling sites except de la Plata River. Furthermore, the Pb concentration was below the recommended maximum value (1 μg L$^{-1}$) only for the Chasicó Lake.

Even higher levels of As in surface and groundwater were recently reported for Chasicó Lake, reaching up to 166 and 413 μg L$^{-1}$, respectively (Puntoriero et al., 2014a,b). High concentrations of As in water and sediments were also reported for streams and rivers near Chasicó Lake, ranging from 181 to 1134 and 246 to 1031, respectively (Rosso et al., 2011, 2013).

Pb levels in the de la Plata River vary widely among seasons (2 to 58 μg L$^{-1}$). This is likely due to seasonal fluctuations in industrial activity because this area is one of the most densely populated in the country (Guerrero and Kesten, 1994). Regardless, high levels of Pb (120–200 mg kg$^{-1}$) have been observed in sediments of tributaries to de la Plata River (Ronco et al., 2008).

According to the Canadian Guidelines for the Aquatic Biota Protection, the concentrations of Hg, Zn and Cd in water from all sampling sites were above the recommended maximum levels (0.02 μg L$^{-1}$ for Hg; 30 μg L$^{-1}$ for Zn; 1.2 μg L$^{-1}$ for Cd) (Table 1).

The high Hg concentration in water samples from de la Plata River may be a consequence of the large cities located by the river (e.g. Buenos Aires, La Plata and Rosario in Argentina and Montevideo in Uruguay) since a high concentration of this element (1.9–6.8 mg kg$^{-1}$) was found in sediments from different tributaries to de la Plata River (Ronco et al., 2008). Mercury is especially concerning because its inorganic form is biologically transformed in aquatic environments into methylmercury (MeHg), a lipophilic organic compound that bioaccumulates and biomagnifies as it moves up the aquatic food chain (Carrasco et al., 2011; Olmedo et al., 2013). As a result, human populations with an elevated dietary intake have the highest potential exposure to MeHg and are at an increased risk for developing neurotoxic effects. This is a particularly important issue for children, pregnant women and breast-feeding mothers (Olmedo et al., 2013). High concentrations of trace elements related to adverse health effects were recently reported by several authors in different water bodies (Table 2). The levels of As found in Lake Chasicó were extremely high compared to other anthropogenically contaminated environments. Hg levels were also very high compared to other systems such as the Llobregat Delta (Spain) and Lancang River (China). The Lancang River is associated with elevated concentrations of Hg, Pb and Zn due to recent industrial development in China (Zhao et al., 2012). Pb levels in the study sites were relatively low compared to other water bodies as Lancang River

**Table 2**

Concentrations of As, Pb, Hg, Cd and Zn in surface water samples ($\mu g\ L^{-1}$) at each site in this study and concentrations reported for previous studies.

| Water body | Country | *Date | As | Pb | Hg | Cd | Zn | Reference |
|---|---|---|---|---|---|---|---|---|
| De la Plata Estuary | Argentina | 2011 | 3.9 | 2.4 | 0.07 | 0.32 | 81.2 | This paper |
| Adela Lagoon | Argentina | 2011 | 28.4 | 1.4 | 0.13 | 0.17 | 59.5 | This paper |
| Barrancas Lagoon | Argentina | 2011 | 42.9 | 1.5 | 0.29 | 0.21 | 71.3 | This paper |
| Chasicó Lake | Argentina | 2011 | 367 | 0.9 | 0.63 | 1.0 | 90.0 | This paper |
| Chascomus Lagoon | Argentina | 2011 | 22.8 | 8.22 | – | 0.28 | 27.3 | Schenone et al., 2014 |
| Canyon Lake | USA | 2008–2009 | – | 32 | – | – | 9.5 | VanLandeghem et al., 2012 |
| Timok River | Serbia | 2006–2009 | – | 0.05 | 0.11 | 0.11 | 0.01 | Brankov et al., 2012 |
| Borska River | Serbia | 2006–2009 | – | 0.04 | 0.15 | 0.86 | 0.14 | Brankov et al., 2012 |
| Lancang River | China | 2005 | 5.3–21 | 11–32 | 0–0.2 | 0–3.9 | <706 | Zhao et al., 2012 |
| Tigris River | Turkey | 2009 | 23.8 | 5.6 | – | – | 101 | Varol and Şen, 2012 |
| Ganga River | India | 2005–2006 | – | 120 | – | 5 | 60 | Aktar et al., 2010 |
| Pardo River | Brazil | 2011–2012 | 1.93 | 4.10 | <0.2 | 0.06 | 13.14 | Alves et al., 2014. |
| Llobregat Delta | Spain | 2007–2008 | – | 6.06 | 0.37 | 5 | – | Teijon et al., 2010. |
| Bílina River | Czech Republic | 2004–2008 | 14.1 | 7.9 | 0.1 | 0.21 | 35.2 | Kohušová et al., 2011 |

* Date: sampling time.

(China), Ganga River (India) and Canyon Lake (USA), as well as with other lakes located in the region of study (Chascomus Lagoon). The levels of Cd and Zn found in this study far exceed those reported for other systems as Timok River (Serbia), Pardo River (Brazil), and Bílina River (Czech Republic), but are lower compared to Lancang River (China).

For the rest of the elements related to human health (Co, Cr, Ni, Mn, Fe, Se, Ag), no significant differences ($p > 0.05$) were found among the study areas for Co and Cr concentrations in water samples (Table 1). Similar concentration of Cr was previously reported in nearby Adela and Barrancas Lagoons (Schenone et al., 2014). The concentration of Ni was significantly higher at Barrancas Lagoon, but there were no differences between de la Plata River, Chasicó and Adela Lagoon. The Mn and Fe concentrations were highest ($p < 0.05$) in water from de la Plata River in relation to the other sampling sites. It has been observed that the Fe also varies among seasons (280–1310 $\mu g\ L^{-1}$), presumably due to changes in industrial activity (Guerrero and Kesten, 1994). The Table 1 shows that the concentrations of Ag and Se were significantly higher at Chasicó Lake than at de la Plata River. However, there were no differences between the lagoons and Chasicó Lake or with de la Plata River.

No significant differences were found among the study areas for concentrations of Bi (all sites) and Ga (Río de la Plata, Barrancas and Chasicó)($p > 0.05$).

The Ti concentration was below detection limit of the equipment. The Sn concentration for Chasicó was below detection limit, whereas there were no significant differences between the other three sites (Table 1).

The Na concentration for Chasicó Lake was higher than the other three sites and exceeded the upper detection limit of the equipment. Concentration of elements such as B, Ba, Ca, K, Li, Mg, Mo, Rb, Sr, U and V were lowest in de la Plata River, highest in the Chasicó Lake,

**Table 3**

Concentration (means ± SD in $mg\ kg^{-1}$ and $g\ kg^{-1*}$ wet wt) of trace elements in tissues.

| Element | Río de la Plata River | Adela Lagoon | Barrancas Lagoon | Chasicó Lake |
|---|---|---|---|---|
| Ag | $0.03 \pm 0.03^{a}$ | $0.05 \pm 0.08^{a}$ | $0.04 \pm 0.05^{a}$ | $0.08 \pm 0.07^{a}$ |
| As | $0.03 \pm 0.01^{a}$ | $0.09 \pm 0.03^{b}$ | $0.08 \pm 0.03^{b}$ | $0.76 \pm 0.29^{c}$ |
| B | $0.11 \pm 0.04^{a}$ | $0.25 \pm 0.06^{b}$ | $0.30 \pm 0.12^{b}$ | $1.47 \pm 0.40^{c}$ |
| Ba | $0.51 \pm 0.20^{a}$ | $0.62 \pm 0.27^{a}$ | $0.52 \pm 0.09^{a}$ | $0.15 \pm 0.06^{b}$ |
| Bi | $1.32E^{-03} \pm 3E^{-04a}$ | $1.33E^{-03} \pm 4^{-04a}$ | $1.77E^{-03} \pm 3E^{-04a}$ | $1.84E^{-03} \pm 5E^{-04a}$ |
| Ca* | $1.54 \pm 0.82^{b}$ | $1.51 \pm 0.44^{b}$ | $1.15 \pm 0.20^{a}$ | $0.55 \pm 0.24^{a}$ |
| Cd | $0.01 \pm 0.01^{a}$ | ND | ND | $0.02 \pm 0.02^{b}$ |
| Co | $0.01 \pm 0.01^{a}$ | $0.03 \pm 0.01^{b}$ | $0.03 \pm 0.01^{b}$ | $0.01 \pm 0.01^{a}$ |
| Cr | $0.10 \pm 0.05^{a}$ | $0.09 \pm 0.03^{a}$ | $0.05 \pm 0.03^{a}$ | $0.04 \pm 0.02^{a}$ |
| Fe | $7.53 \pm 1.06^{a}$ | $4.06 \pm 0.98^{b}$ | $3.92 \pm 0.37^{b}$ | $4.56 \pm 1.41^{b}$ |
| Ga | ND | ND | ND | ND |
| Hg | $0.30 \pm 0.06^{ab}$ | $0.04 \pm 0.02^{a}$ | $0.03 \pm 0.00^{a}$ | $0.42 \pm 0.05^{b}$ |
| K* | $1.45 \pm 0.82^{a}$ | $1.58 \pm 0.08^{a}$ | $1.36 \pm 0.36^{a}$ | $0.59 \pm 0.33^{a}$ |
| Li | $0.02 \pm 0.00^{a}$ | $0.02 \pm 0.00^{a}$ | $0.02 \pm 0.00^{a}$ | $0.02 \pm 0.01^{a}$ |
| Mg* | $0.73 \pm 0.03^{a}$ | $0.61 \pm 0.07^{b}$ | $0.46 \pm 0.11^{b}$ | $0.73 \pm 0.32^{a}$ |
| Mn | $0.32 \pm 0.09^{a}$ | $0.35 \pm 0.09^{a}$ | $0.41 \pm 0.03^{a}$ | $0.25 \pm 0.04^{a}$ |
| Mo | $0.03 \pm 0.01^{a}$ | $0.04 \pm 0.01^{a}$ | $0.03 \pm 0.01^{a}$ | $0.03 \pm 0.01^{a}$ |
| Na* | $1.95 \pm 0.44^{b}$ | $1.61 \pm 0.24^{a}$ | $1.22 \pm 0.23^{a}$ | $2.59 \pm 0.47^{c}$ |
| Ni | $0.07 \pm 0.07^{a}$ | $0.07 \pm 0.03^{a}$ | $0.08 \pm 0.06^{a}$ | $0.06 \pm 0.03^{a}$ |
| Pb | $0.19 \pm 0.08^{a}$ | $0.09 \pm 0.09^{ab}$ | $0.06 \pm 0.19^{ab}$ | $0.04 \pm 0.03^{b}$ |
| Rb | $20.5 \pm 1.70^{a}$ | $3.25 \pm 0.53^{b}$ | $3.00 \pm 0.94^{b}$ | $3.92 \pm 0.45^{b}$ |
| Sb | $0.01 \pm 0.00^{a}$ | $0.01 \pm 0.00^{a}$ | $0.01 \pm 0.01^{a}$ | $0.00 \pm 0.00^{a}$ |
| Se | $0.55 \pm 0.04^{a}$ | $0.44 \pm 0.08^{a}$ | $0.41 \pm 0.08^{a}$ | $5.28 \pm 0.66^{b}$ |
| Sn | $0.01 \pm 0.00$ | ND | ND | ND |
| Sr | $1.93 \pm 1.12^{a}$ | $3.62 \pm 0.98^{ab}$ | $4.53 \pm 2.41^{ab}$ | $8.02 \pm 4.51^{b}$ |
| Te | ND | ND | ND | ND |
| Ti | $2.59E^{-03} \pm 1E^{-03b}$ | $6.33E^{-06} \pm 1E^{-05a}$ | $3.54E^{-05} \pm 3E^{-05ab}$ | $1.14E^{-05} \pm 1.58E^{-05a}$ |
| U | $8.86E^{-04} \pm 5.70E^{-04a}$ | $2.22E^{-03} \pm 5.76E^{-04b}$ | $2.47E^{-03} \pm 4.49E^{-04b}$ | $2.47E^{-03} \pm 1.77E^{-03b}$ |
| V | $0.02 \pm 0.01^{a}$ | $0.03 \pm 0.01^{a}$ | $0.03 \pm 0.01^{a}$ | $0.18 \pm 0.07^{b}$ |
| Zn | $12.0 \pm 2.90^{a}$ | $14.0 \pm 4.2^{a}$ | $12.6 \pm 2.60^{a}$ | $18.4 \pm 3.40^{a}$ |

a, b, c: Different letters indicate statistical significant differences ($p < 0.05$); ND: not detected.

* = mg L– 1

and intermediate in the Adela and Barrancas Lagoons (Table 1). High concentrations of V in surface water were reported for Chasicó Lake, reaching up to 366 to 413 μg $L^{-1}$ (Puntoriero et al., 2014b).

In the absence of industrial pollution, the presence of As, Mo, V and U in Chasicó Lake cannot be attributed to anthropic sources. Alternatively, these trace elements originate from volcanic glass from Pampean loess (Teruggi, 1957). Weathering of sediments of Chasicó Lake and the evaporation–crystallization process could be one of the factors controlling the chemical composition of the water of the Lake (Puntoriero et al., 2014b). Further studies are required in order to analyze the geochemical processes determining the chemical composition of the Chasicó Lake.

### 3.2. Toxic metals and trace elements in muscle

High concentrations of trace elements related to adverse health effects, such as As, Pb and Hg (Table 3), were found in the silversides. These elements are classified as toxic due to their lack of metabolic functions and the harm they cause in humans when ingested over a long period, even at low concentrations (Somers, 1974). The muscle sample results showed significant concentrations of As and Hg in silversides from the Chasicó Lake as well as significant concentrations of Pb in silversides from de la Plata River (Table 3). Considerably high levels of Zn (all sites) and Cd (Chasicó Lake) were also found (Table 3). The trends of As, Cd, Hg and Pb observed in muscle are consistent with those found in water (Table 1).

The concentrations of B, V and Se in silversides from the Chasicó Lake were higher ($p < 0.05$) than in fish from the other water bodies (Table 3). The U concentrations were low in specimens from de la Plata River as well as in fish from the lagoons and Chasicó Lake.

The geological origin (volcanic) of the study area may explain the high concentrations of As, Se, B, V and U found in water samples from the Chasicó Lake, resulting in a high concentration of these elements in silverside muscle (Rosso et al., 2011, 2013).

The Ca concentration was highest in silversides from de la Plata River and lagoons ($p > 0.05$), while low in fish from Chasicó Lake ($p < 0.05$). The Rb and Fe concentrations were highest in silversides from de la Plata River ($p < 0.05$) as well as in fish from the lagoons and the Chasicó Lake ($p > 0.05$). No significant differences ($p > 0.05$) were found among the study areas in the content of Ag, Sb, Bi, K, Ni, Mn, Mo, Cr and Zn in muscle samples. The concentration of Co and Mg in muscles was higher for fish from the Adela and Barrancas Lagoons, while Ti and Sn concentrations were higher in silversides collected from de la Plata River. The concentration of Ga and Te in muscle samples was low in all the studied sampling points. The concentration of Ba was lowest in fish from the Chasicó Lake. The levels of Sr were lowest in de la Plata River, highest in the Chasicó Lake and intermediate in the Adela and Barrancas Lagoons (Table 3).

Our results were compared with those reported by other authors for *O. bonariensis*, other species from the same genus (*Odontesthes*) and other species from related environments (Table 4). As concentration in Chasicó Lake was within the values obtained for *O. bonariensis* in Salado River, but was lower than that of Quequén Salado River, Argentina (Rosso et al., 2013). Both rivers are located in the vicinity of Chasicó Lake and are affected by the same volcanic phenomenon (Rosso et al., 2013). Furthermore, the As concentration in *O. bonariensis* from Chasicó Lake was similar to the values obtained for *Rhamdia quelen* in Quequen Salado River (Rosso et al., 2013). On the other hand, the As concentration in *O. bonariensis* from Río de la Plata, Adela and Barrancas was similar to the values obtained for *O. microlepidotus* in Negro River (Viana et al., 2005), which is not naturally contaminated with this element.

The Chascomús Lagoon is connected to Adela and Barrancas Lagoons in high water periods. The concentration of Pb in *Parapimelodus valenciennis* from Chascomús Lagoon was higher than our results in *O. bonariensis* while the concentration of As was found to be lower (Schenone et al., 2014). Hg concentration in *O. bonariensis* from De la Plata estuary and Chasicó Lake were within the values obtained for *Odontesthes sp.* in de la Plata River estuary (Viana et al., 2005) and for *O. microlepidotus* in Río Negro River (Arribere et al., 2003). In *Mugil platanus* inhabiting de la Plata River estuary, we observed similar concentrations of Hg in this study (Marcovecchio and Moreno, 1993).

### 3.3. Toxic elements and consumption effect

The main adverse effects reported to be associated with long term ingestion of inorganic arsenic in humans are skin lesions (e.g. chronic arsenicism), cancer, developmental toxicity, neurotoxicity, cardiovascular diseases, abnormal glucose metabolism and diabetes (EFSA, 2009). As early as the beginning of the century physician noted an increased incidence of clinical skin alteration and high risk of lung and kidney cancers in patients from Argentina (e.g. Córdoba Province). The high arsenic content of drinking water from wells in this region was found to be the cause (Steinmaus et al., 2010; Pou et al., 2011; Aballay et al., 2012).

On the other hand, lead-associated neurotoxicity was found to affect central information processing, cause psychiatric symptoms, and impair manual dexterity. In children, high lead levels in blood are inversely associated with a reduced intelligence quotient (IQ) (EFSA, 2010). The adverse effects associated with the ingestion of mercury and methylmercury are ataxia, dysarthria, constriction of visual fields, impaired hearing, cerebral palsy and severe developmental retardation in prenatally exposed children as well as sensory disturbance as a symptom of fatal methylmercury poisoning in exposed laboratory workers (EFSA, 2012).

The levels of As, Pb and Hg were below the recommended maximum levels established by the Argentinean Food Codex (1 mg $kg^{-1}$ for As,

**Table 4**

Concentration of trace elements in muscle (mg $kg^{-1}$ wet wt) of species of commercial importance from genus *Odontesthes* and other genus from related water bodies.

| Genera *Odontesthes* | | *Date | As | Pb | Hg | Reference |
|---|---|---|---|---|---|---|
| *O. bonariensis* | De la Plata estuary, Argentina | 2011 | 0.03 | 0.19 | 0.30 | This paper |
| *O. bonariensis* | Adela Lagoon, Argentina | 2011 | 0.09 | 0.09 | 0.04 | This paper |
| *O. bonariensis* | Barrancas Lagoon, Argentina | 2011 | 0.08 | 0.06 | 0.03 | This paper |
| *O. bonariensis* | Chasicó Lake, Argentina | 2011 | 0.76 | 0.04 | 0.42 | This paper |
| *O. bonariensis* | Salado River, Argentina | 2008–2009 | 0.8 | – | – | Rosso et al., 2013 |
| *O. bonariensis* | Quequen Salado River, Argentina | 2008–2009 | 1.23 | – | – | Rosso et al., 2013 |
| *Odontesthes sp.* | De la Plata estuary, Uruguay | 1998–1999 | – | – | 0.24 –0.49 | Viana et al., 2005 |
| *O. microlepidotus* | Negro River, Argentina. | – | 0.056–0.12 | – | 0.07–0.38 | Arribere et al., 2003 |
| Other genus | | | | | | |
| *Mugil platanus* | De la Plata estuary, Argentina | 1987–1988 | – | – | 0.4 | Marcovecchio and Moreno, 1993 |
| *Rhamdia quelen* | De la Plata estuary, Argentina | 1987–1988 | – | – | 0.13 | Marcovecchio and Moreno, 1993 |
| *Rhamdia quelen* | Quequén Salado River, Argentina | 2008–2009 | 0.7 | – | – | Rosso et al., 2013 |
| *Parapimelodus valenciennis* | Chascomús Lake | 2011 | 0.04 | 0.13 | – | Schenone et al., 2014 |

* Date: sampling time.

0.5 mg $kg^{-1}$ for Hg, and 0.3 mg $kg^{-1}$ for Pb, on wet weight basis), the European Commission (EC) (0.3 mg $kg^{-1}$ for Pb, and 0.5 mg $kg^{-1}$ for Hg, on wet weight basis), and the US Environmental Protection Agency (1.3 mg $kg^{-1}$ for As, on wet weight basis) (Eisler, 1994). However, according to the European Food Safety Authority (EFSA, 2010), the lower limit on the benchmark dose for a 10% response (BMDL) values associated with health risk for As ranged from 4.9 to 24 μg $kg^{-1}$ of body weight (b.w.) per week. Considering these values, a 60 kg person must consume more than 0.38–1.9 kg of silverside muscle from Chasicó per week to have health problems. Nevertheless, this consumption rate may be actually lower in order to affect health since the BMDL values correspond to inorganic arsenic, which represents a small percentage of the total arsenic concentration.

Furthermore, the EFSA CONTAM Panel has established a Tolerable Weekly Intake (TWI) of 4 μg $kg^{-1}$ b.w. for inorganic mercury, which is in agreement with JECFA (FAO, 2010). On this basis, a 60 kg person would have to consume more than 0.05 kg of silverside muscle from the Chasicó Lake per week to obtain health issues. According to the EFSA (2010), the recommended maximum intake of Pb ranges from 4.34 to 29.4 μg $kg^{-1}$ b.w. per week. Considering these values, a 60 kg person would have to consume more than 1.5–9.3 kg of silverside muscle from de la Plata River per week to obtain health problems. In Argentina, the average fish consumption rate (0.1–0.2 kg per week, FAO, 2012) is similar to the rates calculated above for As and Hg in relation to silverside consumption from Chasicó Lake. However, the consumption rate is much lower than that calculated for Pb from de la Plata River. The result can be especially severe for the lagoon-side and riverside communities that may consume greater amounts of fish, leading to higher risk of chronic exposure to contaminants.

## 4. Conclusions

The results from water and muscle samples showed high concentration of toxic elements from natural and anthropic sources highlighting the human effect over the aquatic environment. A fraction of the population may consume more fish contaminated with Hg (de la Plata River and Chasicó Lake) and As (Chasicó Lake) than the recommended maximum intake values, which would leading to more vulnerable communities.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.10.119.

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