UNIVERSIDADE FEDERAL DO RIO GRANDE
CURSO DE PÓS-GRADUAÇÃO EM
OCEANOGRAFIA FÍSICA, QUÍMICA E GEOLÓGICA

CONEXÕES E INTERAÇÕES ENTRE A ÁGUA SUPERFICIAL E SUBTERRÂNEA NA COSTA NORTE DO RS (CNRS), BRASIL

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Rio Grande
2018

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Tese apresentada à comissão de curso de Pós-Graduação em Oceanografia Física, Química e Geológica da Universidade Federal do Rio Grande, como requisito parcial para a obtenção do título de Doutor em Oceanografia Física, Química e Geológica

Orientador: Prof. Dr. Luis Felipe Hax Niencheski
Coorientador: Prof. Dr. Eduardo Guimarães Barboza

Rio Grande
2018

RESILIÊNCIA (s. f.)

é ir à guerra e voltar;

é sentar com seus demônios numa mesa de bar e... conversar;

é apanhar de todo lado e levantar

é ter espírito boxeador, dar ganchos de direita nas

dificuldades e nocautear a própria dor;

é trabalhar para estudar e estudar para trabalhar;

é ter uma alma-água, que se adapta ao co(r)po em que estiver,

da melhor forma que puder.

AGRADECIMENTOS

Particularmente, percebo o “tempo do doutorado” como uma fase, no mínimo, interessante e desafiante da vida, a qual todos deveriam experimentar. Não por suas benesses ou pelo título que gera, mas por todas as dificuldades que esse “tempo” nos impõe, fazendo-nos crescer sobremaneira. A dita resiliência que defini anteriormente. É o “tempo” de descobrir do que se é capaz e o que vale a pena.

Eu não teria vencido esta etapa tão desafiadora e de autoconhecimento sem as balizas e influências dos meus orientadores, por isso tenho muito a lhes agradecer, Felipe e Eduardo! Serão para sempre figuras de inspiração e base, pela iniciativa, clareza, paciência e amor à ciência.

É uma fase tão decisiva, especialmente no âmbito psicológico, que o amor e compreensão da minha família não teria sido mais importante senão agora. Poucos talvez admitam, mas os momentos em que cogitei desistir de tudo existiram e não foram poucos, mas a força que meus pais e irmã mostraram haver dentro de mim, me fez ir adiante. Muito obrigada, família! Sempre juntos!

Tive um companheiro por praticamente todo este caminho, uma alma com a qual trilhei muitas escolhas e esteve sempre solidário e atuante, inclusive nos muitos consertos, nas saídas a campo, nos dias chuvosos, nos finais de semana e noites de trabalho. Anderson, foste essencial e agradecerei para sempre!

Somado a tudo que um doutorado nos impõe, isoladamente, optei por não abandonar meu trabalho e meus ideais junto ao Ceclimar, trabalhando e estudando, estudando e trabalhando. Foram muitos os quilômetros percorridos e essa escolha teve seu ônus e bônus.

Ônus de não alcançar talvez uma tese “mais” perfeita (isso nunca existirá, hoje eu sei!), mas não maior que a falta de convivência com os amigos e colegas da Hidroquímica. Sentimento que sempre foi uma constante, uma vontade, algo que ficou fora do alcance. Assim, agradeço aos colegas, amigos, professores, servidores por seu apoio, acolhimento e compreensão!

Isso foi compensado pelo bônus de não ver um dos trabalhos mais importantes da minha vida andar para trás, o laboratório ao qual me dedico desde 2008. Junto dele, muitos projetos valiosos tiveram continuidade e procurei disseminar o conhecimento, criando lá uma célula de tudo que aprendi na Oceano Química, Física, Geológica e (porque não) Biológica. Agradeço então aos meus amigos, alunos e colegas de Ceclimar, por acompanharem tudo se desdobrar, por torcer pelo melhor, por partilharem inúmeras coletas, incontáveis análises e gratificantes oportunidades aproveitadas! Sua paciência, empenho e eterna disposição de enfrentar o (sempre) curto tempo, nos fez ir mais longe, tendo hoje um lindo reconhecimento.

E por fim, a experiência de fechamento com chave de ouro foi o semestre junto aos mestres Herb Windom e Cliff Buck, entre outros incríveis pesquisadores do *Skidaway Insitute of Oceanography*. Que oportunidade! Que honra! Muito obrigada pela carinho, acolhida, interesse, paciência, parceria, aprendizado, segurança e aventura daqueles lindos e quentes dias de Savannah!

Essa mesma Savannah que oportunizou uma vivência mais próxima com a linda, querida e inteligentíssima colega Mariele. Obrigada, Mari, pela amizade, tolerância, conversas e todo apoio de “irmã de tese”! Por dividir a (tão americana) normalidade de atravessar o estado da Geórgia fugindo de um furacão, que nos enlouqueceu um bocado e testou nossas estruturas internas!

Acredito que o aprendizado deste “tempo” está no caminho que traçamos, nos obstáculos que contornamos, em tudo que se constrói de um ponto ao outro, e não no “chegar” propriamente dito. De longe o que mais importa para mim são as pessoas que compartilham esse “passeio” conosco, pessoas igualmente imperfeitas, que se esforçam muito e estão ali para

apoiarem-se mutuamente. Amigos, obrigada pelos abraços, palavras, encontros, cafés, cervejas, jantas, sorrisos, lágrimas e tudo mais que, certamente, encoraja a seguir em frente!

A Academia, assim como muitas outras posições, pode ser muito cruel, cobrando números, resultados, produtos, sendo a batalha principal manter a sanidade e a coragem. Todos ao meu redor de alguma forma tornaram esse "tempo" mais positivo, produtivo, construtivo, também mais doce, engraçado, prazeroso e gratificante!

O "tempo de doutorado" nada mais é do que uma etapa da vida, como muitas outras, que muda tua percepção e que me fez ser mais grata por tudo que tenho, pelo que sou e por quem está ao meu lado.

Muito obrigada!!

SUMÁRIO

RESUMO GERAL

Este trabalho teve como objetivo geral compreender as relações de comunicação entre águas superficiais e subterrâneas na região costeira nordeste do Rio Grande do Sul (CNRS), considerando a geologia e a hidroquímica local. Para tanto, foi mapeada e estabelecida a descarga de água subterrânea (DAS) na região entre as lagoas costeiras e a plataforma adjacente através de 14 transectos, assim como a influência da geologia local sobre este processo. Empregou-se o imageamento via radar de penetração (GPR), que mapeia em subsuperfície a arquitetura deposicional, assim como foram feitas medidas do isótopo natural radônio (^{222}Rn), reconhecido traçadores das águas subterrâneas, através do equipamento RAD-7. Com base nos dados obtidos foram compreendidas interações e influências geológicas na DAS e o papel da barreira arenosa nestes fluxos. Diferenças relevantes no comportamento da DAS foram estabelecidas, apresentando-se, aproximadamente, 10 vezes maior na área progradacional comparada à retrogradacional, sendo ainda magnificados pelos efeitos climáticos quando comparadas duas temporadas amostrais, verão austral 2016 e 2017. Foram estimados as concentrações e os fluxos de nutrientes e elementos traços para a CNRS, correlacionando-os com os padrões de empilhamento, com feições e descontinuidades geológicas, demonstrando a influência destes nos padrões de distribuição, de forma a traçar comparativos com outras áreas altamente relevantes. Além de qualificar e quantificar o papel da geologia enquanto *driver* na DAS e nos fluxos associados, foram estimados efeitos nos processos de fertilização das águas costeiras oceânicas demonstrando ser a região CNRS altamente beneficiada pelo processo.

Palavras chave: progradacional, retrogradacional, DAS, GPR, ^{222}Rn, nutrientes, elementos traço, Litoral Norte.

ABSTRACT

The aim of this work was to understand the relations between surface water and groundwater in the Northeast Coastal region of Rio Grande do Sul (CNRS), regarding local geology and hydrochemistry. Submarine Groundwater Discharge (SGD) was established in the barrier between the coastal lagoons and the coastal inner shelf in 14 transects, as well as the influence of local geology on this process. The Ground-Penetration Radar (GPR) was used, which illustrates the depositional architecture in subsurface, coupled with measurements of the natural radon isotope (^{222}Rn), recognized groundwater tracer, applying the RAD-7 equipment. The influence of geology on DAS was clarified, being established the role of the sandy barrier in these fluxes. Significant differences were observed in the DAS behavior, presenting 10 times higher in the progradational area compared to the retrogradational. The weather conditions magnified the DAS when comparing two sampling seasons, austral summer 2016 and 2017. Concentrations and fluxes of nutrients and trace elements were estimated for the CNRS, correlating them with the stacking patterns, geological features and discontinuities. It influences on the nutrient and trace element pattern distribution allowing to establish comparisons with other highly relevant areas. In addition to qualify and quantify the geology as a driver in the DAS and associated fluxes, effects on oceanic coastal water fertilization were estimated, pointing out the CNRS region as a region highly benefited by the process.

Keywords: progradational, retrogradational, SGD, GPR, ^{222}Rn, nutrients, trace elements, North Coast.

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ACRÔNIMOS

AAG – afloramento do Aquífero Guarani

BLP – Barreira da Lagoa dos Patos

CNRS – Costa Nordeste do Rio Grande do Sul

DAS – Descarga de Água Subterrânea

ENSO – *El Niño South Oscillation*

GIS - *Geographic Information System*

GPR – *Ground-Penetrating Radar*

LNSO – *La Niña South Oscillation*

MSL - *mean sea level*

NCR – *Northern Coast of Rio Grande do Sul state*

ND – *not detectable*

NIT – nitrogênio inorgânico total

PLB – *Patos Lagoon Barrier*

RS – estado brasileiro do Rio Grande do Sul

SGD – *Submarine Groundwater Discharge*

TDIN – *total dissolved inorganic nitrogen*

INTRODUÇÃO GERAL

A descarga de água subterrânea (DAS, do inglês *Submarine Groundwater Discharge* - SGD) foi, por muitos anos, negligenciada cientificamente pela dificuldade de avaliação e da percepção de que este processo não era importante (Burnett et al., 2003). Entretanto, atualmente, este processo é reconhecido mundialmente, contextualizado nos balanços hidrológicos e, portanto, cada vez mais estudado. São muitos os trabalhos desenvolvidos na última década (Anderson and Emanuel, 2010; Bejannin et al., 2017; Bishop et al., 2015; Burnett et al., 2007; Lee et al., 2009; McCormack et al., 2014; Russoniello et al., 2016; Sugimoto et al., 2016; Tovar-Sánchez et al., 2014; Viso et al., 2010) e, muitos destacaram a importância deste processo ao longo da planície costeira do sul do Brasil (Andrade, 2010; Attisano, 2012; Lima, 2014; Niencheski et al., 2007; Paiva, 2014; Souza, 2015; Windom et al., 2006) que evidenciam a relevância deste processo para a produtividade biológica.

A DAS oriunda dos aquíferos costeiros exporta grandes volumes de água doce para os oceanos mundiais (Attisano et al., 2013; Charette et al., 2013; Lee et al., 2009; Windom et al., 2007) e, atualmente, é tida como um grande transportador de matéria dissolvida entre a terra e o mar (Weinstein et al., 2011), sendo via de ciclagem de nutrientes e elementos traço para o oceano costeiro (Santos et al., 2009a).

Estas águas subterrâneas contém micro, macronutrientes e contaminantes, com importância algumas vezes comparáveis às descargas superficiais, pelo fato de terem um elevado tempo de residência junto aos sedimentos permeáveis, que desencadeiam reações biogeoquímicas resultando em elevadas concentrações, ou mesmo atuando como fonte principal em regiões mais empobrecidas nutricionalmente (Rodellas et al., 2015; Santos et al., 2009b), garantindo suporte à produção primária.

Em um dos primeiros estudos do Brasil, no estado do RS, a DAS foi avaliada ao longo de toda barreira costeira da Laguna dos Patos (BLP), por meio das concentrações de nutrientes (Niencheski et al., 2007) evidenciando a significância desse processo e seus efeitos sobre a produtividade costeira. A partir deste trabalho, a avaliação da DAS passou a ser investigada em quase toda a costa brasileira, estendendo-se para o Uruguai e Argentina (Paiva and Niencheski, 2018).

Segundo Burnett *et al.* (2003), essas descargas de águas provindas do subterrâneo oscilam muito, sendo variáveis com o tempo, desiguais e difusas em vista de múltiplas forças. Assim, quanto mais variadas às técnicas de verificação deste aporte, maiores as chances de cobrir significativo percentual destas manifestações. Existem diversas formas para detecção da ocorrência da DAS, como por exemplo o emprego de isótopos de rádio – ^{223}Ra, ^{224}Ra, ^{226}Ra e ^{228}Ra (Breier and Edmonds, 2007; Kim et al., 2008; Moore, 1996a), de radônio - ^{222}Rn (Peterson et al., 2010; Santos et al., 2008c; Wu et al., 2004) e *seepage meter* (Lee, 1977; Rocha, 2014).

Andrade (2010) aplicou algumas destas técnicas nos canais de irrigação na Lagoa Mangueira e detectou importante fluxo contribuindo para aquele sistema lagunar. Na sequência, Paiva (2011) realizou um estudo pioneiro nas lagoas costeiras da região nordeste do RS, utilizando o ^{222}Rn como traçador geoquímico, onde foi verificada DAS em sete delas. Numa continuidade de esforços na região, diferentes metodologias indicaram significativa diferença entre a DAS nas margens lagunares próximas das elevações da Serra Geral (margem oeste) daquelas próximas da linha de costa (margem leste) (Rocha et al., 2015). Paralelamente, a região de encosta e de tributários que servem a estas lagoas foi monitorada com resultados de descarga estimados na casa de milhões de metros cúbicos ao ano, respondendo por aproximadamente 20 % do volume de água destes corpos hídricos (Rocha, 2014).

Tão expressiva DAS, ainda que interna ao continente, enseja o questionamento de como se expressa esse processo na região costeira nordeste do Rio Grande do Sul (CNRS) para sua plataforma adjacente, qual seu volume e efeito. Estudos restritos a região de Torres, divisa do estado, evidenciaram a riqueza daquela região visto a influência do Aquífero Guarani em contato direto com o oceano (Paiva, 2014). Entretanto, ao longo de toda barreira da CNRS, aproximadamente 150 km, não existem ainda informações. Possivelmente a DAS se expresse como o observado ao sul do estuário da Lagoa dos Patos, na barreira que separa as Lagoas Mirim e Mangueira do Oceano Atlântico (Attisano, 2012), mas a CNRS possui características geológicas diferenciadas, oportunizando estudos mais amplos sobre os efeitos associados entre a hidrogeologia e a DAS, uma vez que esta área costeira apresenta-se sob dois processos evolutivos costeiros, barreira progradacional e retrogradacional.

Uma técnica que pode contribuir muito para avaliações de acamadamentos e ilustrar os processos subterrâneos é o radar de penetração no solo (do inglês *Ground-Penetrating Radar* - GPR), técnica geofísica não invasiva que detecta descontinuidades elétricas no subsolo raso (<50 m) e que o faz mediante a geração, transmissão, propagação, reflexão e recepção de pulsos discretos de alta frequência. Este método vislumbra a geometria deposicional da sedimentação do passado e suas relações ambientais, facilitando a maior compreensão hidrogeológica (Neal, 2004).

A união de técnicas de imageamento subterrâneos aos métodos de rastreio da DAS poderiam esclarecer parte dos fluxos que se desenvolvem na região costeira e os efeitos da geologia como uma das mais importantes forçantes, já que se trata do meio em que a água subterrânea sofre os processos biogeoquímicos que dão sua caracterização importante e ao mesmo tempo distinta da água superficial. Além disso, a conjunção dessas técnicas na CNRS proporcionaria uma oportunidade única de avaliar, espacialmente, a zona de mistura entre as águas doce e marinha no estuário subterrâneo, considerando inclusive processos de mistura entre vários aquíferos costeiros de profundidades variáveis e, os efeitos que a maré meteorológica e a intrusão salina exercem sobre a DAS.

Assim, o presente trabalho se faz necessário para a avaliação e compreensão do estuário subterrâneo e sua ligação e dependência com os corpos de água superficiais e as consequentes contribuições para a plataforma continental adjacente, provocando o incremento da produção primária.

O uso de técnicas bastante modernas pode definir assim, o papel das formas de deposição ali estabelecidas e a DAS. A hipótese é de que a DAS é quantitativamente elevada e qualitativamente relevante para o oceano costeiro na CNRS em vista de seus volumes e concentrações. Mais do que isso, acredita-se que a DAS esteja condicionada à deposição sedimentar costeira, sendo assim facilitada pela geologia local, ao passo que em outros pontos o efeito possa ser inverso, permitindo a penetração da cunha salina mais facilmente.

Desta forma, objetivou-se estabelecer a relação de conexão entre as águas superficiais e subterrâneas na CNRS, através da hidroquímica e da geologia local. E para responder este objetivo geral, esta tese está organizada em três artigos principais.

No primeiro artigo são apresentados dois perfis imageados por meio de radar de penetração (GPR) que representam as duas barreiras costeiras estabelecidas na CNRS, com arquiteturas deposicionais distintas, e sua correlação com a atividade do geotraçador radônio e a respectiva DAS estimada.

O segundo artigo expande a avaliação para 13 linhas perpendiculares à costa, como evidenciado pela Figura 1, cobrindo a barreira costeira nas regiões, progradacional e retrogradacional, a fim de testar a hipótese de que a DAS e a geologia estão ligadas, avaliando inclusive o fluxo de nutrientes associados para a plataforma costeira.

O terceiro artigo é dedicado a compreender os efeitos de características geológicas específicas, estabelecidas na região progradante e retrogradante da CNRS, sobre a DAS e o fluxo de elementos traço para a região costeira.

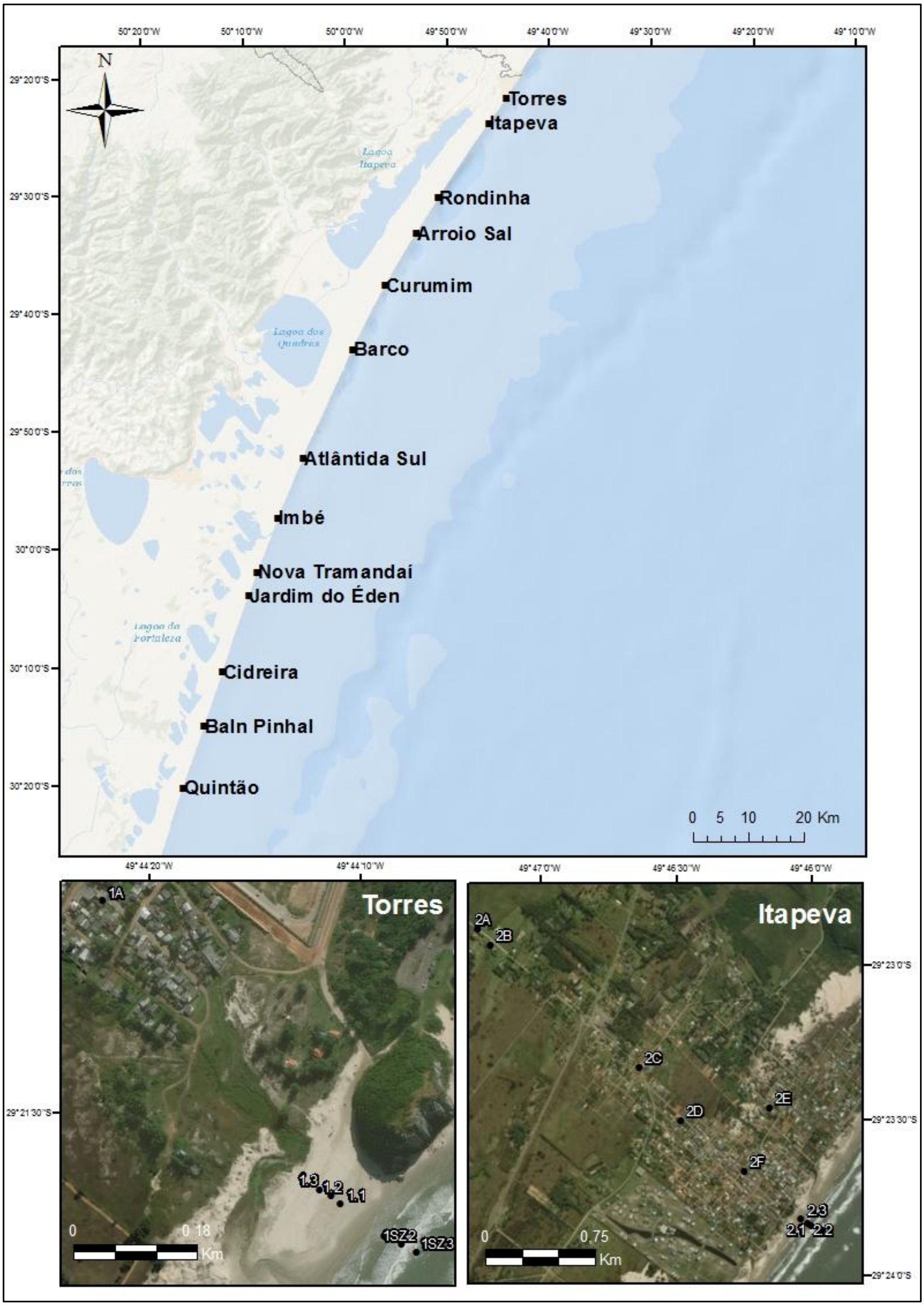
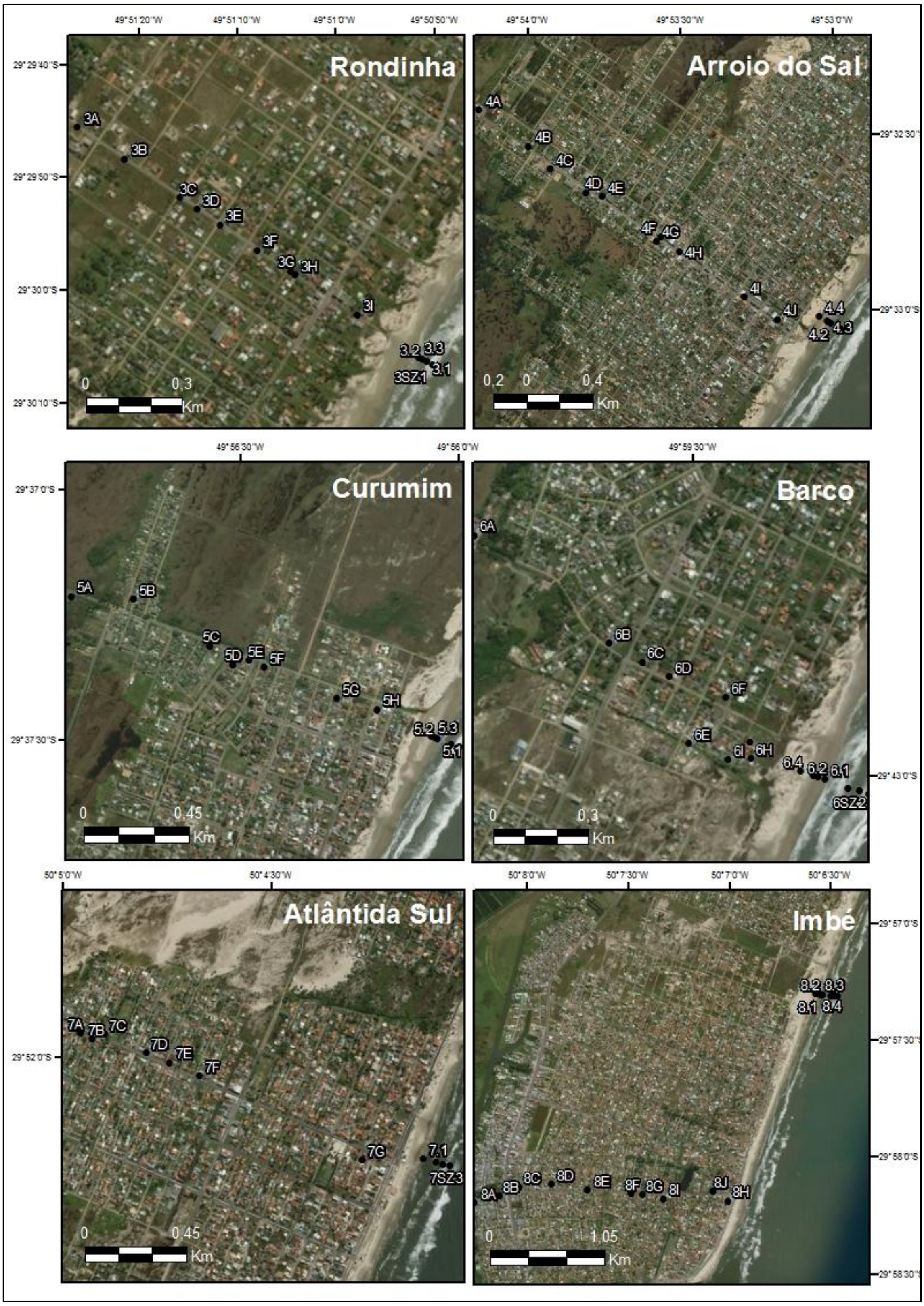


Figura 1 – Mapa geral da amostragem dos 13 transectos distribuídos dentro das áreas progradacional e retrogradacional, evidenciando nos detalhes cada um dos transectos com os pontos amostrais em poços permanentes (identificados por letras), piezômetros (identificados por números) e na zona de surfe (identificados pela sigla SZ).



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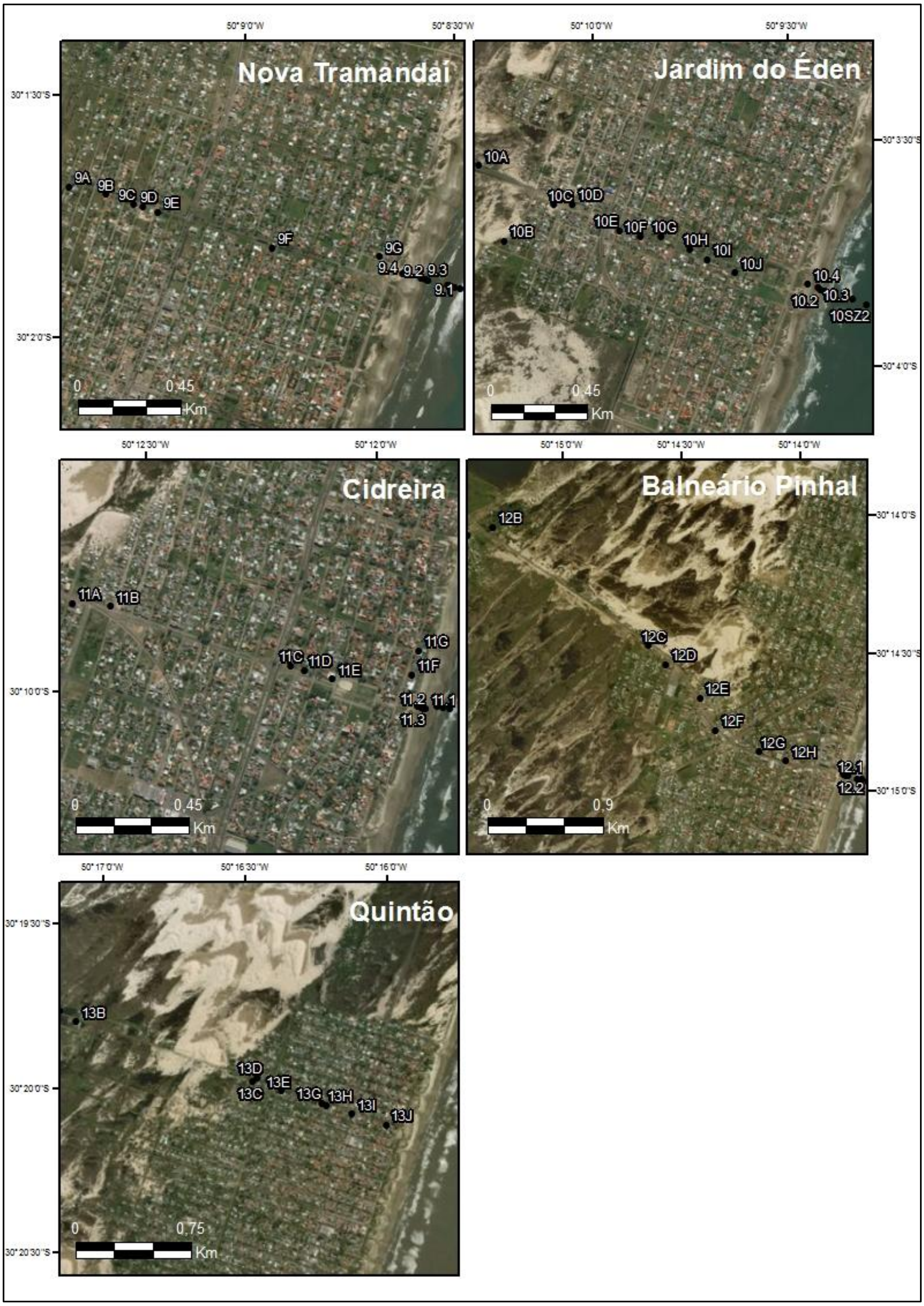

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ARTIGO 1 – RADON ACTIVITY AND SUBMARINE GROUNDWATER DISCHARGE IN DIFFERENT GEOLOGICAL REGIONS OF A COASTAL BARRIER IN SOUTHERN BRAZIL

Este artigo foi aceito para publicação no jornal *Environmental Earth Sciences*/Springer e segue as respectivas normas de formatação. Disponível: https://doi.org/10.1007/s12665-018-7711-0

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ABSTRACT

Submarine groundwater discharge (SGD) is still a relatively new topic and when associated with geological techniques like Ground-Penetrating Radar (GPR) it can be considered innovative. The present work aims to better understand the influence of geology in this important process of fluid flow from the continent to the coastal environment, with regards to radon isotope flux and hydrological budgets in Southern Brazil. An area that presents two different stacking patterns - progradational and retrogradational - was chosen and GPR scans were applied to these in an attempt to obtain stratigraphic deposition details. The derived discharge rates were calculated as well as the spatial budgets for the water fluxes from the coastal environment. The water moving across the sediment-water interface in these two geological regions is different, despite having the same main driving forces such as rainfall, tide and wave setup. The water flux is 13 times more expressive in progradational stacking (46.2 ± 2.4 cm/day) and radon activity is correlated with depth due to the different sedimentological layers. In the region dominated by retrogradational stacking the discharge is lower (3.6 ± 0.1 cm/day) and the average radon activity distribution is lower which is influenced by sediment type. Also, the estimated SGD in each region is 969.3 ± 51.2 10^3 m^3/d and 59.7 ± 1.7 10^3 m^3/d for 70 and 55 km of coastline, respectively. These significant differences ($p=0.004$) in radon activities and water fluxes certainly are defined by the geological issue being the result of depositional architecture, permeability and porosity.

KEYWORDS: SGD; GPR; sedimentary facies; progradational; retrogradational; Northern Coast.

INTRODUCTION

The submarine groundwater discharge (SGD) from coastal aquifers is an important process due to the great volume of freshwater exported to the ocean around the world (Attisano et al., 2013; Charette et al., 2013; Lee et al., 2009; Windom et al., 2007). This water contains nutrients and contaminants, sometimes comparable to superficial discharge (Santos et al., 2009b).

Since the late 20th century, numerous works about SGD have been published, especially in Southern Brazil (Andrade 2010; Attisano 2012; Lima 2014; Niencheski et al. 2007; Rocha 2014; Santos et al. 2008 a, b; Souza 2015; Windom and Niencheski 2003) which indicate a large volume of water arriving in the coastal shelf. It is especially important to support the chemical oceanographic processes there and around the world.

There are different ways to measure the SGD: seepage meter (Lee, 1977), nutrients (Andrade et al., 2011; Rutkowski et al., 1999) and, geotracers such as radium (^{226}Ra) and radon (^{222}Rn) (Santos et al., 2008c). We used ^{222}Rn for estimating total groundwater advection rates. It is an excellent natural tracer for identifying SGD because its half-life (3.8 d) is comparable to the timescale of most coastal processes. Also, it is two to four orders of magnitude more concentrated in groundwater than seawater, and it is conservative (Cable et al., 1996). In contrast to surface estuaries, subterranean estuaries are usually characterized as a non-conservative salinity mixing process.

^{222}Rn is easy to measure, making it possible to quantify groundwater discharge to coastal waters as shown in previous studies (Burnett et al. 2010; Burnett and Dulaiova 2006; Cable et al. 1996; Corbett et al. 1999; Dugan et al. 2011; Hussain et al. 1999; Moore and Shaw 1998; Moore 1996a, b; Peterson et al. 2010; Peterson et al. 2008; Santos et al. 2010). However, ^{222}Rn activity in the coastal zone, as well as the SGD, are governed by many elements and processes resulting in a discharge to the coastal ocean (Corbett et al., 1999; Swarzenski et al., 2007). The existence of an unconfined aquifer, the sampling depth, the tidal effect, the rainfall and seasons, and the types of sediments (Rocha 2014; Santos et al. 2009) are some of the features that control the SGD. Also, sediment deposition is important (Russoniello et al., 2013) since it is the pathway for the discharge.

The understanding of the coastal geologic system features could reveal a pattern and provide knowledge about its relationships with sedimentary facies. In our study area, the typical depositional stacking pattern is progradational and retrogradational, according to sea level changes and sediment balance (Barboza et al., 2011). These stacking patterns could potentially be landward or seaward reflectors (Barboza et al., 2011), which could be used as an alternative to facilitate the water movement, as well as the succession of facies that are reported for this and other regions.

We hypothesized that regions with different stacking patterns and sediment deposition present distinct ^{222}Rn activities and SGD.

The association of two approaches, Ground-Penetrating Radar (GPR) and SGD, is interesting since, until now, no studies have coupled them. If it is possible to demonstrate that the GPR images can somehow indicate the coastal subterranean discharge processes, this will result in an easier and didactical understanding of the influence of the geological structure on the SGD.

The main goal of this pioneering work is to couple GPR and geotracers to evaluate the influence of the geological feature on the SGD in different stacking pattern regions.

MATERIAL AND METHODS

a) Study area

The study area is located in the Northern Coast of Rio Grande do Sul state (NCR), Southern Brazil, between latitude 29º19'S and 30º25'S and longitude 49º42'W and 50º19'W, and 150 km long, as presented in Fig. 1. This area is a physiographic feature corresponding to the onshore portion of the Pelotas Basin. It was formed during the Quaternary by the juxtaposition of sedimentary deposits of two lagoon-barrier systems, designated III (Pleistocene) and IV (Holocene) (Villwock et al., 1986). The Holocene barrier system corresponds to a high-frequency depositional sequence stratigraphy (Rosa et al., 2017, 2011). It was built with a large volume of sand from the long-term northward littoral drift and sand transfer from the adjacent shelf (Dillenburg and Barboza, 2014). This barrier is composed of permeable sediments that form deposits known as transgressive dune sheets and foredunes, backshore/foreshore and upper shoreface (Barboza et al., 2013; Dillenburg et al., 2017).

About 40 % of the NCR is covered by coastal lagoons where the verified SGD is 150,000 m^3/d, approximately 20 % of the volume of these bodies of water (Paiva, 2011; Rocha, 2014). Since this is a significant value, a SGD is also foreseen for the coastal oceanic region as well. In addition, this was reinforced by the data obtained for the restinga of the Patos Lagoon (Windom et al., 2006) and Mangueira (Attisano, 2012), which were of the order of $8.5\ 10^7$ m^3/d and $6.7\ 10^7$ m^3/d, respectively.

The NCR coastal lagoons are separated from the ocean by a non-uniform sand barrier which presents some differences in geological structure, sediment composition (Dillenburg et al. 2017; Dillenburg et al. 2013; Dillenburg et al. 2011; Dillenburg et al. 2009; Rosa et al. 2017) and hydrogeology (Profill, 2005).

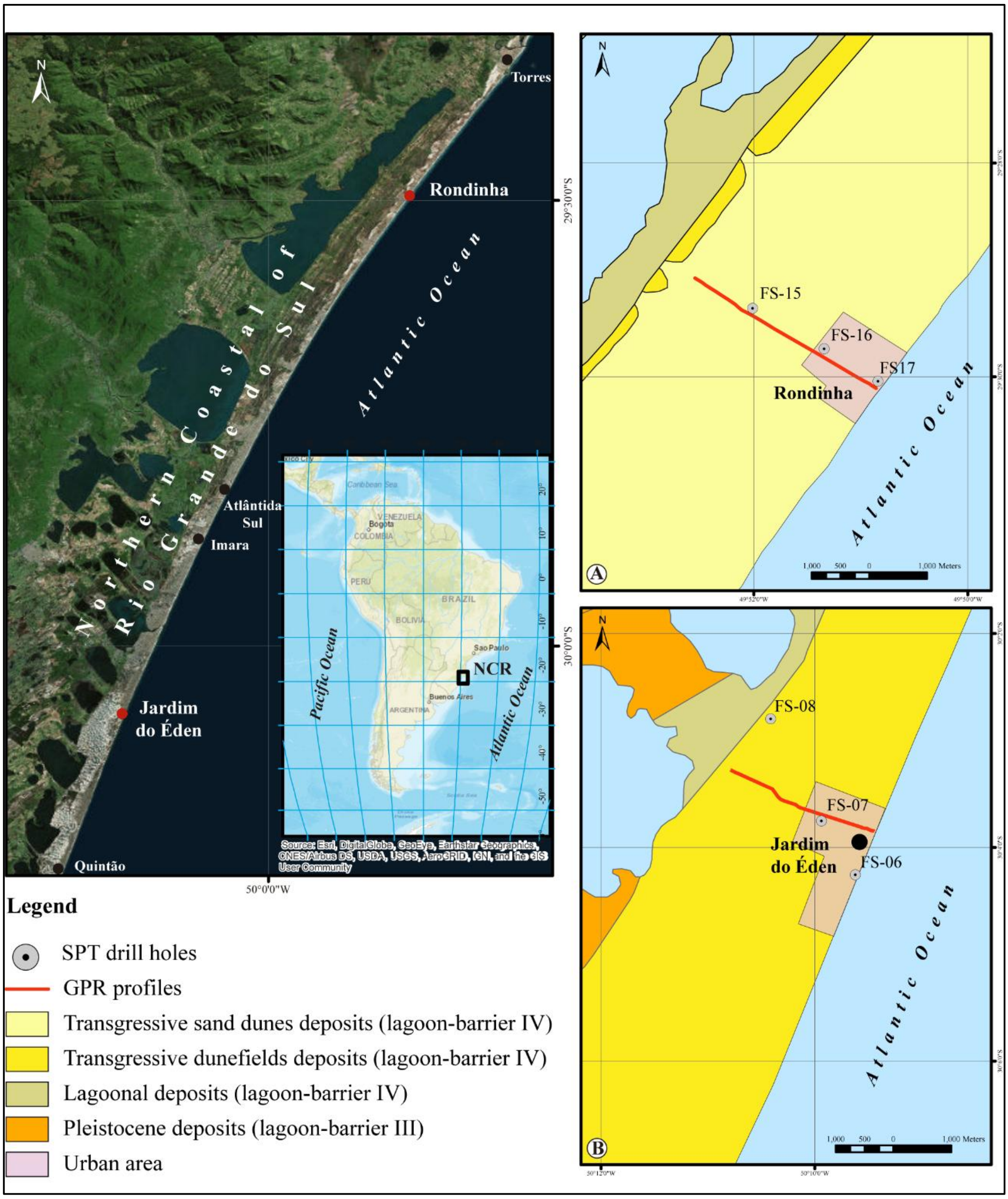

Fig. 1 Geologic map of the study area in NCR, Southern Brazil, showing GPR and drilling profile location (adapted from CPRM, 2006). Highlighted by sampling stations in Rondinha and Jardim do Éden beaches, representative of progradational (A) and retrogradational (B) stacking patterns and the drill holes (FS), respectively, based on Lima (2013) and Travessas et al. (2005).

Based on the geological background, (Fig. 2) we have split the study area into two regions with distinct stacking patterns: A) progradational (or regressive) and B) retrogradational (or transgressive) coastal evolution (Barboza et al., 2011; Lima, 2013; Travessas et al., 2005). Our sampling strategy was based on two transects perpendicular to the shoreline, one in each mentioned stacking pattern. The Rondinha Transect, which extends for 3100 m and represents the progradational region, and the Jardim do

Éden Transect, located in a region dominated by the retrogradational evolution process, which extends for 2400 m.

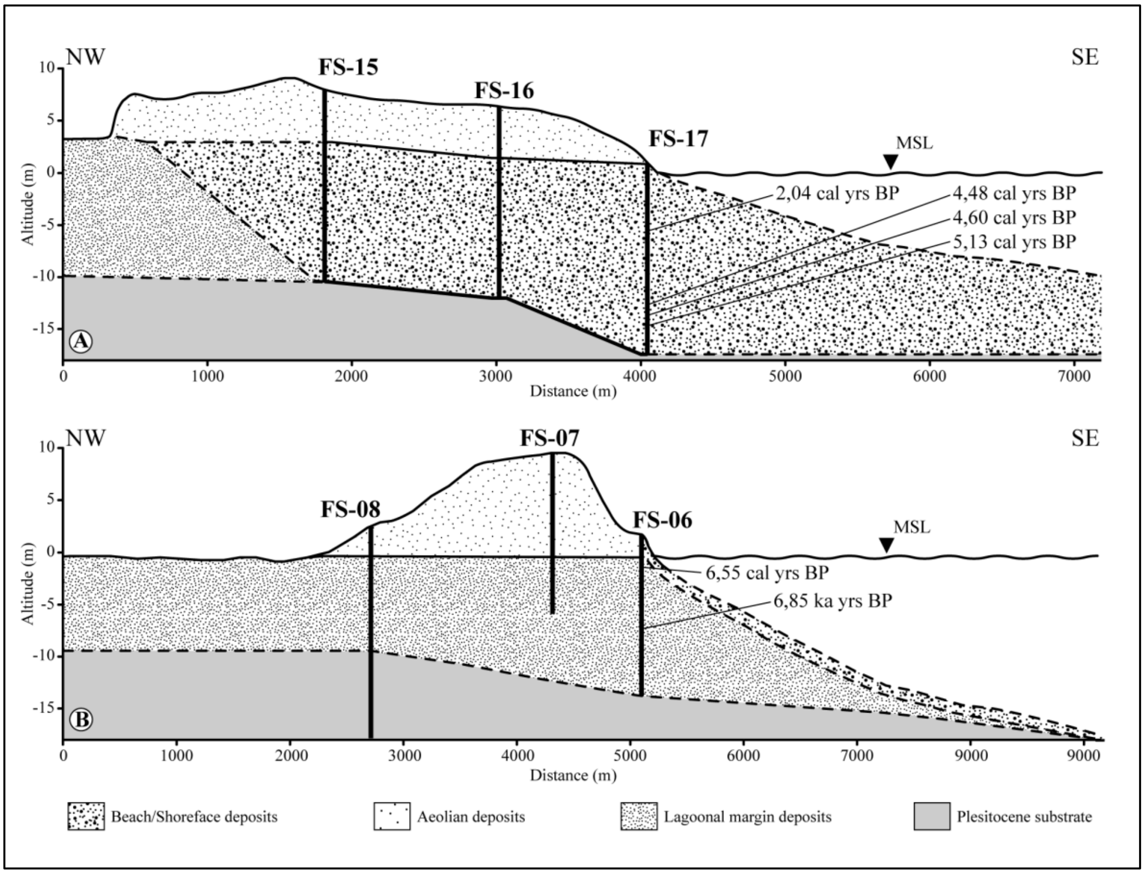

Fig. 2 Stratigraphic cross section of the A) progradational barrier at Rondinha, based on Lima (2013) and B) retrogradational barrier at Jardim do Éden, modified from Travessas et al. (2005) with drill holes (FS) (see drilling profile location in Fig. 1). MSL - mean sea level.

Geologically, both transects are located in a Holocene Barrier area with aeolian deposits composed of well-selected medium to fine sand (CPRM, 2006). In general, such Coastal Deposits occur along the entire coastline of Southern Brazil and correspond to more than 17700 km², with widths from 8 to 80 km (Machado and Freitas, 2005a). It is a flat region with very good soil infiltration (CPRM, 2015), since sands and sandstones are generally excellent aquifers with lateral and vertical continuity forming large groundwater reservoirs, with porosity up to 40 %.

Both sampling sites are located in a Coastal Quaternary Aquifer System region, with poor multilayer (unconsolidated) cementation of different origins. It is presented as an unconfined, semi-confined to confined aquifer due to sandy and sandy-silty deposits, with considerable aquifer potential (Lisboa et al., 2004; Machado and Freitas, 2005b).

b) GPR profiles

GPR has been used since it is a useful tool in the identification of local stratification, in order to know the vertical and horizontal variations. Although dominated by sandy sediments, the barrier is complex and the GPR can illustrate the compartments. We expect to associate the depositional geological differences with distinct SGD results, because the images produced by the GPR are provided in a continuous form, which makes it possible to identify the depositional architecture.

The GPR system used was composed of a Cobra Plug-In GPR (Radarteam Sweden AB) with a Subecho SE-70 monostatic antenna (80 MHz – center frequency) and Subecho SE-150 monostatic antenna (124 MHz – center frequency). This configuration allowed for the acquisition of two-way travel time (TWTT) with a range of 550/570 ns, with depth penetrations of -25/-27 m. The method used for GPR acquisition was reported by Barboza et al. (2014).

The data were post-processed with the Radan™, Reflex-Win® and Prism2® software packages. A dielectric constant of 10 for wet sand was used to convert travel-time to depth, which represents a velocity of 0.09 m/ns (Daniels et al., 1995). This constant was validated using lithological data obtained from SPT drill holes (Dillenburg et al., 2011; Lima, 2013; Travessas et al., 2005). The GPR profiles were topographically corrected using GNSS post-processed elevation data points collected along the profile lines at interval times of 1 second. These data were acquired using a GNSS Trimble® ProXRT (datum: WGS84) and analyzed in a Geographic Information System (GIS). The interpretation was based on the seismostratigraphy method (Mitchum Jr. et al., 1977) adapted to GPR (Neal, 2004). The method was based on termination (onlap, downlap, toplap and truncations), geometry and reflection patterns (Abreu et al., 2010; Barboza et al., 2013, 2011, 2009; E G Barboza et al., 2014; Catuneanu et al., 2009; Mitchum Jr. et al., 1977).

c) Submarine groundwater discharge (SGD)

1. Sampling ^{222}Rn activity

The ^{222}Rn activity sampling was conducted at the Rondinha and Jardim do Éden transects. Respectively, were taken nine and 10 samples in permanent wells, three and four samples in piezometers and three samples in the surf zone at both transect, during the Austral Summer of 2016 (Fig. 3).

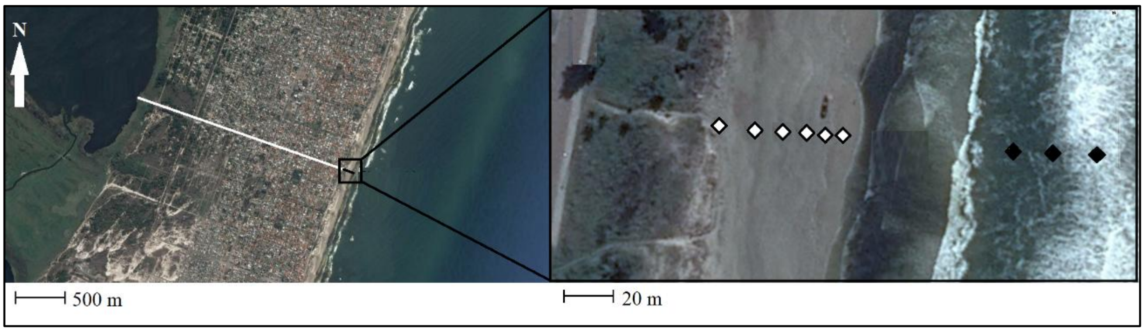

Fig. 3 Sample grid strategy, between lagoon terrace and ocean, applied to Rondinha and Jardim do Éden stations. The permanent wells were distributed over the white line. The piezometers and surf zone samples are distributed as indicated by the white and black diamonds, respectively (satellite image from Google™ Earth).

What we call permanent wells are the existing wells on private properties, used for household use and irrigation and the watering of animals. Our objective in using these wells of opportunity is to provide a better representation of the activities of ^{222}Rn, indicating the probable relationship that may exist between the sedimentary facies (progradational and retrogradational) existing between the coastal lagoons and the ocean, and the superficial aquifers of that region.

In the beach region, where the underground estuary is located, the salinity gradient and, consequently, the mixing process between fresh and salt water, is verified by drilling several surface piezometric wells. The mixing process is accompanied by the activity gradient of ^{222}Rn versus the salinity. Based on the activity data of ^{222}Rn from the beach wells, it is possible to establish the regional end member, which is a condition absolutely necessary for the knowledge of the geochemical processes that take place in the saline mixture of the underground estuary. The oceanic end members are defined as the concentration, in this case the ^{222}Rn activity, in the freshwater and at highest salinity (Niencheski et al., 1999).

The piezometers were drilled -1 m deep on the face of the beach where the salt mixing process is typically located. The permanent wells, with varying depths (-6 to -35 m), were chosen to cover the greatest vertical/horizontal variation of the station. In permanent wells, water was pumped out using the pre-existing pump in each well. The pushpoint system was used for piezometers, which consists of a thin empty stick that pulls the water from the ground with the aid of a peristaltic pump (Fig. 4a).

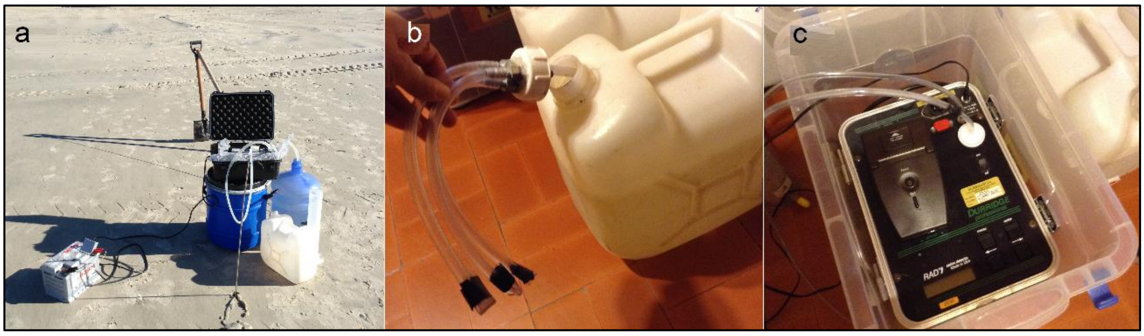

Fig. 4 Equipment for sampling groundwater in beachface a) pushpoint and peristaltic pump and equipment for ^{222}Rn determination b) Big Bottle and c) RAD7 monitor (Durrigde Inc.)

RAD7 portable monitor equipment (Durridge Co., Inc) was used to measure the ^{222}Rn activity with the Big Bottle Method (Fig. 4b and 4c), where a 10 L-bottle was used to sample 8 L of groundwater. Considering the short half-life of ^{222}Rn, it was necessary to perform the measurement of the samples as soon as possible.

Surface water from the surf zone is applied exclusively to assess the SGD in each region, progradational and retrogradational. Groundwater was sampled (8 L) at 3 points as a transect towards the ocean, 10-20-30 m from the shoreline. The ^{222}Rn activity is the proxy for obtaining the mass balance and ^{222}Rn flows determined by a mass balance approach, presented by (Corbett et al., 1997).

2. Mass Balance and fluxes

The calculation method, previously presented by a number of authors (Burnett and Dulaiova, 2003; Corbett et al., 1997; Paiva, 2014; Peterson et al., 2008; Rocha, 2014; Santos et al., 2008c), measure a ^{222}Rn gradient in surf zone samples. Diffusive flow from the sediment, atmospheric loss flow, mix loss flow and the decay from ^{226}Ra and ^{222}Rn in the water column are assessed (Eq. 1). In this study, the mix loss was calculated using a conservative element, the conductivity, as established by the Glover calculation (2005).

$$F_{dif} + F_{SGD} + {}^{226}Ra.\lambda_{222} - {}^{222}Rn.\lambda_{222} - F_{atm} - F_{hor} = 0 \qquad (1)$$

where:

F_{dif} – diffusion flow from radium concentration in sediment that decay to ^{222}Rn, in dpm/m² d;

F_{SGD} – ^{222}Rn flow, in dpm/m² d;

$^{226}Ra.\lambda_{222}$ – ^{222}Rn excess from ^{226}Ra father decay, in dpm/m² d;

$^{222}Rn.\lambda_{222}$ – ^{222}Rn inventory in the water column, in dpm/m² d;

F_{atm} – atmospheric flow or atmospheric loss, in dpm/m² d;

F_{hor} – flow horizontal mixing or loss by horizontal mixing, in dpm/m² d.

The ^{222}Rn flow (F_{SGD}) is established and applied in Eq. 2. ^{222}Rn activity groundwater of end member (Rn_{pw}) is the typical and representative value determined in a subterranean environment where the flow originates at the site of interest. In our study, the end member was obtained considering the average of the ^{222}Rn activity results of the piezometers sampled on the beach for each transect. It is recommended to measure the end member for each region in order to avoid overestimating/underestimating the SGD.

$$W = F_{SGD} \div Rn_{pw} \qquad (2)$$

where:

W – submarine groundwater discharge rate, in m/d;

F_{SGD} – ^{222}Rn flow, in dpm/m² d;

Rn_{pw} – ^{222}Rn activity in groundwater or porewater, in dpm/m³;

From the SGD rate (W) it is possible to estimate the SGD volume in the studied region, according to Eq. 3. The gradient distance corresponds to the distance where samples of the ^{222}Rn gradient were taken, for this study, 30 m. The lengths from the beach (L) used were70 km and 55 km for the Rondinha transect and the Jardim do Éden transect, respectively.

$$Vol = W \; x \; D_{grad} \; x \; L \qquad (3)$$

where:

Vol – discharge volume for specific area, in m³/d;

W – submarine groundwater discharge rate, in m/d;

D_{grad} – gradient distance, in m;

L – length of the beach, in m.

The results provided by the mass balance calculation, based on the box model and with consideration for a steady state, provide water flow or SGD rate (m/d) and discharge per unit length (m^3/m d) data (Burnett and Dulaiova, 2003). Both give the dimension of daily water flow or SGD flux (m^3/d), exported by each region (progradational and retrogradational), in view of the spatial representativeness of each region of coastal evolutionary process. This daily water flow is called SGD.

RESULTS

a. GPR profiles

The profiles obtained, shown in Fig. 5 and Fig. 6, respectively, represent the Rondinha and Jardim do Éden transects, covering the lagoon terrace to the beach line along the coastal barrier. As for the altimetry of the GPR sections, the data were acquired on level roads in both transects.

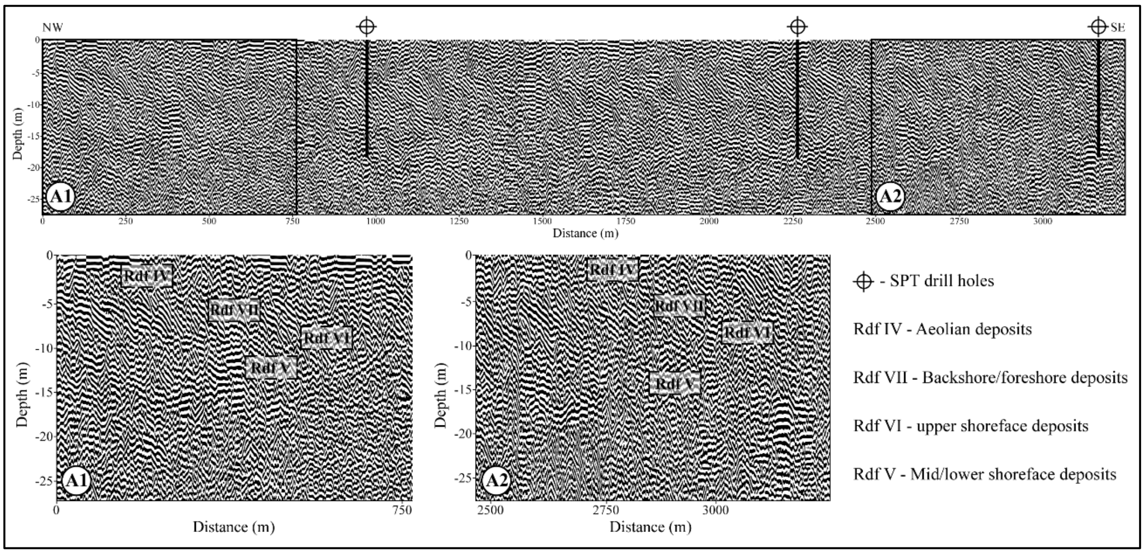

Fig. 5 GPR image section of Rondinha transect, 3.1 km from northwest to southeast and -25 m deep. The highlight for subsection in blackline areas - A1 and A2 – showing cores (FS-15, FS-16 and FS-17) and radarfacies (Rdf) characterizing a progradational region.

Fig. 5 shows that the Rondinha profile is influenced by aeolian deposition and the coastal conformation with progradation features and reflectors dipping seawards. This barrier started to prograde at around 7,000-8,000 years before the present during a rising sea-level condition (Dillenburg et al., 2013; Watanabe, 2016). Also, the figure shows that two regions have detached structures, which were extracted and presented as Fig. 5 A1 and 5 A2.

Subsection A1, closer to the lagoon terrace, presents the deposition units described below, from the lower level to the top:

- an attenuation of the signal is recorded under -18 m depth caused by a clay layer which results in insulation;

- in depth between -15/-18 m there are reflectors from the middle/lower shoreface unit (radarfacies V) and above this (-15/-10 m depth) there is the shoreface upper unit (radarfacies VI), that is the sinuous and planar shapes the difference between the two layers;

- the layer from -5 to -10 m deep is characterized by planar reflectors with a low angle being identified as the backshore/foreshore unit (radarfacies VII);

- and close to the surficial interface (0/-5 m deep) the aeolian deposition is presented (radarfacies IV) as foredune ridges and/or transgressive dune sheets (Barboza et al., 2013).

Subsection A2 presents a similar profile from subsection A1, added to the reflectors characteristic of the ridge/swale (foredunes ridges – between -4/-7 m deep).

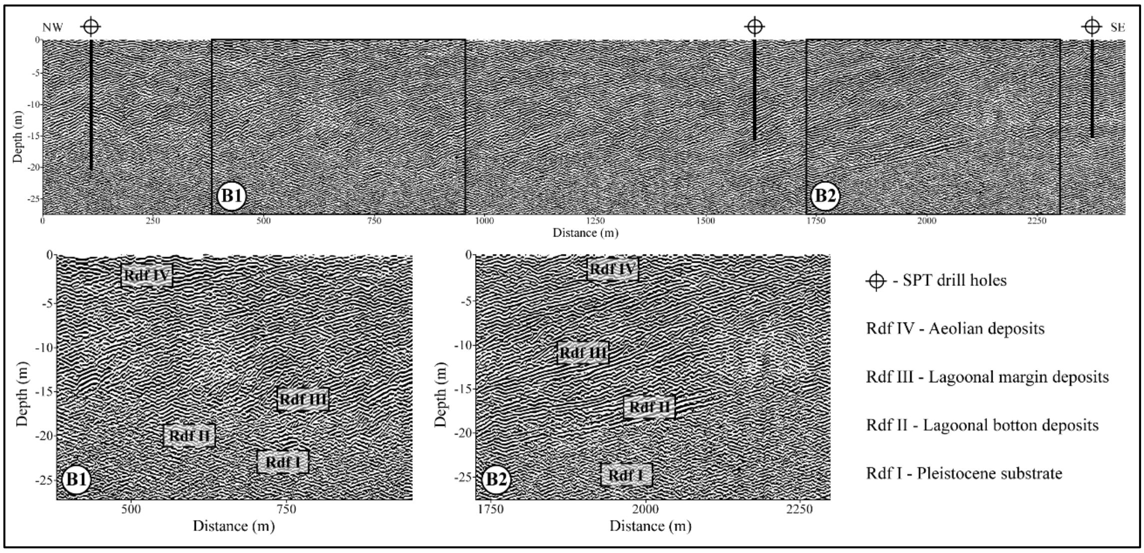

Fig. 6 GPR image section of Jardim do Éden transect with 2.4 km from northwest to southeast and -27 m deep. The highlight for subsection in blackline areas – B1 and B2 – showing cores (FS-06, FS-07 and FS-08) and radarfacies (Rdf) characterizing a retrogradational region.

Fig. 6 presents a classical retrogradational barrier, the Jardim do Éden transect, dominated by transgressive dune sheets (Dillenburg et al., 2013; Watanabe, 2016). Subparallel reflectors have been observed due to aeolian deposition (at the top of the profile), covering the mud from a paleolagoonal system. Also, Fig. 6 shows that two areas have detached structures (B1 and B2).

Subsection B1, closer to the terrace lagoon, presents 4 deposition units from bottom to the top, being:

- under -25 m deep there are signals (attenuated) coming from the Pleistocene substrate (radarfacies I);

- at -20/-25 m deep, the layer formed by clay (radarfacies II) which acts as insulation, causing attenuation in the GPR signal;

- at a depth of -15/-20 m, sigmoidal reflectors were recorded corresponding to deltas formed during the aggradation process in lagoon margins (radarfacies III) as mentioned by Rosa et al. (2016);

- at -5/-10 m depth the aggradation reflectors (radarfacies III) are presented.

Subsection B2 presents two marked areas with oblique reflectors dipping landward over lagoonal inter-barrier depression reflectors (between -5 and -23 m depth). The first (-10 m deep) are muddy reflectors dipping landward which correspond to the bottom paleolagoon (radarfacies II). The second, above the previous, is aggradation in the lagoonal tract (-5 to -15 m, radarfacies III).

The continuous lines reach up to the coastline providing strong evidence of a retrogradational stacking pattern region. These lines should be lagoonal muds outcropping (radarfacies II) along the current backshore/foreshore due to erosion processes as already recorded in previous sedimentary studies (Barboza et al., 2011; Dillenburg et al., 2004a; Martinho et al., 2008; Travessas et al., 2005). At the top, aeolian deposition (radarfacies IV) through transgressive dune sheets can be observed (0/-4 m deep).

The geological characteristics observed in these transects (sediment composition, compaction, depositional architecture) produce different conditions among the regions that, consequently, govern ^{222}Rn activity. Thus, our sampling strategy was developed in such a way, so that each of these geological units was analyzed for ^{222}Rn activity.

b. ^{222}Rn activities

Table 1 presents ^{222}Rn activities obtained in the permanent wells, piezometers and surf zone. These data have a non-normal distribution (Shapiro-Wilks significance 0,090 for progradational and 0,001 for the retrogradational region) and the averages were 52.5 and 29.0 dpm/L for the Rondinha and Jardim do Éden transects, respectively.

This shows that the progradational system has almost twice as much ^{222}Rn activity as the retrogradational region It should be noted, however, that in both regions there are important variations in the ^{222}Rn activity values. This can be explained by the fact that the permanent wells are opportunity wells, where their depths and locations along the transects are variable.

Table 1 ^{222}Rn activities in Rondinha and Jardim do Éden stations during Austral Summer 2016

Location	Samples	Depth (m)	Radon activity (dpm/L)	Salinity	Distance from the ocean (m)
Rondinha	permanent well	22	136.67 ± 2.38	0.05	1,160
	permanent well	6	21.22 ± 0.91	0.05	1,000
	permanent well	12	60.92 ± 1.59	0.06	820
	permanent well	20	85.19 ± 1.96	0.08	765
	permanent well	28	120.25 ± 2.37	0.06	690
	permanent well	24	198.32 ± 3.02	0.06	560
	permanent well	24	98.75 ± 2.14	0.04	460
	permanent well	35	44.04 ± 1.48	0.05	440
	permanent well	12	37.00 ± 1.36	0.05	240
	piezometer	1	15.72 ± 0.97	5.24	15
	piezometer	1	19.04 ± 0.89	6.50	10
	piezometer	1	18.66 ± 1.05	12.25	5
	surf zone	0	9.4 ± 0.8	29.82	-10
	surf zone	0	3.8 ± 0.5	31.55	-20
	surf zone	0	1.2 ± 0.3	31.65	-30
Jardim do Éden	permanent well	22	269.09 ± 3.14	0.18	1,515
	permanent well	7	101.93 ± 1.97	0.08	1,310
	permanent well	12	79.72 ± 1.73	0.06	1,170
	permanent well	17	102.32 ± 1.99	0.07	1,105
	permanent well	7	65.73 ± 1.60	0.06	880
	permanent well	17	45.46 ± 1.39	0.12	795
	permanent well	7	30.69 ± 1.14	0.07	715
	permanent well	24	27.21 ± 1.09	0.08	585
	permanent well	18	17.01 ± 0.94	0.14	505
	permanent well	6	17.03 ± 0.95	0.13	380
	piezometer	1	13.71 ± 0.92	6.23	65
	piezometer	1	19.40 ± 1.00	10.16	30
	piezometer	1	25.69 ± 1.05	20.37	20
	piezometer	1	19.12 ± 0.98	23.13	10
	surf zone	0	1.3 ± 0.2	30.86	-10
	surf zone	0	0.7 ± 0.2	31.68	-20
	surf zone	0	0.4 ± 0.1	36.34	-30

The ^{222}Rn activities at the Rondinha and Jardim do Éden transects were plotted over respective GPR sections (Fig. 7 A2 and B2). The activities measured in the permanent wells of the coastal barrier are used to evaluate the process of mixing groundwater in the unconfined surface aquifers, which are a function of the stacking profiles.

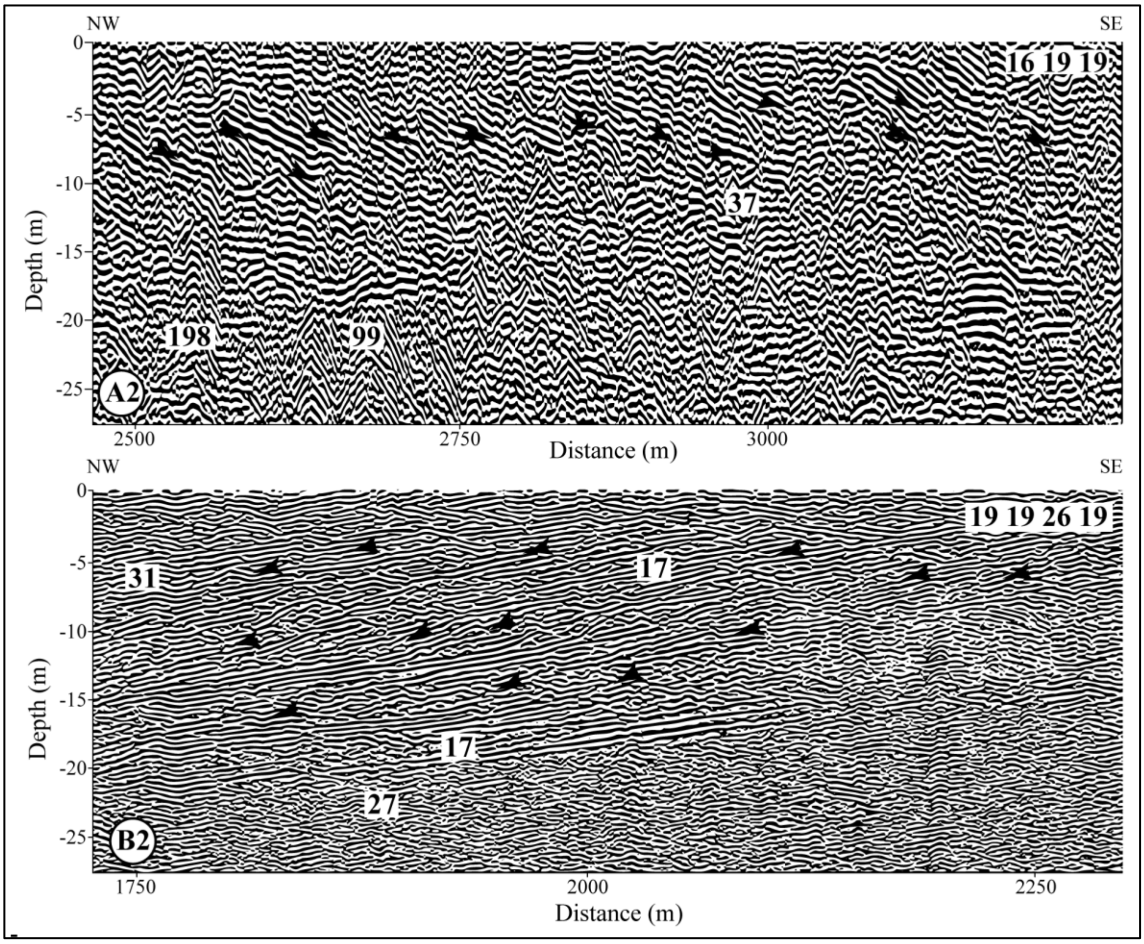

Fig. 7 Subsections of GPR images from Rondinha (A2) and Jardim do Éden (B2) transects. A2 presents ^{222}Rn activity (in dpm/L), plotted on the progradational stacking pattern units. The black arrows indicate the dive of the reflectors seaward. B2 presents ^{222}Rn activity (in dpm/L), plotted on the retrogradational stacking pattern units. The black arrows indicate the dive of the reflectors landward

In the Rondinha profile, the activity range was between 16 to 198 dpm/L, and it shows to slightly increase with depth (r^2=0.65). Subsection A2 presents higher ^{222}Rn activities for the samples obtained at depths under -18 m due to the insulation observed at this depth. At this point it is possible to find a different compartment composed of a Pleistocene substrate. Semi-confined and confined aquifers presented restrictions in mixing waters since the groundwater had little or no influenced on freshwater (from recharge) or marine water (from recirculation). The intermediary layers (-10/-18 m) present medium ^{222}Rn activities. These layers are composed of sandy deposits which allow for this partial mixing. Surficial layers have presented low ^{222}Rn activities due to the unconfined aquifer where the groundwater has been submitted to an intense mixing and evasion process.

In the Jardim do Éden profile, the ^{222}Rn activity range was 14 to 269 dpm/L, but without correlation with depth (r^2=0.42). The results show horizontal gradient slightly rises from ocean side to the lagoonal terrace side of the barrier (r^2=0.50). In subsection B2, the layer between 0 to -15 m of depth presents low activity due to the insulation. ^{222}Rn activity was higher in deeper wells because they are located below the bottom lagoonal unit and close to the Pleistocene, a confined compartment, allowing for layers enriched in ^{222}Rn.

c. SGD

The results presented in Table 2 indicate the difference in calculated SGD rate (W) and flux between progradational and retrogradational regions (p = 0.004; p = 0.004).

Table 2 Summary of ^{222}Rn and SGD data for Rondinha and Jardim do Éden transects during Austral Summer 2016

Location	^{222}Rn activity Rn_w (dpm/m³)	^{222}Rn flow F_{SGD} (dpm/m² d)	End member Rn_{pw} (dpm/m³)	SGD rate W (cm/d)	SGD flux (m³/m d)
Rondinha	1,866 ± 97	10,323 ± 521	21,341 ± 1,109	46.15 ± 2.4	13.85 ± 0.73
Jardim do Éden	1,940 ± 100	900 ± 29	26,160 ± 1,370	3.62 ± 0.1	1.09 ± 0.03

Considering that the salinity range of the piezometric wells did not reach zero salinity, defined as end member, this was established as the base for the extrapolation of the ^{222}Rn activities of the piezometric wells and surf zone, as shown in Fig. 8.

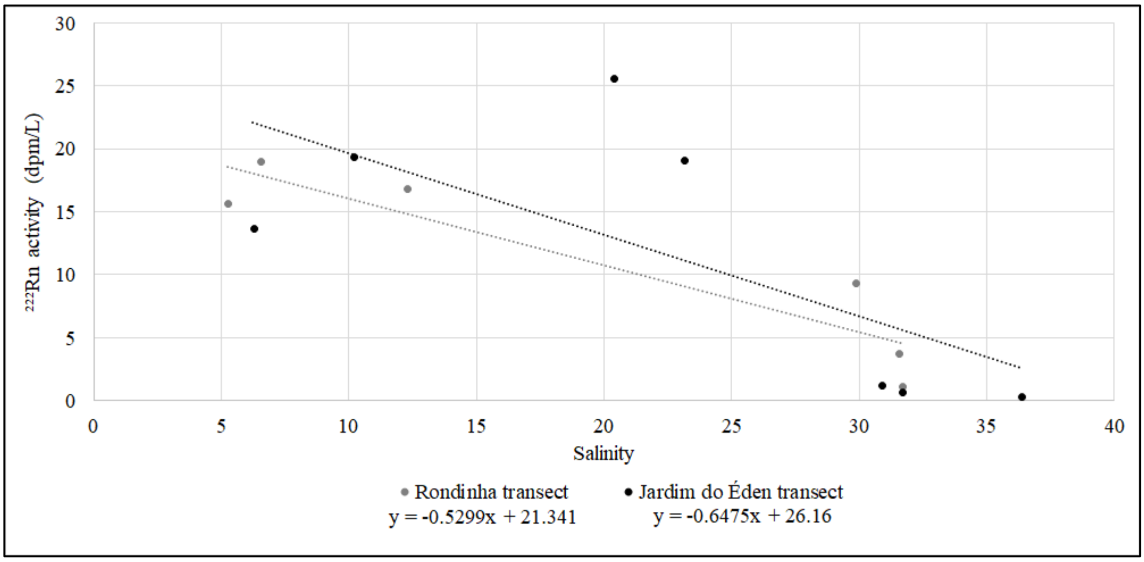

Fig. 8 Distribution of ^{222}Rn activities to beach wells (piezometers) and surfing zone of the respective sampled regions, Rondinha and Jardim do Éden. The extrapolation of the trend curves indicates the value prospected at zero salinity (end member)

The SGD for each region has been estimated at 969.3 ± 51.2 10³ m³/d in the progradational region and 59.7 ± 1.7 10³ m³/d for the retrogradational region. These values represent an order of magnitude and, if this difference is applied to the northern coastal region which these transects represent, it will be extremely significant. For instance, the Rondinha transect represents 70 km of coastline, which extends from Torres to the Atlântida Sul Beach, while the Jardim do Éden transect is representative of the stacking process that occurs from Imara Beach to Quintão, on 55 km of coastline. This gives an indication that the coastal regions with progradational processes have a great influence on the nutrient supply for biological production.

DISCUSSION

The evaluation of the images from the two GPR sections indicates that the stacking patterns (progradational and retrogradational) are different with reflexes in the ^{222}Rn activity. It is evident that no horizontal and vertical homogeneity has been pointed out. Vertical and horizontal variations of stratification and facies occur in this system due to the different depositional environments that have succeeded spatially and temporally but are still closely interconnected (Lisboa et al., 2004).

The characterization of the reflectors for the progradational region is arranged towards the sea, demonstrating a process of coastal growth. These reflectors (Fig. 7 A2 - black arrows) in agreement with ^{222}Rn activity indicate a mixing process with marine water closer to the ocean. The vertical gradient of ^{222}Rn activities observed in the Rondinha transect clarifies the mixing process between recharged waters with the deeper compartments, in view of the horizontal layerings, which result in creating compartmentalization.

Meanwhile, the reflectors in the retrogradational region (Fig. 7 B2 - black arrows) are arranged transversally to the continent, demonstrating a process of retrogradation/erosion of the coast, fortifying saline wedge effects and mixing more internalized when compared to the subterranean estuary of the progradational region. The horizontal gradient of the Jardim do Éden transect (Table 1) indicates the extent of influence of the SGD or even the salt wedge in the opposite direction. The absence of a vertical gradient demonstrates the existence of units that act as semi or non-permeable, as seen in the paleolagoonal bottom reflectors.

So, based on numerous studies that were carried out to this date (Barboza et al., 2011; Dillenburg et al., 2017, 2011; Dillenburg and Barboza, 2009; Lima, 2013; Rosa, 2012; Silva, 2011; Travessas et al., 2005; Watanabe, 2016), the stacking features, compaction and composition for progradational and retrogradational barriers can be summarize as presented in Table 3.

It demonstrates that, in general, both are very similar in climatic and hydrogeological influences (macro compartments), but locally there are some characteristics that can have diverse effects under water properties and processes.

Such reflectors are a historical result and they indicate how the deposition occurred as well as the layers that guide the water pathway. Although the layers in this region are quite permeable, the moderate degree of cementation allows for easy pathways, being oriented towards the ocean in the progradational region and to the continent in the retrogradational region. This characteristic alters, albeit minimally, the hydraulic gradient between the studied regions, since it creates a moment of incline or slope in the coastal barrier.

Table 3 Lithofacies and radarfacies of regressive and transgressive barriers in NCR, Southern Brazil

Barrier	Radarfacies	Stacking units	Compactation	Composition
Progradational	IV	aeolian	very low	fine and very fine sands
	VII	backshore/foreshore	moderate/high	fine and very fine sands
	VI	upper shoreface	moderate	fine sands
	V	lower shoreface	moderate	fine sands intercalated by mud
	I	Pleistocene substrate	moderate/high	fine muddy sands
Retrogradational	IV	aeolian	very low	fine and very fine sands
	III	lagoonal beach	low	fine and very fine sands
	III	aggradational	moderate	fine sands intercalated by mud
	II	lagoonal bottom	moderate	very fine muddy sands
	I	Pleistocene substrate	moderate/high	fine muddy sands

The driven forces that operate in this region are many, but most are equivalent or very similar within the two stations (tide, wave set up, rainfall, altimetry, seasons). Therefore, it is possible to assume that the difference in ^{222}Rn activity and SGD are due to the stacking pattern, playing the regulatory role, since it defines the different compositions, the stacking order, isolation conditions, formation of reservoirs, among other features of the coast geology.

A remarkable characteristic of our study site is the sharp gradients of salinity in beach wells (Table 1). For example, salinity ranged from 6 to 23 within a 65 m horizontal distance in Jardim do Éden Beach and from 5 to 12.5 within 15 m in Rondinha Beach.

However, for the unconfined permanent wells the salinity range was 0.05 to 0.06 and 0.06 to 0.14 for Rondinha and Jardim do Éden, respectively. It indicates a "lower" subterranean estuary dominated by freshwater in both regions, while the "upper" subterranean estuary (beach face) provides evidence of the saline mixing process, on a scale of a meter. Salinity data (although showing non-conservative behavior) indicates that this region has a surficial aquifer which is recharged locally. Although the behavior is dissimilar between "lower" and "upper" subterranean estuaries, these are closely connected and the activities observed within the sandy barrier were decisive for ^{222}Rn activities in the beach wells in each region.

The ^{222}Rn application revealed the water distribution in the coastal barrier and indicated that the region is dominated by several aquifers as it was mentioned by Niencheski and Windom (2015). The definition of the end member for each of the regions is an important result of this work, since it is often a difficult piece of data to obtain due to the high sample size required. However, the small difference between the end-member values observed in these regions (Table 2) highlights that both have a similar source of groundwater recharge, since they are part of the same coastal aquifer system (Lisboa et al., 2004; Machado and Freitas, 2005b), differing only in their depositional architectures.

Based on these findings, a significant difference is recorded in SGD rate between the progradational and retrogradational region. The SGD flux reaches more than 15-fold in the progradational region, an expressive variation.

In practice, variations in SGD flows have repercussions on the productive process of the coastal area. The associated nutrient fluxes (although not determined in this study) make up an important (if not major) source in the fertilization of coastal waters, being more expressive in the progradational region when compared to the retrogradation region.

CONCLUSION

The results obtained with the combination of the ^{222}Rn tracer and the GPR technique are innovative and prove that the GPR technique is a very useful tool for SGD studies, since it provides information on coastal depositional geological structures continuously. This is different from conventional testimonial methods, which, despite providing material for complete geological studies, are punctual and small in number.

In environments with differentiated structures such as CNR, GPR images are fundamental for understanding the stacking pattern and how, consequently, SGD occurs. Since this a pioneer study, the geological structures detected by GPR were validated using sedimentological data that was available for the region, proving to be a completely reliable method.

The heterogeneities of the barrier are sufficient to influence the SGD, due to the stacking, sediment, shape and depositional architecture characteristics. Thus, the geology of the progradation region gives 15 times more expressive SGD than in the retrogradational region.

Although it was not the objective of this work, it is possible to assume that a greater impact is produced in the flow of micro and macronutrients to the coastal zone in the progradational region. The SGD data for the progradational region are an order of magnitude higher than the values found for the retrogradational region. This indicates that it must have more prominent coastal biological productivity.

Finally, the more prospecting lines that are obtained with GPR, the more knowledge there is of the region. This makes it possible to optimize the definition of sampling design for future groundwater collections, either by choosing between pre-existing wells or by drilling new wells.

For this investigated region, which is 125 km long, future studies on the existence of traces of paleochannels in the barrier that are connected with the geological features observed on the continental shelf (Melo, 2017) are recommended. This may be preferred routes for SGD to reach locations farther from the coast. In addition, time series and more in-depth assessments of the beach strip should be developed.

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ARTIGO 2 - THE INFLUENCE OF GEOLOGICAL STACKING PATTERNS IN THE SUBMARINE GROUNDWATER DISCHARGE AND NUTRIENTS FLOW, SOUTHERN BRAZIL

Este artigo foi submetido ao jornal *Journal of Hydrology*/Elsevier, seguindo as respectivas normas de formatação.

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ABSTRACT

The influence of drivers in the Submarine Groundwater Discharge (SGD) has been extensively discussed as well as the coastal geology, the main focus of the present work. Both progradational and retrogradational stacking are evolution patterns which contribute differently to SGD due to their different composition and deposition features. These stackings compose the coast in Southern Brazil, covering 125 km of coastline, which were evaluated to determine the SGD and nutrient fluxes during the summers of 2016 and 2017. Samples from permanent wells, beach wells and surf zone in 13 sampling stations have indicated significant differences between progradational and retrogradational areas in radon activity ($\bar{x}_{REG}$ = 1,950±237 dpm/m³; $\bar{x}_{TRA}$ = 1,896±249 dpm/m³), SGD rate ($\bar{x}_{REG}$ = 32.9±2.8 cm/d; $\bar{x}_{TRA}$ = 6.9±0.8 cm/d), as well as for end member values ($\bar{x}_{REG}$ = 22,468±997 dpm/m³; $\bar{x}_{TRA}$ = 27,006±1,120 dpm/m³). Moreover, each sampling campaign was on the effect of ENSO and LNSO phenomena, typical in this region. Thus, there was a significant difference in the means of comparison tests between Season 1 and Season 2 results allowing to evaluate the combined effects of geology and weather. The freshwater component represents 73 e 56% of SGD in the progradational and retrogradational region, respectively, discharging more freshwater than ocean water recirculated. The "new" input represents 99% of the phosphate, 93% of the TDIN and 99% of the silicate to the continental shelf in the progradational area and 89% (phosphate), 87% (TDIN) and 99% (silicate) in the retrogradational area. The complexity of the process was reaffirmed once the nutrient enrichment is unequal.

Keywords: subterranean estuary; fluxes; transgressive; regressive; North Coast; LNSO; ENSO.

1. INTRODUCTION

The nutrient flux associated with the submarine groundwater discharge (SGD) has been studied by many authors (Andrade, 2010; Attisano, 2012; Bishop et al., 2015; Burnett et al., 2007; Lee et al., 2009; Lima, 2014; McCormack et al., 2014; Niencheski et al., 2007; Paiva, 2014; Rocha et al., 2018; Rodellas et al., 2014; Russoniello et al., 2016; Souza, 2015; Sugimoto et al., 2016; Tovar-Sánchez et al., 2014) in order to evaluate the impact of this important and well known process. The nutrient flux is a complex water movement described by Niencheski et al. (2007) according to the conceptual model, which is composed by five different fluxes. These flow from the continent to the coastal zone, recirculating through the permeable sediments before leaving the surf zone and going across the shelf to the open ocean. It describes a subterranean estuary which shows the same conditions for a typical estuary, mixing freshwater and seawater, being equivalent in volume of discharge and even with higher nutrient concentration (Burnett and Dulaiova, 2006; Rodellas et al., 2015; Simmons, 1992; Wang et al., 2017).

Even though most of the researchers agree with the conservative behavior model (Windom et al., 1999) where the concentration of nutrients and trace elements are correlated with salinity, these fluxes vary widely as observed in Southern Brazil and others countries (Attisano, 2012; Niencheski et al., 2007; Paiva, 2011; Wang et al., 2017; Weinstein et al., 2011). In fact, a number of factors are responsible for controlling and influencing these nutrient levels, like continental influence, anthropogenic uses, water recharge, sediment permeability, rainfall and seasonality (Andrade, 2010; Charette et al., 2003; Rocha et al., 2015; Sadat-Noori et al., 2015; Santos et al., 2012a), amongst others.

Although the composition and physical characteristics of the sediment have been considered, the role of coastal geology is not yet fully understood. This issue was already addressed by Rocha et al. (2018) in a study of radon (^{222}Rn) activities, and SGD has registered significant differences between two regions with progradational and retrogradational behavior in Southern Brazil (Figure 1). However, such a study is a preliminary evaluation based on only two sampling stations, leaving questions about the extrapolation of such behavior to large areas, such as CNRS with 150 km. This study area presents a stacking pattern characterized of lagoon/barrier depositional system, to the north with progradational behavior, and to the south with retrogradational behavior (Dillenburg and Barboza, 2014). This type of depositional system represents almost 15% of the world coastlines (Barnes, 1980) as well as areas of permeable sediments, which cover 40% of coastal areas worldwide (Riedl et al., 1972). Hence, understanding what role coastal sedimentology plays in the magnitude of SGD is a very important issue, which is still unknown.

In general, the region with progradational stacking showed SGD almost five times bigger than area dominated by retrogradational, resulting, in the same way, in higher volumes of water (Rocha et al., 2018). The difference is justified by the stacking structure, presenting layers slightly sandy and more compacted in the progradational area, stacked seaward, while the retrogradational region is slightly more muddy, lesser compacted and stacked landward (Rocha et al., 2018), which would favor the SGD.

Considering this hypothesis, it is possible to understand the forcing factor that plays an important role in the water movement from continental fluxes to seaward that defines the water flows through sandy beds. Lower flows mean bigger residence time of water inside of the barrier, moreover, it also means the contact water-sediment being longer and, consequently, it provides conditions to be enriched on nutrients and other elements (Santos et al., 2012b). Thus, even though some areas could show lower water discharge, it could be compensated with the exportation of higher nutrient levels.

The objective of this paper is to emphasize the relationship between geological stacking and the SGD and to assess the influence that the two more important geological features of the Northern Coast of Rio Grande do Sul state (NCR), progressive and transgressive can cause in the nutrient transport associated with SGD.

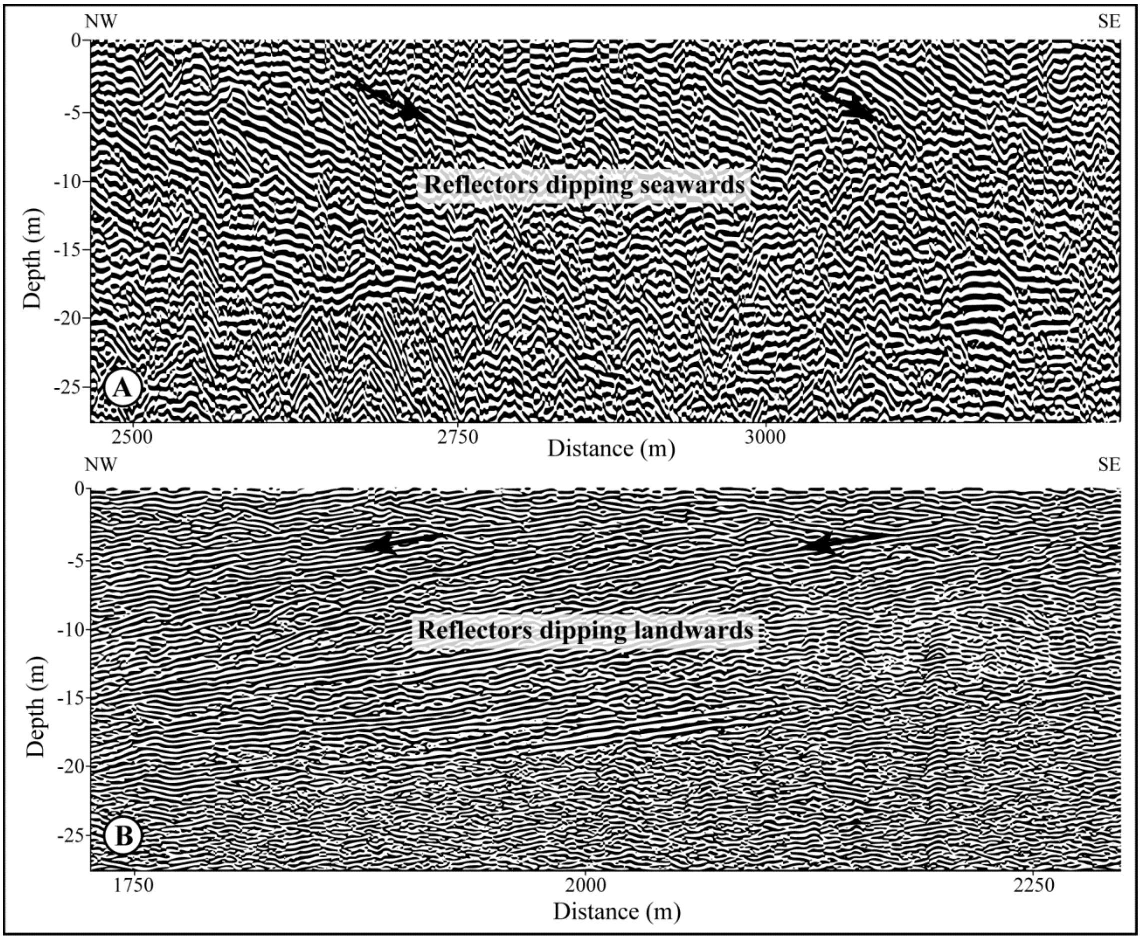

Figure 1 – Sections of GPR (Ground Penetrating Radar) images from Rondinha – progradational barrier area (A) and Jardim do Éden – retrogradational barrier area (B). The progradational area is represented by reflectors dipping seawards, while retrogradational area is represented by reflectors dipping landwards. Modified from Rocha et al. (2018).

2. MATERIAL AND METHODS

2.1. Study area

The NCR presents long Holocene lagoon/barrier depositional system, which has been extensively studied in the recent years, in particular its geology evolution (Clerot et al., 2001; Dillenburg et al., 2004b; Dillenburg and Barboza, 2009; Hesp et al., 2007, 2005, Martinho et al., 2010, 2008; Travessas et al., 2005). This lagoon/barrier depositional system was formed during the last glacio-eustatic transgressive cycle as a complex system (Villwock and Tomazelli, 1995), and the maximum sea level was

6-5 kyr with more 2-3 m, which has since fallen (Angulo et al., 2006; Barboza and Tomazelli, 2003). According to Rosa (2017, 2011), this Holocene barrier system corresponds to high-frequency depositional sequence stratigraphy.

The climate of the region, according to the classification of Köppen (Alvares et al., 2013), is subtropical constantly humid (Cfa), without a dry season, with hot summer. The average annual temperature is between 18 and 20 º C. Average annual precipitation is between 1,600 and 1,900 mm, with maximum values in summer months and lowest in winter.

This region is flat presenting eolian deposits of permeable sediments composed by fine and medium sand, well selected, with geomorphology dunes represented for barchan chains, isolated barchans, transverse chains, transgressive sand sheets and foredunes (Barboza et al., 2013; Dillenburg et al., 2017). These substrates ensure very good soil infiltration capacity (CPRM, 2015), aquifers with lateral and vertical continuity allowing large groundwater reservoirs with a porosity of up to 40%. Although these deposits are very similar, there are important variations in vertical and horizontal stratification, as well as in facies. Unequal depositional environments succeeded both spatially and temporally, but which are nevertheless closely interconnected (Lisboa et al., 2004).

Dillenburg et al. (2000) presented the two different types of barrier that occurs in this region, being the progradational and retrogradational barriers. A more recent study (Watanabe, 2016) reviews a large area of this stratigraphy as well as the evolution process of this rich and complex depositional system (Rosa, 2012).

The sediment supply and sea level combined to result in a depositional architecture, which according to Rosa (2012) when the supply is higher than accommodation space, allows for a progradation process called regressive behavior of the coastal line; and when the supply is lower, the system retrogradational being referred as transgressive behavior of the coastal line.

The progradational area presents itself in the embayments, and through the positive sediment, balance shows growth of coast with foredune ridges (Dillenburg and Barboza, 2009). Also, it shows reflectors dipping seaward (Rosa, 2012) which has prograded up to 40 m (Toldo Jr et al., 2005).

The retrogradational area presents itself in coastal projections and retrogrades eroding, including feeding the dunefields (Dillenburg and Barboza, 2009). In this region, the erosion was responsible for more than 300 m since last maximum sea level (Travessas et al., 2005) and it shows reflectors dipping landward (Figure 1B), which are justified by sediment migration from beach face to coastal lagoons (Rosa, 2012).

Considering that this 600 km barrier has been studied by many authors in the South, on the influence of Patos Lagoon (Attisano et al., 2008; Lima, 2014; Niencheski et al., 2007; Niencheski and Windom, 2015; Souza, 2015; Windom et al., 2007) and in the North, with the Guarani Aquifer Outcrop (Paiva, 2014), the present work considers the coast between both, which represents 125 km, being half in retrogradational and half in progradational behavior (Figure 2), an excellent place to test our hypothesis.

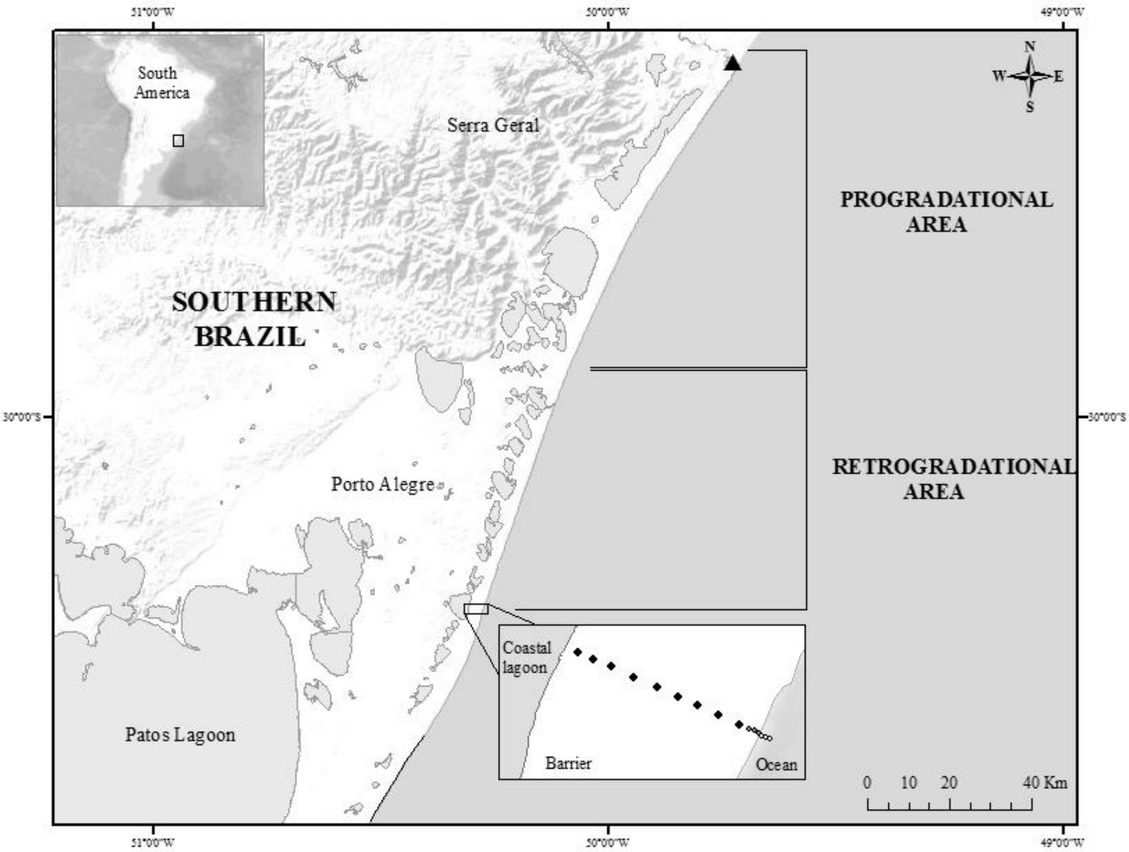

Figure 2 – Southern Brazil study site showing the transect applied in each beach, shows permanent wells (●), beach wells and surf zone (○), in regressive and transgressive areas, located between Guarani Aquifer Outcrop (▲) and Patos Lagoon Barrier (│).

The progradational region showed a mix/recirculation area closer to the sea, which facilitates the SGD, while in the retrogradational region the mix/recirculation area is located more inland allowing for less SGD.

The study area was defined as 1) progradational barrier, from Station 1 - Torres up to Station 7 - Atlântida Sul Beach, and 2) retrogradational barrier, from Station 8 - Imbé up to Station 13 - Quintão Beach. In each sampling location, water was sampled in permanent wells, piezometers and surf zone surface during the Austral Summer of 2016 (December 2015 to April 2016) and 2017 (January to April 2017), denominated here as Season 1 and Season 2, respectively, during the same climatic pattern with random wind and tide conditions.

2.2. Samplings and Analysis

The permanent wells were randomly chosen on particular properties with perpendicular distribution to the shoreline. These wells present varied depth on the varied distance to the shoreline. However, all area between the lagoon terrace and the beach face have been covered (Figure 1). The sampling procedure was done with the pre-existing pump in each well. A sample of 8 L was collected in a closed system plastic 10-L-bottle to analyze ^{222}Rn activity, and the second sample of 3 L, used to analyze pH, conductivity, salinity, and nutrients (phosphate, ammonium, nitrate, nitrite, TDIN, and silicate, following protocols described by Baumgarten et al. (2010).

Piezometers have been drilled at beach face, and the sampling procedure was the same described above. These wells were drilled 1 m deep, covering the beach face from the dune up to swash zone, using a pushpoint equipment and a peristaltic pump. Surf zone surface water was sampled in a transect with three points far from the beach 10, 20 and 30 m. In all sampling campaigns, complete meteorological data were obtained.

The choice of using ^{222}Rn instead of another geotracer is due to the fact that results can be obtained instantaneously and are easy to be quantified and handle. In this case, only a small volume of sample is required, which can be very useful when sampling is done in restricted or private areas. Also, when a sample cannot be measured *in locus*, the measurement can be performed *in route* from one station to another.

The ^{222}Rn activity measurement to define the ^{222}Rn inventory ($^{222}Rn.\lambda_{222}$) was made by the RAD7 monitor (Durrigde Inc.) with the Big Bottle system, which uses the internal pump of the monitor to disperse ^{222}Rn from 8 L volume of sample and circulate it to the counter for measurement. Before water sampling, the wells were pumped out to put away the water that was standing on the system; then, water was sampled in plastic bottles and stored until the analysis.

The main principle of using ^{222}Rn measurements to obtain SGD rates is based on isotope inventory regarding losses due to atmospheric evasion and mixing with lower concentration waters, converting all processes in fluxes, being dominated by the advective transport of groundwater through the sediment. With the assessment of the ^{222}Rn fluxes is easy to estimate water fluxes (Burnett and Dulaiova, 2003), being the F_{SGD} measured based on Equation 1, defined by Corbett et al. (1997).

$$F_{dif} + F_{SGD} + {}^{226}Ra.\lambda_{222} - {}^{222}Rn.\lambda_{222} - F_{atm} - F_{hor} = 0 \quad (1)$$

where:
F_{dif} – diffusion flow from ^{226}Ra concentration in sediment that decay to ^{222}Rn, in dpm/m² d;
F_{SGD} – ^{222}Rn flow, in dpm/m² d;
$^{226}Ra.\lambda_{222}$ – ^{222}Rn excess from ^{226}Ra father decay, in dpm/m² d;
$^{222}Rn.\lambda_{222}$ – ^{222}Rn inventory in the water column, in dpm/m² d;
F_{atm} – atmospheric flow or atmospheric loss, in dpm/m² d;
F_{hor} – flow horizontal mixing or loss by horizontal mixing, in dpm/m² d.

F_{dif} emanates from the sediment, from the ^{226}Ra concentration that decays to ^{22}Rn, and it is established by measurements of ^{22}Rn in sediment samples after an equilibration-time (Corbett et al., 1998) or, alternatively, measurements of ^{226}Ra directly from the sediment. So, the excess of ^{22}Rn from the decay of ^{226}Ra in the water column, detailed in Equation 1, was based in data from Windom et al. (2006) along the surf zone in Patos Lagoon barrier, close region, which is in average 79,7 ± 9,4 dpm/m³.

Similarly, there is the excess of ^{222}Rn from the ^{226}Ra decay in the water column which is also disregarded due to the same reason, high ^{226}Ra decay time.

The F_{atm} is based on the ^{222}Rn transfer estimate from water to air, considering the ^{222}Rn concentrations in the water column and in the air, as well as the water temperature, air temperature, wind speed, water salinity and the solubility of the ^{222}Rn gas (Burnett et al., 2006; Dulaiova and Burnett, 2006; Wanninkhof, 1992).

The horizontal mixing loss, F_{hor}, is based on the concept of steady-state advection-diffusion (Craig, 1969; Glover et al., 2005), which evaluates the ^{222}Rn activity and its decay constant, ^{222}Rn from ^{226}Ra decay, the turbulent diffusion coefficient and the horizontal advection. Another conservative tracer is taken, in this case, the conductivity, to establishes a mixing relation between ^{222}Rn and conductivity to find the radon flux in the horizontal mixing process (loss), using the transect distance applied in the measurements.

Although the SGD estimation based on the ^{222}Rn mass balance in steady-state is criticized due to some forces, tidal dynamics and wave set up; this study area presents quite similar micro-tidal between sample points being disregarded once it is smaller than 30 cm (Motta, 1969), near an amphidromic point. In wave height case, the mean is from 1.50 for 15-20 m depth (Motta, 1969) with variations of the order of 10% (1.40 to 1.56 m) (Lima et al., 2013). The effect is reduced at a depth up to 1 m, where the transect is developed.

Thus, the result in Equation 1 is the ^{222}Rn flux, F_{SGD}, with which is assessed the SGD rate by Equation 2, where the end member was based on ^{222}Rn activity in piezometric beach wells (salinity zero).

$$W = F_{SGD} \div Rn_{pw} \qquad (2)$$

where:

W – SGD rate, in m/d;

F_{SGD} – ^{222}Rn flow, in dpm/m² d;

Rn_{pw} – ^{222}Rn activity in groundwater or porewater, called end member, in dpm/m³;

To avoid overestimation, each end member is applied for the respective transect. How higher is the number of piezometers sampled, more robust is the F_{SGD} data. In this study, with 205 piezometers, an evaluation of the difference between each area and period was allowed.

The daily flux for each transect is estimated based on SGD rate (Equation 3).

$$Vol = W\ x\ D_{grad}\ x\ L \qquad (3)$$

where:

Vol – discharge volume for a specific area, in m³/d;

W – SGD rate, in m/d;

D_{grad} – gradient distance, in m;

L – length of the beach, in m.

The gradient distance corresponds to the transect distance sampled, for this study, 30 m. The lengths from the beach (L) used were 70 and 55 km for the progradational and retrogradational areas, respectively.

Based on the SGD rate and nutrient concentrations in each sample group (permanent wells, piezometers, and surf zone) were estimated the outcome in the nutrient delivery from continent to coastal waters (Equation 4, 5, 6 and 7) (Niencheski et al., 2007).

$$F_1 = [C]_{fw} \times Q_{fw} \quad (4)$$

$$F_2 = [C]_{bgw} \times SGD \quad (5)$$

$$F_3 = [C]_{sz} \times Q_{sw} \quad (6)$$

$$F_4 = F_2 - F_3 \quad (7)$$

where:

F_1 – fresh groundwater moving toward the ocean, in mol/d;

$[C]_{fw}$ – nutrient concentration in permanent wells, in mol/L;

Q_{fw} – freshwater component of SGD, in m³/d;

F_2 – SGD entering the ocean, in mol/d;

$[C]_{bgw}$ - nutrient concentration in beach wells, in mol/L;

SGD – submarine groundwater discharge, in m³/d;

F_3 – seawater recirculating through permeable sediments, in mol/d;

$[C]_{sz}$ - nutrient concentration in surf zone, in mol/L;

Q_{sw} – seawater component of SGD, in m³/d;

F_4 – flux to the inner continental shelf, in mol/d.

$[C]_{fw}$, $[C]_{bgw}$ and $[C]_{sz}$ are the nutrient concentrations in permanent wells, piezometers and surf zone, respectively. Q_{fw} and Q_{sw} are defined according to SGD salt balance (Equation 8 e 9) according to Niencheski et al. (2007):

$$SGD = Q_{fw} + Q_{sw} \quad (8)$$

$$Q_{fw} = \frac{SGD \times (Sal_T - Sal_{fw})}{Sal_T} \quad (9)$$

where:

Sat_T – total salinity expected, usually 36;

Sal_{fw} – average salinity in piezometer samples.

The ^{222}Rn determination in water and sediments were procedure at Ceclimar/UFRGS, as well as nutrients and physicochemical parameters for which was followed APHA (2012).

The results were evaluated statistically applying SPSS® software to establish the difference between progradational and retrogradational areas.

3. RESULTS

^{222}Rn activity and SGD

The results for ^{222}Rn activities and fluxes, end members and SGD rates and fluxes during Season 1 and Season 2 are presented in Table 1.

Evolution coastal	Station	^{222}Rn activity Rn_w (dpm/m³)	^{222}Rn flux F_{SGD} (dpm/m² d)	End member Rn_{pw} (dpm/m³)	SGD rate W (cm/d)	SGD flux SGD (m³/m d)
Season 1						
Progradational	1	856 ± 196	1,019 ± 224	38,830 ± 1,456	2.62 ± 0.58	0.70 ± 0.15
	2	2,161 ± 105	2,504 ± 102	20,349 ± 1,021	12.31 ± 0.50	3.18 ± 0.13
	3	1,866 ± 97	10,284 ± 516	18,662 ± 972	55.11 ± 2.77	4.33 ± 0.22
	4	678 ± 169	1,674 ± 410	14,805 ± 862	11.30 ± 2.77	1.45 ± 0.36
	5	988 ± 240	545 ± 123	30,695 ± 1,346	1.78 ± 0.40	0.33 ± 0.07
	6	1,995 ± 95	3,888 ± 165	38,521 ± 1,532	10.09 ± 0.43	2.65 ± 0.11
	7	406 ± 144	193± 66	105,225 ± 1,162	0.18 ± 0.06	0.06 ± 0.02
Retrogradational	8	1,402 ± 275	5,626 ± 661	11,526 ± 648	48.81 ± 5.74	2.05 ± 0.24
	9	447 ± 152	0 ± 0	35,147 ± 1,514	0.00 ± 0.00	0.00 ± 0.00
	10	1,940 ± 100	861 ± 24	19,256 ± 986	4.47 ± 0.13	0.43 ± 0.01
	11	3,790 ± 481	4,779 ± 818	23,254 ± 1,243	20.55 ± 3.52	2.07 ± 0.35
	12	2,050 ± 293	943 ± 120	32,230 ± 1,210	2.93 ± 0.37	0.43 ± 0.06
	13	624 ± 173	246 ± 61	23,096 ± 1,121	1.07 ± 0.27	0.09 ± 0.02
Season 2						
Progradational	1	2,058 ± 246	4,000 ± 462	9,630 ± 633	41.54 ± 4.80	12.42 ± 1.43
	2	3,623 ± 345	2,767 ± 240	5,699 ± 448	48.54 ± 4.20	6.86 ± 0.59
	3	8,906 ± 557	52,534 ± 3,263	48,331 ± 1,240	108.70 ± 6.75	51.84 ± 3.22
	4	14,940 ± 754	65,537 ± 3,284	24,588 ± 920	266.54 ± 13.36	48.18 ± 2.41
	5	10,328 ± 581	20,624 ± 1,140	4,157 ± 414	496.16 ± 27.43	12.57 ± 0.69
	6	13,693 ± 551	50,850 ± 2,463	33,306 ± 1,189	152.68 ± 7.39	27.07 ± 1.31
	7	3,991 ± 341	6,215 ± 683	12,038 ± 663	51.63 ± 5.67	14.23 ± 1.56
Retrogradational	8	25,902 ± 740	36,297 ± 1,648	6,411 ± 497	566.13 ± 25.70	83.78 ± 3.80
	9	3,381 ± 303	7,276 ± 679	30,759 ± 926	23.66 ± 2.21	2.78 ± 0.26
	10	249 ± 88	432 ± 150	19,937 ± 865	2.17 ± 0.75	0.69 ± 0.24
	11	4,284 ± 370	8,665 ± 730	41,032 ± 1,118	21.12 ± 1.78	8.00 ± 0.67
	12	1,880 ± 280	4,918 ± 719	5,739 ± 519	85.70 ± 12.53	3.77 ± 0.55
	13	8,133 ± 542	4,760 ± 298	42,279 ± 1,211	11.26 ± 0.71	5.53 ± 0.35

Table 1 - Data of ^{222}Rn activity and flux, end member and SGD rate and flux for each station in Southern Brazil during Seasons 1 and Season 2.

The F_{dif} results were estimated close to 58 dpm/m² d once the ^{226}Ra decay is low due to the low time of measure.

The F_{atm} was variable due to wind regime influence (ranging between 1 to 5 m/s during both seasons), air temperature (range 17.4 and 30º C for both seasons), and low ^{222}Rn air concentration (<2 dpm/m³), resulting in losses of up to 81x10³ dpm/m² d.

The F_{hor} varied between 0.2 and 11 dpm/m² d during Season 1 and between 0.2 and 270 dpm/m² d during Season 2, once the mixing coefficients vary between 165 and 16,050 for both periods.

These data show the contribution and relevance of each factor (losses and entrances) in the mass balance, being dominated by advection, mixing loss and atmospheric loss.

The end member data evidenced 10-fold variations between the sampling stations, for Season 1 and Season 2. The end member results are presenting a not normal distribution reinforcing the assessment based on its respective end member values, avoiding overestimation of SGD results.

Differences between each transect and also between the two sampling periods have been observed. When considering the results between progradational and retrogradational areas, it was possible to confirm the difference in SGD. Data from Station 8 was ruled out because this specific place is on the influence of Tramandaí River mouth, showing quite diverse results from other stations. Even with the exclusion, both areas have sampling stations at every nine kilometers. The regressive coastline is represented by 70 km, while the retrogradational area has 55 km of beaches.

Table 2 summarizes the results by areas, where the SGD rate and the SGD flux are higher in progradational area than in retrogradational region during both seasons.

Area	^{222}Rn activity Rn_w (dpm/m³)	^{222}Rn flux F_{SGD} (dpm/m² d)	End member Rn_{pw} (dpm/m³)	SGD rate W (cm/d)	SGD flux SGD (m³/m d)
Progradational	1,950 ± 237	3,294 ± 370	22,468 ± 997	32.41 ± 2.79	3.70 ± 0.37
Retrogradational	1,896 ± 249	2,488 ± 248	27,006 ± 1,120	6.87 ± 0.73	1.37 ± 0.25
Season 1					
Progradational	1,320 ± 136	1,233 ± 127	30,695 ± 1,162	7.02 ± 0.50	0.90 ± 0.13
Retrogradational	1,382 ± 197	702 ± 61	23,254 ± 1,210	2,93 ± 0.27	0.35 ± 0.02
Season 2					
Progradational	6,594 ± 509	20,624 ± 1,140	12,038 ± 663	95.63 ± 6.86	14.76 ± 1.43
Retrogradational	3,162 ± 303	4,760 ± 450	30,759 ± 926	21.12 ± 1.41	3.77 ± 0.39

Table 2 – Data of ^{222}Rn activity and flux, end member and SGD rate and flux for the progradational and retrogradational area in Southern Brazil during both seasons, Season 1 and Season 2.

An important forcing factor for SGD is the rainfall. The pluviometry was evaluated for Season 1 and Season 2, and the results were 78 and 174 mm/month, respectively.

To assess the fluxes of SGD and the components to determine how much freshwater has been delivered to the ocean, the salt balance it applied and the result are then calculated by the area (Table 3).

Evolution coastal	Salinity beach groundwater	SGD (10^4 m³/d)	Q_{fw} (10^4 m³/d)	Q_{sw} (10^4 m³/d)
Progradational	10.1	54.94 ± 5.49	40.14 ± 4.01	14.80 ± 1.48
Retrogradational	15.3	11.41 ± 1.14	6.45 ± 0.64	4.96 ± 0.50
	Season 1			
Progradational	10.5	6.55 ± 0.94	4.65 ± 0.67	1.90 ± 0.27
Retrogradational	15.0	2.05 ± 0.13	1.20 ± 0.08	0.85 ± 0.05
	Season 2			
Progradational	9.7	103.32 ± 10.04	75.62 ± 7.35	27.70 ± 2.69
Retrogradational	15.7	20.76 ± 2.15	11.70 ± 1.21	9.06 ± 0.94

Table 3 – Estimated SGD flux, freshwater and seawater components for progradational and retrogradational areas during both seasons, Season 1 and Season 2.

Nutrient fluxes

The evaluation of these components in the nutrient flux context allows a better understanding of how the input of nutrients and the potential transport for the inner shelf and offshore is. This assessment is based in nutrient concentrations in each area (in permanent wells, piezometers and surf zone) and can explain the frequently algae bloom and limiting nutrients (Table 4).

Evolution coastal area	Season	N	Salinity	PO_4^{3-} (µM)	SiO_2 (µM)	NH_4^+ (µM)	NO_2^- (µM)
Permanent wells							
Progradational	average	88	0.07	0.37	77.11	16.01	0.03
	1	50	0.06	0.20	157.49	34.36	0.03
	2	38	0.11	0.50	77.11	8.81	0.04
Retrogradational	average	88	0.11	0.72	269.98	14.44	0.07
	1	50	0.10	2.94	354.28	16.31	0.05
	2	38	0.11	0.67	174.11	7.52	0.05
Piezometric wells							
Progradational	average	99	10.45	2.17	53.12	9.42	0.04
	1	43	10.94	4.94	49.39	10.50	0.05
	2	56	8.36	0.29	56.84	8.61	0.04
Retrogradational	average	92	13.79	0.97	47.82	9.53	0.03
	1	40	11.89	3.29	34.30	54.25	0.07
	2	52	16.06	0.26	124.78	9.97	0.02
Surf zone							
Progradational	average	38	31.43	0.10	1.34	2.49	0.11
	1	17	29.24	29.33	1.34	4.47	0.12
	2	21	32.36	ND	1.34	2.26	0.10
Retrogradational	average	34	30.70	0.25	1.34	2.79	0.06
	1	16	27.84	24.85	1.34	8.30	0.05
	2	18	32.68	ND	1.34	2.26	0.05

ND: not detectable (<0,1 µM).

Table 4 – Nutrient concentrations in permanent wells, piezometric wells and surf zone for progradational and retrogradational areas during both seasons, Season 1 and Season 2.

Complementary data from these samples are available in Appendix 1 and 2 at the end of this paper.

4. DISCUSSION

^{222}Rn activity and SGD

Regarding the subterranean estuary results, the first approach for SGD evaluation was to present data station by station (Table 1) to indicate ranges and averages in SGD in different sampling areas and times, especially concerning end member values, scarce discussed in the literature.

The results of the ^{222}Rn activity, SGD flux and rate showed the difference between the sampling stations (1 to 13) and between the areas (progradational and retrogradational) within the same season, as well as between periods (Season 1 and 2). The data range between progradational and retrogradational areas overlap, however, the SGD in the progradational area is more expressive that retrogradational area. The end member for this area was also higher, indicating a higher SGD potential in this region, a result of the local geology.

When comparing SGD rates and fluxes during Seasons 1 and 2, it is evident that the geological differences are reinforced by the unequal rainfall during both periods. The effects are also observed on the end member results when these are reduced by the higher water recharge.

Although very clear, these data were evaluated statistically to understand how significant and non-random this difference between regions and periods would be (Table 5). These comparisons confirmed significant differences between the data obtained in the previous established geological features, progradational and retrogradational areas, for both seasons.

Comparisons	SGD rates averages (cm/d)	Significance
Spatial		
Progradational (P) x Retrogradational (R)	(45.04 ± 3.49) x (7.87 ± 0.73)	p = 0.009
Progradational 1 (P1) x Retrogradational 1 (R1)	(7.02 ± 0.50) x (2.93 ± 0.27)	p = 0.043
Progradational 2 (P2) x Retrogradational 2 (R2)	(95.63 ± 6.86) x (21.12 ± 1.41)	p = 0.043
Temporal		
Progradational 1 (P1) x Progradational 2 (P2)	(7.02 ± 0.50) x (95.63 ± 6.86)	p = 0.018
Retrogradational 1 (R1) x Retrogradational 2 (R2)	(2.93 ± 0.27) x (21.12 ± 1.41)	p = 0.138

Table 5 – Multiple comparison tests of SGD rates averages for progradational and retrogradational areas during Season 1 and Season 2.

When all data obtained in this work were considered together, the statistical tests indicated a significant difference (p=0.009) in the SGD between progradational and retrogradational areas, which confirmed the hypothesis that the geological features are determinant for the SGD.

In spite that both samplings were collected during the summer season, being one in dry period (2016) and another in the wet period (2017). This situation is a typical climatologic behavior in the study area, which is frequently influenced by *El Niño* Southern Oscillation (ENSO) and *La Niña* Southern Oscillation (LNSO) phenomenon. Thus, Season 2 has an average of 23% more monthly rainfall than Season 1.

According to other studies in the same region and even worldwide (Andersen et al., 2007; Gonneea and Charette, 2014; Niencheski et al., 2007; Sadat-Noori et al., 2015; Santos et al., 2012b, 2008b; Swarzenski et al., 2007), is important to assess the temporal effects on SGD, mainly in this region known by rainfall oscillations, as an indicative of a) geological influence to be annulled or to contribute by the weather phenomenon, and of b) which is the temporal influence for each geological feature, understanding if the effects will be the same or not.

It is important to emphasize that when the decision to do this work in two consecutive summers was made, access to all the permanent wells chosen *a priori* was guaranteed, since the majorities of the properties that should be visited are second houses, opened only during the summer season. However, fortunately for the development of this work, the two summers presented very different characteristics of rainfall, due to the effects of ENSO and LNSO.

Hence:

a) when the two sample periods were considered separately (P1 *versus* R1 and P2 *versus* R2), the difference in SGD kept significant both in Season 1 ($p=0.043$) and Season 2 ($p=0.043$), which indicate that the geological influence is determinant both in the dry and wet period, and

b) when the SGD was assessed by time, the results were different for each geological feature, being different significantly ($p=0.018$) for the progradational area (P1 *v.* P2), while for the retrogradational area (R1 *v.* R2) it was not ($p=0.138$).

In the studied area, the progradational area presents a progradational stacking pattern dipping seawards, with reflectors also dipping seawards and with sandier layers than the retrogradational area (Rocha et al., 2018), resulting in higher permeability and porosity, allowing the high SGD.

The retrogradational area presents retrogradational stacking pattern composed by sand layers intercalated by mud and reflectors dipping landwards (Rocha et al., 2018), which restricts the SGD due to reduced permeability and the lower water movement among the geological units.

These geological features also explain the temporal effect of SGD between dry and wet periods. In progradational regions, the higher rainfall increases the hydraulic gradient. That, in addition to the high permeability and the arranged seaward, results in higher SGD. In the retrogradational area, the water displacement to the ocean has more limitations and the SGD is decreased, even with higher rainfall, once the stacking is to landwards and the layers have lower permeability.

The results above mentioned indicating that the geological influence predominates in this region being magnified by the weather influence.

The flow in the progradational area is, in average, five folds higher in the retrogradational region and the difference in salinity beach groundwater evidence the higher influence of freshwater in progradational portion (Table 3).

Also, it is relevant to consider that even the extent of the barrier being smaller in the retrogradational region, this does not favor the higher DAS, indicating the importance of the depositional architecture.

The flow of Patos Lagoon in the summer (dry season) is 1,400 m^3/s (Niencheski and Windom, 1994) and 75 m^3/s for Tramandaí Estuary (Motta, 1965), both estuarine systems located closest and responsible by surficial drainage to the ocean. Therefore, during Season 2, when the SGD was more expressive in the whole study area, it represents 1 and 19% of Patos and Tramandaí's flows, respectively.

The hydraulic gradient acting on the PLB is a response to the water level in Patos Lagoon, 3 m above sea level (Windom and Niencheski, 2003). The same effect occurs on the CNRS barrier since the water level in coastal lagoons also is positive. For example, Itapeva and Quadros lagoons present 1.3 m and 0.8 m water level, respectively (Bitencourt, 2015; Lima, 2013), which reinforces the hydraulic action through the permeable barrier.

Although lower flow, the freshwater component (Q_{fw}) is more expressive, representing 71-73% of SGD in the progradational area and 56-58% of SGD in the retrogradational area, while in Patos Lagoon Barrier, for example, the freshwater component represents only 36% (Niencheski et al., 2007). That means the SGD in this study area is composed mainly by new waters, which introduces new nutrients in the coastal system, and added to recirculated nutrients from seawater component (Qsw), could explain the frequent algae blooms registered in this region, recognized as nutrient-rich even far from superficial drainages points.

Nutrient fluxes

Given this, another goal of this work is to know the nutrient fluxes associated with SGD and, ultimately, the primary production for each area.

In general, the nutrients concentrations (Table 4) present differences. For permanent wells, phosphate and silicate are higher in retrogradational, and nitrate is higher in the progradational area. The piezometric wells exhibit somehow an opposite behavior: phosphate higher in the progradational area and nitrate higher in the retrogradational area. In surf zone, the concentrations were quite similar, being phosphate more concentrated in the progradational area, nitrate more concentrated in the retrogradational area and silicate concentrations were the same.

Evolution coastal	Nutrients (10^3 mol/d)	F_1	F_2	F_3	F_4
Progradational	Phosphate	**0.15**	1.19	0.01	1.18
	Ammonium	6.42	**5.18**	0.37	**4.81**
	Silicate	30.95	29.18	**0.20**	28.98
Retrogradational	Phosphate	**0.05**	0.11	0.01	**0.10**
	Ammonium	0.93	**1.09**	0.14	0.95
	Silicate	17.41	5.45	**0.07**	5.39
	Season 1				
Progradational	Phosphate	**0.01**	0.32	0.56	**0.00**
	Ammonium	1.60	**0.69**	0.09	0.60
	Silicate	7.33	3.24	**0.03**	3.21
Retrogradational	Phosphate	0.04	**0.07**	0.21	**0.00**
	Ammonium	**0.20**	1.11	0.07	1.04
	Silicate	4.24	0.70	**0.01**	0.69
	Season 2				
Progradational	Phosphate	**0.38**	**0.30**	**0.00**	**0.30**
	Ammonium	6.66	8.90	0.63	8.27
	Silicate	58.31	58.73	0.37	58.36
Retrogradational	Phosphate	0.08	**0.05**	**0.00**	**0.05**
	Ammonium	**0.88**	2.07	0.21	1.86
	Silicate	20.37	25.90	0.12	25.78

Table 6 – Estimated nutrient fluxes (averages) for the progradational and retrogradational area in southern Brazil during Season 1 and Season 2. Limiting nutrients are indicated in bold.

Although the nutrient concentrations were different, there was not a pattern between each area and for that reason, it is not possible to confirm the hypothesis that retrogradational area is enriched in nutrients concentrations, considering lower SGD. In regards to the fluxes, based on each flow and concentration, it was possible to estimate each nutrient flux (Table 6). Limiting nutrients are indicated according to Redfield ratio to estimate the primary production potential in each area, discussed ahead.

These nutrient fluxes are spread over 125 km, being 70 km of progradational area and 55 km of retrogradational area, and they showed lower values when compared with Patos Lagoon barrier (ca. 10^6 mol/d) located further south (Niencheski et al., 2007). For this study, however, it is necessary to take into account that the considered area is smaller, with lower SGD and lower nutrient concentration when compared with Patos Lagoon barrier.

Considering the difference between F_1, F_2 and F_3 it is possible to understand that the input of TDIN, phosphate, and silicate to the nearshore (F_4) is expressive, especially in the progradational areas.

For a temporal evaluation, differences for the general inputs (F1 up to F4) were identified between the seasons, being higher during the wet season (Season 2) than the dry season (Season 1). Moreover, in the phosphate case, during Season 1, F_3 is higher than F_2, pointing to seawater recirculated as the main source.

Furthermore, the continental flux is in general considered as the main source of phosphate, TDIN, and silicate. While the Season 2 (wet) presented same pattern for progradational and retrogradational areas, during the Season 1 (dry) the seawater recirculated flux became the source for phosphate, which is observed for both areas. During the dry period, the hydraulic gradient is lowered and thus, become less significant, which causes the seawater to be the main source of phosphate. This process is likely to be influenced by the currents features in this area, some summers on Brazil Current and others on Malvinas Current, which are known nutrient-poor and nutrient-rich currents, respectively.

The phosphate flux F_1 is governed by the pH and salinity of the environment, which regulates the phosphate available. Inland the pH means is lower when compared by beach wells and surf zone samples, which allows for higher phosphate adsorption onto iron-manganese (hydr)oxides. When the pH is higher and lower salinity, as observed in beach wells, the phosphate finds conditions to be released (Gonneea and Charette, 2014), agreeing with higher phosphate concentrations observed in F_2.

As a result of the absorption to primary production, the silicate flux F_3 is lower while F_1 is higher than F_4, as registered in others studies (Attisano, 2012; Lima, 2014; Niencheski et al., 2007; Souza, 2015), except during Season 2, when the higher rainfall volume increases the silicate flux F_4. This phenomenon contradicts a study by Souza (2015), which described a dilution process with high level in Patos Lagoon.

Hence, overall the nutrient flux is higher in progradational areas, which oppose the previously proposed hypothesis where lower SGD in the retrogradational region allows longer residence time and more enriched fluxes. Even when considering the proportion of the coastal length for each coastal evolution process, the progradational area has higher nutrient fluxes. As observed previously, the freshwater/seawater mix zone location influences on nutrient discharge (Rocha et al., 2018). So, in the progradational area where the mix/recirculation zone is located closer to the sea, the result is a higher influence of freshwater with higher nutrient concentrations, while in the retrogradational region, the seawater influence prevailing once the mix/recirculation area is located more inland, resulting in lower nutrient concentrations.

Although every flux to continental shelf (F_4), except phosphate in Season 1, was positive and showed an average of 95% of F_2 value, indicating that a fair amount of "new" nutrients are arriving in the coastal system. In progradational areas, the "new" input means 99, 93 and 99% for P, TDIN, and Si, respectively, while for retrogradational the values are 89, 87 and 99%, respectively. Thus, F4 is the amount that escapes, meaning that surf zone is a source to the continental shelf.

The primary production is affected by these unequal fluxes for both seasons. Based on Redfield ratios, the limiting nutrient changes (Table 6). In a single evaluation of each season, most of the fluxes are P limiting (68%), opposed to Patos Lagoon Barrier and others areas (Niencheski and Windom, 2015) where the nutrient limiting is TDIN or nitrogen compounds.

Also, the temporal effects determined that rainfall keeps the phosphate fluxes, in general, and increases the TDIN and silicate fluxes in an average 6 and 20 times, respectively.

The phosphate flux presented low results in F_1 different than what was observed by Souza (2015) in Patos Lagoon Barrier, where a registered low F_3 allowed F_4 to be closer than F_1. The present study, however, registered higher values in F_2 than in F_3, allowing similar results between F_4 and F_2, also registered by Lima (2014) in Patos Lagoon Barrier in 2006. So, these variations in concentrations, SGD and fluxes showed the complexity of these systems.

It is also interesting to consider the difference between results from the coastal lagoons and the barrier. The mean concentration of nutrients in coastal lagoons in this area is ca. 2.6 and 16.1 µM for total phosphate and ammonium (Rocha, 2013), higher than most of the permanent wells, beach wells or surf zone concentrations. The nutrient flux, based on average of freshwater flow in some of these lagoons, described by Rocha et al. (2015), is 7.4 10^3 mol/d and 45.9 10^3 mol/d for phosphate and ammonium, respectively. Furthermore, in coastal lagoon system, the limiting nutrients are nitrogen compounds, and this result indicates that the coastal lagoons act like a nutrient trap and the barrier is geologically different, which gives particular features to groundwaters.

At last, the nutrient fluxes associated with SGD in this study area allows a carbon production of 72 and 19 g/m² by year for the progradational and retrogradational area, respectively. This supports an expressive annual production of 3.4 10^8 g C for 125 km of coastline. During Season 1 was 10 and 19 g C/m² y, for progradational and retrogradational, respectively, while Season 2 were 66 and 15 g C/m² y. These results showed that the rainier periods have different effects on the primary production, especially because of the change in the nutrient ratios. Also, an increase in nutrient fluxes delivery could be expected for the winter season. Since the winter season, it is the period with recurrent algal blooms, it was not included in this study.

5. CONCLUSIONS

The present study emphasizes that geological stacking in NCR, Southern Brazil, represent a decisive forcing factor, driving the SGD, according to geological features.

The evaluation based in nutrients concentration and fluxes in different geological conditions did not represent a pattern of enrichment in areas with lower SGD, being recommended an additional evaluation during ENSO and LNSO influence, as well when both are absent.

The flux of nutrients observed in NCR is expressive, and it is evidence that this region is important. Although it delivers lower nutrient flux than Patos Lagoon Barrier or Guarani Outcrop, it is a coastal area in Southern Brazil considered as enriched by SGD and ecologically productive.

ACKNOWLEDGMENTS

The authors wish to acknowledge the excellent support from the staff of the CECLIMAR/Federal University of Rio Grande do Sul (UFRGS) who helped ensure that this experiment was successfully carried out. A grant from the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) provided financial support for this research, Instituto Nacional de Ciência e Tecnologia em Ciências do Mar (INCT Mar-COI 565062/2010-7) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq 303672/2013-7, 302822/2017-8 and 302822/2017-8).

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ARTIGO 3 – CARACTERÍSTICAS GEOLÓGICAS DA CNRS, BRASIL: DRIVERS NO FLUXO DE ELEMENTOS 3

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RESUMO

A influência da geologia na Descarga de Água Subterrânea (DAS) e na distribuição de nutrientes vem sendo esclarecida usando como área teste a Costa Norte do Rio Grande do Sul (CNRS). A discussão mais específica dos efeitos sobre os elementos traço e seu transporte para plataforma costeira, foco deste trabalho, evidencia diferenças na distribuição, bem como nos fluxos resultantes. Amostragens em cinco transectos de praia (poços rasos) mostraram faixas de concentrações diferentes para os elementos manganês, estrôncio, alumínio, vanádio, molibdênio, ferro, níquel, cobre e chumbo. Os gradientes salinos também são desiguais e não há um padrão de aporte continental ou marinho entre elementos, assim como não há entre transectos. Os drivers que governam a DAS e tais alterações são equivalentes entre as estações, restando à geologia com suas feições e descontinuidades geológicas a justificativa para tamanhas variações. Transectos onde se registram o Afloramento do Aquífero Guarani (AAG), a desembocadura do Estuário Tramandaí e o afloramento de lama paleolagunar na faixa de praia, comparados com outras regiões onde estes não ocorrem demonstraram variações no pH e no processo de mistura. No transecto de Jardim do Éden, inclusive, redunda em uma compartimentação em vista de uma unidade confinante. A CNRS se destaca pelas concentrações de elementos traços maiores que as registradas na Barreira da Laguna dos Patos e na região do Afloramento do Aquífero Guarani, ainda que seus fluxos sejam reduzidos.

Palavras-chave: estuário subterrâneo; regressivo; transgressivo; Litoral Norte; afloramento do Aquífero Guarani.

INTRODUÇÃO

O conceito do estuário subterrâneo (Moore, 1999, 1996a) esclareceu o importante processo de descarga de água doce submarina) para o ambiente marinho. Este mecanismo é semelhante à descarga superficial de um estuário. A mistura entre água subterrânea e água marinha na zona subsuperficial, com intenso contato entre a água e o substrato sedimentar, permite uma gama de processos biogeoquímicos e importante fluxos de nutrientes, elementos traço e contaminantes para a região costeira (Gonneea and Charette, 2014; Niencheski et al., 2007). A exposição prolongada à rocha-mãe produz características de altas concentrações de alguns nutrientes na descarga de água subterrânea (DAS) (Kroeger et al., 2007; Moore et al., 1996; Santos et al., 2008c; Slomp and Van Cappellen, 2004; Weinstein et al., 2011) e o seu efeito sobre a produção biológica pode ser comparado ao escoamento superficial.

Estudos ao longo dos últimos vinte anos mostraram como a DAS e os fluxos de elementos químicos associados são importantes em sistemas costeiros em todo o mundo (Attisano, 2012; Charette et al., 2003; Niencheski et al., 2007; Peterson et al., 2008; Stewart et al., 2015; Wang et al., 2017; Windom et al., 1999). Em alguns casos, a DAS representa a principal entrada de nutrientes para esses sistemas costeiros (Kwon et al., 2014; Rodellas et al., 2014; Sospedra et al., 2017). No sul do Brasil, numerosos estudos em lagoas costeiras (Andrade, 2010; Paiva, 2011; Rocha, 2014; Rocha et al., 2015; Santos et al., 2008b) e ao longo do litoral (artigo 1; Windom et al. 1999; Attisano 2012; Lima 2014; Paiva 2014; Niencheski and Windom 2015; Souza 2015) demonstraram a magnitude da DAS e os fluxos associados de nutrientes e elementos traço.

A distribuição da DAS não é homogênea ao longo de um determinado trecho da costa e a heterogeneidade pode ser influenciada por diferentes fatores (Russoniello et al., 2013; Stieglitz et al., 2008). Moore (1999) compara algumas características desses sistemas complexos (como a zona de mistura, tempo de residência, impacto humano, fluxo, íons e fauna associada) com estuários superficiais, assim como estudos posteriores sugerem fatores variados que poderiam influenciar a DAS (Andrade et al., 2011; Charette et al., 2003; Corbett et al., 1999; Rocha et al., 2015; Sadat-Noori et al., 2015; Santos et al., 2009a, 2008c; Swarzenski, 2007). Em muitos casos, a geologia em torno do estuário subterrâneo determina os constituintes químicos e taxa de descarga (Russoniello et al., 2013; Santos et al., 2012b; Ullman et al., 2003), porém mais estudos sobre as características sedimentares são ainda necessários, sendo, em alguns casos, recomendado uma melhor cobertura espacial, em vez de amostragens frequentes nas mesmas localizações (Beck et al., 2007).

A Costa Norte do Rio Grande do Sul (CNRS), no sul do Brasil, é caracterizada por duas áreas distintas de configuração geológica com histórias evolutivas que, certamente, impactam a DAS nesta região. Estas são áreas com distintos padrões de empilhamento, progradacional (regressivo) e retrogradacional (transgressivo), apresentando estratigrafia e composição específicas (Barboza et al., 2011, 2009; Dillenburg and Barboza, 2014; Rosa, 2012). Ambas estão incluídas em uma barreira que foi formada por ciclos regressivo/transgressivo (Villwock and Tomazelli, 1995). Com base no suprimento de sedimentos e no nível do mar, cada área desenvolveu uma arquitetura deposicional específica. Na área progradacional, o balanço de sedimentos foi positivo com o crescimento da costa (Rosa 2012) enquanto que na área retrogradacional, o balanço foi negativo, havendo migração de sedimentos da face da praia para as lagoas costeiras (Rosa, 2012) e, erosão no campo de dunas (Dillenburg and Barboza, 2009).

Em estudo anterior na região (artigos 1 e 2 deste trabalho), foram verificadas diferentes atividades de radônio (^{222}Rn) e taxas de DAS em ordem de magnitude diferente entre as áreas progradacional e retrogradacional, produzindo variações significativas tanto nas concentrações de nutrientes como de fluxos para a plataforma interna.

Este estudo tem como objetivo determinar o papel dessas duas importantes características geológicas da CNRS quanto ao mecanismo de transporte de elementos traço para a plataforma costeira do Oceano Atlântico Sul, bem como os fluxos resultantes.

MATERIAL E METÓDOS

Área de estudo

A área de estudo está localizada na CNRS, sul do Brasil, entre 29º 21 'S e 30º 25' S e 49º 43 'O e 50º 20' O, na porção terrestre da Bacia de Pelotas, formada durante repetidos eventos de transgressão marinha, formando os sistemas Lagoa-Barreira durante o Pleistoceno e Holoceno (Villwock et al., 1986).

Estabelecidos estes sistemas, a costa se desenvolveu com o balanço de sedimentos positivos e negativos e, de acordo com isso, a região costeira evoluiu em dois formatos, apresentando parte de área com processo costeiro progradacional e parte com comportamento retrogradacional. As principais características da área progradante é estar localizada nos embaiamentos e apresentar padrão de empilhamento com refletores de georadar mergulhando em direção ao mar, enquanto a região retrogradante está localizada principalmente em projeções da costa apresentando refletores de georadar mergulhando em direção ao continente (Barboza et al., 2011; Dillenburg et al., 2017, 2011; Rosa, 2012; Silva, 2011; Watanabe, 2016). Além disso, a composição e compactação das camadas são diferentes, sendo mais arenosas e mais compactadas na área progradacional quando comparadas com a região retrogradacional (artigo 1), características que poderiam influenciar na composição da água subterrânea.

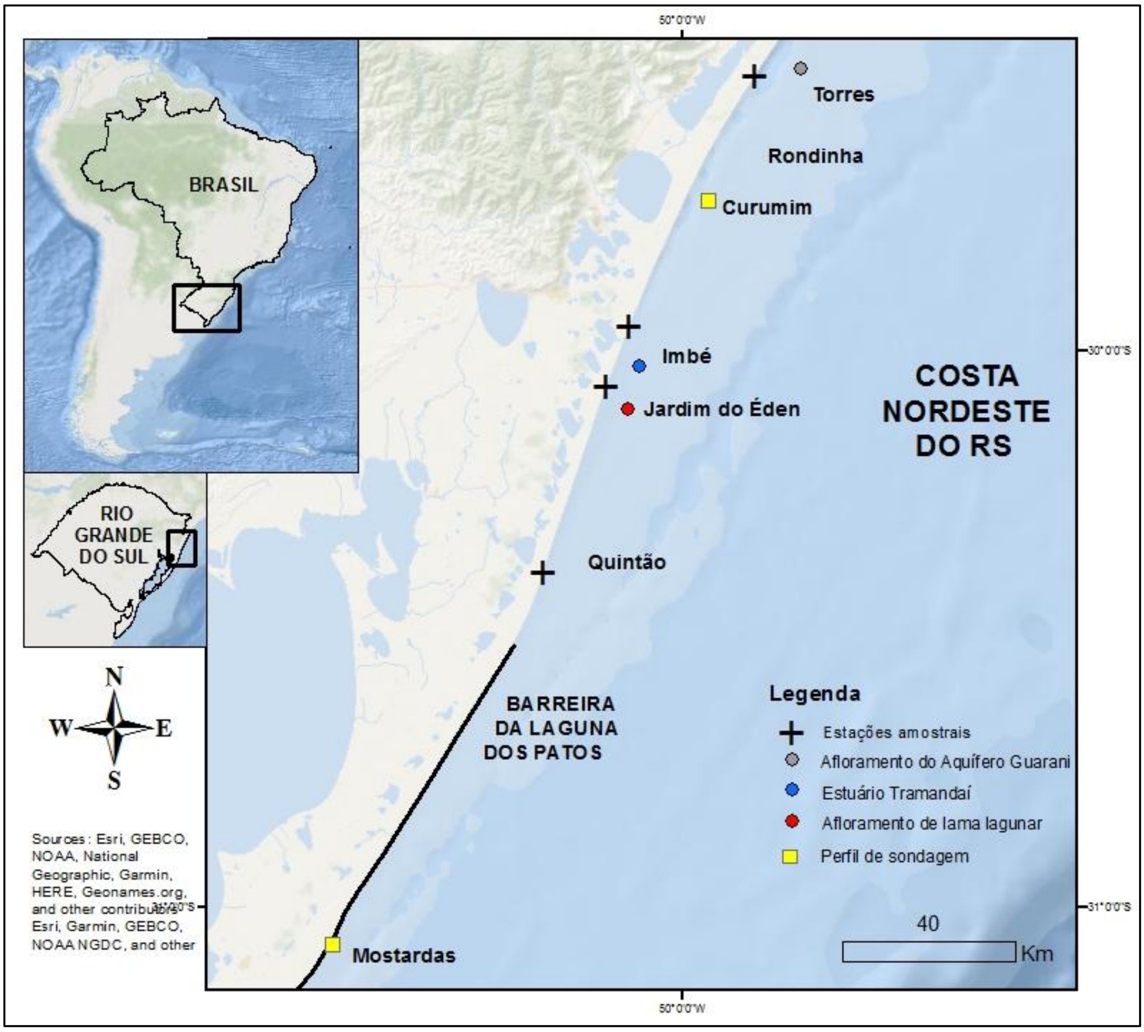

Figura 1 - Área de estudo na CNRS, sul do Brasil, com cinco estações de amostragem e destaque para as características geológicas.

Além disso, esta área pode estar submetida ao mesmo tempo sob a influência da Barreira da Laguna dos Patos (BLP) e do afloramento do Aquífero Guarani (AAG), uma vez que está localizada entre essas feições, conforme observado na Figura 1. Da mesma forma, outros elementos interessantes (arquitetônicos, exposições, registros históricos) estão localizados nesta área, como a foz do Estuário de Tramandaí e um afloramento do terraço lagunar (Dillenburg et al., 2004a; Tomazelli et al., 1998).

Foram definidas cinco estações de estudo, que correspondem as praias de Torres, Rondinha e Imbé (área progradacional) e Jardim do Éden e Quintão (área retrogradacional). Torres situa-se na área de influência do AAG, Imbé localiza-se na foz do Estuário de Tramandaí e a estação Jardim do Éden apresenta afloramentos lamacentos na faixa de praia. A amostragem foi desenvolvida no verão austral de 2017, onde a água subterrânea foi amostrada a partir de poços de praia, aplicando um sistema *pushpoint*, em transectos perpendiculares a linha de praia, avaliando dessa forma o gradiente subterrâneo de salinidade.

Amostragem e análise

A influência das águas subterrâneas nos sistemas marinhos pode ser caracterizada usando a salinidade da faixa de praia, reconhecido proxy para área de mistura. Todos os poços de amostragem foram perfurados a 1 m de profundidade ao longo de um transecto compreendido desde a duna frontal até a zona de varrido, de forma a estabelecer um gradiente de salinidade, partindo do *end member* de água doce indo até o *end member* água marinha costeira. Salinidade, condutividade e pH foram determinados *in loco*. Para a coleta de elementos traço foram usados frascos de 50 mL, as amostras foram filtradas (0,45 µm) e preservadas (HCl destilado 0,024 M), segundo metodologia padrão do Programa GEOTRACES, descrita por Cutter (2014).

Os elementos traço determinados foram: manganês (Mn), estrôncio (Sr), alumínio (Al), vanádio (V), molibdênio (Mo), ferro (Fe), níquel (Ni), cobre (Cu) e chumbo (Pb). As análises destes elementos foram feitas no *Skidaway Institute of Oceanography*/UGA usando um equipamento ICP-MS Perkin Elmer Nexion quadripolo 300d. Não foram necessários tratamentos prévios das amostras, sendo apenas diluídas 100 vezes para as leituras de Mn e 10 vezes para a leitura dos demais elementos. O instrumento foi executado usando amônia (NH_3) como gás de reação para reduzir as interferências poliatômicas e o índio (In) foi usado como um padrão interno para monitorar a deriva do instrumento. Dois materiais de referência padrão (NASS 6 e NIST 1643f) foram analisados (dados nos ANEXOS 3 A e 3B) com cada lote de amostras para determinar a exatidão e a precisão da análise. As recuperações de todos os elementos estiveram dentro de 98 % do valor de referência. O método analítico foi calibrado usando padrões externos de elementos mistos.

RESULTADOS

A Tabela 1 apresenta as características dos poços amostrados como também as faixas de pH e salinidade que expressam melhor as variações do gradiente salino em cada região considerada.

Tabela 1 - Dados dos poços da praia em cinco estações na CNRS e faixas de pH e salinidade.

Estação	Latitude Longitude	Comprimento de praia (m)	N° de poços amostrados	pH	Salinidade
Torres	29°21’33,3’’ 49°44’11,5’’	33	05	7,85-8,17	0,97-12,61
Rondinha	29°03’03,6’’ 49°50’52,4’’	50	06	7,15-8,15	0,06-30,71
Imbé	29°57’19,0’’ 50°06’36,3’’	54	08	7,63-7,87	6,22-31,22
Jardim do Éden	30°03’51,5’’ 50°09’26,0’’	41	07	6,44-7,28	2,65-24,55
Quintão	30°23’59,9’’ 50°17’18,1’’	45	07	7,61-8,00	5,38-26,26

A distribuição espacial de salinidade apresentou variações significativas entre os transectos, não sendo possível obter gradientes completos (de 0 a 35 de salinidade). A correlação entre salinidade e distância da zona de varrido foi elevada (r²> 0,90), exceto para a estação Jardim do Éden (r²<0,58), que apresentou um poço, cerca de 20 m da zona de varrido, com um valor distinto de qualquer tendência, que é reflexo da geologia local e que será explicado na sequencia deste trabalho.

As concentrações dos elementos traço para os respectivos transectos são apresentados na Figura 2 e 3. Destaque para Jardim do Éden e Quintão que apresentaram concentrações muitas vezes superiores aos demais transectos.

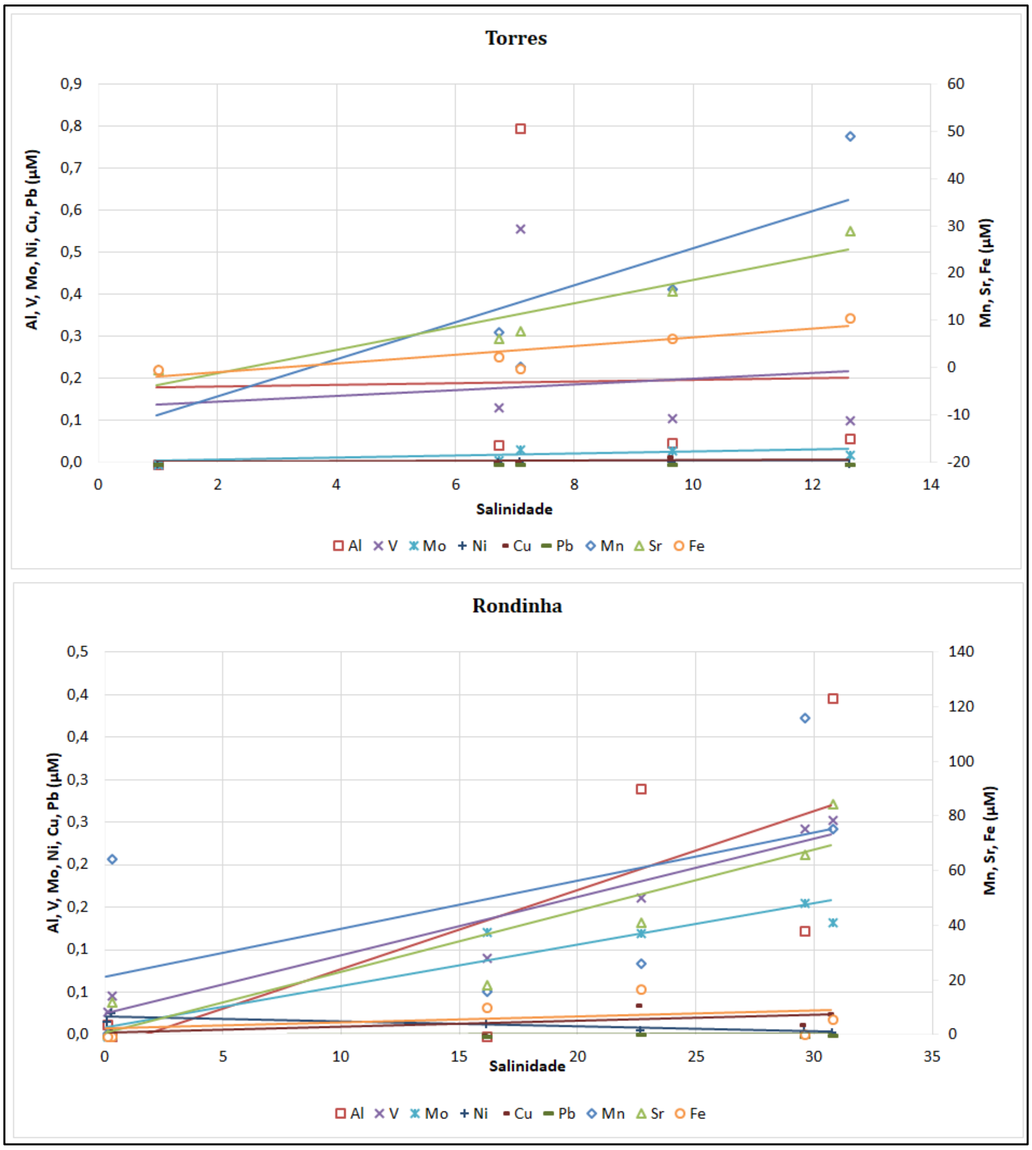

Figura 2 – Concentrações de elementos traço (Mn, Sr, Al, V, Mo, Fe, Ni, Cu e Pb) plotados no gradiente salino nas estações Torres e Rondinha, com suas respectivas linhas de tendência.

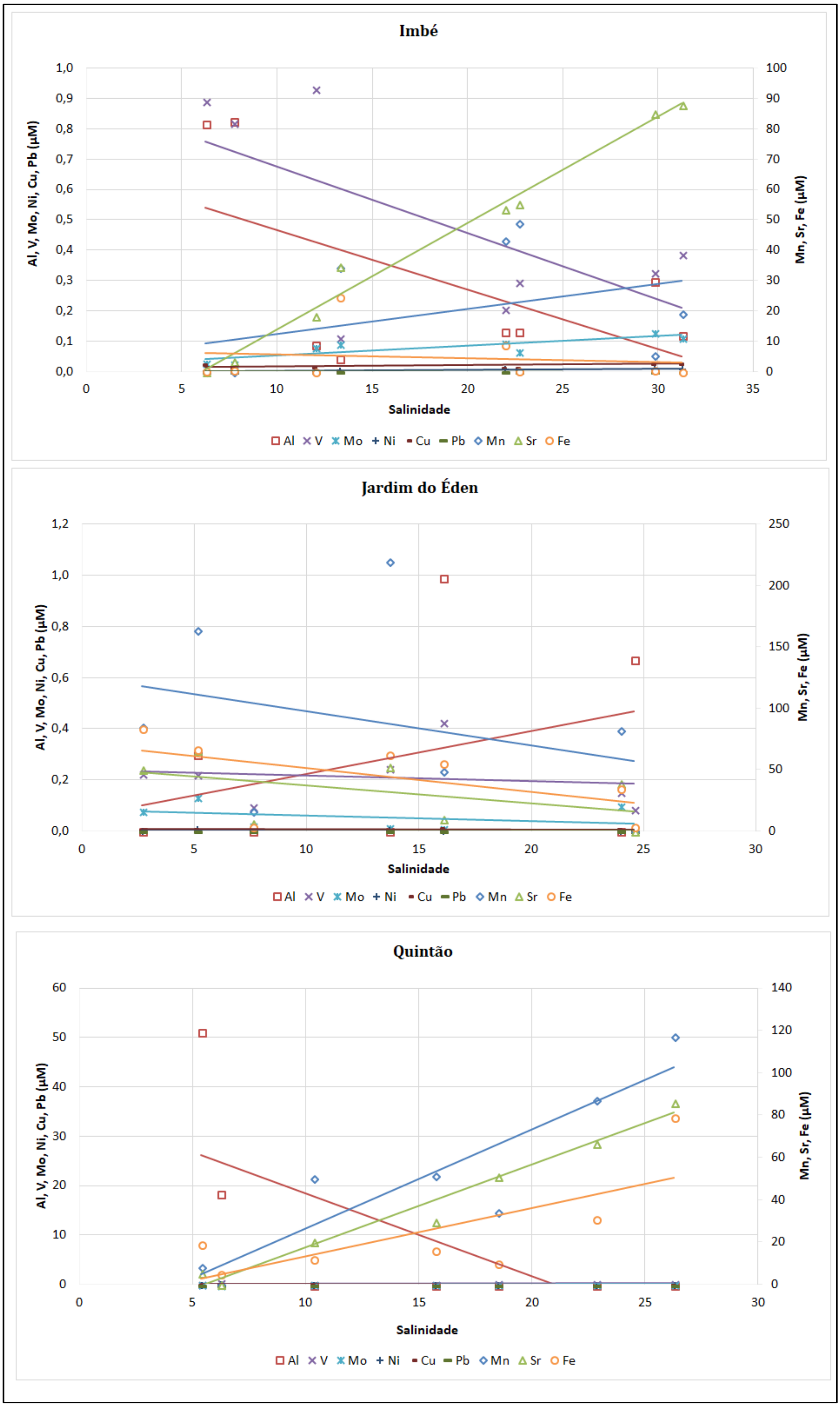

Figura 3 – Concentrações de elementos traço (Mn, Sr, Al, V, Mo, Fe, Ni, Cu e Pb) plotados no gradiente salino nas estações Imbé, Jardim do Éden e Quintão, com suas respectivas linhas de tendência.

A distribuição dos elementos traço mostrou tendências variáveis em relação à salinidade, dos quais Sr apresentou aporte marinho conservativo, exceto na estação Jardim do Éden, onde a maioria dos elementos apresentou aporte continental.

Com base nas concentrações obtidas neste estudo e do fluxo de águas oriundos do compartimento subterrâneo estimado por Rocha et al. (artigo 2) foi possível estimar o fluxo de elementos traço para a plataforma costeira (Tabela 2). Como tratam-se de regiões específicas, sob influência de fatores geológicos de ação limitada espacialmente, foram tomadas a DAS e os fluxos dos elementos para comprimentos de costa de 1000 m.

Tabela 2 – Fluxos de elementos traço por transecto de praia amostrado na CNRS, verão austral 2017.

Estação		Torres	Rondinha	Imbé	Jardim do Éden [b]	Quintão
DAS [a] (10^3 m³/d)		12.5±1.5	51.9±3.2	83.9±3.8	0.8±0.3	5.6±0.4
Fluxos (mol/d) [a]	Mn	100.6±11.64	2386±148.0	1037±47.12	24.79±8.26	281.2±17.64
	Sr	101.3±11.72	1572±97.62	3704±168.3	6.37±2.12	167.0±10.47
	Al	0.64±0.07	3.59±0.22	11.1±0.5	0.26±0.09	ND
	V	1.36±0.16	6.64±0.41	29.99±1.36	0.13±0.04	1.04±0.07
	Mo	0.27±0.03	6.34±0.39	7.17±0.33	ND	0.31±0.02
	Fe	33.7±3.9	188.3±11.69	41.39±1.88	22.36±7.45	90.12±5.65
	Ni	ND	0.53±0.03	0.39±0.02	ND	ND
	Cu	ND	0.54±0.03	1.4±0.06	ND	0.17±0.01
	Pb	ND	ND	ND	ND	ND

[a] Estimados para comprimentos de costa de 1000 m; [b] compartimento B.

DISCUSSÃO

Salinidade e pH

A faixa de salinidade variável verificada para os transectos já evidencia o diferenciado processo de mistura, havendo casos em que a água doce tem mais influência e outros em que a água do mar predomina.

O transecto de Torres apresentou domínio de águas doces de forma significativa, tendo a salinidade alcançado apenas 12,61 de valor máximo ao longo das amostras coletados. o que evidencia o efeito do Aquífero Guarani que aflora naquela região, exportando sob a forma de DAS, elevados volumes de água doce do continente para o mar (Paiva, 2014).

Para os transectos de Jardim do Éden e Quintão a salinidade máxima ficou próxima de 25. Para estes casos, deve ser considerado a influência de chuvas e de todo o consequente escoamento superficial. Para o caso do transecto de Imbé, deve ser considerado, também, a existência de paleocanais que potencializam o aporte de água doce. Deve ser mencionado que, em nenhum dos locais amostrados, foi alcançado o valor da salinidade marinha, o que é uma clara indicação de quão importante é a DAS nesta região do litoral sul do Brasil.

A avaliação dos resultados de pH também indica diferenças entre as estações amostrais, sendo por exemplo a estação de Torres a que apresenta faixa de pH mais elevada, em resposta a composição ígnea do Aquífero Serra Geral que ali se apresenta (Alloway, 2013). Já a estação de Imbé, influenciada pelo estuário superficial da Lagoa de Tramandaí/Armazém, mostra uma faixa de pH restrita. A estação de Jardim do Éden é a mais discordante, por sua faixa de pH estar bem abaixo das demais, devido a composição sedimentar da faixa de praia, que apresenta afloramento de lama proveniente do terraço paleolagunar do sistema laguna/barreira IV, em consequência do processo transgressivo observado na região (Dillenburg et al., 2004a; Tomazelli et al., 1998). Isto é também a explicação para ter sido encontrado um ponto fora de qualquer tendência, mas que não pode ser considerado como um outlier, pois indica um perfil dividido em dois compartimentos isolados entre si por uma unidade confinante, ou seja, um ponto isolante que neste caso faz com que haja 2 gradientes independentes, como ilustrada pela Figura 4.

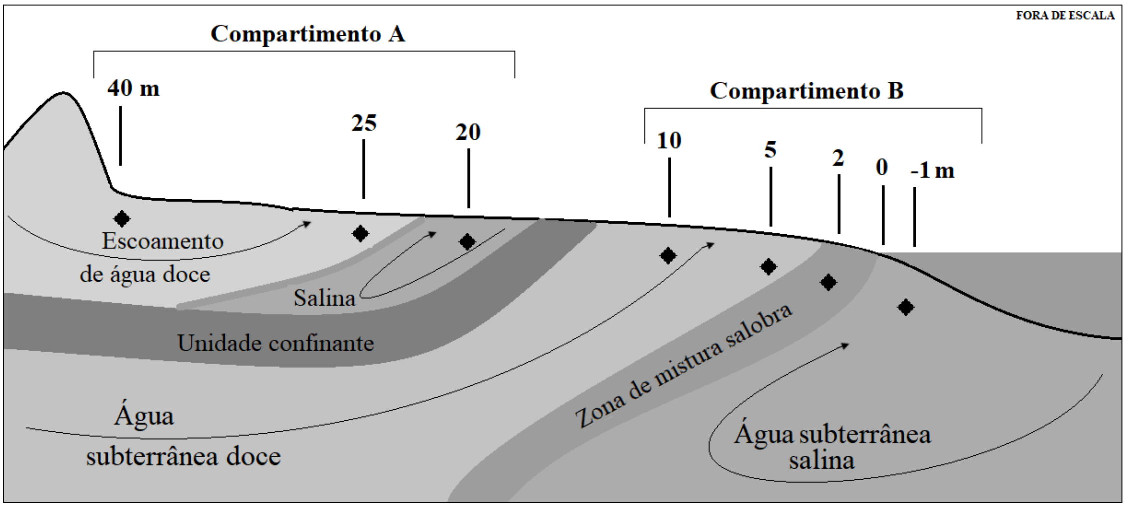

Figura 4 - Diagrama esquemático da DAS na estação Jardim do Éden, ilustrando o efeito da unidade de confinamento (isolamento) em poços de praia (♦ 1m de profundidade) estabelecendo dois compartimentos.

Por fim, a estação Rondinha é a que apresentou faixa de salinidade e pH mais amplas, por ser a região que não apresenta feições ou descontinuidades geológicas.

Como cada uma das áreas consideradas possui características que lhe são particulares, estas devem atuar como forçantes para alterar ou diferenciar o comportamento e fluxo de elementos traços nestes transectos.

Elementos traço

A distribuição dos elementos traço no gradiente salino foi bastante varável entre as diferentes estações de coleta, bem como entre os próprios elementos, de forma que foram evidenciados casos de aporte do continente para o mar e do mar para o continente, o que se denomina de intrusão salina.

Nas concentrações de Mo, Sr, Mn e Fe observadas nos transectos de praia foram destacados os resultados na estação Jardim do Éden, com comportamento diferenciado de aporte continental. Enquanto outras quatro estações apresentaram aumento das concentrações de elementos traço com o aumento da salinidade, ou seja, ocorrendo um aporte marinho, as concentrações em Jardim do Éden foram iguais ou até mesmo decrescentes. Além disso, as concentrações médias de Mn e Fe foram maiores no Jardim do Éden (88,3 e 44,5 µM, respectivamente) em comparação com outras estações (média de 33,6 e 9,8 µM). Possivelmente, a matéria orgânica dos sedimentos lamosos permite uma remineralização mais intensa desses elementos (Reckhardt et al., 2017).

Nas concentrações de Al, Cu e Pb, o destaque foi para os resultados da estação do Quintão, que ora comportaram-se como aporte continental, ora marinho. As concentrações na estação Quintão também foram maiores para os elementos Al (10,0 µM), Cu (26,8 nM) e Pb (1,1 nM), do que nas demais estações (médias de 0,2 µM Al; 11,4 µM Cu e 0,4 µM Pb).

A distribuição de V apresentou comportamento distinto, e o destaque é a alta concentração na estação de Imbé (0,5 µM) comparado com as demais estações (média 0,2 µM). A distribuição de Ni bastante baixa em comparação aos demais elementos, sendo as maiores concentrações verificadas na estação de Rondinha (11,9 nM) enquanto as demais estações tiveram média de 3,9 nM, apresentando correlações ora positivas, ora negativas com gradientes de salinidade.

Os elementos Pb, Cu e Ni apresentaram comportamento estável (sem incremento ou decréscimo nas concentrações) entre as amostras nos diferentes piezômetros. Já os demais elementos traço quantificados apresentaram variações entre comportamento de aporte continental e aporte marinho, bem como entre os transectos, não havendo padrão na distribuição, embora as concentrações médias dos elementos traço sejam 2,5 vezes maiores na área que apresenta processo costeiro de evolução retrogradacional (Imbé, Jardim do Éden e Quintão). Mesmo assim, não há correlação das concentrações de elementos traço com o gradiente de salinidade, como observado em trabalho pretérito na BLP (Niencheski and Windom, 2015).

Muitas destas variações são justificadas pelas características de pH e salinidade, que consequentemente governam efeitos oxi-redox, dessorção, liberação, entre outros processos geoquímicos. Assim, com base nos resultados é possível perceber a não uniformidade no comportamento dos elementos traço, produto da influência das características geológicas intrínsecas a cada um dos transectos.

Este tipo de interferência geológica não havia sido abordado anteriormente e, responde negativamente a ideia de homogeneidade da costa do sul do Brasil. As 3 regiões que se estabelecem – BLP, CNRS e AAG são diferentes em termos de hidroquímica local. A diversidade de composição da barreira é mais expressiva na região norte, onde há maior número de compartimentos (aquíferos) que contribuem para a riqueza geoquímica das águas. Proporcionalmente, a BLP apresenta características menos diversificadas geologicamente e, como mencionado, menos rica em elementos traço, por exemplo. No geral, a BLP apresenta um enorme gradiente hidráulico e, portanto, intensa DAS (Windom et al., 2006). Já a CNRS apresenta forças motrizes diferentes, assim como a hidrogeologia e os reservatórios, que resultam em aporte mais marinho do que continental, ainda que o percentual de água doce subterrânea (Q_{fw}) seja maior na composição da DAS (artigo 2). Assim, a BLP, CNRS e AAG mostram-se relevantes cada um com sua especificidade, visto que fluxos menores podem ser compensados por concentrações de nutrientes e elementos traço maiores, como é o caso da CNRS, conferindo a cada uma delas papel importante nos processos costeiros.

O agrupamento de alguns elementos traço por seu comportamento, observado por Niencheski & Windom (2015), não é percebido neste trabalho. Inclusive, a perspectiva de que a litologia poderia responder pela diferença entre as concentrações observadas nos 5 transectos analisados não foi confirmada. A distribuição é bastante variada, influenciada pelas características geológicas já apontadas, mas sem um gradiente definido entre elas.

Entretanto, percebe-se a influência da litologia quando se comparam tais resultados da CNRS com concentrações obtidas na BLP, na região sob influência do AAG e com dados obtidos em outras regiões costeiras espalhadas pelo mundo, como indicado na Tabela 3.

Tabela 3 - Compilação de elementos traço medidos no sul do Brasil e outras áreas de estudo.

Área de estudo deste trabalho	Salinidade	Elementos traço								
		Mo (nM)	Sr (µM)	Mn (µM)	Fe (µM)	Al (µM)	Cu (nM)	Pb (nM)	V (nM)	Ni (nM)
Progradacional [a]	12.4	57.9	26.0	34.3	5.1	0.2	9.7	0.4	158	7.5
Retrogradacional [a]	15.6	61.7	37.5	50.9	23.7	3.4	18.0	0.6	311	4.4
Barreira da Laguna dos Patos [b c d]	11.1	41.1		4.3	27.8		1.1		10.6	
	22.9	45.2		4.4	24.1		2.3		9.5	
				10.0 [i]	24.0		1.0 [i]			
		1.0 [i]		10.0 [i]	24.0		1.0 [i]		10.0 [i]	10.0 [i]
Afloramento do Aquífero Guarani [e]					91.7		38.3	11.1		
Long Island, NY [f]	0.0			0.01		0.08	1.9	0.01		2.7
Costa espanhola [g]					0.24		25	0.5		15
Costa alemã do Mar do Norte [h]	31	101		0.03	0				21	

[a] Este estudo, médias do gradiente salino; [b] Windom et al. (2007); [c] Windom et al. (2006); [d] Niencheski et al. (2015); [e] Paiva (2011); [f] Beck et al. (2007); [g] Trezzi et al. (2016); [h] Reckhardt et al. (2017); [i] valores aproximados baseados em dados gráficos.

Para os elementos Mo, Sr, Mn, Al e V, os maiores resultados são registrados na área de estudo amostrada neste artigo. Fe, assim como Cu e Pb, apresentaram maiores concentrações na área do AAG, enquanto para a BLP, houve destaque apenas as concentrações de Ni.

A diferença nas concentrações dos elementos traço entre os transectos deste trabalho e da BLP se justificam pelos diferentes compartimentos subterrâneos que os formam, sendo a CNRS composta por três compartimentos - Quaternário/Serra Geral/Botucatu, enquanto a BLP apresenta 2 compartimentos - Quaternário/Mioceno. Esses dados referem-se a perfis de sondagem em Curumim (Machado and Freitas, 2005a) e Mostardas (ANP, 1964) até 410 m de profundidade. O Aquífero Serra Geral é uma formação ígnea (máfica) que se apresenta elementos traço (Mo, Mn, Cu, Pb, V e Ni), aproximadamente, 7 vezes mais concentrados que os aquíferos de arenitos (Alloway, 2013), além de ser enriquecido em Fe (Reginato, 1996). Assim, este maior número de compartimentos garante mais diversa contribuição geológica de elementos para as águas subterrâneas na CNRS, bem como mais concentradas quando comparadas a PLB.

Além disso, as elevadas concentrações destacadas neste trabalho para vários elementos analisados, quando comparadas a outras regiões, evidenciam a relevância dessa região como fonte de elementos traços para o oceano costeiro adjacente e, ainda, preenchem a lacuna de dados de elementos traços havia até então entre a região da BLP (mais ao sul) e o AAG (mais ao norte).

É importante considerar que, os dados obtidos na BLP não são oriundos de um gradiente salino, como o produzido neste estudo, e sim de poços pontuais localizados na barreira costeira. Desta forma, o *end member* (salinidade zero) das amostras deste estudo tendem a ser ainda mais elevados, o que traz um destaque ainda maior da região considerada neste estudo sobre a BLP.

Fazendo uso das concentrações médias obtidas e da DAS, estudo complementar a este deve ser feito a fim de extrapolar a estimativa de fluxo de elementos traço para toda a região da CNRS, como foi realizado por Niencheski et al. (2007) para os nutrientes, na BLP. Entretanto, sabendo da especificidade geológica desta região considerada (Vide artigo 2) e da significativa variação observada neste estudo entre os 5 transectos, entende-se não ser válida a extrapolação destes dados para toda a CNRS (150 km), como forma de validar fluxos totais. Baseados em nossos conhecimentos atuais, consideramos como estimativa segura a aplicação em um comprimento de costa de 1000 m para cada um dos transectos, e a medida que os estudos avançarem nesta importante região, poderão ser aplicados cálculos para regiões cada vez mais amplas.

CONCLUSÃO

Este estudo evidencia que feições e descontinuidades geológicas de menores proporções espaciais tem efeitos sobre a distribuição, transporte, mistura e fluxos de elementos traço na região da CNRS.

A avaliação dos cinco transectos com condições geológicas diferenciadas mostraram não haver um padrão de aporte continental ou marinho definido, e embora similares em algumas características, apresentam variação de pH, do gradiente de salinidade, bem como da faixa de concentração dos elementos traço. Destaca-se o observado enriquecimento na região do Jardim do Éden, transecto este que é influenciado pelo afloramento de lama lagunar.

Este trabalho possui um grande valor por indicar de forma pioneira que a região da CNRS apresenta concentração de elementos traço elevadas, superiores as regiões próximas, como a BLP e a região do AAG, ambas essas regiões consideradas como referência em termos de fluxo de elementos químicos para plataforma costeira adjacente.

Os resultados deste trabalho lançam dados iniciais para o entendimento do fluxo de elementos traço e efeitos geológicos de descontinuidade, mas são necessários maiores estudos para melhor compreender a (re)mobilização e transporte dos elementos traço em vista das características da barreira costeira, hidroquímica local e ocupação humana, bem como das áreas entre transectos.

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AGRADECIMENTOS

Os autores gostariam de agradecer o excelente apoio do Skidaway Institute of Oceanography (UGA/USA) e da equipe do CECLIMAR/Universidade Federal do Rio Grande do Sul (UFRGS) que garantiram que este trabalho fosse realizado com sucesso. Bolsista da Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) que concedeu apoio financeiro a esta pesquisa, e ao Conselho Nacional de Desenvolvimento Científico e Tecnológico (INCT Mar-COI 565062/2010-7) e ao Conselho Nacional de Desenvolvimento Científico e Tecnológico. (CNPq 303672/2013-7, 302822/2017-8 e 302822/2017-8).

ANEXOS

ANEXO 1A

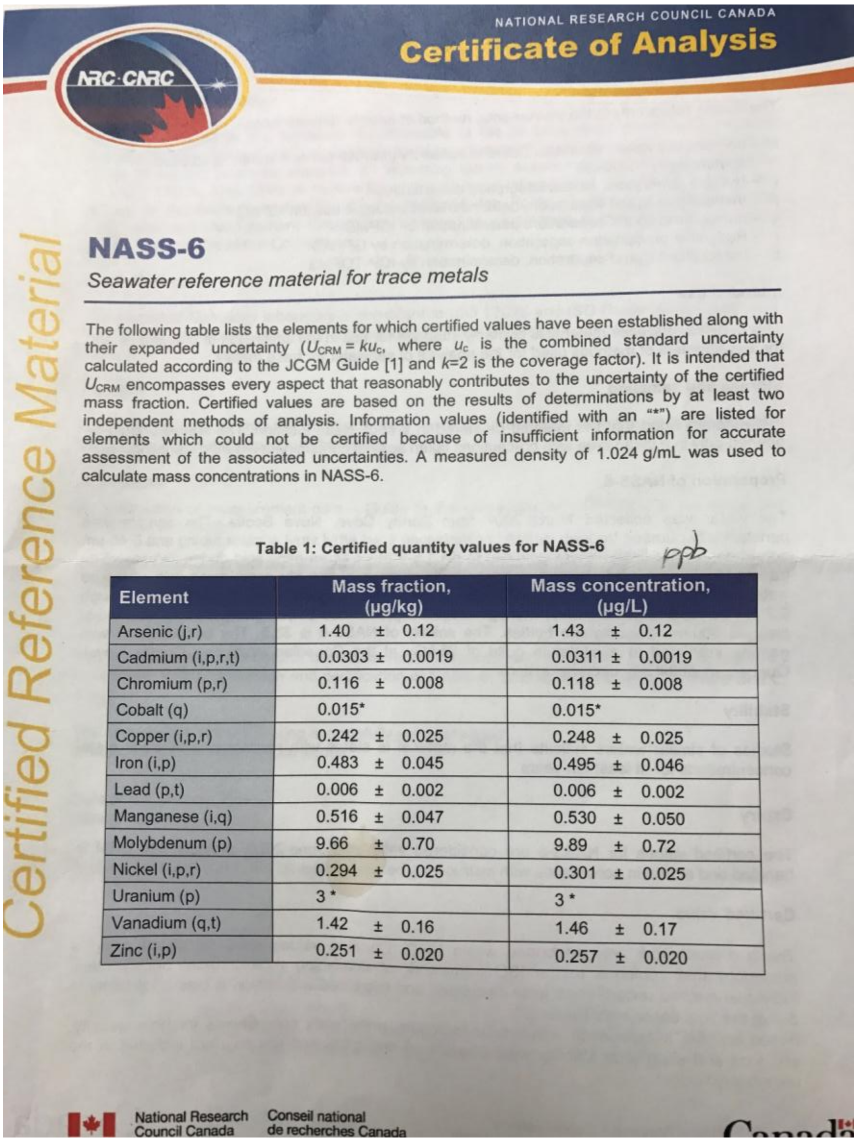
ANEXO 1B

ANEXO 2A

ANEXO 2B

ANEXO 3A

Material de Referência NASS6 (fotocópia – Skidaway Institute of Oceanography).



NATIONAL RESEARCH COUNCIL CANADA

Certificate of Analysis

Certified Reference Material

NASS-6

Seawater reference material for trace metals

The following table lists the elements for which certified values have been established along with their expanded uncertainty ($U_{CRM} = ku_c$, where u_c is the combined standard uncertainty calculated according to the JCGM Guide [1] and k=2 is the coverage factor). It is intended that U_{CRM} encompasses every aspect that reasonably contributes to the uncertainty of the certified mass fraction. Certified values are based on the results of determinations by at least two independent methods of analysis. Information values (identified with an "*") are listed for elements which could not be certified because of insufficient information for accurate assessment of the associated uncertainties. A measured density of 1.024 g/mL was used to calculate mass concentrations in NASS-6.

Table 1: Certified quantity values for NASS-6 ppb

Element	Mass fraction, (µg/kg)	Mass concentration, (µg/L)
Arsenic (j,r)	1.40 ± 0.12	1.43 ± 0.12
Cadmium (i,p,r,t)	0.0303 ± 0.0019	0.0311 ± 0.0019
Chromium (p,r)	0.116 ± 0.008	0.118 ± 0.008
Cobalt (q)	0.015*	0.015*
Copper (i,p,r)	0.242 ± 0.025	0.248 ± 0.025
Iron (i,p)	0.483 ± 0.045	0.495 ± 0.046
Lead (p,t)	0.006 ± 0.002	0.006 ± 0.002
Manganese (i,q)	0.516 ± 0.047	0.530 ± 0.050
Molybdenum (p)	9.66 ± 0.70	9.89 ± 0.72
Nickel (i,p,r)	0.294 ± 0.025	0.301 ± 0.025
Uranium (p)	3 *	3 *
Vanadium (q,t)	1.42 ± 0.16	1.46 ± 0.17
Zinc (i,p)	0.251 ± 0.020	0.257 ± 0.020

National Research Council Canada   Conseil national de recherches Canada

ANEXO 3B

Material de Referência NIST1643f.

National Institute of Standards & Technology

Certificate of Analysis

Standard Reference Material® 1643f

Trace Elements in Water

This Standard Reference Material (SRM) is intended primarily for use in evaluating methods used in the determination of trace elements in fresh water. A unit of SRM 1643f consists of approximately 250 mL of acidified water in a polyethylene bottle, which is sealed in an aluminized plastic bag to maintain stability. SRM 1643f simulates the elemental composition of fresh water. The solution contains nitric acid at a volume fraction of approximately 2 %, equivalent to an amount of substance concentration (molarity) of approximately 0.32 mol/L.

Certified Values: The certified values for elements in SRM 1643f are listed in Table 1. All values are reported both as mass fractions (µg/kg) and as mass concentrations (µg/L). A NIST certified value is a value for which NIST has the highest confidence in its accuracy in that all known or suspected sources of bias have been investigated or taken into account [1]. The certified mass fraction values are consensus estimates that blend the results of the gravimetric preparation value and a value determined by either inductively coupled plasma mass spectrometry (ICP-MS) or inductively coupled plasma optical emission spectrometry (ICP-OES) [2]. The certified mass concentration values are derived from the certified mass fraction values using the measured density of SRM 1643f. Additional information about the certification of SRM 1643f is given under "Certification of Material".

The expanded uncertainty for each certified value is calculated as

$$U = ku_c$$

where k is the coverage factor for a 95 % confidence interval and u_c is the combined standard uncertainty calculated through the application of the Monte Carlo method described in the ISO/JCGM Supplement 1 [3]. The value of u_c for the certified mass fraction values is intended to represent, at the level of one standard deviation, the combined effect of uncertainty components associated with the gravimetric preparation, the ICP-MS or ICP-OES determination, method bias, and stability. Additionally, the uncertainty evaluations associated with the certified mass concentration values assume that the temperature at which the material will be measured is between 15 °C and 25 °C.

Expiration of Certification: This certification of **SRM 1643f** is valid, within the measurement uncertainty specified, until **31 October 2023**, provided the SRM is handled and stored in accordance with instructions given in this certificate (see "Instructions for Use"). This certification is nullified if the SRM is damaged, contaminated, or modified.

Maintenance of SRM Certification: NIST will monitor this SRM over the period of its certification. If substantive technical changes occur that affect the certification before the expiration of this certificate, NIST will notify the purchaser. Registration (see attached sheet or register online) will facilitate notification.

Coordination of the NIST technical measurements was under the direction of T.A. Butler, J.L. Molloy and M.R. Winchester of the NIST Chemical Sciences Division. The density, ICP-MS and ICP-OES analyses were performed by T.A. Butler and J.L. Molloy.

Statistical analysis of the experimental data was performed by A.M. Possolo of the NIST Statistical Engineering Division.

Support aspects involved in the issuance of this SRM were coordinated through the NIST Office of Reference Materials.

Carlos A. Gonzalez, Chief
Chemical Sciences Division

Gaithersburg, MD 20899
Certificate Issue Date: 18 August 2015

Robert L. Watters, Jr., Director
Office of Reference Materials

Table 1. Certified Values, Expanded Uncertainties, and Coverage Factors (k) for Elements in SRM 1643f

Element	Mass Fraction (µg/kg)			k	Mass Concentration (µg/L)			k
Aluminum (Al)	132.5	±	1.2	1.9	133.8	±	1.2	1.9
Antimony (Sb)	54.90	±	0.39	1.9	55.45	±	0.40	2.0
Arsenic (As)	56.85	±	0.37	2.0	57.42	±	0.38	2.0
Barium (Ba)	513.1	±	7.3	2.1	518.2	±	7.3	2.1
Beryllium (Be)	13.53	±	0.11	2.1	13.67	±	0.12	2.1
Bismuth (Bi)	12.50	±	0.10	1.9	12.62	±	0.11	1.9
Boron (B)	150.8	±	6.6	2.2	152.3	±	6.6	2.2
Cadmium (Cd)	5.83	±	0.13	2.2	5.89	±	0.13	2.2
Calcium (Ca)	29 140	±	320	2.1	29 430	±	330	2.1
Chromium (Cr)	18.32	±	0.10	2.0	18.50	±	0.10	2.1
Cobalt (Co)	25.05	±	0.17	2.0	25.30	±	0.17	2.0
Copper (Cu)	21.44	±	0.70	2.1	21.66	±	0.71	2.2
Iron (Fe)	92.51	±	0.77	2.1	93.44	±	0.78	2.1
Lead (Pb)	18.303	±	0.081	2.0	18.488	±	0.084	2.1
Lithium (Li)	16.42	±	0.35	2.2	16.59	±	0.35	2.2
Magnesium (Mg)	7 380	±	58	1.9	7 454	±	60	2.0
Manganese (Mn)	36.77	±	0.58	2.1	37.14	±	0.60	2.2
Molybdenum (Mo)	114.2	±	1.7	2.1	115.3	±	1.7	2.1
Nickel (Ni)	59.2	±	1.4	2.2	59.8	±	1.4	2.2
Potassium (K)	1 913.3	±	9.0	2.0	1 932.6	±	9.4	2.1
Rubidium (Rb)	12.51	±	0.12	2.0	12.64	±	0.13	2.0
Selenium (Se)	11.583	±	0.078	2.0	11.700	±	0.081	2.0
Silver (Ag)	0.9606	±	0.0053	2.0	0.9703	±	0.0055	2.0
Sodium (Na)	18 640	±	240	2.1	18 830	±	250	2.1
Strontium (Sr)	311	±	18	2.1	314	±	19	2.2
Tellurium (Te)	0.9672	±	0.0082	2.0	0.9770	±	0.0084	2.0
Thallium (Tl)	6.823	±	0.034	1.9	6.892	±	0.035	2.0
Vanadium (V)	35.71	±	0.27	2.0	36.07	±	0.28	2.0
Zinc (Zn)	73.7	±	1.7	2.1	74.4	±	1.7	2.1

[a] The measurand is the total mass fraction for each element. Metrological traceability is to the SI unit for mass, expressed as micrograms per kilogram and micrograms per liter.

Preparation of Material: SRM 1643f was prepared at NIST using only high purity reagents. A polyethylene cylindrical tank was filled with deionized water and sufficient nitric acid to bring the nitric acid amount of substance concentration (molarity) to approximately 0.32 mol/L. Known masses of the matrix elements (sodium, potassium, calcium, and magnesium) were added to the tank as solutions prepared from the same materials used to prepare the SRM 3100 series of single element solutions. Known masses of the other elements were then added to the tank solution using weighed aliquots of the SRM 3100 series. After mixing thoroughly, the tank solution was transferred into the acid-cleaned, 250 mL, polyethylene, SRM bottles and immediately sealed in individual aluminized plastic bags.

Certification of Material: Each of the certified elements was determined using either ICP-MS or ICP-OES. The final total mass of the tank solution prior to bottling was determined from the sum of the mass fraction values of the elements in Table 1 and the sum of the known masses of those elements added to prepare the solution, therefore allowing calculation of the gravimetric preparation mass fraction for each element. Certified mass fraction values were calculated by combining the gravimetric preparation values with the ICP-MS or ICP-OES values, as described under Certified Values. Certified mass concentration values were calculated using the measured density of 1.0101 g/mL ± 0.0012 g/mL, where the uncertainty is expressed at a confidence level of approximately 95 %, within the temperature range of 15 °C to 25 °C.

INSTRUCTIONS FOR USE

Precautions: The SRM should be shaken before use because of possible water condensation on the inner surfaces of the bottle. To prevent possible contamination of the SRM, **DO NOT** insert pipettes into the bottle. Samples should be decanted at a room temperature of 15 °C to 25 °C. After use, the bottle should be recapped tightly and returned to the aluminized plastic bag, which should be folded and sealed with sealing tape. This safeguard will protect the SRM from possible environmental contamination and long-term evaporation.

The accuracy of trace element determinations, especially at the micrograms per liter level, is limited by contamination. Apparatus should be scrupulously cleaned and only high purity reagents employed. Sampling and manipulations, such as evaporations, should be done in a clean environment, such as a Class-100 clean hood.

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[2] DerSimonian, R.; Laird, N.; *Meta-Analysis in Clinical Trials*; Control. Clin. Trials, Vol. 7, pp. 177–188 (1986).

[3] JCGM 101:2008; *Evaluation of Measurement Data — Supplement 1 to the "Guide to the Expression of Uncertainty in Measurement" — Propagation of Distributions using a Monte Carlo Method*; Joint Committee for Guides in Metrology (JCGM) (2008); available at http://www.bipm.org/utils/common/documents/jcgm/JCGM_101_2008_E.pdf (accessed Aug 2015).

Users of this SRM should ensure that the Certificate of Analysis in their possession is current. This can be accomplished by contacting the SRM Program: telephone (301) 975-2200; fax (301) 948-3730; e-mail srminfo@nist.gov; or via the Internet at http://www.nist.gov/srm.

CONSIDERAÇÕES E RECOMENDAÇÕES

A aplicação do isótopo radioativo ^{222}Rn como geotraçador para águas subterrâneas combinado com a técnica de imageamento de subsuperfície com GPR é inovador e inédita e, gerou resultados que permitiram avançar substancialmente no conhecimento das DAS na CNRS.

A partir das imagens produzidas nos diferentes transectos da CNRS ficam reforçadas as diferenças na arquitetura deposicional das regiões tanto progradacional como retrogradacional, oportunizando uma avaliação vinculada à tais características e seus efeitos sobre a DAS. Foram elencados elementos estruturais que caracterizam cada um dos tipos de barreira costeira, sendo possível correlacioná-los com a atividade do traçador ^{222}Rn e compreender os diferentes acamadamentos geológicos que compõem cada região.

Este estudo quantificou a DAS em 14 diferentes pontos ao longo de, aproximadamente 150 km, preenchendo uma importante lacuna do conhecimento, demonstrando ser esta região responsável pela descarga de volumes expressivos de águas subterrâneas enriquecidas em micro e macronutrientes, o que esclarece a ocorrência de frequentes florações de algas, ainda que distantes das desembocaduras de rios e estuários.

Além disso, comprovou e quantificou o fluxo de elementos traço e suas variações entre transectos com características geológicas distintas, reforçando a evidência de que a geologia é uma importante forçante, embora muitos trabalhos a desconsiderem.

As heterogeneidades da barreira na CNRS mostraram-se suficientes para influenciar a DAS, tanto pela composição quanto pela forma em que a deposição sedimentar ali ocorre, evidenciando a necessidade de obtenção de maiores conhecimentos, inclusive lançando mão de uso rotineiro de GPR em trabalhos futuros envolvendo a avaliação de água subterrânea, o que facilitaria, agilizaria e tornaria extremamente correta a tomada de amostras, tendo em vista que estas seriam executadas seguindo as informações pretéritas sobre as estruturas geológicas.

Entretanto, as tantas respostas obtidas com o desenvolvimento deste trabalho, abriram outras lacunas de conhecimento indicando que os estudos na CNRS, nas suas regiões limítrofes e em outras regiões constituídas por sedimentos permeáveis devem ser realizadas cada vez mais.

Uma vez evidenciado, ainda que preliminarmente, o papel da geologia sobre a DAS, ficam recomendados estudos sobre o efeito dos sedimentos sobre a difusão do ^{222}Rn, de forma a obter dados exclusivos da atividade de ^{222}Rn do sedimento, tanto da faixa de praia, regiões lagunares e da barreira em diferentes profundidades.

Como a avaliação tanto dos nutrientes quanto dos elementos traço apontou a ausência de padrões de concentração e fluxos, maiores estudos são encorajados a serem desenvolvidos em diferentes pontos da CNRS, de forma especial que sejam contemplados estudos em diferentes profundidades, seja ao longo da barreira arenosa como na faixa de praia, procurando compreender os processos biogeoquímicos e os principais atores responsáveis. Por exemplo, realizar estudos em diferentes condições meteorológicas, especialmente quando o sistema está sob efeito de fenômenos como ENSO e LNSO e, quando estes não se fazem sentir sob o ambiente costeiro.

Ainda, a respeito de avaliações ao longo do tempo, são recomendados estudos com uso de séries temporais de ^{222}Rn, como forma de esclarecer as oscilações vinculadas a subida e descida de marés, que atualmente são consideradas como desprezíveis em função da proximidade de um ponto anfidrômico, bem como efeitos de forçantes vinculadas ao ritmo nictemeral e variações de regime de ventos.

A metodologia escolhida para o desenvolvimento deste trabalho, tendo como base o decaimento natural do ^{222}Rn mostrou-se extremamente eficaz e, continua sendo recomendada, inclusive pela possibilidade de avaliação de corpos hídricos costeiros com diferentes características (rios, lagoas, mar), mas faz-se importante recomendar que avaliações concomitantes de isótopos de rádio sejam executadas, pois trazem informações complementares importantes, principalmente quanto a determinação do tempo de residência da água subterrânea.

Durante a realização de cruzeiros oceanográficos do Instituto do Milênio – FURG, os sinais de sonar de varredura lateral de 3,5 KHz registraram 10 paleocanais na costa adjacente à região CNRS, bem como na BLP. Estes paleocanais podem atuar como caminhos preferenciais de escoamento da água subterrânea, potencializando-a em direção ao mar aberto. Dessa forma, é recomendada a continuidade destes estudos acoplados a ferramenta GPR no sentido de mapear a região costeira e verificar a existência de traços destes paleocanais na barreira arenosa, o que poderia significar uma DAS amplificada e ainda mais efetiva como fonte de macro e micronutrientes para a plataforma costeira adjacente.

Considerando, também, a importância do fluxo de micro e macronutrientes para a produção biológica primária e secundária, já vinculada à DAS em outros locais do sul do Brasil, ficam recomendados estudos sobre as comunidades planctônica e bentônica (densidade, distribuição e composição) associadas a DAS.

O efeito antrópico sobre a DAS deve ser considerado para uma avaliação rigorosa, principalmente porque a CNRS sofre ocupação maciça nos períodos de veraneio, onde a população aumenta uma ordem de magnitude e, o sistema de esgotamento sanitário (sistemas fossa-sumidouro) é totalmente defasado, causando intensificação dos aportes de nutrientes e contaminantes, provocando efeitos nocivos às águas subterrâneas desta planície sedimentar, que é completamente permeável.

Por fim, elencadas algumas das oportunidades de estudos ainda pendentes, verifica-se mais uma vez a importância deste trabalho para a CNRS e, recomenda-se fortemente, que estudos envolvendo as conexões e interrelações entre as águas de superfície e subterrânea sejam continuadas, pois somente desta forma será possível entender os processos biogeoquímicos entre a barreira arenosa e sua região costeira adjacente, uma das regiões mais ricas em produção biológica do Brasil.

CONCLUSÃO GERAL

O presente trabalho estabeleceu, através do uso de duas técnicas bastante atuais, o GPR e o isótopo ^{222}Rn, as interações entre águas superficiais e subterrâneas na região da CNRS. Ficaram esclarecidas as arquiteturas deposicionais em duas regiões com diferentes processos evolutivos costeiros, progradacional e retrogradacional, bem como a DAS e os fluxos de micro, macronutrientes e elementos traço associados.

Foi comprovada a interdisciplinaridade do estudo da DAS e a necessidade de avaliações amplas que contemplem:

- a geologia da região, uma vez que foram comprovados os efeitos dos compartimentos geológicos sobre a DAS, onde variações de compactação, estrutura, granulometria e descontinuidades (afloramentos, estuários e unidades isolantes) produziram DAS cerca de 15 vezes maior na região progradante do que na retrogradante;

- a meteorologia local, que influencia no processo de recarga de águas subterrâneas e de mananciais superficiais, é submetida aos efeitos temporais que atuam de forma tão intensa resultando em uma DAS de, aproximadamente, 10 vezes maior entre o verão influenciado pelo ENSO e o verão subsequente, dominado pelo LNSO;

- a química dos traçadores radioativos, uma vez que este trabalho evidenciou a pertinência do uso do ^{222}Rn, bem como reconhece a contribuição de outros isótopos e metodologias, sempre considerando a região de estudo e o que se quer responder;

- a hidroquímica da DAS, tendo sido evidenciada neste trabalho a sua expressividade, seus efeitos e variações da DAS nos diferentes compartimentos analisados, refletindo ambientes sobre domínio de processos biogeoquímicos diversos;

- a produtividade biológica suportada pela DAS e os efeitos oceanográficos mundiais, em vista dos expressivos fluxos já mencionados e as consequências positivas sobre a produção primária, secundária e os ecossistemas associados;

- a hidrologia da região, conhecida mais extensivamente por este estudo, inclusive indicando que a DAS responde por 20 % do volume de corpos hídricos continentais (lagoas e rios) na região a CNRS, bem como por uma descarga costeira expressiva, que se assimila à cerca de 1 %, 6 % e 19 % DAS calculadas para a Laguna dos Patos, Lagoa Mangueira e Estuário Tramandaí, respectivamente, reconhecidos mananciais superficiais.

Fica, assim, evidenciada a influência decisiva da geologia entre os compartimentos superficial e subterrâneo na CNRS, região diversa, importante e possivelmente única, que é um "*driving force*" da DAS, com grau de importância semelhante aos efeitos climatológicos de grande escala, como ENSO e LNSO.