A reevaluation of submarine groundwater discharge along the southeastern coast of North America

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[1] New data of fluxes of radium isotopes and concentrations of radium in coastal groundwater enable an estimation of fluxes of submarine groundwater discharge (SGD) to a large section of the South Atlantic Bight (SAB). The new fluxes are considerably greater than SGD fluxes to this region estimated earlier. The annual average total SGD fluxes, which represent a range of salinity, are 3 times greater than the river fluxes. The highest fluxes of radium and SGD occur in the summer off the coast of Georgia. When scaled per kilometer of shoreline, the SGD flux is the same for the coastlines of Onslow Bay, NC (140 km), a section of the South Atlantic Bight from Onslow Bay, NC, to Cresent Beach, Florida, United States (600 km), and the entire Atlantic Ocean (75,000 km). The fact that each independent estimate is based on different methods and assumptions gives confidence to the results. These SGD fluxes are not restricted to the shoreline but occur throughout the continental shelf. They are important for supplying not only radium but also nutrients, carbon, metals, and freshwater to these coastal waters.

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1. Introduction

[2] Submarine groundwater discharge (SGD) is the flow of water on continental margins from the seabed to the coastal ocean, regardless of fluid composition or driving force [*Burnett et al.*, 2003]. Thus, SGD may be salty or fresh and may be driven by hydraulic forcing from inland aquifers or by ocean forces of waves, tides, storms, and buoyancy (including heat flow). *Moore* [2010] has argued that bioirrigation and shear flow, which affect the exchange of pore water in the upper 2–10 cm of the seabed, not be included in the definition of SGD. This proposed definition would require SGD processes to operate over scale lengths of meters or greater.

[3] An important aspect of SGD is the chemical reaction of mixed fresh and salt water that occurs in coastal aquifers. Biogeochemical processes in this subterranean estuary release some materials from the aquifer framework to solution and sequester other materials. From an oceanographic perspective, the SGD flux is important because it carries nutrients, carbon, and metals to the coastal waters [*Moore*, 1999]. Because groundwater typically contains higher concentrations of nutrients than surface water, it can contribute significantly to nutrient budgets in estuarine and coastal systems [*Valiela et al.*, 1990; *Paerl*, 1997; *Burnett et al.*, 2003; *Kroeger and Charette*, 2008].

[4] The distribution of long-lived radium isotopes in coastal water has served as a proxy for the flux of SGD to the coastal ocean. This technique was developed by *Moore* [1996] and refined by *Moore* [2000], *Windom et al.* [2006], *Hancock et al.* [2006], and *Moore et al.* [2008]. Three important requirements for this technique are knowledge of (1) the ^{226}Ra and/or ^{228}Ra flux into or out of the system, (2) other sources of ^{226}Ra and/or ^{228}Ra to the system, and (3) the ^{226}Ra and/or ^{228}Ra concentration in SGD. With this knowledge a mass balance of one or both of these Ra isotopes can be converted to a flux of SGD.

[5] In 1994 the estuary-shelf-ocean exchange experiment (ESOE) was conducted to develop tracers that could determine the rate of exchange of estuarine and near-shore waters with the open ocean. The experiment was designed to explore the use of ^{223}Ra ($t_{1/2}$ = 11.4 days) and ^{224}Ra ($t_{1/2}$ = 3.66 days) as an index of the exchange time. ESOE was carried out in the South Atlantic Bight (SAB) off the coasts of North and South Carolina in July 1994, during a prolonged period of vertical stratification on the shelf. This vertical stratification isolated surface waters from bottom waters except on the inner continental shelf [*Moore et al.*, 1998]. Samples were collected along five shore-perpendicular transects. Two of these transects were repeated 3 times, and one was repeated 4 times during 10 days.

[6] An unexpected and important result of ESOE was that the SGD flux was estimated to be at least 40% of the entire river flux during July 1994 [*Moore*, 1996]. This result has been widely cited [e.g., *Li et al.*, 1999; *Prieto and Destouni*, 2005; *Destouni et al.*, 2008]. This SGD flux was estimated by adopting the highest value measured for the concentration

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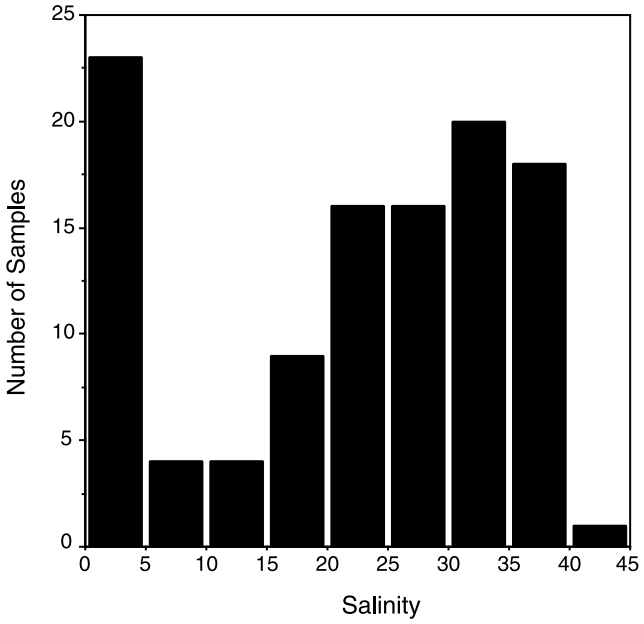

Figure 1. The distribution of salinity in samples of coastal groundwater from the South Atlantic Bight does not follow a normal (in the statistical sense) distribution.

of ^{226}Ra in a few samples of coastal groundwater, a procedure that made the SGD estimate very conservative. This was prudent at the time because only a few samples of salty groundwater had been analyzed for radium. Now this has changed. We have over 100 sites along the SAB where radium isotopes have been measured in groundwater; many of these sites have been measured multiple times [*Moore et al.*, 2008]. Now we can select a more appropriate SGD end-member for calculating the SGD input based on radium isotopes. Additionally, there are now data from four more cruises in these coastal waters that have determined radium fluxes during the summer, early fall, winter, and spring from North Carolina to northern Florida [*Moore*, 2007].

2. Radium Fluxes to the South Atlantic Bight

[7] Based on the ESOE data set, *Moore* [2000] estimated offshore fluxes of ^{226}Ra and ^{228}Ra from the 320 km coastline of the central SAB (Cape Fear, NC, to Savannah, GA). In a larger study *Moore* [2007] provided an assessment of the seasonal distribution and flux of radium isotopes to the SAB based on studies conducted during late summer and early fall 1998, winter 2000, and spring 1999. The study area extended 600 km from Onslow Bay, NC, to Crescent Beach, Florida. By quantifying the inventories of ^{224}Ra and ^{223}Ra in the coastal water, *Moore* [2007] used their decay constants to determine the rate at which each inventory decays. At steady state the decay of ^{224}Ra and ^{223}Ra inventories in the coastal water must be balanced by new inputs of these isotopes. These inputs include (1) riverine, both dissolved and desorbed from particles; (2) regeneration and release from fine-grained particles; and (3) SGD. By evaluating fluxes due to 1 and 2, *Moore* [2007] estimated fluxes of ^{224}Ra and ^{223}Ra due to SGD. This study did not estimate SGD water fluxes.

[8] The long-lived Ra isotopes do not decay significantly in coastal waters; however, their inventories are continually diluted by mixing with the ocean. Therefore, to maintain steady state the observed enrichments of these isotopes in coastal waters must be supplied from the continent or continental shelf. *Moore* [2007] concluded that most of the excess ^{226}Ra and ^{228}Ra in these coastal waters was derived from SGD. This was based in part on new studies of riverine inputs of radium isotopes to the study area [*Moore and Shaw*, 2008]. *Moore* [2007] determined average annual fluxes of ^{226}Ra of 3.0×10^{14} dpm yr^{-1} and ^{228}Ra = 5.8×10^{14} dpm yr^{-1} to this 600 km coastline of the SAB due to SGD.

[9] The excess inventories and fluxes of ^{226}Ra and ^{228}Ra also provided an estimate of the residence time of water on the shelf. These residence times ranged from 30 to 60 days with a mean of about 40 days, similar to the value adopted by *Moore* [1996] based on earlier studies.

[10] During the 1998–2000 cruises, activities of ^{226}Ra and ^{228}Ra were highest off the coast of Georgia. In the summer, high activities extended throughout the study area; but during early fall, spring, and winter, they decreased markedly off the coast of SC and NC. The primary source of excess ^{226}Ra and ^{228}Ra (that is activities in excess of open ocean values) was shown to be SGD. Because the activities of these isotopes in monitoring wells differed little with season, the lower excess activities off South Carolina implied lower rates of SGD during the early fall, spring, and winter, but specific fluxes of SGD were not calculated. In this paper I will combine the offshore ^{226}Ra and ^{228}Ra fluxes from the 1998–2000 studies with concurrent studies of the concentrations of ^{226}Ra and ^{228}Ra in coastal groundwater to estimate SGD fluxes.

3. Concentration of ^{226}Ra and ^{228}Ra in SGD

[11] *Moore et al.* [2008] compiled data on the concentrations of salinity, ^{226}Ra and ^{228}Ra in samples of coastal groundwater taken from monitoring wells and PushPoint samplers throughout the Atlantic margin. About half of these sites were along the coast of the SAB. These data are available as an online supplement to *Moore et al.* [2008]. Each number in this database represents a monitoring well or PushPoint location. When data on multiple samples are available from a single well or PushPoint location, these are averaged to provide one datum per site. Two hundred twenty-six sites are represented in the Atlantic database, with 111 from the SAB. The SAB data represent the following sites: salt marshes (25), beaches (10), islands (19), sounds and embayments (20), sands and shallow limestone on the continental shelf (17), and the Floridan limestone aquifer near the shore (20). With the exception of the U. Floridan wells, most SAB samples were collected 1–4 m below the sediment surface; they cover a range of salinities from 0 to >40. The salinity distribution has a peak in the 0–5 range and another between 30 and 35 (Figure 1). There is no significant correlation between ^{226}Ra and salinity (R^2 = 0.006).

[12] Concentrations of radium isotopes in these groundwater samples are skewed, that is, there are many more

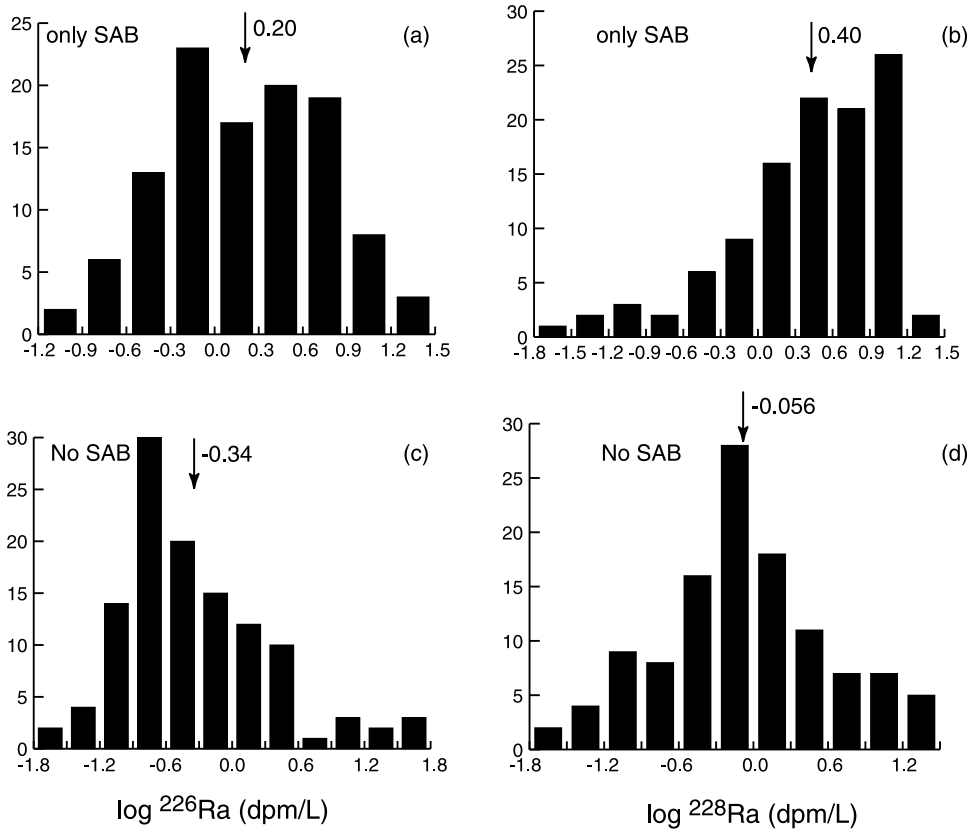

Figure 2. Frequency distribution of ^{226}Ra and ^{228}Ra (log dpm/L) in (a and b) samples collected within the SAB compared to (c and d) samples collected from the rest of the Atlantic margin. The geometric means for each data set are indicated by an arrow. Data from [*Moore et al.* 2008].

samples with low concentrations than with high. In this case it is appropriate to use the log transform of the data to determine the most representative value to use for estimating the groundwater end-member concentration. Figure 2 shows the log transform of the radium isotope distribution at the sites within the SAB compared to the distribution for the remainder of the Atlantic data set. Two trends are apparent: (1) the Atlantic data with SAB samples removed displays a relatively normal peaked distribution and (2) the SAB samples comprise a flatter distribution, with significantly more high values. The geometric means of these data sets are significantly different. Here concentrations are in dpm/L, and the ranges represent ±2 standard errors. For the SAB samples the mean ^{226}Ra is 1.58 with a range 1.24–2.01; the other Atlantic samples have a mean of 0.46 and a range of 0.35–0.62. For ^{228}Ra, the SAB samples have a mean of 2.51 and a range of 1.90–3.31; remaining Atlantic samples have a mean of 0.88 with a range 0.66–1.19. Thus, the SAB samples are about threefold enriched in ^{226}Ra and ^{228}Ra compared to samples from other Atlantic regions. These SAB values are about twentyfold enriched for ^{226}Ra and one hundredfold enriched for ^{228}Ra compared to North Atlantic surface water.

[13] Because of the effect of temperature on the distribution coefficient of radium [*Rama and Moore*, 1996], there could be differences in the concentration of radium in SGD between colder and warmer months. Some of the monitoring wells in the SAB were sampled repetitively over several years during different seasons. Two offshore monitoring wells and eight wells in the Okatee river estuary were sampled enough times to provide a seasonal assessment of possible changes in radium concentration. The offshore wells have been described by *Moore et al.* [2002] and *Moore and Wilson* [2005]; the Okatee wells have been described by *Moore et al.* [2006]. Table 1 summarizes the seasonal data from these locations. There are adequate data to divide the offshore samples into seasonal bins. Samples collected in the winter have a slightly lower mean activity of ^{226}Ra and ^{228}Ra; however, the differences are not significant at a 2 σ standard deviation. There are more wells but fewer sampling campaigns for the Okatee. Here I compare samples taken from December through April (cold months) with the total data set from each Okatee well. There are no statistical differences (at 1 σ) in radium activity in any single well (n = 8) during the colder months.

4. Discussion

4.1. Sources of Radium

[14] Some workers [*Prieto and Destouni*, 2005; *Destouni et al.*, 2008] have argued that the source of the excess ^{226}Ra to coastal waters could be unmonitored freshwater streams

Table 1. Seasonal Averages and Standard Deviations of Salinity and Radium in Offshore and Okatee River Estuary Wells[a]

	Season	Number of Samples	Salinity	^{226}Ra	^{228}Ra
			Offshore Wells		
Well 1	summer	14	35.12 ± 0.54	5.23 ± 1.83	4.94 ± 2.32
Well 1	fall	15	35.09 ± 0.07	4.58 ± 2.09	3.74 ± 2.14
Well 1	winter	4	35.68 ± 0.05	4.13 ± 0.90	3.82 ± 1.24
Well 1	spring	6	35.42 ± 0.04	5.96 ± 1.33	5.77 ± 1.14
Well A	summer	10	34.83 ± 0.31	5.86 ± 1.06	8.14 ± 1.57
Well A	fall	12	35.06 ± 0.16	5.50 ± 0.75	7.55 ± 1.13
Well A	winter	4	35.75 ± 0.17	3.15 ± 0.44	4.45 ± 0.63
Well A	spring	6	35.20 ± 0.09	4.96 ± 1.46	7.24 ± 2.15
			Okatee Wells		
Well 1A	all	8	33.81 ± 3.60	1.65 ± 0.24	12.50 ± 2.08
	winter	3	36.90 ± 0.42	1.67 ± 0.37	13.10 ± 1.97
Well 2	all	9	41.16 ± 5.23	1.42 ± 0.41	14.11 ± 4.09
	winter	4	42.23 ± 3.73	1.33 ± 0.12	13.18 ± 1.51
Well 4	all	9	29.71 ± 3.38	3.15 ± 0.79	11.80 ± 3.60
	winter	4	30.80 ± 2.34	3.14 ± 0.71	11.76 ± 2.80
Well 5	all	10	28.12 ± 1.38	4.69 ± 2.06	9.64 ± 4.09
	winter	5	27.95 ± 1.91	3.62 ± 2.27	7.68 ± 4.97
Well 6	all	8	33.68 ± 2.35	5.28 ± 1.13	13.16 ± 2.87
	winter	3	34.27 ± 0.58	5.13 ± 0.99	12.44 ± 1.61
Well 07	all	9	27.14 ± 0.58	2.87 ± 1.16	6.76 ± 2.85
	winter	3	27.53 ± 0.12	2.05 ± 1.02	4.86 ± 2.71
Well 10	all	8	27.20 ± 0.72	4.57 ± 0.72	12.70 ± 2.13
	winter	3	27.27 ± 0.45	4.51 ± 0.87	12.35 ± 1.81
Well 11	all	9	12.09 ± 1.34	3.72 ± 0.54	35.57 ± 4.69
	winter	5	11.42 ± 1.15	3.77 ± 0.57	35.96 ± 3.33

[a]Seasonal averages and standard deviation are in dpm per liter. The offshore wells have been described by [*Moore and Wilson* 2005] and the Okatee wells by *Moore et al.* [2006].

from coastal catchments. For these to be a viable source of radium to the SAB, the unmonitored sources must constitute ~20% of the monitored flow and the ^{226}Ra concentration must be on the order of 4 dpm L^{-1} [*Destouni et al.*, 2008]. Such high concentrations of ^{226}Ra are unreasonable. No freshwater streams in the region that have been measured contain more than 0.8 dpm L^{-1}; most are <0.1 dpm L^{-1} [*Moore and Shaw*, 2008]. The primary riverine input of radium is due to desorption from suspended particles carried to the estuary [*Li et al.*, 1977]. Rivers that originate in the coastal plain have very low concentrations of suspended solids compared to those that originate in the Piedmont and thus contribute little radium [*Moore and Shaw*, 2008]. Therefore, unmonitored coastal plain streams are expected to provide only a small fraction of the excess radium to these coastal waters.

[15] *Moore* [1996] assumed that the concentrations of ^{226}Ra in SGD were due largely to salt water intrusion into coastal aquifers and desorption of radium. *Moore* [2007] reevaluated the sources of radium and concluded that the desorption mechanism was not adequate to support the measured fluxes unless the average intrusion rate was on the order of 100 m yr^{-1}. *Moore* [2007] concluded that the additional sources of radium were (1) diagenetic reactions with aquifer solids releasing radium to the pore fluids as evidenced by increased alkalinities in the pore fluids [*Burt*, 1993] and (2) regeneration from thorium parent isotopes on aquifer solids. In this case the residence time of water in the aquifer must be long enough to allow ^{226}Ra to regenerate significantly before being flushed; over this time interval, ^{228}Ra reaches secular equilibrium with ^{232}Th and increases no more. Leakage of fluids from the limestone aquifer comprising the continental shelf could enrich the pore waters of overlying sandy sediments in radium as well as other components. This pore water reservoir may release radium continuously or episodically during storms or temperature inversions [*Moore and Wilson*, 2005]. This mechanism is illustrated in Figure 3 and discussed further in section 4.4.

4.2. Fluxes of SGD

[16] Using the Ra fluxes from *Moore* [2000, 2007] and the analysis of the groundwater Ra concentrations from the SAB (assuming the SAB groundwater database represents the concentrations of radium that are discharged as SGD), we can calculate the flux of SGD to the SAB using each isotope. Because a component of the groundwater radium is from seawater that entered the aquifer, the groundwater data must be adjusted to account for this radium. The average ^{226}Ra and ^{228}Ra activities measured on the shelf (0.09 dpm L^{-1} for each) have been subtracted from the groundwater concentrations to estimate the "new" radium added by SGD (Table 2).

[17] The data sets for the offshore and Okatee wells indicate that radium activities in these groundwaters are not statistically different during the winter compared to other months; therefore, a single value is used to represent each well. The SGD flux results are given in Table 2. To compare these fluxes with other studies, I normalize to discharge per kilometer of coastline; the lengths used here are total distances between points as a ship would record sailing along the coast, not the detailed shoreline. This normalization

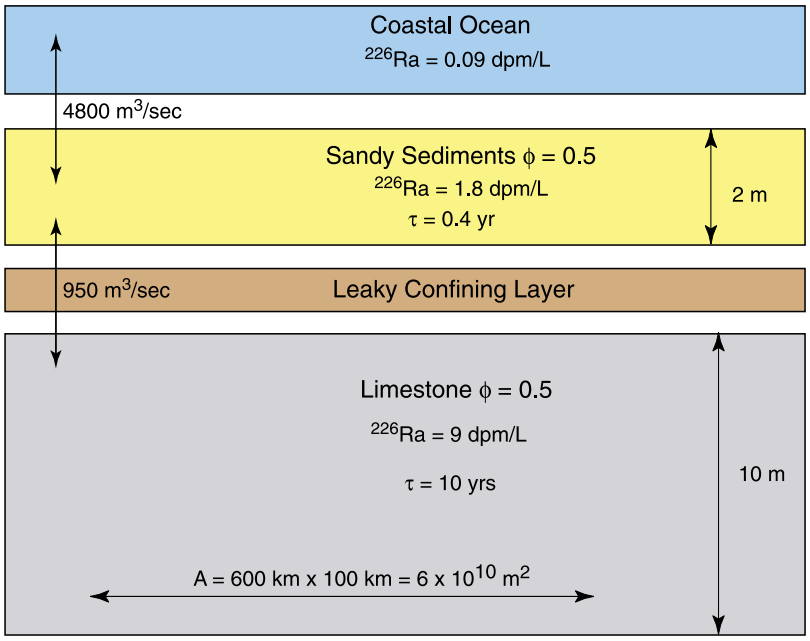

Figure 3. Estimates of fluid exchange from the shelf necessary to balance the ^{226}Ra budget from Cape Fear, NC, to Jacksonville, Florida. Decay of ^{230}Th in continental shelf limestone generates ^{226}Ra, which dissolves into pore fluids. These fluids leak from the limestone and are stored temporarily in the overlying sandy sediments. Periodically, these pore fluids are released to the coastal ocean by storms or thermal inversions.

should not imply that SGD only occurs along the shoreline; there is compelling evidence that it occurs throughout the continental shelf [*Moore and Shaw*, 1998; *Moore et al.*, 2002; *Moore and Wilson*, 2005; *Moore*, 2010]. The average annual fluxes for the 600 km SAB coastline (Onslow Bay to Crescent Beach) calculated from the 1998–2000 data are $(2.01^{+0.59,-0.45}) \times 10^{14}$ L yr^{-1} based on ^{226}Ra and $(2.26^{+0.76,-0.56}) \times 10^{14}$ L yr^{-1} based on ^{228}Ra, values that do not differ statistically. For this discussion, I will use the average of $(2.1^{+0.7,-0.5}) \times 10^{14}$ L yr^{-1}. These SGD fluxes (adjusted for coastline length) are 10 times greater than the initial conservative values estimated by *Moore* [1996] for Cape Fear to Savannah. The difference is explained by two factors: (1) the choice of ^{226}Ra concentration in SGD (to be conservative, *Moore* [1996] adopted 7 dpm L^{-1} compared to 1.58 dpm L^{-1} based on a statistical analysis of the much larger data set used here.) and (2) higher Ra fluxes along the coast of Georgia and northern Florida, an area not included in the study by *Moore* [1996, 2000].

[18] The fluxes of radium change seasonally; but, the concentrations of radium in the groundwater change little, if at all (Table 1). These observations imply that SGD fluxes must also change seasonally, with highest fluxes occurring during the summer and lowest fluxes during the late fall and winter (Table 2). Similar patterns have been found in Rhode Island Sound [*Kelly and Moran*, 2002], the Okatee estuary, SC [*Moore et al.*, 2006], and the salt marsh of North Inlet, SC [*Krest et al.*, 2000].

Table 2. Fluxes of SGD to the South Atlantic Bight Based on Fluxes of ^{228}Ra and ^{226}Ra Due to SGD and on ^{228}Ra and ^{226}Ra Concentrations in SGD[a]

Date	Coast Length (km)	SGD Flux of ^{228}Ra	^{228}Ra in SGD[b]	SGD Flux Based on ^{228}Ra	SGD Flux of ^{226}Ra	^{226}Ra in SGD[b]	SGD Flux Based on ^{226}Ra
Jul 1994	320	1.78[c]	$2.42^{+0.80,-0.61}$	$7.4^{+2.5,-1.8}$	1.00[a]	$1.49^{+0.43,-0.34}$	$6.7^{+2.0,-1.5}$
Sep 1998	600	3.50[d]	$2.42^{+0.80,-0.61}$	$14.5^{+4.9,-3.6}$	1.83[b]	$1.49^{+0.43,-0.34}$	$12.3^{+3.6,-2.8}$
Oct 1998	600	2.50[d]	$2.42^{+0.80,-0.61}$	$10.3^{+3.5,-2.6}$	1.28[b]	$1.49^{+0.43,-0.34}$	$8.6^{+2.5,-1.9}$
Apr 1999	600	3.00[d]	$2.42^{+0.80,-0.61}$	$12.4^{+4.2,-3.1}$	1.67[b]	$1.49^{+0.43,-0.34}$	$11.2^{+3.3,-2.5}$
Feb 2000	600	1.28[d]	$2.42^{+0.80,-0.61}$	$7.1^{+1.8,-1.3}$	0.68[b]	$1.49^{+0.43,-0.34}$	$4.6^{+1.3,-1.0}$

[a]The total SGD fluxes are $2.26^{+0.76,-0.56} \times 10^{14}$ L yr^{-1} based on average ^{228}Ra fluxes from 1998 to 2000 and $2.01^{+0.59,-0.45} \times 10^{14}$ L yr^{-1} based on average ^{226}Ra fluxes during 1998–2000.
[b]Average radium activities have been adjusted for seawater component.
[c]From [*Moore* 2000].
[d]From *Moore* [2007].

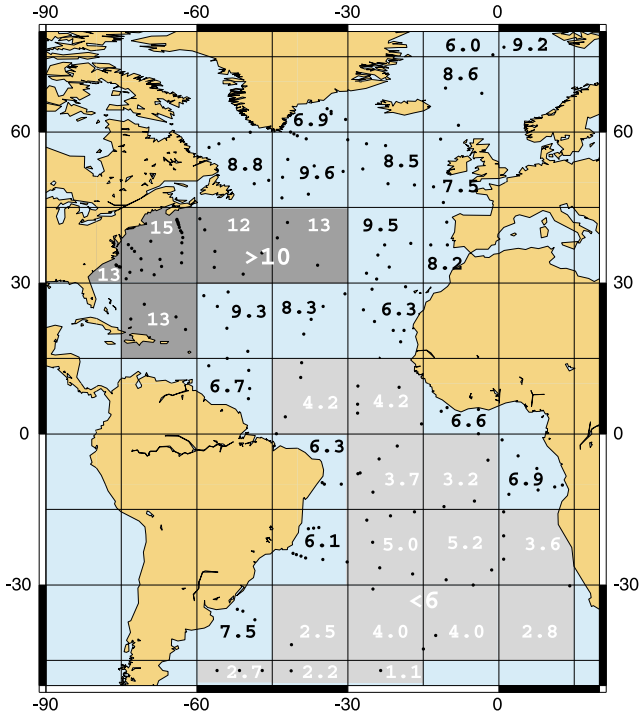

Figure 4. Inventories of ^{228}Ra (10^3 dpm m^{-2}) in the upper km of the Atlantic Ocean increase from south to north, reaching highest values off the coast of North America. Here the values are double the average Atlantic inventory of 6900 dpm m^{-2}. Note that values along continental margins are higher than central ocean inventories, but values off the Amazon River mouth are not greater than other values along the South American margin. The total inventory has been used to estimate total SGD flux to the Atlantic Ocean [*Moore et al.*, 2008].

[19] H. Li et al. (Submarine groundwater discharge through deep multilayered aquifer systems beneath the seafloor, submitted to *Water Resourses Research*, 2010) have proposed a model that separates SGD from surficial and confined aquifers. They conclude that the SGD flux from the confined aquifer, which in their model contributes most of the ^{226}Ra, varies seasonally with a delayed response to changing potentometric heads. Their model is consistent with the patterns of excess ^{226}Ra reported by *Moore* [2007].

4.3. Comparison of These SGD Fluxes to Other Estimates

[20] We can compare the average SAB SGD estimate to other estimates of SGD by dividing our new fluxes by a 600 km coastline to yield $(3.6^{+1.1,-0.9}) \times 10^{11}$ L km^{-1} yr^{-1}. *McCoy et al.* [2007] estimated total SGD fluxes to Onslow Bay, NC, based on ^{222}Rn mass balance models. For total SGD, *McCoy et al.* [2007] estimated 1320 m^3 s^{-1} for 140 km long Onslow Bay or 3.0 x 10^{11} L km^{-1} yr^{-1}, remarkably similar to the estimates given here, when normalized to the shoreline length.

[21] To maintain the inventory of ^{228}Ra in the upper km of the entire Atlantic Ocean, *Moore et al.* [2008] estimated SGD fluxes to the Atlantic of $(3.5 \pm 1.2) \times 10^{11}$ L km^{-1} yr^{-1}, a value indistinguishable from the fluxes to the SAB.

[22] To establish the Atlantic inventory of ^{228}Ra, *Moore et al.* [2008] mapped the distribution of ^{228}Ra in the upper 1000 m of the Atlantic Ocean. Figure 4 shows a new version of this inventory map (expressed as average inventories, 10^3 dpm m^{-2}, in 15° × 15° boxes) accentuating regions having $<6 \times 10^3$ dpm m^{-2} and $>10 \times 10^3$ dpm m^{-2} inventories. It is clear that the South Atlantic and Southern oceans contain considerably less ^{228}Ra than does the North Atlantic. The south to north increase in inventory tracks the pattern of the upper ocean conveyor that carries surface water from south to north in the Atlantic. To maintain steady state, the northward moving surface water must receive ^{228}Ra from the continents at a rate greater than the decay rate of ^{228}Ra (12% per year), causing the inventory to increase to the north. Inventories in the North and South Atlantic are generally greater in the boxes intersecting coastal regions. Interestingly, the inventory in the box intersecting the Amazon River mouth is not significantly greater than in boxes intersecting other parts of the South American continent. This supports the conclusion of *Moore et al.* [2008] that riverine and sedimentary input of ^{228}Ra are relatively minor sources compared to SGD.

[23] The highest inventories of ^{228}Ra are found off the coast of North America; these are about a factor of 2 greater than the average Atlantic value of 6.9×10^3 dpm m^{-2}. Some of the increase is explained by the fact that these surface waters have received ^{228}Ra during their passage through the Atlantic. Another factor must be the higher concentrations of ^{228}Ra in submarine groundwater discharging in this region, considering that the average SAB fluid fluxes are similar to the average Atlantic flux.

4.4. A Conceptual Model of SGD on the South Atlantic Bight

[24] Is the magnitude of these very large SGD fluxes realistic? If all of this water flowing to the SAB passed through the shoreline to a depth of 10 m, the flux would be 90 m^3 m^{-2} d^{-1}, implying a flow rate (assuming 50% porosity of the aquifer) of 180 m d^{-1}. Such widespread flow in permeable sediments is not realistic. Instead, consider vertical flow through the continental shelf. The width of the 600 km SAB shelf considered here is about 100 km and is covered by about 2 m sand. Thus, the water reservoir within the sand (assuming 50% porosity) is 6×10^{13} L. To provide a SGD flux of 2×10^{14} L yr^{-1}, the sand pore water must be replaced about 3 times per year (average vertical flow = 5 m yr^{-1}). This is reasonable as episodic flushing of the sand because of storms and temperature inversions is well documented [*Moore and Wilson*, 2005]. This does not rule out some shoreline flow; it is likely that the total SGD flux is composed of shoreline and shelf fluxes.

[25] If the sandy shelf sediments provide the reservoir for much of the SGD, their pore fluids must contain the requisite concentrations of radium to explain these fluxes. Because ^{226}Ra regenerates very slowly, there is not enough time within the sand reservoir to produce significant activity on a flushing time scale of months. There must be another

source of ^{226}Ra to these pore fluids. Could this source be the underlying limestone foundation?

[26] I constructed a conceptual model to determine if the required flux of ^{226}Ra could be sustained by regeneration from the decay of ^{230}Th contained in shelf limestone and then flushed through the sands to the coastal ocean as SGD. Figure 3 illustrates the features of the model; note that the Ra activities and time scales are not well constrained by data but are forced to be consistent in the model. The ^{226}Ra is generated by decay of ^{230}Th present in the limestone that makes up the shelf platform. Other sources, such as dissolution of the limestone or accessory minerals, are neglected. Taking a density of 2.5 g cm^{-3}, a porosity of 0.5, and using average values for U and Th in limestone [*Gascoyne and Schwartz*, 1982], the limestone contains about 1 dpm ^{230}Th cm^{-3}. In short term, this will generate ^{226}Ra at the rate of 4.3×10^{-4} dpm cm^{-3} yr^{-1}. Thus, a 1 cm^2 × 10 m section of the limestone will generate ^{226}Ra at a rate of 0.43 dpm cm^{-2} yr^{-1}. At steady state the radium produced will desorb to pore fluids (volume of a 10 m section = 0.5 L cm^{-2}) and the ^{226}Ra concentration will increase at the rate of 0.9 dpm L^{-1} yr^{-1}. After 10 years, the pore fluid concentration will be 9 dpm L^{-1}. This is near the upper end of concentrations we have measured in offshore monitoring wells set into the limestone [*Moore et al.*, 2002]. If we take the residence time in the upper limestone to be 10 years, the pore fluids must exchange to a depth of 10 m at a rate of 0.5 m^3 m^{-2} yr^{-1}. Thus, the ^{226}Ra flux will be 4500 dpm m^{-2} yr^{-1}. If this occurs over the entire shelf (6×10^{10} m^2), the flux will be 2.7×10^{14} dpm yr^{-1}, close to the estimated ^{226}Ra flux of 3×10^{14} dpm yr^{-1} [*Moore*, 2007].

[27] In this model the radium released from the limestone is stored temporarily in the overlying sandy sediments. If the water volume of the sand reservoir is 6×10^{10} m^3 (2 m thick × 6×10^{10} m^2 × 0.5 porosity) and the residence time is 0.4 years, the average ^{226}Ra concentration in the pore water due to leakage from the limestone will be 1.8 dpm L^{-1}. This is in the lower range of values we measure in pore water extracted from shelf sands [*Moore and Wilson*, 2005] and only slightly higher than the geometric mean (1.6 dpm L^{-1}) for the SAB groundwater database. This sand pore water will exchange with the overlying ocean at a rate of 2.5 m^3 m^{-2} yr^{-1} (i.e., the volume will be flushed 2–3 times per year), yielding a flux that balances the ^{226}Ra flux from the limestone, 2.7×10^{14} dpm yr^{-1}.

[28] In this simple model I assume that most of the water flushed from the sand and limestone is replaced by seawater with a near zero ^{226}Ra concentration. This is probably a good assumption for the sand pore water. The limestone could also be receiving water from terrestrial sources or from water stored deeper in the formation. These additional sources would reduce the amount of ^{226}Ra required from regeneration. The point here is that the ^{226}Ra flux can be balanced realistically by regeneration in the limestone, although this certainly does not eliminate other sources.

4.5. Comparison of SGD to Surface and Subsurface Freshwater Fluxes

[29] About 80% of the SAB groundwater samples have a salinity below the lowest normal SAB seawater value of 34% and 65% are below salinity 29. Because the distribution of salinity in these samples is bipolar (Figure 1), it is not possible to assign a probable value to the salinity of SGD in this region. If most of the SGD flux is from the shallow shelf limestone, the freshwater component is probably low. Nevertheless, the fact that so many samples are considerably below the salinity of the SAB implies that there is likely a significant freshening of SGD entering the SAB.

[30] *McCoy et al.* [2007] estimated both total and freshwater SGD fluxes to Onslow Bay, NC, based on ^{222}Rn mass balance models. For the freshwater component of SGD, they estimated 260 m^3 s^{-1} or 5.9×10^{10} L km^{-1} yr^{-1}, 3 times greater than the river flux to Onslow Bay. This value is about 20% of the value they estimated for the average total SGD flux, implying that the average salinity of the SGD entering Onslow Bay is 20% fresher (salinity = 28) than the seawater that entered the aquifer. The fact that 65% of samples in the SAB SGD database are less than 29 provides support for the fresh SGD estimate of *McCoy et al.* [2007].

[31] There have been other attempts to estimate the regional freshwater component of SGD. Using what they call an integrated hydrologic-hydrogeologic approach, *Zektser et al.* [2007] estimated fluxes of fresh groundwater into the ocean. This approach assumes that the groundwater input to rivers (L km^{-1} yr^{-1}) that drain specific hydrogeologic provinces is similar to groundwater discharge to the ocean (L km^{-1} yr^{-1}) from these provinces. They estimated the groundwater input to the rivers based on published studies and scaled the expected discharge per kilometer of river with the shoreline length of each province to provide groundwater fluxes from each province to the ocean. They note that this approach only includes fluxes from the upper (shallow) zones of the provinces (the same zones that drain into rivers) and may miss fluxes from deeper zones (confined aquifers) that drain into the ocean. *Zektser et al.* [2007] provide a detailed list of discharge and water composition from each province into individual ocean basins. The water compositions are in almost all cases <1 g L^{-1} total dissolved solids; thus, their estimate is essentially freshwater SGD. *Zektser et al.* [2007] estimate the flux of fresh groundwater to the Atlantic Ocean for the hydrologic province between the Hudson and Savannah rivers to be 6.3×10^9 L km^{-1} yr^{-1} and for the Florida Peninsula to be 10.6×10^9 L km^{-1} yr^{-1}. Since the SAB extends over both of these provinces, I will take an intermediate value of 8×10^9 L km^{-1} yr^{-1}. This is less than 15% of the freshwater SGD flux to Onslow Bay estimated by *McCoy et al.* [2007], which includes additions from deeper aquifers. If the estimate of the freshwater SGD flux by *Zektser et al.* [2007] is correct, this would imply that the average salinity of the SGD is only reduced by 0.85, i.e., from 35.00 to 34.15. This is not consistent with the large number of samples in the SAB groundwater database having salinity less than 34. I conclude that *Zektser et al.* [2007] underestimated the freshwater flux to the SAB.

[32] The average total river flux to the SAB is 6.6×10^{13} L yr^{-1} [*Pomeroy et al.*, 1993]. This is about one third the total SGD flux to the region considered here (Table 2). Furthermore, SGD usually contains higher concentrations of

nutrients [*Paerl*, 1997; *Moore et al.*, 2002; *Burnett et al.*, 2003; *Kroeger and Charette*, 2008], metals [*Shaw et al.*, 1998; *Windom et al.*, 2006; *Beck et al.*, 2007], and carbon [*Cai et al.*, 2003; *Goni and Gardner*, 2003] than surface waters. Thus, SGD fluxes are probably much more important than rivers in controlling fluxes of these materials to the coastal ocean in this region.

5. Conclusions

[33] Regional estimates of total SGD to (1) Onslow Bay, NC (140 km coastline, *McCoy et al.* [2007]), (2) the South Atlantic Bight from Onslow Bay to Crescent Beach, Florida (600 km coastline, this study), and (3) the entire Atlantic Ocean (75,000 km coastline, *Moore et al.* [2008]) are indistinguishable when scaled to a kilometer of coastline. This remarkable agreement from three independent techniques gives confidence that total SGD fluxes to the ocean are large and important. The magnitude of the flux requires that a significant fraction must pass through sandy sediments on the continental shelf. This sandy reservoir may release fluids continuously or episodically. Leakage of fluids from the underlying limestone may provide a source of radium, nutrients, metals, and carbon to the sandy sediments.

[34] The revised total SGD flux to the SAB is 3 times greater than the river flux. Because the concentrations of nutrients, metals, and carbon are typically greater in SGD compared to rivers, the SGD source is almost certainly more important in supplying these materials to this and to other coastal regions. Although the freshwater component of the SGD flux is not resolved for the entire SAB, ^{222}Rn studies [*McCoy et al.*, 2007] and the distribution of salinity in coastal groundwater (this study) imply that significant freshwater enters the SAB from SGD.

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