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1 Detection and quantification of low submarine groundwater discharge flows by

2 radionuclides to support conceptual hydrogeological model of porous aquifers

- 3 S. Santoni^{1,2,*}, E. Garel^{1,2}, A. Mayer³, O. Radakovitch^{4,5}, Y. Travi³, F. Huneau^{1,2} 4 5 6 ¹Université de Corse Pascal Paoli, Faculté des Sciences et Techniques, Département 7 d'Hydrogéologie, Campus Grimaldi, BP 52, F-20250 Corte, France. 8 ²CNRS, UMR 6134, SPE, F-20250 Corte, France. 9 ³Avignon Université, UMR 1114 EMMAH, UAPV, 301 rue Baruch de Spinoza, BP 21239, F-84916 10 Avignon, France. ⁴ Aix-Marseille Univ, CNRS, IRD, INRA, Coll France, CEREGE, 13545 Aix-en-Provence, France. 11 12 ⁵Institut de Radioprotection et de Sureté Nucléaire (IRSN), PSE-ENV/SRTE/LRTA, BP 3, 13115 Saint-13 Paul-Les-Durance, France. 14 15 ^(*) Corresponding author: Sébastien Santoni, Département d'Hydrogéologie, UMR CNRS SPE 6134 16 Université de Corse Pascal Paoli, Campus Grimaldi, F-20250 Corte, France. Contact: 17 santoni_s@univ-corse.fr or santonisebastien17@gmail.com 18 19 Highlights 20 • Low and diffuse submarine groundwater discharge flows are investigated. Groundwater flow velocity from ²²²Rn and from Darcy law are in agreement. 21 ٠ 22 Discharge rates are in agreement with recharge rates solving aquifer water • 23 balance.
- 24

25 Abstract

26 Submarine groundwater discharge (SGD) is considered as a good indicator of the natural 27 behavior of coastal aquifers especially in the case of intense groundwater withdrawal or 28 when the aquifer recharge conditions are limited. Indeed, a decrease in SGD flows would 29 result in saline intrusion compromising the groundwater reservoir use. The need for SGD 30 studies in Mediterranean and semi-arid environments have long been recognized and little is 31 known on low and diffuse flows. For this reason, the detection and quantification of low and 32 diffuse SGD flows have been attempted in a Mediterranean porous aguifer with low potential 33 of recharge and high touristic pressure from using a multi-tracing approach combining aerial 34 thermal infrared images (TIR), temperature (T), electric conductivity (E.C.), major ions, selected trace elements, ²H, ¹⁸O, ²²²Rn, ²²³Ra and ²²⁴Ra. The TIR images allowed the 35 36 detection of slight temperature variations at the seawater surface. Contrasted lithology and 37 strong water-rock interactions due to groundwater residence time of many decades favor the ²²²Rn, ²²³Ra and ²²⁴Ra contents in groundwater that reach the seashores. Low temperature 38 39 and E.C, as well as high radon and radium activities, evidenced SGD flows in agreement 40 with the known flow conditions within the aquifer. The quantification of SGD flows revealed in

41 very good agreement with the known recharge rates of the aquifer that validates its 42 conceptual hydrogeological model. For the first time in the Mediterranean or in semi-arid 43 context, a natural tracing experiment based on geochemistry and isotope hydrology tools 44 allowed the quantification of SGD flows to validate the hydrogeological conceptual model and 45 to solve the water balance at the whole aquifer scale. Such a methodology could be applied 46 worldwide to other similar coastal aquifers with low and diffuse SGD flow conditions.

47

48 Keywords

49 Diffused SGD flows, Low Recharge, TIR imagery, Radon, Radium, Aquifer Water Balance.

50

51 **1. Introduction**

52 To ensure sustainable management of groundwater resources, it is necessary to rely 53 on detailed conceptual hydrogeological models. This includes the quantification of all 54 water balance terms at the aquifer scale. For coastal aquifers, an important water 55 balance term is the submarine groundwater discharge (SGD) (Burnett et al., 2006; 56 Baudron et al., 2015). The aquifer recharge areas contributing to SGD flows being 57 frequently densely populated, SGD flows are then recognized to be a source of 58 nutrients, contaminants and pollutants that may affect the coastal hydrosystems 59 functioning (Kaleris, 2006; Tomasky-Holmes et al., 2013; Rodellas et al., 2015). Furthermore, the amount of fresh SGD flows can reduce or even disappear due to 60 61 over-pumping, causing seawater intrusion into coastal aguifers compromising the 62 groundwater reservoir use (Moujabber et al., 2006; Mahlknecht et al., 2017; Nlend et 63 al., 2018).

The issue of seawater intrusion is obviously stronger for coastal aquifers displaying large interface with the sea that can be worsened by pumping rates (McCoy and Corbett, 2009; Mahlknecht *et al.*, 2017) and/or by increasing climate change effects such as seawater rise (Sherif and Singh, 1999; Barlow and Reichard, 2009; Werner *et al.*, 2013) or modifications in aquifer recharge conditions governing groundwater

69 levels (Holding et al., 2016; Meixner et al., 2016; Santoni et al., 2018). Bokuniewicz 70 et al. (2003) propose a typology for coastal groundwater and provides order of 71 magnitude for SGD flows rates in porous media from various location and climate 72 conditions highlighting very low values around 1 - 2 litter/day/linear meter in arid 73 regions. Such coastal aquifers displaying low SGD flows are then more vulnerable to 74 saline intrusion due to the precarious balance between freshwater and seawater of the salt wedge. Dependant of climate change, coastal drainage geometry and 75 76 groundwater withdrawal for coastal population, irrigation, and industrial activities, the presence of SGD flows is consequently considered as a good indicator of the natural 77 78 behavior of aquifers (Sawyer et al., 2017).

79 Coastal aquifers of the Mediterranean area are known to be particularly vulnerable facing climate and anthropogenic pressures, so much so saline intrusions are already 80 81 common (Richts et al., 2011; Pachauri et al., 2014). Destouni and Prieto (2003) 82 made a numerical modelling of whole-aquifer water balance terms including SGD 83 flows in a selection of porous aquifers in Israel, Greece and Cyprus for several 84 management conditions. Later, Mazi et al. (2014, 2016) used an analytical approach 85 to optimize groundwater exploitation conditions in a selection of porous aquifers 86 located in the Nile Delta, Israel, and Cyprus based on water balance components 87 quantification at the whole aquifer scale too. They also define the concept of 88 sustainable management space to prevent any major seawater intrusion.

In addition to numerical modelling and analytical solutions, experimental methodologies have been developed to study SGD flows. TIR Imagery be used to localize them, based on the detection of warm or cold groundwater inputs into the sea with a contrasted temperature (Rapaglia *et al.*, 2015; Tamborski *et al.*, 2015). This technique has been employed worldwide and may be combined with *in situ*

94 measurements of T, E.C. and/or geochemical tracers including natural radioactive 95 isotopes, particularly radon and radium isotopes (Mulligan and Charette, 2009; Mejías et al., 2012; Povinec et al., 2012; Lee, 2014; Rapaglia et al., 2015; Tamborski 96 97 et al., 2015). These radionuclides are produced in the aquifer or poral waters of the 98 sediment by the decay chain of uranium and thorium within the aquifer rocks and 99 sediments, and are released in coastal waters by advection or diffusion, making them 100 useful tracers of SGD flows (Moore, 2000; Burnett et al., 2006; Beck et al., 2010). 101 Radium isotopes are indeed relevant tools thanks to their wide range of half-lives (from 3.63 days for ²²⁴Ra to 1600 years for ²²⁶Ra). They can also be used to describe 102 103 SGD processes in discriminating freshwater inputs from rivers or aquifers with those 104 of recirculated seawater (Moore, 1996; Povinec et al., 2006; Stieglitz et al., 2010; 105 Bejannin et al., 2017). In the current literature, SGD flows are widely investigated 106 through experimental methodologies in mainly large diffuse or channelized flows 107 provided by karst, fractured or porous aquifers (Burnett et al., 2003; Burnett et al., 108 2006; Lamontagne et al., 2008; Povinec et al., 2008; Mejías et al., 2012; Santos et 109 al., 2012; Post et al., 2013; Montiel et al., 2018). However, to our knowledge, 110 experimental methodologies have never been used to quantify diffuse SGD flows 111 necessary to solve the water balance at the whole aquifer scale in Mediterranean 112 and semi-arid areas.

An experimental multi-tracing approach combining TIR imagery, T, E.C., major ions, selected trace elements, ²H, ¹⁸O, ²²²Rn, ²²³Ra and ²²⁴Ra was attempted on the well documented Mediterranean coastal aquifer of Bonifacio (Corsica Island, France). Even if no major seawater intrusion is yet detected, the Bonifacio aquifer is highly vulnerable facing seawater intrusion because more than a half of its boundaries conditions are the Mediterranean Sea (Santoni, 2016). In addition, the equilibrium

119 between groundwater and seawater is fragile since SGD flows are expected to be 120 low due to limited recharge conditions typical of the Mediterranean area (Stigter et 121 al., 2012; Ghiglieri et al., 2014). Previous studies already demonstrated its low 122 potential of recharge (Chesnaux et al., 2018; Santoni et al., 2018) and highlighted its 123 porous matrix composition (Orsini et al., 2010; Reynaud et al., 2012). As a 124 consequence, the Bonifacio coastal aguifer is an ideal study site to evaluate the 125 potential of TIR, geochemical and isotope tools to detect and quantify low and diffuse 126 SGD flow conditions. For this aguifer, recharge and groundwater withdrawal rates 127 being already known, the SGD flows quantification is intended to validate its 128 conceptual hydrogeological model as well as to solve the water balance at the whole 129 aquifer scale.

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131 **2. Study site**

The Bonifacio aquifer (Corsica Island, France) is part of a coastal plateau located in the western Mediterranean (Figure 1). The study site is under a typical Mediterranean climate with mean annual precipitation and temperature about 605 mm and 16.8 °C respectively (calculated from 01/2000 to 01/2012 at the Bonifacio Meteo France station). The sector of Bonifacio is known for its wind blowing up to 300 days per year, often with gusts above 60–80 km/h especially in the extreme south of the plateau.

The Bonifacio plateau is constrained in equal proportions by the Mediterranean Sea at the South and Northeast and by granites on East and West. Its shape is triangular and it extends over only 25 km² with a mean elevation of 80 m asl. It is made of Miocene sediments filling a fractured Hercynian granitic depression up to 270 m deep continuing in the surface at an altitude of 243 m at the Monte Corbu (west) and of

144 113 m at the Punta di San Mulari (Rossi and Cocherie, 1991; Orsini *et al.*, 2010). The 145 3,000 inhabitants of the Bonifacio city reside mainly along the St. Julien and the St. 146 Jean valleys and also around the harbour. All the Bonifacio municipality coastlines 147 constitute a Natural marine reserve of National importance. Many campgrounds and 148 villas spread all over the plateau are crowded during the summer due to tourism 149 activities equivalent to 1.8 million people each year.

150 The hydrogeology of the Bonifacio plateau consists of two aquifer levels separated 151 by an aquitard (Santoni, 2016). The two aquifers levels are mainly found in the 152 Bonifacio and Balistra geologic formations (Reynaud et al., 2012), forming an upper 153 and a lower aquifer level, respectively. They are separated by an aquitard 154 corresponding to a silty layer of the Cala di Labra geologic formation. Both aquifer 155 levels are composed of sandy calcarenites and calcareous sandstones representing 156 a very compacted material with a cemented granular structure whose effective 157 porosity is on average 7% (Dörfliger et al., 2002; Chesnaux et al., 2018). 158 Potentiometric levels and geochemical tracers highlighted a recharge area on the 159 plateau (autochthonous recharge) and another within the surrounding fractured 160 granitic massifs (allochthonous recharge) from which two main flow lines are 161 identified in direction of the Bonifacio Harbour and of the Sant'Amanza Bay (Figure 162 1). The Canali River is the only permanent river and it has very low flow. It takes its 163 source at the West within granitic massifs before flowing on the plateau and draining 164 the upper aquifer until the Sant'Amanza bay (Santoni, 2016).

The carbonate plateau of Bonifacio is in continuity with the large Miocene sedimentary basin developing between Corsica and Sardinia islands. The sedimentary deposits of the Bonifacio plateau are conditioned by the granitic substratum and the thickness of the sediments is a function of the palaeo-

morphology of this one. Since no karst morphology is observed and considering the
porous matrix of the aquifer, the SGD flows are expected to reach the sea very close
to the shorelines and according to the salt wedge position (Figure 1).

172 The groundwater residence time was estimated from dissolved anthropogenic gases 173 CFCs and SF₆ concentrations in groundwater (Santoni *et al.*, 2016b). Weighted ages 174 range from 0 to 50 years in the upper aguifer and from 35 to 60 years in the lower 175 aquifer implying a low dynamic of renewal and a long residence time amplifying water-rock interactions. An approach using ⁸⁷Sr/⁸⁶Sr validated groundwater flow 176 177 pattern and residence time by computing water-rocks interaction kinetics and mixing 178 rates between the granites, the upper and the lower aquifer (Santoni et al., 2016a). 179 Recently, Chesnaux et al. (2018) and Santoni et al. (2018) highlighted favourable 180 aquifer recharge rates about respectively 23 and 26 % of annual rainfall that 181 suggests the presence of low but detectable SGD flows near the coastal shores.

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183 **3. Method**

184 The investigation strategy consisted in three main steps: (i) the acquisition of airborne TIR images of the coastlines on the 9th September 2011 to detect and 185 186 quantify SGD flows; (ii) the measurement of physicochemical parameters, short halflives radionuclides (222Rn, 223Ra, 224Ra), major ions, trace elements and stable 187 188 isotopes of the water molecule in one stream and nine groundwater samples (Figure 1) on the 27th, 28th and 30th October 2012. A fixed measurement for ²²²Rn in position 189 A in the Harbour was also made; (iii) the measurement of temperature, E.C., ²²²Rn 190 ²²³Ra and ²²⁴Ra in seashores to validate and refine the SGD flows localisation and 191 also to try their quantification on the 8th, 9th, and 10th January 2014. The objective 192 193 of the groundwater and river sampling was to identify the main hydrochemical 194 features to ensure the presence of sufficient radon and radium activities in195 groundwater to detect SGD flows later in the sea.

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3.1.1. Thermal Infrared imagery

TIR images were acquired along the carbonated rocks shores on the 09th September 198 199 2011 using a FLIR thermal system B20HSV. This period was chosen to provide the best thermal contrast between groundwater and seawater. The flight acquisition of 200 201 airborne TIR images was performed by the service-provider company "l'Avion 202 Jaune". The flight was conducted at 2200 m altitude from 10:30 am to 11:00 am 203 without any cloud and during low wind (7.8 m/s, ESE direction at the Meteo France 204 weather Station of Bonifacio, Pertusato). The atmospheric temperature during the 205 flight was 24.2°C and no significant rain event was observed during the last 6 days. 206 The TIR camera wave-length range from 7.5 to 14 µm with 16384 grey levels set on 207 a temperature palette between -40 to +150°C with an accuracy of ± 1°C. At 2200 m 208 altitude, the pixel field of view is about 2.0×2.0 m.

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3.1.2. Geochemistry and stable isotopes of the water molecule

E.C. and T were directly measured in the field using a WTW Cond 3310 (WTW GmbH, Weilheim, Germany). Bicarbonates were also determined in the field with a HACH digital titrator (HACH Company, Loveland, USA). Major ions samples were filtrated through 0.45 mm nitrocellulose membranes in two 50 mL polyethylene bottles before storage at 4°C. One bottle was acidified using ultrapure nitric acid for cation analysis. A Dionex ICS 1100 chromatograph (Thermo Fischer Scientific, Waltham, USA) was used to determine ionic concentrations at the EMMAH

218 laboratory of the University of Avignon, France. The analysis quality was validated for219 ionic balance error of less than 5%.

220 Samples for trace elements were filtered through 0.20 µm nitrocellulose membranes 221 and acidified with ultrapure nitric acid in 50 mL polyethylene bottles before storage at 222 4°C. A Thermo Fisher Q-ICP MS X-series II was used to determine trace elements 223 concentrations at the AETE technical platform of the University of Montpellier, 224 France. Samples for the analysis of stable isotopes of the water molecule were 225 collected in 20 mL amber glass bottles with no head-space. A liquid-water stable 226 isotope analyzer DLT-100 (Los Gatos Research, San Jose, USA) was used in 227 accordance with the analytical scheme recommended by the International Atomic 228 Energy Agency (Penna et al., 2010) at the EMMAH laboratory of the University of 229 Avignon, France. The analysis quality was checked with standard deviation conditions up to 0.1‰ for δ^{18} O and 1‰ for δ^{2} H. 230

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3.1.3. ²²²Rn, ²²³Ra and ²²⁴Ra activities measurements

The ²²²Rn activity was measured in groundwater and in seawater along the 233 coastlines and within the Bonifacio Harbour in six fixed positions by using a RAD7 234 235 radon monitor combined with a RADAQUA water exchanger (Durridge Co. Inc.). Prior 236 to the radon measurements, the wind was moderate (from 4.0 to 19.6 m/s, ESE direction) from the 27th to the 30th October 2012 and low (from 3.8 to 5.0 m/s, SW 237 238 direction) from the 8th to the 10th January 2014. Each sampling campaign were preceded by slight to moderate rain events (45 mm on the 23rd October 2012; 20 mm 239 on the 05th January 2014). Since seawater typically displays low activity, the 240 241 sensitivity of the system was increased using two RAD7 monitors connected with the 242 RAD-AQUA in closed loop (Mayer et al., 2016). The water was pumped with a 4.5

I/min flow rate for injection into the RADAQUA water exchanger. The ²²²Rn activity 243 measure is determined from the decay of daughter isotope ²¹⁸Po when the 244 245 equilibrium in the air-loop between the gas-exchanger and the two RAD7 monitors is reached (Dimova et al., 2009). The ²²²Rn activity of water is then calculated from that 246 247 of air knowing the partition Ostwald coefficient, which depends on salinity and 248 temperature (Schubert et al., 2011). For the coastlines survey, all devices were 249 installed on a semi-rigid vessel equipped with GPS, E.C. and T data loggers. The 250 system allowed continuous measurements during navigation or fixed position. For 251 more detailed information, sampling and analytical process are consistent with those 252 outlined in (Mayer et al., 2016).

The ²²³Ra and ²²⁴Ra activities were measured using Mn-fibers by filtration of surface 253 water, groundwater, and seawater samples. Sample positions for the Bonifacio 254 Harbour are the same as for ²²²Rn. The fibers were analysed using RaDeCC 255 256 counters systems at the EMMAH and CEREGE laboratories (Moore and Arnold, 1996). Calibration was made by using in-house mono-isotope standards of ²²⁷Ac and 257 ²³²Th (parents of ²²³Ra and ²²⁴Ra respectively) and multi-isotope standards, 258 containing ²²⁷Ac, ²³²Th, and ²²⁶Ra. All standards were prepared according to the 259 260 technique of Scholten et al. (2010). Efficiencies changes with time are within the error 261 range and thus not significant. Errors were calculated according to Garcia-Solsona et 262 *al.* (2008).

To assess whether the diffusive release of radionuclides from sediments could be a significant radon source, five sediment samples were collected from the sea beaches of Paraguanu, Pertusatu, San Mulari, Sant'Amanza and Canettu (Figure 1). To estimate ²²²Rn diffusion from sediment, activities in pore water were analysed following the equilibration method after Corbett et al. etc (Corbett *et al.*, 1998; Burnett

et al., 2007; Kluge *et al.*, 2012). Experiments were performed with 500 g of dry sediment equilibrated in closed glass bottles with 900 mL of Ra-free seawater, previously filtrated through Mn-fiber. Seawater and sediment equilibrated in closed bottles over one month, according to the protocol of Corbett et al 1998.

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273 4. Results and discussion

274 *4.1.SGD flows detection from airborne TIR images*

275 The seawater surface temperature obtained in September 2011 through the TIR 276 image varies from 19.7 to 23.5°C along the coast (Figure 2). In the Harbour, red 277 hotspots correspond to pontoons and moored boats. The most common value range 278 between 21.2°C and 23.5°C due to summer conditions, but the temperature may be 279 less than 21.2°C locally. This is especially the case in the Sant'Amanza Bay and in 280 the Bonifacio Harbour where a discreet but detectable decrease in temperature is 281 observed. This low thermal contrast is obviously the result of a high thermal diffusion 282 of groundwater in the sea. No plumes are visible unlike on other SGD flows studies 283 (Tamborski et al., 2015; Bejannin et al., 2017), but this weak and progressive 284 decrease in temperature is a first clue arguing in favour of low and diffuse SGD flows 285 as expected for this Mediterranean porous aquifer. The slight decrease in 286 temperature is detectable over a length of around 350 m in the Sant'Amanaza bay 287 and 140 m in the Bonifacio Harbour. Furthermore, cooler temperatures less than 288 21.2°C are also observed at the extreme south of the plateau. This is probably due to 289 the wind occurring in this specific area during the flight (7.8 m/s, ESE direction 290 measured at the Pertusato Meteo France weather station).

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293 4.2. Groundwater-rock interactions as a source of radionuclides in SGD

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295 Groundwater from the upper aquifer (Bonifacio geologic formation) and stream water 296 were investigated to characterize their hydrochemical and isotopic signatures and to 297 identify mineralization processes that may further help the SGD flows detection 298 (Table 1). Groundwater temperatures are close to the average annual atmospheric 299 temperature at Bonifacio (+16.8°C) involving a low dynamic of renewal that favour 300 water-rocks interactions within the aquifer (Santoni et al., 2016b). The EC is rather 301 high for all samples with values from 990 to 2040 µS/cm, and is mainly influenced by 302 HCO₃, CI, Ca and Na contents. The pH values as well as the high contents in HCO₃ 303 and Ca indicate interactions with the aquifer carbonate matrix. The high Na, Cl and Mg contents denote an influence from the sea, but the negative $\delta^2 H$ and $\delta^{18} O$ 304 305 signatures exclude any large seawater intrusion into the aquifer and rather suggest 306 the influence of sea sprays in the mineralization (Santoni et al., 2016a). Groundwater 307 NO₃ contents comply with those found in natural waters, except for three samples 308 above 20 mg/L which highlight locally the vulnerability of the aquifer influenced by human activities. SiO₂ and U contents are noticeable with values up to 17 mg/L and 309 310 1.5 μ g/L, respectively, with a correlation coefficient r² about 0.81. These are elements 311 sourced by granitic rocks surrounding the aquifer or sourced by the calcarenites of 312 the aquifer that include sediments from the granite weathering.

The ²²²Rn, ²²⁴Ra and ²²³Ra activities were measured in stream and groundwater (Table 2). The ²²³Ra activity is low (less than 1.5 Bq/m³) compared to the ²²²Rn and ²²⁴Ra activity, varying from 1199 to 27853 Bq/m³ and from 8.8 to 34.3 Bq/m³, respectively. Radon is fairly well correlated to dissolved uranium in groundwater, (r² = 0.79, Table 1), suggesting that the main sources of radon are the Herynian granites. The higher activities are found in groundwater sampled at the interface with the fractured granites (REP-B18 and CPO-B10). Stream water CANR-R1 and groundwater from the plateau display lower radon activities, between 1199 and 6081 Bq/m³. Bonifacio groundwater and stream water have therefore radon activities sufficiently elevated to provide a radon signature in seawater expected to be detectable using our Rad7 system near the discharges points.

324 Figure 3 displays binary scatter plots combining hydrochemical and isotope data of 325 groundwater along the flow path from the recharge areas until the coastal outlets 326 where the SGD flows measurement is carried out. The (Na+Cl)/(Ca+HCO₃) vs Na 327 graph (Figure 3-A) shows the influence of both marine context and carbonated 328 lithology on the groundwater mineralization through the Na+Cl and Ca+HCO₃, 329 respectively. From the recharge areas to the aguifers outlets, the Na content coming 330 from sea sprays decrease by dilution due to a mixing of flow lines originated from 331 different recharge process such as focused recharge (fast transfert of rainwater to 332 the water table) or irrigation recharge (Santoni et al., 2018). In the same time, the 333 (Na+Cl)/(Ca+HCO₃) ratio decreases, highlighting an increase in the (Ca+HCO₃) 334 content due to the calcite dissolution. This is well supported by the increase in the 335 saturation indexes (SI) of calcite plotted in Figure 3-B. Moreover, in the same figure, 336 the increase in the SI of strontianite with calcite suggests the control of the magmatic 337 and sedimentary lithologies on groundwater mineralization along the flow path from 338 the granites to the carbonate aquifer as also observed in Santoni et al. (2016a). 339 Indeed, the high correlation coefficient between saturation indexes of calcite and 340 strontianite highlights dissolution of granites and of calcarenites that includes calcite 341 and granitic alteration products. The groundwater mineralization is consequently

influenced by the groundwater origins, by the contrasted lithologies found along theflow path and finally by complex mixing processes.

344 This is also true for the groundwater radon activity: the groundwater flow line 345 originating from the granite surroundings (allochthonous recharge area) displays the 346 higher U concentration and radon activity (Figure 3-C). This feature slightly 347 decreases toward the Stentinu (Canali valley, Sant'Amanza Bay) outlet, probably due 348 to mixing with surface water with groundwater from the calcarenites. This is well supported by the δ^{18} O enrichment suggesting the progressive addition of evaporated 349 350 surface water (Santoni et al., 2018). In addition, the groundwater flow line originating 351 from the plateau (autochthonous recharge area) displays lower U concentration and radon activity that remains relatively high and guite stable until the outlet of the Saint 352 353 Julien valley (Bonifacio Harbour). The SGD flows detection from ²²²Rn survey along 354 the coastlines is then expected to be fruitfull in this context.

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356 4.3. SGD flows detection from in situ T, E.C. and ²²²Rn survey along the 357 coastlines

358 The *in situ* seawater temperatures measured along the coastline during the survey 359 are mainly comprised between 14.5 and 15.0°C (Figure 4-A), typical for winter in the 360 western Mediterranean. The local decrease in temperature can be probably related 361 to temperature equilibrium with the cold atmosphere, especially in shallow areas. In 362 addition, thermal contrast between the groundwater and the sea or the atmosphere is 363 expected to be very limited, especially in case of diffuse outflow during winter when 364 the difference of temperature between the sea and the groundwater is the smallest. It 365 is then more relevant to refer to the Figure 2 for discussing temperature contrasts, 366 more visible and relevant during summer. Nevertheless, TIR images and in situ temperature give insights on coastal water bodies structure, with differentiated water
bodies in the Stentinu and in the Maora areas of the Sant'Amanza bay as well as
within the Bonifacio Harbour. These water bodies does not instantly mix with external
seawater, suggesting low advection processes.

371 To detect the presence of continental freshwater inputs to the seashores, in situ E.C. 372 was also measured during the survey (Figure 4-B). The highest values are between 373 58 and 61 mS/cm and are found along the eastern granitic part of the study site as 374 well as between the Bonifacio Harbour and Pertusato. In opposition, lower values 375 between 55 and 58 mS/cm are found especially in the northern, southern and 376 western part of the Bonifacio plateau coasts including the Bonifacio Harbour and the 377 Sant'Amanza Bay (Stentinu). Since the E.C. variation is independent of 378 environmental temperature, it is then reasonable to relate a decrease in E.C. to a 379 continental water inflow even if it does not allow to distinguish between surface and 380 groundwater inputs.

381 The Figure 4-C displays the radon activity distribution along the coastline. The 382 eastern granitic part and the northern plateau coasts display intermediate radon activity, comprised between 0 and 40 Bq/m³. The punctual increase observed along 383 384 the eastern coast may be explained by very local inputs of fresh groundwater through 385 the intense fracturation of the Hercynian granites (Orsini et al., 2010; Santoni et al., 386 2016a), but in any case they cannot be considered as Bonifacio aquifer outlets. The 387 coasts surrounding the Bonifacio plateau display no or low radon activity at the south 388 and the west, except in the harbour and in the Sant'Amanza bay where the values may be greater than 50 Bg/m³, especially in the Maora and Stentinu areas. The low 389 390 E.C. value and low radon activity found at the south and west part of the plateau can 391 be explained by a weak input of surface water. In opposition, the low E.C. and high

392 radon activity found in the Bonifacio Harbour and in the Sant'Amanza Bay (including 393 the narrow Stentinu area of 70 m wide) reveals the presence of SGD flows. This is in 394 very good agreement with the potentiometric and ageing groundwater flow line 395 directions from Santoni (2016) and with the slight decrease in seawater temperature 396 observed from the TIR images displayed in Figure 2.

The Figure 5 displays E.C. box plots as a function of ²²²Rn activity classes for all the 397 coastline survey data. The correlation coefficient r² between E.C. and ²²²Rn activity is 398 399 about 0.53, hihlighting a relation between these two variables: the lower is the E.C., 400 higher is the radon activity. This means that the E.C. may be relevant for the 401 detection of SGD flows along the coastlines. Nevertheless, SGD flows may have contrasted effects on coastal seawater E.C and ²²²Rn activity according to the 402 403 groundwater origins (*i.e.* recharge areas). The Figure 3-C clearly shows a wide range of ²²²Rn activity in groundwater, with the highest values for groundwater coming from 404 405 the granites area and reaching the Stentinu (Figure 1, Figure 4-C). As a consequence, low SGD flow rates may provide a high ²²²Rn activity with no major 406 407 influence on E.C. This observation may explain the moderate r² value (Figure 5). 408 Even if each parameter individually presents its limits, their combination allows here 409 to clearly detect the presence of SGD flows.

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411 **4.4.** Estimates of SGD seepage velocities from radon and radium activities

An estimate of SGD seepage velocities using radon and radium activities from the fixed measurements carried out over the 1800 m length of the Bonifacio Harbour (Figure 1) is here attempted. As suggested by *in situ* temperature repartition along seashores (Figure 4A) highlighting different water masses, this calculation assumes that the mixing with external seawater is negligible. This assumption was tested evaluating the eddy diffusion (D_{eddv}) coefficient in the harbour's water as in Moore

418 (2000). Under the assumption of isotropic medium and steady state condition, the419 eddy diffusion coefficient is given by equation 1:

420
$$D_{Eddy} = \sqrt{\frac{\lambda}{\frac{\partial \ln(C)}{\partial x}}} \text{ (equation 1)}$$

421 where C is the activity of tracer (radon or radium isotope), x the distance from the 422 input point and lambda the decay constant of the tracer. The computed distance vs. 423 logarithm activities regression line and eddy diffusion coefficients are reported in 424 Figure 6 and Table 4, respectively. The very good correlation between logarithm of 425 radionuclide activities and distance (Figure. 6), as well as the low eddy diffusion 426 coefficients deduced from them suggests that advection is minimal, diffusion and 427 radioactive decay from groundwater inputs in the inner side of the harbour are the 428 dominant processes governing the activity of these tracers in this setting and-at the time scales of the half lives of these tracers (3.8 days for ²²²Rn, 11.3 days for ²²³Ra, 429 and 3.6 days for ²²⁴Ra). This is probably due to the reduced volume and narrow 430 431 configuration of the Harbour.

432 The wind was low to moderate during the surveys (cf 3.1.3), and the narrow 433 configuration of the Bonifacio Harbour clealry limits the radon degassing from 434 seawater in this area. The radon degassing was therefore not taken into account in 435 the SGD seepage velocity calculation. The incertitude introduced by neglecting the degassing term is discussed later. To evaluate the potential influence of ²²²Rn 436 437 diffusion from the sediment (not associated to the SGD flows from the aquifer), a 438 selection of near-shore sediments were sampled over the study area and analysed for the ²²²Rn activity produced by their equilibration with seawater (Figure 1, Table 3). 439 The ²²²Rn diffusive flux obtained from these sediments is very low, in the range from 440

441 1.4 to 6.2 Bq/m²/day. These marine sediments thus cannot constitute a source of
442 ²²²Rn capable to prevent the detection of SGD flux.

In order to calculate SGD seepage velocities, the ²²²Rn, ²²³Ra, and ²²⁴Ra inventories 443 444 in the harbour have been determined. Radon and radium activities in function of 445 distance have been fitted by exponential curves that are then integrated over the 1800 m length of the harbour (Figure 6). The general pattern of ²²²Rn, ²²³Ra and 446 447 ²²⁴Ra data clearly shows an exponential decrease in their activities with distance, 448 implying that the dilution of the isotopic signature with external seawater is negligible. 449 The good exponential data-fit also suggests that the activities of these tracers are 450 probably in a steady state equilibrium condition between sources (groundwater 451 advection) and sinks (diffusions and decay). In the case of the present study, the 452 narrow morphology of the harbour and the piezometric gradient organisation implies 453 major flowlines converging into the internal part of the Bonificio harbour (Santoni, 454 2016). For these reasons, SGD seepage directly through the bottom sediments of the 455 harbour are expected to be low. Assuming horizontal SGD flows as constrained by 456 the geomorphology and the salt wedge position (i.e. horizontal convergence of 457 groundwater flow lines in Figure 1), the radionuclides activities in groundwater inputs 458 are considered as vertically constant on the innermost face of the harbour made of 459 concretes and limestone. Then, the integrals of the activity in harbour water over the distance correspond to the horizontal tracer's inventories for 1 m² surface units 460 461 flowing perpendicularly to the innermost face of the harbour and in direction toward 462 the sea (sampling point A to the sampling point F in Figure 1). Parameters of this 463 calculation (distances, activities, exponential regression equations, inventories and resulting seepages for ²²²Rn, ²²³Ra and ²²⁴Ra) are reported in Table 4. Under these 464

465 assumptions, the calculated horizontal inventories are independent of the water466 depths in the harbour.

From the horizontal tracer inventories, the activity flux (Bg/m²/day) of ²²²Rn, ²²³Ra 467 and ²²⁴Ra necessary to maintain the diffusive – radioactive decay profile may be 468 469 calculated for a unit surface perpendicular to the length of the harbour (Table 4), 470 assuming a steady state condition. The groundwater seepage supporting this flux is 471 then calculated dividing the activity flux by the groundwater activity C_{GW} as measured 472 in the sampled point ORE-W2 (Figure 1, Table 4). The computed SGD seepage 473 velocities necessary to compensate for diffusion and decay thus result about 2.24 and 3.19 m/day for ²²²Rn (for 2012 and 2014, respectively), 22.32 m/day for ²²³Ra 474 and 23.67 m/day for 224 Ra. 475

The same approach was applied in the Sant'Amanza Bay, second outlet of the 476 aquifer. From using the in situ 222 Rn in the Stentinu and considering C_{GW} as the 477 478 sampled point CPO-B10, the computed SGD flows seepage velocity necessary to 479 compensate for the diffusion and decay is about 1.84 m/day (Table 4). However, this 480 estimation is probably impacted by degassing process during flow within the Canali 481 Valley until the Stentinu. To avoid this, a computation may also be performed 482 considering C_{GW} as the canali river sample point CAN-R1. The computed SGD flows 483 seepage velocity is then about 18.6 m/day (Table 4).

The SGD seepage velocites calculated from ²²²Rn, ²²³Ra and ²²⁴Ra are in good agreement with other similar contexts in the Mediterranean as obtained in the Port-Miou Calanques, southern France (0.1 to 0.4 m/s), in open Mediterranean sea in the Gulf of Lion, southern France (23 m/s,) by Bejannin *et al.* (2017). Nevertheless, in

view of the fairly large range of SGD flows seepage velocities obtained, it appears
necessary to validate and better constrain these results.

490 **4.5.** Validation of SGD flows velocities by groundwater Darcy velocity

491 The eddy mixing coefficients deduced from the good exponential fit of radionuclides 492 activities over the 1800 m of the Bonifacio Harbour allow the application of the first 493 Fick's law to calculate the SGD freshwater seepage velocity necessary to dilute the 494 seawater salinity in the Harbour as observed in the E.C. profile. The flux calculated 495 using the first Fick's law assumes a freshwater fraction gradient in the harbour of -496 1.89E⁻⁰⁵ (m⁻¹). This gradient is calculated from 3.5% freshwater dilution, obtained from 497 the E.C. difference between inner harbour and offshore waters, over a total length of 498 1800 m (Figure 1). Assuming a steady state condition and E.C. conservative, the 499 seepage velocity of SGD flows of the "pure" freshwater component, necessary to 500 sustain the diffusive profile observed for E.C., is between 0.44 and 2.06 m/day (Table 4), using respectively the D_{eddy} obtained from ²²³Ra and ²²⁴Ra profiles. 501

Unlike for E.C., the SGD seepage velocities based on ²²²Rn, ²²³Ra, and ²²⁴Ra 502 503 corresponds to the sum of two flows, fresh groundwater and recycled seawater. The 504 quantification of the seawater recycling is however particularly difficult using radium 505 and radon isotopes because these tracers may be partially scavenged from seawater 506 when intruding a coastal aquifer, yielding in this case to an underestimation of total 507 SGD (Zhang et al., 2017). In other setting, the recycled seawater flux may constitute 508 a net tracer input in seawater inventory, yielding to an overestimation of the real 509 "continental" water SGD flux (Rodellas et al., 2015).

511 Nevertheless, by comparing the total SGD seepage velocity obtained from the radon 512 mass balance (2.25 m/d) and the freshwater SGD seepage velocity obtained from 513 the salinity profile with the first Fick's Law (2.04 m/d), we may roughly estimate that 514 the recycled seawater component represents about 10 % of the total SGD. The fraction of recycled seawater estimated by comparing ²²⁴Ra or ²²³Ra seepage 515 516 velocities and freshwater SGD seepage velocity from salinity profile is instead much larger (seawater recycling would be about 10 times the freshwater SGD). We tend to 517 518 prefer the estimate based on ²²²Rn because sediment equilibration experiments 519 showed that radon diffusion from sediment is very low. Seawater recycling through 520 these sediments would therefore not affect significantly the total input flux of radon 521 and so the estimate of total SGD. Conversely, for the radium isotopes, since radium 522 diffusion from near shore sediments is unknown, the effect of seawater recycling on 523 the total SGD calculation cannot be estimated. The total SGD estimate based on ²²²Rn (and the radon-based seawater recycling fraction) was also preferred because 524 525 the seepage velocities values obtained in 2012 and 2014 yielded similar results 526 (Table 4).

527 In order to validate the SGD velocities estimates, they are now compared with the 528 Darcy velocity deduced from hydrodynamic modelling and aquifer recharge rate 529 estimations previously published.

Table 5 shows Permeability (K) for the Bonifacio aquifer from existing literature and associated Darcy velocity (q) computed between EB-B1 and ORE-W2 in the Saint Julien Valley considering $\Delta L = 1173$ m and $\Delta H = 48$ m. The q values deduced from Proton Magnetic Resonance and pumping tests are between 0.46 and 1.06 m/d. These values are slightly lower but closest and in the same order of value to SGD

535 seepage velocities from radon and from the Fick law (Table 4). Uncertainties 536 nevertheless remain on q values because they are computed from permeability 537 coefficients K issued from different method as well as from various location over the 538 plateau. Furthermore, the Darcy groundwater velocities assume a steady state and 539 isotropic porous media, which is the case at the Saint Julien Valley's scale 540 (Chesneaux et al., 2018) but not the case at the whole aquifer scale (Santoni et al., 541 2018).

542 The SGD seepage velocities from radon and from the Fick law are also in very good 543 agreement with values of groundwater Darcy velocity within the Bonifacio aquifer 544 from combining analytical solution with geochemical and dating tracers (Table 5). 545 Considering a steady-state Dupuit-type flow in the Saint Julien Valley, the analytical solution of Chesnaux et al. (2018) applied between the sampling points EB-B1 and 546 547 ORE-W2 and assuming groundwater transit time of 30 years, an effective porosity of 7%, and a hydraulic conductivity from 1.3 to 3.0 \times 10⁻⁴ m/s, the recharge was 548 549 calculated to be of 115 mm/year i.e. 23 % of the annual rainfall. This value is in very 550 good agreement with the aquifer functioning and with the 132 mm/year (26%) 551 obtained previously for this aquifer using 8 other methods by Santoni et al. (2018). 552 This highlights the strong relationship of groundwater flow velocity with groundwater 553 residence time, effective porosity, permeability and recharge for this aquifer. From 554 this steady-state Dupuit-type flow in the Saint Julien Valley, the Darcy groundwater 555 velocity computed considering the permeability range give values between 0.46 and 556 1.06 m/day, that is in the same order of value for the SGD flows seepage velocity calculated from ²²²Rn and from the Fick law, allowing the SGD seepage velocities 557 558 from radon to be considered as robust.

559 **4.6.SGD** seepage flow and water balance quantification of the Bonifacio

560

Mediterranean porous aquifer

561 As shown through TIR images and in situ measurements, the aquifer outlet 562 generates diffuse outflow in the Bonifacio Harbour and within the Sant'Amanza Bay 563 including the Stentinu area. The computed SGD seepage flow velocities for surface 564 units of 1 m² being now considered as robust, SGD seepage flow rates can be 565 computed by multiplying SGD seepage flow velocities with the discharge area at the 566 outlets. The computation of SGD seepage flow rates assumes an outlet width of 140 567 m and 1.4 m depth in the Bonifacio Harbour, a outlet of 350 m width and 1.5 m depth 568 in the Sant'Amanza bay and a outlet of 70 m width and 1.5 m depth in the Stentinu. The ²²²Rn seepage velocity considered is 2.24 m/d for the Bonifacio Harbour, 1.84 569 570 m/d in the Sant'Amanza Bay and 18.60 m/d in the Stentinu. The total seepage flow 571 rate obtained is then 38.9 L/s, with 5.1 L/s in the Bonifacio Harbour, 11.2 L/s in 572 Sant'Amanza and 22.6 L/s in the Stentinu (including the Canali River flow).

573 The SGD seepage flow rates can now be related to the hydrogeological functionning 574 of the Bonifacio aquifer. Considering a recharge about 132 mm/y in a 33 Km² 575 watershed, i.e. 138 L/s (Santoni et al., 2018) and a public water supply about 174,638 m³/y, *i.e.* 5.5 L/s (Santoni, 2016), the expected SGD seepage flow rate is 576 577 expected to be 132.5 L/s. In these conditions, the total 38.9 L/s SGD seepage flow 578 rate obtained represents 29 % of the expected flow, *i.e.* 28 % of the aquifer recharge 579 rate. According to this observation, 68 % of the recharge water are missing at the 580 identified aquifer outlets.

Assuming that the aquifer recharge value is robust (Chesnaux *et al.*, 2018; Santoni *et al.*, 2018), many hypothesis may be formulated on this result : (1) the field campaign corresponds to a low water level period, underestimating the SGD seepage flow rate

584 due to a loss of water pressure within the aquifer ; (2) due to diffuse SGD flow 585 conditions, the major part of the SGD flows is not detected ; (3) the major part of the 586 recharge water in the upper aquifer level supplies the lower aquifer level by downward drainance. The first hypothesis can be rejected because ²²²Rn 587 588 measurements have been performed in the Bonifacio Harbour during October 2012 589 at low water level and during January 2014 at high water level without this causing 590 any major change in the SGD seepage flow velocities (2.24 m/d in Oct. 2012 and 591 3.19 m/d in Jan. 2014; Table 4) that is in relation with the known inertial behavior of 592 the aquifer (Santoni, 2016). The second hypothesis cannot be refuted as it stands but 593 the third hypothesis seems to be the most probable. Indeed, according to Santoni et 594 al. (2016a), the recharge of the lower aquifer by downward drainance is estimated by a combined $H_2^{18}O - {}^{86}Sr/{}^{87}Sr$ approach to be 65 – 70 %. This observation implies 595 596 that the outlet of the lower aquifer is different from those of the upper one. The first 597 should be governed by the paleomorphology of the underlying fractured granites 598 whereas the second one may be directly dependant of the present surface 599 morphology organized in valleys WSW-ENE oriented explaining the Bonifacio 600 Harbour and Sant'Amanza outlets. The outlet(s) of the lower aquifer is(are) probably 601 located further offshore and at greater depth, which explains its(their) non-detection 602 during the survey at 1 m depth performed for this study.

The SGD seepage flow being now solved, it is possible to propose a water balance budget for the whole Bonifacio Mediterranean porous aquifer (Figure 7). From an aquifer recharge rate about 132 mm/y, 32 % of the recharge water flows within the upper aquifer until the Bonifacio Harbour (4 %), the Sant'Amanza Bay (8%) and until the Stentinu as SGD flows and river flow (16 %). The public drinking water supply also represents an outlet to the upper aquifer (4%). Finally, 68 % of the aquifer

recharge rate supply the lower aquifer level by downward drainance from the upper
aquifer. This water balance then validate and improve the conceptual
hydrogeological functioning of the Bonifacio aquifer.

612 From a management point of view, the detection and localization of SGD flows 613 suggest a natural behavior of the aquifer excluding major over-pumping and 614 seawater intrusion despite the limited potential of recharge, the large interface with 615 the sea, the touristic pressure and the changes in the climate. This seems to be 616 related to a low withdrawal pressure yet exerted on the aquifer as well as to a 617 predominant focused recharge process implying a fast transfer of rainwater to the 618 water table that may be favored by the increase in high rainfall events due to climate 619 change in the Mediterranean (Hartmann et al., 2017; Santoni et al., 2018).

620

621 5. Conclusions

622 This study allowed to validate and improve the conceptual hydrogeological model of 623 a Mediterranean porous aquifer with low potential of recharge and high vulnerability 624 due to a large interface with the sea, the tourism pressure and the changes in the 625 climate. The experimental approach coupling TIR, T, E.C., majors ions, selected trace elements, ²H, ¹⁸O, ²²²Rn, ²²³Ra and ²²⁴Ra proved to be relevant in the detection 626 627 and quantification of low and diffuse SGD flows, highlighting guite natural flow 628 conditions and excluding any major over-pumping. The acquisition of TIR images is 629 able to detect slight temperature variations at the seawater surface. Such imagery 630 nevertheless loses effectiveness in windy areas. The *in situ* geochemical and isotopic 631 investigations allows ensuring the presence of sufficient radon and radium activities 632 in groundwater to detect SGD flows along the seashores. Favourable lithology and 633 strong water-rock interactions involved by long groundwater residence time favours

634 the radon content in coastal aquifer outlets. Most of the time, the lower is the 635 temperature and the E.C., the higher are the radon and radium activities. The SGD flow velocity required for flow rates calculation from ²²²Rn is validated with 636 637 groundwater Darcy velocity in the aquifer. The guantification of SGD flows rates 638 revealed in very good agreement with the known recharge rates and flow conditions 639 within the aquifer. Such a methodology could be applied to other similar coastal aquifers with low and diffuse SGD flow conditions worldwide. For the first time in the 640 641 Mediterranean or in semi-arid context, a natural tracing experiment based on 642 geochemistry and isotope hydrology tools allowed the quantification of SGD flows to 643 validate the hydrogeological conceptual model and to solve the water balance at the 644 whole aquifer scale. This study finally helped to better define the scope in terms of 645 detection limits for TIR images associated with *in situ* geochemical and isotopic tools 646 in the detection of low and diffuse SGD flows.

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Figure 2 : Airborne TIR images of the study site's coastlines (on the 9th September 2011).





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Figure 4 : measured T, E.C., and ²²²Rn activity along the coastlines (January 2014).



Figure 5 : box plots of E.C. (B) values vs activity classes of ²²²Rn from all the coastline survey. Mean E.C. values are also represented with blue squares as well as their corresponding regression line in blue dotted line for each activity class of ²²²Rn.





Figure 6:



Figure 7: 222Rn, 224Ra and 223Ra activities from the 6 fixed measurement carried out within the Bonifacio Harbor as function of distance



947 Tables

	т	рН	EC	HCO 3	CI	NO ₃	SO ₄	Na	к	Mg	Ca	SiO ₂	U	Sr	δ²H	δ ¹⁸ Ο
	°C		µS/c m	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µg/L	µg/L	‰	‰
CPO-B10	16. 8	6.7 3	2040	478	374. 5	37.6	55.9	170.0	3.9	21.8	213.1	12.7	1.9	297.4	-36.9	-5.96
ARA-W4	18. 4	7.0 3	1200	409	163. 0	9.8	28.6	88.3	2.6	12.3	140.8	10.0	0.6	181.3	-38.2	-6.32
CANR-R1	14. 0	7.6 4	1160	351	181. 4	1.1	46.1	98.0	2.8	15.6	128.8	13.7	2.8	216.0	-36.0	-5.97
CUC-B6	18. 0	7.2 9	1280	357	205. 5	5.5	28.1	113.3	2.6	15.8	116.6	10.5	0.5	189.9	-35.6	-5.84
EB-B1	16. 6	6.9 1	1650	432	278. 7	28.9	58.8	147.3	3.3	27.0	157.0	10.2	1.5	410.4	-36.3	-5.77
ORE-W2	17. 5	7.0 7	990	338	132. 3	21.3	25.3	65.5	1.8	9.3	122.1	11.2	0.7	138.6	-38.2	-6.17
GEN-W3	20. 7	6.8 0	1082	396	137. 9	7.2	32.9	70.1	3.7	11.0	140.7	16.9	0.7	196.9	-37.7	-6.24
REP-B18	16. 5	6.8 8	1215	387	174. 1	8.4	43.2	108.2	3.2	14.1	121.3	16.1	2.8	237.1	-38.0	-6.15
VB-B5	16. 9	7.4 7	1066	395	140. 3	0.3	0.5	77.0	3.1	13.2	104.2	9.0	0.0	286.0	-37.9	-6.16

949Table 1: physicochemical parameters, major ions, selected trace elements and stables isotopes of the water950molecule of the stream (CANR-R1) and groundwater samples (October 2012).

	^{222Rn} (Bq/	m3)	^{224Ra} (Bo	q/m3)	^{223Ra} (Bq/m3)	
		Std.		Std.		Std.
CPO-B10	10823	173	33.9	2.0	1.4	0.3
ARA-W4	6081	133	13.3	0.8	0.6	0.1
CANR-R1			10.7	0.9	0.1	0.0
CUC-B6	3528	95	15.7	1.3	1.0	0.1
EB-B1	4092	97	28.3	1.7	1.5	0.3
ORE-W2	3393	89	16.3	0.9	0.4	0.1
GEN-W3	3714	93	15.2	1.3	1.1	0.1
REP-B18	27853	340	34.3	1.6	0.5	0.1
VB-B5	1199	68	8.8	0.5	0.1	0.0

956 Table 2 : Radon and Radium radionuclides activity of stream (CANR-R1) and groundwater (October 2012).

958 Table 3: Radon activities for 6 sand sediment samples spread all over the study area (January 2014).

Beach sediment sample	²²² Rn (Bq/m ³)		
		Std.	
Canettu	2.3	0.6	
Sant'Amanza	3.1	0.5	
Paraguano	3.5	0.7	
San Mulari	6.2	1.0	
Pertusato	1.4	0.8	

Table 4 :

	Bonifacio Harbour	Sant'Amanza Bay		
	²²² Rn	²²³ Ra	²²⁴ Ra	²²² Rn
Distance vs. logarithm activities regression line equation	y = -0.0013x + 4.1722	y = -0.0013x + 1.8632	y = -0.0016x + 0.5507	y = -0.002x + 5.5393
Isotope half-lifes (day)	3.86	11.8	3.6	3.86
lsotope lambda (day ^{⋅1})	0.180	0.059	0.193	0.180
Computed seawater mixing coefficients, D _{eddy} (m²/s)	1.23	0.40	1.32	1.84
Distance vs. activites exponential regression equation	$y = 57.6776e^{-0.0012x}$	$y = 0.4553e^{-0.0010x}$	y = 5.3360e ^{-0.0012x}	y = 254.497e ^{-0.00199x}
Isotope horizontal inventories for 1 m^2 of water parcel flowing from A to F (\mbox{Bq}/\mbox{m}^2)	42268	380	3934	110954
Isotope fluxes from groundwater input in the harbor (Bq/m²/day)	7590	22	757	19924
isotope activities in groundwater, C_{GW} (Bq/m ³)	3393	0.4	16.3	10823
Computed SGD seepage velocities (m/day)	2.24	55.81	46.47	1.84
Computed SGD seepage flow (L/s)	2.5	63.3	52.7	24.6