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1 Temporal variability of lagoon-sea water exchange and seawater circulation through a  
2 Mediterranean barrier beach

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## 32 **ABSTRACT**

33 The subterranean flow of water through sand barriers between coastal lagoons and the sea, driven  
34 by a positive hydraulic gradient, is a net new pathway for solute transfer to the sea. On the sea-  
35 side of sand barriers, seawater circulation in the swash-zone generates a flux of recycled and new  
36 solutes. The significance and temporal variability of these vectors to the French Mediterranean  
37 Sea is unknown, despite lagoons constituting ~50% of the coastline. A one-dimensional  
38  $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra}$  reactive-transport model was used to quantify water flow between a coastal lagoon  
39 (La Palme) and the sea over a six-month period. Horizontal flow between the lagoon and sea  
40 decreased from ~85 cm d<sup>-1</sup> during May 2017 (0.3 m<sup>3</sup> d<sup>-1</sup> m<sup>-1</sup> of shoreline) to ~20 cm d<sup>-1</sup> in July  
41 and was negligible in the summer months thereafter due to a decreasing hydraulic gradient.  
42 Seawater circulation in the swash-zone varied from 10 – 52 cm d<sup>-1</sup> (0.4 – 2.1 m<sup>3</sup> d<sup>-1</sup> m<sup>-1</sup>), driven  
43 by short-term changes in the prevailing wind and wave regimes. Both flow paths supply minor  
44 DSi fluxes on the order of ~3 – 10 mmol Si d<sup>-1</sup> m<sup>-1</sup>. Lagoon-sea water exchange supplies a net  
45 DIC flux (320 – 1,100 mmol C d<sup>-1</sup> m<sup>-1</sup>) two orders of magnitude greater than seawater circulation  
46 and may impact coastal ocean acidification. The subterranean flow of water through sand barriers  
47 represents a significant source of new DIC, and potentially other solutes, to the Mediterranean  
48 Sea during high lagoon water-level periods and should be considered in seasonal element  
49 budgets.

## 50 INTRODUCTION

51 Subterranean water exchange between coastal lagoons and the coastal ocean remains  
52 severely understudied, despite coastal lagoons representing ~13% of the global coastline (Barnes  
53 1980; Santos et al. 2012). This underground lagoon-sea water exchange depends on the hydraulic  
54 gradient between the two water bodies (pressure) and the properties of the barrier which separates  
55 them, including sediment porosity and permeability. Separately, tides and waves can drive large  
56 volumes of seawater into permeable beaches, resulting in a seawater circulation cell (Li et al.  
57 1999; Robinson et al. 2007; Xin et al. 2010). Pore waters from either flow path can obtain a  
58 unique geochemical signature from early diagenetic nutrient mineralization (Anschutz et al.  
59 2009; Santos et al. 2009; Charbonnier et al. 2013; Goodridge and Melack 2014) and/or sediment  
60 weathering (Schopka and Derry 2012; Ehlert et al. 2016). Lagoon-sea water exchange and  
61 seawater circulation flow paths are therefore a potential source of both recycled and new  
62 chemical elements to the coastal ocean; thus, it is necessary to understand the time-scale of each  
63 flow path to properly evaluate chemical fluxes (Tamborski et al., 2017). Each of these flow paths  
64 may be characterized as a type of submarine groundwater discharge (SGD), broadly defined as  
65 “any and all flow of water on continental margins from the seabed to the coastal ocean, regardless  
66 of fluid composition or driving force” (Burnett et al. 2003).

67 Lagoon-sea water exchange has been documented along sandy barrier beaches in Fire  
68 Island, New York (USA) (Bokuniewicz and Pavlik 1990), Florida Bay (USA) (Corbett et al.  
69 2000; Chanton et al. 2003), Sapelo Island, Georgia (USA) (Evans and Wilson 2017), Malibu Bay,  
70 California (USA) (Dimova et al. 2017), Venice Lagoon (Italy) (Rapaglia et al. 2010), Patos-  
71 Mirim Lagoon (Brazil) (Windom et al. 2006; Niencheski et al. 2007) and the Great Barrier Reef  
72 (Australia) (Santos et al. 2010). Chemical fluxes associated with this SGD have only been

73 reported for Malibu Bay, Venice Lagoon, Patos-Mirim Lagoon and the Great Barrier Reef  
74 (Windom et al. 2006; Niencheski et al. 2007; Rapaglia et al. 2010; Santos et al. 2010; Dimova et  
75 al. 2017). All of these study sites are situated along areas with significant tidal ranges to produce  
76 water level, and therefore pressure, variations. For example, the tidal range of Venice lagoon  
77 (neap = 0.3, spring = 1.2 m), situated along the northern Adriatic Sea, was found to drive SGD  
78 flow rates up to  $140 \text{ cm d}^{-1}$  through highly permeable paleochannels (Rapaglia et al. 2010).  
79 However, little to no information exists on the magnitude and temporal variability of this lagoon-  
80 sea water exchange flow path to microtidal coastal environments. This is the case of the French  
81 Mediterranean coastline, where tides are insignificant ( $\sim 20 - 30 \text{ cm}$ ) and there is a striking  
82 succession of coastal lagoons of various sizes that are separated from the sea by only narrow sand  
83 barriers, constituting  $\sim 50\%$  of the coastline (Stieglitz et al. 2013).

84 Seawater circulation through permeable sandy beaches is primarily driven by tides and  
85 waves (Li et al. 1999; Santos et al. 2012) and may be regarded as a net zero water flux. Such a  
86 process can induce large volumetric fluxes of circulated seawater in high-energy environments,  
87 as observed for example, in Long Island Sound, NY (Tamborski et al., 2017) and the French  
88 Aquitanian Coast (Charbonnier et al. 2013). Tides are minor along the French Mediterranean Sea,  
89 such that wave-setup and water level fluctuations are hypothesized to drive a net-seaward flowing  
90 circulation cell beneath the swash-zone (Sous et al. 2016). The temporal variability of seawater  
91 circulation as a vector for solute transport to the French Mediterranean Sea is largely unknown  
92 (Tamborski et al. 2018).

93 The suite of naturally occurring Ra isotopes ( $^{223}\text{Ra} = 11.4 \text{ d}$ ,  $^{224}\text{Ra} = 3.66 \text{ d}$ ,  $^{226}\text{Ra} = 1,600$   
94  $\text{y}$ ,  $^{228}\text{Ra} = 5.75 \text{ y}$ ) and  $^{222}\text{Rn}$  (3.83 d) have unique half-lives, which enable tracing pore water flow  
95 paths of varying time-scales (Rodellas et al. 2017). These radionuclides are supplied to pore

96 fluids by the decay of their surface-bound parent isotope through alpha-recoil, and are thus  
97 dependent upon sediment grain-size, U/Th content (Porcelli and Swarzenski 2003; Porcelli 2008),  
98 and Fe/Mn-oxide surface coatings (Dulaiova et al. 2008; Beck and Cochran 2013). Ra isotopes  
99 are primarily adsorbed onto sediment surfaces at low ionic strengths and are partially released  
100 into solution at higher ionic strengths due to surface-competition displacement from cations  
101 (Webster et al. 1995; Gonnee et al. 2013). Radionuclides can be used in simplified systems to  
102 estimate one-dimensional advection or diffusion (horizontal or vertical), when the above-  
103 mentioned variables are well-constrained (Krest and Harvey 2003; Smith et al. 2008; Michael et  
104 al. 2011).

105         In this study, we use the term “lagoon-sea water exchange” to refer to the lateral transport  
106 of pore waters through a permeable barrier beach, mainly driven by water level differences across  
107 the barrier. Separately, we use the term “seawater circulation” to refer to coastal Mediterranean  
108 seawater which has been driven into permeable sediments by physical forcing mechanisms,  
109 including waves. Using radionuclide tracers and physical observations, we assess the relative  
110 magnitude and monthly temporal variability of (1) lagoon-sea water exchange through the barrier  
111 beach and (2) seawater circulation through the swash-zone. In addition, we provide a first-order  
112 approximation of solute fluxes (DIC, DSi, Ba) driven by these two separate processes.

### 113 **Study Site**

114         La Palme lagoon is a small coastal lagoon (~500 ha) situated along the French  
115 Mediterranean coastline, adjacent to the barrier beach La Franqui (**Figure 1**). The southern basin  
116 of La Palme lagoon is a shallow sandy basin (20 – 50 cm water depth) that has limited surface  
117 water exchange with the northern basin of the lagoon through a constructed railway dike and  
118 bridge and is often partially dry during the summertime. The lagoon may exchange with the

119 Mediterranean Sea via a small inlet which is naturally opened from high-energy storm-events.  
120 Tides in this region are minor (~20 cm), such that when the inlet is closed, water level differences  
121 between the southern basin of La Palme lagoon and the Mediterranean Sea are mainly driven by  
122 changes in precipitation and evaporation. Precipitation is highest during the fall and spring (440  
123 mm total in 2017); evaporation rates typically exceed precipitation rates during the summer  
124 (Stieglitz et al. 2013). The beach sediment of La Franqui is a well-sorted medium sand, with a  
125 mean grain-size between 200 – 500  $\mu\text{m}$ .

## 126 **METHODS**

### 127 **Field Methods**

128 The inlet to the Mediterranean Sea was closed months prior to the start of sampling. A  
129 shore-perpendicular transect (T1) was sampled monthly between the southern basin of La Palme  
130 lagoon and the Mediterranean Sea during May – September 2017 (**Figure 1**). Transect T1 was  
131 additionally sampled in November 2017, four days after the beach was breached by a storm-  
132 event, which resumed surficial exchange between the lagoon and the sea. A second transect (T2)  
133 was sampled during May 2017 only. Samples along the first transect were evenly spaced 10 m  
134 apart; samples from the second transect (~250 m north) were evenly spaced 20 m apart to assess  
135 long-shore spatial variability (**Figure 1**).

136 During May, transects were sampled from boreholes that intersected the saturated zone of  
137 the beach; the interstitial water that filled the borehole was immediately sampled. For the  
138 remaining sampling campaigns (June – November), pore waters were collected using a shielded-  
139 screen (Mesh #50 = 297  $\mu\text{m}$ ) drive-point piezometer (Charette and Allen 2006) attached to a  
140 vacuum hand pump. Samples were acquired at depth intervals of ~50, 100 and 150 cm below

141 grade (150 cm for select stations only). Boreholes were dug adjacent to each piezometer sampling  
142 location to measure the depth to the water table for all sampling events. During each campaign,  
143 lagoon water and Mediterranean Sea surface water endmembers were sampled directly using a  
144 hand pump. Salinity, temperature, pH and dissolved oxygen (DO) were immediately measured  
145 for all samples in the field using a calibrated WTW hand-probe (Multi 3430). Bulk sediment  
146 samples were collected from several boreholes during May 2017 (~50 cm depth). Shore-  
147 perpendicular beach topography was surveyed in June 2017 using a real time kinematic GPS  
148 (Trimble). We assume that the June 2017 surveyed topography is representative of May, July,  
149 August and September, a period in which storms (and thus beach erosion and accretion) were  
150 minimal. Beach topography was not surveyed in November 2017.

## 151 **Analytical Methods**

### 152 *Sediment*

153 Sediment samples were dried at 80 °C for 48 h and water content loss was measured  
154 (triplicate measurements) to calculate sediment porosity, assuming a mineral grain density of  
155 2.65 g cm<sup>-3</sup> and corrected for sea salt. Bulk sediment samples were sealed for >3 weeks and  
156 counted on a semi-planar Canberra Ge detector at 352 keV (<sup>214</sup>Pb), 583 keV (<sup>208</sup>Tl) and 911 keV  
157 (<sup>228</sup>Ac) to determine the total (surficial + lattice-bound) activity of solid-phase <sup>226</sup>Ra, <sup>228</sup>Th and  
158 <sup>228</sup>Ra, respectively. IAEA 300 and NIST Material 4350B sediment were used to determine the  
159 semi-planar detector counting efficiencies.

160 Sediment samples were additionally measured in a radium delayed coincidence counter  
161 (RaDeCC) system (Moore and Arnold 1996), to determine the amount of surface-exchangeable  
162 <sup>223</sup>Ra and <sup>224</sup>Ra produced by <sup>227</sup>Th and <sup>228</sup>Th present on sediment surfaces (Sun & Torgersen,  
163 1998b). The number of alpha-decays to produce Ra and Rn isotopes are different; this can result



164 in a slightly different rate of recoil supply of Ra ( $\epsilon$ ) to that of Rn. In the ensuing analyses, we  
165 assume  $\epsilon = 1.0$  (Krishnaswami et al. 1982, 1991). Therefore, we assume that all  $^{219}\text{Rn}$  and  $^{220}\text{Rn}$   
166 diffused into the circulating He gas loop of the RaDeCC system are produced by  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$   
167 decay on sediment surface-exchangeable sites. Radon emanation was measured on the sediment  
168 using the column cartridges traditionally used for Mn-fiber measurements. Using the traditional  
169 column cartridge, we are able to measure a larger volume of sediment ( $>150$  g) in comparison to  
170 the chamber developed by Cai et al. (2012). The disadvantage of the traditional cartridge is that  
171 its cylindrical geometry is only suitable for sandy sediment, so that the circulating He gas flow is  
172 unobstructed. Fine-grained sediment (silts, clays) could impede the He flow, making this method  
173 unusable. Bulk sediment samples were partially dried in an oven until a water:sediment ratio of  
174 approximately 0.1 was reached, a ratio previously determined to be sufficient for counting  
175 sediment samples on the RaDeCC (Cai et al., 2012; Sun & Torgersen, 1998a). Cartridges were  
176 plugged on each end with raw acrylic fiber to prevent any sediment from entering the system.  
177 The He flow rate was continuously monitored throughout the measurement period and matched  
178 that of typical Mn-fiber measurements ( $\sim 6$  L  $\text{min}^{-1}$ ). A replicate set of homogenized sediment ( $n$   
179 = 8) was spiked with 3.8 – 57 dpm  $^{224}\text{Ra}$  in equilibrium with  $^{228}\text{Th}$  (standard addition; triplicate  
180 measurements) to determine the detector counting efficiency (Cai et al., 2012; **Figure 2**).  
181 Sediment column  $^{224}\text{Ra}$  counting efficiency was equivalent to that of Mn-fiber measurements  
182 (Sun & Torgersen, 1998b) and therefore we assume the same detector efficiency as Mn-fiber  
183 measurements for both  $^{224}\text{Ra}$  and  $^{223}\text{Ra}$ . The importance of sample load was tested to determine  
184 the effect of helium “channeling” (Cai et al. 2012), with varying sediment loads of 29 – 167 g ( $n$   
185 = 7; triplicate measurements).

186 Ra distribution coefficient ( $K_D$ ) experiments were run on sediment samples from two  
187 locations (5-PW5-A and 5-2-PW3-A) to determine the Ra partition coefficient ( $K$ ) for water  
188 salinities of 27 and 38 (representative of salinities measured in beach pore waters), following  
189 established protocols for such measurements (e.g. Fisher et al. 1983; Roberts et al. 2009). Briefly,  
190 these involve suspending the material of interest (in this case, beach sand) in a solution  
191 containing the element for which the distribution coefficient is to be determined, under conditions  
192 of in-situ pH and salinity. After equilibration, generally within ~48 h, the fractions of tracer on  
193 the sediments and in solution are measured. Sediments were first gamma counted (as noted  
194 above) to determine the initial activity of  $^{226}\text{Ra}$  from the 352 keV  $^{214}\text{Pb}$  photopeak. For each  
195 sediment sample, a  $^{226}\text{Ra}$  tracer (~20 dpm) was added to 100 mL of Ra-free water containing ~11  
196 g (salinity of 27) or ~20 g of sediment (salinity of 38). The pH of each solution was ~7.5 after  
197 addition of the  $^{226}\text{Ra}$  tracer and no adjustment was made to alter pH for the experiments. Samples  
198 were allowed to equilibrate for ~5 days with frequent stirring. At the end of the experiment, the  
199 water was decanted off and the sediment was rinsed with small portions of distilled water to  
200 remove any residual tracer solution. The sediment was dried and recounted to determine the new  
201  $^{226}\text{Ra}$  activity. The activity of  $^{226}\text{Ra}$  adsorbed onto the sediment was calculated as the excess  
202  $^{226}\text{Ra}$  (above the intrinsic  $^{226}\text{Ra}$  activity in the sample) multiplied by the mass of sediment used in  
203 the experiment (Eq. 1).  $K_D$  was calculated as:

$$204 \quad K_D = \left[ \frac{\text{dpm } ^{226}\text{Ra adsorbed}}{\text{dpm } ^{226}\text{Ra in solution}} \right] / C_p \quad (\text{Eq. 1})$$

205 where  $C_p$  = g sediment per mL of solution.  $^{226}\text{Ra}$  was used for these experiments and because the  
206 sediments contained natural  $^{226}\text{Ra}$ , this fraction was subtracted to determine the partitioning of Ra  
207 between solution and solid at the end of the experiment. The solid-phase at the start of the

208 experiment comprised several fractions of “natural”  $^{226}\text{Ra}$ : 1) in the mineral structure, 2)  
209 originally adsorbed onto the sediments and 3) originally dissolved in wet sediment pore water  
210 (and subsequently adsorbed onto the sediments after drying). All three fractions were effectively  
211 measured via the gamma spectrometric measurements of  $^{226}\text{Ra}$  before the start of the sorption  
212 experiments.

213 Radium and radon equilibration experiments were also conducted to estimate the  
214 production and partitioning of  $^{223}\text{Ra}$ ,  $^{224}\text{Ra}$  and  $^{222}\text{Rn}$  from sediments. For  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$ , ca.  
215 1,000 g of sediment was added into a 6 L plastic container, to which ca. 400 mL of water was  
216 added, for a water salinity of 27 and 38 (representative of salinities measured in beach pore  
217 waters). The containers were sealed and after >7 weeks the wet sediment was vacuum filtrated to  
218 isolate the pore water from the sediment.  $\text{MnO}_2$  coated acrylic fibers were subsequently  
219 introduced to the extracted solution, partially rinsed, and analyzed on a RaDeCC system, as  
220 previously described. For  $^{222}\text{Rn}$ , ca. 200 g of dry sediment was added into a 500 mL gas tight  
221 bottle, which was then completely filled with Ra-free lagoon water (salinity of 38; triplicates of  
222 the same sediment sample were conducted). The bottles were hermetically sealed for >1 month  
223 and periodically shaken. The  $^{222}\text{Rn}$  concentration in water was measured using the RAD7  
224 coupled to the gas extraction accessory for bottles and corrected to the specific ratio of pore  
225 water/solids in sediments following Stieglitz et al. (2013).

### 226 *Surface & Pore Waters*

227 Between 0.5 – 2 L of pore water was collected for Ra analysis via vacuum hand-pump  
228 into plastic containers, for all sampling dates. The plastic containers were left for several hours  
229 (or overnight) to allow particles to settle to the bottom of the container. Afterwards, the water  
230 overlying the particles was decanted off into a graduated cylinder and volume was recorded; this

231 water was then filtered through raw acrylic fiber (Mn-free) to further remove any particles. In the  
232 laboratory, hypoxic ( $\text{DO} < 3 \text{ mg L}^{-1}$ ) pore water samples were aerated for ~20 minutes; after  
233 aeration, Mn-fibers were added directly to the water sample and periodically stirred ( $< 1 \text{ hr}$ ) to  
234 quantitatively adsorb all Ra from the solution. Two water samples were subjected to a second  
235 Mn-fiber addition; these second fibers did not produce any counts (above background) and thus  
236 we assume a 100% initial Ra yield. Large volume ( $> 10 \text{ L}$ ) endmember samples were  
237 traditionally filtered through cartridges ( $< 1 \text{ L min}^{-1}$ ). Mn-fibers were triple-rinsed, partially dried  
238 (Sun & Torgersen, 1998a) and counted using a RaDeCC to measure the short-lived  $^{224}\text{Ra}$  isotope;  
239 samples were counted again one week after collection to determine  $^{223}\text{Ra}$  (Moore and Arnold  
240 1996). Mn-fibers were recounted approximately three weeks after sample collection to measure  
241  $^{228}\text{Th}$ , to calculate excess  $^{224}\text{Ra}$  (hereafter denoted  $^{224}\text{Ra}_{\text{ex}}$ ). Detector efficiencies were determined  
242 using a  $^{232}\text{Th}$  and  $^{227}\text{Ac}$  standard; analytical counting uncertainties were calculated by standard  
243 rules of error propagation and the results were decay corrected back to the sample collection time  
244 (Garcia-Solsona et al. 2008).

245 For  $^{222}\text{Rn}$  analysis, pore waters from May, June and November were collected into 250  
246 mL air-tight bottles, which were coupled to the piezometer tubing to minimize water-air contact,  
247 via vacuum hand-pump. Lagoon and Mediterranean Sea endmembers were collected in 2-L air-  
248 tight bottles using a small submersible pump and submerging the bottle beneath the water surface  
249 to minimize gas loss.  $^{222}\text{Rn}$  samples were analyzed using the radon-in-air monitor RAD7 coupled  
250 to a gas extraction accessory for bottles (DurrIDGE Co.) and decay corrected back to the sample  
251 collection time; uncertainties are presented as  $\pm 1$  standard deviation.

252 Water samples for solute and carbon measurement were syringe filtered using  $0.45 \mu\text{m}$   
253 cellulose acetate filters into rinsed 60 mL PDPE vials. Water samples were diluted 10x with

254 deionized water and analyzed for dissolved Ba and Si using an ICP-OES (Horiba Jobin Yvon  
255 Ultima2®); analytical precision is  $\leq 10\%$ . Dissolved inorganic carbon (DIC) and dissolved  
256 organic carbon (DOC) were analyzed using a Shimadzu® and results are expressed in molar  
257 quantities of carbon; analytical precision is  $\leq 5\%$ .

## 258 **Ancillary Parameters**

259 To investigate the driving mechanisms of seawater circulation through swash-zone, we  
260 obtained (1) hourly wind data (speed and direction) from the nearby meteorological station  
261 “Leucate” from Météo France, the French meteorological service; and (2) wave sensor data (half-  
262 hour intervals) from the nearby buoy “Leucate” (ID 01101) from CEREMA. Wind and waves  
263 contribute to local sea-levels and lead to a dynamic interaction between seawater and pore waters.  
264 In this study we separate total water levels at the coast into wind and wave contributions as  
265 respectively wind-driven setup and wave run-up at the beach. The wind setup formulation ( $S_{wind}$ )  
266 (Dean and Dalrymple 2004) relates measured wind speeds, direction relative to the shore-normal  
267 and distance over which the wind blows (fetch) to a local increase in water levels (Eq. 2)  
268 presuming that the wind is present for a long enough time to reach an equilibrium state.

$$269 \quad S_{wind} = h * \left( \sqrt{1 + \frac{A_s x}{L}} - 1 \right) \quad (\text{Eq. 2})$$

270 In which  $A_s$  is

$$271 \quad A_s = \frac{2n\tau_s L}{\rho_w g h^2} \quad (\text{Eq. 3})$$

272 Where

$$273 \quad \tau_s = \rho_w c_t W^2 \quad (\text{Eq. 4})$$

274 In (Eq. 2), (Eq. 3) and (Eq. 4),  $h$  is the mean water depth over the fetch length  $L$ ,  $x$  is the position  
275 along the fetch (in this case  $x = L$  since we are at the coast),  $A_s$  is a dimensionless parameter

276 containing  $n$ , in which  $n = 1 - (\tau_b / \tau_s)$  to account for bottom ( $\tau_b$ ) and wind shear stress ( $\tau_s$ ). Here  $n$   
 277 is set as 1.15,  $\rho_w$  represents water density,  $g$  is the gravitational acceleration,  $c_t$  is a friction  
 278 coefficient, and  $W$  is the wind speed (shore-normal), usually measured at 10 m elevation.

279 The wave contribution to total sea level at the coast is calculated using a statistical bulk  
 280 run-up formula (Stockdon et al., 2006). Stockdon et al. (2006) relates offshore measured wave  
 281 conditions such as significant wave height relative to the shore-normal and peak period to a  
 282 statistical value for wave run-up ( $R_{wave}$ ) at the beach (2% exceedance) following:

$$283 \quad R_{wave} = 1.1 \left( 0.35\beta_f \sqrt{H_0 L_0} + \frac{\sqrt{H_0 L_0 (0.563\beta_f^2 + 0.004)}}{2} \right) \quad (\text{Eq. 5})$$

284 Wherein  $\beta_f$  is the beach face slope,  $H_0$  is the offshore wave height (shore-normal) and  $L_0$   
 285 represents the offshore wave length. The superposition of the wind and wave contribution results  
 286 in a time-varying total water level.

287 The physical impact of wind and waves on seawater circulation in the swash-zone might  
 288 be far from instantaneous; for example, a certain adaptation time or lag may exist between  
 289 seawater circulation and the occurring wind and wave conditions. To investigate the importance  
 290 of this adaptation time lag, mean wind and wave data variables were binned into seven different  
 291 time-intervals: 2 weeks, 1 week, 72 h, 48 h, 24 h, 12 h and 6 h prior to each sampling. These time  
 292 intervals were chosen to represent the range in time-scale that is traced by short-lived  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$   
 293 and  $^{224}\text{Ra}_{\text{ex}}$ . Binned averages for different wind and wave parameters were compared to  
 294 radionuclide-derived advection velocities (see Discussion) to investigate any potential temporal  
 295 variability caused by wind setup and wave setup. Multiple linear regression analysis for  
 296 uncorrelated variables was performed using XLSTAT 2018.

## 297 RESULTS

### 298 Sediment

299 Total (surficial and lattice-bound)  $^{226}\text{Ra}$  (0.50 – 0.58 dpm g<sup>-1</sup>),  $^{228}\text{Ra}$  (0.41 – 0.66 dpm g<sup>-1</sup>),  $^{228}\text{Th}$  (0.56 – 0.79 dpm g<sup>-1</sup>) and sediment porosity ( $0.38 \pm 0.1$ ) showed little variability across  
300 the beach transect (**Table 1**). RaDeCC detectors exhibited a linear increase in  $^{224}\text{Ra}$  cpm with  
301 increasing  $^{224}\text{Ra}$  added (solution spike and sediment mass), suggesting that the RaDeCC is  
302 suitable for measuring the surface-exchangeable Ra of beach sands (**Figure 2**). Surface-  
303 exchangeable Ra was similar between sediment samples, with a mean ( $\pm$  STD; four replicate  
304 measurements) activity of  $0.0025 \pm 0.0002$  dpm g<sup>-1</sup> (n=4) for  $^{223}\text{Ra}$  and  $0.041 \pm 0.006$  dpm g<sup>-1</sup>  
305 (n=4) for  $^{224}\text{Ra}$  (**Table 2**). Thus, as compared to the bulk  $^{228}\text{Th}$ , the recoil efficiency of  $^{224}\text{Ra}$  into  
306 pore fluid was between ~5 and 7%, as expected for coarse-grained sands (Copenhaver et al. 1993;  
307 Porcelli 2008). We assume that all  $^{222}\text{Rn}$  input is similar across the beach transect, as evidenced  
308 by the solid-phase  $^{226}\text{Ra}$  distribution (**Table 1**). The production rate ( $P$ ; dpm cm<sup>-3</sup>) of Ra was  
309 estimated as  
310

$$311 \quad P = \frac{(1-\varphi)}{\varphi} * \rho_{sed} * Ra_{surf} \quad (\text{Eq. 6})$$

312 Where  $\varphi$  is the sediment porosity (0.38),  $\rho_{sed}$  is the grain density (2.65 g cm<sup>-3</sup>), and  $Ra_{surf}$  is the  
313 activity of surface-exchangeable Ra (dpm g<sup>-1</sup>). For the beach transect samples, the production rate  
314 ( $P$ ) with zero partitioning is equal to  $10.6 \pm 1.0$  dpm L<sup>-1</sup> for  $^{223}\text{Ra}$  and  $176 \pm 25$  dpm L<sup>-1</sup> for  $^{224}\text{Ra}$ .  
315 The resulting theoretical dissolved equilibrium  $^{224}\text{Ra}/^{223}\text{Ra}$  activity ratio is thus  $16.6 \pm 2.8$  and is  
316 likely not varying across the beach transect (**Table 2**). Note that the difference between the above  
317 activities ( $P$ ) and the actual rate of Ra production (dpm L<sup>-1</sup> d<sup>-1</sup>) is simply a function of the Ra  
318 isotope decay constant ( $\lambda$ ).

319 Mean  $K_D$  values equal  $1.7 \pm 0.9 \text{ cm}^3 \text{ g}^{-1}$  at a salinity of 38 (Mediterranean seawater) and  
320  $2.7 \pm 0.7 \text{ cm}^3 \text{ g}^{-1}$  at a salinity of 27 (average of two sediment samples; **Table 1**). Considering  
321 sediment porosity and grain density,  $K$  is equal to  $7.5 \pm 3.9$  at a salinity of 38 and  $11.6 \pm 3.0$  at a  
322 salinity of 27 [ $K = K_D * \{(1-\phi) * \rho_{sed}/\phi\}$ ]. An estimate can be made of the magnitudes of the pore  
323 water  $^{226}\text{Ra}$  and adsorbed  $^{226}\text{Ra}$  included with the sediments based on the determined  $K_D$  values  
324 ( $1.7 - 2.7$ ). For a porosity of 0.38, there was  $0.23 \text{ cm}^3_{\text{water}}/\text{g}_{\text{dry sediment}}$ . The experiments used 11 –  
325 20 g of sediment, and the pore water  $^{226}\text{Ra}$  in the beach sand was measured as 74 dpm  $100 \text{ L}^{-1}$ .  
326 Pore water  $^{226}\text{Ra}$  that would have been present in the two sediment aliquots at the start of the  
327 sorption experiments would have been 0.002 and 0.003 dpm, respectively. The in-situ adsorbed  
328  $^{226}\text{Ra}$  would have thus been 0.019 and 0.024 dpm for the 11 and 20 g experiments, respectively.  
329 Thus, the total  $^{226}\text{Ra}$  contributed from dissolved and adsorbed fractions at the start of the  
330 experiment was 0.02 – 0.03 dpm. These are small fractions of the total  $^{226}\text{Ra}$  activity of the solid  
331 phase determined by gamma spectrometry before the  $K_D$  experiments ( $0.5 - 0.6 \text{ dpm g}^{-1}$ , or  $\sim 6 -$   
332  $12 \text{ dpm total}$  contributed by the 11 – 20 g aliquots) and, as noted above, would have been  
333 included in the “starting”  $^{226}\text{Ra}$  used to calculate  $K_D$  after the sorption experiments (**Table 1**). We  
334 also note that the natural “adsorbed”  $^{226}\text{Ra}$  would have contributed at most 10% of the sorbed Ra  
335 at the conclusion of the  $K_D$  experiment, and thus no correction was made for it in calculating  $K_D$ .  
336 The  $K_D$  values measured here ( $1.7 - 2.7$ ) are comparable to that determined for beach sand by  
337 Colbert and Hammond (2008) ( $K_D = 1.6$ ) and Beck and Cochran (2013) ( $K_D = 1.14 - 1.60$ ), using  
338 a different method.

339 Sediment incubation experiments represent an equilibrium Ra activity in pore water  
340 (production = decay) controlled by the ionic strength of the solution (via  $K$ ) and may additionally  
341 include inputs of Ra from sediment dissolution (i.e. weathering). The equilibrium pore water



342  $^{223}\text{Ra}$  activity was equal to  $2.5 \pm 0.3$  and  $3.0 \pm 0.2$  dpm  $\text{L}^{-1}$  for a salinity of 27 and 38,  
343 respectively; equilibrium  $^{224}\text{Ra}_{\text{ex}}$  activity was equal to  $29.0 \pm 3.0$  and  $52.5 \pm 4.0$  dpm  $\text{L}^{-1}$  over the  
344 same salinity interval, and the resulting  $^{224}\text{Ra}/^{223}\text{Ra}$  ratios are thus  $11.6 \pm 1.8$  and  $17.5 \pm 1.8$  for  
345 salinities of 27 and 38, respectively (**Table 3**). Pore water  $^{222}\text{Rn}$  activity in equilibrium with  
346 sediments was  $160 \pm 20$  dpm  $\text{L}^{-1}$  (average of 3 samples).

347 Changes in salinity (ionic strength) drive changes in the partitioning ( $K$ ) of Ra between  
348 dissolved and solid-phases (Webster et al. 1995; Gonnee et al. 2008). The activity ratio of  
349 mobile  $^{224}\text{Ra}/^{222}\text{Rn}$  is approximately equivalent to the ratio of the emanation efficiency of  $^{222}\text{Rn}$   
350 and the recoil efficiency of  $^{224}\text{Ra}$  in fresh groundwater systems when steady-state is achieved  
351 (Porcelli and Swarzenski 2003), under the assumption that all alpha-decay radionuclides are  
352 supplied at similar rates to that of  $^{222}\text{Rn}$  (Krishnaswami et al. 1982).  $^{222}\text{Rn}$  is an inert, noble gas  
353 whose distribution in pore fluid is supplied chiefly by alpha recoil from sediment surfaces, thus  
354  $^{222}\text{Rn}$  is not partitioned between solid and dissolved phases like Ra. Bulk solid-phase  $^{228}\text{Th}/^{226}\text{Ra}$   
355 activity ratios, the parent isotopes of  $^{224}\text{Ra}/^{222}\text{Rn}$ , are between 1.05 – 1.37 (**Table 1**); therefore,  
356 pore waters in equilibrium with sediment surfaces should have  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios within  
357 this range if there is no partitioning of Ra between sediment surfaces and solution. Pore water  
358  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios determined from the equilibration experiments are between  $0.18 \pm 0.03$   
359 for a salinity of 27 and  $0.33 \pm 0.05$  for a salinity of 38 (**Table 3**). Assuming that all  $^{222}\text{Rn}$  is input  
360 at a constant rate and considering a mean surface-exchangeable  $^{224}\text{Ra}$  activity of  $0.041 \pm 0.006$   
361 dpm  $\text{g}^{-1}$  ( $176 \pm 25$  dpm  $\text{L}^{-1}$ ), then the total (dissolved + surface-bound)  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratio is  
362  $1.28 \pm 0.22$  for a salinity of 27 and  $1.43 \pm 0.24$  for a salinity of 38 (**Table 3**), approximately  
363 within the range of the bulk  $^{228}\text{Th}/^{226}\text{Ra}$  activity ratios.

## 364 **General Water Parameters**

365           Precipitation largely occurs during fall and spring months, with little rainfall during the  
366 six-month sampling interval (May – November 2017; **Figure 3**). Total precipitation for the week  
367 prior to each sampling event equaled 3.4, 0.0, 0.2, 0.2, 2.4 and 0.8 mm for May, June, July,  
368 August, September and November, respectively. Wind speed and direction are dominated by two  
369 modes, blowing predominantly from either the sea ( $\sim 90^\circ$ ) towards land or from land ( $\sim 315^\circ$ )  
370 towards the sea (**Figure 3**). Significant wave height and direction follow a similar pattern to that  
371 of wind speed (**Figure 3**).

372           Lagoon surface water salinity increased between May and September 2017 from 26 to 52,  
373 due to low precipitation and high temperatures (**Figure 4**). In May, pore waters sampled between  
374 the lagoon and Mediterranean Sea primarily reflected the salinity of the lagoon endmember (26).  
375 Pore waters with a slightly lower salinity ( $\sim 24$ ) were likely derived from past lagoon conditions  
376 (April or earlier). During the spring, lagoon water levels were at a maximum (**Table 4**), and the  
377 May sampling period therefore experienced the greatest hydraulic gradient between the lagoon  
378 and sea, further evidenced by the second transect (T2), sampled over 250 m away (**Figure 4**).

379           In June and July, pore waters sampled closest to the lagoon were similar in salinity as the  
380 lagoon endmember (**Figure 4**). During this time period, pore water salinities decreased  
381 continuously with increasing distance from the lagoon towards the sea, reflecting a mixture  
382 between recent infiltration of the actual lagoon endmember and older low-salinity ( $\sim 24$ ) lagoon  
383 waters. During August and September, pore waters sampled next to the lagoon were significantly  
384 lower in salinity than the lagoon endmember (33 vs. 45 on August 30<sup>th</sup>; 33 vs. 52 on September  
385 25<sup>th</sup>). High evaporation rates and low precipitation during August and September (**Figure 3**)  
386 resulted in a diminished hydraulic gradient between the lagoon and the sea, corresponding to a

387 period in which lagoon surface water levels were relatively low (**Table 4**). Consequently, the  
388 high salinity lagoon waters did not penetrate into the sand barrier during that period, resulting in a  
389 large contrast in salinity between the lagoon waters and the pore waters close to the lagoon. In  
390 November, strong winds created an opening in the beach spit, forming a natural outlet to the sea,  
391 where the barrier beach was approximately half of the width (~40 m) that it had been in previous  
392 months. Because of the input of seawater (salinity = 38) into the lagoon, the salinity of the lagoon  
393 waters decreased to reach a salinity of 39 (in comparison to 52 in September).

#### 394 **Radionuclide Distribution**

395 Pore waters sampled directly adjacent to the lagoon (1 – 3 m) in May, June and July,  
396 which were similar in salinity to that of the lagoon (**Figure 4**), have elevated  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$  and  
397  $^{224}\text{Ra}_{\text{ex}}$  over the lagoon. This pattern indicates relatively recent lagoon water infiltration and  
398 subsequent radionuclide ingrowth from contact with the sediment (**Figures 5 & 6**).  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$   
399 and  $^{224}\text{Ra}_{\text{ex}}$  activities reach maximum activities within a ~30 m distance from the lagoon. Within  
400 the first ~5 – 15 m of the Mediterranean shoreline, pore water salinities reflected that of the  
401 Mediterranean Sea (~38), representing a zone in which the beach face was exposed to constant  
402 wave-setup.  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$ , and  $^{224}\text{Ra}_{\text{ex}}$  increased with increasing distance from the shoreline in this  
403 seawater salinity zone (**Figure 5**). Vertical profiles show generally increasing activities of pore  
404 water  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$  and  $^{224}\text{Ra}_{\text{ex}}$  with increasing depth, near both the lagoon and the sea (**Figure 7**).

405 In-situ pore water  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios were between 0.2 – 0.4 in samples taken in  
406 the center of the beach transect (n=14), which are assumed to be in equilibrium with sediment  
407 surfaces, since pore water  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios determined from the sediment equilibration  
408 experiments were 0.18 – 0.33. Considering a mean surface-bound  $^{224}\text{Ra}$  activity ( $0.041 \pm 0.006$   
409  $\text{dpm g}^{-1}$ ; **Table 2**) and assuming that all labile  $^{222}\text{Rn}$  is in solution, then the total (dissolved +

410 surface-bound) in-situ  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratio is equal to  $2.2 \pm 0.7$  (n=14). It is important to  
411 note that we do not consider  $^{222}\text{Rn}$  produced from dissolved  $^{226}\text{Ra}$  and any potential absolute  
412 differences in the  $^{238}\text{U}/^{232}\text{Th}$  activity ratio of the sediment (Luo et al. 2000).

### 413 **Dissolved Solutes**

414 DOC was depleted by  $\sim 0.1 - 0.4$  mM in beach pore waters, with respect to theoretical,  
415 two-endmember linear mixing between lagoon surface and Mediterranean Sea endmembers  
416 (depending upon position and season; **Figure 8**). Pore water DIC was enriched over both surface  
417 endmembers, increasing in concentration toward the center of the beach (**Figure 8**). There was  
418 more DIC observed than expected from DOC consumption ( $\sim 2 - 3$  mM), suggesting that there  
419 was DIC input to the pore waters from the weathering of carbonate minerals. Consumption of  
420 POC (if present in the sand barrier) may also contribute to the DIC enrichment in pore waters.  
421 DSi transects were similar to DIC, reaching a relative maximum ( $100 \mu\text{M}$  in August) in the center  
422 of the beach (**Figure 8**), indicating that dissolution of Si minerals takes place within the sand  
423 barrier and releases DSi into the pore water (Anschutz et al. 2009; Tamborski et al. 2018).  
424 Vertical DIC and DSi profiles sampled closest to the Mediterranean Sea increased in  
425 concentration with increasing depth, similar to  $^{222}\text{Rn}$  and  $^{223,224}\text{Ra}$ , for all six sampling seasons  
426 (**Figure 7**). Pore waters at 50 cm depth were enriched over surface water concentrations between  
427  $25$  and  $110 \mu\text{M}$  for DIC, and between  $4$  and  $11 \mu\text{M}$  for DSi. Transects of dissolved Ba, a  
428 chemical analog to Ra, primarily followed two-endmember mixing between surface water  
429 endmembers (**Figure 8**), while vertical profiles taken near the sea were relatively constant in  
430 concentration with depth (**Figure 7**). Therefore, these profiles suggest that pore water Ra  
431 distributions were significantly impacted by ingrowth from water-sediment interaction (**Figure**  
432 **5**). All surface and pore water data are summarized in the **Supplemental Information**.

433 **DISCUSSION**

434 **Reactive-Transport Model**

435 *Theory*

436 The change in Ra isotope activity with time may be described with a one-dimensional  
437 reactive-transport model. Here we use a horizontal reactive-transport model to describe the Ra  
438 activity of the beach transects as a function of advective lagoon-sea water exchange. In parallel,  
439 we use a vertical reactive-transport model to describe the Ra activity of the profiles taken  
440 adjacent to the Mediterranean Sea, as a function of seawater circulation. Neglecting  
441 hydrodynamic dispersion and assuming steady-state conditions (Krest and Harvey 2003; Michael  
442 et al. 2011), the advection of Ra in pore water can be described as:

443 
$$A(x) = \frac{P}{1+K} + \left( A_o - \frac{P}{1+K} \right) * e^{-\Delta x \lambda v^{-1}} \quad (\text{Eq. 7})$$

444 Where  $A(x)$  is the Ra activity (dpm L<sup>-1</sup>) at distance  $x$  (m) from a defined boundary condition,  $A_o$  is  
445 the initial Ra activity at a  $x = 0$ ,  $P$  is the Ra isotope production rate (dpm L<sup>-1</sup>),  $K$  is the  
446 dimensionless Ra partition coefficient (adsorbed exchangeable Ra on the solid/dissolved Ra),  $\lambda$  is  
447 the Ra isotope decay constant (d<sup>-1</sup>) and  $v$  is the advective pore water velocity (m d<sup>-1</sup>). Under  
448 homogeneous conditions in which groundwater salinity is constant and the initial Ra  
449 concentration is zero, Kiro et al. (2012) note that Eq. 7 may be further reduced if  $x > \frac{vt}{1+K}$  (where  
450  $t$  is time) to:

451 
$$A(x) = \frac{P}{1+K} * (1 - e^{-\Delta x \lambda v^{-1}}) \quad (\text{Eq. 8})$$

452 Considering <sup>224</sup>Ra<sub>ex</sub>/<sup>223</sup>Ra activity ratios, the solution to Eq. 7 becomes:

$$\frac{{}^{224}\text{Ra}}{{}^{223}\text{Ra}} = \frac{\frac{P_{224}}{1+K} + \left(A_{224o} - \frac{P_{224}}{1+K}\right) * e^{-\Delta x \lambda_{224} v^{-1}}}{\frac{P_{223}}{1+K} + \left(A_{223o} - \frac{P_{223}}{1+K}\right) * e^{-\Delta x \lambda_{223} v^{-1}}} \quad (\text{Eq. 9})$$

Equations 7, 8 and 9 may be modeled using a numerical curve-fitting approach, by adjusting the advective velocity term ( $v$ ) to minimize the root mean square error of the model with respect to the observed data. We have explicitly measured  $A_o$ ,  $P$  (**Table 2**) and  $K$  (via  $K_D$  and  ${}^{224}\text{Ra}/{}^{222}\text{Rn}$  activity ratios; **Tables 1 & 3**) to be used in the model calculations.

${}^{224}\text{Ra}_{\text{ex}}/{}^{223}\text{Ra}$  activity ratios were chosen to model the horizontal advection of lagoon-sea water exchange (Eq. 9). Small-scale sediment heterogeneity may result in variable (equilibrium) pore water  ${}^{223}\text{Ra}$  and  ${}^{224}\text{Ra}$  activity distributions over the ~100 m long beach transects (**Figure 5**). Sediment-exchangeable  ${}^{224}\text{Ra}/{}^{223}\text{Ra}$  activity ratios were relatively constant across the beach transect, indicating a homogeneous sediment distribution (**Table 2**); therefore,  ${}^{224}\text{Ra}/{}^{223}\text{Ra}$  activity ratios are better suited to identify disequilibrium between pore waters and sediment surfaces (Eq. 9) over absolute activities (Eq. 7 & 8) at such a spatial scale. Here, the horizontal boundary condition is set as the lagoon surface water endmember for May – July, to represent lateral flow through the barrier beach, driven by a positive hydraulic gradient between the lagoon and the sea (**Table 4**). We cannot assume steady-state conditions for August – September (because of the diminished hydraulic gradient) and November (the spit was recently breached); therefore, we do not model these months. The horizontal model is limited to shallow pore water samples only, where we could collect sediment samples. Shallow pore water samples closest to the lagoon are lower in  ${}^{223}\text{Ra}$  and  ${}^{224}\text{Ra}_{\text{ex}}$  activity compared to deeper samples (**Figure 7**); therefore, we consider these shallow samples as representative of this active water exchange zone, here defined as 1.0 m depth.

474 To estimate seawater circulation in the swash-zone, radionuclide activities were modeled  
475 for the pore water profiles sampled closest to the Mediterranean Sea ( $A_o$ ) (Eq. 7), where pore  
476 water salinities were equivalent to seawater (**Figure 7**). The vertical advective velocity is a rate  
477 of flushing exchange (net zero water flux), in which Ra atoms are lost from pore waters, similar  
478 to pore water exchange creating a (total)  $^{224}\text{Ra}$  deficit relative to its surface-bound parent  $^{228}\text{Th}$   
479 (Cai et al. 2014), but here we only consider the dissolved Ra pool. The short-lived radionuclide  
480 flux is not necessarily driven by a net water (discharge or recharge) flux; rather, the Ra flux can  
481 be sustained by seawater circulation (net zero water flux) (Cook et al. 2018a). The deficit of  
482 short-lived  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$  and  $^{224}\text{Ra}_{\text{ex}}$  does not reflect instantaneous seawater circulation for the  
483 exact time of sampling; rather, the radionuclide deficit is integrating across the time-scale of  
484 radionuclide ingrowth and decay.  $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra}$  activity ratios were not used for the vertical model  
485 because the range of the  $^{224}\text{Ra}/^{223}\text{Ra}$  activity ratios (and its relative uncertainty) were much lower  
486 compared to the larger range in activity ratio observed for the horizontal transects ( $\sim 10 - 35$ ).  
487 Terms used in the one-dimensional horizontal and vertical reactive transport models are  
488 summarized in **Table 5**.

### 489 *Sensitivity & Assumptions*

490 Reactive-transport model sensitivity to changes in production rate ( $P$ ) and Ra partition  
491 coefficient ( $K$ ; or the “retardation factor”  $R = 1 + K$ ) have been summarized in detail elsewhere  
492 (Tricca et al. 2001; Krest and Harvey 2003; Michael et al. 2011; Kiro et al. 2015; Tamborski et  
493 al. 2017). Michael et al. (2011) note that the ratio of Ra production to retardation is difficult to  
494 quantify and should be done so carefully in order to use the one-dimensional reactive-transport  
495 model in any quantitative, meaningful way. Tamborski et al. (2017) showed that radionuclide-  
496 derived pore water residence times ( $\tau = v/d$ ) within tidal beach environments are sensitive to

497 values of  $P$  for long residence times, but less sensitive when residence times are low  
498 (approximately less than one day). It is important to note that flow velocity is spatially and  
499 temporally variable, and is not as simple as the depiction of the one-dimensional model due to  
500 subsurface heterogeneity (Michael et al. 2011).

501 Sediment equilibration-derived  $K$  values (from  $^{224}\text{Ra}/^{222}\text{Rn}$ ; **Table 3**) for two different  
502 salinity solutions (27 and 38) were not significantly different when considering surface-bound  
503  $^{224}\text{Ra}$ , with respect to their range in uncertainties. In-situ determined  $K$  values ( $2.2 \pm 0.7$ ) were  
504 slightly higher than those determined from the sediment equilibration experiments and exhibited  
505 greater variability, as evidenced by a large standard deviation. All  $K$  values estimated from  
506  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios are lower than experimentally determined values from the sorption  
507 experiments (7.5 – 11.6). This discrepancy may be explained by an experimental particle  
508 concentration effect, as observed for the highly particle-reactive  $^{234}\text{Th}$  (Honeyman et al. 1988). In  
509 theory,  $K_D$  should remain constant regardless of the particle-to-solution ratio used  
510 experimentally; in practice, measured  $K_D$  values are greater for experiments with smaller particle-  
511 to-solution ratios, which can result in a factor of  $\sim 10$  difference in  $K_D$  (Honeyman et al. 1988). In-  
512 situ particle-to-solution ratios are an order of magnitude greater than particle-to-solution ratios  
513 typically used for  $K_D$  experiments. Indeed, Cochran et al. (1986) determined that the  $K_D$  for Th  
514 was  $\sim 10^5 \text{ cm}^3 \text{ g}^{-1}$  in nearshore (Buzzards Bay, MA, USA) sediment pore waters, in contrast to  
515 values two orders of magnitude greater ( $\sim 10^7 \text{ cm}^3 \text{ g}^{-1}$ ) in the deep sea (Bacon and Anderson  
516 1982; Honeyman et al. 1988).

517 Considering the range in  $K$  values noted above, we elect to use values derived from in-situ  
518  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios for samples assumed to be in equilibrium with sediment surfaces, taken  
519 in the center of the beach transect ( $2.2 \pm 0.7$ ;  $n=14$ ). We use a range in  $K$  determined from the  
520 standard deviation, to represent a minimum  $K$  (1.5) for salinity 38 and a maximum  $K$  (2.9) for



521 salinity 27. This range in  $K$  is qualitatively validated from the sorption  $K_D$  experiments (despite  
522 the particle-concentration effect noted above), which provides additional confidence to this  
523 approach. The uncertainty of the sediment equilibration  $K$  values prevents us from accurately  
524 distinguishing between salinities of 27 and 38, despite observable differences in experimental  
525 (**Table 3**) and in-situ (**Figure 5**) dissolved  $^{223,224}\text{Ra}$  activities.

526 Ra desorption from estuarine sediments is a non-linear process chiefly governed by the  
527 availability of Ra on particle surfaces and the major ion present in solution, where non-linearity is  
528 typically greatest between a salinity of 0 and ~15 (Webster et al. 1995). Changes between  $K$  and  
529 salinity for relatively high ionic strength solutions (i.e. > 30% seawater composition for seawater  
530 salinity of 36) may be approximated as linear (Webster et al. 1995; Kiro et al. 2012, 2013). Here  
531 we assume a linear relationship between  $K$  and salinity along the horizontal groundwater flow  
532 path for salinity 27 ( $K = 2.9$ ) and salinity 38 ( $K = 1.5$ ), based on the observed pore water salinity  
533 distribution (**Figure 4**). We further extrapolate to a salinity of 22 and 42 to match the observed  
534 minimum and maximum salinity. Bulk  $^{228}\text{Th}$  (**Table 1**) and surface-exchangeable  $^{224}\text{Ra}$  (**Table 2**)  
535 are relatively uniform throughout the beach. We assume that the mean value of  $P$  is  
536 representative of the beach transect in the horizontal direction. Values of  $P$  were measured for  
537 surficial sediments (~0 – 50 cm depth) along the investigated transect; values of  $P$  may change  
538 with depth in the vertical model due to differing amounts of  $^{228}\text{Th}$  present on sediment surfaces.  
539 Spatially variable Ra and Rn production can occur if there are significant differences in the Fe  
540 and Mn (hydr)oxide coatings on sediment surfaces (Dulaiova et al. 2008; Beck and Cochran  
541 2013), or barite ( $\text{BaSO}_4$ ) precipitation (Kiro et al. 2012), which we do not consider at present. For  
542 the vertical reactive-transport model, the term  $[P/(I+K)]$  is simply constrained from the sediment  
543 incubation experiments for seawater salinity (**Table 3**).

544 *Model Results*

545 Results from the horizontal model (Eq. 9) for lagoon-sea water exchange are presented in  
546 **Figure 9**. In general, there was an abrupt increase in the observed  $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra}$  activity ratio  
547 within a few meters of the lagoon boundary, due to the relatively more rapid ingrowth of  $^{224}\text{Ra}$   
548 with respect to  $^{223}\text{Ra}$ , followed by an abrupt decrease in the activity ratio, in which  $^{224}\text{Ra}$  reached  
549 equilibrium with sediment-surfaces while  $^{223}\text{Ra}$  continued to be produced, until the dissolved  
550 activity of both isotopes was balanced by production and decay ( $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra} = 16.7$ ; **Table 2**).  
551 Model best-fit horizontal advective velocities decreased from 85 to 30 to 20  $\text{cm d}^{-1}$  for May, June  
552 and July, respectively (**Figure 9**), following the decreasing hydraulic gradient (**Table 4**). The two  
553 transects sampled in May (over 250 m apart) have equal advective velocities from the modeled  
554  $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra}$  activity ratios, suggesting that the flow of lagoon water through the beach was  
555 prevalent along La Palme lagoon and the beach of La Franqui during this high-water level period.  
556 Therefore, during May, pore water residence times were on the order of hours close to the lagoon  
557 edge ( $\sim 1$  m), several days within 10 m of the lagoon and several weeks in the center of the beach.  
558 Advective linear velocities are converted to water fluxes considering a mean sediment porosity of  
559 0.38 (**Table 5**) and an exchange zone depth of 1.0 m (**Figure 7**). Volumetric water fluxes for  
560 May, June and July are therefore between  $0.1 - 0.3 \text{ m}^3 \text{ d}^{-1} \text{ m}^{-1}$  of shoreline.

561 We can qualitatively compare our horizontal one-dimensional reactive-transport model  
562 results to the observed changes in pore water salinity with time (**Figure 4**). Changes in pore water  
563 salinity are explained by mixing between current high-salinity lagoon waters and older (fresher)  
564 lagoon waters, driven laterally toward the sea from a positive hydraulic gradient (**Table 4**). The  
565 pore water salinity profile observed on June 22<sup>nd</sup> persisted 14 days later to July 6<sup>th</sup>, as the lagoon  
566 water level decreased (**Table 4**) and increased in salinity (37 to 39). On July 6<sup>th</sup>, the lower  
567 salinity endmember from May ( $\sim 25$ ) was observed up to 42 m away from the lagoon, an absolute

568 difference of 6 m (seaward) from June 22<sup>nd</sup>. The difference in the lower salinity plumes  
569 horizontal position across the beach corresponds to an integrated horizontal flow velocity of ~43  
570 cm d<sup>-1</sup> (6 m/14 d). From August 30<sup>th</sup> to September 25<sup>th</sup> the low salinity plume reversed direction;  
571 the plume was displaced 11 m toward the lagoon (~40 cm d<sup>-1</sup>).

572 Vertical profiles (Eq. 7; derived from <sup>223</sup>Ra) displayed significant temporal variability,  
573 with model best-fit vertical advective velocities equal to 30, 14, 10, 25, 55 and 52 cm d<sup>-1</sup> for May,  
574 June, July, August, September and November, respectively (e.g. **Figure 10**). Estimates derived  
575 from <sup>222</sup>Rn equal 30 and 90 cm d<sup>-1</sup> for June and November (**Figure 10**), in relative agreement  
576 with <sup>223</sup>Ra, despite integrating over different time-scales. <sup>224</sup>Ra<sub>ex</sub> model results are linearly  
577 correlated to estimates from <sup>223</sup>Ra (slope = 0.92). Scaling the <sup>223</sup>Ra best-fit vertical advective  
578 velocities to the width of the swash-zone impacted by seawater circulation (5 m into the beach-  
579 face and 5 m into the sea) and considering a mean porosity of 0.38 results in volumetric seawater  
580 circulation fluxes between 0.4 – 2.1 m<sup>3</sup> d<sup>-1</sup> m<sup>-1</sup> of shoreline. We note that the seawater circulation  
581 fluxes for September and November may easily be a factor of two larger, considering the width  
582 of the beach impacted by wave-setup, from the observed pore water salinity distribution (**Figure**  
583 **4**).

584 The Mediterranean beach studied here has a minor tidal range (~20 – 30 cm); therefore,  
585 we can compare the above results to other studies conducted in areas with significant tidal ranges  
586 to qualitatively assess the role of tidal pumping as a driver of both lagoon-sea water exchange  
587 and seawater circulation. Lagoon-sea water exchange fluxes (0.1 – 0.3 m<sup>3</sup> d<sup>-1</sup> m<sup>-1</sup> of shoreline)  
588 are an order of magnitude lower than those obtained from higher-energy tidal environments  
589 subject to exchange through a permeable barrier (e.g. 2.6 – 5.7 m<sup>3</sup> m<sup>-1</sup> of shoreline per tide for  
590 Cabretta Beach, Georgia) (Evans and Wilson 2017). The advective linear velocity estimated for  
591 May is similar to rates in a porous limestone coral-reef environment (100 cm d<sup>-1</sup>) (Santos et al.

592 2010). Larger tidal ranges produce greater water level differences between coastal lagoons and  
593 the coastal sea, effectively increasing the hydraulic gradient and therefore SGD rate.

594 The volumetric flux of seawater circulation estimated here for the swash-zone (0.4 – 2.1  
595  $\text{m}^3 \text{d}^{-1} \text{m}^{-1}$  of shoreline) is lower than modeling estimates of wave-swash induced infiltration to  
596 the unsaturated zone of a sandy beach ( $1.6 \text{ m}^3 \text{ m}^{-1}$  of shoreline per tidal cycle) (Heiss et al. 2014),  
597 and is an order of magnitude lower than the highly energetic Aquitanian French coastline ( $\sim 15.2$   
598  $\text{m}^3 \text{ m}^{-1}$  of shoreline per tidal cycle), driven by tidal pumping and wave-setup (Charbonnier et al.  
599 2013). Large tidal variations drive greater amounts of seawater into permeable sediments; tidal  
600 pumping is likely more significant than wave or wind-setup in tidal environments with limited  
601 fetch (Bokuniewicz et al. 2015).

## 602 **Physical Drivers of Exchange**

### 603 *Lagoon-Sea Water Exchange*

604 Lagoon-sea water exchange is driven by the pressure induced hydraulic gradient between  
605 the water level of the lagoon and the water level of the Mediterranean Sea. It is apparent that as  
606 the lagoon water level decreased from spring through summer, the hydraulic gradient between the  
607 lagoon and the Mediterranean Sea decreased (**Table 4**), resulting in a slower flow of water  
608 through the beach. Qualitative flow estimates from Darcy's law ( $Q = -k_h * i$ ), considering a  
609 hydraulic conductivity ( $k_h$ ) on the order of  $10^{-2}$  to  $10^{-3} \text{ m s}^{-1}$  for clean sands (Freeze and Cherry,  
610 1979) and the hydraulic gradient between the lagoon and the sea ( $i$ ; **Table 4**) results in horizontal  
611 flow velocities on the order of  $\sim 100 \text{ cm d}^{-1}$ . The absolute value of the Darcy flux should not be  
612 overly interpreted here, as we do not have detailed information on water table elevation (few  
613 point measurements) and sediment hydraulic conductivity (assumed values). Nevertheless, this

614 exercise is useful to constrain the relative magnitude of the horizontal flow velocity to that of the  
615 horizontal reactive-transport model.

### 616 *Seawater Circulation*

617 Vertical flushing velocities were highly variable over the six-month sampling campaign  
618 (**Figure 10**). Variability in vertical flushing can change with wave height, wave period and wave  
619 direction, all of which are a function of the prevailing wind speed and direction. According to the  
620 results from the multiple linear-regression model (Section *Ancillary Parameters*), wind speed and  
621 direction significantly explain the temporal variability in vertical flushing velocities. Regression  
622 coefficients are strongest for the 48 h and 72 h binned averages ( $R^2 = 0.87$  and  $0.92$ ,  $F = 0.02$  and  
623  $0.05$ ,  $df = 3$ ) (**Table 6**). Regression coefficients are weaker for the 6 – 24 h binned averages ( $R^2 =$   
624  $0.64 - 0.79$ ,  $F \geq 0.1$ ,  $df = 3$ ); no correlation is observed for the 1-week and 2-week binned  
625 averages ( $R^2 = 0.2$ ). The significant relationships over the 48 and 72 h intervals display the  
626 “memory effect” that Ra and Rn isotopes have in integrating water exchange fluxes over the  
627 time-scale of the radionuclide half-life. In contrast, wind-setup and wave-runup (Eq. 2 – 5)  
628 significantly explain the temporal variability in vertical flushing for only the 6 h binned average  
629 ( $R^2 = 0.87$ ,  $F = 0.05$ ,  $df = 3$ ) (**Table 6**). Several hours may be needed for winds to produce waves  
630 in the Mediterranean Sea; however, only a few hours may be needed for waves to affect pore  
631 waters, both of which are highly dependent upon direction.

### 632 *Biogeochemical Significance*

633 Lagoon-sea water exchange and seawater circulation flow paths drive early diagenetic  
634 nutrient mineralization (Charbonnier et al. 2013) and sediment weathering reactions (Ehlert et al.  
635 2016). Here we provide a first-order estimate of solute fluxes driven by lagoon-sea water  
636 exchange during high water conditions, assuming that flow through the barrier beach was

637 ultimately supplied to the Mediterranean Sea. Lagoon-sea water exchange is a net water source  
638 and may therefore be a net solute flux to the coastal Mediterranean Sea, depending upon  
639 biogeochemical reactions (Cook et al. 2018b). Solute concentrations measured at ~20 m distance  
640 from the sea (**Table 7**) are used as the endmember to estimate lagoon-sea water exchange solute  
641 fluxes. At this distance, we assume negligible dilution from mixing with seawater (**Figure 4**) and  
642 that most of the biogeochemical transformations have occurred (**Figure 8**). Corresponding DSi,  
643 DIC and Ba fluxes equal  $10 \text{ mmol Si d}^{-1} \text{ m}^{-1}$ ,  $1,100 \text{ mmol C d}^{-1} \text{ m}^{-1}$  and  $70 \text{ } \mu\text{mol Ba d}^{-1} \text{ m}^{-1}$  of  
644 shoreline during May 2017 (**Table 7**). Solute fluxes decrease from June to July (**Table 7**) as a  
645 consequence of reduced flow (**Figure 9**).

646         The lagoon-sea water exchange DIC flux is  $\sim 70 - 200 \text{ mmol m}^{-2} \text{ d}^{-1}$  assuming a 5 m wide  
647 seepage-face; this flux is within the global range of DIC inputs from SGD ( $\sim 10 - 2000 \text{ mmol m}^{-2}$   
648  $\text{d}^{-1}$ ) summarized by Sadat-Noori et al. (2016). DIC inputs from lagoon-sea water exchange can  
649 lead to coastal ocean acidification (Robinson et al. 2018), which may explain the relatively low  
650 pH values measured in the coastal Mediterranean Sea during the spring (pH = 8.03 in May, pH =  
651 8.20 in June and pH = 8.25 in September; **Supplemental Information**). With regard to Ba,  
652 dissolved concentrations are typically  $\sim 40 \text{ nM}$  in the Mediterranean Sea (Jacquet et al. 2016).  
653 Local Ba inputs from lagoon-sea water exchange may alter local Ba biogeochemical cycling;  
654 further work is required to understand the role of SGD as a source of Ba to the Mediterranean  
655 Sea.

656         Adjacent to the Mediterranean Sea, pore waters with the same salinity as seawater were  
657 enriched in DIC ( $25 - 110 \text{ } \mu\text{M C}$ ) and DSi ( $4 - 11 \text{ } \mu\text{M Si}$ ) over surface waters, while Ba  
658 concentrations were relatively uniform (**Figure 7**). Linear Ba profiles suggest that seawater  
659 circulation in the swash-zone is not a net source of Ba to the coastal sea. We may estimate solute

660 fluxes in the swash-zone by multiplying the net solute concentration (enrichment over  
661 Mediterranean seawater) by the volume of seawater circulation (Cook et al. 2018b). Seawater  
662 circulation-driven DIC fluxes through the swash-zone are between 14 – 220 mmol C d<sup>-1</sup> m<sup>-1</sup> of  
663 shoreline (**Table 7**), possibly as a consequence of carbonate mineral weathering, in addition to  
664 POC and DOC consumption (**Figure 7**). Seawater circulation-driven DSi fluxes through the  
665 swash-zone are between 3 – 8 mmol Si d<sup>-1</sup> m<sup>-1</sup> of shoreline (**Table 7**).

666         During November 2016, SGD was estimated to supply a DSi flux of  $2.4 \pm 1.4$  mol Si d<sup>-1</sup>  
667 m<sup>-1</sup> of shoreline for La Franqui, based on offshore transects that were conducted in the coastal  
668 seas (Tamborski et al. 2018). This DSi flux was hypothesized to be driven by both lagoon-sea  
669 water exchange (relatively high-water levels) and seawater circulation (Tamborski et al. 2018);  
670 this flux is 2 – 3 orders of magnitude larger than the DSi fluxes calculated here for the swash-  
671 zone (**Table 7**). The DSi flux of Tamborski et al. (2018) may include additional, unaccounted for  
672 flow paths between the swash-zone (this study) and 300 m offshore, where the surface water  
673 transects to quantify SGD originated (Figure 6 of Tamborski et al., 2018). Water depth is  $\leq 5$  m  
674 over 300 m from shore; seawater circulation through this shallow zone, over a large area ( $\sim 3$   
675 km<sup>2</sup>), may thus be an adequate source of DSi (and Ra) offshore of La Franqui. To reconcile the  
676 difference in the DSi flux from this study and Tamborski et al. (2018), we hypothesize that  
677 offshore seawater circulation may supply a DSi flux on the order of 1 – 2 mol Si d<sup>-1</sup> m<sup>-1</sup> of  
678 shoreline. While seawater circulation is relatively small in supplying DSi from the narrow swash-  
679 zone, this flow path may be significant over the larger area of the shallow coastal zone; future  
680 research is required to better constrain these coastal solute fluxes.

## 681 CONCLUSIONS

682 Various flow paths make up the term submarine groundwater discharge (SGD) (Santos et al. 2012). Individual driving mechanisms are seldom quantified with respect to absolute water  
683 fluxes and its associated chemical load to the sea (Robinson et al. 2018), despite increasing  
684 recognition that individual flow paths may have unique geochemical signatures (Tamborski et al.  
685 2017; Rodellas et al. 2018). In this study we present a simple Ra isotope methodology, which  
686 builds upon preexisting analytical measurements and theoretical models to explicitly quantify (1)  
687 lagoon-sea water exchange through a permeable sand barrier and (2) seawater circulation in the  
688 swash-zone of the sand barrier. Multiple methods are recommended to determine the production  
689 ( $P$ ) and partitioning ( $K$ ) of Ra between sediment surfaces and solution in order to properly model  
690 1-D Ra transport. This methodology provides a framework where future researchers may  
691 discriminate between unique SGD flow paths, which in turn will help improve solute flux  
692 estimates and our understanding of coastal biogeochemical cycles.

694 Repeated measurements at a Mediterranean barrier beach site show that lagoon-sea water  
695 exchange varies in response to the changing hydraulic gradient between the lagoon and the sea.  
696 This flow path is a net water flux which supplies the greatest DSi ( $10 \text{ mmol d}^{-1} \text{ m}^{-1}$ ), DIC ( $1,100$   
697  $\text{mmol d}^{-1} \text{ m}^{-1}$ ) and Ba ( $70 \text{ } \mu\text{mol d}^{-1} \text{ m}^{-1}$ ) flux to the coastal Mediterranean Sea during high lagoon  
698 water-level periods. The DIC flux from lagoon-sea water exchange is comparable to other SGD  
699 studies and suggests that this vector may enhance coastal ocean acidification in the  
700 Mediterranean Sea (Sadat-Noori et al. 2016). We conclude that coastal lagoons represent a  
701 significant source of new DIC and Ba to the Mediterranean Sea during high water-level periods  
702 and are likely an important source for other dissolved chemical elements as well. Solute fluxes  
703 may significantly decrease during the dry season (summer), when lagoon surface water-levels



704 decrease. As a result, this seasonal vector should be considered in future coastal zone chemical  
705 budgets.

706 Seawater circulation through the swash-zone varies in response to short-term (hours to  
707 days) changes in regional wind and wave regimes. This is one of the first studies to quantitatively  
708 link a seawater circulation flux to its physical driving mechanism(s), tidal pumping  
709 notwithstanding. This vector is relatively minor in transferring DSi and DIC to the Mediterranean  
710 Sea during the studied period (May – November 2017), while Ba inputs are negligible. It remains  
711 to be seen how significant these individual flow paths are in transferring other chemical elements,  
712 such as N and P, to the coastal Mediterranean Sea. Further work is required to understand the  
713 importance of seawater circulation along the shallow nearshore coastal zone (~0 – 300 m  
714 offshore) to the Mediterranean Sea.

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## 926 **FIGURE LEGENDS**

927 **Figure 1.** The study site of La Franqui, located between La Palme lagoon and the Mediterranean  
928 Sea (Southern France, inset a; black square). The shore-perpendicular transect (Transect 1) was  
929 sampled monthly from May to September 2017, and again in November 2017. Transect 2 was  
930 sampled during May 2017 only. Google Earth imagery from May 14<sup>th</sup> 2017 is located  
931 approximately at the position of the white star (inset b). Lagoon surface water elevation  
932 measurement location is depicted by a circle (inset b; data from Parc Naturel Régional de la  
933 Narbonnaise en Méditerranée).

934 **Figure 2.** Sediment counting efficiency of the RaDeCC system determined from a <sup>224</sup>Ra standard  
935 addition spike, where the counting efficiency is equal to the slope of the cpm vs. dpm linear  
936 regression (A). <sup>224</sup>Ra sediment activity as a function of sample load (B). Note that the error bars  
937 are generally smaller than the symbol size.

938 **Figure 3.** Precipitation, wind speed and wave height during 2017. Wind and wave direction are  
939 plotted by color. Wind speed data is from “Meteo France” and wave height data is from  
940 CEREMA. Sampling periods are indicated by vertical gray lines. The dominant winds in the  
941 region include wind from the NW (i.e. “Tramontane”,  $\sim 315^\circ$ , yellow color on the plot) and wind  
942 from the sea (E;  $\sim 90^\circ$ , blue color on the plot).

943 **Figure 4.** Spatial distribution of shallow pore water salinity (50 cm depth; hollow circles) with  
944 respect to lagoon surface water (filled triangle) and Mediterranean Sea (filled square)  
945 endmembers. During May, a second transect (Transect 2) was sampled and is depicted by  
946 diamonds connected with a dashed-line; note T1 is offset during May for clarity. Note the y and  
947 x-axes differ between plots.

948 **Figure 5.** Spatial distribution of shallow (50 cm depth) pore water  $^{223}\text{Ra}$  (filled diamonds) and  
949  $^{224}\text{Ra}_{\text{ex}}$  (hollow circles) with respect to lagoon surface water (triangle) and Mediterranean Sea  
950 (square) endmembers.

951 **Figure 6.** Spatial distribution of shallow pore water (50 cm depth; hollow circles)  $^{222}\text{Rn}$  with  
952 respect to lagoon surface water (filled triangle) and Mediterranean Sea (filled square)  
953 endmembers.  $^{222}\text{Rn}$  was only sampled during May, June and November. Note the x-axes differ  
954 between plots.

955 **Figure 7.** Vertical distribution of salinity,  $^{223}\text{Ra}$ ,  $^{224}\text{Ra}_{\text{ex}}$ , DSi and Ba in pore water samples  
956 collected closest to the Mediterranean Sea (top) and closest to the lagoon (bottom), arranged by  
957 sampling season.

958 **Figure 8.** Spatial distribution of shallow pore water (50 cm depth; circles) DOC, DIC, DSi and  
959 Ba with respect to lagoon surface water (filled triangle) and Mediterranean Sea (filled square)



960 endmembers. Note that November is not included and only a limited number of samples were  
961 analyzed for DOC in May, August and September.

962 **Figure 9.** Distribution of shallow pore water ( $\leq 50$  cm)  $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra}$  activity ratios and  
963 corresponding results from the  $^{224}\text{Ra}_{\text{ex}}/^{223}\text{Ra}$  one-dimensional horizontal reactive-transport model.  
964 MED SW = Mediterranean surface water endmember; LW = lagoon surface water endmember.  
965 Note that transect 2 is included in May, with samples depicted as diamonds and model results as  
966 gray lines (a); T1 is offset during May for clarity. The solid black line is the model run with the  
967 lagoon water set as the boundary condition ( $A_o$ ). Dashed lines represent  $\pm 15\%$  model  
968 uncertainty. Model results do not extend farther than 40 m from the lagoon endmember.

969 **Figure 10.** Vertical distribution of  $^{222}\text{Rn}$ ,  $^{223}\text{Ra}$  and  $^{224}\text{Ra}_{\text{ex}}$  in pore water samples collected  
970 closest to the Mediterranean Sea. The thick dashed-line is the one-dimensional vertical reactive  
971 transport model result for June ( $^{222}\text{Rn } \nu = 30 \text{ cm d}^{-1}$ ;  $^{223}\text{Ra } \nu = 14 \text{ cm d}^{-1}$ ;  $^{224}\text{Ra } \nu = 14 \text{ cm d}^{-1}$ ) and  
972 the thin dashed-line is the vertical model result for November ( $^{222}\text{Rn } \nu = 90 \text{ cm d}^{-1}$ ;  $^{223}\text{Ra } \nu = 52$   
973  $\text{cm d}^{-1}$ ;  $^{224}\text{Ra } \nu = 35 \text{ cm d}^{-1}$ ).

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979 **TABLES**

980 **Table 1.** Bulk solid-phase radionuclide activities and measured distribution coefficients ( $K_D$ ) for  
 981 a salinity of 27 and 38.

ID	Distance from sea	$^{226}\text{Ra}$	$^{228}\text{Ra}$	$^{228}\text{Th}$	$^{228}\text{Th}/^{226}\text{Ra}$	$K_D$ (S = 27)	$K_D$ (S = 38)
	<i>m</i>	<i>dpm g<sup>-1</sup></i>	<i>dpm g<sup>-1</sup></i>	<i>dpm g<sup>-1</sup></i>		<i>cm<sup>3</sup> g<sup>-1</sup></i>	<i>cm<sup>3</sup> g<sup>-1</sup></i>
5-PW3-A	23	0.57 ± 0.02	0.66 ± 0.04	0.67 ± 0.01	1.17 ± 0.04	-	-
5-2-PW3-A	44	0.58 ± 0.04	0.41 ± 0.07	0.79 ± 0.07	1.37 ± 0.15	3.1	2.1
5-PW5-A	43	0.58 ± 0.02	0.47 ± 0.03	0.61 ± 0.02	1.05 ± 0.05	2.4	1.4
5-PW7-A	63	0.50 ± 0.02	0.55 ± 0.04	0.56 ± 0.01	1.12 ± 0.05	-	-

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995 **Table 2.** Sediment surface-exchangeable  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities, determined via RaDeCC.

<b>ID</b>	<b>Distance from sea</b> <i>m</i>	$^{223}\text{Ra}$ <i>dpm g<sup>-1</sup></i>	$^{224}\text{Ra}$ <i>dpm g<sup>-1</sup></i>	$^{224}\text{Ra}/^{223}\text{Ra}$
5-PW3-A	23	0.0022 ± 0.0003	0.034 ± 0.001	15.7
5-PW5-A	43	0.0025 ± 0.0005	0.038 ± 0.002	15.2
5-PW6-A	53	0.0024 ± 0.0005	0.050 ± 0.002	20.9
5-PW7-A	63	0.0028 ± 0.0003	0.041 ± 0.001	15.0
<b>AVG ± STD</b>		<b>0.0025 ± 0.002</b>	<b>0.041 ± 0.006</b>	<b>16.7 ± 2.4</b>

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1009 **Table 3.** Sediment equilibration results. Total  $^{224}\text{Ra}/^{222}\text{Rn}$  activity ratios consider a mean surface-  
 1010 exchangeable  $^{224}\text{Ra}$  activity of  $0.041 \pm 0.006 \text{ dpm g}^{-1}$  ( $176 \pm 25 \text{ dpm L}^{-1}$ ).

	$^{222}\text{Rn}^*$ <i>dpm L<sup>-1</sup></i>	$^{223}\text{Ra}$ <i>dpm L<sup>-1</sup></i>	$^{224}\text{Ra}$ <i>dpm L<sup>-1</sup></i>	$^{224}\text{Ra}/^{222}\text{Rn}$ <i>dissolved</i>	$^{224}\text{Ra}/^{222}\text{Rn}$ <i>total</i>
<b>Salinity 27</b>	$160 \pm 20$	$2.5 \pm 0.3$	$29.0 \pm 3.0$	$0.18 \pm 0.03$	$1.28 \pm 0.22$
<b>Salinity 38</b>	$160 \pm 20$	$3.0 \pm 0.2$	$52.5 \pm 4.0$	$0.33 \pm 0.05$	$1.43 \pm 0.24$

1011  $^{222}\text{Rn}$  equilibration results are only for a salinity of 38 but assumed to be representative for all salinities (minor  
 1012 influence of water salinity on  $^{222}\text{Rn}$  partitioning).

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1025 **Table 4.** Lagoon surface water elevation and water table elevation above sea level. Water table  
 1026 elevation measurements are taken closest to the lagoon edge, approximately 73 m from the  
 1027 Mediterranean Sea and are corrected for the elevation of the beach surface (measured during  
 1028 June). November is not included. Lagoon surface water elevation data is from the Parc Naturel  
 1029 Régional de la Narbonnaise en Méditerranée monitoring station Coussoules (Fig. 1).

	<b>Surface Water Elevation</b>	<b>Water Table Elevation</b>
	<i>m</i>	<i>m</i>
<b>May</b>	0.85	0.36
<b>June</b>	0.75	0.29
<b>July</b>	0.60	0.24
<b>August</b>	0.53	0.19
<b>September</b>	0.45	0.13
<b>November</b>	0.55	-

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1039 **Table 5.** Parameters used in the one-dimensional reactive-transport model. Note that the term  $K$   
 1040 is only interpolated for the horizontal model. The vertical model uses the term  $[P/(I+K)]$   
 1041 determined from sediment equilibration experiments for seawater salinity. The radionuclide  
 1042 activity of the Mediterranean Sea and lagoon surface endmember is taken as the value measured  
 1043 for each respective sampling campaign.

Term	Definition	Value	Uncertainty	Units
$\phi$	Porosity	0.38	0.01	-
$\rho$	Sediment Density	2.65	-	g cm <sup>-3</sup>
$P_{222}$	<sup>222</sup> Rn Production	160	20	dpm L <sup>-1</sup>
$P_{223}$	<sup>223</sup> Ra Production	10.6	1.0	dpm L <sup>-1</sup>
$P_{224}$	<sup>224</sup> Ra Production	176	25	dpm L <sup>-1</sup>
$K$	Ra Partition Coefficient	linear interpolation from salinity distribution*	-	-
$\Delta x$	Change in distance	measured from boundary condition	-	m
$\lambda_{222}$	<sup>222</sup> Rn Decay Constant	0.181	-	d <sup>-1</sup>
$\lambda_{223}$	<sup>223</sup> Ra Decay Constant	0.061	-	d <sup>-1</sup>
$\lambda_{224}$	<sup>224</sup> Ra Decay Constant	0.193	-	d <sup>-1</sup>
$A_{sea}^{222}$	<sup>222</sup> Rn coastal Mediterranean Sea	4 - 9	20-40%	dpm L <sup>-1</sup>
$A_{sea}^{223}$	<sup>223</sup> Ra coastal Mediterranean Sea	0.1 - 0.3	7-9%	dpm L <sup>-1</sup>
$A_{sea}^{224}$	<sup>224</sup> Ra coastal Mediterranean Sea	3.4 - 8.3	5-9%	dpm L <sup>-1</sup>
$A_{lagoon}^{223}$	<sup>223</sup> Ra Lagoon Surface Water	0.1 - 1.3	7 - 14%	dpm L <sup>-1</sup>
$A_{lagoon}^{224}$	<sup>224</sup> Ra Lagoon Surface Water	3.1 - 26	6 - 14%	dpm L <sup>-1</sup>
$v$	1-D advection velocity	modeled from curve-fitting	15%	m d <sup>-1</sup>

1044 \* $K$  is linearly interpolated between values of 1.5 – 2.9 for salinities of 27 – 38.

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1051 **Table 6.** Summary of Fisher’s F test ( $Pr>F$ ,  $df = 3$ ) multiple linear regression model results for  
 1052 seawater circulation through the swash-zone, with corresponding regression coefficients ( $R^2$ ).  
 1053 Seawater circulation advective velocities (dependent variable) are determined from the  $^{223}\text{Ra}$   
 1054 reactive-transport model. Wind speed and wind direction are the variables used in the “Wind”  
 1055 regression; wind-setup and wave-runup are the variables used in the “Waves” regression.  
 1056 Statistically significant model results are indicated in bold.

Time (h)	Wind - $R^2$	Wind - $Pr>F$	Waves - $R^2$	Waves - $Pr>F$
6	0.78	0.10	<b>0.87</b>	<b>0.05</b>
12	0.79	0.10	0.75	0.12
24	0.64	0.21	0.67	0.19
48	<b>0.92</b>	<b>0.02</b>	0.09	0.86
72	<b>0.87</b>	<b>0.05</b>	0.20	0.86
168	0.20	0.71	0.38	0.49
336	0.20	0.71	0.24	0.66

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1065 **Table 7.** Summary of water exchange, DSi ( $\text{mmol m}^{-1} \text{d}^{-1}$ ), DIC ( $\text{mmol m}^{-1} \text{d}^{-1}$ ) and Ba ( $\mu\text{mol m}^{-1} \text{d}^{-1}$ ) fluxes, arranged by sampling  
 1066 season. Lagoon-sea water exchange is only considered for May, June and July. Swash-zone seawater circulation flushing rates are  
 1067 derived from  $^{223}\text{Ra}$  profiles; Ba fluxes are not estimated for seawater circulation.

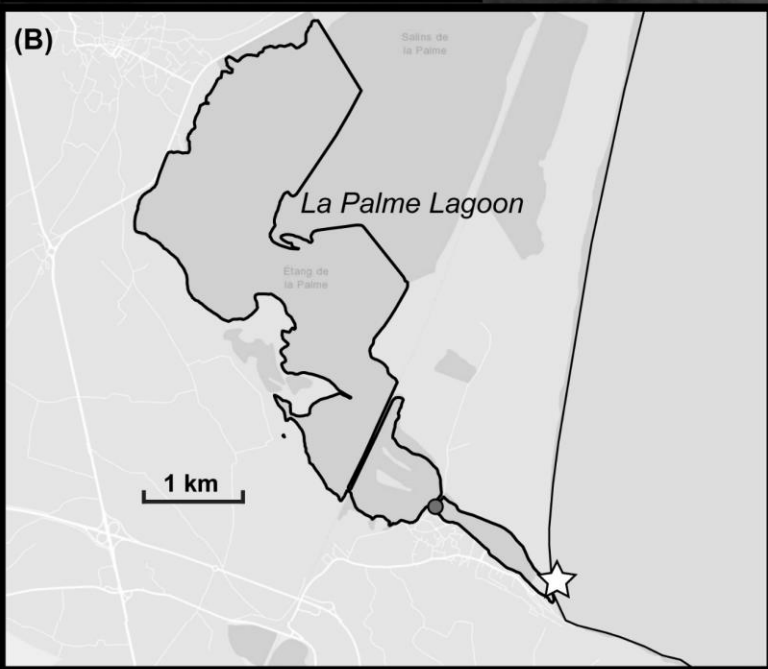
Date	Flushing Rate		Concentration	Si Flux	Concentration	C Flux	Concentration	Ba Flux
	$\text{cm d}^{-1}$	$\text{m}^3 \text{d}^{-1} \text{m}^{-1}$	$\mu\text{M Si}$	$\text{mmol m}^{-1} \text{d}^{-1}$	$\mu\text{M C}$	$\text{mmol m}^{-1} \text{d}^{-1}$	nM Ba	$\mu\text{mol m}^{-1} \text{d}^{-1}$
<i>Horizontal Transport (lagoon)*</i>								
9-May	85	0.3	31	10	3,280	1,100	220	70
22-Jun	30	0.1	35	4	3,610	410	120	10
6-Jul	20	0.1	44	3	4,260	320	150	10
<i>Seawater Circulation^</i>								
9-May	30	1.1	4	5	25	29	-	-
22-Jun	14	0.5	11	6	27	14	-	-
6-Jul	10	0.4	9	3	53	20	-	-
30-Aug	25	1.0	9	8	52	50	-	-
25-Sep	55	2.1	-	-	-	-	-	-
17-Nov	52	2.0	4	7	110	220	-	-

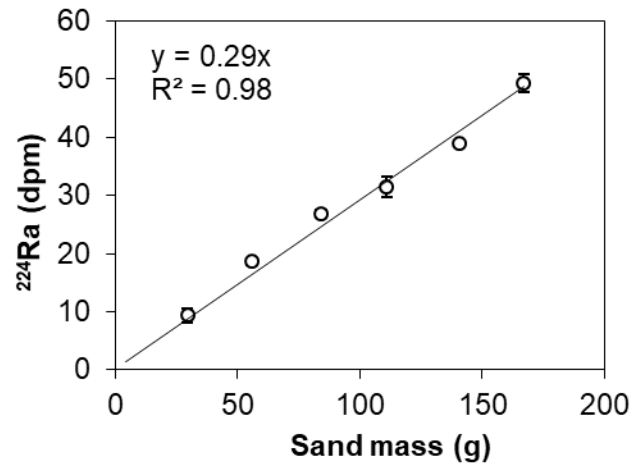
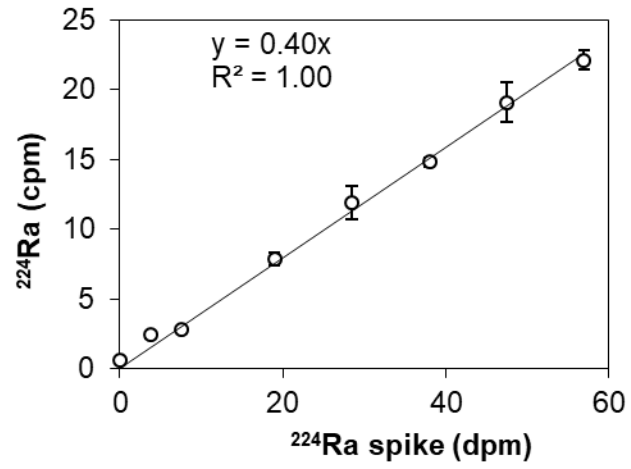
1068 \*Exchange zone depth = 1 m; DSi, DIC and Ba concentrations taken at  $\sim x=20$  m from the sea.

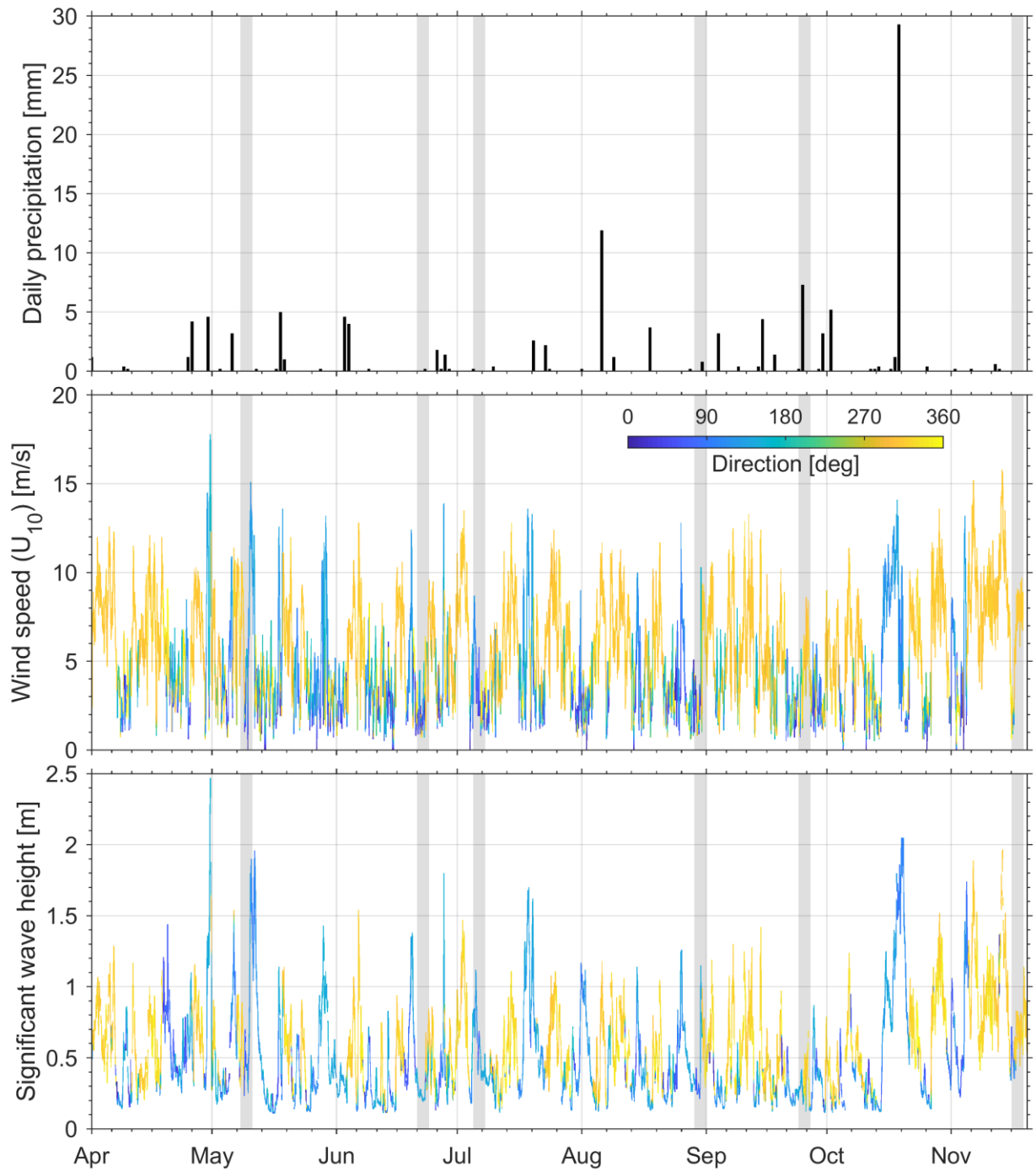
1069 ^Swash-zone width = 10 m; DSi and DIC concentrations are net enrichments over coastal Mediterranean Sea surface endmembers.

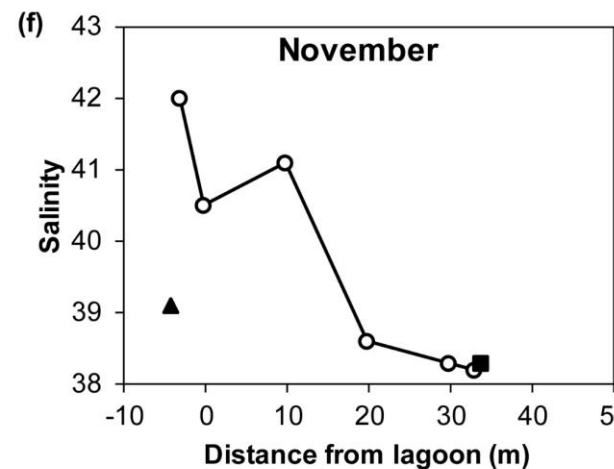
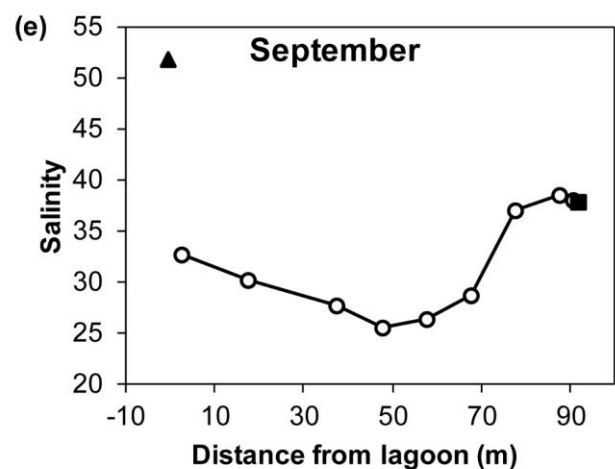
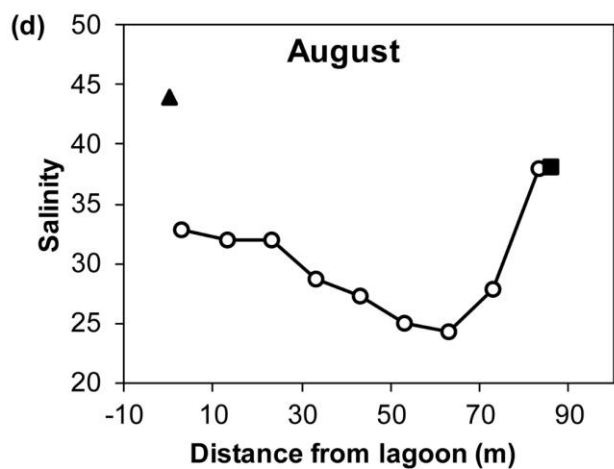
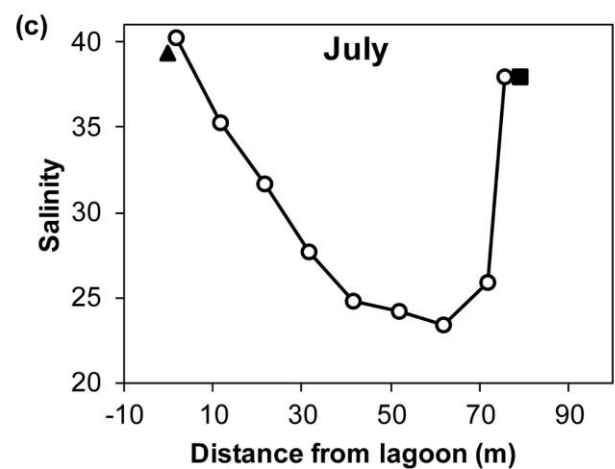
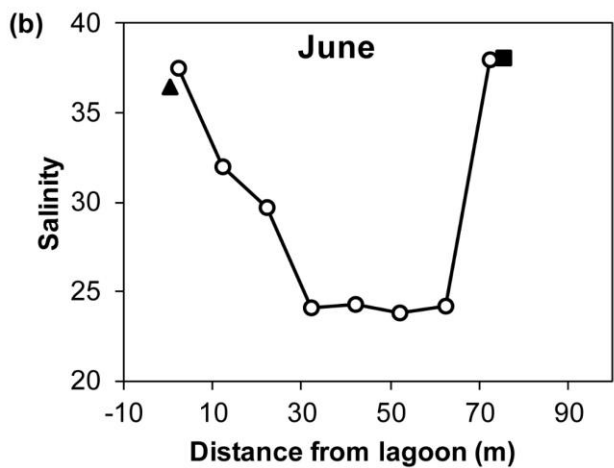
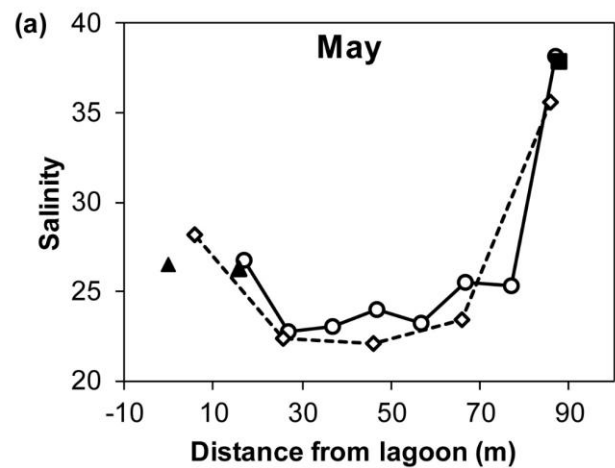
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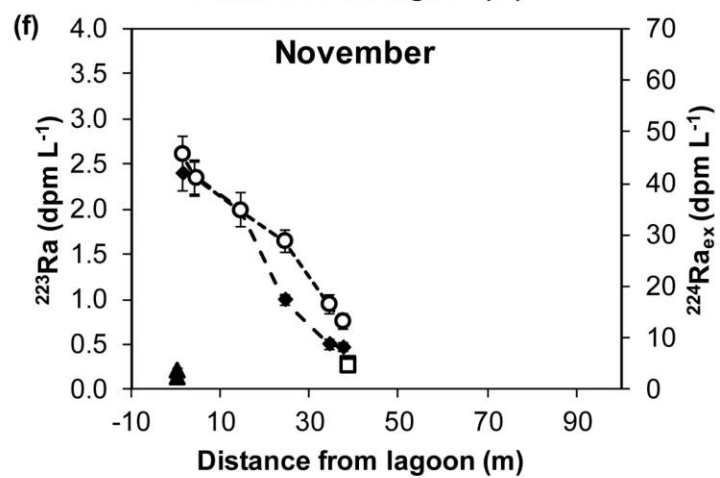
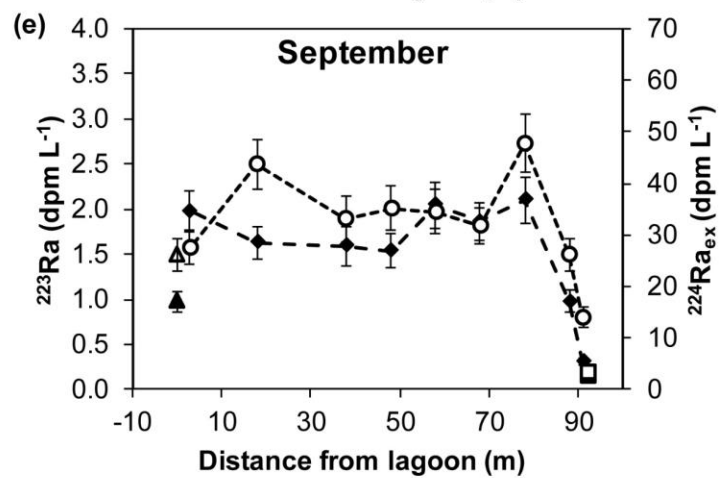
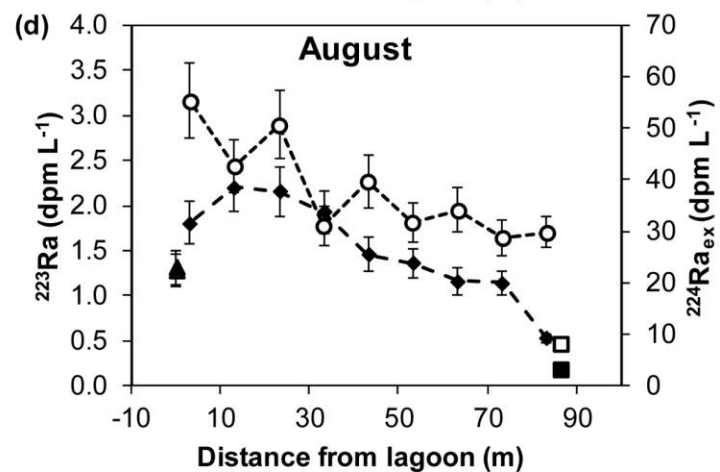
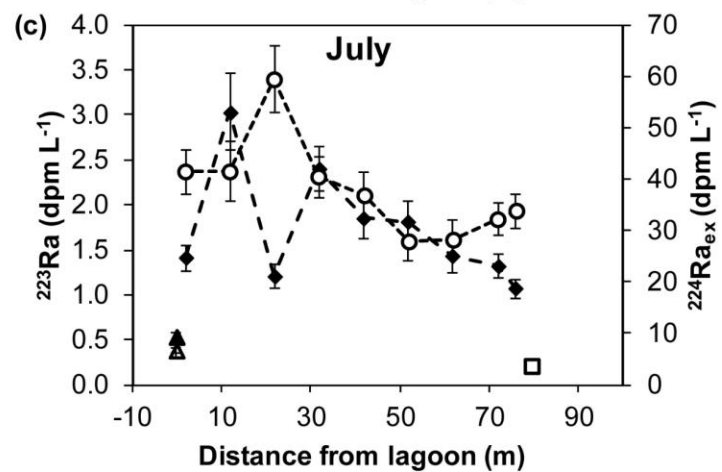
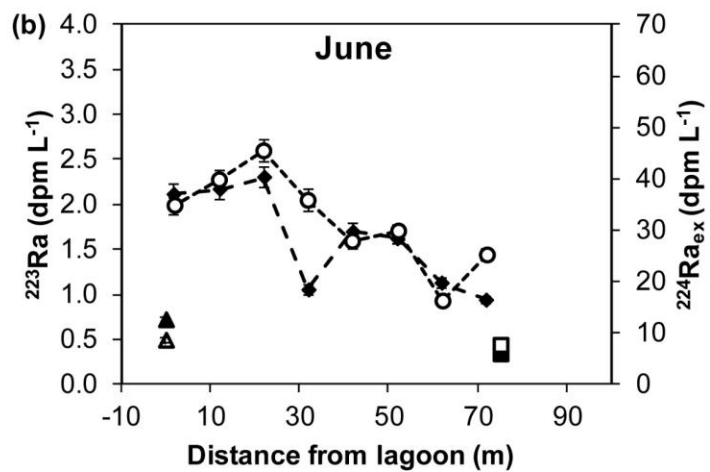
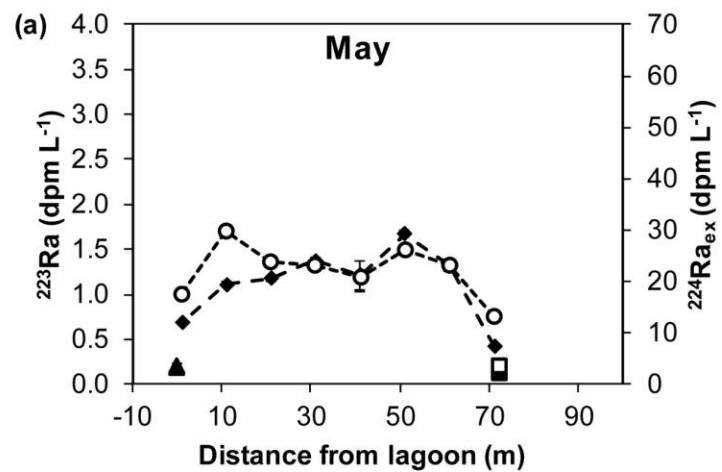


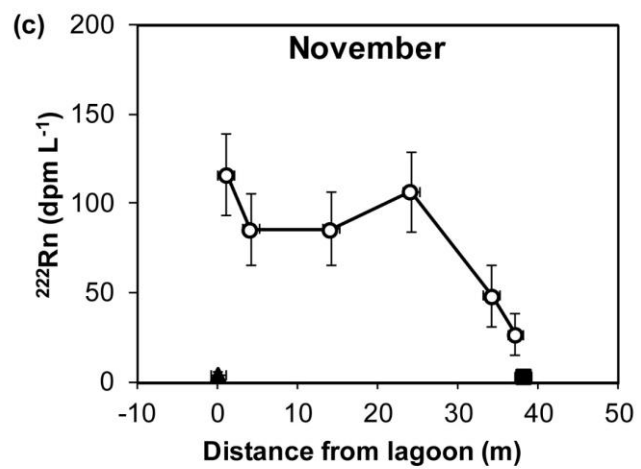
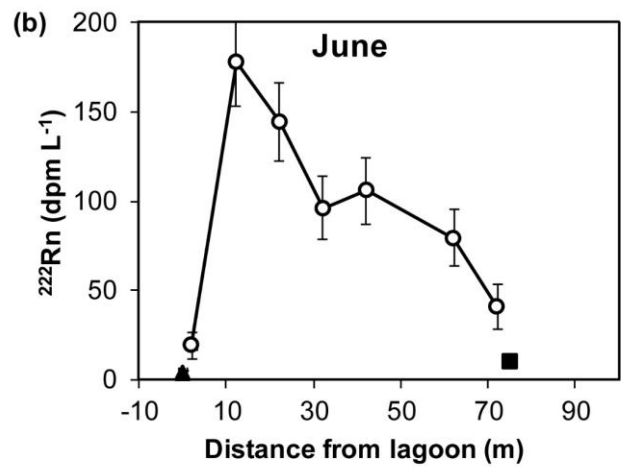
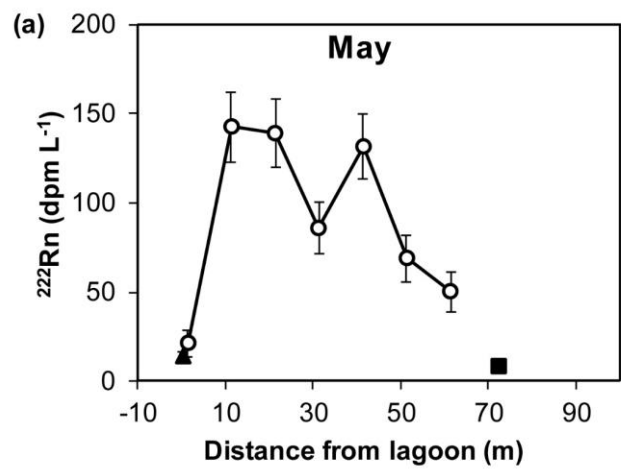


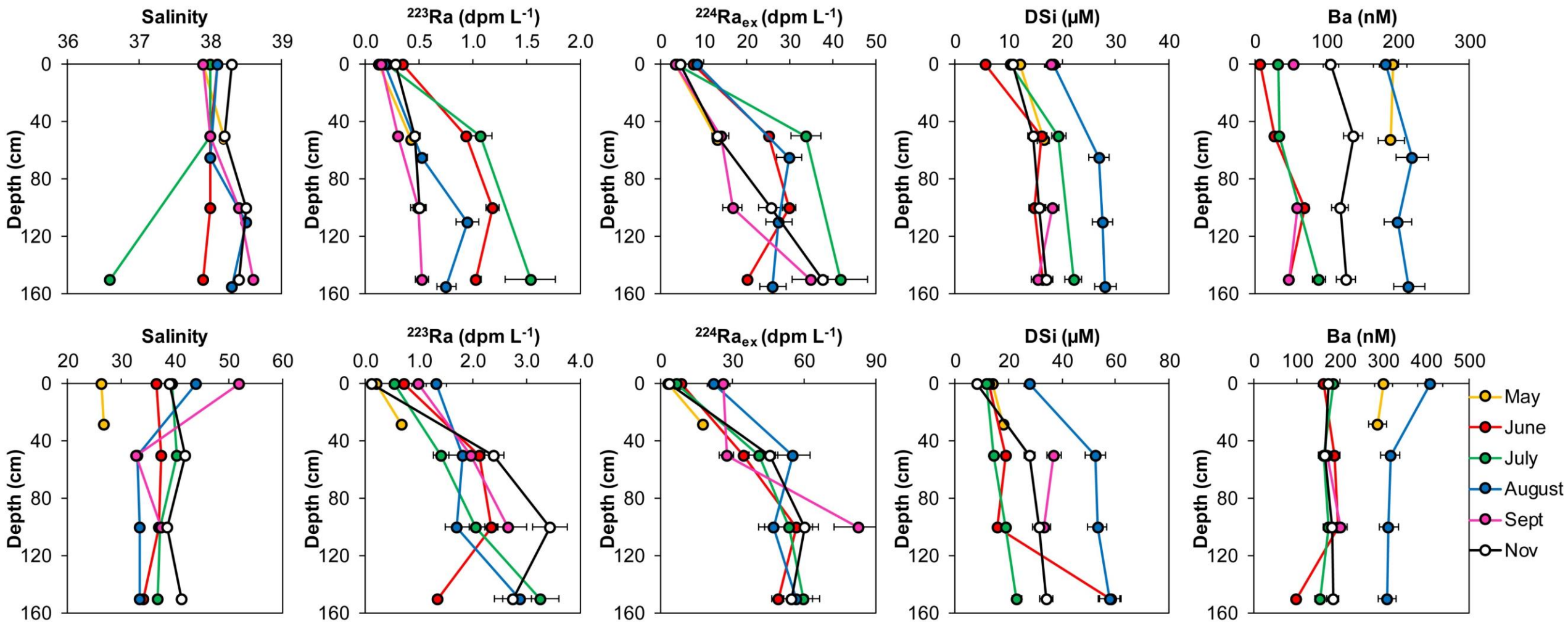


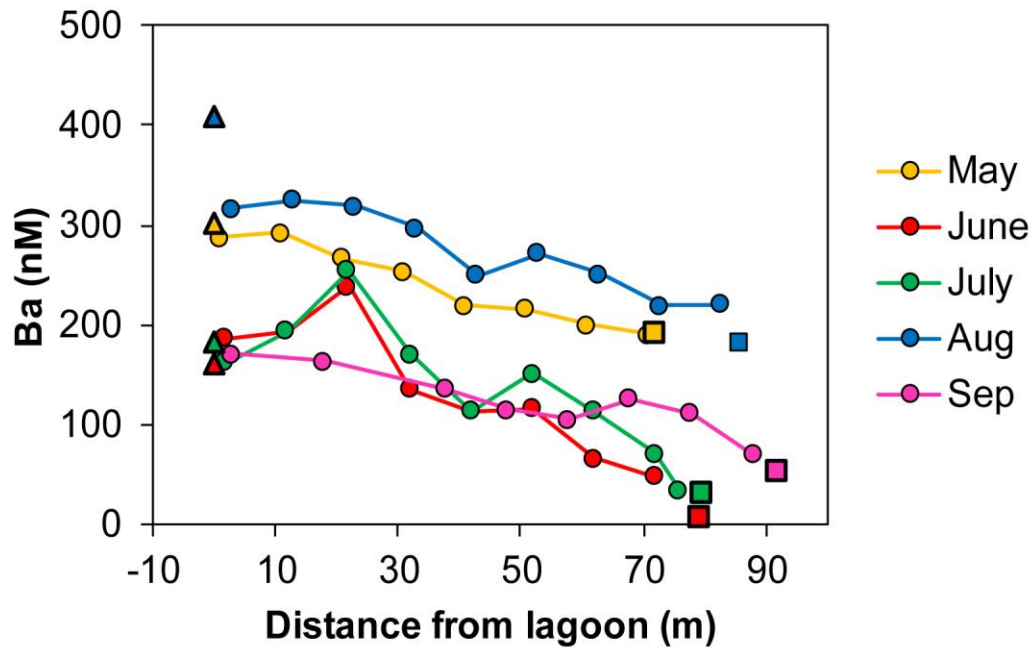
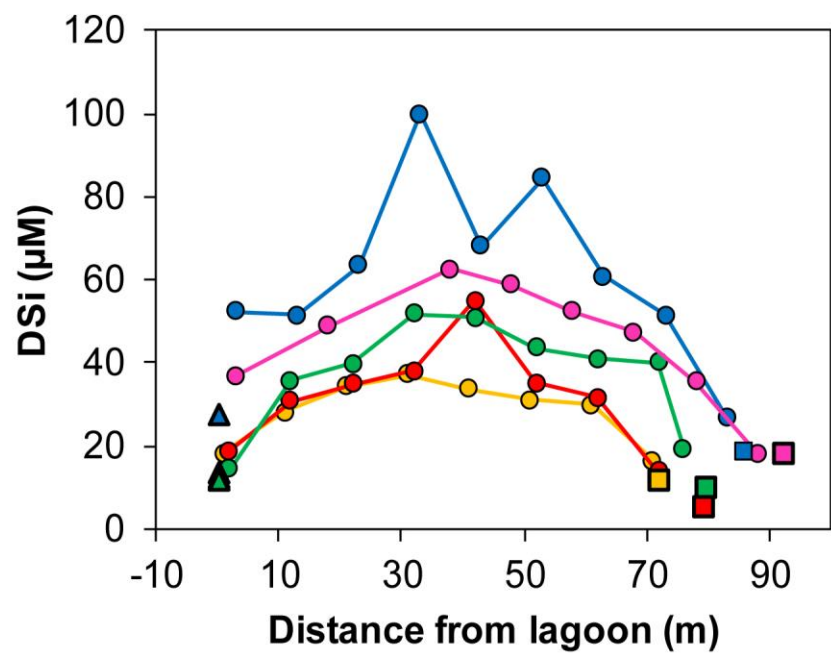
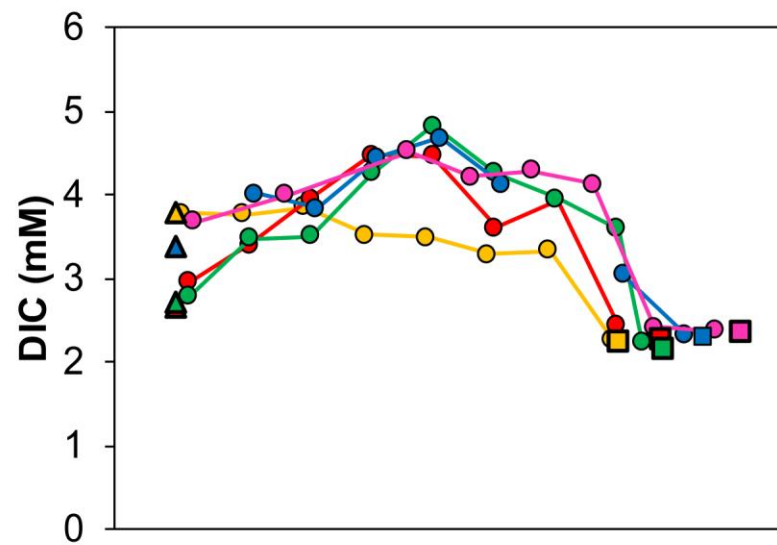
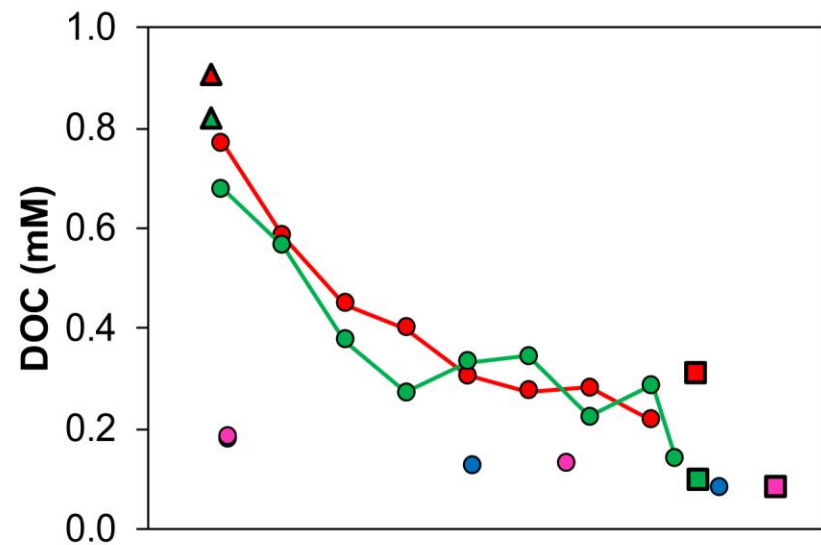






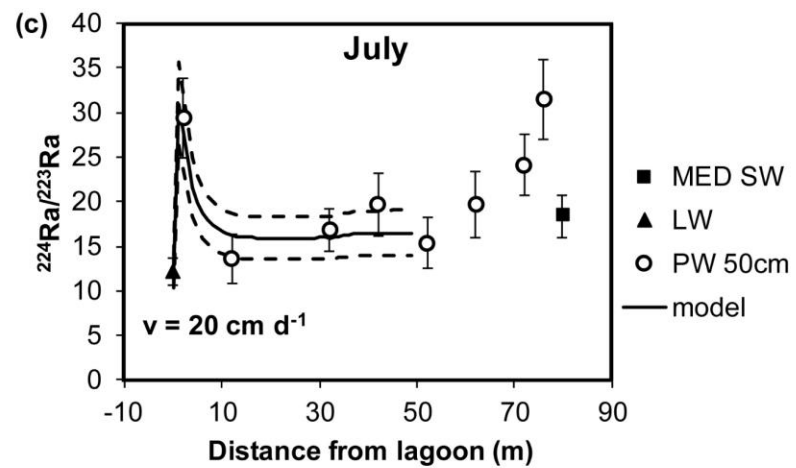
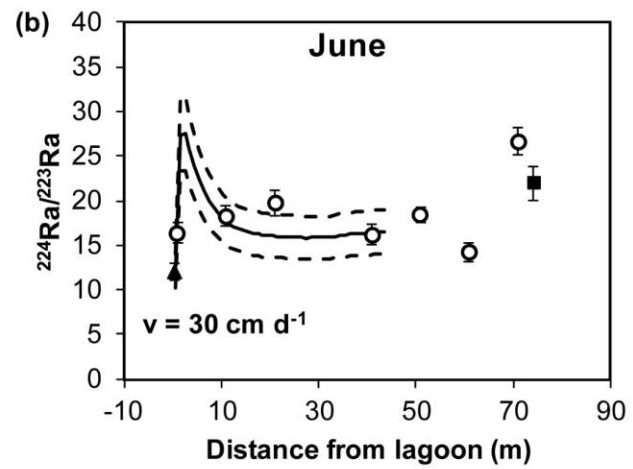
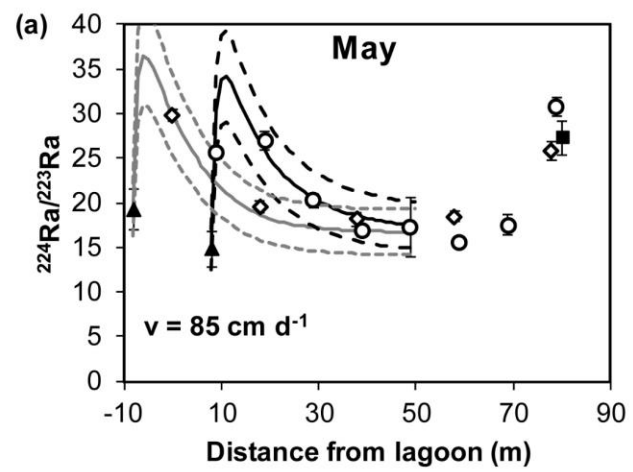


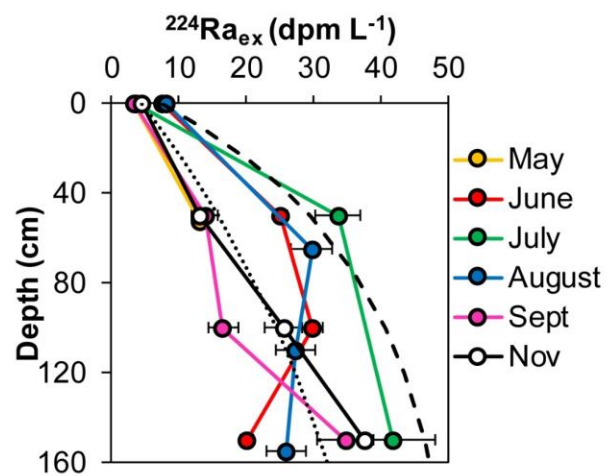
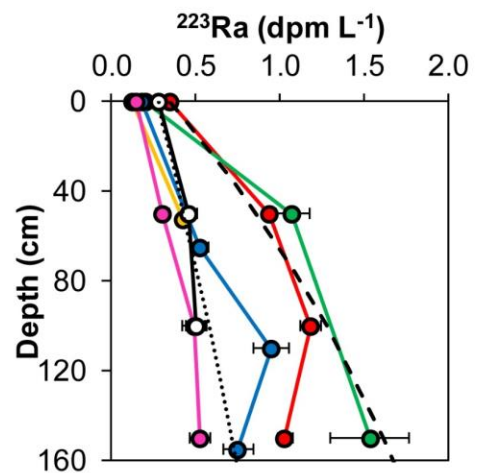
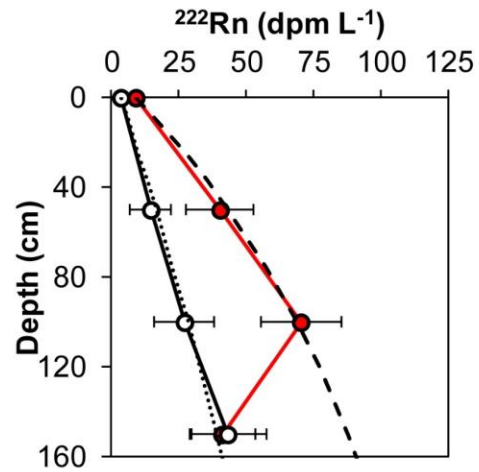




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