DETERMINATION OF SUBMARINE GROUNDWATER DISCHARGE (SGD) VIA NATURAL RADIONUCLIDES IN A REGION NEAR THE MOUTH OF THE YELLOW RIVER

WILLIAM C. BURNETT[1], Richard Peterson[1], Isaac Santos[1], Makoto Taniguchi[2], and Tomotoshi Ishitobi[2]

1 Department of Oceanography, Florida State University, Tallahassee, Fl 32306, USA
2 Research Institute for Humanity and Nature (RIHN), Kyoto, 603-8047, Japan

Abstract: Naturally occurring radionuclides have been shown to be useful tracers of submarine groundwater discharge due to their high concentrations in groundwater and relatively low concentrations in surface seawater. We present here an assessment of SGD rates using ^{222}Rn and Ra isotopes from an area of high SGD to the south of the Yellow River. The data were collected during a 24-hour stationary time series analysis in September 2006. Our results based on a radon mass balance model indicate average SGD rates of ~ 37 cm/day. Average rates based on short-lived radium isotopes are similar, at 40 and 37 cm/day for ^{223}Ra and ^{224}Ra, respectively. While all of these estimates depend upon assumptions involving residence times and end-member values, the results are internally consistent and agree well with values reported for an automatic seepage meter deployed nearby (av = 44 cm/day).

Key words: Submarine groundwater discharge, radium, radon, tracers

Introduction: Submarine groundwater discharge (SGD) can be an important pathway for biogeochemical constituents to the coastal zone. There is a mounting degree of evidence that the Bohai Sea has been increasing in its inorganic nitrogen species over the past several decades (Zhang et al., 2004). It is possible that subterranean inputs, often greatly enriched in nitrogen relative to phosphate, may be part of the explanation. It is well known that groundwater N:P ratios often exceed those of surface waters by substantial amounts (Slomp and Van Cappellen, 2004).

Study Site and Methods: We occupied a station (A-1) about 1 km offshore, just a few kilometers south of the Yellow River delta, over a 24-hour period during September 21-22, 2006. This site was within about 50 m of an automated seepage meter (Taniguchi and Iwakawa, 2001) that was deployed from September 18-21, 2006. Salinity and temperature were recorded continuously from outside the seepage meter chamber. We collected 100-L samples once per hour for analysis of short-lived radium isotopes by methods described in Moore and Arnold (1996). Radon was measured continuously over the 24-hour period using an automated radon detector (Burnett et al., 2001).

Results and Discussion: The concentrations of ^{223}Ra ($T_{1/2}$=11.2 days) and ^{224}Ra ($T_{1/2}$=3.6 days) show systematic changes over the 24-hour time series (Fig. 1). In order to use this information to estimate the total (saline + fresh water) SGD, we need to make assumptions about the groundwater end-member concentrations and the residence time of the water being sampled. We can then estimate the flow rates from the following equation:

$$SGD\,(\text{cm/day}) = \frac{\text{Excess}\ {}^{224}Ra\,(dpm/m^3) \bullet Depth\,(m)}{\tau\,(\text{days}) \bullet {}^{224}Ra_{gw}(dpm/m^3)} \bullet \frac{100\,cm}{m} \qquad (1)$$

A similar equation can be written for ^{223}Ra. The "excess" in this case refers to the activity of the radium isotope above that present in Bohai Sea water (^{223}Ra ~20 dpm/m^3, ^{224}Ra ~360 dpm/m^3; n=4). The residence time, τ, can be estimated by a relationship between radium isotope activity ratios in the source waters (assumed to be SGD in this case) and the same ratios in the overlying receiving waters (Moore et al., 2006):

$$\tau = \frac{(^{224}Ra/^{223}Ra)_{gw} - (^{224}Ra/^{223}Ra)_{sw}}{(^{224}Ra/^{223}Ra)_{sw} \bullet (\lambda_{224} - \lambda_{223})} \qquad (2)$$

Based on the assumptions one makes concerning the end-member ratio, the calculated residence times are in the range of 0.7-2.2 days. We chose to estimate the residence time of the waters at our station at 1 day, the length of a complete tidal cycle. The final term in equation 1, ^{224}Ra$_{gw}$, refers to the activity of the radium isotope in the advecting groundwater. We have chosen to use activities of 120 and 3600 dpm/m^3 for the groundwater values of ^{223}Ra and ^{224}Ra, respectively. These values are both somewhat higher (within about 20%) than the activities measured in a sample collected from a seepage meter in 2005.

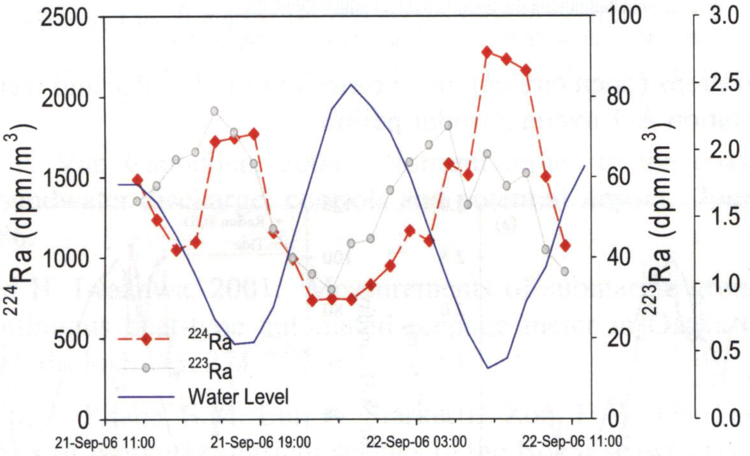

Figure 1 Activities of ^{223}Ra and ^{224}Ra over a 24-hour period at station A-1.

Using average values for the two isotopes of radium during the time series and the assumed residence time and end-member values, we estimate SGD advection rates at 40 and 37 cm/day for ^{223}Ra and ^{224}Ra, respectively. The average advection recorded by the seepage meter over the same time interval was 44 cm/day.

The results of the continuous radon measurements (Fig. 2) show that while the concentrations of ^{222}Rn did not vary more than about 10%, there were huge changes in the inventories (concentration x water depth). After correcting for atmospheric evasion and losses due to mixing, the changes in the inventories per unit time, can be converted to radon fluxes to achieve a mass balance. Thus, if one knows or can estimate the concentration of radon in the fluids advecting into the system, one can estimate the water flux (Burnett and Dulaiova, 2003).

We did not have an analysis of ^{222}Rn from seepage meter water so we estimated the end-member radon by multiplying the average high ^{222}Rn/^{224}Ra activity ratio (AR) in the water column (AR~11) by the end-member ^{224}Ra (3,600 dpm/m^3) for an estimated ^{222}Rn end-member (36,900 dpm/m^3). We feel this approach is appropriate since the advecting pore waters are likely the major source of both these tracers. Our model results show a pattern and magnitude of SGD very close to what was observed by the automatic seepage meter (Fig. 3). The seep meter average and standard deviation for this period was 44±20 cm/day while the radon model average result is 37±30 cm/day. It should be pointed out that the "±" in this case is not an uncertainty but rather an indication of the actual variation of the flow. Clearly the SGD in this case, as in many other cases, is not steady-state but highly variable with a phase that suggests a relationship to tidal forcing.

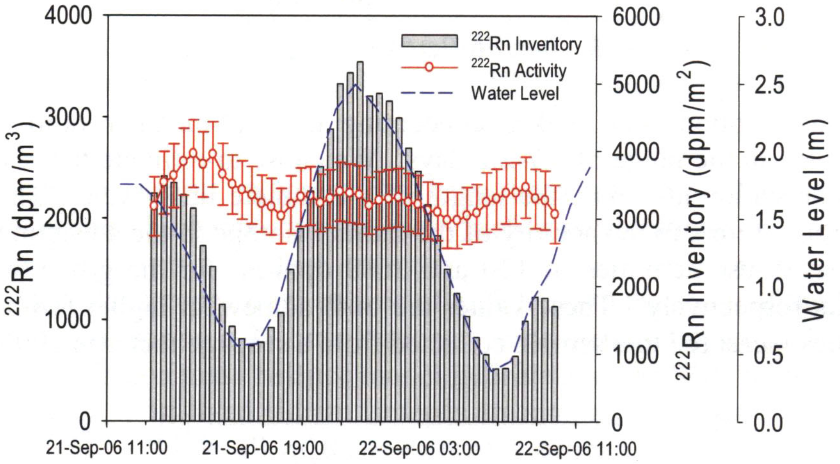

Figure 2 Concentrations (open circles), inventories (bars) of ^{222}Rn, and water level (dashed line) at station A-1 over a 24-hour period.

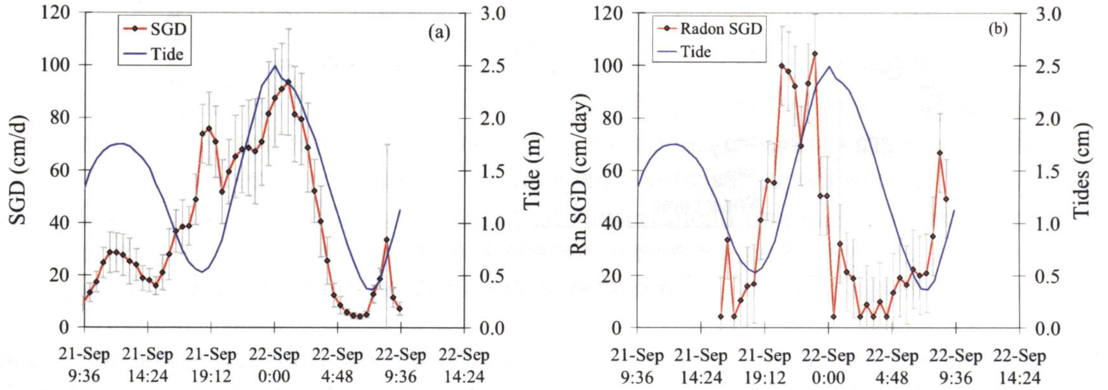

Figure 3 SGD variations as measured by an automatic seepage meter (a) and as calculated by the mass balance radon model (b). Both plots are shown on the same scale to make comparisons easier.

Conclusions: Our calculations suggest average rates of total groundwater (saline + fresh water) of 40 and 37 cm/day based on ^{223}Ra and ^{224}Ra, respectively. We also calculate an average rate of 37 cm/day based on continuous ^{222}Rn measurements. These estimates compare well to the 44 cm/day average rate from an automatic seepage meter deployed nearby. While we have made some assumptions concerning the residence time and end-member values of these isotopes in the groundwater, the results are internally consistent within all 3 tracers as well as the automated seepage meter.

It is important to note that the "groundwater" flow in this case appears to be dominated by recirculated seawater. Thus, calculation of nutrient fluxes requires analysis of the offshore marine groundwater (porewater rather than the terrestrial fresh groundwater).

Acknowledgments: The authors are grateful to all the members of the "Delta" and "Bohai Sea" research groups from the "Yellow River Project" who shared data and provided assistance in the field. We thank Prof. Y. Fukushima of the Research Institute for Humanity and Nature (RIHN) for allowing us to become involved in this research. Additional support was provided by the National Science Foundation (OCE0350514).

References:

Burnett, W.C., G. Kim, and D. Lane-Smith, 2001. A continuous radon monitor for assessment of radon in coastal ocean waters. Jour. Radioanal. Nucl. Chem., 249, 167-172.

Burnett, W.C. and H. Dulaiova, 2003. Estimating the dynamics of groundwater input into the coastal zone via continuous radon-222 measurements. Jour. Environmental Radioactivity, 69, 21-35.

Moore, W. S., J.O. Blanton, and S. Joye. 2006. Estimates of flushing times, submarine groundwater discharge, and nutrient fluxes to Okatee River, South Carolina. Journal of Geophysical Research-Oceans 111, doi:10.1029/2005JC003041.

Moore, W. and R. Arnold, 1996. Measurement of ^{223}Ra and ^{224}Ra in coastal waters using a delayed coincidence counter. Jour. Geophys. Res., 101, 1321-1329.

Slomp, C.P. and P. Van Cappellen, 2004. Nutrient inputs to the coastal ocean through submarine groundwater discharge: controls and potential impact. Journal of Hydrology, 295(1-4): 64-86.

Taniguchi, M. and H. Iwakawa, 2001. Measurements of submarine groundwater discharge rates by a continuous heat-type automated seepage meter in Osaka Bay, Japan. Jour. Groundwater Hydrology, 43, 271-277.

Zhang, J., Z.G. Yu, T. Raabe, S.M. Liu, A. Starke, L. Zou, H.W. Gao, and U. Brockmann, 2004. Dynamics of inorganic nutrient species in the Bohai seawaters. Journal of Marine Systems, 44(3-4), 189-212.