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Use of radon-222 to assess the groundwater inflow in a phreatic lake of a dune field (San Luis, Argentine)

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ABSTRACT

In the southeastern portion of San Luis Province (33°53’10”-34°19’00” S and 65°42’00”-65°20’00” W), the sand dune landscape hosts a lacustrine system with more than 200 water bodies, where the water table reaches the surface and fills the deepest depressions. The aim of this study is to analyze surface-groundwater interactions using the radioactive isotope radon-222 (^{222}Rn) in a lake known as “Los Pocitos”. During September 2017, *in situ* ^{222}Rn determinations were performed in the air; in the water/sediment interface and in surface waters at 6 sampling stations, as well as in 4 groundwater samples collected in the dune and the lake mudflat using the RAD-7 equipment (Durridge Co.). Surface waters are of the HCO_3^- - Na^+ - K^+ type, with mean pH and electrical conductivity in Lake Los Pocitos of 8.7 and 1232 µS cm^{-1}, respectively, whereas the groundwater is of the HCO_3^- - Ca^{+2} type, with variable pH and electrical conductivity values. In the northern portion of the lake, concentrations of ^{222}Rn in surface water were ~ 70 Bq m^{-3}, one order of magnitude greater than those of the southern sector, which registered values < 5 Bq m^{-3}. By means of a ^{222}Rn mass balance model it was possible to determine that the groundwater discharge occurs in the northern sector, with an inflow discharge rate of about 185.3 ± 39.1 m^3 d^{-1}, whereas in the southern sector, an outflow from the lake to the surrounding aquifers can be detected. The intense groundwater inflow into these lakes may explain their relatively low salinity under a semi-arid climate in which evaporation by far exceeds direct rainfall input.

Keywords: aquifer, environmental tracers, geochemistry, lake, radioactive isotopes.

Uso de radón-222 para determinar el flujo de agua en una laguna freática de la llanura medanosa de San Luis (Argentina)

RESUMEN

En el sector sureste de la provincia de San Luis (33°53’10”-34°19’00” S y 65°42’00”-65°20’00” O), sobre un paisaje medanoso, se desarrolla un sistema lagunar compuesto por más de 200 depresiones someras en donde los niveles freáticos alcanzan la superficie. El propósito de este trabajo es analizar las interacciones agua superficial-subterránea utilizando el isótopo radioactivo radón-222 (^{222}Rn*) en una de estas lagunas, conocida como “Los Pocitos”. Durante septiembre de 2017 se realizaron, in situ, determinaciones de* ^{222}Rn *utilizando el equipo portátil RAD-7 (Durridge Co.), en el aire, en la interfase agua/sedimento, en 6 estaciones de muestreo alrededor de la laguna, así como también en 4 muestras de agua subterránea tomadas en la duna y en las márgenes de la laguna. Los resultados obtenidos indican que las aguas superficiales son del tipo* HCO_3^- *-* Na^+

- K^+ y tienen un pH y una conductividad eléctrica promedio de 8,7 y 1232 µS cm^{-1}, respectivamente, mientras que las aguas subterráneas son HCO_3^- - Ca^{+2} y tienen un pH y una conductividad eléctrica variable. En el sector norte se presentan las mayores concentraciones de ^{222}Rn en el agua superficial, de aproximadamente 70 Bq m^{-3}, un orden de magnitud mayor que en el sector sur, donde se registraron concentraciones < 5 Bq m^{-3}. Mediante un modelo de balance de masa de ^{222}Rn se estimó que la descarga de agua subterránea ocurre en el sector norte, con un caudal de 185,3 ± 39,1 m^3 d^{-1}. Por el contrario, en el sector sur hay un aporte de agua desde la laguna hacia los acuíferos circundantes. Este flujo de agua subterránea hacia las lagunas podría explicar su relativa baja salinidad en un clima semiárido en el que la evaporación supera a las precipitaciones en el balance hídrico.

Palabras clave: acuífero, geoquímica, isótopos radiactivos, lago, trazadores ambientales.

Introduction

In the last decades, the agricultural frontier of the Argentine Pampean region has been expanding as a result of technological advances, irrigation systems and the use of agrochemicals. Areas that were clearly destined for cattle production have been replaced by crop activity (Viglizzo *et al.*, 2010). This causes variations in the water table which, in turn, modify the dynamic and chemical interrelations with surface water bodies. Shallow lakes in this region are very important to wildlife, as they include a considerable diversity of aquatic fauna, unusual in other environments of this semi-arid region, and they also offer important watering holes for livestock.

When estimating lake water balance, groundwater contribution is usually assumed to be insignificant due to the difficulty of quantifying the groundwater component (e.g., Rosenberry *et al.*, 2015), which in turn, introduces an important bias in the hydrological characterization. In closed-basin lakes, a negative water balance results in low water levels and the concentration of dissolved salts (e.g., Córdoba et al., 2014; Fritz, 1990). Identifying water exchange between the groundwater and surface water bodies leads to the understanding of the possible responses of the sys-

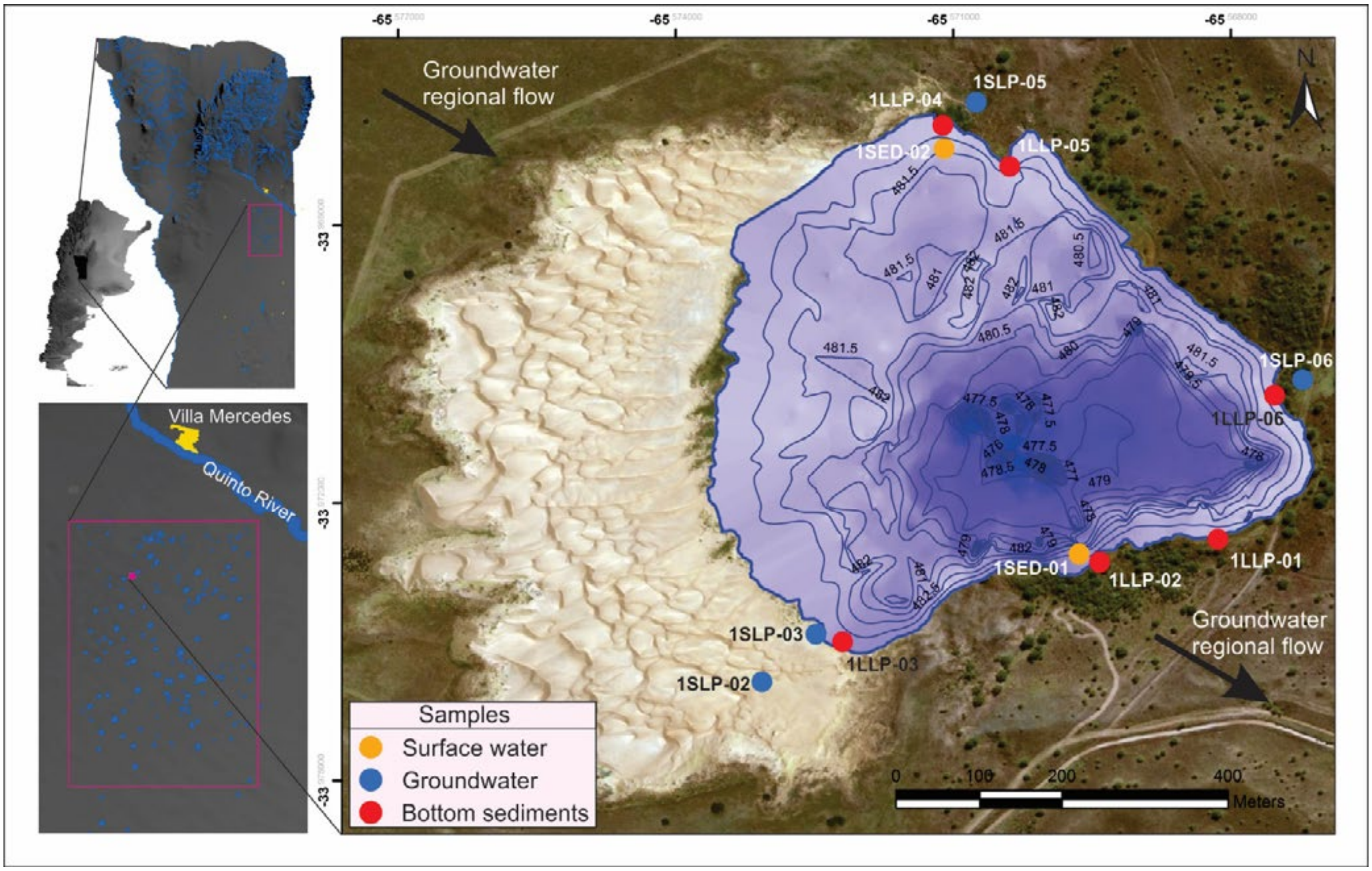

Figure 1. Location of the study area and sampling points at Lake Los Pocitos.
Figure 1. *Ubicación del área de estudio y puntos de muestreo en la Laguna Los Pocitos.*

tem to climate and land use changes, and also helps to improve water resource management policies. During the past decades, several studies have been carried with aim of analyzing the flow of water and solutes across the sediment–water interface of the lakes One of the most common approaches used to investigate groundwater-lake water interactions, which are often patchy and diffuse and therefore difficult to evaluate using traditional methods, is the use of geochemical tracers, such as ^{222}Rn. The source of ^{222}Rn is ^{226}Ra, which belongs to the U disintegration chain, and is included in rock forming minerals, and also in soils and sediments. Thus, the amount of ^{222}Rn in the aquifers is determined by their lithology and geochemical composition. The activity of ^{226}Ra in rainfall is negligible. For these reasons, its occurrence in surface waters is derived mostly from groundwater discharge, but due to a relatively short half-life of ^{222}Rn (3.8 days) and the rapid gas exchange to the atmosphere, high ^{222}Rn activities are measured in surface waters when active groundwater discharge occurs. Several studies have examined the surface groundwater interactions in lake systems by means of ^{222}Rn, such as Arnoux *et al.* (2017), Dimova *et al.* (2013), Kluge *et al.* (2007), Schmidt *et al.* (2010), Tuccimei *et al.* (2005), to mention a few.

In order to investigate why the lakes in the region neither reduced their areas nor are as salty as they should be with high evaporation rates, the aim of this current study is to identify the spatial distribution and magnitude of groundwater discharge into Lake Los Pocitos by means of a mass balance model.

Study area

The eastern region of San Luis province in Argentine shows an eolian landscape referred to as the “Pampean Sand Sea” (Iriondo and Kröhling, 1995; Tripaldi and Forman, 2016). This Quaternary dune field contains eolian sandy sediments characterized by parabolic dunes (NE-SW orientation) and more than 200 water-filled blowout depressions of variable size, most of them of permanent regime (Tripaldi *et al.*, 2013; Vilanova *et al.*, 2015). The area presents an incompletely integrated surficial drainage system, with numerous endorheic basins with no evidence of rivers or streams (Tripaldi *et al.*, 2013, Viglizzo *et al.*, 2009). The only important permanent water course is the Quinto River, located in the north of the study area.

Local soils have low water retention capacity and surface horizons with low organic matter content. Regarding the vegetation cover, Calden forest and grasslands prevail (Colazo, 2012). Climate in the study area is mesothermal, sub-humid to semi-arid, with a mean annual temperature of 15.6 °C, in winter 8 °C and in summer 24 °C (AD 1981-2010, Servicio Meteorológico Nacional). Rainfall occurs mainly in spring and summer, between October and March, with a dry winter. The mean annual rainfall is 618 mm, with a minimum of 230 mm and a maximum of 1,205 mm (Villa Mercedes, series 1903 to 2016, Fig. 2). During the warm months, evaporation rates are significant, frequently generating a negative water balance.

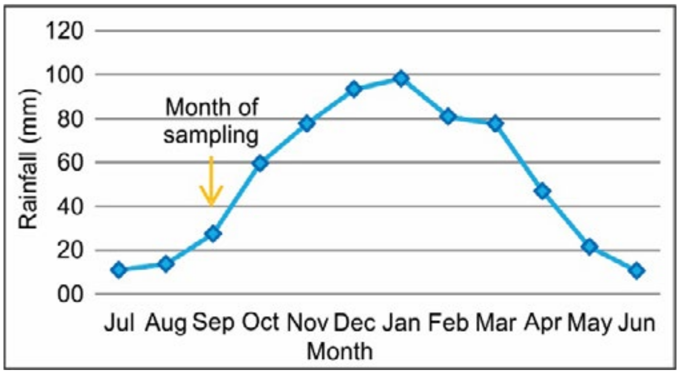

Figure 2. Monthly mean rainfall series in the city of Villa Mercedes (1903-2016 record period)
Figure 2. *Serie precipitaciones media mensuales Villa Mercedes 1903-2016.*

The surfaces and depths of the lakes can range from 20 to 100 ha and from 2 to 12 metres, respectively. The groundwater regional flow follows a NW-SE direction, which agrees with the general land slope (Ceci and Coronado, 1981). Lake Los Pocitos is one of these numerous permanent water bodies, located 45 km south of Villa Mercedes city (Fig. 1), with an area of 25 ha and a mean depth of 2 m.

Material and methods

Sampling and analyses

Surface water was collected from Lake Los Pocitos and the groundwater was sampled by hand-dug wells of 0.8 to 2.2 m below the surface around the lake during the dry season in September 2017. Groundwater was sampled from the sand dunes and from modern sediments and soils. The dunes are composed of aeolian sands which correspond to lithic feldsarenites and feldspathic litharenites (Tripaldi *et al.*, 2010). Modern sediments present deep soils with a shallow surficial horizon (Colazo, 2012). Field determinations consisted of temperature (T), pH, oxidation reduction potential (ORP), electrical conductivity (EC), total dissolved solids (TDS), and alkalinity. Hydrochemical characterization was made on 6 samples collected from Lake Los Pocitos and 4 groundwater samples. The samples were filtered *in situ* through 0.22 μm cellulose acetate membrane filters (Millipore Corp.) and divided into two aliquots. The major anions (Cl^- and SO_4^{-2}) were analyzed by UV spectrometry (Metrhom IC 850 Professional) at CNEA laboratory (Bariloche, Argentina). The other aliquot was acidified (pH <2) with ultrapure HNO_3 for the analytical determination of the major cations by inductively coupled plasma-mass spectrometry (ICP-MS, Activation Laboratories Ltd., Ancaster, Ontario, Canada).

The lake perimeter was measured with a Garmin GPS equipment. The lake area, volume and mean depth were calculated with a geographical information system software. The Thiessen polygon method was used to estimate the sub-areas. The depths were measured over the entire area of the lake, approximately every 3 metres by digital sensor NAV-MAN 4433 sonar.

Measurements of ^{222}Rn were made with the portable detector RAD-7 (Durridge Inc.), which provides precise measurements *in situ* and in real time. RAD-7 measures the activity of ^{222}Rn by counting its alpha-emitting daughters (^{218}Po and ^{214}Po). The surface water was pumped with a bilge pump at 6 points located ~ 1.5 m from the lake margin. To measure the ^{222}Rn activity in the lake water, an air-water exchanger (AQUA, Durridge Inc.) was used. AQUA equipment consists of a plastic cylinder where the water flow is continuous and ^{222}Rn outgasses from water until the solubility equilibrium is reached. The duration of each time series was 80 minutes, and a new ^{222}Rn concentration was obtained every 20 minutes. The obtained data were corrected for humidity and temperature. The first correction is carried out automatically by the equipment, whilst the second was performed using the equilibrium coefficients, since the partition of ^{222}Rn between the liquid and gas phases is controlled by temperature. For this purpose, continuous water temperature measurements were carried out every 5 minutes using a HOBO data logger (Onset Co.). On the other hand, ^{222}Rn concentrations in groundwater samples were measured with the RAD H20 (Durridge Inc.) in 250 mL glass bottles. The measurement is made in a closed air circuit between the water sample and the RAD-7.

Diffusion of ^{222}Rn from the bottom sediments to the overlying water column can be an important source of ^{222}Rn. Thus, an estimation of this diffusive benthic flow is necessary. Two bottom sediment samples (northern and southern sectors of the lake) were collected for this purpose. These measurements were done following the procedure described by Corbett *et al.* (1998). ^{222}Rn in the air above the lake surface was also measured in order to estimate the exchange between the air-water interface, which represents a ^{222}Rn potential loss. The wind speed was measured every 30 minutes with a Digital Anemometer (Digital Instruments).

To estimate the ^{222}Rn that results from the decay of the dissolved ^{226}Ra in the water column, approximately 150 L of lake water was circulated through a cartridge filled with acrylic fibers impregnated with manganese oxide, which retains the radium dissolved in the water (Moore and Reid, 1973). The average concentration of ^{222}Rn resulting from the ^{226}Ra decay in the lake water was determined using the RAD-7 following the measurement protocol described by Peterson et al. (2009).

^{222}Rn mass balance

The mass balance model used to determine groundwater-surface water interactions includes all input and output flows of ^{222}Rn from the water body (Tuccimei *et al*, 2005). The flux terms that need to be considered include: (1) flux of ^{222}Rn in the sediment-water interface (*Jbenthic*), which in turn is composed of the diffusion of ^{222}Rn from bottom sediments in the lake water (*Jdiffusion*) and the advective groundwater through sediments (*Jadvective*); (2) production of ^{222}Rn in the water column by ^{226}Ra decay (*Jproduction*); (3) ^{222}Rn loss to the atmosphere through the air-water interface (*Jatm*); and (4) ^{222}Rn decay in the water column (*Jdecay*).

^{222}Rn mass balance is calculated following the general formula:

$$Jbenthic + Jproduction - Jatm - Jdecay = 0 \quad \text{(Equation 1)}$$

Advective flow of groundwater can be calculated according to:

$$Jadvective = Jbenthic - Jdiffusion \quad \text{(Equation 2)}$$

Velocity of groundwater flow is obtained according to Corbett et al. (2000):

$$Gvel = Jadvective / \text{Concentration of } ^{222}Rn \text{ in groundwater} \quad \text{(Equation 3)}$$

Groundwater discharge (*Dsub*) can be estimated as follows:

$$Dsub = Gvel * \text{Lake area} \quad \text{(Equation 4)}$$

Results and discussion

Lake Los Pocitos has an area of 25 ha with a mean depth of 2 m and a maximum depth of 11 m (Fig. 1). The parameters measured in the lake water and groundwater from the wells are shown in Table 1. The lake water temperature is homogeneous, with an average value of 16 °C. In the groundwater, the temperature is higher, reaching values of ~25 °C in the shallowest wells located in the dune zone. The lake pH is relatively constant, ranging from 8.5 to 8.8, indicating alkaline waters. On the other hand, the groundwater is slightly less alkaline with pH values of between 7.8 and 8.4. The average electrical conductivity of the lake water is 1231 µS cm^{-1}, indicating that it is fresh to brackish, concentrated due to evaporation processes. The groundwater is less concentrated. Low electrical conductivities in the dune wells (i.e., 1SLP-02 and 1SLP-03) are interpreted as fresher water, which allows us to infer a recharge by atmospheric precipitations in the surrounding dune.

According to the Piper diagram, the lake waters are of the HCO_3^- - Na^+ - K^+ type, with a slight variation

Samples	1LLP-01	1LLP-02	1LLP-03	1LLP-04	1LLP-05	1LLP-06	1SLP-02	1SLP-03	1SLP-05	1SLP-06
Depth (m bs)							2.2	0.85	1.83	2.21
T (°C)	15.50	15.80	15.50	16.30	16.60	16.70	25.20	24.80	19.80	-
pH	8.81	8.56	8.55	8.60	8.85	8.82	8.43	7.86	8.02	-
Eh (mV)	349.99	334.68	376.99	296.17	377.87	360.77	380.10	394.50	396.60	-
TDS (mg L^{-1})	611	611	618	587	609	668	89	256	507	826
EC (µS cm^{-1})	1219	1218	1234	1169	1216	1333	177	511	1014	1651
Na^+ (mg L^{-1})	200.00	181.00	198.00	169.00	178.00	200.00	18.10	31.90	51.00	200.00
Mg^{+2} (mg L^{-1})	27.80	24.90	27.20	28.70	24.90	29.10	9.37	22.10	31.80	21.80
K^+ (mg L^{-1})	16.80	15.10	16.70	16.30	15.40	17.40	4.87	7.42	26.90	33.70
Ca^{+2} (mg L^{-1})	32.70	32.60	32.80	33.00	32.50	32.90	25.30	38.90	150.00	78.00
Cl^- (mg L^{-1})	76.00	87.00	88.00	94.00	64.00	61.00	2.20	1.40	6.90	150.00
SO_4^{-2} (mg L^{-1})	150	159	164	177	125	117	4	2	5	130
CO_3^{-2} (mg L^{-1})	21.59	20.39	27.59	31.19	26.39	93.57	12.00	0.00	0.00	0.00
HCO_3^- (mg L^{-1})	282.98	295.18	296.40	286.64	285.42	289.08	131.73	340.31	689.76	497.04
^{222}Rn (Bq m^{-3})	2.2 ± 1.2	6.1 ± 0.7	6.9 ± 3.7	76.6 ± 14.0	63.6 ± 12.1	0.8 ± 0.8	10082 ± 122	3023 ± 205	7372 ± 435	8782 ± 875

Table 1. Depth groundwater, physico-chemical parameters and activity of ^{222}Rn measured *in situ* in the lake water and groundwater.
Tabla 1. *Profundidad de los pozos, parámetros físico-químicos y actividad de ^{222}Rn medidos in situ en la laguna y aguas subterráneas.*

in the anionic composition to a mixed classification (Fig. 3). On the other hand, the groundwater is of the HCO_3^- - Ca^{+2} type, with the exception of sample 1SLP-06, which shows intermediate hydrochemical characteristics between both reservoirs.

The results of the ^{222}Rn mass balance are shown in Table 2, where the uncertainties for all the terms of the mass balance equation (different sources and sinks)

Samples	Jatm	Jbenthic	Jadvective	Gvel	Dsub
		Bq m^{-2} day^{-1}		cm day^{-1}	m^3 day^{-1}
1LLP-01	0.8 ± 0.5	1.8 ± 0.8	-1.0 ± 0.8	-0.01 ± 0.01	-1.9 ± 1.3
1LLP-02	2.2 ± 0.2	3.3 ± 0.5	0.5 ± 0.6	0.01 ± 0.01	3.4 ± 5.5
1LLP-03	0.9 ± 0.5	1.9 ± 0.8	-0.9 ± 0.9	-0.01 ± 0.01	-8.5 ± 6.7
1LLP-04	9.8 ± 1.8	16.3 ± 2.6	16.2 ± 2.9	0.20 ± 0.02	83.6 ± 20.5
1LLP-05	8.2 ± 1.6	14.6 ± 2.4	14.6 ± 2.6	0.18 ± 0.05	100.2 ± 25.0
1LLP-06	0.3 ± 0.3	1.3 ± 0.6	-1.5 ± 0.7	-0.02 ± 0.01	-7.1 ± 1.6

Table 2. Parameters of ^{222}Rn mass balance equation and results of groundwater discharge at Lake Los Pocitos.
Tabla 2. *Parámetros de la ecuación del balance de masa con ^{222}Rn y resultados de descarga de agua subterránea en la Laguna Los Pocitos.*

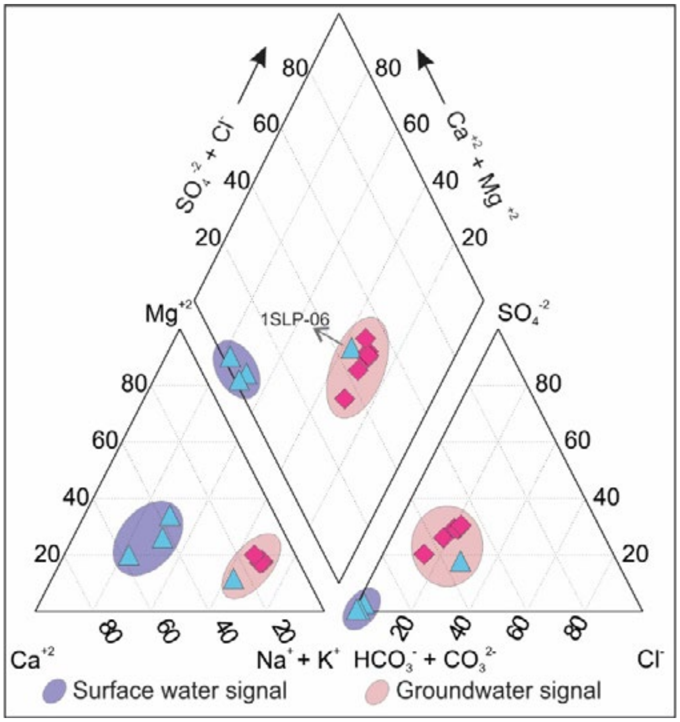

Figure 3. Piper diagram showing the major chemical composition of the groundwater and lake samples.
Figure 3. *Diagrama de Piper mostrando la composición química de los elementos mayoritarios en muestras de aguas subterráneas y de la laguna.*

are also reported. When comparing the ^{222}Rn concentration measurements at the 6 sampling points in the lake, a heterogeneous distribution is observed (Fig. 4), dividing the lake into two sectors: south (1LLP-01, 1LLP-02, 1LLP-03 and 1LLP-06) with lower ^{222}Rn concentrations (< 7 Bq m^{-3}); and north (1LLP-04 and 1LLP-05) with higher ^{222}Rn concentrations (~ 70 Bq m^{-3}).

Because there are no surface streams in this area, the balance of ^{222}Rn is controlled only by the groundwater flow *(Dsub),* the ^{222}Rn production resulting from the ^{226}Ra decay *(Jproduction),* the ^{222}Rn loss to the atmosphere (Jatm) and the ^{222}Rn decay in the water column *(Jdecay).* The estimated *Jproduction* is $2.9 \times 10^{-4} \pm 2.8 \times 10^{-5}$ Bq m^{-2} day^{-1}, whereas the *Jdecay* and *Jdiffusion* were calculated for each sector of the lake. In the southern sector the *Jdecay* is 1.0 ± 0.3 Bq m^{-2} day^{-1}, whereas *Jdiffusion* is 2.8 ± 0.1 Bq m^{-2} day^{-1}; whilst in the north it is 6.4 ± 0.8 Bq m^{-2} day^{-1}, and 0.1 ± 0.3 Bq m^{-2} day^{-1}, respectively.

^{222}Rn concentration in groundwater was measured in 4 shallow wells near the lake, with a mean depth of 1.8 m. The ^{222}Rn concentration in groundwater ranges between 3023.2 ± 205.1 Bq m^{-3} and 10081.6 ± 122.2 Bq m^{-3}, with a mean value of 7314.8 ± 829.2 Bq m^{-3}, i.e. up to three orders of magnitude greater than the ^{222}Rn concentration measured in the surface water.

From the mass balance model (equation 4), it can be estimated that groundwater discharge occurs in the northern sector of the lake with a flow rate of 185.3 ± 39.1 m^3 day^{-1}, coinciding with the groundwater regional flow. On the contrary, in the southern sector, the model generally gives negative values, suggesting a contribution from the lake to the aquifer (-13.4 ± 14.8 m^3 day^{-1}). It was also estimated that the groundwater

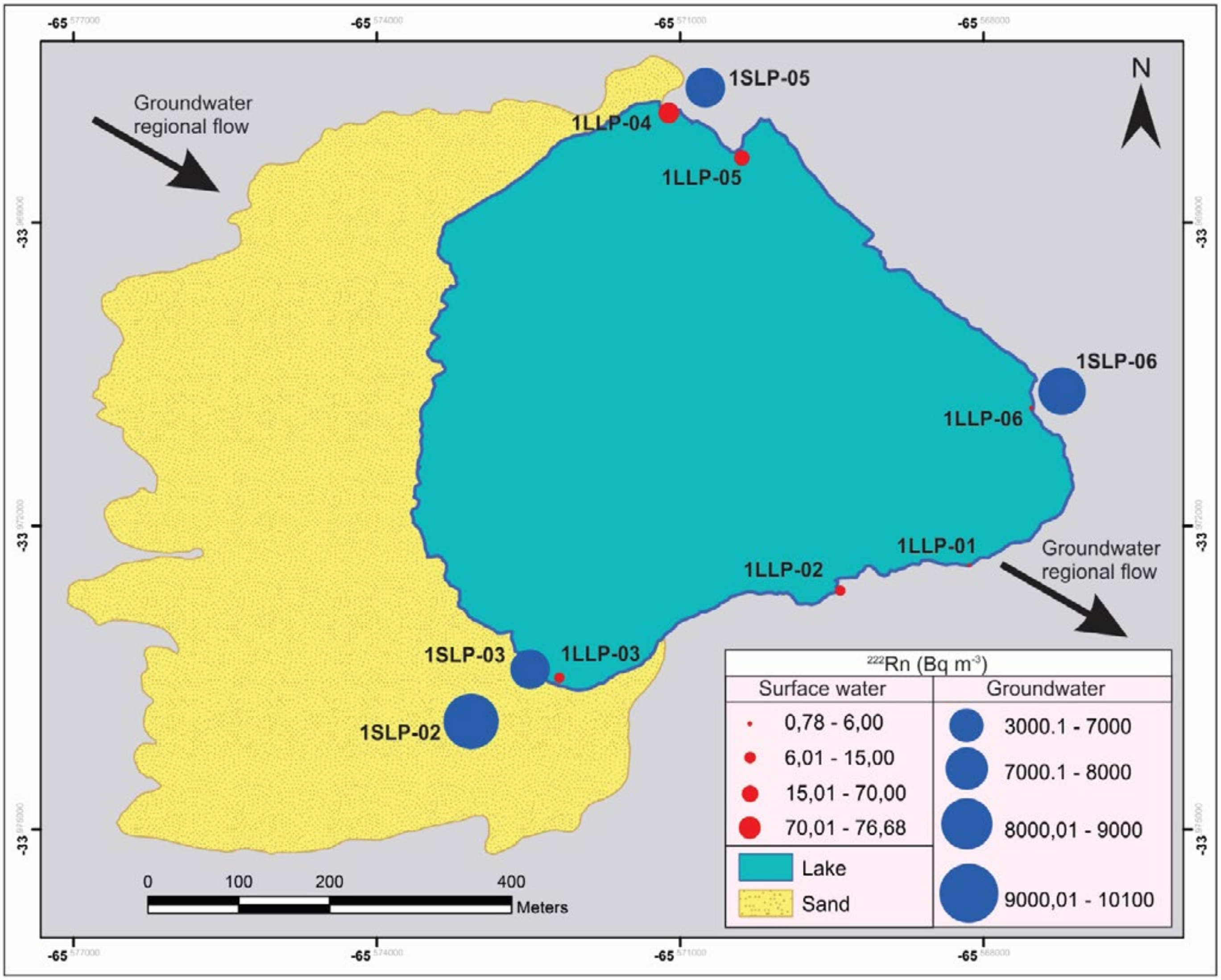

Figure 4. Distribution of ^{222}Rn concentration in Lake Los Pocitos and the associated groundwater.
Figure 4. *Distribución de la concentración de ^{222}Rn en la Laguna Los Pocitos y aguas subterráneas asociadas.*

flow in the northern sector has a velocity of 0.2 ± 0.04 cm day^{-1}.

Moreover, as expected, the greatest uncertainties in the model are observed in the southern sector of the lake, where ^{222}Rn activity was the lowest. This uncertainty is given by the ^{222}Rn concentrations of the groundwater endmembers, since the ^{222}Rn activity of groundwater can vary significantly at small spatial scales. These high groundwater inflow rates obtained for Lake Los Pocitos also reflect the importance of quantifying this term of the hydrological balance.

It is well known that in semi-arid areas with high evaporation rates, lakes tend to become salty and reduce their surface area when the hydrological balance is negative. However, in Lake Los Pocitos the surface area has remained relatively constant during the last 10 years (observed from satellite images, not included in this paper) and it presents moderate salinity. This can be explained by the continuous lake groundwater inflow to the lake. Although the lake is not as salty as expected for this hydrological setting, it is slightly saltier than the groundwater and it shows a significantly higher salinity than the aquifers located in the dune.

The analysis of the hydrochemistry reveals that two main geochemical processes are occurring in the lake related to the groundwater inflow. One is the evaporation, evidenced by the concentration of dissolved ions such as Na^+, Cl^-, and SO_4^{2-}; whereas the second process is carbonate precipitation. This is supported by Ca^{2+} and alkalinity depletions and also by the positive saturation indexes obtained for calcite, aragonite and dolomite (calculated with PHREEQC, not shown in this paper).

Conclusions

The identification and quantification of groundwater flows discharging into surface water bodies are essential for water resource management, mainly in semi-arid areas. For this purpose, the environmental tracer ^{222}Rn was used to evaluate groundwater discharge at Lake Los Pocitos, located in the sandy plain of the south-eastern region of San Luis Province. This water body has total dissolved solids concentrations of ~ 600 mg L^{-1} and an alkaline character. The lake water is of the HCO_3^- - Na^+ - K^+ type, whilst the groundwater is predominantly of the HCO_3^- - Ca^{+2} type.

The ^{222}Rn concentrations measured in the lake are three orders of magnitude lower than those measured in the surrounding shallow groundwater. The ^{222}Rn mass balance model used in this study shows that during the dry season, Lake Los Pocitos exhibits a differential hydrogeological behaviour along its perimeter. In accordance with the regional groundwater flow, in the northern sector there is groundwater discharge into the lake, giving it an influent character of the aquifer with respect to the lake, whilst in the southern sector, the lake is influent with respect to the aquifer. This is consistent with what was observed in the hydrochemical data, where the groundwater of the southern sector the surface water signal was observed.

The importance of this study is that, since the dissolved species concentrations (solutes in general, nutrients and pollutants in particular) in groundwater are generally different from those in surface waters, these volumes of groundwater entering the lake in the northern sector can have important biogeochemical effects, increasing or decreasing the biodiversity of the lake.

In arid to semi-arid environments with low rainfall and high evaporation rates, existing lakes, such as Los Pocitos, are expected to be supplied by groundwater throughout their perimeter. The results of this work show that environmental tracers such as ^{222}Rn are very useful for detecting and quantifying groundwater discharge or recharge in lake systems.

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