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Influence of volcanic activity and anthropic impact in the trace element contents of fishes from the North Patagonia in a global context

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Abstract The elemental contents in salmonid muscle and liver tissues from different lakes around the world were investigated. Fish from pristine areas were compared with those fishes from impacted environments, both by volcanic and anthropogenic activities. Within the data, special attention was given to fishes from the Andean Patagonian lakes in two contexts: local and global. The local evaluation includes geological and limnological parameters and diet composition which were obtained through a data search from published works. The volcanic influence in Andean Patagonian lakes was mainly observed by an increase of cesium (Cs) and rubidium (Rb) concentrations in fishes, influenced by calcium (Ca) and potassium (K) water contents. Zinc (Zn), selenium (Se), iron (Fe), silver (Ag), and mercury (Hg) contents in fishes showed the effect of the geological substratum, and some limnological parameters. The diet composition was another factor which affects the elemental concentration in fishes. The analyzed data showed that the fishes from Andean Patagonian lakes had elemental content patterns corresponding to those of pristine regions with volcanic influence. Selenium and Ag contents from Andean Patagonian fishes were the highest reported.

Keywords Elements · Salmonids · Andean Patagonian lakes · Pristine area · Anthropic area · Volcanic influence

Introduction

The elements in freshwater ecosystems are incorporated mainly through biogeochemical cycles. These involve transfer processes of substances from rocks and soils through weathering and runoff (Bailey et al. 1978). Moreover, the human activities which alter the land, biodiversity, and hydrology systems intensify these processes increasing the availability of the elements. The global transport also represents element sources which are associated to natural dust or gaseous products as volcanic or anthropogenic compounds from remote areas of the world.

The elements enter to the aquatic ecosystems by dry and wet precipitation, dissolve in water, sediment deposits, or suspended matter in the water column (Eisler 1987; Alma 1983). Physical and biogeochemical characteristics of the freshwater ecosystems determine the element bioavailability to the organisms (Newman and Jagoe 1994). In this sense, mixing processes, pH, dissolved organic carbon (DOC), redox, and elemental concentrations are the main factors involved (Newman and Jagoe 1994). Among environmental pollutants, metals are of particular concern, due to their potential toxic effects and liability to be bioaccumulated. Heavy metals, including both essential

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and non essential elements, have a particular significance in ecotoxicology since they are persistent and all have the potential to be toxic to living organisms (Kwaansa-Ansah et al. 2012). The water and diet are the metal entry routes for aquatic organisms and the trophic position is one of the main factors that influence the reached concentration levels (Bury et al. 2003). The fishes include a variety of trophic levels and have a long life cycle; this makes them good indicators of long-term effects and habitat conditions (Plafkin et al. 1989). The elemental bioconcentrations depend on the chemical form, e.g., Hg which can be biomagnified as methylmercury (Wang 2012), while others such as arsenic (As), cadmium (Cd), and lead (Pb) can be diluted (Leeves 2011; Revenga et al. 2012).

In the last years, elemental contents in fish tissues from lakes of the Northern Patagonia Andean Range have been studied (Ribeiro Guevara et al. 2005; Arribére et al. 2006; Arribére et al. 2008; Rizzo et al. 2010 and therein references), where high concentrations of several elements, e.g., Se, Ag, Rb, and Cs, were found, respectively, to other fishes from freshwater ecosystems of the world.

The lakes of the Northern Patagonia Range limit with the Andes Mountains to the west, which includes several active volcanoes known as the Southern Volcanic Zone (SVZ) (Petit-Breuilh Sepúlveda 2004). The orography and the prevailing westerly winds determine the rainfall regime, with a strong gradient from West to East. Due to these conditions, the volcanic activity from SVZ has impacted over the Northern Patagonia region. The lakes are the glacial origin, mostly ultraoligotrophic, which are in an area with low human development and historically protected under the National Park Administration (Díaz et al. 2007). The lakes have native fish species, e.g., Creole perch *Percichthys trucha*, inaga *Galaxias maculatus*, and introduced salmonids as rainbow trout *Oncorhynchus mykiss*, brown trout *Salmo trutta*, brook trout *Salvelinus fontinalis*. This fish group originated from the North hemisphere and has been artificially widely distributed around the world (Wegrzyn and Ortubay 2009). In Patagonia, the salmonids were introduced at the end of the nineteenth century, and at the beginning of the twentieth century, adapted well in these environments (Ferriz, et al. 1998).

The aims of this work are the following: (a) to investigate how the regional characteristics affects the bioaccumulation processes of elements (K, sodium (Na), Rb, Zn, Fe, bromine (Br), Cs, Se, Hg, and Ag) in muscle and liver tissues of rainbow trout; (b) to investigate if the trends observed in the local study are similar to those found in other salmonids from ecosystems with volcanic influence; and (c) to investigate how the anthropic influence affects the bioaccumulation patterns.

To achieve these goals, an exhaustive search from the scientific works published from 1968 to 2013 was done. This range of time was considered for including more amounts of elements due to the lack of reported data in salmonids. The local study, involved the evaluation of the elemental contents in fish related to elemental concentrations of the geological substratum (aquatic sediments), water chemistry, and rainbow trout diet composition. At a global scale, the elemental contents in salmonid species were compared to those from pristine regions with and without volcanic influence and anthropic impacted areas.

Material and methods

Local study

Rainbow trout was the chosen specie for the elemental composition study from the Patagonian Andes due to its wider distribution and abundance in this area (Wegrzyn and Ortubay 2009). The elemental contents in muscle and liver were taken from Bubach D. Ph.D. (2010).

Elemental Analyses Aliquots of about 100 mg of dried homogenized and powdered sample were sealed in SUPRASIL AN® quartz ampoules, irradiated for 24 h, and analyzed by instrumental neutron activation analysis (INAA) at the RA-6 nuclear research reactor at Centro Atómico Bariloche. The elements analyzed were as follows: K, Na, Rb, Zn, Fe, Br, Cs, Se, Hg, and Ag, and corresponded to 183 adult individuals analyzed mainly in pools. Gamma ray spectra were collected using an intrinsic high-purity germanium (HPGe) n-type detector, 12.3 % relative efficiency, and a 4096-channel analyzer. Spectra were analyzed by using the GAMANAL routine included in the GANAAS package, distributed by the International Atomic Energy Agency (IAEA). Corrections for spectral interferences were included when necessary. The concentrations reported are referred to dry weight basis. Standard reference materials from the National Research Council of Canada (NRCC)-DORM-2 dogfish muscle, (NRCC)-TORT-2 lobster, (NRCC)-DOLT-2 dogfish liver, as well as sample replicates, were analyzed to check on the quality of analysis. The analyses of the standard reference materials showed good agreement with certified and informed values, and replicate samples were consistent These data were reported by Ribeiro Guevara et al. (2006).

Study area

Figure 1 shows the sampled lakes: Traful, Espejo Chico, Nahuel Huapi, Moreno (Atlantic drainage basin), and Guillelmo (Pacific drainage basin) all belonging to Nahuel Huapi National Park (NHNP) and Rivadavia and Futalaufquen (Pacific drainage basin) from Los Alerces National Park (LANP) located at Northeast of the Andean Patagonia (The main characteristics of the lakes are shown on Table 1). These lakes are glacial origin, ultraoligotrophic, with very dilute calcium, bicarbonate, and silica dominance (Pedrozo et al. 1993) and they have been classified as warm monomictic, with a summer stratification period (Quirós and Drago 1985). The largest human settlement of the study site is around lake Nahuel Huapi, mainly by the city of San Carlos de Bariloche (circa 110,000), Dina Huapi (6500 habitants), and Villa La Angostura (7600 habitants); other settlements have less than 500 habitants (INDEC 2001). The primary economic activity is tourism, which attracts hundreds of people per year from Argentina and abroad.

Geological substratum

The geological substratum was evaluated through the Ag, Br, Cs, Fe, Hg, Na, Rb, Se, and Zn contents of stream sediments from the principal lakes tributaries. These data were obtained from Ferpozzi et al. (2001, 2004). The lake Traful tributary data correspond to sediments from streams located in both the North and South margins, which were systematically taken 1 every 10 from more than 1000 data finally reduced to 554. The lake Espejo Chico tributaries correspond only to the North margin. The lakes Espejo Grande and Correntoso were considered as Nahuel Huapi tributaries; Lakes Cholila and Menéndez were treated as tributaries of the lakes Rivadavia and Futalaufquen from LANP. In general, the total data of these lakes were $10<N\leq 22$.

Limnological data

Magnesium (Mg), Na, K, Ca, total phosphorous (TP), soluble reactive phosphorous (SRP), bicarbonate (HCO3=) contents, and pH were the chosen parameters for characterizing the aquatic environment. The data were taken from Pizzolón et al. (1994), Reissig (2005), and Díaz et al. (2007), corresponding to seasonal values of at least 1 year.

Fish diet

Diet composition was estimated from the stomach contents in a summer period, which was performed through the identification and count of the food categories and prey using a stereomicroscope. The volume of each food category per stomach was measured by water displacement in a graduated cylinder (Macchi 2004). These data were obtained from Bubach (2010).

Data for the global study

A database of elemental concentrations in liver and muscle tissues from scientific publications from 1968 to 2013 was performed. The genera considered for this work included the following: *Salmo*, *Oncorhynchus*, *Salvelinus*, *Coregonus*, and *Thymallus*. The data published were taken when the contents were given either in dry weigh (DW) basis or wet weight (WW) with the humidity percentage informed.

The classification of the regions was made according to development degree or considering the type of impact close to the lake area, namely:

Pristine region (PR): a place with low population (less than 100,000 habitants), very small industrial or farming activities.

Pristine region with volcanic influence (PRV): when the area is pristine and has volcanic or hydrothermal activity.

Anthropic impacted region (AIR): area impacted by urban waste discharges, industrial development, mining regions, hydrocarbons extraction, or intensive agriculture.

The elemental contents in the tissues of fishes from these three types of regions were compared with those from the North Andean Patagonian (AP) aquatic ecosystems used in the local study included other salmonids such as brown trout (N=64) and brook trout (N=56), wherein data were taken from Bubach (2010).

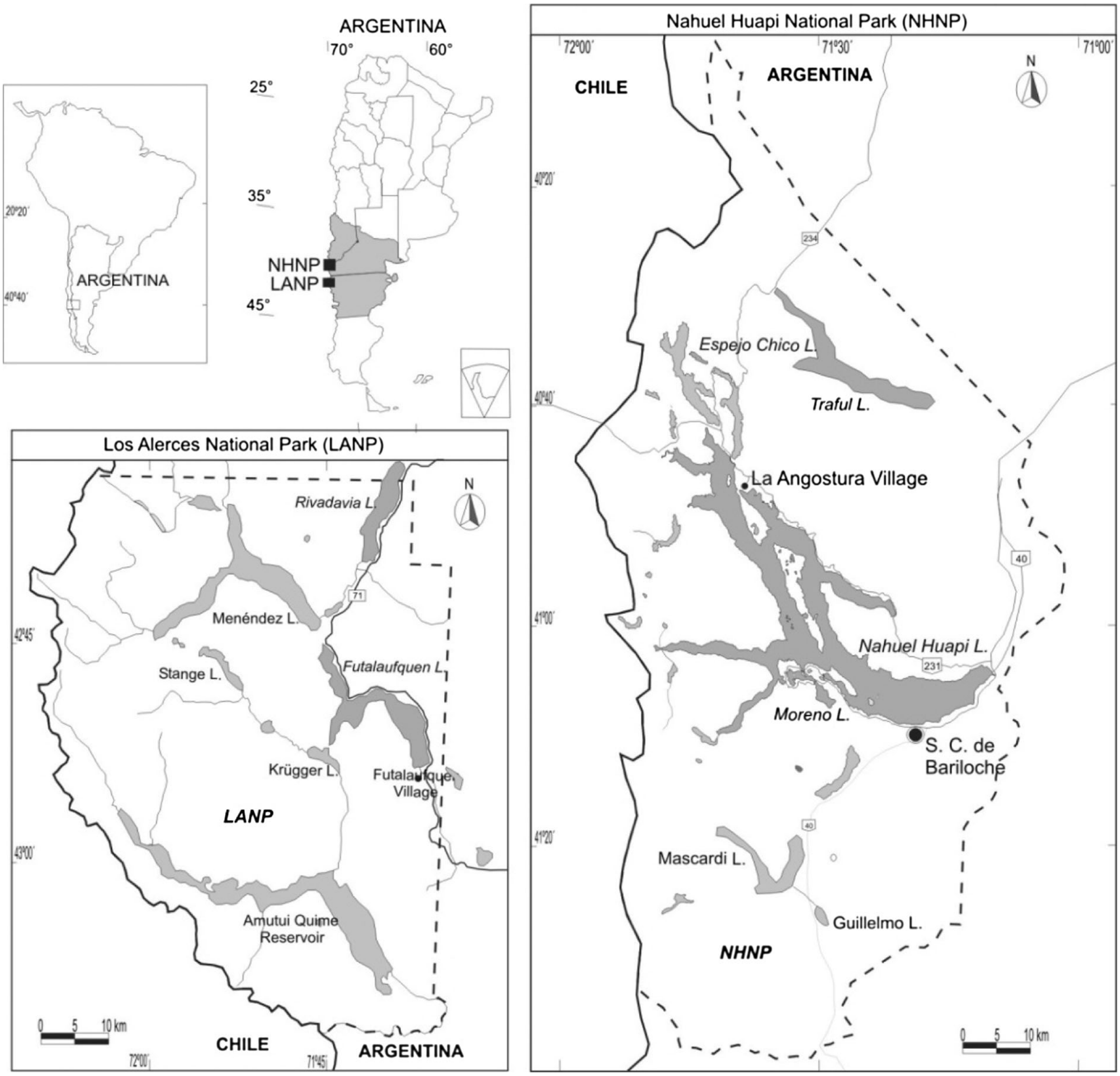

Fig. 1 Study area

Statistical analysis

The statistical analysis was performed with the program XLSTAT version (7.5.3) copyright 1995–2005 Addinsoft. The significance level considered in all statistical tests was (α) ≤0.05. Principal components analysis (PCA) was performed for the local study where elemental contents in fishes and averages of tributary sediments and limnological parameters were used. The differences among areas at global level were checked by ANOVA and Kruskal-Wallis tests and post hoc Fisher's LSD test were used to discriminate similar groups.

Results

Evaluation at local scale

In this section, we present the results of the different PCA obtained from AP lakes mentioned in the "Study area" section and the rainbow trout diet composition. The PCA of rainbow trout muscle samples produced three independent components explaining 73 % of the variance and 66 % for livers. Figure 2a shows the component principal 1 (CP1) vs component principal 2 (CP2) and Fig. 2b, CP2 vs component principal 3 (CP3).

Table 1 Geographic coordinates and physical parameters of the study lakes

National park	Lake	Location	Altitude (msm)	Mean depth (m)	Maxim depth (m)	Area (km^2)	Watershed area (km^2)	Secchi disk (m)
Nahuel Huapí	Traful	40° 30′ S, 71° 17′ O	750[a]	173[b]	200[a]	75[a]	–	15[a]
National park	Espejo Chico	40° 34′ S, 71° 44′ O	750[a]	–	68[a]	1.9[a]	–	15[a]
	Nahuel Huapi	40° 50′ S, 71° 30′ O	765[a]	157[c]	464[a]	557[a]	4260[a]	7[a]
	Moreno	41° 5′ S, 71° 33′ O	765[a]	–	180[a]	11[a]	–	7[a]
	Guillelmo	41° 23′ S, 71° 29 ′O	826[a]	61[d]	100[a]	5.4[a]	–	11[a]
Los Alerces	Rivadavia	42°3 6′ S 71° 39′ O	527[e]	104[e]	147[e]	22[e]	1647[e]	12[e]
National park	Futalaufquen	42° 49′ S, 71° 43′ O	518[a]	101[d]	168[f]	45[a]	2920[f]	14[a]

[a] Díaz et al. (2007)
[b] Vigliano et al. (2002)
[c] Pedrozo and Vigliano (1995)
[d] Quirós (1988)
[e] Pizzolón and Arias (1995)
[f] Pizzolón (1995)

The CP1 and the CP3 are explained by Se, Br, Zn, Na, Fe, and Hg, while Cs and Rb are explained by the second component (CP2) in both plots. The observations are distributed in two groups in agreement with Rb–Cs–Br, Zn–Fe–Hg–Na, and Se–Na vectors which show significant correlations (Table 3, Complementary data). The first group encloses fishes from lakes Espejo Chico, Traful, Nahuel Huapi, Moreno, in a gradient distribution related to Rb–Cs (Fig. 2a), distinguishing the lake Espejo Chico on the top vector. The second cluster includes lakes Futalaufquen, Rivadavia, and Guillelmo due mainly to the Se and Zn contributions. Figure 2b shows enclosed lakes Nahuel Huapi and Moreno by vectors Fe and Hg. The significant correlations among the elements in liver (Table 4, Complementary data) are the same as those observed for muscle. Figures 3 shows the PCA for the liver data, where a similar pattern to muscles is observed. The only difference is given by Ag and Hg in liver; both elements determine a gradual separation of lakes Nahuel Huapi and Moreno from the other lakes (Fig. 3b).

Figure 4 shows the PCA results using the data of the stream sediments (Fig. 4a) and limnological parameters (Fig. 4b). The lakes Rivadavia, Futalaufquen, and Menéndez in Fig. 4a are separated from lakes Correntoso, Cholila, Espejo Chico, and Traful. The first group of lakes is mainly enclosed by K, Se, Ag, Cs, Br, and also Rb; Na vector is associated with the other four lakes, while the Espejo Grande is distinguished by Hg. The same assemblage is also observed in Fig. 4b, characterized by higher pH, Ca, PRS, HCO_3, and K. The lakes Moreno and Nahuel Huapi are in the same direction of Na and TP and opposite to pH vector, indicating a more acidic pH respect to the other lakes.

The rainbow trout diet composition is presented in Fig. 5. This shows a similar composition among groups of lakes as was observed in the PCA (Figs. 2 and 3). In the lakes Guillelmo, Rivadavia, and Futalaufquen, the diet includes larvae and adult insects and plankton while insect larvae, amphipods, and organic matter are the dominant items from fish of lakes Espejo Chico and Traful and inanga *G. maculatus*, a native fish, for lakes Moreno and Nahuel Huapi, which represented more than the 60 % of consumption.

Evaluation at global scale

Table 2 shows mean and standard deviation of the elemental concentrations in the muscle and liver of fishes of different regions of the world and the probability level (p) of the ANOVA and Kruskal-Wallis test for each element among areas. An extended version of these data is found on Tables 1 and 2 in the Complementary data. Table 3 reports post hoc Fisher's LSD test for the elements that had significant differences.

The database is composed at least by 450 values from each area depending on the element; in general, the muscle data (2237 individual fishes) is more complete than the liver (1892 individuals). A few data are reported by Ag in muscle and Br in both tissues.

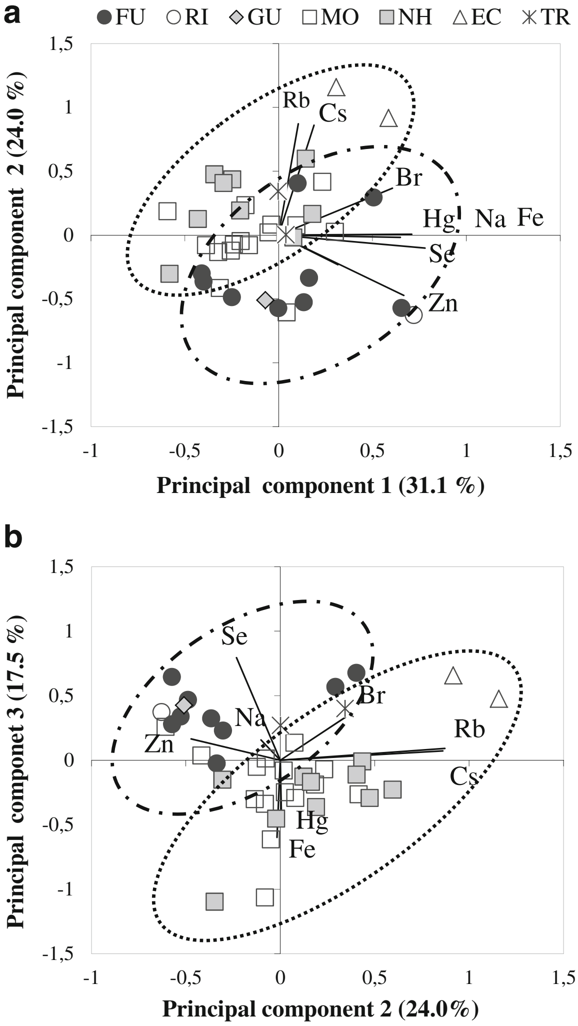

Fig. 2 Principal component analysis of elements in rainbow trout muscle from lakes Futalaufquen (*FU*), Rivadavia (*RI*), and Guillelmo (*GU*) *within dotted circle* and Moreno (*MO*), Nahuel Huapi (*NH*), Espejo Chico (*EC*), and Traful (*TR*) *within lined and dotted circle*. **a** Principal component 1 vs 2. **b** Principal component 2 vs 3

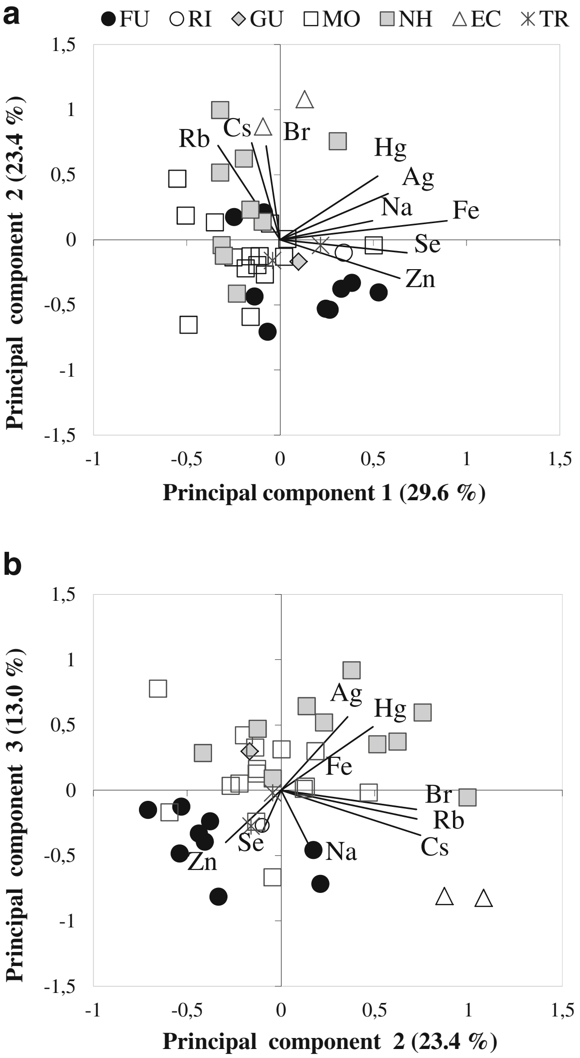

Fig. 3 Principal component analysis of elements in rainbow trout livers from lakes Futalaufquen (*FU*), Rivadavia (*RI*), Guillelmo (*GU*), Moreno (*MO*), Nahuel Huapi (*NH*), Espejo Chico (*EC*), and Traful (*TR*). **a** Principal component 1 vs 2. **b** Principal components 2 vs 3

The differences between muscle and liver samples basically are due to the different physiology of the tissues (Yamazaki et al. 1996 and Barber et al. 2003).

The elemental concentrations in the database decreases consistently with their natural abundance, biological function, and essentiality as shown on Table 2. The concentrations in muscle decreases from (18,000 to 0.024 μg/g) in order: K>>>Na≫Fe~Rb~Br~Zn>Se~Hg~Cs>Ag. The element sequence for the livers shows a shift of Br and Rb, which are allocated between Zn and Se.

Rubidium, Cs, and Na in both tissues and Hg in liver showed significant differences among areas ($p<0.05$). Those were given mainly for PRV which concentrations were one to two orders of magnitude higher than in the other three regions (AP, PR and AIR, Table 2). Moreover, significant differences ($p<0.05$) in both tissues of AP fishes were observed among PRV; PR and AIR for Cs, and also, between PRV and PR for Rb (post hoc Fisher's LSD test, Table 3). The concentrations of these elements were twice higher than AP in respect to PR and AIR (Table 2). Sodium and Hg from AP presented significant differences with PRV in both tissues

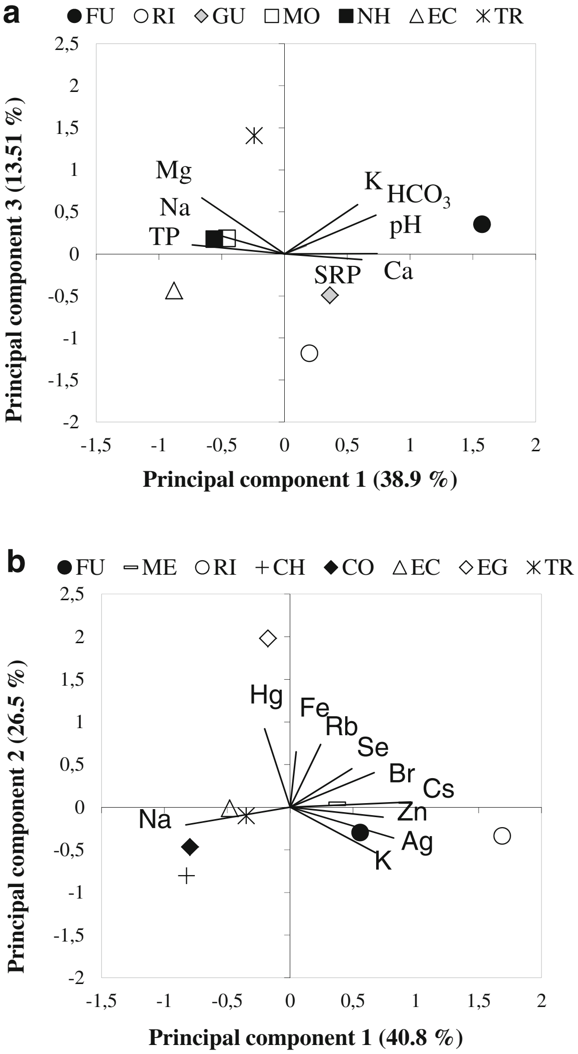

Fig. 4 Principal component analysis in **a** elements in tributary sediments and **b** water chemistry. Lakes of Pacific drainage basin: Guillelmo (*GU*), Futalaufquen (*FU*), Menéndez (*ME*), Rivadavia (*RI*), Cholila (*CH*). Lakes of Atlantic drainage basin: Correntoso (*CO*), Espejo Chico (*EC*), Espejo Grande (*EG*), and Traful (*TR*). Total phosphorus (*TP*) and soluble reactive phosphorus (*SRP*)

($p<0.05$, post hoc Fisher's LSD test, Table 3), and the highest values were in PRV (Table 2).

Although, the elements K, Zn, Fe, and Ag did not show significant differences among the areas due to their great dispersion, it was observed in some tendencies (Table 2). The highest values were mainly observed in PRV for K and Zn in both tissues, in addition to Se in muscle and Fe in liver. On the other hand, Se and Ag were markedly higher, around double, in liver of AP fishes respect to those from the other areas (Table 2).

Discussions

Water chemical properties as well as geological substratum influence on diverse factors like vegetation development, elemental bioavailability, and food web assemblage in freshwater ecosystems (Newman and Jagoe 1994; Weatherhead and James 2001; Marziali et al. 2008). The trophic structure and dietary composition are factors which also play an important role in the trace elements bioaccumulation in aquatic organisms (Soto-Jiménez 2011; Wang 2012).

Our results showed that the elemental contents in fish tissues were grouped according to the lake of origin (Figs. 2 and 3). The relevant result was the agreement in the patterns observed in all PCA (fish tissues, limnological parameters, and tributary sediment composition). The elements Rb, Cs, Br, Na, Se, and Zn mainly distinguished the lakes Rivadavia-Futalaufquen-Guillelmo; from Moreno-Nahuel Huapi-Espejo Chico, also in some cases Espejo Chico-Traful. The lakes Rivadavia, Futalaufquen, and Menéndez drain to the Pacific Ocean, while the others lakes (Espejo Chico and Traful) drain towards the Atlantic.

The rainbow trout diet composition (Fig. 5) provides some explanation for the elemental content patterns. The diet of fishes from lakes of Pacific drainage was mainly composed by plankton and insects (larvae and adults), while the diet of fish from lakes of Atlantic drainage were more varied. These assemblages by drainage basin are also observed in limnological parameters (Fig. 4b).

Alkaline elements are typically found in igneous rocks and volcanic products like ashes and gases (Ruiz and Cebriá Gómez 1990; Grassom et al. 1999; Llyinskaya 2007). Thus, the high Rb, Cs, and Na values could be explained by a volcanic source. The area of the AP lakes has been affected by SVZ (Petit-Breuilh Sepúlveda 2004). The geochemical characterization of volcanic products may provide a fingerprint which enables to identify the volcano provenance, associating terrestrial and lacustrine tephra deposits with specific volcanic events. Some investigations about the volcanic product inputs in lakes from NHNP have been performed by Daga et al. (2010, 2014). Chaitén volcano and Puyehue Cordón Caulle (PCC) volcanic complex had two of the most recent eruptions occurred in 2008 and 2011, respectively. The first is located close to lake Futalaufquen in LAP while PCC volcanic complex is near to lakes Espejo Grande, Espejo Chico, Brazo

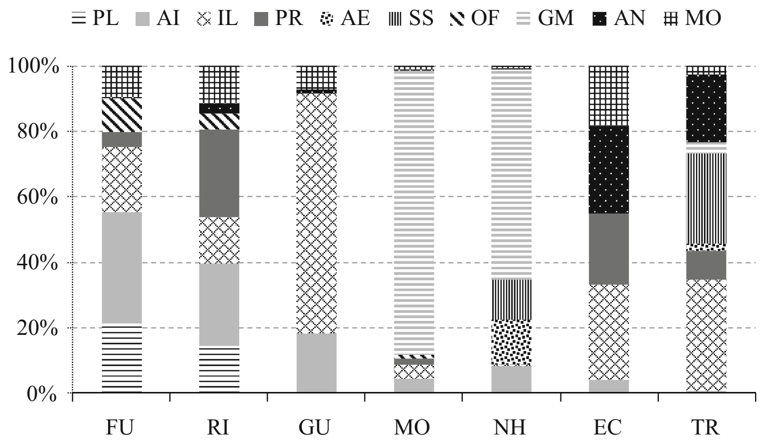

Fig. 5 Rainbow trout diet from lakes Futalaufquen (*FU*), Rivadavia (*RI*), Guillelmo (*GU*), Moreno (*MO*), Nahuel Huapi (*NH*), Espejo Chico (*EC*), and Traful (*TR*). Plankton (*PL*), adult insect (*AI*), insect larvae (*IL*), plant remains (*PR*), *Aegla* sp. (*AE*), *Samastacus* sp. (*SS*), other fishes (*OF*), *Galaxias maculatus* (*GM*), amphipods (*AN*), and mollusks (*MO*)

Rincón from Nahuel Huapi, and Traful (Fig. 1). In particular, Daga et al. (2014) reported elemental concentration differences according to the volcano sources, e.g., Rb and Cs in glass shards, with larger concentration for Chaitén while Fe, Br, and Zn for PCC. After the initial impact, the prolonged exposure of the pyroclastic products to weathering may cause the slow release of elements from the structure of constituent mineral phases and glassy matrix to soils and waters. Furthermore, a bioindication study in the same AP area after the last PCC eruption showed a positive correlation among Rb, Cs, and Br with the distance to PCC volcano (Bubach et al. 2012, 2014). Therefore, the results in fish tissues may indicate a strong relation among Rb, Cs, and also Br, with the distance to the volcano. Moreover, the absence of concordance of fishes with sediments in the PCA (Figs. 2, 3, and 4a) and the Daga et al. (2014) results may be the consequence of the weathering and runoff processes that produce dissimilar element bioavailability due to limnological parameters, geological substratum, and drainage slope differences (Fig. 4b). The water, in addition to the diet, is another pathway of elements to fishes mainly through the gills, which is a metabolically active target organ for metal accumulation (Yilmaz et al. 2007; Jarić et al. 2011; Poleksić et al. 2010; Višnjić-Jeftić et al. 2010). This route seems important for alkaline elements and dissolved gases. Relations between Rb and Cs with other alkaline elements in fresh water environments were reported by Rowan and Rasmussen (1994), Avery (1996), Hagström (1999). These authors have found negative correlations between K concentration in water and ^{137}Cs in fish. Sonesten (2001) verified that ^{137}Cs contents in fish were lower when the water hardness increased, probably due to Rb and Cs competition with K absorption in the gills (Oughton and Salbu 1992). This can explain the difference observed in the lakes patterns around the Rb and Cs vectors where the lakes are grouped as Espejo Chico, Nahuel Huapi-Moreno-Traful, separated from Guillelmo-Rivadavia and Futalaufquen (Figs. 2, 3 and 4).

Rubidium and Cs had the highest concentrations in the fishes from Yellowstone National Park NS lakes Bolserna and Bracciano, included into the PRV and AP lakes, respectively, to the fishes of the other areas (Table 2, and Tables 1 and 2 from Complementary data). Yellowstone is an environment with thermal springs. Lakes Bolserna and Bracciano, originating from volcanic craters, are located in central-west Italy, in the Vulcini volcanic and Sabatini complexhttp://en.wikipedia.org/wiki/Italy.

High Na values were observed in fish tissues from PRV. Besides the eventual Na and K volcanic contribution, the Na concentration in fishes could also be influenced by Ca concentrations in water. Wurts (1987) and Ratte (1999) showed an Na influx increase in the fish gills when Ca concentration in water is low. Verbost et al. (1989) revealed the water chemistry importance in the Na^+/Ca^{2+} transepithelial exchange between gills and water. The Na concentration in AP fishes (2300±420 μg/g) were higher than in Bolserna and Bracciano (710±240 μg/g); Orban et al. 2006, Table 1 and Table 1 in Complementary data) while both Na concentrations in water were similar (~1–2 mg/L, Pizzolón 1995; Mosello et al. 2004; Díaz et al. 2007) The Ca concentration in water of AP lakes (3–8 mg/L; Pizzolón et al. 1994, 1995; Díaz et al. 2007) is one order of magnitude lower than lakes Bolserna and Bracciano

Table 2 Number of fishes analyzed (*n*), means of elemental concentrations (μg/g DW±SD) in muscle and liver of salmonids from the Andes of Northern Patagonia (AP), pristine areas without (PR) and with volcanic influence (PRV) and anthropic impacted areas (AIR). Probability levels of ANOVA and Kruskal Wallis test (p)

Areas	*n*	K (wt. %)	Na	Rb	Zn	Fe	Br	Cs	Se	Hg	Ag	Reference
Muscles												
Areas	*n*	K (wt. %)	Na	Rb	Zn	Fe	Br	Cs	Se	Hg	Ag	Reference
AP	303	1.8±0.2	2301±420	47±12	22±7	22±6	26±3	1.1±0.4	1.2±0.3	0.74±0.27	<0.080	1
PR	669	1.8±0.3	875±428	20±19	22±7	30±16	11	0.38±0.34	1.5±0.6	0.41±0.22	0.0014≤0.10	2–9
PRV	64	*4.4±0.1*	*7571±580*	*184±24*	*56±42*	<50	–	*76±10*	*3.5±0.8*	*1.8±0.4*	–	10
	24	1.80±0.07	708±237	*110±9*	24±3	15±2.0	–		1.1±0.4	0.10±0.10	–	11
AIR	1480	1.7±0.1	1598±867	23±10	21±15	*63±56*	*42±4*	0.060±0.035	2.1±1.9	*1.5±1.7*	0.024±0.010	12–20
p[a]		0.23	0.036	0.00010	0.059	0.22	–	0.00010	0.24	0.47	–	
AP	303	1.0±0.2	*4647±590*	33±7	122±36	877±402	56±11	0.58±0.13	22±25	0.72±0.34	3.0±3.4	1
PR	450	1.3±0.5	3744±1759	32±20	106±23	635±459	60	0.12±0.11	12±10	0.50±0.33	1.2±0.8	2–7, 21–23
PRV	64	1.2±0.2	*5400±929*	*68±16*	108±13	*1057±120*	–	*17±5*	9±2	*1.8±0.38*	<0.020	10
AIR	1378	1.0±0.1	2596±436	24	145±54	507±202	–	0.070±0.040	18±17	0.74±0.36	0.84	12–13, 15–17
p[a]		0.60	0.17	0.042	0.13	0.37	–	0.0010	0.70	0.0010	0.56	

Major concentrations are presented in italics

[a] Probability value of ANOVA and Kruskal-Wallis test. Significance level ($p<0.05$)

1 Fishes from AP, *2* Allen-Gil et al. (1997), *3* Mueller et al. (1996), *4* DeBeers (2002), *5* Rancitelli et al. (1969), *6* Munn et al. (1995), *7* Matz et al. (2005), *8* Campbell et al. (2005), *9* Van der Velden et al. (2013), *10* Chaffee et al. (2007), *11* Orban et al. (2006), *12* Crock et al. (2000), *13* Higgins (2006), *14* Palace et al. (2004), *15* Amundsen et al. (1997), *16* Amundsen et al. (2006), *17* Kashulin et al. (2006), *18* Alcalde and Gil (2000), *19* Mierzykowski (2013), *20* Fallah et al. (2011), *21* Peterson and Boughton (2000), *22* Burton (2002), *23* Yancheva (2010), *24* Kidd et al. (2004), *25* Lamas et al. (2007)

Table 3 Probability values of the post hoc Fisher test of ANOVA. Andes of Northern Patagonia (AP), pristine areas without (PR) and with volcanic influence (PRV) and anthropic impacted areas (AIR)

Areas	Muscle			Liver		
	Rb	Cs	Na	Rb	Cs	Hg
PRV-PR	*<0.00010*	*<0.00010*	*0.0087*	*0.023*	*0.00013*	*<0.00010*
PRV-AIR	*<0.00010*	*0.00017*	*0.013*	*0.023*	*<0.00010*	*0.00087*
PRV-AP	*<0.00010*	*0.0033*	*0.036*	*0.012*	*0.00022*	*0.00047*
AP-PR	*0.042*	*0.0011*	0.28	0.52	*0.010*	0.14
AP-AIR	0.063	*0.045*	0.53	0.94	*0.0053*	0.92
AIR- PR	0.96	0.22	0.58	0.60	0.72	0.26

$p<0.05$ are presented in italics

(21 mg/L; Mosello et al. 2004). Sodium in fishes and Ca in water comparisons between Bolserna and Bracciano and our results indicate that the Cs, Rb, and Na concentrations in AP fishes may be explained by the exchange of these ions through the gills (Fig. 4b).

Bromine sources have been identified in marine aerosols, volcanic products, and anthropic compounds such as esters of polybrominated biphenyls (PBDEs) and methyl bromide (Sturges and Harrison 1967; Wit 2002; Martin et al. 2012; Bubach et al. 2012, 2014). In AP fishes, the correlation between Br and Rb and Br and Cs could confirm its volcanic source; but at a global level, it cannot be certain because the data are insufficient (Table 2, Figs. 2 and 3, Tables 1 and 2 in Complementary data).

The element chemical availability in the freshwater system is the result of the interaction of several variables which include pH. The Fe, Zn, Hg, and Ag availability is easier in acidic pH, in contrast to other elements like Se, which is most favored at basic pH (Eisler 1987, 1993, 1996 and Ezoe et al. 2001). The rainbow trout distributions (see Figs. 2a, b and 3b) in relation to Fe, Hg, Se (muscle and liver) and Ag (liver) vectors is according to pH like is shown in Fig. 4a. This distribution assembles the Nahuel Huapi and Moreno fishes by the Hg and Ag vectors, and Guillelmo, Rivadavia, and Futalaufquen by Se (Fig. 3b). This agreement between element and pH is not observed for Zn, where its concentration in fishes is higher in the lakes Guillelmo, Futalaufquen, and Rivadavia, which are more alkaline than the other lakes. A probable explanation for this could be the chelation reactions between PRS and Zn (Fig. 4b) which solubility is higher at basic pH (Wetzel 1981). Likewise, the assemblage observed in relation to this element and also Se in the fishes reflects the diet composition (Figs. 2, 3, and 5). At a global level, the great variation in Zn, Se, and Fe content observed in Table 2 gave the lack of significant differences among areas. This great variation was more important in the AIR and PRV areas, mainly by Yellowstone National Park with the highest concentrations (~50 % of average, Table 2; and Table 1 and 2 in Complementary data). However, Zn, Se, and Fe are regulated by the organisms; the bioconcentration of those elements due to their high availability exceeds probably the excretion capacity of the fishes from those areas.

The Ag contamination occurs naturally in the earth's crust and in mining deposits (Purcell and Peters 1998; Hein et al. 1999) and also sewage wastes (Ratte 1999; Nichols et al. 2006) linked to a global and local transport. The higher Ag content in AP livers could be due to the presence of mineral deposits in the region (Giacosa et al. 1999) and Ag availability may be modified for anthropic activities. Ribeiro Guevara et al. (2005) attributed the Ag presence to the release of photography reagents from the city waste effluents, which were dumped directly into lake Nahuel Huapi. However, Ag in photographic wastes is in a chemical form (as a metal) that cannot be bioconcentrated by the organisms (Eisler 1996; Ratte 1999). Silver has high affinity to sulfur ligands and halogens in water and sediments (Bell and Kramer 1999), and may be accumulated in the benthic food chain as described by Ratte (1999) and Larissa et al. (2006). The rainbow trout diet from the lakes Nahuel Huapi and Moreno is mostly composed for inanga (40–60 %). This native species has benthic feeding habits (Wegrzyn and Ortubay 2009), which could provide an explanation to Ag content observed in AP fishes.

On the other hand, Ribeyre et al. (1995) observed a 30 % Ag increase in zebrafish when metals like Cu, Zn, Hg, and Se, were present. The same results were reported in fishes by Howe and Dobson (2002). Selenium and Ag concentrations in liver from AP fishes (see Table 2) showed the highest values reported in the literature. In consequence, higher Ag and Se contents may be a signal of the environmental exposition to the divalent elements.

Increases in Hg accumulation rates have been observed in the uppermost layers of lake sediment cores including remote places (Lindberg et al. 2007). The background Hg concentrations are assumed to represent the "pre-industrial" accumulation rates. These increases have been attributed to an enhancement of the Hg global load in the environment due to direct inputs from mining, land use, and other industrial activity also from natural sources such as Hg deposition from volcanic origin (Martin et al. 2011, 2012). This Hg load in the environment was also reflected in the Hg contents in fishes at global and local scale being highest in PRV due probably to volcanic contribution, following by AIR and AP (Table 2). In particular, sediment sequences in different AP lakes not only showed increments of Hg concentrations corresponding to the second half of the twentieth century but also high concentrations in time periods in the past millennium (Ribeiro Guevara et al. 2010). This was ascribed as the result of the natural sources, e.g., volcanic activity of the Andes or extended fires. Thus, the Hg in AP fishes could be the consequence of that observed in the sediment, in addition to the presence of the metals in the divalent form as was mentioned earlier by Ag and Se.

Conclusions

At a local level, we found similarities in the elemental concentrations in fishes among lakes Espejo Chico-Traful, Nahuel Huapi-Moreno on the one hand; and Guillelmo, Rivadavia, and Futalaufquen on the other, as result of several factors that included volcanic sources, geological substratum, chemistry of the water, characteristics associated to drainage basin (Atlantic vs. Pacific), and diet composition.

In particular, Rb, Cs, and Br correlation indicated volcanic sources mainly associated to the PCC complex distance and their bioavailability due to Ca and K water contents. Iron, Se, Hg, and content in rainbow trout were associated to a major availability due to pH and Zn by PRS.

At a global scale, Se and Ag in livers of AP fishes showed the higher concentrations. The wide concentration range of elements usually associated with anthropic activities (Zn, Fe, and Hg) of the database did not allow us to find significant differences among areas. The elemental content patterns, especially the Rb, Cs, and Na concentrations, showed that the fishes from Andean Patagonian lakes correspond to those of pristine region with volcanic influence.

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