

## Distribution patterns of salinity and $^{222}\text{Rn}$ in Yatsushiro Inland Sea, Kyushu, Japan

Y. NIKPEYMAN<sup>1</sup>, M. ONO<sup>2</sup>, T. HOSONO<sup>1</sup>, H. YANG<sup>1</sup>, K. ICHIYANAGI<sup>1</sup>,  
J. SHIMADA<sup>1</sup> & K. TAKIKAWA<sup>3</sup>

<sup>1</sup> Kumamoto University, GSST, Japan  
[yaser.nikpeyman@yahoo.com](mailto:yaser.nikpeyman@yahoo.com)

<sup>2</sup> The National Institute of Advanced Industrial Science and Technology (AIST), Japan

<sup>3</sup> Centre for Marine Environment Studies, Kumamoto University, Japan

**Abstract** Submarine Groundwater Discharge (SGD), as a way through which solutes and nutrients travel from terrestrial areas towards coastal areas, is part of the hydrological cycle. Various methods are used to locate SGD at different scales. Among them,  $^{222}\text{Rn}$  has been developed with the viewpoint of accurate local estimations of SGD points indirectly. This research aims to identify SGD areas in the Yatsushiro Sea, southwest Japan, using the  $^{222}\text{Rn}$  method, while considering rivers with high  $^{222}\text{Rn}$  concentration in the study area. The area is an inland sea with high tidal fluctuations and there is a large contribution between the sea and groundwater, which are greatly affected by rivers. A multi-detector  $^{222}\text{Rn}$  survey has been carried out simultaneously with sea water electrical conductivity (EC) and temperature. In addition, several river grab samples were analysed for  $^{222}\text{Rn}$  concentration. Considering the sea water radon distribution and river characteristics, several points were selected for future SGD volume estimations.

**Key words** Submarine Groundwater Discharge (SGD); Yatsushiro Sea, Japan;  $^{222}\text{Rn}$ , multiple – detector

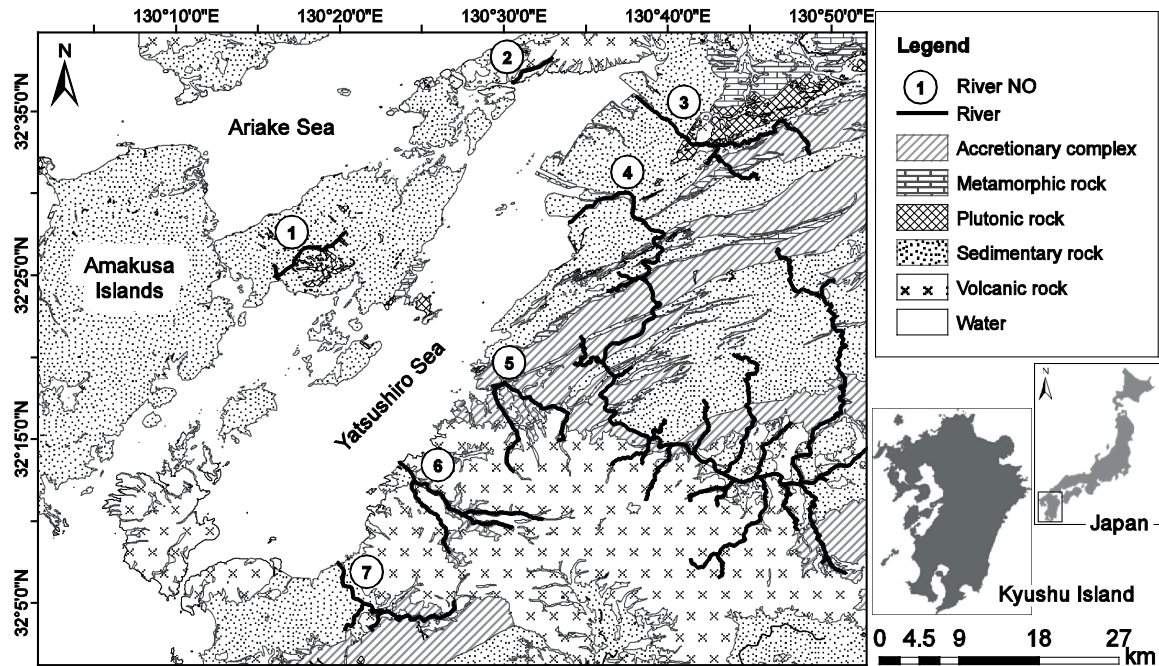
## INTRODUCTION

The submarine groundwater discharge (SGD) is a process through which nutrients, materials and pollutants travel from land to ocean (Burnett *et al.* 2003). Identification of SGD locations and the estimation of the groundwater flow magnitudes are skimpy due to the complexity of measurements and invisible land–sea pathway issues (Moore 1996, 1999, Zektser 1996). The various methods are used to locate SGD are not trivial and salinity is the most obvious SGD tracer in coastal areas. This is while the sea water salinity highly affected by freshwater river discharge in estuarine areas. During the last decade, natural radioactive elements such as  $^{222}\text{Rn}$  and radium isotopes ( $^{223}\text{Ra}$ ,  $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$ ) have been applied to trace SGD toward estuarine areas and coastal zones (e.g. Cable *et al.* 1996, Moore 1996, Krest *et al.* 2000, Charette *et al.* 2003, Dulaiova *et al.* 2005). Among the mentioned isotopes,  $^{222}\text{Rn}$  assessment needs fewer corrections since it is an inert gas and there is no need to consider biogeochemical reactions (Dulaiova *et al.* 2005). The conventional sampling and analysis methods for  $^{222}\text{Rn}$  assessment to locate SGD on a regional scale were developed by Dulaiova *et al.* (2005). They applied a multi-detector system to monitor the  $^{222}\text{Rn}$  continuously off the Florida State University Marine Laboratory (FSUML) along the northeast coastline of the Gulf of Mexico by connecting a set of three RAD7 (DurrIDGE Co.) radon detectors and improved the measurement resolution to about 1 record every 10 minutes. The spatial resolution of monitored radon depends on the equipment boat speed in this method.

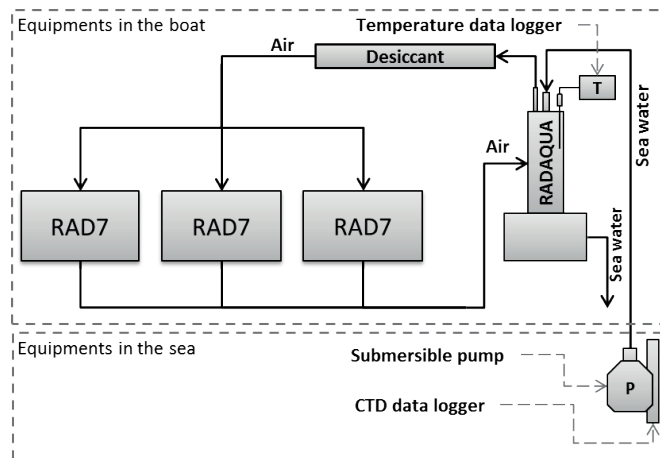
There are many rivers in the world with high  $^{222}\text{Rn}$  concentration discharging to coastal areas (Al-Masri and Blackburn 1999, Cook *et al.* 2003, Giap 2003, Marques *et al.* 2004, Mukherjee *et al.* 2007, Pourhabib *et al.* 2011). In order to exclude the effect of high radon concentration rivers on the Dulaiova *et al.* (2005) approach for measuring SGD, EC time series were merged with radon concentrations monitored to locate SGD signals along the coast line of the Yatsushiro Sea, Kyushu Island, southwest Japan.

## STUDY AREA

The research was conducted along the coast of the Yatsushiro Sea, southwest Japan. The Yatsushiro Sea perimeter is approximately 220 km long with freshwater inflow from 58 rivers. Among the rivers, seven major surface streams were selected as having runoff representative for the study area (Fig. 1).



**Fig. 1** The Yatsushiro Sea study area, Kyushu Island, south west Japan. Rivers NO are: ① Kawachi, ② Koriurakawa, ③ Hikawa, ④ Kuma, ⑤ Yunoura, ⑥ Minamata, ⑦ Komenotsu.



**Fig. 2** Sketch of the multi-detector system to monitor  $^{222}\text{Rn}$  continuously in sea water. Modified from Dulaiova et al. (2005).

These streams are well-distributed over the study area and are the biggest discharging rivers with the highest  $^{222}\text{Rn}$  concentration. The Yatsushiro Sea is linked to the Kyushu mainland from the east coastline while the western coasts are related to the Amakusa Islands, which consist of several small islands. Although the aquifers consist of Quaternary fractured volcanic rocks (andesite lava and tuff breccia) and pyroclastic beddings, marine sediments form dominant aquifer layers in the study area. The geologic settings of the Amakusa Islands are mostly uniform, containing sedimentary rocks with a high diversity of fracture direction influencing groundwater flow direction. While the geologic features in the eastern part are very complicated, consisting of an accretionary complex, metamorphic, plutonic, sedimentary and volcanic rock types, in which the fracture directions and bedding planes are mostly toward the Yatsushiro Sea resulting in groundwater flow uniformity. The land use in the basins includes citrus farming, paddy fields and bamboo–cedar forests. The Yatsushiro Sea is the largest inland sea in Japan. The land to ocean dynamics are usually considered from a nutrients discharge perspective.

The average annual precipitation and temperature in the study area are about 2008 mm and 16.8°C respectively. Maximum average precipitation occurs during June and July, while the temperature reaches its highest level during July and August. Note, the precipitation and catchment size in the Amakusa Islands is smaller compared to other parts of the study area, which results in smaller surface streams.

## METHODS

The technique applied in this research was developed by Dulaiova *et al.* (2005). In this method, a boat was equipped with a multi-detector continuous monitoring system (Fig. 2). The system uses three RAD7 detectors (DURRIDGE Co.) to monitor  $^{222}\text{Rn}$  concentration in the air, which circulates in a closed air loop through RAD7s and RADAQUA. The RADAQUA is fed with a constant stream of sea water by a submersible pump (Rule 800 GPH Submersible Bilge Pump, USA) and the  $^{222}\text{Rn}$  reaches an equilibrium condition between sea water and the air in the closed air loop within the RADAQUA. The RAD7 monitors  $^{222}\text{Rn}$  in the air by measurement of  $\alpha$ -emitting isotopes ( $^{214}\text{Po}$  and  $^{218}\text{Po}$ ). As the sea water temperature was monitored by a T-logger (HOBO temp External Sensor Logger H08-002-02, USA), the  $^{222}\text{Rn}$  concentration in the sea water was calculated using equation (1):

$$a' = 0.105 + 0.405e^{-0.0502T} \quad (1)$$

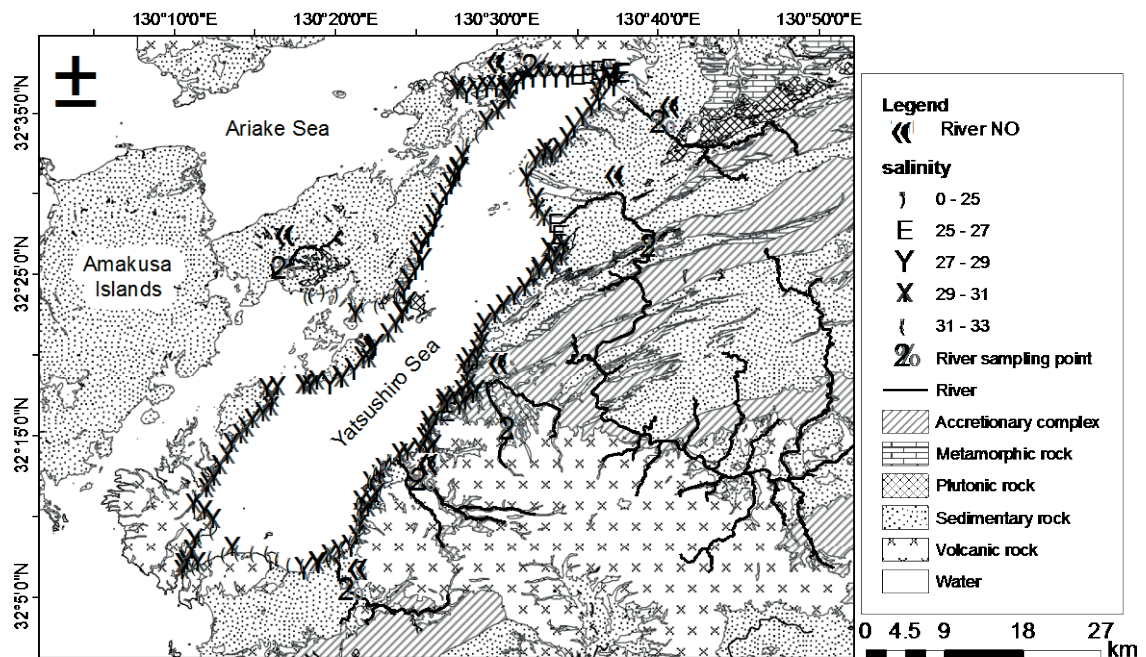
where  $a'$  illustrates the concentration ratio of water to air and  $T$  is the sea water temperature in °C. The sea water EC was recorded during studies using a CTD data logger (CTD-Diver, DI219, Schlumberger Water Services, USA) which was connected to a submersible pump. Considering the sea water temperature and EC, the salinity was calculated. The sea water studies were conducted during 14–16 September 2010, 24–28 October, 2011 and 3–4 August 2012. The detectors cycle was set to measure the  $^{222}\text{Rn}$  concentration in air every 10 minutes. Also, the equipment boat was driven as slowly as 7–8 km/h to increase the data resolution to a level of 1 record per 1.5 km.

To eliminate the rivers' noise in measurements, seven representative rivers discharging into the Yatsushiro Sea were analysed from the viewpoint of  $^{222}\text{Rn}$  concentration. The rivers are well-distributed along the coastline and are the biggest discharging rivers within the study area. Grab samples were taken during 22–28 November 2012 and analysed for  $^{222}\text{Rn}$  concentration immediately after the samples were taken. Furthermore, flow rate, temperature and EC for each river were measured simultaneously, and the river water salinity calculated using EC and temperature, as well as the method applied for the sea water (Table 1).

## RESULTS

The calculated sea water salinity fluctuated between almost 0 and 33 ppt (Fig. 3). There are several big rivers on the eastern coastline (e.g. Kuma and Komenotsu rivers). Also, the sea water salinity fluctuates significantly at the lowest level (0–25 ppt) at the estuary area of Kawachi and Minamata rivers to maximum values (31–33 ppt) around the estuary area of Komenotsu River. The sea water salinity fluctuations along the eastern coastline reflect the magnitude of the rivers' effect on sea water salinity values in these areas. Further, the sea water salinity values are almost moderate (27–31 ppt) along the western coastline.

The sea water  $^{222}\text{Rn}$  concentration in the study area varies from 0 to 180 Bq/m<sup>3</sup> (Fig. 4). Along the western coastline, the results were low (zero), while maximum values were recorded along the eastern areas, especially around the Komenotsu River estuary area which is almost 180 Bq/m<sup>3</sup>. The distribution pattern of radon concentration along the eastern coastline represents background values of about 10–40 Bq/m<sup>3</sup>, with considerable high signals near the river estuary areas. The most significant increase of sea water radon concentration was around the Komenotsu River estuary area, while the Minamata River estuary area represents a very small effect on sea water radon concentration with a diluting effect.



**Fig. 3** The distribution map of sea water salinity in the study area.

**Table 1** Location and results of measurements of the studied streams.

NO	Name	Coordinates Longitude	Latitude	Q (m <sup>3</sup> /day)	Salinity (ppt)	Rn (Bq/m <sup>3</sup> )
1	Kawachi	E 130° 16' 38.892"	N 32° 25' 18.430"	13 038	0.050	340
2	Koriurakawa	E 130° 32' 15.900"	N 32° 37' 46.639"	1 137	0.102	370
3	Hikawa	E 130° 40' 08.976"	N 32° 34' 19.880"	61 257	0.071	1610
4	Kuma	E 130° 39' 35.460"	N 32° 26' 40.459"	—	0.057	680
5	Yunoura	E 130° 30' 50.148"	N 32° 15' 19.670"	41 262	0.065	1030
6	Minamata	E 130° 25' 19.884"	N 32° 12' 09.230"	34 478	0.066	770
7	Komenotsu	E 130° 20' 52.296"	N 32° 05' 19.540"	554 299	0.055	580

## DISCUSSION

As can be seen, the sea water radon concentration along the western coast line (Amakusa Islands coastal area) is almost zero (Fig. 4). The only part of the Amakusa Islands which discharges a high volume of low salinity and moderate <sup>222</sup>Rn concentration freshwater is the Kawachi River, (Table 1). Furthermore, the decreasing trend of sea water salinity and increasing trend of <sup>222</sup>Rn concentration toward the Kawachi River estuarine area (Fig 3), represents the dominance of the effect of the Kawachi River on sea water characteristics, with no SGD.

There are high <sup>222</sup>Rn concentration signals in the southeastern part of the study area. Also, the background value of radon in sea water of the southeast parts of the Yatsushiro Sea is 10–120 Bq/m<sup>3</sup>. Moreover, an accretionary trend toward the Komenotsu River estuary is apparent in which the radon concentration is the highest sea water radon concentration value recorded during measurements (180 Bq/m<sup>3</sup>). Considering the low salinity (0–31 ppt) and high discharge rate of the Komenotsu River (Table 1), very low salinity was expected in the estuarine area, while the monitored sea water salinity represents a small moderate-salinity domain around the estuary area of the Komenotsu River (Fig. 3). Consequently, the sea water high radon concentration in the southeastern part of the Yatsushiro Sea is due to strong SGD.



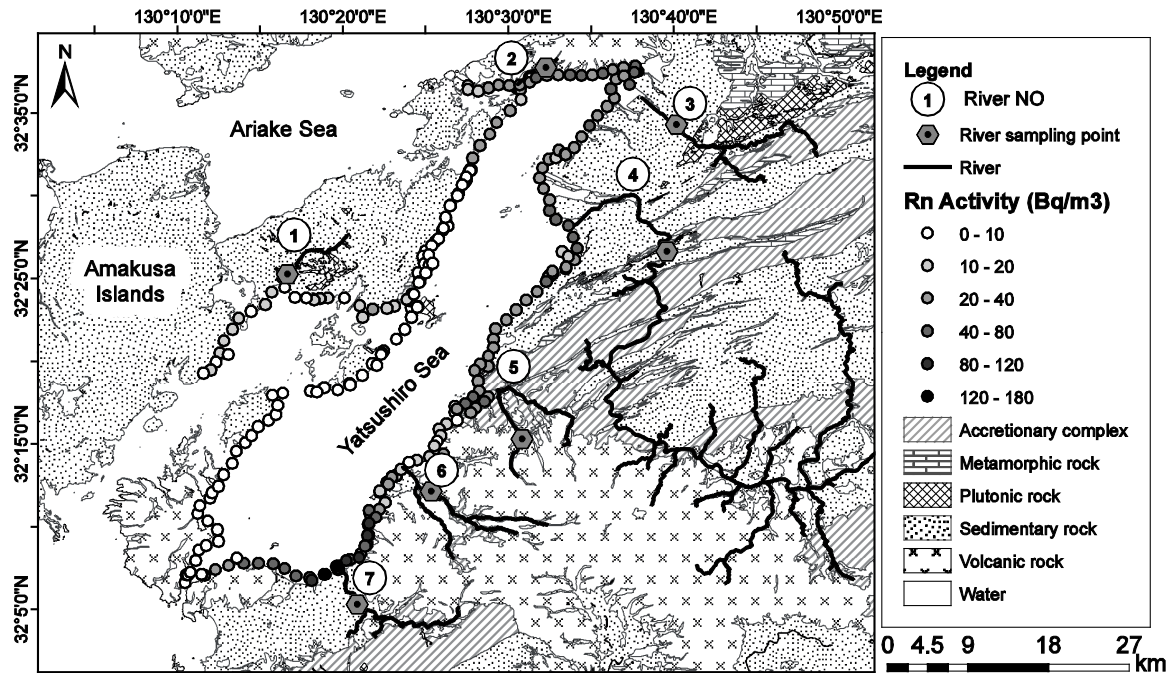


Fig. 4 The distribution map of  $^{222}\text{Rn}$  concentration in sea water in the study area.

Moderate to high sea water salinity range (29–31 ppt) around the estuarine areas of the Minamata and Yunoura rivers represents the non-significant influx of rivers at the eastern coastline of the Yatsushiro Sea (Fig. 3). Moreover, although the  $^{222}\text{Rn}$  concentration of these rivers' grab samples are relatively high (Table 1), the radon noise due to river discharge has very little effect on locating SGD signals. This means that the monitored sea water  $^{222}\text{Rn}$  concentration along the eastern coast line is mostly due to discharge of groundwater.

The Kuma River discharging at the eastern coast of the Yatsushiro Sea is one of the enormous rivers within the study area. This river discharges a huge volume of water with low salinity and moderate  $^{222}\text{Rn}$  concentration (Table 1). The low salinity sea water domain (25–27 ppt) at the estuary area of the Kuma River (Fig. 3) represents the noteworthy radon noise due to river discharge around the northeastern part of the Yatsushiro Sea (Fig. 4). Further, the sea water radon concentration increases to approximately 50 Bq/m<sup>3</sup> around the estuary area of the river from a background value of about 30 Bq/m<sup>3</sup>. This means that although the SGD proposition is valid for the northeastern part of the Yatsushiro Sea, supplementary data are necessary to make the final decision.

Although, the sea water salinity distribution along the northern coast line is highly affected by several discharges by small rivers, our evidence confirms a considerable volume of SGD. There are not only coastal springs, but also data obtained from a monitoring well in the northern part of the study area indicate an upward groundwater pressure head, little seasonal trend and daily fluctuations with reverse correlation to the tidal movements (Shimada *et al.* 2007). So, supplementary data are proof of the SGD along the northern coastal area of the Yatsushiro Sea, while the radon noise due to rivers discharge makes it complicated to discuss and conclude about the probability of SGD within the area.

To sum up, the sea water salinity distribution (Fig. 3) and  $^{222}\text{Rn}$  concentrations along the coastal area (Fig. 4) represent probable SGD areas around the Komenotsu and Minamata rivers. In addition, evidence confirms the submarine groundwater discharge in the northern part of the study area. Also, the radon studies are ambiguous in northern and northeastern areas of the Yatsushiro Sea. This is due to low to moderate sea water salinity (25–29 ppt) and intermediate  $^{222}\text{Rn}$  concentration in sea water in, for example, the Kuma River estuary area. However, it is apparent that the sea water radon survey was likely to not be an accurate method of SGD estimation in the Yatsushiro Bay, while it is a very useful tool with supplementary datasets such as river

characteristics, monitoring borehole records and residents' observations and reports. Also, given the structural geologic characteristics of the study area, it is more obvious that the probability of SGD along the Amakusa Islands coastline is very low due to the non-oriented fractures which disperse groundwater flow lines. Furthermore, the lineaments uniformity in the eastern coastal aquifers conducts the groundwater flow lines toward the coastline and discharge into the Yatsushiro Sea. Besides, to confirm the geology relic it is necessary to consider various aspects such as rock–groundwater interactions more accurately.

To conclude, the Yatsushiro inland sea interacts highly with the eastern coastline groundwater. As is apparent, the link between Amakusa Islands and the Yatsushiro Sea is not as great as in the eastern coastal areas due to small catchment size and precipitation. In addition, the disseminated fractures without unique seaward direction minimize the groundwater–seawater interaction on the western coastline. Above all, it is very important to consider more detailed data in the areas with low sea water salinity and intermediate  $^{222}\text{Rn}$  concentration in sea water.

**Acknowledgements** The authors would like to extend sincere appreciation and gratitude to the members of the fishing community in Kumamoto and Kagoshima Prefectures for their cooperation during field measurements. Also, we are pleased to express a special thank you to Mr Sakai for accompanying us during the collection of data for sea water  $^{222}\text{Rn}$  survey. In addition, thanks go to the Yatsushiro Sea project authorities for their financial support.

## REFERENCES

- Al-Masri M. S. and Blackburn R. (1999) Radon-222 and related activities in surface waters of the English Lake District. *Applied Radiation and Isotopes* 50, 1137–1143.
- Burnett, W. C., et al. (2003) Groundwater and pore water inputs to the coastal zone. *Biogeochemistry* 66, 3–33.
- Cable, J., et al. (1996) Estimating groundwater discharge into the north eastern Gulf of Mexico using radon-222. *Earth and Planetary Science Letters* 144, 591–604.
- Charette, M., et al. (2003) Salt marsh submarine groundwater discharge as traced by radium isotopes. *Marine Chemistry* 84, 113–121.
- Cook P. G., et al. (2003) Determining natural groundwater influx to a tropical river using radon, chlorofluorocarbons and ionic environmental tracers. *Journal of Hydrology* 277, 74–88.
- Dulaiova, H., et al. (2005) A multi-detector continuous monitor for assessment of  $^{222}\text{Rn}$  in the coastal ocean. *Journal of Radioanalytical and Nuclear Chemistry* 263(2), 361–365.
- Giap T. V. (2003) Use of radon-222 as tracer to estimate groundwater infiltration velocity in a river bank area. *Nuclear Science and Technology* 2 (2), 12–17.
- Krest, J., et al. (2000) Marsh nutrient export supplied by groundwater discharge: evidence from Ra measurements. *Global Biogeochemical Cycles* 14, 167–176.
- Marques, A. L., Santos, W. and Geraldo, L. P. (2004) Direct measurements of radon activity in water from various natural sources using nuclear track detectors. *Applied Radiation and Isotopes* 60, 801–804.
- Moore, W. (1976) Sampling  $^{228}\text{Ra}$  in the deep ocean. *Deep Sea Research and Oceanographic Abstracts* 23, 647–651.
- Moore, W. (1996) Local groundwater inputs to coastal waters revealed by  $^{226}\text{Ra}$  enrichments. *Nature* 380, 612–614.
- Moore, W. (1999) The subterranean estuary: a reaction zone of groundwater and sea water. *Marine Chemistry* 65, 111–125.
- Mukherjee, P. K., et al. (2007) A stream sediment geochemical survey of the Ganga River headwaters in the Garhwal Himalaya. *Geochemical Journal* 41, 83–95.
- Pourhabib, Z., Binesh, A. and Mohammadi, S. (2011) Determination of radon and radium in springs, wells, rivers and drinking water samples of Ramsar in Iran. *International Archive of Applied Sciences and Technology* 2(1), 32–36.
- Schwartz, M. (2003) Significant groundwater input to a coastal plain estuary: assessment from excess radon. *Estuarine, Coastal and Shelf Science* 56, 31–42.
- Shimada, J., et al. (2007) Basin-wide groundwater flow study in a volcanic low permeability bedrock aquifer with coastal submarine groundwater discharge. In: *A New Focus on Groundwater–Seawater Interactions* (ed. by W. Sanford et al.). IAHS Publ. 312, 75–85. IAHS Press, Wallingford, UK.
- Zektser I. S. (1996) Groundwater discharge into the seas and ocean: state of the art. In: *Groundwater Discharge in the Coastal Zone* (ed. by R. W. Buddemeier). *Russian Academy of Sciences* 122–123.