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Distribution of ^{222}RN on the Continental Shelf in Southern Brazil and its Relation with Groundwater Discharge

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Abstract

^{222}Rn has been widely used as a tracer ofsubmarine groundwater discharge (SGD) processes because it offers certain advantages, such as being an inert gas that is found at high concentrations in groundwater. This studyshows, for the first time, the distribution of this element on the continental shelf adjacent to Rio Grande do Sul state, Brazil. It describes its relation with sources and oceanographic processes, as well as its contribution to the profile of SGD along the state coast that stretches over 400 km. Since it also aims to fill gaps in the understanding of the SGD process, it answers the following question: what are the constancy and the scope of the process along this coastal system? SGD quantificationwas made from ^{222}Rn mass balance. ^{222}Rn activity ranged from values below the detection limit to 1.23 ± 0.32 dpm.L^{-1}. Perpendicular to the coastline, ^{222}Rn activity decreased offshore at a rate of 0.5 dpm.L^{-1}.km^{-1}up to approximately 10 km from the coast, where it was constant and below 0.4 dpm.L^{-1}. Groundwater fluxes were 5.66 ± 1.30 (southern region), 2.06 ± 1.02 (central region), 5.50 ±1.50 (northern region) and 6.63 ± 1.03 cm.day^{-1}(northern region). Variability in advection rates found by this study shows that, incoastal systems which are dominated by a certain type of sediment, some factors, such as local hydrodynamics and geology, can alter SGD quantitatively.

Keywords: Radioactivity; Geochemical Tracer; Permeable Sediment; Radon; South AtlanticOcean; Patos Lagoon

1. INTRODUCTION

Radon is a noble radioactive gas which has 36 radioactive isotopes, but only three of them occur naturally and are produced at a known rate, i. e.,^{222}Rn ($T_{1/2}$ = 3,823 days)fromthe ^{239}U radioactive series, ^{219}Rn ($T_{1/2}$ = 3.96 seconds)fromthe ^{235}U radioactive series and ^{220}Rn ($T_{1/2}$ = 55.6 seconds)from the ^{232}Th radioactive series. Isotope ^{222}Rn, which is treated as radon in this study,has been widely used as a tracer in the environment [1].

Radon is found in all types of rocks and soils, despiteits different ranges of concentration. Because it is abundant and there are no other sources or sinks, other than the decay of its parent ^{226}Ra and its own decay, radon is potentially important in coastal groundwater studies [2,3].

In the submarine groundwater discharge (SGD) process, one of current approach is the use of tracers[4-7].

SGD refers to the flow of water through continental and insular margins from the seabed to the coastal ocean, regardless of fluid composition or driving force [8,9]. This process is recognized as an important transport pathway of dissolved elements to marine environments, such as nutrients, metals and carbon [10-12]. There are two components of SGD, fresh groundwater and recirculated saline water. These two components together can contribute with dissolved materials many times greater than river inputs [9].

The use of ^{222}Rn as a tracer of SGD is advantageous because it provides an integrated signal when it arrives in the water column through several sources [10]. Widespread use of this isotope is mainly due to its high concentration in groundwater (on average, 1,000 times higher than in surface water) and to its completely inert character, that is, its activity is not altered by biochemical reactions, thus making it an ideal tracer [13] Currentlyradon and radium isotopes remain as the most popular geochemical tracers to identify and quantify SGD processes [9].

The study was carried out in the southern region in Brazil, mainly on the coast of Rio Grande do Sul (RS) state, where a group of researchers have conducted studiesof SGD processes [14-17]. It is due to the existence of many coastal lagoons in the region and because it is separated from the ocean by permeable sediments, which influence the flow of groundwater to the ocean directly. According to [18] about 13% of the coastal zone all over the world is occupied by coastal lagoons.

To understand the significance of SGD on a global scale, studies must be done in representative systems, within a finite number of land-seaboundary types [19]. These studies would enable results to be extrapolated to similar systems. In this case, the scopeof the study represents those areas occupied by coastal lagoons and separated from the ocean by a permeable barrier.

The use of ^{222}Rn has already been considered promising in the quantification of groundwater flows to some lagoons in the RS coast [20-22]. As for the ocean, this elementhas successfully used to quantify SGD onbeaches in northern RS [23]. However no further radon evaluations have been made in regions separated from the beach.

Many gaps still need to be filled so that complete understanding of the interaction between surface and groundwater in environments can be reached.They include questions, such as: are flows constant along the barrier or are they influenced by other factors? If so, which are these factors? Considering the applicability of radon, this study shows, for the first time, an evaluation of its distribution along 400 km parallel to the beach line and in regions separated from the beach (offshore), in a total area of approximately 12,000 km². Therefore, this study aims to evaluate the distribution of ^{222}Rn activity in relation to oceanographic sources and processes, as well as its contribution to the SGDprofile along the continental shelf adjacent to RS,Brazil.

2. MATERIALS AND METHODS

2.1 Study Area

Thestudy areais the coast of RS, in southern Brazil, between latitude 29°17'S - 32°09'S and longitude 52° 04'W - 49° 20'W, within the geomorphological unit called the Coastal Plain of RS. This region was formed in the Quaternary period of the Cenozoic era, the most recent of the formation of Earth. It is a large area of lowland (33,000 km2), mostly occupied by a huge system of coastal lagoons, especially the Mirim-Mangueira-Patos lagoon complex [24]. The only connection between this lagoon complex and the ocean is a800 m wide mouth, the estuary of the Patos Lagoon, located near Rio Grande city.

The RS coastline, which stretches over 625 km, is made up of a barrier system formed during the post-glacial marine transgression of the Holocene. This sandy barrier is composed of coarse clastic permeable sediments, a mixture of shells and finer pond deposit grains [25]. Studies have shown that the main source of recharge of the surface aquifers of this barrier system is precipitation [26].

Previous studies have shown that SGDprocesses are important sources of iron and other nutrients (nitrogen, phosphate and silicate) to the platform adjacent to the 240 km barrier that separates the Atlantic Ocean from the Patos Lagoon [16,17].In the south, adjacent to the Mangueira Lagoon, paleochannels facilitates this process [14, 15]. In general, groundwater flow towards the ocean, but, depending on precipitation conditions, the subterranean estuary, formed when fresh water meets sea water, may cause water to flow to a region farther away from the beach, into the mainland.

The climate in the region is considered mild mesothermic and superhumid, without anydry season. In general, the RS coastline has increase in rainfall, which varies between 1,200 and 2,000 mm, in the south-north direction.It is due to the basaltic plateau in the extreme north of the state, which facilitates the rise of air masses, thus, forming clouds and increasing precipitation [27]. The highest precipitation occurs in winter, between July and August, when there is high superficial discharge of fresh water to the ocean [28]. However, in dry periods, a reverse flow may occur, as a consequence of low water levels in the Lagoon.

2.2 Field procedures

Physicochemical and radioactive analyses of subsurface water, groundwater and sediment were performed. Subsurface water analyses were carried out in approximately 400 km of beach line and up to 30 km offshore, in approximately 12,000 km². Radon activity was determined at 42 stations, which were located parallel to the beach line and in four transects perpendicular to the beach line. Figure 1 showstransects A, B, C and D (average length is 33 km). Stations were located from the port in Rio Grande city to the mouth of the Mampituba River, on the border of RSand Santa Catarina (SC) state. The choice of transect sites was based on operability and equidistance. Transects C and D were close to each other because there are different hydrodynamic processes in that region, a topic which is discussed in item 3.1.

Analyses were carried out aboard the research vessel *AtlânicoSul*on an oceanographic cruise as part of the INCTMar-IOC Integrated Oceanography and Multiple Uses of the Continental Shelf project in July 2014 (austral winter). Activity of ^{222}Rn was measured in situ by the portable RAD-7 radon monitor (DurridgeCompany Inc.), which was adapted by using the RAD-AQUA accessory [29]. This monitor determines ^{222}Rn activity in the air by countingα emissions of the daughter isotope, ^{218}Po. Activity of ^{222}Rn in the air is then converted into water activity through a temperature and salinity-dependent solubility coefficient [30]. Physico-chemical parameters of the water column (temperature and salinity) were measured by a Sea-Bird CTD profiler.

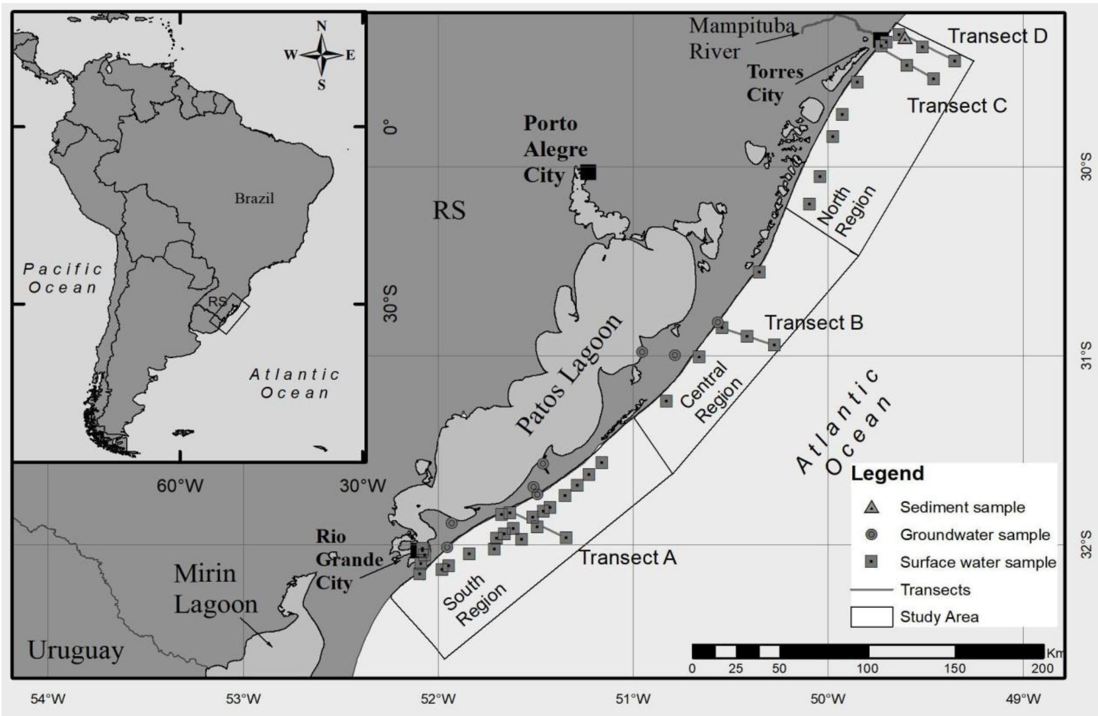


Figure 1. Location of the area and sampling stations under study

Groundwater sampling campaigns (advective fluid) were done in multi-level wells at the quaternary barrier. ^{222}Rn activity was also determined by the portable radon monitor RAD-7 (DurridgeCompany Inc.). In addition, a sediment sample was collected at transect D by a Van Veen dredge.Samples were kept under refrigeration. An experiment was carried out in the laboratory to determine radon fluxes from the sediment, in agreement with the methodology described by [31].

2.3 Data analysis

Data on ^{222}Rn activity in surface water,collected at 42 sampling stations, were interpreted together.It was done in order to evaluate the spatial variation of this element in the studyarea. In SGD quantification, only data on the three transects perpendicular to the beach line were used, as well as^{222}Rn activity in sediment and groundwater. The following section, 2.3.1, details the method of SGD quantification.

2.3.1 Quantification of SGD

For SGDquantification, the studyareawas divided into three regions, which are shown in Figure 1: south (Transect A), covering 120 km of beach line, center (Transect B), covering 170 km of beach line and north (Transects C and D), covering110 km of beach line.

SGDquantification was carried out in each region (south, center and north) from^{222}Rn mass balance in the water columm, where all sources (inputs) and sinks (outputs) of the tracerare considered in the area under study. Afterwards, values were converted into fluxes[32]. The following equation was used:

$$F_{dif} + F_{SGD} + (I_{226}\,\lambda_{222}) - (I_{222}\,\lambda_{222}) - F_{atm} - F_{mix} = 0 \qquad \text{(Eq.1)}$$

where F_{dif} is the diffusive flux of ^{222}Rn from the sediment, in dpm.m^{-2}.day^{-1}; F_{SGD} is the radon flux assigned to SGD, in dpm.m^{-2}.day^{-1} (value to be found); $I_{226}$is the inventory of ^{226}Ra, that is, the activity of ^{226}Ra in dpm.L^{-1}, multiplied by the depth, in meters (m); λ_{222} is the radon decay constant, in days; $I_{222}$is the inventory of ^{222}Rn, that is, the activity of ^{222}Rn in dpm.L^{-1}, multiplied by the depth, in meters (m); F_{atm} is the atmospheric evasion flux, in dpm.m^{-2}.day^{-1} and $F_{mix}$is the horizontal mixing flux., in dpm.m^{-2}.day^{-1}. Details of calculations of each element of the mass balance are described in sections 2.3.1.1 to 2.3.1.3.

Rate of advection of groundwater was calculated from the result of F_{SGD} by the following equation:

$$W = F_{SGD} / Rn_{pw} \qquad \text{(Eq.2)}$$

where W is the advection rate, in m.day^{-1}; $F_{SGD}$is the radon flux assigned toSGD in dpm.m^{-2}.day^{-1} and $Rn_{pw}$is the activity of ^{222}Rn in groundwater.

The following equation was used to transform the rate of advection into volume of advected water (SGD):

$$V = W.D.L \quad \text{(Eq.3)}$$

where V is the volume of advected water (SGD) in the regionunder study, in $m^3.day^{-1}$; W is the advection rate, in $m.day^{-1}$; D is the offshore distance of the transect, in m; and L is the length of the beach line, in m.

Sections 2.3.1.1 to 2.3.1.3 detail calculations of ^{222}Rn input and output fluxes in the environment, which compose Equation 1, from^{222}Rn mass balance. It is worth emphasizing that calculation of these fluxes in every region in the world, where SGD processes are studied, is inviable, since they can be extremely variable and lead to errors that overestimate/underestimate SGD rates. However, due to lack of data on the activity of this element, many studies have only used estimates for this calculation.

2.3.1.1 Diffusive flux

In mass balance,sediment is considered an influxof ^{222}Rn into the system, to the extent that the parent ^{226}Ra adsorbed into the sediment decays. To obtain ^{222}Rn flux from the sediment (F_{dif}), the method described by[31]adapts the first law of Fick. It is represented by the following equation:

$$F_{dif} = (\lambda_{222}\ D_s)^{1/2}(C_{eq}-C_w) \quad \text{(Eq.4)}$$

where λ is the decay constant of ^{222}Rn, in days; $D_s$is the diffusion coefficient in the sediment in $m^2.day^{-1}$; $C_{eq}$is the concentration of ^{222}Rn in equilibrium with the sediment in $dpm.L^{-1}$; and $C_w$is the activity of ^{222}Rn in the overlying water layer, in $dpm.L^{-1}$. The value of D_s was obtained by the following equation:

$$D_s = \varphi.e^{\left[-\left\{\left(\frac{980}{T}\right)+1,59\right\}\right]} \quad \text{(Eq.5)}$$

where T is the water temperature (° K) and φ is the porosity of the sediment, in%.To obtain C_{eq}, an experiment described by [33] was carried out, i. e., samples of sediment and local water were confined andkept under agitation for 4 weeks. Then,^{222}Rn activity was measured.

2.3.1.2 Atmospheric Evasion

Radon is a noble gas that tends to evade at the water/air interface. It can represent the loss of a large amount of the element when mass balance is made. Atmospheric evasion (F_{atm}) was calculated by the following equation [34]:

$$F_{atm} = K_{600.}\ (C_w-\alpha C_{air}) \quad \text{(Eq.6)}$$

where K_{600} corresponds to the gas transfer rate, in $m.min^{-1}$, which depends on wind speed and water salinity [35];C_w is the activity of Rn in water, to reduce radon activity in the air; and α is the solubility coefficient of Ostwald ($\alpha = 0.105 + 0.405e^{-0.0502T}$, where T is temperature in ° C).

2.3.1.3 Horizontal mixing flux

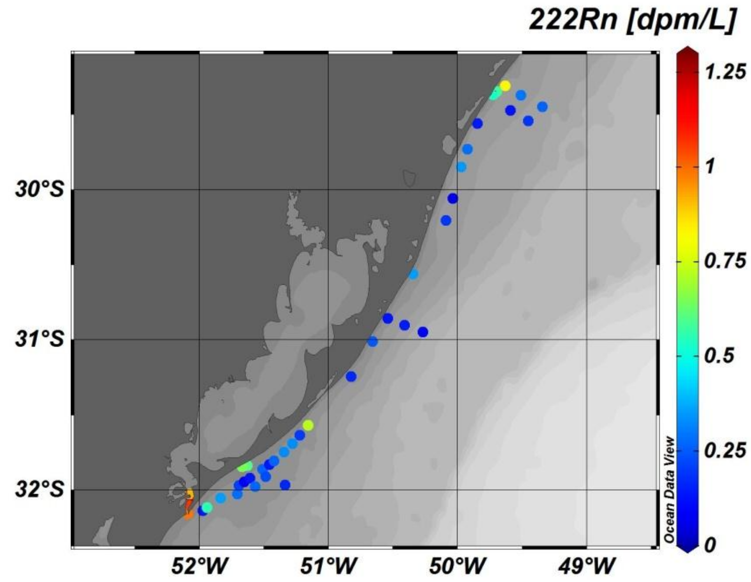
^{222}Rn is also lost by horizontal mixing with less concentrated waters. Thus, this loss must be calculated in the area under study. Based on the measurement of ^{222}Rn activity and specific water conductivity, horizontal advection and mixing coefficients were calculated by the method described by [36].

3. RESULTS AND DISCUSSION

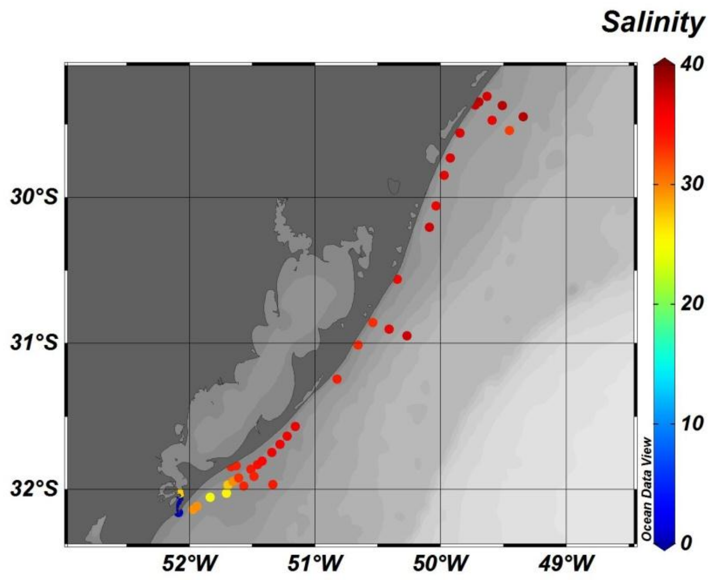
3.1 Spatial distribution of ^{222}Rn

General distribution of ^{222}Rn activities, as well as salinity, is shown in Figure 2.The lowest values of salinity, below 1, were found in the mouth of the Patos Lagoon, due to the spring fresh water supply. At this site, positive correlation with ^{222}Rn activity was also found. These results are within the expected range of collections taken at thistime of year (winter), when precipitation is high, pond and river levels increase and there is high freshwater discharge to the ocean. Data from the Brazilian National Institute of Meteorology [37] shows that accumulated rainfall in the month prior to collection was 191.37 mm.

^{222}Rn activityat all sampling stationsranged from values below the detection limit to 1.23 ± 0.32dpm.L^{-1}. Major activities were found at the mouth of the Patos Lagoon and in the northernmost region of the area under study(Figure 2a). The northernmost regionhas been described as one of discharge sites for South American groundwater reservoirs, the Serra Geral and Guarani Aquifer Systems [38]. It may explain why^{222}Rn activity is higherin this region than the one found in the other sites in the area under study. In the same region, the mouth of the Mampituba River (Figure 1), a small body of water with an average flow of only 19m^3.s^{-1}[39], can give small contributions to^{222}Rn activity during flood periods. However, this activity probably reaches only a few meters from its mouth. Therefore, it is inferred that ^{222}Rn activity in this place originates from subterranean discharges of the aquifers. The mouth of the Patos Lagoon represents a site that accumulates the drainage of several elements of continental origin in approximately 201,626 km^2. In addition, it is a place with intense port activity. It may lead to high concentrations of suspended material which generally contain absorbed ^{226}Ra, thus, increasing^{222}Rn activity in its vicinity. In freshwater environments (rivers and groundwater), Ra is mostly adsorbed at the suspended particles, whereas in sea water, this element is mainly dissolved [40]. When particles of suspended material meetsalt water, there is desorption of Ra from its surface. Average concentration of total suspended matter is estimated to be70 mg.L^{-1} in the estuary region of the lagoon, considering periods of drought and rain[41]. In the literature, each gram of suspended matter desorbsabout 2 dpm of ^{226}Ra in the mixing zone [40]. Based on this value and considering concentration of total suspended matterof 70 mg.L^{-1}in the estuary, the average contribution of ^{226}Ra from the suspended matter is approximately 0.1dpm.L^{-1}.



(a)



(b)

Figure 2. a)distribution of ^{222}Rn activity; and **b)** distribution of salinity at the sampling stations parallel to the coastline

In the north of RS[23] found different activities of two sedimentary stacking patterns in the surf zone. The authors found activities between 0.4 and 1.3 dpm.L^{-1}at a site with a retrograde (transgressive) stacking pattern and higheractivity, between 1.2 and

9.4 dpm.L^{-1}, at a site with a regressive stacking model.It shows that heterogeneities of the geological formation of the coastal barrier influence SGD.

Data collected by[23] added to those found by this study, show that distribution of ^{222}Rn follows a downward trend with the beach line spacing at an average rate of 0.5 dpm.L^{-1}.km^{-1}. In the west-central platform of Florida,[42] found activities from 7.2 dpm.L^{-1}, near the beach line, to 0.2 dpm.L^{-1},40 km offshore, which corresponds to a rate of approximately 0.17 dpm.L.km $^{-1}$. [43] in Iles-de-la-Madelein (sandy environment affected by melting ice), found activities from 4 dpm.L^{-1}, near the beach line, to 0.3 dpm.L^{-1}, 0.5 km offshore, which corresponds to a decrease rate of approximately 7.4 dpm.L^{-1}.km^{-1}.

Assuming that the main source of ^{222}Rn is SGD, it is clear that ^{222}Rn activity tends to decrease offshore. However, this rate can vary much, depending on the region, as exemplified in the previous paragraph.

This study evaluated concentrations of ^{222}Rn in regions far away from the beach to verify the reach of SGD on the continental shelf and to refine ^{222}Rn mass balance. Many studies that use ^{222}Rn as a tracer of SGD do so by using time series at points very close to the beach line. However, it is known that SGD can also occur farther offshore. For example, south of the Patos Lagoon, in a region about 100 km from its mouth,[15] analyzed nutrients, radio isotopes and bodies of waterand showed that SGD may occur near a paleocanal 50 km from the coast. Distribution of ^{222}Rn in the four transects perpendicular to the beach line are shown in Figure 4. Data on all transects showed that, approximately 10 km from the beach, activities tend to be constant, below 0.4 dpm.L^{-1}.[17]considered the occurrence of the SGD process up to this distance (10 km). Thus, it is inferred that 0.4 dpm.L^{-1} is the radon value supported by its ^{226}Ra precursor, presumably attached to mineral/ organic particles. Inprevious monitoring, ^{226}Ra activity found in this region was relatively homogeneous,i. e., 0.09 ± 0.01 dpm.L^{-1},on average [44]. In addition, farther from the beach line (deeper water), even if there is entrance of ^{222}Rn via SGD, this element is blocked due to the existence of pycnoclines (data not shown);therefore, itdoes not reach the superficial part where samples were collected.It should be highlighted that SGD may occur at different scales, such as the Nearshore, Embayment and Shelf ones [45]. This study only considered the Embayment Scale.

Distribution of ^{222}Rn was studied in few places in Brazil. In astudy carried out in Todosos Santos Bay (northeastern Brazil) [46],most radon measurements ranged between 1 and 2 dpm.L^{-1}andsalinity varied from 29 to 35. The authors interpret that, at this location, there could be high atmospheric losses due to high speedwinds (between 5 and 8 ms^{-1}). The authors also evaluated ^{223}Ra and ^{224}Ra activities, whose datasets reached a value of SGD that is three-fold the average discharge of the Paraguaçu River, which ends in this bay. This is evidence of the fact that there is significant contribution of groundwater in this location. [47] evaluated the spatial distribution of ^{222}Rn and found variation in the activity, whosevalues ranged from below 0.5 to 3 dpm.L^{-1} in the southeast of Brazil, in Ubatuba city, and in the Bays ofFlamengo and Fortaleza. They also carried out a temporal evaluation at a station located 200 m from the beach line, where the activity ranged from 2 to 6 dpm.L^{-1}. Values about 50-fold this value were found at the same site by [48]. thislarge difference was attributed to the distance from the coast, since it was only 15m. It should be highlighted that, in this study, the minimum distance from the coast where ^{222}Rn activity was measured was approximately 1 km. It explains the low activity that was found when both studieswere compared. In the south of South America, on the coast of Argentina, [49] found high values, between 1 and 6 dpm.L^{-1}, which the authors suggest to be associated with intense SGD.

Salinity values were lower near the beach line and tended to increase in the offshore direction, with the exception of transect C. [17] have already described that SGD from the permeable barrier which separates the Patos Lagoon from the ocean is able to reduce ocean salinity along the coastline. In addition, salinity and ^{222}Rn activity had a negative correlation at transects A (r^2 = 0.71) and D (r^2 = 0.87), which is expected when there are SGD processes with predominance of fresh water. At transect A, salinity was lower than the one of the other transects.It may be due to the influence of the Patos Lagoon plume, which, depending on the wind and discharge conditions of the Lagoon, can go north [50]. At transect C, salinity exhibited a different distribution from the other three transects, remaining practically constant in the three seasons of this transect. There is a small island called Lobos Island very close to the first station of this transect.Since it is the only one on the coast of RS, this island changes the local hydrodynamics;therefore,it causes difference in salinity.

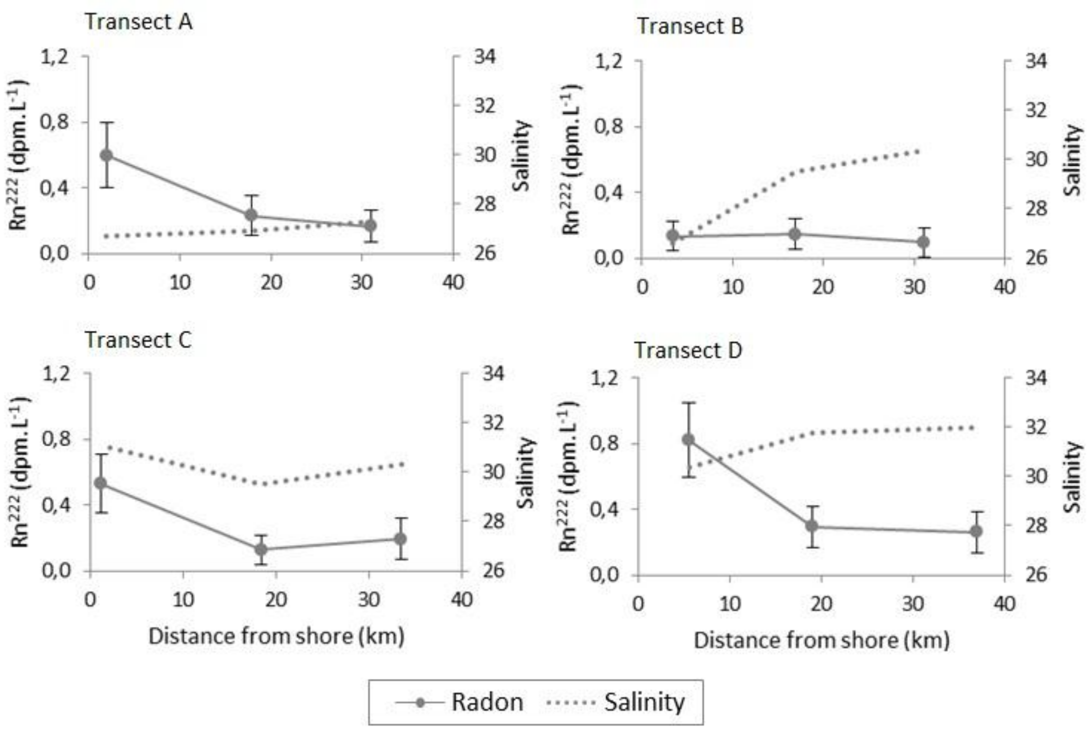


Figure 3. Distribution of ^{222}Rn activities and salinity at the transects perpendicular to the coastline

In order to determine whether the sites with the highest gradients (A, C and D) are effectively supplied by a large flow of SGD, linked to the spatial and geological differences and to the presence of large lagoon bodies, the box model (the balance of inputs and outputs) was applied.It was proposed[32], based on data on ^{222}Rn activity, and was described in section 2.3. Data on SGD are shown in the next section (3.2).

3.2 Submarine groundwater discharge (SGD)

As described in section 2.3, the study area was divided into three regions: south (transect A), center (transect B) and north (transect C and D).

SGD quantification involved the definition of ^{222}Rn sources and sinks. Results of the fluxes are shown in Table 1.

Table 1. ^{222}Rn influx and outflow in the regions under evaluation, where F_{dif} is the diffusive flux of ^{222}Rn from the sediment, F_{mix} is the horizontal mixing flux and F_{atm} is the atmospheric evasion flux.

Region	Transect	Influx	Outflow	
		F_{dif} (dpm.m^{-2}.day^{-1})	F_{mix} (dpm.m^{-2}.day^{-1})	F_{atm} (dpm.m^{-2}.day^{-1})
South	A	-	$23.3.10^{-6}$	1030
Center	B	-	$94.5.10^{-6}$	473
North	C	-	$8.8.10^{-6}$	1086
North	D	0.08	$11.6.10^{-6}$	1064

Besides SGD, F_{dif} and ^{226}Ra, the parent of ^{222}Rn, were the sources under evaluation. Several authors who calculated F_{dif} in other regions found very low fluxes and considered this entrance insignificant, in relation to the advection of groundwater [22, 52].Even so, in this study, this contribution was analyzed. To carry it out, a station of transect D was chosen.The F_{dif} value was 0.08 dpm.m^{-2}.day^{-1}.The comparison between F_{dif} and F_{SGD} values (Table 2) showed that the contribution of the sediment can be considered negligible. Therefore, this value could be extrapolated to the other parts of the area under study. The considered value of ^{226}Ra was 0.09 dpm.L^{-1}.It was determined by a previous monitoring[44].

The more atmospheric losses (F_{atm}) and losses by mixing (F_{mix}) are considered, the more ^{222}Rn sinks. These losses occur in greater or lesser proportions, depending on the wind speed. In the period under study, wind speed was 3.6, 4, 4 and 3 m.s^{-1} at transects A, B, C and D, respectively.

F_{mix} was found by the method described by[36]; it was based on the measurement of the spatial distribution of ^{222}Rn in a gradient perpendicular to the coastline (transects). Calculated results of loss per mixture are considered low, a fact that can be explained by the extension of the transects. For example, at transect A, ^{222}Rn activity found 5 km from the coast was 1.09 dpm.L^{-1}. At the most distant point from the beach (37 km), the value was 0.32 dpm.L^{-1}, that is, variation (loss of ^{222}Rn) is small when a 32-km stretch is considered.

From the determination of the flows of inputs and outputs (sources and sinks) of ^{222}Rn, the flux (F_{SGD}) corresponding to SGD could be found. In order to calculate advection rates (W), an endmember value had to be defined; it corresponds to ^{222}Rn activity in the groundwater that gives rise to the fluxes under investigation. In the northern region in RS,[23], through an extensive sampling network, found an endmember activity of 26.16 ± 1,37 dpm.L^{-1}. This value was used in this study for transects C and D. In order to find this value for the southernmost region, collections were taken from 9 wells off the coastal barrier at different depths (n = 20). Results were ^{222}Rn activity value of 25.93 ± 4.64 dpm.L^{-1}, which was the value used for transects A and B.

Results of groundwater flows are shown in Table 2.

Table 2.^{222}Rn fluxes and SGD in the regions under evaluation

Region	Transect	Total Area (m^2)	^{222}Rn - F_{SGD} (dpm.m^{-2}.day^{-1})	SGD rate – W (cm.day^{-1})
South	A	1.2 x 10^9	1467±338	5.66±1.30
Center	B	1.7 x 10^9	534±266	2.06±1.02
North	C	1.1 x10^9	1438±394	5.50±1.50
North	D	1.1 x 10^9	1735±271	6.63±1.03

Recorded advection rates (W) varied, with higher values in the south and north (embayments) and lower values in the central region (coastal projection). [17]used radio isotopes and found a rate of 3.5 cm.day $^{-1}$in the area adjacent to the Patos Lagoon in summer.

[23]used^{222}Rn and found different rates in two regions in northern RS, i. e.,46,15cm.day^{-1} (progradation stacking pattern) and 3,62cm.day^{-1}(retrograde pattern). Thus,a progradation stacking pattern facilitates the advection process, resulting in higher volumes of SGD by comparison with regions with a retrograde pattern. In a study carried out by[53] on the coast of RS, areas where coastal projections occur (prominent areas), such as the case of the area of transect B, the coast is predominantly retrogradational. It may explain the low rate (2.06cm.day^{-1}), when it is compared to the other three transects.

Conversion of advection rates by volume were calculatedby Eq. 3, in which the rate (W) is multiplied by the length of the beach line (L) and by the offshore distance of the transect (D). In this calculation, the offshore distance of 10 km was considered, instead of the total distance of each transect, because, as discussed in section 3.1, from this distance,^{222}Rn activities tend to be constant, below 0.4 dpm.L^{-1}, which is inferred to be the radon value supported by its ^{226}Ra precursor, i.e., the reach of the SGD process would be approximately 10 km. In a previous study developed in the same region, [17] also considered the occurrence of the SGD process up to this distance. Thus, the total volume found for the 400 km coastline (sum of the SGD volume of the southern, central and northern regions) was 2035 ± 514m3s^{-1}. In this calculation, rates were transects A, B and D, since salinity of transect C hasa different trend from the expected one. It did not affect the results, since two transects were made in the north region (C and D). It is worth mentioning that, in this study, no differentiation was made between both SGD components, fresh groundwater (Q_{fw}) and recirculated saline water (Q_{sw}), but [16], through a salt balance, found a flow of fresh water (Q_{fw}) to this region which corresponds to approximately 35% of the total volume of SGD. By using this percentage, it is estimated that the volume of fresh water that reaches the coast of thestudy area is approximately 610 m^{3}s^{-1}.It is estimated that the average freshwater discharge of the Patos Lagoon reaches 2,400 m^3.s^{-1} [54]that is, freshwater contribution via SGD may correspond to approximately 25% of the discharge of the Patos Lagoon. By adding both components (Q_{fw}) and (Q_{sw}), this contribution reaches 80%, which is an expressive value, when considering the contribution of dissolved elements associated with SGD.

4. CONCLUSIONS

This study showed that fluxes of groundwater are not constant along the continental shelf in the South of Brazil, a fact that may be associated with differences in hydrodynamics and geology.In a perpendicular assessment of the coastline, distribution of ^{222}Rn followed a similar behavior throughout the areaunder study, with decrease in offshore direction at an average rate of 0.5 dpm.L^{-1}.km^{-1}. In addition, approximately 10 km offshore, ^{222}Rn activity tends to remain constant and below 0.4 dpm.L^{-1}, which is inferred to be the value supported by its ^{226}Ra precursor. The expressive total volume of SGD, which corresponds to approximately 80% of the superficial discharge of the Patos Lagoon, indicates that SGD can be one of the main sources of dissolved elements in this region.Future studies should evaluate the vertical distribution of ^{222}Rn activity in the water column to understand the interaction at the sediment-water interface.

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