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Water geochemistry of shallow lakes from the southeastern Pampa plain, Argentina and their implications on mollusk shells preservation

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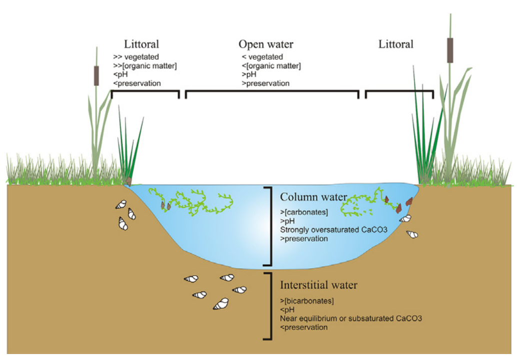
HIGHLIGHTS

- Littoral (LIT) areas have less favorable physico-chemical conditions for shell preservation than open waters (OW).
- In sediments, aragonite and calcite indices are in equilibrium or slightly subsaturated in both LIT and OW settings.
- Mollusk shells within sediments would be subject to dissolution.
- The water column in is strongly oversaturated with regard to carbonates.
- Mollusk shells in contact with the column water are not likely to be dissolved.

GRAPHICAL ABSTRACT



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ABSTRACT

A seasonal sampling of sediments, column and interstitial water for physico-chemical analysis were performed in littoral and open water areas in three freshwater shallow lakes (Nahuel Rucá, Las Mostazas and Los Carpinchos) from Southeastern Pampa plain, Argentina. The main objective of the present study is to evaluate how the characteristics of the depositional environments could be affecting mollusk shell preservation. These lakes are very shallow (2 m) and are characterized by an extensive littoral area, dominated by the emergent macrophyte *Schoenoplectus californicus*, which forms a complete ring around the lake, and an open water area, in general free of vegetation. Five samples of sediments in each compartment were extracted for analysis of pH, moisture, organic matter and carbonates content using a gravity corer, while five samples from column and interstitial water were extracted for chemical analysis (pH, conductivity, major ions, minor ions and hardness). Besides, calcite and aragonite saturation indices and the redox potential were calculated for each lake. The results show the significant impact of water chemistry and redox conditions on the preservation potential of freshwater mollusk and consequently in the quality of paleonvironmental reconstruction based on the biological record from the study region. The higher concentration of organic matter and lower pH registered in the littoral area, mainly during warm months (autumn and summer), suggest worst environments for mollusk preservation, compared to open waters. Moreover, water geochemistry analysis showed aragonite and calcite indices near equilibrium or slightly subsaturated in interstitial water associated with more acid pHs, while column water is strongly

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oversaturated related to alkaline pHs. These results suggest that carbonate remains within sediments will be subject to dissolution affecting negatively their preservation potential. However, mollusk shells in contact with the column water are not expected to be dissolved.

1. Introduction

In aquatic environments, most taphonomic loss is thought to occur at and below (first centimeters) the sediment–water interface, a zone known as the Taphonomically Active Zone (TAZ; Davies et al., 1989). Dissolution appears to be the main factor affecting preservation within the TAZ, owing to the metabolic activity of organisms, mainly by bacterial decomposition of organic matter, which regulate the state of redox within sediments and affect the saturation state of calcium carbonate (Aller, 1982; Walker and Goldstein, 1999; Best et al., 2007; Cherns et al., 2008). The microbial zone and the saturation state of the pore waters with respect to calcium carbonate will be the most important aspects of any taphonomic study that examines post-burial preservation of skeletal hard parts (Walker and Goldstein, 1999).

In marine settings different redox environments have been recognized in the sedimentary milieu (Berner, 1981) in relation to increasing reducing conditions, which can be identified through the species that reduces during the organic matter oxidation. On one hand, in the aerobic zone bioturbation produces acidity through respiration (because of the increment of CO_2) and sulfide oxidation. On the other hand, as long as we go deeper into the anoxic sediments, alkalinity increases due to sulfate reduction which adds HCO_3^- and H_2S. These sulfides may react with iron minerals to produce iron sulfide phases avoiding potential acid conditions. Below the sulfate reduction milieu, there is a zone of methane production which diminish pH, but do not produce carbonate subsaturation unless little alkalinity accumulate during sulfate reduction (Canfield and Raiswell, 1991; Cherns et al., 2011). In freshwater sediments, the decomposition of organic matter follows the same pattern, but with the important difference that sulfate reduction is much less significant due to its lower concentration in these environments. Large organic matter concentrations can be degraded by methanogenesis, which releases CO_2 and sediments can quickly subsaturate with respect to $CaCO_3$. Although sulfate reduction is much less significant than in marine environments, it produces large amounts of iron oxides which are available to be reduced, directly generating alkalinity due to production of HCO_3^-. This process tends to neutralize the acid CO_2 formed by methanogenesis. Since bacterial activity is higher during warmer months, preservation would be affected by seasonality (Walker, 2001). Thus, preservation potential of calcium carbonate hard parts is highly variable, depending on the initial composition of water and the relative importance of methanogenesis and iron reduction, and to a less degree to sulfate reduction (Canfield and Raiswell, 1991).

At present studies that integrate taphonomic analysis and water chemistry composition are scarce in freshwater environments (Cummins, 1994; Nielsen et al., 2008). In particular, it is unknown how pH, organic matter, carbonates, and water chemistry composition affect mollusks preservation below the sediment-water interface in freshwater shallow lakes from the Pampa region. Specifically, the freshwater malacofauna of the region is characterized by low taxonomic diversity (richness vary between 1 and 11) and is dominated by epifaunal gastropods, mainly *Heleobia parchappii* and *Biomphalaria peregrina*. Most of the species represented in the area display a wide range of ecological tolerance, i.e., most species are found in different kinds of water bodies (for details see Tietze and de Francesco, 2010, 2012), and have been widely used for paleoecological and paleoenvironmetal reconstruction of the area (Prieto et al., 2004; de Francesco et al., 2013; Steffan et al., 2014; Pisano et al., 2015; as examples). However, the fossil record formation depends not only on ecological processes that affect life assemblages but also on taphonomic processes which modify death assemblages during and after their deposition (Beherensmeyer et al., 2000). Therefore, the knowledge about the dynamic and processes occurring within modern depositional environments will have a significant impact in the improvement of paleoenvironmental reconstruction quality based on the biological record.

The aim of the present work is to analyze the chemical composition of sediments, column and interstitial water in shallow lakes from the Pampa plain in order to highlight their relative influence on mollusk shells preservation. Results are compared with previous taphonomic studies carried out in the same study region (Cristini and De Francesco, 2012; Cristini, 2016; Tietze and de Francesco, 2012, 2014, 2017). In particular, the present contribution aims to compare (a) patterns between different compartments of the lakes (littoral versus open water) and (b) calcium carbonate saturation indices with respect to aragonite and calcite between column and interstitial water as well as to estimate potential redox from sediments.

2. Materials and methods

2.1. Study area

The region is a vast grassy plain that covers the central area of Argentina, which is characterized by a quite uniform relief, except for the presence of two mountains ranges (Tandilia and Ventania) towards the southeast (Diovisalvi et al., 2014) (Fig. 1). Soils of the area are generally fertile with a high content of nutrients, composed mainly of loess and with a marked capacity for cationic interchange, predominantly involving calcium (Rodrigues Capítulo et al., 2010). The climate is temperate humid or sub-humid with a mean annual temperature of 15 °C and a mean annual precipitation of 1100 mm (Feijoó and Lombardo, 2007). Precipitation patterns also display large variability, both geographically and inter-annually. This large interannual variability in combination with poorly developed drainage systems results in recurrent and extensive floods, alternating with drought periods (Fig. 2). All these processes affect the lake water residence time, the water content of soils, and the depth of the water table (Diovisalvi et al., 2014).

The gently slope of the landscape promotes the development of numerous permanent and temporary shallow lakes, which are very shallow (2 m) without thermal stratification except for short periods of time (Quirós and Drago, 1999; Fernández Cirelli and Miretzky, 2004). These lakes are characterized by an extensive littoral area, dominated by the emergent macrophyte *Schoenoplectus californicus* (C. A. Mey) Soják, which forms a complete ring around the lake, and an open water area, in general free of vegetation except for the submerged macrophytes *Myriophyllum elatinoides* and *Ceratophyllum demersum* (Stutz et al., 2010, 2012). These general characteristics are common for the three studies lakes. They have several environmental functions, among them charge and discharge of aquifers, flood control, water provision, climate regulation, recreational use, sportive fishing, and waste disposal (Fernández Cirelli and Miretzky, 2004), being naturally eutrophic or hypertrophic with most of them turbid due to the high amount of algae, while few are clear macrophyte-dominated lakes (Quirós et al., 2002; Quirós et al., 2006). Their degree of salinity is varied, ranging from

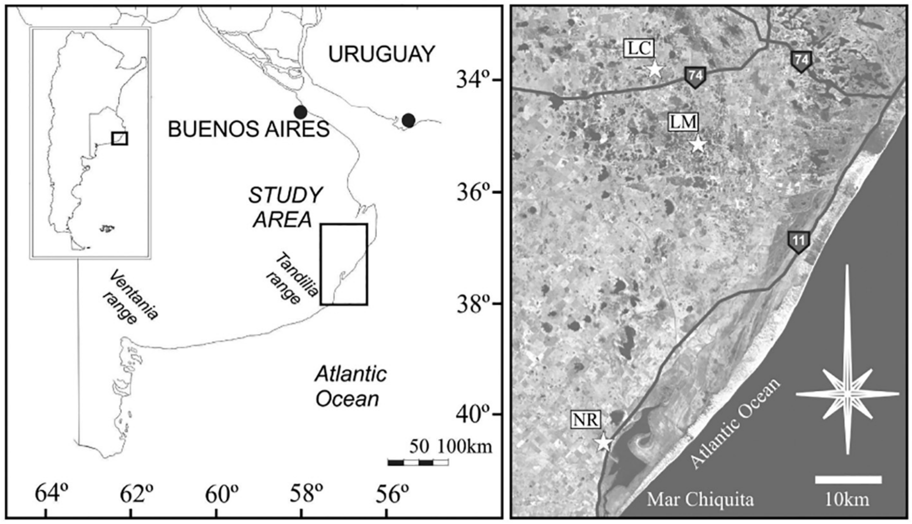


Fig. 1. Location of study area. NR = Nahuel Rucá, LM = Las Mostazas, LC = Los Carpinchos.

oligohaline to hyperhaline. Chemical compositions include sodium chloride bicarbonate or sodium bicarbonate chloride type, oligosulfate to sulfate type, and hypomagnesium to hemimagnesium type. The water column is almost always thermally homogeneous, promoting a high concentration of suspended particulate matter and low transparency, saturated with dissolved oxygen and with a high content of dissolved organic matter. Upper sediments are sandy silt, silt and clayey silt, with calcium carbonate content between 1% and 20%, and organic matter lower than 15% (Fernández Cirelli and Miretzky, 2004).

The study was conducted in three freshwater shallow lakes from the Southeastern Pampa plain, Argentina: Nahuel Rucá (NR) 37°37′21″S; 57°25′42″W; Las Mostazas (LM) 37°9′57″S; 57°14′50″W and Los Carpinchos (LC) 37°3′34″S; 57°19′56″W (Fig. 1). The physical characteristics as well as the connectivity to the surrounding streams vary among lakes. LM and LC are closed water bodies while NR is an open lake. Surface area is lower in LC (0.53 km^2) than in LM (1.61 km^2) and NR (2.45 km^2). Depth varies among lakes and within compartments of lakes, being shallower in littoral areas (littoral mean depth: 40 cm, 73.3 cm and 57.4 cm for NR, LM and LC respectively; open water mean depth: 78.7 cm, 108.9 cm and 92.7 cm for NR, LM and LC). The three lakes exhibit vegetated littoral areas and open areas generally free of vegetation. In fact, the percentage of macrophyte coverage of littoral rings is similar in the three lakes (NR: 50.2%, LM: 55.3% and LC: 54.7%), although LM presents a higher concentration of submerged macrophytes in some moment of the year (Gabriela Hassan pers. com).

2.2. Sampling

In order to fulfill the aims of the study, two types of sampling were performed in the three lakes: (1) a unique sampling of sediments for grain size analysis and (2) a seasonal sampling of sediments, column water (CW) and interstitial water (IW) for physico-chemical analysis. For grain size analysis only one sample of approximately 500 g of sediment was taken in each lake. The sampling of sediment, CW and IW was conducted during autumn (A) and winter (W) 2013 and summer (S) 2014. This sampling schedule was chosen to represent the natural variability in temperature of the water lake, with the warmest and coldest months represented, as well as an intermediate situation. In each lake, sediment, CW and IW samples were obtained in the littoral area (LIT) as well as in open water (OW). Five samples of sediments in each compartment were extracted for analysis of pH, moisture, organic matter and carbonates content using a gravity corer (Table 1). Sediment samples were taken within the first 10 cm below the sediment-water

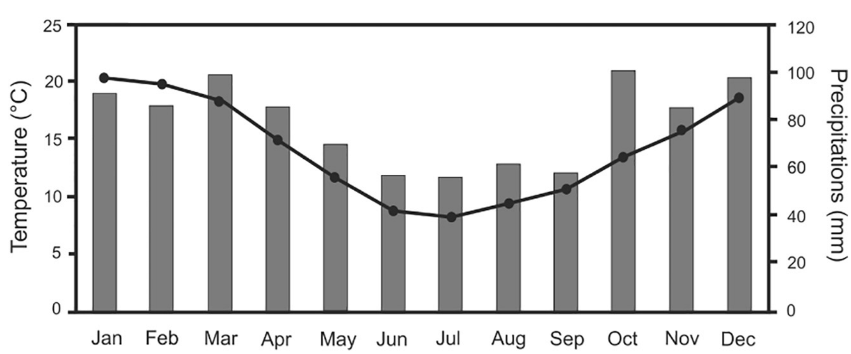


Fig. 2. Mean monthly precipitations (bar) and mean monthly temperature (line) recorded in the study area during the period 1974–2014. Data obtained from Meteorological National Service, Mar del Plata station.

Table 1
A, Summary of the sampling design showing the number of samples recorded in each type of sampling: grainsize (not seasonal) and sediments, column and interstitial water (seasonal), and in each compartment: littoral (LIT) and open water (OW). **B**, summary of the determination of environmental variables measured in sediments, column and interstitial water samples.

A.

	Grain size	Sediment		Interstitial water		Column water	
		LIT	OW	LIT	OW	LIT	OW
Autumm	1	5	5	5	5	5	5
Winter		5	5	5	5	5	5
Summer		5	5	5	5	5	5

B.

	Sediment		Interstitial water		Column water	
	LIT	OW	LIT	OW	LIT	OW
pH paste	X	X	–	–	–	–
Moisture	X	X	–	–	–	–
Organic matter	X	X	–	–	–	–
Carbonates	X	X	–	–	–	–
pH	–	–	X	X	X	X
Conductivity	–	–	X	X	X	X
Hardness	–	–	X	X	X	X
CO_3^{-2}	–	–	X	X	X	X
HCO_3^-	–	–	X	X	X	X
Cl^-	–	–	X	X	X	X
NO_3^-	–	–	X	X	X	X
NO_2^-	–	–	X	X	X	X
SO_4^{-2}	–	–	X	X	X	X
S^{-2}	–	–	X	X	–	–
Fe^{+3}	–	–	X	X	X	X
Ca^{+2}	–	–	X	X	X	X
Mg^{+2}	–	–	X	X	X	X
Na^+	–	–	X	X	X	X
K^+	–	–	X	X	X	X

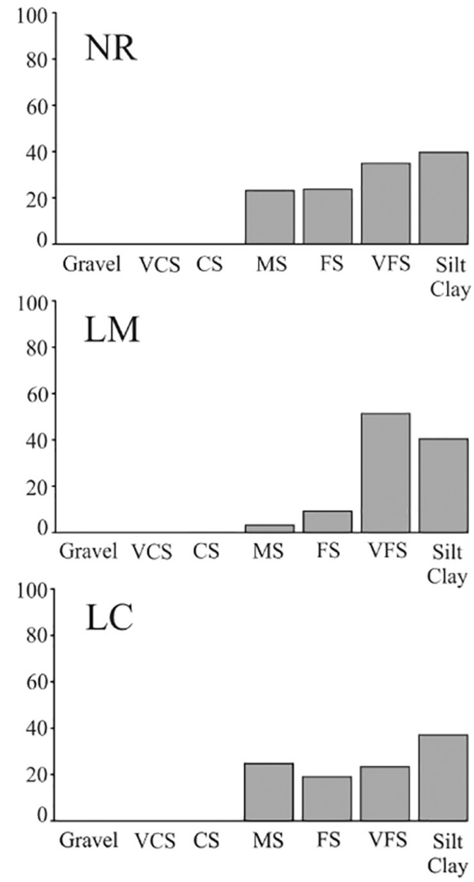


Fig. 3. Bar plot showing grain size of NR, LM and LC lakes. Values are expressed in percentage (%) (VCS = very coarse sand, CS = coarse sand, MS = medium sand, FS = fine sand, VFS = very fine sand).

interface. According to previous work (Cristini and De Francesco, 2012) the first 10 cm represents the zone below the sediment-water interface where the greatest taphonomic destruction takes place (TAZ).

Five samples of CW and IW were extracted for chemical analysis. In both CW and IW pH, conductivity, major ions ($Ca^{+2,}$ Mg^{+2}, Na^+, K^+, Cl^-, CO_3^{-2}, HCO_3^-, SO_4^{-2}), minor ions (NO_3^-, NO_2^-, S^{-2}, Fe^{+3}) and hardness ($CaCO_3$) were determined (Table 1).

IW was obtained from centrifugation of sediments (sampled with a gravity corer) at 2500 rpm for 25 min. CW was sampled with a sterile bottle of 1 l volume. Water samples were stored in ice until they were analyzed in the laboratory. For determining sulfurs, samples were fixed in the field with zinc acetate 2 N 4 drops each 100 ml, while for determining NO_3^- and total iron samples were fixed with concentrated sulfuric acid until pH was lower than 2 and stored in refrigerator until determination.

2.3. Laboratory analysis

Grain size was analyzed using the dry-sieving technique of Folk (1968). Categories of grain size included gravel (>2 mm), very coarse sand (2–1 mm), coarse sand (1–0.5 mm), medium sand (0.5–0.25 mm), fine sand (0.25–0.125 mm), very fine sand (0.125–0.0625 mm), and mud (silt and clay, <0.0625 mm).

For sediment samples, pH (in sediment paste) was measured and moisture, organic matter and carbonates content were determined. Moisture was calculated by drying 20 g of sediment for 24 h at ca.105 °C. Organic matter and carbonates content was calculated using the loss-on-ignition method (LOI) burning dry sediment for 4 h at 550 °C and 2 h at 1000 °C, respectively (Heiri et al., 2001).

CW and IW samples were analyzed in the Hydrogeochemistry and Isotopic Hydrology Laboratory of University of Mar del Plata within 48 h. During the first 24 h IW samples were centrifuged under 2500 rpm during 25 min to extract interstitial water from the sediment. Conductivity and pH were recorded and ions calcium (Ca^{+2}), magnesium (Mg^{+2}), sodium (Na^+), potassium (K^+), chloride (Cl^-), carbonates (CO_3^{-2}), bicarbonates (HCO_3^-), nitrate (NO_3^-), nitrite (NO_2^-), sulfate (SO_4^{-2}), sulfur (S^{-2}) were measured as well as hardness (mg/l of CaCO3) and total iron (Fe^{+3}).

2.4. Data analysis

With the aim of assessing if there exist seasonal and areal differences in sediment physico-chemical variables, a two-way permutational multivariate analyses of variance (PERMANOVA Anderson et al., 2008) was performed for each lake with 9999 permutations at $\alpha = 0.05$, using Euclidean distance (Legendre and Birks, 2012). Physico-chemical

Table 2
Seasonal mean values of physico-chemical variables measured in sediments from Nahuel Rucá (NR), Las Mostazas (LM) and Los Carpinchos (LC).

	Nahuel Rucá			**Las Mostazas**			**Los Carpinchos**		
	A	**W**	**S**	**A**	**W**	**S**	**A**	**W**	**S**
pH	7.6	7.6	7	7.9	8.3	7.6	7.6	7.9	7
Moisture (%)	70.4	66.9	58.1	66.2	65.9	71.1	69.5	81.4	74.9
Organic matter (%)	14.4	16.2	15.3	16.7	9.6	24.3	25.7	37	39.8
Carbonates (%)	2	2	3.4	1.2	1.1	1.7	2.7	2.5	2.8

variables were previously standardized in order to bring to some common scale previous any ordination process (Legendre and Birks, 2012; Clarke and Gorley, 2006). The PERMANOVA design consisted of two factors: *lake area*, (with two levels: LIT and OW) and *season*, (with three levels: A, W and S)·When main effects or interactions were significant, a posteriori comparisons were explored (Anderson, 2001). When significant differences were detected a Similarity Percentage analysis (SIMPER) was performed in order to identify the variables with higher

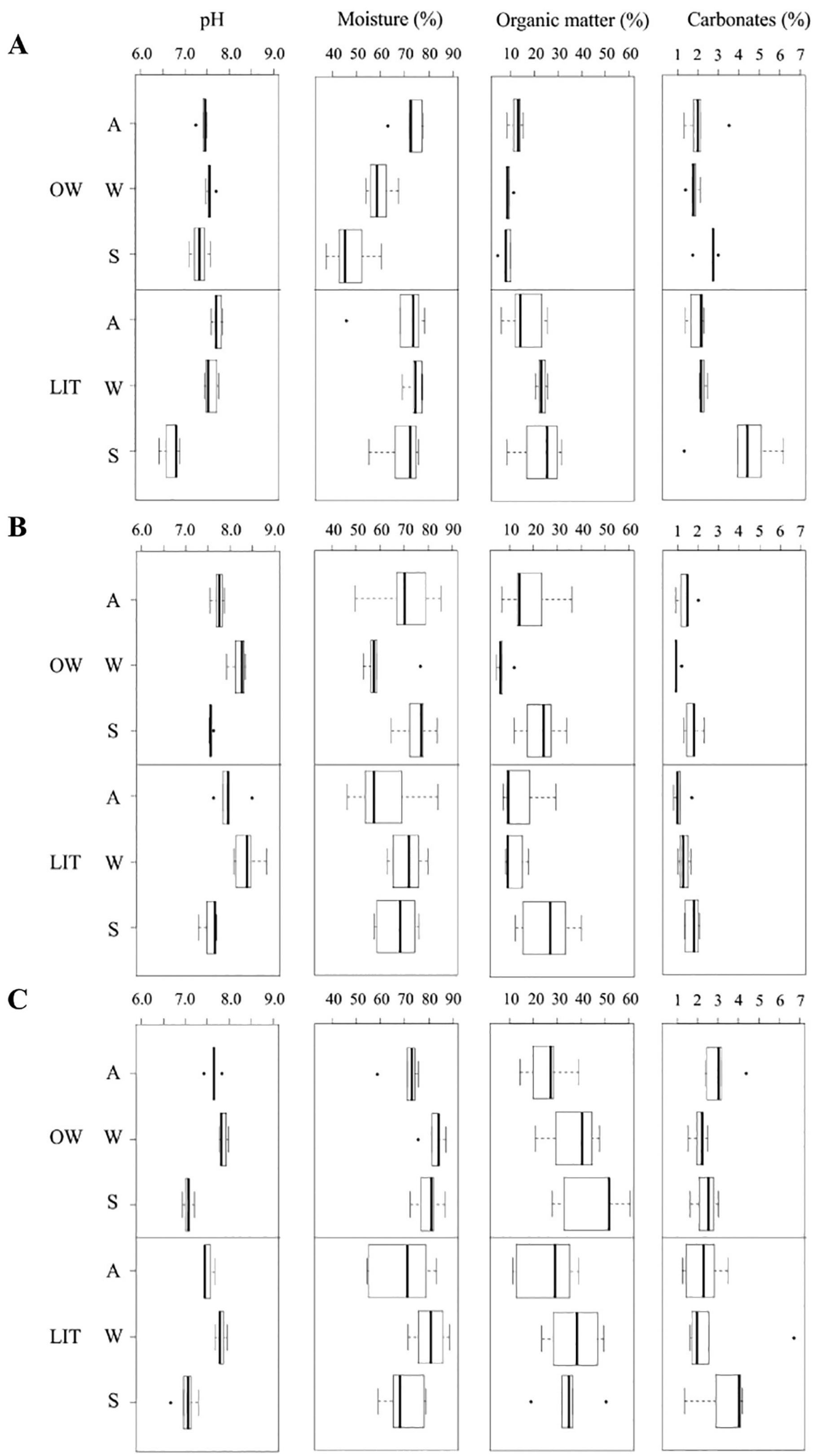


Fig. 4. Boxplot showing physico-chemical variables measured in sediments among seasons and between lake areas. **A**: NR, **B**: LM; **C**: LC (A = autumn, W = winter and S = summer, LIT = littoral, OW = open water).

contribution to the differences found with the PERMANOVA (Clarke, 1993). A non-metric multidimensional scaling graph (NMDS) was used to visualize the multivariate pattern among observations.

In order to characterize the type of water of the three lakes Piper diagram were performed. Piper is a simple and useful tool to the analysis of chemical water evolution as well as identification and classification of type of water.

To evaluate if there exist seasonal differences in chemical variables of IW and CW and between areas of the lakes a three-way PERMANOVA design (with 9999 permutations at $\alpha = 0.05$) was performed, using Euclidean distances (Legendre and Birks, 2012). The chemical variables were previously standardized in order to bring to some common scale previous to any ordination process (Legendre and Birks, 2012; Clarke and Gorley, 2006). The design consisted of three factors: 1-*season*, (with three levels: A, W and S), 2- *water type*, (with two levels: CW and IW), and 3- *lake area,* (with two levels: LIT and OW). Also SIMPER analysis was used as a posteriori test when significant differences were found. A Principal Component Analysis was used to visualize the multivariate pattern and to observe which variables are affected in the pattern. PCA was originally defined for multinormal distributions; however, deviations from normality do not necessarily produce biases in the analysis (Legendre and Legendre, 1998; and references there in). It is not necessary to fulfill multinormality criterion if the analysis is used with descriptive and exploratory purposes (Jolliffe, 2002). All analyses were performed using Plymouth Routines in Multivariate Ecological Research (PRIMER-e) (Clarke and Gorley, 2006).

Finally, calcite and aragonite saturation indices and the redox potential were calculated for each lake using Phreeqc 3.2.0 (Parkhurst and Appelo, 1999). Redox potential was calculated using redox pair sulfate/sulfur, S (+6)/S (−2). Is it important to highlight that the formation of aragonite or calcite depends on different environmental constraints such as temperature, salinity and Mg/Ca ration (Müller et al., 1972; Morse et al., 1997), and they can be replacing one to the other as these conditions change. Because of that the thermodynamic state of both is simultaneously analyzed by using PHREEQC, considering that the final conclusions are not affected by the specific mineral formed in each situation.

Table 3
PERMANOVA on Euclidean distances for physico-chemical variables measured in sediments: among seasons autumn (A), winter (W) and summer (S) and between lake areas: littoral (LIT) and open water (OW) from Nahuel Rucá (NR), Las Mostazas (LM) and Los Carpinchos (LC). [1]Pair-wise test comparison of interaction "Season × Lake area" for factor "season". [2]Pair-wise test comparison of interaction "Season × Lake area" for factor "lake area". [3]Pair-wise test comparison for factor season. Values in bold indicate significant *p* values at $\alpha = 0.05$.

Source	NR[a]		LM[b]		LC[c]	
	Pseudo-F	p	Pseudo-F	p	Pseudo-F	p
Season	9.36	**0.0001**	8.11	**0.0005**	7.27	**0.0001**
Lake Area	13.6	**0.0003**	0.54	0.5398	1.12	0.3144
SeasonxLake Area	6.39	**0.0006**	1.86	0.1527	1.09	0.354

[a]Comparison[1]	[a]Comparison[2]						
	LIT		OW		LIT vs. OW		
	t	p	t	p		t	p
A vs. W	1.50	0.1256	2.48	0.0233	A	1.29	0.223
A vs. S	2.94	**0.0188**	3.46	**0.0076**	W	6.04	**0.0085**
W vs. S	2.99	**0.0095**	2.61	**0.0086**	S	3.14	**0.0253**

[b]Comparison[3]			[c]Comparison[3]	
	t	p	t	p
A vs. W	1.87	**0.0485**	2.24	**0.0122**
A vs. S	2.04	**0.0455**	2.67	**0.0041**
W vs. S	4.60	**0.0001**	3.13	**0.0002**

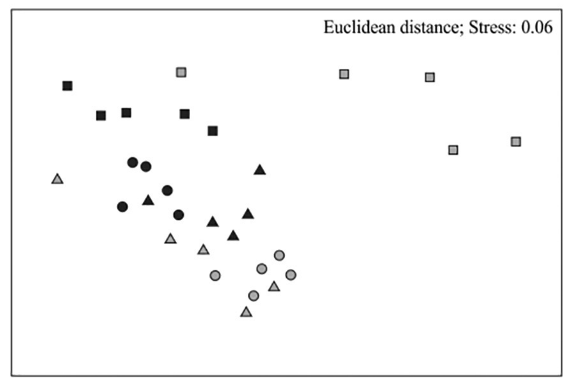
3. Results

3.1. Grain size and sediment characterization

The three lakes were characterized by sediments classified as medium sand, fine sand, very fine sand and mud, recording similar percentages in NR and LC. However, in LM lower values of medium and fine sand and higher values of very fine sand and mud were recorded. In the three lakes coarse sand was also recorded but in low percentages (<0.02%) (Fig. 3).

pH varied between 6.4 and 8.8. LM presented the highest values. Moisture content varied between 37.5% and 88.6% and organic matter between 4.5% and 60.6% with the highest values recorded in LC. Carbonates in sediment represented between 0.8% and 6.7% with lowest values in LM. In the three lakes, the lowest values of pH and the highest percentages of organic matter were recorded during the summer, with the exception of some observations in OW of NR and LIT area of LC,

A



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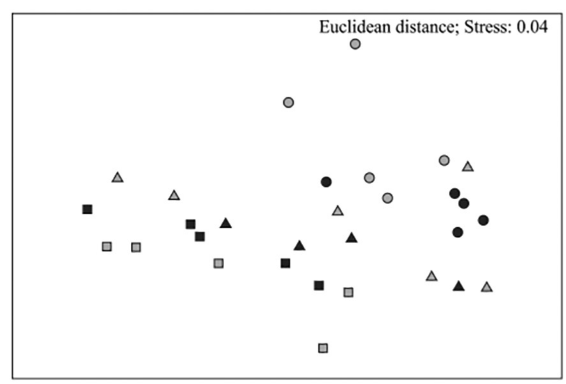

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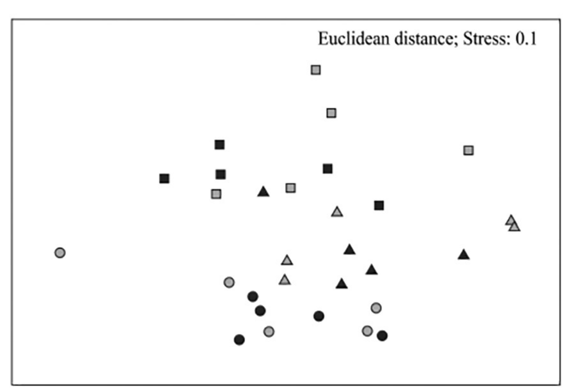

Fig. 5. NMDS of physico-chemical variables measured in sediments among seasons and between lake areas **A**: NR; **B**: LM; **C**: LC (triangle = autumn, circle = winter and square = summer, gray symbols = littoral, black symbols = open water).

Table 4
SIMPER analysis on Euclidean distances for physico-chemical variables measured in sediments, showing different percentages contribution (%) to the differences found among autumn (A), winter (W) and summer (S) and between littoral (LIT) and open water (OW) in Nahuel Rucá, Las Mostazas and Los Carpinchos lakes.

	pH	Moisture	Organic matter	Carbonates
Nahuel Rucá				
A vs. W	10.08	45.59	32.31	12.02
A vs. S	34.75	25.52	12.66	27.07
W vs. S	42.32	14.03	–	35.23
LIT vs. OW	17.68	27.51	35.62	19.19
Las Mostazas				
A vs. W	30.66	30.71	21.19	17.45
A vs. S	14.42	26.81	26.9	31.86
W vs. S	36.49	12.63	25.8	25.08
Los Carpinchos				
A vs. W	–	37.58	25.76	27.67
A vs. S	28.9	23.89	32.17	15.03
W vs. S	47.69	14.68	15.49	22.14

where the highest values of organic matter were recorded during autumn and winter, respectively. The higher values of moisture were mainly recorded during autumn and winter, with the exception of OW of LM where the higher values were recorded during the summer (Table 2, Fig. 4A-C).

In the three lakes physico-chemical variables registered in the sediment (pH, content of moisture, organic matter and carbonates) varied seasonally (NR: $p = 0.0001$, Table 3, Fig. 5-A; LM: $p = 0.0005$, Table 3, Fig. 5-B and LC: $p = 0.0001$, Table 3, Fig. 5-C). Besides, in NR there were significant differences between LIT and OW ($p = 0.0003$) and in the interaction of factors ($p = 0.0006$). A posteriori comparisons showed significant differences between all season (autumn, winter and summer) (Table 3; Fig. 5A-C). The only exception was NR that in LIT did not present significant differences between autumn and winter (Table 3, Fig. 5-A). SIMPER analysis showed that pH was the responsible variable to the seasonal differences found in the three lakes, with more acid values in summer, except in some cases where the rest of the variables had higher contribution than pH (e.g. between autumn and winter in NR and LC, and between autumn and summer in LM) (Table 4). Differences between LIT and OW in NR were mainly due to the higher content of organic matter and moisture in LIT (Table 4) (Fig. 4A-C).

3.2. Chemical composition of water

In general, the three lakes were characterized by neutral to alkaline and very hard waters, registering the highest hardness and conductivity in LM (Table 5). At the same time, water of LM was sodic chlorinated in the three seasons, while in NR and LC varied between sodium bicarbonate and sodium chloride, with more contribution of magnesium in some cases (Fig. 6).

Chemical dissolved species varied seasonally in the three lakes ($p = 0.0001$) and between column and interstitial water ($p = 0.0001$). However, there were no significant differences between LIT and OW ($p > 0.05$), with the exception of summer in NR ($p = 0.0229$) and LM ($p = 0.0539$, marginally significant) (Table 6, Fig. 7). Factor interactions were all significant, with the exception of interaction between water type and lake area in NR. The comparison in that interaction in LM and LC showed that chemical composition of column and interstitial water varied between LIT and OW in LM and only the interstitial in LC (Supplementary material, Fig. 7). The PCA graph illustrates the results obtained in the PERMANOVA (Fig. 7). It is observed a clear separation of the seasons as well as between column and interstitial water, with higher concentrations of carbonates and pH in the column and higher concentration of bicarbonates and lower pH in the interstitial water.

In the three lakes aragonite and calcite saturation indices of interstitial water was near the equilibrium or slightly subsaturated associated to more acidic pHs, while column water with more alkaline pHs was highly oversaturated. Besides, in autumn and winter the aragonite and calcite saturation indices were lower than in summer, with the exception of LM where the inverse pattern was observed in column water (Fig. 8). All redox potentials calculated from the redox pair SO_4^{-2}/S^{-2} in the interstitial water indicated reducing environment (Fig. 9).

4. Discussion

The three lakes studied in the present contribution exhibited differences in physico-chemical variables measured in the sediment as well as chemical composition of CW and IW. Yet, some common patterns could be recognized among lakes. For instance, the seasonal variation detected in sediments is related to the higher concentration of organic matter and consequently more acidic pH in warmest seasons (summer and autumn) and in the LIT area. This pattern was expected according to the abundant vegetation in the LIT area and the highest productivity recorded during warm seasons. It is known that LIT is a highly vegetated area with their own macroinvertebrates fauna associated (González Sagrario and Ferrero, 2013). Hence, LIT constitutes the greater source of organic matter available for decomposition and consequently the worst conditions for carbonate remains preservation. This idea is supported by results from Cristini (2016), who found that shell recovered from LIT displayed a worst state of preservation than those recovered from OW. Furthermore, this is linked to pH which diminishes as

Table 5
Seasonal mean values (mg/l) of ions measured in water from Nahuel Rucá (NR), Las Mostazas (LM)and Los Carpinchos (LC) lakes.(A = autumn, W = winter, S = summer, Cond. = Conductivity, Cl^- = chloride, CO_3^{-2} = carbonate, HCO_3^- = bicarbonate, NO_3^- = nitrate, NO_2^- = nitrite, SO_4^{-2} = sulfate, S^{-2} = sulfur, Fe^{+3} = total iron, Ca^{+2} = calcium, Mg^{+2} = magnesium, Na^+ = sodium and K^+ = potassium).

	Nahuel Rucá			Las Mostazas			Los Carpinchos		
	A	W	S	A	W	S	A	W	S
pH	8.14	8.11	8.52	8.52	8.37	8.62	7.96	7.91	8.39
$CaCO_3$	—	254.1	457.8	—	902.6	1499.7	503.5	384.2	817.2
Cond. (uS/cm)	1183.9	1139.2	1138.7	7550	7438.7	6697.8	2221	2623	4004.6
CO_3^{-2}	12.8	2.8	26.1	111.1	137.9	108.6	0	0	69.8
HCO_3^-	414.3	511.5	747.3	1059.1	1126.4	948.4	801.5	596.3	944
Cl^-	165.6	186.3	193.8	2057.4	1645.9	1894	439.9	595.6	957.6
NO_3^-	—	12.4	32.6	163.3	96.9	28.9	50.9	12.2	23.9
NO_2^-	31	—	—	0.93	—	—	0.43	—	—
SO_4^{-2}	96.9	149.5	163	503.7	457	401.5	205.5	577.5	569.1
S^{-2}	6.8	8	2.6	23.6	14.7	15.8	19.8	15.3	5.2
Fe^{+3}	—	0.35	1.8	4.7	—	1.2	0.5	3.1	1.2
Ca^{+2}	47.5	46.3	43	231.5	208.3	127.2	92	117.4	102.6
Mg^{+2}	18.8	33.1	84.3	136.6	91.3	283.6	65.6	22.9	134.6
Na^+	121.5	260.5	246.5	1140	1493.5	1406	341	469.3	340.5
K^+	12.6	22.8	24.2	82.5	75.4	50.4	30.4	31.3	35.2

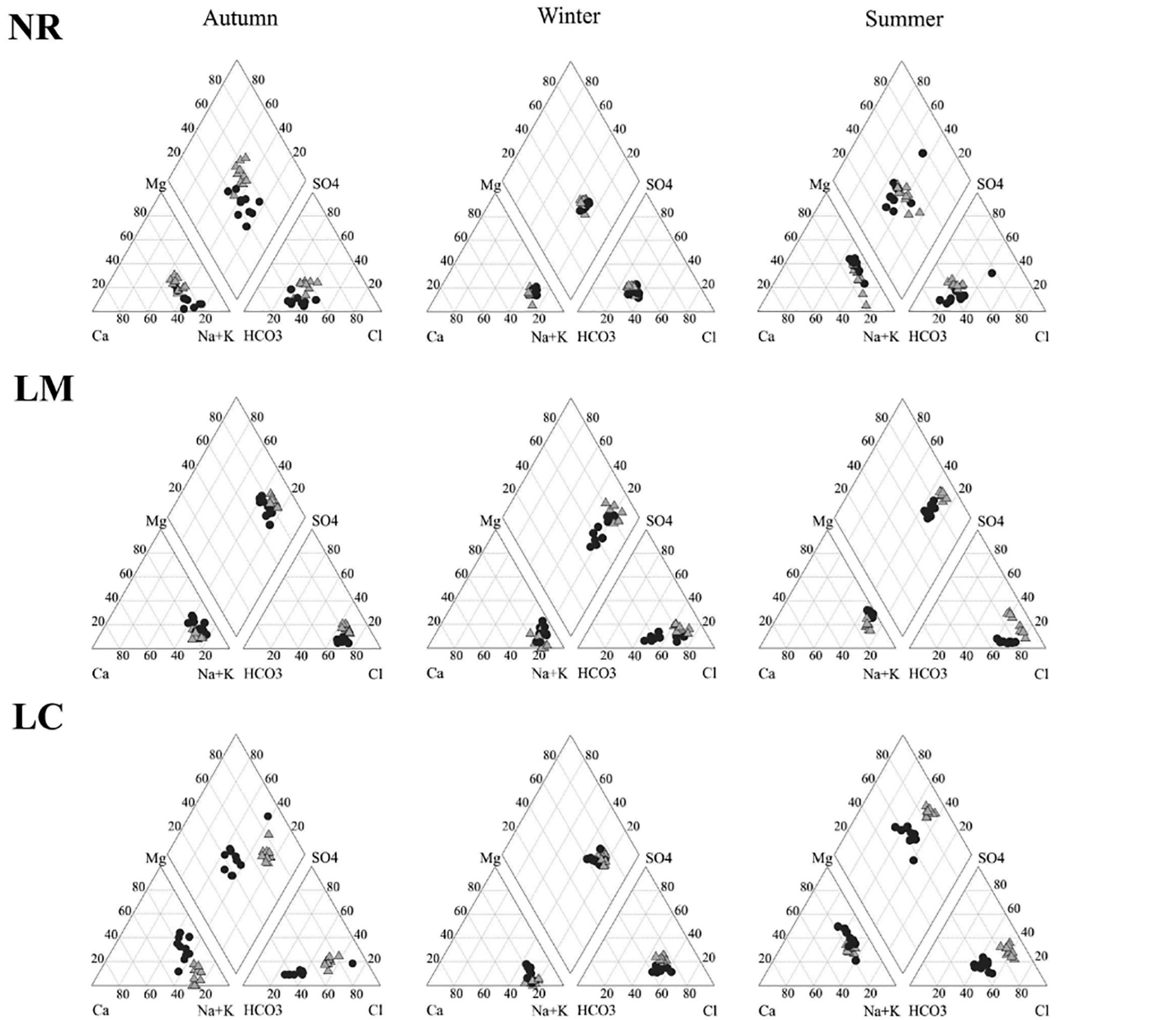

Fig. 6. Piper plot showing characterization of water column as well as interstitial water from NR, LM and LC. Circles indicate interstitial water and triangle water column, respectively.

consequence of organic matter degradation (Canfield and Raiswell, 1991; Cherns et al., 2011) and with the higher microbial activity during summer (Walker, 2001).

As it was previously explained, LIT is a more productive area than OW, but this pattern was only reflected in the physico-chemical variables measured in sediments from NR. There was not a unique variable measured in sediments that explain this observation, due to they were related between each other. The fact that LM and LC did not show differences could be linked to a reduction in lake areas as a consequence of wet and dry periods characteristic of the region (Quirós, 2005; Bohn et al., 2011), causing the lack of differentiation between compartments. Because of their shallowness, the dynamics of Pampean lakes are intricately tied to climate conditions, and the annual precipitation and evaporation volumes are of the same order of magnitude as their hydric volumes (Fernández Cirelli and Miretzky, 2004). In fact, lakes exhibit considerable interannual differences in their areas between floods and drought periods (Bohn et al., 2011; Diovisalvi et al., 2014 and references included there). In the particular case of NR, since it was the only lake that showed differences between LIT and OW, the presence of a floodgate which regulates the water level (maintaining it stable) could be neutralizing the climatic effect in this lake.

The predominance of different types of water among lakes may be tie in with higher evaporation, hydrologic regimen and soil type in the basement of the lakes. A determinant factor is the composition of the input waters. The lakes are fed by groundwater discharge and in some cases by stream water, which is also fed by groundwater discharge upstream. In particular, LM and LC are closed water bodies while NR is an open lake. The supply of the freshwater stream explains the lowest values of conductivity and less concentration of chlorides in NR. Moreover, groundwater in the basin has a changing composition depending mostly on the position in the flowpath and the system flow to which is belonging (Glok Galli et al., 2014) and this variability explains in an important proportion the differences between the lakes. According to soil charters developed by INTA (National Institute of agriculture and livestock technology), in LM there is a soil series named "El Tordillo" which is neither found in NR nor in LC. This soil present similar feature to the "type soil" which was described for a wide plain of ancient tide channels in the "Geomorphologic Subregion Samborombón Bay Marine

Table 6
PERMANOVA on Euclidean distances for chemical variables measured in water: among seasons: autumn (A), winter (W) and summer (S), between water type: column water (CW) and interstitial water (IW) and between lake areas: littoral (LIT) and open water (OW) from Nahuel Rucá (NR), Las Mostazas (LM) and Los Carpinchos (LC). (df = degree of freedom).Values in bold indicate significant p values at $\alpha = 0.05$.

Source	df	Pseudo-F	p (perm)
Nahuel Rucá			
Season	2	39.05	**0.0001**
Water type	1	46.49	**0.0001**
Lake area	1	0.79	0.5673
Season × Water type	2	8.88	**0.0001**
Season × Lake area	2	2.73	**0.0028**
Water type × Lake area	1	1.36	0.2162
Season × Water type × Lake area	2	1.83	**0.0505**
Residual	48		
Total	59		
Las Mostazas			
Season	2	20.52	**0.0001**
Water type	1	39.68	**0.0001**
Lake area	1	1.89	**0.0573**
Season × Water type	2	7.26	**0.0001**
Season × Lake area	2	1.88	**0.0059**
Water type × Lake area	1	1.08	**0.0002**
Season × Water type × Lake area	2	2.88	**0.0006**
Residual	43		
Total	54		
Los Carpinchos			
Season	2	49.19	**0.0001**
Water type	1	33.85	**0.0001**
Lake area	1	1.57	0.1469
Season × Water type	2	11.09	**0.0001**
Season × Lake area	2	2.30	**0.0053**
Water type × Lake area	1	3.07	**0.0045**
Season × Water type × Lake area	2	3.80	**0.0001**
Residual	44		
Total	**55**		

Plain". This series represent marine clay sediments with sodic alkalinity after 30 cm and slightly saline since 10 cm. These sediments that in the past were closely related to the sea, together with the evaporation and close hydrologic regime of the lake, could be the cause of the higher conductivity and chloride concentration in LM. Moreover, LM displayed higher concentration of carbonates, bicarbonates, calcium carbonate and calcium in water, compared to the other lakes. All of these characteristics could be positively affecting carbonate remains preservation since the high alkalinities and high concentrations of bicarbonate discourage dissolution of mollusk shells, thus allowing their preservation (Hagan et al., 1998).

Seasonal differences in chemical variables found in the three lakes would be related to the higher contents of Mg^{+2}, $SO_4^{-2}$and Cl^- during summer, probably as a consequence of the ion concentration caused by a greater evaporation by the increase in temperature. The seasonal pattern of saturation index recorded in NR, LC and in sediments of LM was expected because calcium carbonates present higher solubility at low temperature, expecting lower saturation index during coldest stations (autumn and winter). On the other hand, differences between CW and IW would be mainly linked to the higher concentration of HCO_3^- in IW and CO_3^{-2} in CW. This is expected due to the differences in pH between the two types of water and due to variation in carbonated species according to pH turn (Canfield and Raiswell, 1991; Wetzel, 2001). The higher concentration of organic matter in sediments would acidify the milieu by microbial decomposition, producing a more acid milieu in IW than in CW, causing that carbonated species equilibrium turn to HCO_3^- in IW and to CO_3^{-2} in CW. Consequently, aragonite and calcite saturation indices are near equilibrium or slightly subsaturated in sediments and strongly saturated in CW. This idea is in concordance with previous taphonomic studies conducted in the same area where dissolution was detected in mollusk shells deposited at (Tietze and De Francesco, 2017) and below the sediment-water interface (Cristini, 2016).

The negative values of potential redox obtained in sediments indicate a highly reducing environment. The detection of sulfide in the pore water of the studied environments indicates a redox state belonging to the sulfidic zone in the Berner (1981) zonification. Besides, it is also possible that the methanogenic state was reached in some cases where bubbling gasses were observed (personal observation). Both redox reactions produce acidification of the sediment pore water as it was previously explained in the introduction. In the sulfidic zone sulfate is reduce to sulfide and, if the sulfurs do not react, they accumulate and the solution acidifies, and vice versa (Canfield and Raiswell, 1991). The iron recorded in the sediments makes possible the formation of iron sulfides (solid phase) but the concentration would have not been high enough since saturation indices reported are near to equilibrium or slightly subsaturated in sediments. As it has been said, redox mechanisms are able to affect the saturation state of carbonates due to changes in pH. Consequently, the preservation potential of shell remains could be negatively affected. Indeed, previous taphonomic studies performed in these lakes indicated the existence of destructive processes, such as

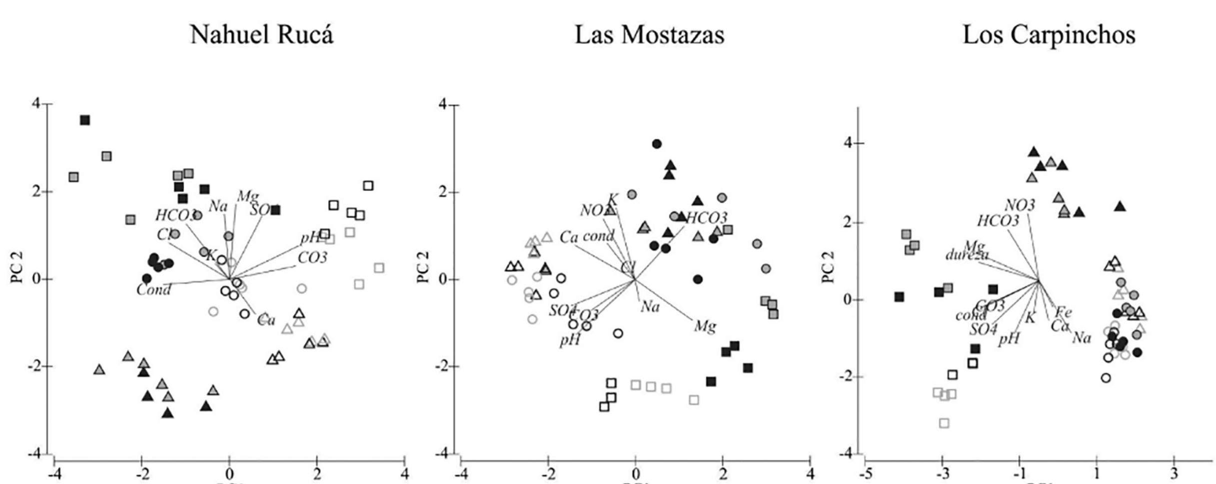

Fig. 7. PCA plot of NR, LM and LC lakes, showing multivariate pattern of column water chemical composition (empty symbols) and interstitial water chemical composition (fill symbols), among seasons (triangle = autumn, circle = winter and square = summer), and between LIT (gray) and OW (black) (Cond.= conductivity, Cl= chloride, CO_3 = carbonate, HCO_3 = bicarbonate, NO_3 = nitrate, SO_4 = sulfate, Fe = total iron, Ca = calcium, Mg = magnesium, Na = sodium and K = potassium).

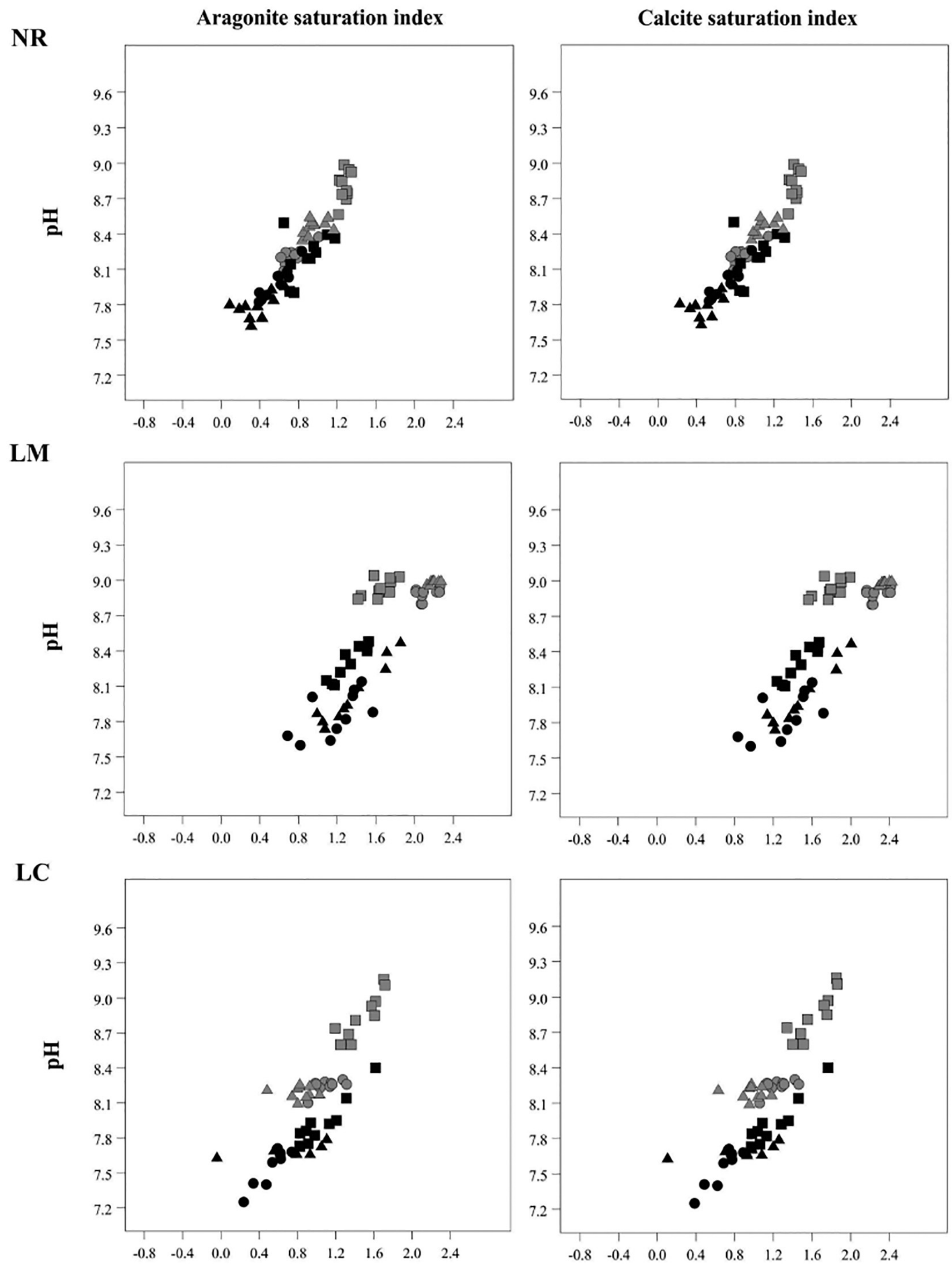

Fig. 8. Scatter plot showing aragonite and calcite saturation index vs. pH of column water (gray symbols) and interstitial water (black symbols) in autumn (triangle), winter (circle) and summer (square) in NR, LM and LC lakes.

dissolution, that diminish the abundance of individuals and the richness of species through depth (Cristini and De Francesco, 2012; Cristini, 2016) compared to those remains placed at the sediment-water interface (Tietze and De Francesco, 2012, 2014).

In general, and based on previously discussed, we are able to say that in these shallow lakes of temperate and humid climate, which characterized Pampa region, carbonate remains preservation is expected to vary depending on the depositional zone. In a horizontal zonation of the lakes, the littoral area is expected to have the worst conditions for preservation due to the higher concentration of macrophytes, which constitutes a greater source of organic matter available for decomposition. The organic oxidation would produce corrosive pore waters due to pH lowering. However, we are able to find high concentration of shell remains since mollusks communities live associated mainly to the emergent and submerged vegetation and shallow depths. Regarding a vertical zonation of the lakes, carbonate remains preservation will vary depending on if they are in direct contact to the column water or below the sediment-water interface, where the milieu is more reducing and acid (Fig. 10). Moreover, these patterns will be subject to variation with the characteristic seasonality of the region

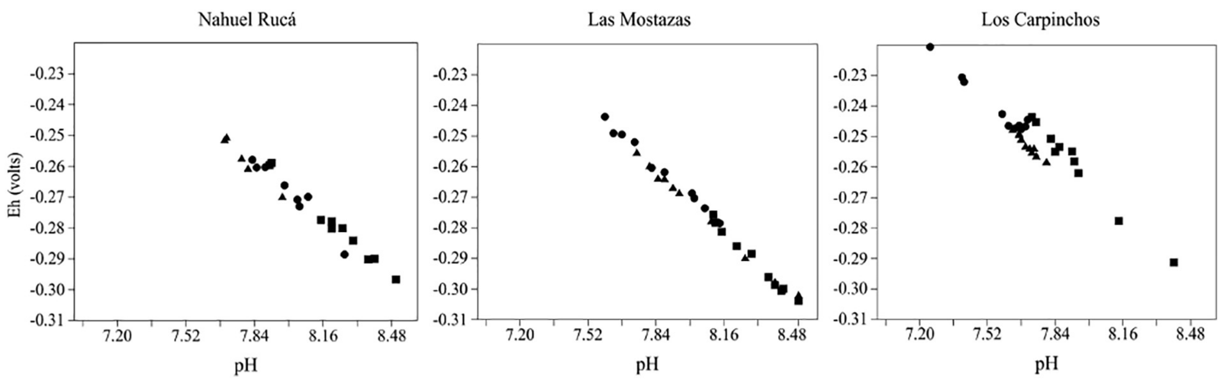

Fig. 9. Scatter plot showing redox potential measured in sediments vs. pH in autumn (triangle), winter (circle) and summer (square) for NR, LM and LC lakes.

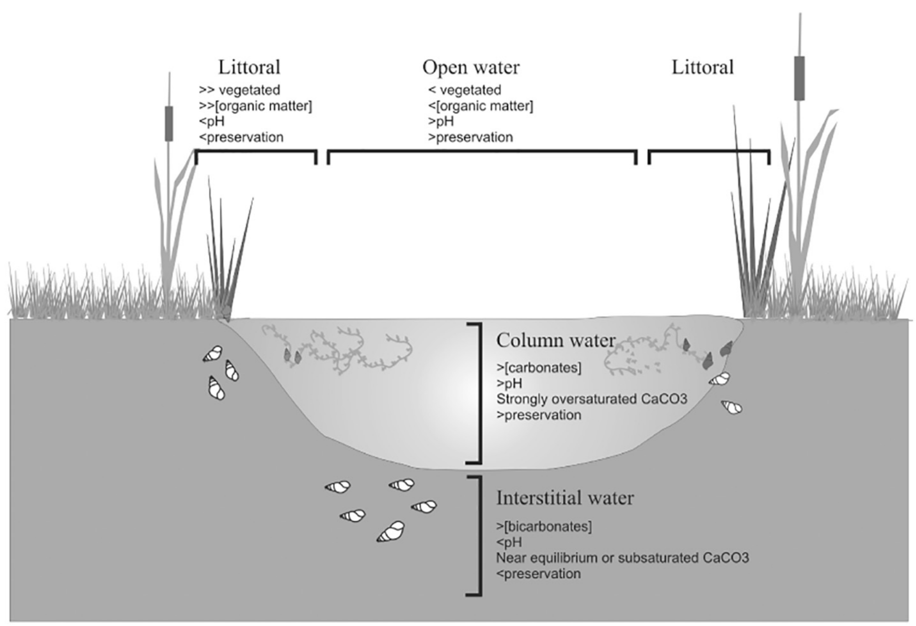

Fig. 10. Graphical abstract of the process taking place in different compartments of the lake (LIT and OW) as well in different water type (CW and IW).

since the destructive conditions are exacerbated during warm months, such as autumn and summer in the present work. Finally, this description can be subject to variation among lakes. In the present study, LM showed higher conductivity, alkalinity and higher concentration of carbonates and probably will exhibit a better mollusk preservation compared to NR and LC.

5. Conclusions

LIT settings has less favorable features to shell preservation than OW ones due to the highest concentration of organic matter and lower pH. This is exacerbated during warmer months (autumn and summer) due to the greater microbial activity.

Sediments from Southeastern Pampa freshwater shallow lakes are highly reducer indicating that organic matter decomposition mainly occur by methanogenesis and sulfate reduction, acidifying the milieu and producing subsaturation of calcium carbonate. This idea is supported by aragonite and calcite indices in equilibrium or slightly subsaturated which lead to think that carbonate remains within sediments would be probably subject to dissolution. Mollusk shells are not expected to dissolve if they are in contact with column water which is strongly oversaturated with respect to calcium carbonate.

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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.2017.06.043.

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