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Inferring coastal processes from regional-scale mapping of ^{222}Radon and salinity: examples from the Great Barrier Reef, Australia

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ABSTRACT

The radon isotope ^{222}Rn and salinity in coastal surface water were mapped on regional scales, to improve the understanding of coastal processes and their spatial variability. Radon was measured with a surface-towed, continuously recording multi-detector setup on a moving vessel. Numerous processes and locations of land–ocean interaction along the Central Great Barrier Reef coastline were identified and interpreted based on the data collected. These included riverine fluxes, terrestrially-derived fresh submarine groundwater discharge (SGD) and the tidal pumping of seawater through mangrove forests. Based on variations in the relationship of the tracers radon and salinity, some aspects of regional freshwater inputs to the coastal zone and to estuaries could be assessed. Concurrent mapping of radon and salinity allowed an efficient qualitative assessment of land–ocean interaction on various spatial and temporal scales, indicating that such surveys on coastal scales can be a useful tool to obtain an overview of SGD locations and processes.

1. Introduction

A multitude of methods are currently employed to study fluxes of water from the land to the ocean from rivers and submarine groundwater discharge (SGD), ranging from river gauging and point flux measurements of SGD with seepage meters, to methods integrating over various spatial and temporal scales such as hydrological models or surface water tracer approaches. Such fluxes are rarely uniformly distributed in space and time. Here we demonstrate the application of regional-scale concurrent mapping of radon and salinity in coastal surface water to improving the understanding of coastal processes and their spatial variability.

On kilometer to hundred kilometer scales, the spatial distribution and regional importance of water input from the land to the ocean can be investigated by mapping relevant tracers in the coastal ocean. One frequently used tracer for this purpose is the radon isotope ^{222}Rn, hereafter referred to as radon (e.g. Hussain et al., 1999). Radon is a powerful indicator of groundwater input to surface water bodies because (a) it is elevated in groundwater relative to surface waters by typically 2–3 orders of magnitude, (b) it has a conservative nature, (c) it can be measured at low concentrations and (d) its half life of 3.82 days is comparable to the time scale of coastal circulation (e.g. Cable et al., 1996). However, groundwater discharge is not the only source of radon to coastal waters. Radon can occur in coastal water through three main mechanisms: (1) river discharge, (2) groundwater discharge, and (3) recirculation of seawater through seabed sediments. A special case of seawater recirculation along tropical coastlines is the tidal pumping of seawater through mangrove forests (Ridd, 1996; Stieglitz, 2005).

Absolute radon activities in seawater will depend upon the rates of seawater pumping, river discharge and groundwater discharge, the production rate within the seafloor sediments, the radon activity in river discharge and groundwater discharge, but also on the location of sampling relative to the ocean and to river mouths, the water depth and the rate of offshore mixing. When

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groundwater containing radon discharges to surface water, radon activities decrease due to gaseous exchange with the atmosphere and radioactive decay. In the case of discharge to the ocean, a decrease in radon activity will also be observed due to mixing with open ocean water, which is typically low in radon.

Dulaiova et al. (2005) demonstrated the principle of continuous radon sampling (Fig. 1a). A preliminary study in the Great Barrier Reef indicated that continuously measured radon along a coastline can be used as an indicator for the presence of SGD (Stieglitz, 2005). Burnett et al. (2008) mapped radon in a coastal embayment along the Ubatuba coastline in Brazil, and Crusius et al. (2005) estimated SGD discharge rates based on concurrently measured radon and salinity. Here a multi-detector field setup is evaluated for the spatial mapping of radon on a still larger spatial scale, and it is discussed how continuous mapping of this tracer, in combination with ocean water salinity, can be used to efficiently infer and interpret locations of radon input to the ocean.

By using a second tracer such as salinity, it is possible to determine the relative importance of some of these different processes, and the relative rate of gas exchange and offshore mixing. Fig. 1b shows a mixing line that would be expected between terrestrial water (which is usually relatively high in radon and low in salinity) and offshore seawater (which is low in radon and high in salinity). If the decrease in radon activity (relative to terrestrial water) is primarily due to mixing between end members, then radon and salinity will be affected equally, and so coastal water should fall on a straight line between the terrestrial and offshore end members. However, if the decrease in radon activity is dominated by either gas loss to the atmosphere and/or radioactive decay, then the decrease in radon activity will be more rapid than the increase in salinity, and coastal water data points will fall below the mixing line. If direct discharge of terrestrial groundwater and river discharge have markedly different radon activities and salinities, then it may also be possible to differentiate between these two terrestrial water sources.

2. Methods

2.1. Continuous radon and salinity sampling

Radon-222 was continuously sampled and counted *in situ* using three electronic radon-in-air monitors Durridge RAD 7 in parallel (Fig. 1a), similar to a setup used by Dulaiova et al. (2005). Seawater was pumped directly through an air–water equilibrator. A non-cavitating positive displacement pump was used in order to avoid potential radon loss to air bubbles in the pipes. The ^{222}Rn-enriched air was circulated in a closed air loop, which passes through the monitors, to establish equilibrium between the circulating air and continuously pumped seawater. The monitors count α-decays of ^{222}Rn daughters, and discriminate different decays in energy-specific windows. Counting interval was 10 min. One sigma uncertainties are reported. The waste water from the exchanger chamber was circulated through a Hydrolab Minisonde water quality sensor, which recorded salinity in 2.5 min intervals. Prior to measurements, the sensor was calibrated to factory standards.

The radon-in-air monitors use a silicon semiconductor detector at ground potential, which attracts positively charged polonium isotopes. The daughter isotope $^{218}Po^{+}$ is counted as a measure of ^{222}Rn in air. ^{218}Po has a half life of 3.1 min, and the polonium activity is in radioactive equilibrium with radon after approx 15 min i.e. five half lives. High resolution energy windows in the detector allow discrimination of all alpha-emitting polonium isotopes, and thus the detectors have a low background. However, the comparatively small detector size results in a somewhat low efficiency of less than 20%. Therefore, three counters were used in parallel, in order to increase the overall efficiency of the counting procedure. The activity and one sigma uncertainty measured with such a multi-detector setup is calculated as:

$$A = \sum c_i \times K_i \pm \mathrm{sqrt}\left(\sum c_i\right)$$

where c_i is counts per detector and K_i the instrument-specific calibration constant. From the radon activity measured in the air loop, the in-water activity is calculated from the temperature-dependant air/water equilibrium Fritz–Weigel equation (Weigel, 1978):

$$^{222}\mathrm{Rn}_{\mathrm{water}} = {}^{222}\mathrm{Rn}_{\mathrm{air}} \times \left(0.105 + 0.405e^{-0.0502T}\right)$$

where $^{222}\mathrm{Rn}_{\mathrm{water}}$ and $^{222}\mathrm{Rn}_{\mathrm{air}}$ are the radon activity in water and air respectively and T is the temperature in °C within the mixing chamber.

2.2. Response of the multi-detector setup to changes in radon activity

The response to variations in ^{222}Rn activity depends on a variety of parameters, chiefly (a) the halflife of ^{218}Po, (b) the detection efficiency, (c) the volume of the degassing chamber and the air loop (total ~4 L), (d) flow rate of air through the air loop (~1 L min^{-1}) and (e) efficiency of transfer of ^{222}Rn from water to air (Dulaiova et al., 2005). Parameters (a) to (d) are system parameters and cannot easily be changed. Parameter (e) is largely determined by the flow rate of water to the mixing chamber. Dulaiova et al. (2005) use a flow rate of ~5 L min^{-1} in the field, and comment that no significant difference in response was observed when flow was reduced to 2 L min^{-1}. Throughout the work presented herein, a water flow rate of >6 L min^{-1} was maintained, in order to ensure that supply of radon from the water to the air loop was not a limiting factor.

To assess the response to variations in radon activity, the multi-detector setup was exposed to alternating high and low activity water in the laboratory. Water was pumped from reservoirs with activities of 50 Bq m^{-3} and 5 Bq m^{-3} for high and low activity respectively, representing typical values encountered in coastal water in the study region, the Great Barrier Reef (GBR). Water reservoirs were switched on different time scales, whereby water from the high-activity reservoir was pumped for 2 h, 20 min, 5 min and 1 min respectively.

In order to verify consistency of this continuous sampling technique method with spot sampling techniques, concurrent water samples were collected for liquid scintillation counting from representative field locations, whilst steaming (Cook et al., 2006). Approximately 1200 mL of water was collected, and shaken with 20 mL of Packard NEN mineral oil scintillant and 30 mL headspace in a 1250 mL plastic bottle, to equilibrate the radon between the water-air scintillant phases. After allowing the scintillant to settle to the top of the bottle (about 1 min), the scintillant

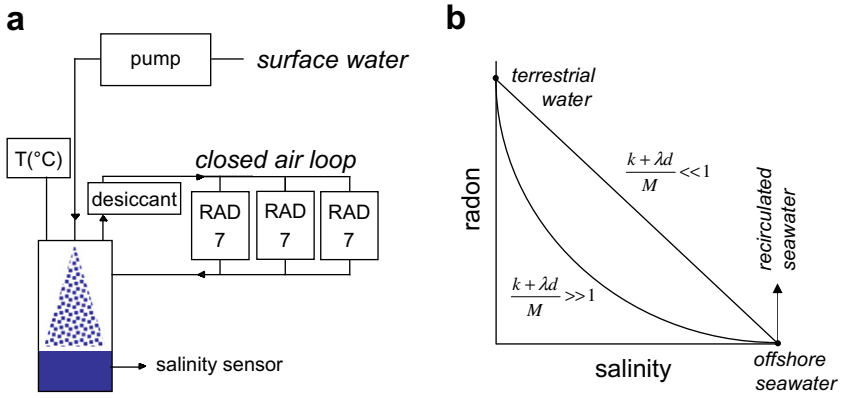

Fig. 1. (a) Multi-detector setup to continuously sample radon from surface water, similar to a setup described in Dulaiova et al. (2005). (b) Mixing plot of radon and salinity. Typical mixing lines for terrestrial water and ocean as well as recirculated seawater are shown. The straight and curved mixing lines represent dominance of mixing M over gas loss k and decay λd and vice versa respectively.