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Key Points:

- We predicted an episode of submarine groundwater discharge (SGD) onto the continental shelf, then found supporting evidence for the event
- The prediction linked episodes of SGD to upwelling-favorable winds, which temporarily lower sealevel
- The enhanced SGD lowered dissolved oxygen concentrations to $<130 \mu\text{M}$ and raised radium activities

Supporting Information:

Supporting Information may be found in the online version of this article.

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Predicted Episode of Submarine Groundwater Discharge Onto the South Carolina, USA, Continental Shelf and Its Effect on Dissolved Oxygen

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Abstract Submarine groundwater discharge (SGD) may directly influence the dissolved oxygen (DO) content of coastal bottom waters. Here, we report a predicted episode of enhanced SGD that caused low DO concentrations on the South Carolina continental shelf. The prediction model linked episodes of SGD to upwelling-favorable winds. The data revealed these waters were a factor of 2–6 higher in ^{226}Ra and ^{228}Ra compared to typical bottom water values and were significantly depleted in DO ($<130 \mu\text{M}$). The tight ^{228}Ra : ^{226}Ra correlation of these data was similar to values during a strong hypoxic event off SC in 2012. Water ages from ^{224}Ra and ^{223}Ra indicated the event occurred 2–9 days before sampling. The success of the prediction lends added credence to the correlation of upwelling-favorable winds—but not necessarily accompanied by upwelling—to episodic SGD events. This prediction from wind data represents a major advance for quantifying SGD in the region.

Plain Language Summary Two decades of studies off the coast of South Carolina have demonstrated that saline groundwater is leaking into the ocean from permeable sediment layers (aquifers) that outcrop on the continental shelf. The leakage—called submarine groundwater discharge or SGD—is periodic: more occurs during the summer. SGD adds nutrients, carbon, metals, and radium isotopes to the coastal waters and may deplete dissolved oxygen (DO) concentrations. The radium isotopes are used as tracers to determine the source and volume of SGD being added. However, sampling cruises are rarely timed to coincide with the periodic releases, causing the signal to be mixed away. We recently determined that periods of high SGD correlated with strong southeasterly winds. These winds that blow along the coastline force surface waters offshore. During the displacement sea level is temporarily lowered. We hypothesized that the lower sea level reduces the pressure on the underlying aquifers, allowing more saline groundwater to escape as SGD. In July 2019, upwelling-favorable winds began blowing along the coast. We immediately arranged offshore sample collections. The resulting water samples displayed high concentrations of radium and low concentrations of DO in the bottom waters, confirming our hypothesis.

1. Introduction

Submarine groundwater discharge (SGD) is recognized as a major source of nutrients, metals, natural radio-nuclides, inorganic and organic carbon, and pathogens to coastal waters (Moore, 2010a). This discharge may occur nearshore or throughout the continental shelf (Bratton, 2010; Michael et al., 2016; Moore & Shaw, 1998). The components of SGD include terrestrial groundwater, seawater that has mixed with freshwater and circulated within the aquifer, and reaction products generated as these fluids react chemically with aquifer solids (Moore, 1999; Moore & Joye, 2021). These reaction products serve as tracers, allowing us to recognize and quantify SGD on regional and global scales (Kwon et al., 2014; Moore, 2006, 2007; Moore et al., 2008). Radium isotopes are especially effective tracers (Moore, 2010a) because they integrate SGD over a variety of space and time scales depending on the half-life of the isotope.

Long-lived radium isotopes provide a link to the source(s) of groundwater on the shelf. These isotopes are released to solution after their formation from uranium and thorium decay in aquifer solids. Because solids have varying abundances of these Ra parents, they produce different relative activities of Ra isotopes, which we refer to as activity ratios (AR's). For example, Moore (2003) used $^{228}\text{Ra}/^{226}\text{Ra}$ AR's on the west Florida shelf to distinguish between groundwater from the surficial sandy aquifer (high ^{228}Ra) and deeper carbonate artesian aquifer (high ^{226}Ra). The surficial aquifer solids were presumably higher in ^{232}Th (parent of ^{228}Ra) and the artesian aquifer higher in ^{238}U (parent of ^{226}Ra). By modeling the relative activities of each long-lived isotope in the

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groundwater and bottom water and the residence time of the bottom water, groundwater fluxes (SGD) may be estimated (Moore, 2003).

Intensive studies of radium isotopes in the South Atlantic Bight (SAB), off the southeastern coast of the US, have confirmed that substantial SGD fluxes occur throughout the continental shelf region (Moore, 2007, 2010b; Moore & Shaw, 1998). These fluxes show distinct seasonal differences with highest fluxes occurring in the summer (Moore, 2010b). The flux of SGD may significantly reduce bottom water oxygen concentrations in the SAB (Peterson et al., 2016) due to the input of reducing substances such as sulfide, ammonia, and methane.

Based on thermal anomalies in instruments retrieved from groundwater monitoring wells, George et al. (2020) realized significant pulses of SGD in the SAB 10–15 km off the coast of South Carolina had occurred during the summer of 2016. These pulses, lasting only a few days, occurred after persistent southeasterly, upwelling-favorable winds caused temporary decreases in sea level. They reasoned that the discharge pulses must issue from confined coastal aquifers that outcrop in the study area and proposed that wind-driven variations in sea level caused these pulses when overlying water pressure was reduced. It is the lowering of sea level, not the upwelling, that causes the SGD pulses. This new mechanism for SGD hypothesized that pulses of SGD, which supply nutrients and reduce bottom water oxygen concentrations, could be predicted from real time wind data. Having a prediction model based on real-time data enables sampling cruises to be launched soon after SGD events and to thus verify effects that are soon lost by biological removal or mixing. George et al. (2020) did not have samples in 2016 to verify enhanced release of radium or to study the effects of the release because no cruise had taken place during or immediately after the groundwater pulses were indicated.

2. Study Area

The study area is located on the 100 km wide, shallow continental shelf of the SAB between Charleston and Myrtle Beach, S.C. (Figure 1). Sandy surficial sediments overlie Tertiary carbonates which outcrop as a seaward-thickening series of stacked aquifers and confining units (Harris et al., 2005; Popenoe et al., 1987). George et al. (2020) described shallow monitoring wells offshore Charleston, S.C., that were used to record thermal profiles throughout the year. The wells serve as sampling sites for groundwaters and bottom waters during cruises. Another well (A) without loggers is approximately 20 km offshore N.C. (Moore et al., 2002).

3. Methods

To test the George et al. (2020) hypothesis, we planned to initiate a bottom water sampling cruise to immediately follow a persistent wind event. Such an event occurred during mid-late July 2019 (Figure 2). This prediction of an SGD event and our decision to focus on Wells 11 and 12, based on prior AR's, is verified through e-mail exchanges prior to the cruise (Email Exchanges in Supporting Information S1).

We monitored continuous records of winds and sea water properties from the Coastal Carolina University monitoring site on Apache Pier at Myrtle Beach, S.C., (<http://hydrometcloud.com/hydrometcloud/sutronehome.jsp?menu=index>) to track the course of this event. After recovering from a significant dissolved oxygen (DO) depletion on 9–10 July (Figure S1 in Supporting Information S1), which corresponded to an earlier lowering of sea level, the bottom water at Apache Pier exhibited another DO decrease on 28 July (Figure S1 in Supporting Information S1). We asked Dr. Richard Peterson of Coastal Carolina University to have his students collect samples to measure radium. They collected surface (−2 m) and bottom (−6 m) samples from Apache pier on 1 August 2019. Meanwhile, we organized a sampling trip for 5–6 August to the area offshore Charleston, S.C., described by George et al. (2020) (Figure 1).

Wind data for the Edisto Buoy and satellite measurements of sea surface elevation near the study site were obtained from the U.S. National Data Buoy Center (NDBC, 1971). These data confirmed that sea surface elevation changed in the network area, but the 10-day resolution of this data set was inadequate for our purposes. Instead, we used sea level data from nearby tide gauges, obtained from the NOAA Tides and Currents database (NOAA, 2020).

Temperature logging “stakes” embedded with multiple waterproof temperature loggers (Hobo MX2204 TidbiT, accuracy 0.2°C) were installed to monitor temperature up to 2 m below the seafloor throughout the Charleston well field during the summer of 2019. The design allowed for direct contact of the loggers with the sediment to

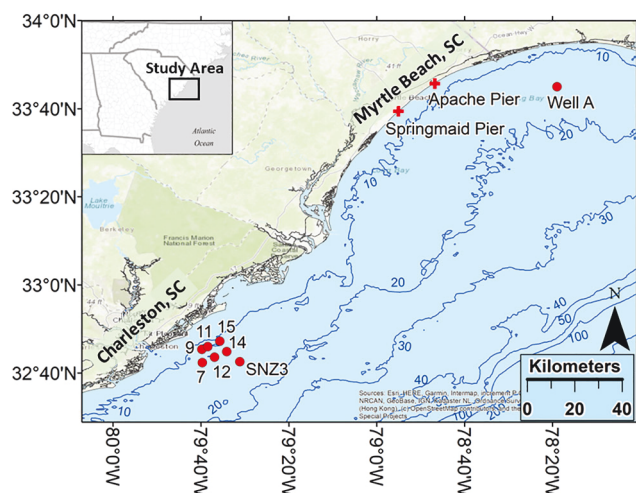


Figure 1. Map of the study area showing locations of wells, sampling stations, and piers.

capture rapid temperature changes and prevent thermal interference between loggers. Groundwater advection was calculated using MATTISI (Wilson et al., 2016), a MATLAB program that estimates groundwater advection based on temperature time series data collected by the loggers. Optimization routines within MATTISI are based on a 1-D heat transport model that simulates heat conduction, advection, and hydrodynamic exchange (Wilson et al., 2016; <https://doi.org/10.5281/zenodo.7082565>).

At each station vertical profiles (surface to bottom) of water column temperature, salinity, pH, and dissolved O_2 were obtained with a YSI model 6820 sonde (<https://www.bco-dmo.org/project/700280>).

Bottom water samples were obtained by lowering a submersible pump to near the seafloor and filling two 26-L carboys. Subsamples for salinity were usually taken from each sample and measured with a YSI conductivity meter to confirm the CTD measurement. Radium samples were filtered through a column of MnO_2 -coated acrylic fiber (Mn-fiber) to quantitatively remove Ra (Moore, 1976). The Mn-fiber was returned to the lab, where short-lived radium isotopes (^{223}Ra , half-life = 11.4 days, and ^{224}Ra , half-life = 3.66 days) were measured using a delayed-coincidence counting system (Moore & Arnold, 1996). After these measurements were complete, Ra and Mn were

stripped from the Mn-fiber and radium was coprecipitated with $BaSO_4$ and transferred to a small tube. ^{226}Ra and ^{228}Ra were measured by gamma spectrometry after ^{222}Rn had equilibrated with ^{226}Ra (Moore et al., 1985).

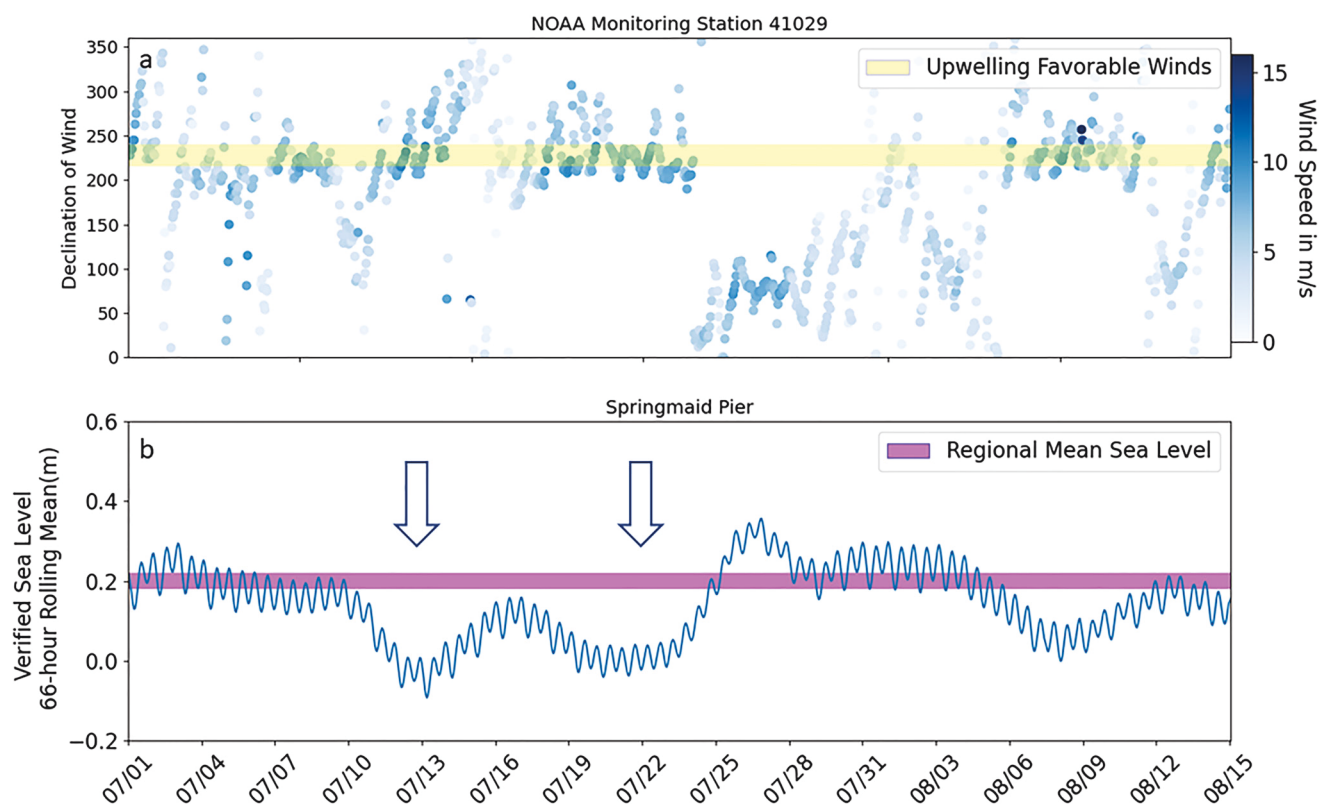


Figure 2. (a) Declination and velocity of wind off the coast of Charleston, S.C., for 1 July–15 August 2019. Yellow shading indicates the declination of upwelling-favorable winds (parallel to the coast). (b) Verified sea level measurements from Springmaid Pier in Myrtle Beach, S.C., (66-hr rolling mean) compared to the summertime regional mean sea level (purple shading). Significant decreases in verified sea level occurred from 10 to 13 and 18 to 23 July; these coincided with upwelling-favorable winds.

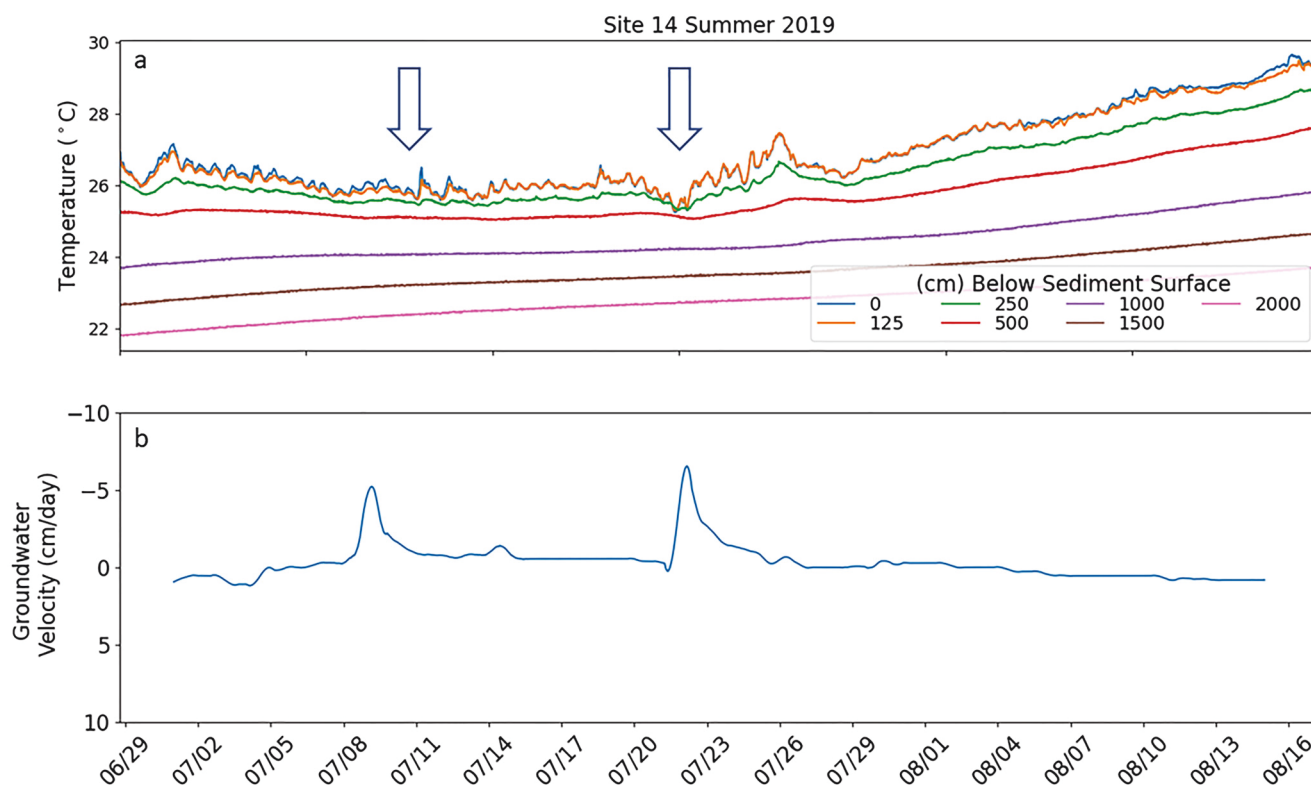


Figure 3. (a) Temperature profiles from site 14 collected at 20-min intervals at multiple depths below the sediment surface. The decrease in temperature at the 0, 12.5, 25, and 50 cm loggers is aligned with the upwelling favorable winds and changes in sea level seen in Figure 2. (b) The temperature data was used in the George et al. (2020) model to identify two pulses of SGD on 9–10 and 22–23 July. Negative velocities indicate discharge from the seabed.

4. Results

During the summer ocean heating causes a positive departure of measured sea level from that predicted from a strictly tidal model (NOAA, 2020). This positive departure is interrupted by periods of sea level lowering that coincide with upwelling-favorable winds (George et al., 2020). In 2019, winds favoring moderate upwelling began the first week of July and coincided with a 25 cm drop in sea level from 10 to 13 July (Figure 2). Around 15 July, the winds slowed and shifted, blowing perpendicular to the shoreline, and causing a brief ~10 cm increase in sea level. From 18 to 23 July, upwelling-favorable winds blew consistently at speeds between 5 and 10 m s⁻¹. These winds again coincided with another drop in sea level accompanied by a sharper decrease in seafloor temperature and discharge of submarine groundwater (Figures 2 and 3).

At Well 14 seawater input (~1 cm/day) to the seabed occurred from the end of June until about 5 July (Figure 3b). This seawater input also drives SGD because the sediments can only temporarily store water (Burnett et al., 2003). The input at Well 14 must later emerge as SGD at Well 14 and elsewhere.

Bottom water samples collected in August 2019 were warm (27.5°C–30°C) and salty (36.0–36.4) in contrast to offshore Charleston samples collected at other times (Table S1 in Supporting Information S1; George et al., 2020; Figure S2 in Supporting Information S1). Two samples with $T \sim 30^\circ\text{C}$ were collected at Apache Pier.

Two of the bottom water samples (W11 and W15) collected off Charleston in August 2019 contained the highest ²²⁶Ra and ²²⁸Ra activities ever measured in the offshore Charleston area (Table S1 in Supporting Information S1). The August 2019 samples from Apache Pier had similar high activities. A plot of ²²⁶Ra versus ²²⁸Ra for the August 2019 data follows a tight linear trend with slope = 1.4 and $R^2 > 0.99$ (Figure 4). The typical value for the Gulf Stream (GS) is shown in blue (Moore, 2007).

The 2019 samples that contained high radium activities also had lower DO concentrations (DO < 130 μM) than samples collected at other times. Figure 5 shows a clear inverse relationship between the bottom water DO

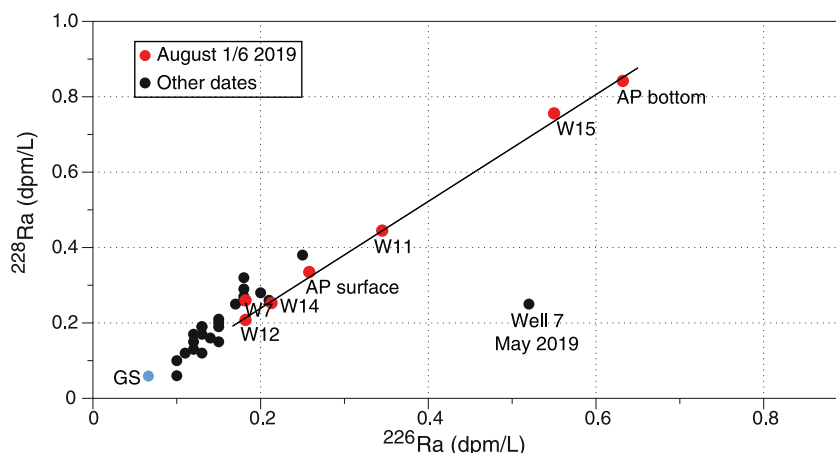


Figure 4. ^{228}Ra versus ^{226}Ra diagram for bottom water samples collected 1–6 August 2019, compared to samples collected at other times. AP is Apache Pier with surface (2 m) and bottom (6 m) samples; site W15 is a relatively shallow eastern location in the Charleston well field (Figure 1); GS is average Gulf Stream data from Moore (2007). In May 2019, a sample enriched in ^{226}Ra but not ^{228}Ra was collected near Well 7. The 2019 data are in Table S1 in Supporting Information S1; earlier data are from George et al. (2020).

content and Ra. The high ^{226}Ra sample collected near Well 7 in May 2019 was also low in DO. Similar relationships between low DO and high Ra were documented by Peterson et al. (2016) at Apache Pier and in the Mississippi Bight by Sanial et al. (2021).

5. Discussion

5.1. Sources of Submarine Groundwater

The high temperatures of the August 2019 bottom water samples (Table S1 in Supporting Information S1) provide convincing evidence that these samples do not represent upwelling from colder regions below the shelf break. Even if the winds caused upwelling, the offshore water did not reach the study area during this investigation. The high salinities of the samples (Table S1 in Supporting Information S1) argue that they do not represent extensions of plumes of freshened water from Charleston Harbor or elsewhere. The only other source of radium enrichment in these bottom waters above sandy sediments is local SGD from shelf aquifers.

George et al. (2020) found differences in ^{228}Ra and ^{226}Ra in water samples from the Charleston offshore wells. Some wells (6, 7, and 9) were much higher in ^{226}Ra indicating they were tapping a source enriched in U (probably carbonate) and others (3, 11, and 12) were higher in ^{228}Ra suggesting the source of Ra in these wells was clastic material more enriched in Th. The relationship between ^{226}Ra and ^{228}Ra in the bottom waters followed a trend suggesting most of the bottom water Ra enrichment was derived from a clastic source with small additions from carbonates (George et al., 2020).

The ^{228}Ra versus ^{226}Ra plot from August 2019 bottom water samples follows a very tight linear trend with a slope of 1.4 and $R^2 > 0.99$ (Figure 4). This is compelling evidence that the radium originated from a clastic source not enriched in uranium. By contrast, the May 2019 bottom water sample near Well 7 indicates this ^{226}Ra enrichment is probably derived from a carbonate source. The lower activity samples from August 2019 intersect the sample array from June 2015 to May 2019, suggesting a common source for radium in these bottom waters throughout the year. August 2019 samples were

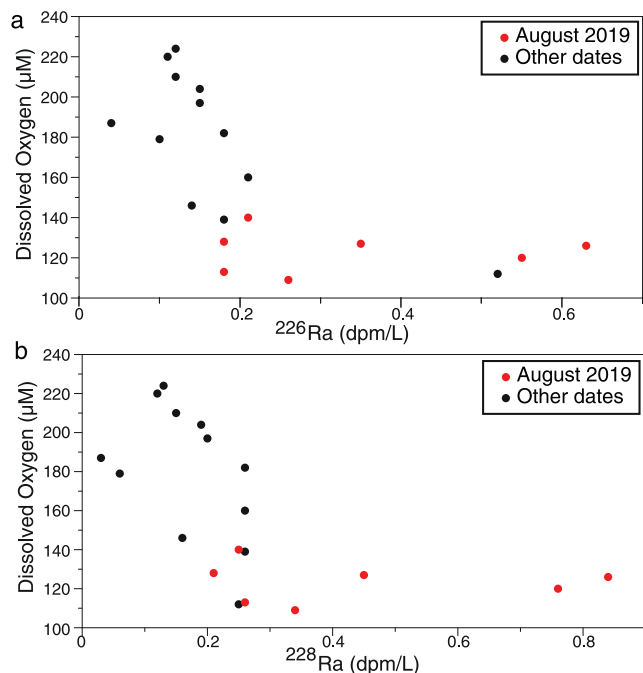


Figure 5. (a) Bottom water dissolved oxygen (DO) versus ^{226}Ra for samples collected 1–6 August 2019 compared to other sampling dates. The May 2019 sample with elevated ^{226}Ra was also depleted in DO. (b) Bottom water DO versus ^{228}Ra for samples collected 1–6 August 2019 compared to other samples.

considerably lower in activity compared to samples reported from Apache Pier in 2012 with a slightly lower slope, 1.4 versus 1.7 (Peterson et al., 2016). There were no bottom water data for the Charleston area during the 2012 event.

5.2. Water Ages

The short-lived Ra isotopes provide an index of water age (Moore, 2000). This age method assumes the Ra-enriched waters have a source with a defined $^{224}\text{Ra}/^{223}\text{Ra}$ AR. Once isolated from the source the AR decreases with time due to radioactive decay. Because decay is exponential and two-component mixing is linear, the ages are affected by mixing with water containing lower Ra activities. Therefore, we call these apparent ages. The equation for calculating ages is

$$T = \frac{\ln \text{AR}_m - \ln \text{AR}_0}{\lambda_{224} - \lambda_{223}} \quad (1)$$

where T is apparent age. AR_m is the measured $^{224}\text{Ra}/^{223}\text{Ra}$ AR, AR_0 is the AR of the source; λ_{224} and λ_{223} are decay constants of ^{224}Ra and ^{223}Ra . To employ this equation, we must determine the AR of the source water (AR_0). The slope of the bottom water $^{228}\text{Ra}/^{226}\text{Ra}$ AR (1.4) is close to that of samples from Well A (slope = 1.35) (Peterson et al., 2016). Assuming groundwater from this formation was the source of the 2019 event, we use the $^{224}\text{Ra}/^{223}\text{Ra}$ AR from Well A (10.6, Figure S3 in Supporting Information S1) as AR_0 to calculate bottom water ages (Table S2 in Supporting Information S1). For the samples from 5 to 6 August, these ages range from 6 to 9 days; for the 1 August Apache Pier samples, the age is 2 days. Thus, the calculated time of the SGD release was 26–30 July 2019, slightly after the modeled SGD release from 22 to 23 July (Figure 3). The difference between the ages probably represents some mixing of radium that lowers the apparent age.

5.3. Flux of SGD

Since 2016, we have continuously logged seabed temperature profiles at the Charleston field site. Observations of groundwater pulses coinciding with cooling events and upwelling favorable winds have occurred multiple times a year (George et al., 2020). Based on the thermal data, submarine groundwater has been calculated to discharge at velocities up to 6 cm/day during periods of continuous offshore prevailing winds and 10 cm/day during major storm events over the field site (George et al., 2020). During these events, discharge velocity varied across wellfield locations and did not seem to be especially localized. It is likely that during the event captured at site 14 based on the July 2019 temperature record (Figure 3) substantial discharge also occurred across the seafloor outside of our study area, as evidenced by the higher radium activities in the eastern samples closer to Myrtle Beach.

Because we do not know the full extent of the bottom water Ra enrichment in August 2019 beyond the Charleston study area, we cannot estimate the total flux of SGD during this event. However, if we assume a 1D system, we can estimate the flux per unit area to the study area for which we have data. Here, we assume there is a small background flux of SGD to the study area, which is enhanced by periodic events. This irregular flux enriches the bottom waters in radium throughout the year. Even without the August 2019 data it is apparent that these bottom waters are enriched in Ra by over a factor of 2 relative to the GS (Figure 4). We want to know how much additional SGD is required to enrich the bottom waters to August 2019 levels, so we average the activities of Charleston bottom water samples collected between June 2015 and May 2019 and subtract this from the average of the activities measured in August 2019 (Table S1 in Supporting Information S1; George et al., 2020). The excess ^{226}Ra is 163 dpm/m³ and ^{228}Ra is 202 dpm/m³. In August 2019 the bottom water thickness based on uniform density was 8.9 m (Figure S4 in Supporting Information S1). Assuming this bottom water was well mixed, the inventory of excess ^{226}Ra in the bottom water was 1,450 dpm/m² and ^{228}Ra was 1,800 dpm/m². Based on the $^{228}\text{Ra}/^{226}\text{Ra}$ AR, we assume the source of the SGD was from an aquifer similar in composition to well A. The median ^{226}Ra activity in well A samples was 5,270 dpm/m³, and for ^{228}Ra it was 7,150 dpm/m³ (Peterson et al., 2016). Thus, the volumes of groundwater required to support the Ra enrichments were 0.27 and 0.25 m³/m² for ^{226}Ra and ^{228}Ra , respectively. Assuming an 8-day residence time for the bottom water (Table S2 in Supporting Information S1), these data translate to an extra SGD flux of 3.4 cm/day for 8 days.

The MATTSI thermal model indicated an SGD flux of 6 cm/day to the Charleston site on 22 July (Figure 3). However the discharge rapidly decreased over the next 3 days to <1 cm/day. To balance the radium enrichment would require an average flux of 3.4 cm/d for 8 days. Thus, it is likely that additional discharge occurred to the east of the Charleston site, perhaps where more permeable sediments outcrop closer to the seabed; then the Ra and DO signals were advected to the west, violating our basic assumption of a 1D system. The higher bottom water Ra activities to the east (Figure 4) reinforce this conclusion.

The 2019 samples that contained high radium activities also contained DO < 130 μ M. As Peterson et al. (2016) have argued, SGD from well A, which is high in ammonia and highly sulfidic, will deplete the DO concentration of the bottom water. We adopt this argument to explain the observed DO depletion in the bottom water and its correlation with high radium. Similar arguments were presented to explain oxygen depletion in the Mississippi Bight by Sanial et al. (2021), who called it a bottom-up mechanism to explain hypoxic bottom waters. Guo et al. (2020) have suggested that SGD contributes to summer hypoxia in the Changjiang (Yangtze) River Estuary.

6. Conclusions

Earlier studies demonstrated a tight connection among persistent offshore winds, sea level and temperature fluctuations in regions of the seabed. George et al. (2020) hypothesized these connections were linked to episodes of enhanced SGD. We tested this hypothesis by sampling bottom waters shortly after such conditions were recognized in late July 2019. The data revealed these waters were a factor of 2–6 higher in ^{226}Ra and ^{228}Ra compared to typical bottom water values and were significantly depleted in DO (<130 μM). There was a tight ^{228}Ra : ^{226}Ra correlation ($R^2 = 0.99$) of these data. Although none of our monitoring wells offshore Charleston had radium AR's matching the AR's in the water column, wells off Myrtle Beach did. Apparent water ages based on ^{224}Ra and ^{223}Ra indicated the event occurred between 26 and 30 July, 2–5 days before collecting the Myrtle Beach samples and 6–9 days before collecting the Charleston samples. Temperature recorders in Well 14 indicated a 6 cm/day discharge event on 22 July; there were no recorders in well A. We conclude the discharge of sulfidic, high radium groundwater at the Charleston site and beyond correlated closely with upwelling-favorable winds. The success of the prediction and verification lends more credence to the correlation of upwelling-favorable winds to episodic SGD events. Our ability to predict these episodic events from easily available wind data represents a major advance for quantifying SGD in the region.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The data on which this article is based are available in Peterson et al. (2016) and George et al. (2020) or are present in the Supplementary Information S1. New radium and CTD data are at <https://www.bco-dmo.org/dataset/882140> and <https://www.bco-dmo.org/dataset/882177>. The MATTSI code is in the Zenodo repository at <https://doi.org/10.5281/zenodo.7082565>.

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