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Groundwater inflow to a shallow, poorly-mixed wetland estimated from a mass balance of radon

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Summary Radon activity within a shallow wetland in southern Australia has been measured on three occasions between May and October 2006. Measured activities within the surface water display a similar pattern of spatial variability on each occasion, suggesting that it is related to the locations of groundwater inflow and mixing processes. The mean groundwater inflow rate has been estimated from the mean radon activity using a mass balance approach. The components of the radon budget are (i) contribution from groundwater inflow, (ii) diffusive flux from wetland bottom sediments (iii) loss due to gas exchange, (iv) loss due to radioactive decay, (v) loss due to groundwater or surface water outflow. Also required to complete the water balance are the surface water inflow rate, direct precipitation on the wetland, and evaporation rate. The radon diffusive flux has been estimated from measurements of radon production within the sediments and a diffusive transport model, calibrated by measurements of radon activity in sealed chambers that can receive radon only from diffusion and lose it only by radioactive decay. Radon loss due to gas exchange is inferred from the loss rate of SF_6, following its injection into isolated areas of the wetland, while the rate of radioactive decay is known. The radon activity in groundwater inflow is measured from sampling piezometers surrounding the wetland. Steady state and transient mass balance approaches yield similar results, with groundwater inflow rates varying between 12 and 18 m^3/day. Estimated groundwater inflow rates are most sensitive to the radon activity of groundwater inflow, the gas exchange velocity, surface water area and the accuracy with which the mean radon activity in the wetland can be measured. Importantly, it is relatively insensitive to the surface water inflow rate, which is poorly known.

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Nomenclature

A	lake area (m^2)	I_g	groundwater inflow (m^3/day)
A_b	area of lake bottom (m^2)	I_s	surface water inflow (m^3/day)
b	Mean lake depth (m)	k	gas exchange velocity (m/day)
c_g	groundwater inflow activity (Bq/L)	P	precipitation falling on lake (m/day)
c_L	mean activity in lake (Bq/L)	Q	surface and ground water outflow (m^3/day)
c_s	surface water inflow activity (Bq/L)	t_r	mean residence time of radon in lake (days)
c_w	mean activity below water surface (Bq/L)	V	lake volume(m^3)
c_Q	mean activity of lake outflow (Bq/L)	ε	sediment porosity (−)
$c(z)$	radon activity in sediment pore water (Bq/L)	γ	radon production rate (Bq/cm^3/day)
D	diffusion coefficient in sediments (m^2/day)	λ	radioactive decay coefficient (day^{-1})
E	evaporation from lake (m/day)	ρ_b	sediment bulk density (g/cm^3)
E_m	emanation rate from sediments (Bq/kg)	ρ_s	sediment particle density (g/cm^3)
F	diffusive flux from lake sediments (Bq/m^2/day)		

Introduction

Groundwater discharge to surface waters is one of the more difficult components of the water balance to measure. Darcian calculations are often limited by our ability to accurately determine the hydraulic conductivity of the sediments (Cey et al., 1998). In the case of lake and wetland systems, they may also be limited by very low hydraulic gradients, which can be difficult to accurately measure. Water balance methods that measure other components of the water balance and calculate groundwater inflow by difference are limited by the errors in the other water balance components, which may be larger than the groundwater inflow term. Seepage meters are greatly affected by spatial variations in hydraulic conductivity, and the difficulty of accurately measuring lateral inflow (as opposed to vertical seepage through the lake or river bed; Langhoff et al., 2006). Environmental tracer methods have therefore proven to be a popular method, and can be used in conjunction with water balances if concentrations of the various sources are well known (e.g., Krabbenhoft and Webster, 1995).

Chloride and the stable isotopes ^2H and ^{18}O are amongst the more widely used environmental tracers in lake water balances studies (e.g., Hunt et al., 1996; Gurrieri and Furniss, 2004). Although they can greatly assist in constraining a water balance, they have two main deficiencies. Firstly, because both of these tracers can enter the lake through precipitation and surface runoff as well as through groundwater inflow, discriminating between these different fluxes is not always straightforward. Secondly, because the residence time of these tracers in some lakes may be long, their concentrations may reflect changes in the lake water balance over an extended period of time. For dynamic systems, measured concentrations may be unrelated to the current water balance. This is particularly the case for chloride, which may accumulate over many years.

Radon-222 is an environmental tracer that has been used for quantifying groundwater inflows to streams and estuaries (e.g., Genereux et al., 1993; Cook et al., 2003) and the ocean (e.g., Cable et al., 1996) for almost two decades. With a half-life of 3.8 days, radon is produced in the subsurface by the radioactive decay of ^{226}Ra, which is part of the ^{238}U decay chain. After groundwater containing radon discharges to surface water bodies, radon activities decrease due to gaseous exchange with the atmosphere and radioactive decay. The radon activity within a lake will therefore reflect the water balance over a period of a few days prior to measurement. Importantly, since radon activities in rainfall are very low, the contribution of precipitation and surface runoff to the lake radon budget will usually be negligible, and so the radon activity will be largely determined by the groundwater inflow rate.

Despite its potential, the only previous studies that have used radon to estimate groundwater inflow to lakes or wetlands are those of Corbett et al. (1997) and Kluge et al. (2007), both of whom studied deep artificial lakes (maximum water depths of 18–20 m). In contrast, this paper examines groundwater inflow to a relatively small, shallow wetland (maximum water depth less than 2 m, and mean depth less than 1 m). Because of its shallow depth, loss of radon to the atmosphere is a much more important component of the radon balance than in the aforementioned studies, and we have estimated this from the loss rate of artificially injected SF_6. Also, because of the shallow depth of the wetland, circulation is limited and so radon activities show significant spatial variation. The pattern of spatial variation yields important information on locations of groundwater inflow, but complicates the calculation of the mass balance.

Theory

Wetland water and solute balance

The surface water budget for a lake or wetland system can be simply expressed

$$\frac{\partial V}{\partial t} = I_s + I_g + PA - Q - EA \quad (1)$$

where V is the water volume (m^3), I_s is the surface water inflow rate (m^3/day), I_g is the groundwater inflow rate (m^3/day), Q is the combined surface water and groundwater outflow rate (m^3/day), P is the precipitation rate (m/day), E is the evaporation rate from the water surface (m/day), A is the surface water area (m^2) and t is time.

The radon activity of the atmosphere, and thus of precipitation is negligible. Although surface runoff will also contain negligible radon, stream inflow may contain radon if the stream receives groundwater inflow. The change in radon mass within a wetland over time can therefore be expressed

$$\frac{\partial c_L V}{\partial t} = I_g c_g + I_s c_s + FA_b - Qc_Q - kAc_w - \lambda V c_L \quad (2)$$

where c_L is the mean radon activity within the surface water (Bq/L), c_w is the mean radon activity immediately below the water surface (Bq/L), c_g is the mean activity in inflowing groundwater (Bq/L), c_s is the mean activity of surface water inflow (Bq/L), c_Q is the mean outflow activity (Bq/L), F is the diffusive flux from the underlying sediments (Bq/m^2/day), A_b is the area of the lake bottom (the sediment-water interface) (m^2), k is the gas transfer velocity (m/day), and λ = 0.18 day^{-1} is the radioactive decay constant. For well-mixed lakes, which show no spatial variability in radon activity, c_L, c_w and c_Q will be equal. More generally, however, these parameters may take different values.

The change in activity with time is calculated by observing that $\frac{\partial c_L V}{\partial t} = c_L \frac{\partial V}{\partial t} + V\frac{\partial c_L}{\partial t}$, and is therefore given by

$$V\frac{\partial c_L}{\partial t} = I_g c_g + I_s c_s + FA_b - Qc_Q - kAc_w - \lambda V c_L - c_L\frac{\partial V}{\partial t} \quad (3)$$

If I_s, I_g, P, E, Q, c_g, c_s, c_Q and k are constant in time, then $\frac{\partial V}{\partial t} = \frac{\partial c_L}{\partial t} = 0$. This is referred to as *steady state* conditions. Substituting for Q using (1) and re-arranging then gives

$$I_g = \frac{I_s(c_Q - c_s) + PAc_Q - EAc_Q - FA_b + kAc_w + \lambda V c_L}{c_g - c_Q} \quad (4)$$

It is often assumed that the wetland is perfectly mixed, so that $c_Q = c_w = c_L$. In this case, the steady state solution becomes

$$I_g = \frac{(I_s + PA - EA + kA + \lambda V)c_L - I_s c_s - FA_b}{c_g - c_L} \quad (5)$$

It is apparent from (5), that if the radon contribution from surface water inflow ($I_s c_s$) and the diffusive flux from the sediments (FA_b) are both negligible and $c_g \gg c_L$, then the groundwater inflow rate will be directly proportional to the radon activity within the wetland.

The mean residence time of radon within a lake (t_r) is the ratio between the mass stored within the lake at any time and the rate of radon loss, and is given by

$$t_r = \frac{c_L V}{Qc_Q + kAc_w + \lambda V c_L} = \left[\frac{Q}{V}\frac{c_Q}{c_L} + \frac{k}{b}\frac{c_w}{c_L} + \lambda\right]^{-1} \quad (6)$$

where b is the mean water depth ($b = V/A$). The residence time therefore reaches a maximum value ($t_r = \lambda^{-1}$ = 5.5 days) when the lake is deep and the rate of outflow, Q, is small. For shallow lakes, the residence time may be very short (<1 day).

The radon diffusive flux

Diffusion of radon into the lake from underlying sediments can be estimated if the sediment properties and radon production rate within the sediment are known. Under steady state conditions, the transport of radon within the underlying saturated sediments can be expressed

$$D\frac{\partial^2 c}{\partial z^2} - \lambda\varepsilon c + \gamma = 0 \quad (7)$$

where c is the radon activity within the sediment pore water (Bq/L) as a function of depth (z), D is the effective diffusion coefficient for radon within the sediments (m^2/day), γ is the production rate per unit volume of the bulk sediment (Bq/cm^3/day), ε is the porosity (dimensionless), and z is depth below the bottom of the lake.

The solution to (7) subject to a constant concentration upper boundary condition has been presented by Martens et al. (1980), and used to estimate the diffusive radon flux from marine sediments. The radon activity within the sediments is

$$c = \frac{\gamma}{\lambda\varepsilon} + \left(c_L - \frac{\gamma}{\lambda\varepsilon}\right)\exp\left(-\sqrt{\frac{\lambda\varepsilon}{D}}z\right) \quad (8)$$

At $z = 0$, $c = c_L$, while as $z \to \propto$,

$$c(z) \to \frac{\gamma}{\lambda\varepsilon} \quad (9)$$

For most reasonable values of D and ε, $c(z)$ will be approximately equal to this equilibrium value below a depth of 5–10 cm. The radon flux into the lake is given by

$$F = D\left.\frac{\partial c}{\partial z}\right|_{z=0} = \left(\frac{\gamma}{\lambda\varepsilon} - c_L\right)\sqrt{\lambda\varepsilon D} \quad (10)$$

Alternatively, (7) can be solved subject to an upper boundary condition representing a well-mixed lake subject to radon loss through radioactive decay and gas exchange. We assume that water within the lake is well mixed vertically. The radon activity $c(0)$ at the top of the sediment column will then be identical to the activity within the lake (c_L). If radon within the lake is derived only from diffusion, then the upper boundary condition can be written

$$A_b D\left.\frac{\partial c}{\partial z}\right|_{z=0} = kAc(0) + \lambda Abc(0) \quad (11)$$

where b is the water depth (m). $A_b D\left.\frac{\partial c}{\partial z}\right|_{z=0}$ is the radon flux into the lake, and $kAc(0)$ and $\lambda Abc(0)$ are the radon loss rates from the lake due to gas exchange and radioactive decay, respectively. Eq. (11) can be derived from (2), by setting $\frac{\partial c_L V}{\partial t}$, I_g, I_s and Q equal to zero (and substituting using $V = Ab$). The solution to (7) subject to (11) and a semi-infinite lower boundary condition is

$$c = \frac{\gamma}{\lambda\varepsilon}\left(1 - \left(1 + \frac{A_b\sqrt{\lambda\varepsilon D}}{kA + \lambda Ab}\right)^{-1}\exp\left(-\sqrt{\frac{\lambda\varepsilon}{D}}z\right)\right) \quad (12)$$

Radon activity in the lake is therefore given by

$$c(0) = \frac{\gamma}{\lambda\varepsilon}\left(\frac{A_b\sqrt{\lambda\varepsilon D}}{kA + \lambda Ab + A_b\sqrt{\lambda\varepsilon D}}\right) \quad (13)$$

and the diffusive flux of radon into the lake (Bq/m^2/day) is

$$F = D\left.\frac{\partial c}{\partial z}\right|_{z=0} = \frac{D\gamma}{\sqrt{\lambda\varepsilon D}}\left(1 + \frac{A_b\sqrt{\lambda\varepsilon D}}{kA + \lambda Ab}\right)^{-1} \quad (14)$$

Site description

Honan Native Forest Reserve is located approximately 16 km west-northwest of Mount Gambier, in the southeast of South Australia (Fig. 1). It consists of 1030 hectares of

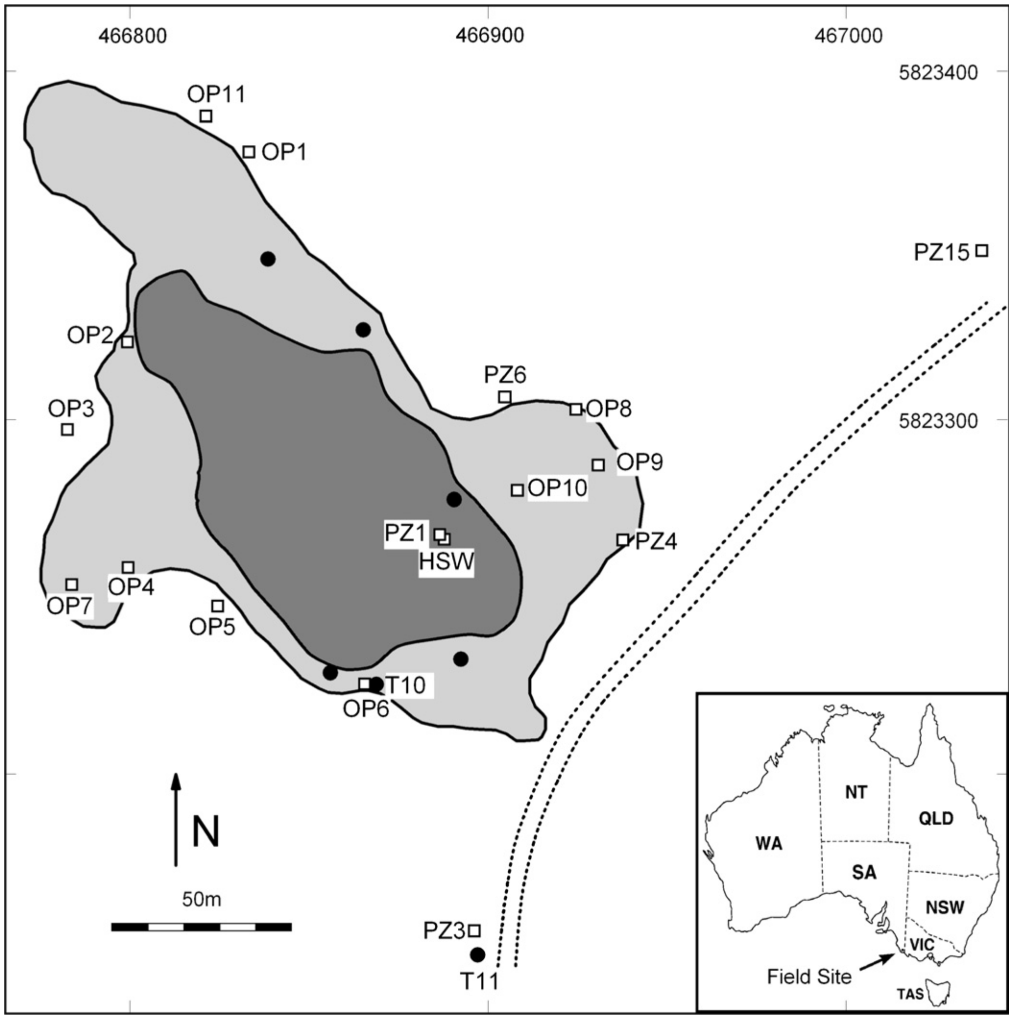

Figure 1 Location map. Open squares denote the locations of permanent piezometers (labelled PZ) and temporary groundwater sampling sites (OP), and circles denote sediment sampling sites (T or unlabelled). The dark grey area denotes the approximate extent of annual inundation, while the light grey denotes the area free of woody vegetation (and which probably reflects the maximum extent of inundation in high rainfall years). The unshaded area is covered with a dense scrub-forest. The broken line represents an access road. Piezometer PZ2 (not shown) is located approximately 180 m ENE of PZ15.

native vegetation and 160 hectares of wetlands, and is the largest area of conserved native forest in this part of South Australia. Mean annual rainfall is approximately 800 mm, and average annual evaporation is approximately 1430 mm. The regional unconfined aquifer occurs in the Gambier Limestone, a sub-karstic dual porosity limestone aquifer (Love et al., 1993). Groundwater salinity is mostly between 500 and 1000 mg/L total dissolved solids, and the aquifer is extensively used for irrigation. In the vicinity of Honan Native Forest Reserve, the Gambier Limestone is overlain by between 5 and 10 m of interbedded sands and clays. In places, the Gambier Limestone aquifer may be partly confined, and perched aquifers may form within the sand layers.

All of the wetlands in Honan Native Forest Reserve are considered to be perennial, although since they are mostly less than 1 m in depth, they vary greatly in size in response to climatic variations, and may dry completely in drought years. Vegetation of the reserve is a mixed closed forest, with an overstorey dominated by *Eucalyptus baxteri* and other eucalptus species and an understorey of heath and bracken. The wetlands themselves are usually surrounded by a dense thicket of *Leptospermum continentale* and various eucalyptus species. Aquatic plants within the regularly inundated area of the wetland include rushes and water lilies. Fig. 1 depicts the wetland chosen for detailed study. No streams enter the wetland, and for most of the year, there is no surface water inflow. During particularly wet periods, however, surface runoff from surrounding areas may enter the wetland. Surface water outflow may also occur during wet periods, but was not observed during the course of the present study. On 24 May 2006, the area inundated by surface water was measured at 5000 m^2, and this increased to 5500 m^2 by 28 September. Over this time, the salinity of the water varied between 550 and 850 mg/L. On 25 July 2006 the water depth was measured at 55 locations, and the mean depth thus determined to be approximately 0.15 m. (The maximum measured water depth was 90 cm.) By 29 November 2006, after a prolonged drought, the wetland was completely dry.

Methods

Water levels and radon activities

A number of piezometers have been installed in the vicinity of the wetland (labelled PZ in Fig. 1). Most piezometers are constructed of 50 mm diameter PVC casing, with screens between 0.5 and 1.0 m in length. Piezometer PZ15, however, is a larger diameter well, with a longer screened interval. (Screen lengths of piezometers are given in Table 3). Water levels in a number of these piezometers have been measured at two-h intervals using pressure transducers and data loggers. The surface water level in the wetland was measured in the same manner using a stilling well (screened above the land surface). Barometric pressure was also measured on-site, and used to convert pressures to water levels.

Groundwater samples were collected from piezometers after first purging three well volumes. Groundwater samples from the shallow perched aquifer were also obtained from 10 cm diameter holes, drilled to between 0.8 and 1.2 m depth using a hand-auger (labelled OP in Fig. 1). Immediately after drilling, the water in these holes was removed using a small submersible pump or bailer. The following day, water which had re-entered the holes was sampled. The holes were then backfilled.

Surface water samples were collected from throughout the wetland on 24 May, 25 July and 9 October 2006. Samples were pumped from 5 to 10 cm below the water surface, and were collected only where the water depth was greater than approximately 10 cm. No attempt was made to collect samples from different depths, which would have been difficult due to the shallow water depth. It is assumed that no significant vertical stratification of radon activity occurs.

Water samples for radon determination are collected in 1250 ml plastic bottles. Within 24 h of sample collection, radon is extracted from these samples as described by Leaney and Herczeg (2006). Approximately 50 ml of water is first removed from the bottles, and then 20 mL of mineral oil scintillant is added from a pre-weighed 22 mL Teflon-coated PTFE scintillation vial. The bottle is shaken for four minutes 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 is returned to the vial, which is sealed and returned to the laboratory. The radon activity is counted by liquid scintillation, on a LKB Wallac Quantulus counter using the pulse shape analysis program to discriminate alpha and beta decay. Corrections are made for radioactive decay between the time of sampling and measurement. Efficiency of radon extraction and counting is approximately 50%, and duplicates are usually within 5%. Water samples obtained from sealed chambers (see below) are collected in 500 ml plastic bottles, which were independently calibrated.

Radon diffusive flux

Sediment bulk density, particle bulk density and porosity were measured at four locations on the edge of the wetland. A piece of steel tubing approximately 20 cm in length (2.5 cm radius) was hammered into the sediment (to approximately 10 cm depth). The core was then carefully dug out, oven-dried and weighed. Bulk density ($\rho_{b,}$ g/cm^3) is calculated as the ratio of the dry weight to the core volume. Particle density ($\rho_{s,}$ g/cm^3) was measured using a pycnometer, as described by Blake and Hartge (1986), and porosity (ε) was calculated from particle density and bulk density (Danielson and Sutherland, 1986).

Measurements of radon emanation were made on sediments collected beneath the wetland using the same method as Cook et al. (2006). Surface samples (0–10 cm) were collected by hand, and samples to 4 m depth were obtained using a hand-auger. Approximately 40 g of oven-dried sediment was sealed in 60 ml brass containers, with 20 ml of mineral oil scintillant. The balance of the volume ($\sim$ 20 ml) was filled with distilled water. After a period of several weeks, the radon activity within the chamber will reach a constant value as the radon production rate from the sediment will be exactly balanced by the radon lost by radioactive decay. After allowing six weeks for this secular equilibrium condition to be reached, the mineral oil was sampled and its radon activity was measured. By using a series of radium standards, the efficiency of this process (percentage of emanated radon that is captured in the scintillant) was determined to be approximately 60%. Analytical error, determine from variation between duplicate samples, is approximately 25%. The radon activity in the mineral oil is used to calculate the total radon emanation rate, E_m (Bq/kg), which is related to the radon production rate, γ (Bq/cm^3/day), by:

$$\gamma = E_{\mathrm{m}}(1-\varepsilon)\rho_{\mathrm{s}}\lambda \tag{15}$$

A number of sealed chambers were installed in the wetland to assist in determining the radon flux to the wetland by diffusion alone. The sealed chambers are a variation on the seepage meter as first used by Lee (1977). The chambers were constructed from 9 kg propane gas cylinders. These cylinders were cut in half, and three fittings were welded to the bottom of the cylinders (the top of the chambers; Fig. 2). These fittings provided points to attach: (a) a plastic tube connected to a 4 L collection bag via a valve, (b) a long narrow brass tube which acted as an exhalation point for gas, and (c) a pump to allow water samples to be collected. The chambers were pushed into the sediments so that they were completed submerged below the water surface (except for the gas exhalation pipe). However, unlike conventional seepage meters, the valve attached to the collection bag (which was pre-filled with 2 L of water) was closed, thus preventing groundwater discharge to the chamber. The chambers were installed in areas of the wetland where the water depth was between 30 and 60 cm. The volume of water contained in the chamber depends on the depth to which the chambers were pushed into the wetland sediments, and ranged between 8.5 and 10.8 L. The chambers are allowed to remain in the wetland undisturbed for 2–3 weeks, to allow sufficient time for radon activities within the chambers to reach secular equilibrium. Water samples were obtained from the chambers for radon analysis by installing a hand operated peristaltic pump to the vacant fitting on the chamber, opening the valve to the collection bag, and then slowly pumping 500 ml of water into a plastic bottle. It is assumed that water that is pumped from the chamber is replenished from the collection bag, but that

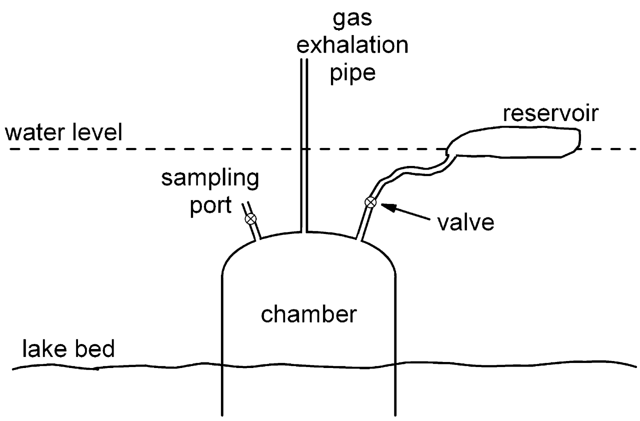

Figure 2 Sealed chamber used for measurement of radon flux into the wetland. The chambers are constructed from 9 kg propane tanks, and are 0.31 m in diameter (covering a surface area of 0.075 m^2). A gas exhalation pipe is attached to the top of the chamber to allow escape of gases. Opening of valves to the sampling port and water reservoir allow samples to be obtained from the chamber.

this does not significantly dilute the radon activity of water in the chamber. The reservoir and sampling ports on the chamber were physically separated to minimise the possibility that water from the reservoir would be sampled. Although not carried out in the current study, it would be possible to use a tracer in the reservoir water to determine its contribution to the sampled water, and hence the magnitude of any dilution of radon activity.

Gas exchange

An injected tracer experiment using SF_6 was carried out from 26 to 29 September 2006, to estimate the rate of gas exchange between the wetland and the atmosphere. Five 200-L steel drums had their ends removed, to create cylinders between 20 and 45 cm in length (diameter 57 cm). These were installed into the bed of the wetland so that they protruded approximately 2 cm above the water level, thus isolating a small part of the wetland. The surface vegetation within the cylinders was estimated, and in some cases totally removed to determine what affect vegetative cover (water lilies) had on gas transfer rates. SF_6 was injected into the water isolated by these cylinders using a diffusive system (Sanford et al., 1996). A 600 ml stainless steel vessel was filled with SF_6 to a pressure of approximately 700 kPa. The tank was then attached to 25 m of 3 mm diameter silicone tubing, via a regulator that maintained the pressure in the tubing at approximately 60 kPa. The apparatus was submerged below the water level within each cylinder for approximately 1 h, during which time SF_6 diffuses through the silicon tubing and dissolves in the water. After the injection apparatus was removed, SF_6 samples were collected from within each cylinder three times per day over the next three days. Approximately 5 ml of water was collected in pre-weighed evacuated vials, by submerging the vial beneath the water surface, and piercing a septum with a needle. SF_6 concentrations were measured on a gas chromatograph equipped with an electron capture detector using a head space method as described by Clark et al. (2004). Technical difficulties prevented local wind speed measurements, although the mean wind speed measured at Mount Gambier airport over the period of the experiment was 5 m s^{-1}. The wind speed would have been lower than this at our wetland, due to the sheltering effect of the surrounding vegetation. Since measurements were made over only a three day period, temporal variations in gas exchange velocity represent a possible source of uncertainty in our analysis. However, the magnitude of this is difficult to determine without a much more detailed study.

Bathymetry

Wetland bathymetry was determined by surveying the wetland bed when it was completely dry, and contouring this data. The topography is relatively flat, so that small changes in water depth can lead to significant variations in surface area. However, because the land surface is uneven, the edge of the wetland can be difficult to define as it grades into a series of poorly connected puddles. Since these puddles were not sampled for radon, and are not well-connected to the main water body, the water volumes and areas presented in this paper relate to areas with water depths greater than 5 cm.

Results

Hydraulics

Fig. 3 compares variations in wetland water level between 9 June and 31 October 2006 with variations in rainfall and pan evaporation at Mount Gambier, and water levels in selected piezometers. As shown, the wetland water level rose steadily until early August, and was then relatively constant until the end of September, after which time the level fell rapidly. At this time, much of southern Australia was undergoing a major drought. The wetland was completely dry by the end of November. Groundwater levels in the vicinity of the wetland vary between 0.5 and 4.0 m below ground surface, and fluctuate in response to seasonal variations in rainfall and evaporation. However, patterns of fluctuations are not well-correlated between individual piezometers, and do not describe a smooth potentiometric surface. The water level in PZ2, located approximately 320 m ENE of the wetland and screened in a shallow perched aquifer, is above the wetland level throughout the year. The water level in PZ15, located 170 m ENE of the wetland and screened in the regional unconfined aquifer is lower than that in PZ2 throughout the year. It is also lower than the wetland water level in mid-July and late October, but above the wetland water level from late July to late September.

Radon in surface water

Fig. 4 depicts the spatial variations in radon activity within the wetland on 24 May, 25 July and 9 October 2006, and water depths measured on 25 July. At each sampling time, radon activities are lowest in the central part of the wetland and towards the northwest, and highest in the southeast and along the eastern margin. The existence of such large spa-

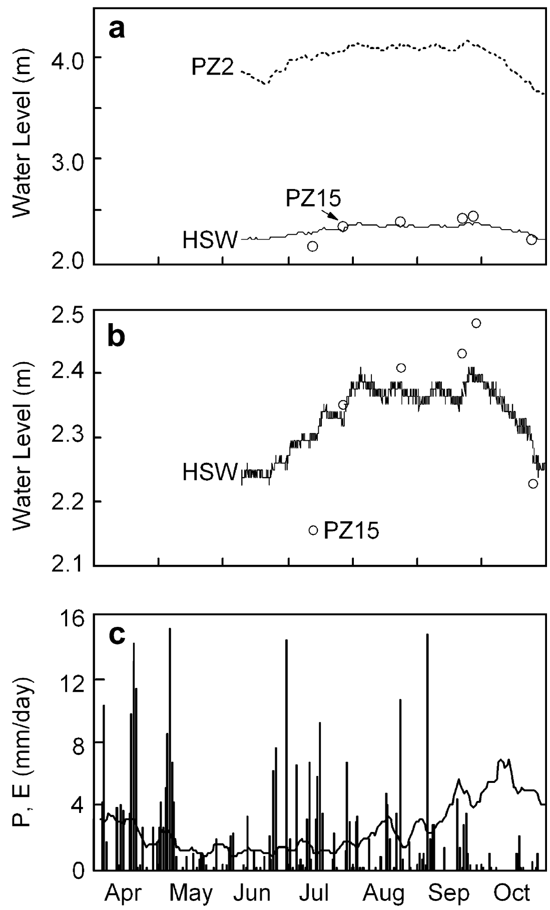

Figure 3 Variations in water levels. (a) Water level in wetland (HSW) and piezometers PZ2 and PZ15, relative to an arbitrary datum. (Levels in HSW and PZ2 were measured at two-h intervals using pressure transducers, whereas only irregular manual readings are available for PZ15.) (b) Detailed comparison of water levels in wetland and piezometer PZ15. (c) Daily precipitation (vertical bars) and the 7-day running mean of pan evaporation (line), measured at Mount Gambier (14 km east of the study site).

tial variations indicates that the timescale for mixing of the surface water is much greater than the residence time of radon. This is not surprising considering the short residence time of the tracer (6). Variations in radon activity are uncorrelated with water depth, which suggests that the spatial variations cannot be due to dilution or variations in the rate of radon loss due to gas exchange, but rather are the result of complex patterns of mixing superimposed upon spatial variations in groundwater inflow rate. The elevated radon activities near the east and southeast edges of the wetland suggest that groundwater flows into the wetland from the east and southeast, and mixes with surface water relatively slowly.

Measurements of the surface water area were made on 24 May and 28 September. Although the area was not measured on 25 July or 9 October, the water level at these times was 6 cm and 4 cm lower, respectively than on 28 September. Wetland surface water areas at these times were therefore determined from lake bathymetry data, and water volume at each sampling time was determined using the same data (Table 1). It is assumed that radon activity within the wetland does not change with depth. The mean radon activity immediately below the water surface (c_w) is there estimated from the measured radon activity, on an area-weighted basis. The mean activity (c_L) is estimated from the mean measured activity on a volume-weighted basis. The volume-weighted mean is slightly lower that the area-weighted mean (Table 1), because the deeper, central parts of the wetland tend to have lower radon activities. Nevertheless, the uncertainty in calculating these mean values is probably greater than the difference between the two numbers. There is no obvious correlation between radon activity and wetland size. The relative uniformity of radon activity over time (±50% variation) indicates a relatively uniform rate of groundwater inflow to the wetland.

Gas exchange

Fig. 5 depicts changes in SF_6 concentration over time, following injection of the tracer into isolated areas of the surface water. SF_6 concentrations decrease approximately exponentially with time, which is reflected as a linear decrease in these diagrams, in which SF_6 concentrations are plotted on a natural logarithmic scale. (The slope of the straight line is equal to the parameter k/b.) As expected, cylinders located in shallower water (G4 and G7) show more rapid gas loss, although there is no obvious relationship with vegetation cover (Table 2). There is also some evidence that the rate of gas loss is higher during the day than during the night (Fig. 5), which would be consistent with diurnal variations in wind speed. When expressed in terms of gas transfer velocity, k, (i.e., normalised for variations in depth), the rates of SF_6 loss show remarkable similarity. Estimated mean gas transfer velocities for each of the cylinders, together with estimated uncertainties (90% confidence interval for the slope of the straight line relationship between natural logarithm of concentration and time) are given in Table 2. The mean for all four cylinders combined is $k = 0.15 \pm 0.003$ m/day. The aqueous diffusion coefficient of ^{222}Rn is approximately 10% greater than for SF_6, which would suggest that its gas exchange rate is between 5% and 10% greater (Genereux and Hemond, 1992). For this reason, a gas transfer velocity of $k = 0.16$ m/day is used for ^{222}Rn in the subsequent analysis.

Radon activities in groundwater

Radon activities measured in groundwater are presented in Table 3. Within the upper 1.2 m of the aquifer system, radon activities range between 8.8 and 31.9 Bq/L, with a mean of 14.2 Bq/L and standard deviation of 7.2 Bq/L. Within the deeper piezometers, values range between 16.6 and 38.3 Bq/L, with a mean of 27.3 Bq/L.

Radon emanation rates

Radon emanation rates measured on surface sediment samples range between 5.9 and 13.7 Bq/kg. The mean bulk density measured in four surface sediment samples was 0.9 g/cm^3 and the mean particle density was 2.5 g/cm^3. The

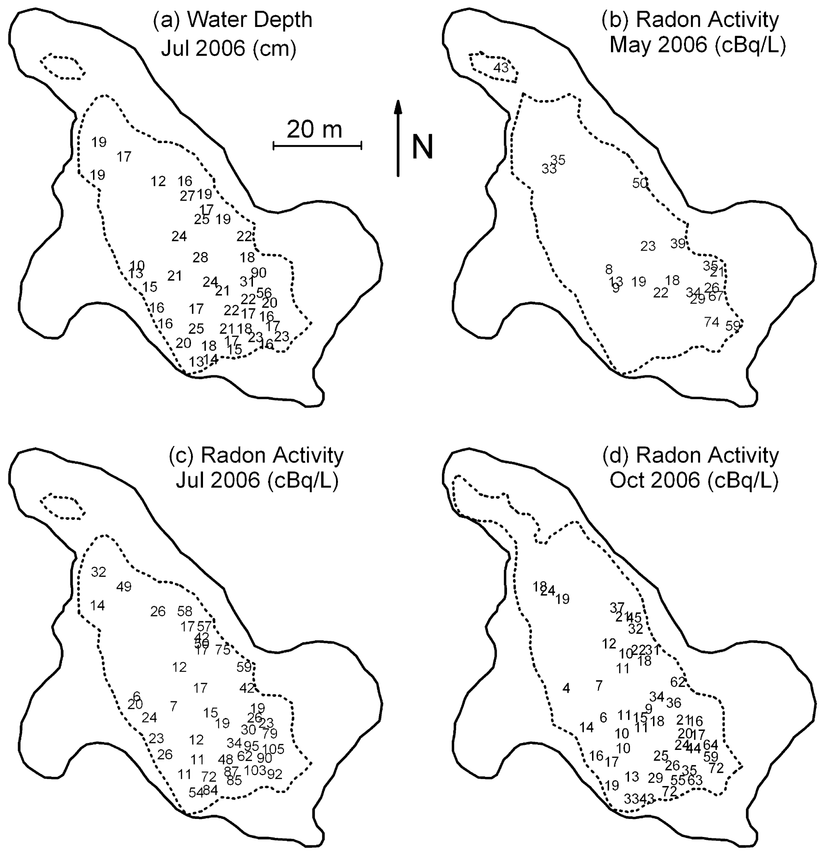

Figure 4 Spatial variations in water depth and radon activity. Numerals denote measured values at each point, although some values in densely sampled regions are not shown. Note that radon activities are reported as centiBecquerels (cBq/L) per litre, for ease of display. Dividing these values by 100 gives radon activities in Bq/L. Uncertainty, based on counting statistics, is approximately ±3 cBq/L (±0.03 Bq/L). The broken lines represent the approximate extent of surface water inundation at each sampling time, although this is difficult to accurately determine because of the very low topographic slope and the undulating ground surface.

Table 1 Temporal variations in radon activity within the wetland, and in wetland size

Date	Radon (Bq/L)				b (m)	A (m^2)	V (m^3)
	Number	Range	c_w	c_L			
24 May	20	0.08–0.74	0.34	0.31	0.11	5000	570
25 July	52	0.06–1.05	0.41	0.36	0.11	4600	510
9 Oct	59	0.04–0.72	0.27	0.23	0.12	5500	650

The number of measurements, range of activities, and area-weighted (c_w) and volume-weighted (c_L) mean activities are shown.

porosity is therefore calculated to be $\varepsilon = 0.64$, and the radon production rate for the surface sediments is estimated to be $\gamma = 1.0–2.2$ mBq/cm^3/day (15). The estimated production rate is consistent with radon activities measured in the shallow part of the aquifer. Measured values of radon activity are mostly between 8 and 21 Bq/L, while the radon activity in equilibrium with this production rate and porosity is estimated to be $c = 8.6–19$ Bq/L (9).

Variations in emanation rate with depth are depicted in Fig. 6. T10, located near the edge of the wetland shows no significant variation with depth, while at T11, emanation rates range between 4.1 and 11.5 Bq/kg. There is no obvious pattern to variations in radon activity with depth, and the variation is of similar magnitude to that observed in surface samples.

Radon flux from sediments

A number of sealed chambers were installed in the wetland to assist in determining the radon flux to the wetland by diffusion alone. Radon contained in these chambers is derived only from diffusion from sediments and is lost only by radioactive decay. Since the decay rate is known, measurement of the radon activity allows direct estimation of the diffusive flux. Radon activities measured in the chambers ranged from 0.19 to 0.73 Bq/L, with an average of 0.44 Bq/L (Table

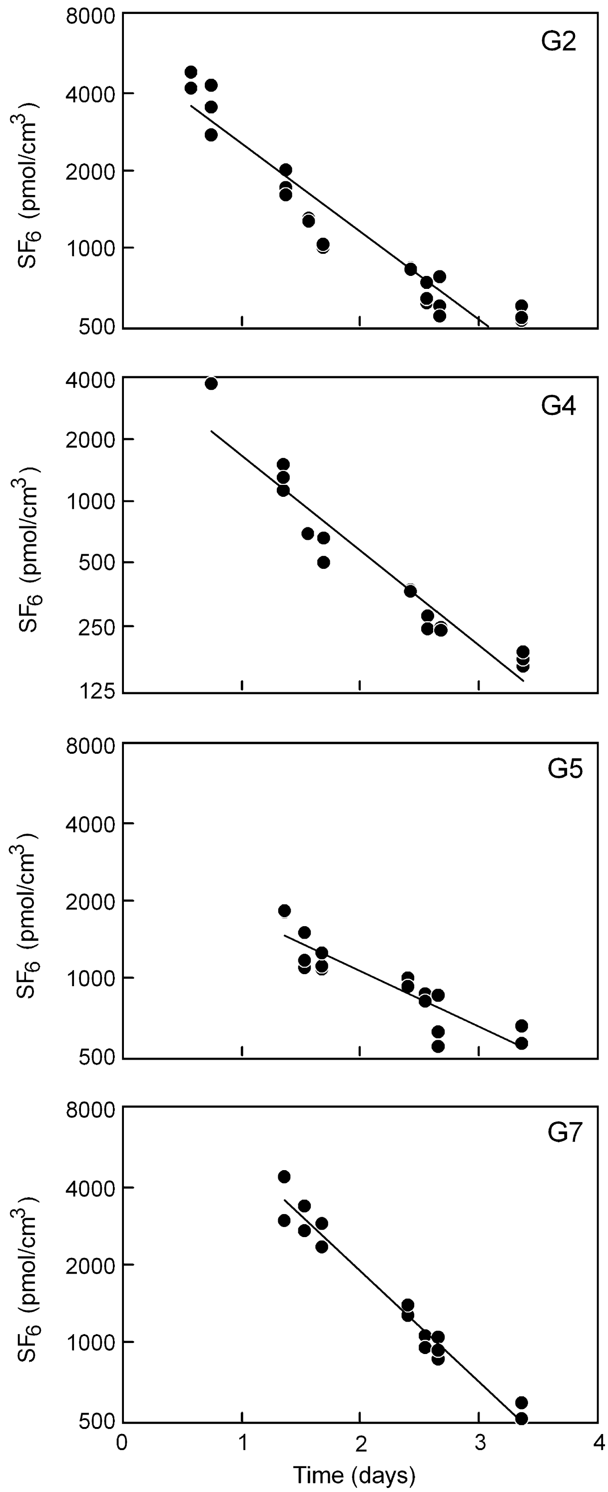

Figure 5 Changes in SF_6 concentrations over time at four sites. SF_6 injection took place on 26–27 September, and time is measured since midnight on 26 September.

4). Of course, because radon is not lost from these chambers by gas exchange, the radon activities measured within the chambers are much higher than in the wetland itself. It is worth noting that radon activities were higher in all samples during the first sampling event, by an average of 28%. The reasons for this are unclear.

Table 2 Gas transfer velocities calculated from SF_6 injection experiments

Cylinder	Depth (m)	Vegetation cover (%)	k/b (day^{-1})	k (m/day)
G2	0.25	35	0.79 ± 0.05	0.20 ± 0.01
G4	0.12	20	1.06 ± 0.05	0.13 ± 0.006
G5	0.25	0	0.50 ± 0.02	0.13 ± 0.008
G7	0.15	0	0.98 ± 0.02	0.14 ± 0.003
Mean				0.15 ± 0.003

Table 3 Radon activity in groundwater

Well No.	Intake Depth (m bgl)	^{222}Rn (Bq/L)
OP1	0–1.0	13.3 ± 0.6
OP2	0–1.0	11.5 ± 0.4
OP3	0–1.0	16.4 ± 0.5
OP4	0–1.0	8.8 ± 0.3
OP5	0–1.0	9.83 ± 0.4
OP6	0–1.2	9.26 ± 0.3
OP7	0–1.0	8.44 ± 0.4
OP9	0–1.0	21.04 ± 0.6
OP10	0–1.0	15.66 ± 0.5
OP11	0–1.0	10.58 ± 0.4
PZ1	1.5–2.0	25.81 ± 1.4
PZ3	3.9–4.9	16.61 ± 0.6
PZ4	3.7–4.2	38.25 ± 1.5
PZ6	0.3–1.0	31.9 ± 0.8
PZ15	4.0–13.6	28.44 ± 1.5

Refer to Fig. 1 for well locations.

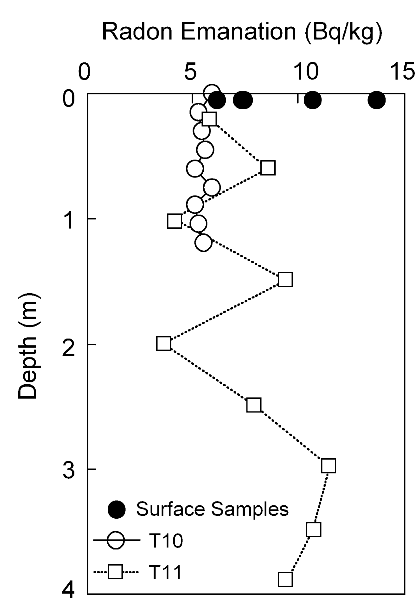

Figure 6 Radon emanation rates from lake sediments. Locations of surface samples and profiles T10 and T11 are shown in Figure 1. Uncertainty of measurements, based on the variation between duplicate samples, is approximately 25%.

The effective diffusion coefficient in the sediment for radon can be calculated from the radon activity within the isolated chambers from re-arrangement of (13), and setting $k = 0$. Using $A = A_b$ and then dividing through by A eliminates the area terms. The required parameters are then the radon production rate, the sediment porosity and the depth of water within the chambers. Using a mean radon activity within the chambers of $c(0) = 0.44$ Bq/L, a mean radon production rate of $\gamma = 1.5$ mBq/cm^3/day, and a mean water depth within the chambers of $b = 0.13$ m, the diffusion coefficient is estimated to be $D = 6.1 \times 10^{-6}$ m^2/day. The value is similar to those obtained by Sogaardhansen and Damkjaer (1987) for moist clay sediments with porosities between 62% and 71%.

The radon activity within the wetland due to diffusion alone can then be calculated by using this diffusion coefficient in the same equation, but using the mean water depth in the wetland rather than the water depth within the chambers and by including gas exchange. Thus, using $b = 0.11$ m and $k = 0.16$ m/day, the radon activity in the wetland that can be attributed to diffusion is estimated to be 0.06 Bq/L. The diffusive flux given by (14) is $F = 10.7$ Bq/m^2/day. It is noteworthy, that estimated radon activity in the wetland due to diffusion alone (13) is very similar to the lowest activity measured within the wetland on the three sampling dates (0.08, 0.06 and 0.04 Bq/L in May, July and October, respectively).

The use of (14) to calculate the radon flux assumes a one-dimensional model with diffusion as the only source of radon. Since radon activities within the wetland are greater than can be attributed to diffusion alone, this model is not strictly correct and will slightly overestimate the diffusive flux. Using (10), however, we can calculate the diffusive flux at the time of sampling by using the observed surface water activity as the upper boundary condition. However, based on $c_L = 0.06$–0.11 Bq/L, an identical value of F is obtained (10.7 Bq/m^2/day), indicating that the form of the solution used is not important.

Wetland water balance

Steady state model

Considering its short residence time, measured radon activities within the wetland are remarkably uniform (in time), with the mean value varying by about 50%. This indicates a relatively uniform rate of groundwater inflow to the wetland. Because of the short residence time, it is reasonable to use a steady state approach to estimate groundwater inflow from the measured radon activity. Since area-weighted and volume-weighted means are equivalent within their respective uncertainties, we will use the mean of these two values for both c_L and c_w. Also, because radon activity of outflow has not been measured (and is very difficult to measure) we will assume that the mean activity of outflow (c_Q) is equal to the mean activity within the surface water. Uncertainty introduced by these assumptions is discussed below. The area of the wetland sediment-water interface (A_b), was not directly measured, although due to the shallow water depth it is assumed equal to the surface water area (A).

In the five days preceding 25 July, the mean precipitation rate was 0.6 mm/day and the mean pan evaporation rate was 0.9 mm/day. Based on a wetland surface water area of 4600 m^2, this gives precipitation inflow of 2.8 m^3/day and evaporative outflow of 4.1 m^3/day. Assuming no surface runoff ($I_s = 0$) and $c_g = 15$ Bq/L, the measured radon activity of 0.385 Bq/L gives a groundwater inflow rate of 18 m^3/day (Eq. (4) and Table 5). Sensitivity of the estimated groundwater inflow rate to variation in input parameters is depicted in Fig. 7. Twenty percent uncertainties in P and E have negligible effect on the estimated groundwater inflow rate. Similar uncertainty in V (or b) and F cause only a 5% uncertainty in I_g, while a 20% uncertainty in A, k, c_L and c_g also causes a 20% uncertainty in I_g. Assuming the uncertainties in P, E, A, V, k, F, c_g and c follow normal distributions with standard deviations equal to 20% of the estimated value (so that there is a 90% probability that the actual values will be within 40% of the estimated value, and a 95% probability that they will be within 60% of this value), then we can calculate the uncertainty in I_g arising from cumulative uncertainty in all parameters using a monte carlo approach. Based on 5000 realisations, the estimated mean value for I_g is 19 m^3/day and the standard deviation is 9 m^3/day.

The estimated groundwater inflow rate (I_g) is also highly insensitive to the estimated surface water inflow rate, with a surface water inflow rate in excess of $I_s = 70$ m^3/day required to change the groundwater inflow rate by more than 10%. (Since surface water inflow occurs as surface runoff rather than stream inflow, its radon activity (c_s) is assumed

Table 4 Radon activities in isolated chambers

Chamber	Volume (L)	^{222}Rn (Bq/L)	
		26/09/2006	9/10/2006
SM1	8.5		0.70
SM3	10.0	0.45	0.30
SM7	10.8	0.23	0.19
SM10	8.5	0.50	0.30
SM13	8.5	0.73	0.57

Chambers were installed 2–3 weeks before the sampling dates indicated.

Table 5 Parameters used in steady state mass balance modelling, and estimated groundwater inflow rates

Parameter	24-May	25-July	9-October
A (m^2)	5000	4600	5500
V (m^3)	570	510	650
PA (m^3/day)	3.0	2.8	1.4
EA (m^3/day)	5.0	4.1	26
I_s (m^3/day)	0	0	0
k (m/day)	0.16	0.16	0.16
F (Bq/m^2/day)	11	11	11
c_g (Bq/L)	15	15	15
c_L (Bq/L)	0.325	0.385	0.25
c_w (Bq/L)	0.325	0.385	0.25
c_Q (Bq/L)	0.325	0.385	0.25
I_g (m^3/day)	16	18	12

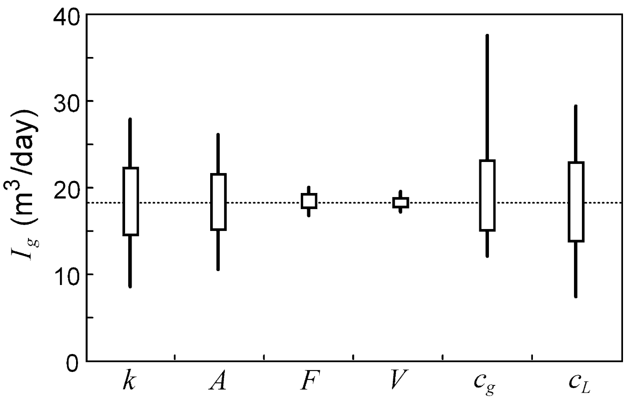

Figure 7 Sensitivity analysis for estimated groundwater inflows for 25 July using the steady state model. Rectangles depict range in I_g from ± 20% uncertainty in each parameter, while vertical lines represent ±50% uncertainty. The broken line represents the value of $I_g = 18.3\ m^3/day$, estimated using the parameters shown in Table 5. Uncertainties in I_g due to uncertainties in P and E are too small to show on this diagram.

to be zero.) The outflow, calculated as the difference between inflows ($I_s + I_g$ + PA) and evaporation (EA) is estimated to be $Q = 17\ m^3/day$. However, although I_g is insensitive to I_s, Q would be very sensitive to surface water inflow, and will increase in direct proportion to I_s (to maintain the water mass balance). Also, because groundwater outflow represents a relatively unimportant loss of radon (gas exchange is the main loss mechanism), the estimated value of I_g is not highly sensitive to c_Q. (For $c_Q = 0$, we get $I_g = 17.9\ m^3/day$, whereas for $c_Q = 0.385$ Bq/L we get $I_g = 18.3\ m^3/day$.) Similarly, the estimate of I_g is insensitive to the difference between area-weighted and volume-weighted radon activities; if we use $c_w = 0.41$ Bq/L and $c_L = 0.36$ Bq/L rather than $c_L = c_w = 0.385$ Bq/L, we get $I_g = 17.2\ m^3/day$ rather 18.3 m^3/day.

Similar inflow rates are calculated on the other sampling dates. On 24 May, the surface water area was estimated to be 5000 m^2, and the measured mean radon activity was 0.325 Bq/L. In the five days preceding radon measurement, the mean precipitation rate was 0.6 mm/day and the mean evaporation rate was 1.0 mm/day, giving a precipitation inflow of 2.1 m^3/day and evaporative outflow of 3.5 m^3/day. I_g is estimated to be 16 m^3/day. In the five days preceding 9 October, the mean precipitation rate was 0.26 mm/day and the mean evaporation rate was 4.8 mm/day, giving PA = 1.4 m^3/day, EA = 26 m^3/day and $I_g = 12\ m^3/day$.

Over the five days prior to 25 July, the water depth in the wetland did not change substantially, and so the steady state assumption may be considered reasonable. However, between 4 and 9 October, the water level was falling at a rate of 5 mm/day, and so the steady state analysis is not strictly valid. (Water level data was not available for 24 May.) However, the total change in water volume over the lifetime of radon is small and so it might be argued that the steady state approach is still likely to yield reasonable results. To confirm this, a transient model simulating changes in water level and radon activity over time was developed.

Transient model

To assess the reasonableness of the groundwater inflow rates calculated using the steady state mass balance, a transient mass balance was carried out using a finite-difference solution to (1) and (3). Daily precipitation and evaporation data were obtained from Mount Gambier airport, and the diffusive gas flux, gas exchange velocity and groundwater radon activity were assumed to be the same as in the steady state analysis. Surface water inflow, groundwater inflow and wetland outflow were adjusted manually, to reproduce observed water depths, wetland areas and radon activities. (The radon activity of surface water inflow was assumed to be zero.) Changes in surface water volume were converted to changes in water depth (d) and area (A) using bathymetry data. The model used a temporal discretisation of 0.1 days, and was performed using an EXCEL spreadsheet.

Since the radon activity in the wetland is relatively insensitive to surface water inflow and wetland outflow, calibration of the model involved manually adjusting I_s and Q to fit the observed water level and wetland area data, and then adjusting I_g to fit the observed radon activities (while making compensating adjustments to I_s and Q to preserve the water balance). Fig. 8 depicts the results of the calibrated model, which fits observed data very well.

The calibrated model uses groundwater inflows of 14.5 m^3/day on 24 May, 18 m^3/day on 25 July and 13 m^3/day on 9 October, which agree well with results of the steady state model. It should be noted that measured radon activities are only sensitive to groundwater inflow rates immediately prior to measurement. Values at other times are poorly constrained, and identical model fits could be produced by increasing or decreasing I_g while making compensating adjustments to I_s or Q to preserve the water balance. It is also important to note that because the mass of radon lost in wetland outflow is relatively small, increasing I_s produces a similar effect to decreasing Q. It should also be noted that simulation of area and depth is highly sensitive to the bathymetry, particularly in the case of a shallow wetland with a very gently sloping bed.

Discussion

In this paper, groundwater inflow to a shallow, poorly-mixed wetland has been estimated from a radon mass balance. The components of the radon budget of the wetland are (i) radon diffusive input from underlying sediments (ii) radon contribution from groundwater inflow (advection), (iii) radon loss due to gas exchange, (iv) radon loss due to radioactive decay, and (v) radon loss due to groundwater or surface water outflow. The radon activity in groundwater inflow is measured from sampling piezometers surrounding the wetland, and the groundwater inflow rate is then determined by mass balance.

There are a number of assumptions with the method. Firstly, it is assumed that the radon activity of precipitation and surface water inflow is negligible. The radon activity of atmospheric air is typically 10^3–10^4 mBq/m^3 (Zahorowski et al., 2004), and so precipitation in equilibrium with this air should have an activity of approximately 0.3–3 mBq/L. This compares with measured values of radon in groundwater that typically range between 5 and 500 Bq/L (Cecil and

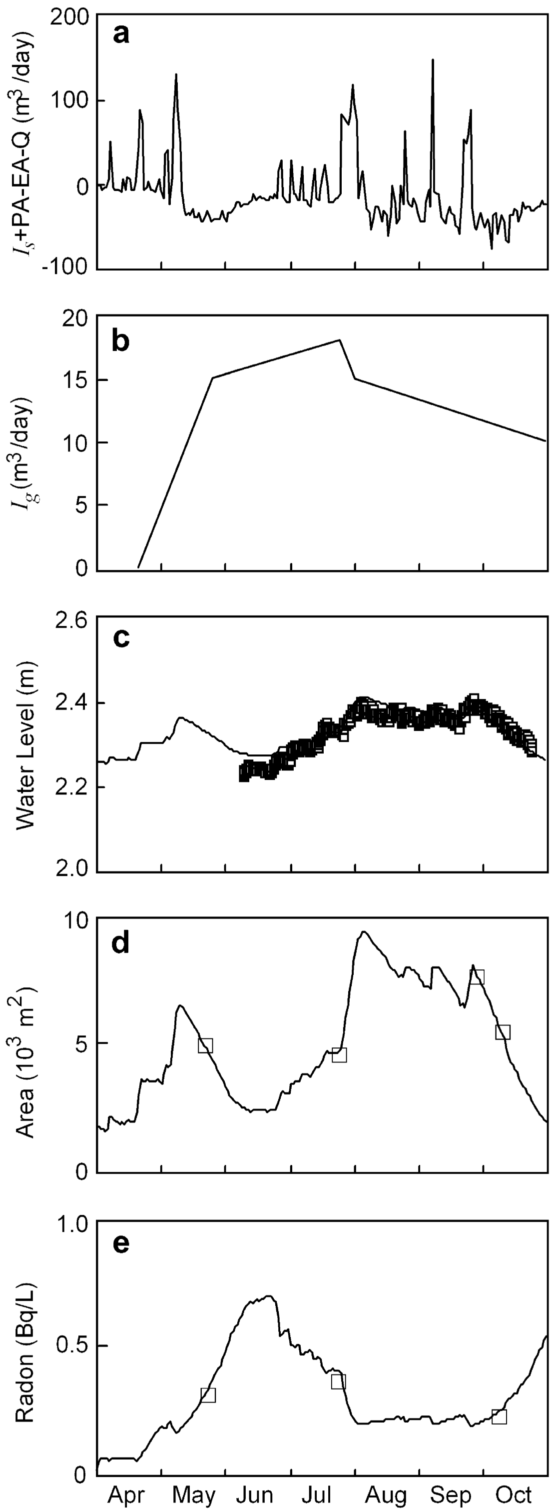

Figure 8 Results of transient mass balance model, run between 1 April and 31 October 2006. (a) Net lake inflow excluding groundwater. (b) Modelled groundwater inflow rate. (c) Modelled and measured lake water level. (d) Modelled and measured lake area. (e) Modelled and measured mean radon activity within the lake. In all cases, curves represent model data and squares denote measured values.

Green, 2000). Although conceptually, surface runoff might attain some radon through contact with sediments, it is easy to show that this will also be negligible. Eq. (13) can be used to estimate the radon activity of runoff, where b is depth of the surface water film. Using $b = 10^{-3}$ m (and other parameters as previously defined) gives $c = 0.07$ Bq/L, which is still insufficient to significantly alter the radon budget. The model also assumes that there is no advective water exchange between the wetland and the underlying sediments. Potentially, such flow could be driven by atmospheric pressure variations or wave action, and could be enhanced by biotic activity. This process is important in marine and river systems (Martens et al., 1980; Cook et al., 2006), but is likely to be less important in small wetlands, because of the reduced fetch and generally low permeability of bed sediments. It is possible that advective water exchange contributes to the radon activity measured in the isolated chambers, and if this is the case then this exchange would be incorporated in the diffusive flux term (FA_b), and lead to an inflated diffusion coefficient.

A third assumption is that the surface water is perfectly mixed vertically. This has not been verified, although the extreme shallowness of our wetland makes it highly likely, and also would make vertical profiling difficult (without inducing mixing during sample collection). Stratification of deep lakes is commonly observed, and vertical radon gradients have been documented in lakes more than 15 m deep (Kluge et al., 2007; Imboden and Joller, 1984). If vertical variation in radon activity occurs, then calculation of the mean radon activity of the lake (c_L) and the mean activity below the lake surface (c_w) will require a more complex sampling strategy. However, the equations developed in this paper are still valid, provided that these averages are calculated appropriately. Alternatively, one could write radon balance equations for different water layers in a stratified layer, and specify exchange parameters for mixing between the layers (Kluge et al., 2007).

The groundwater inflow rate estimated from the radon balance has not been validated using other methods. However, there are two important internal checks on the methodology that was used. These are (i) that the estimated radon production rate from the sediments is consistent with the measured radon activity in the groundwater, and (ii) the radon activity that is calculated to occur in the wetland if there was no groundwater inflow (and diffusion from the sediments was the only source) is very similar to the lowest measured activity within the surface water. These internal checks give confidence in the approach, and hence in the estimated groundwater inflow rates.

It is worth considering the sensitivity of estimates of groundwater inflow derived from a radon balance with sensitivities of other methods. The groundwater inflow rate estimated using radon is most sensitive to the gas exchange rate, the radon activity in groundwater inflow and lake surface area. It is also highly insensitive to the estimated surface water inflow rate, precipitation rate or lake evaporation rate. In contrast, if the groundwater inflow rate is estimated from a surface water balance, then the result would be highly dependent on these other water balance components. The main advantage of radon over other environmental tracers is that the radon activity in surface water inflow and in precipitation can be assumed to be zero, and so these are not important elements of the mass balance. This is not the case with most other environmental tracers.

However, the sensitivity of groundwater inflow rate to the gas exchange rate means that this parameter needs to be accurately estimated. For deeper lakes, however, the sensitivity to the gas exchange rate will decrease, and the main mechanism of radon loss will be radioactive decay, which is more easily quantified. On the other hand, if the groundwater inflow rate is very low, then diffusive flux from the sediments may become an important radon source.

The relatively short residence time of radon in the lake also creates important differences between this and other tracers. Firstly, it means that it is less likely that the lake will be well mixed, and so significantly spatial variations in concentration are likely to occur. This makes estimation of the outflow concentration more problematic. In our case, because lake outflow was a relatively minor component of the radon budget, estimation of the radon activity of outflow was not important. This may not always be the case. Where groundwater outflow represents a significant fraction of tracer loss, the determination of solute balances of poorly mixed lakes will be more difficult. On the other hand, the short residence time of radon means that the use of a steady state model is more easily justified, because uniform hydrological conditions are more likely to exist for short periods of time. It should be noted, of course, that measurements of radon activity in the lake reflect groundwater inflow over the preceding days, whereas concentrations of chloride or other ions may reflect the water balance over many years. Finally, it should be stressed that this method does not necessarily permit differentiation between regional groundwater inflow, and inflow from a shallow perched aquifer. Differentiation between multiple possible groundwater sources will only be possible by using a number of tracers, and even then only if the different aquifers are chemically distinct.

Conclusions

Environmental tracer methods provide a valuable means for quantifying the groundwater input to lakes and wetlands, particularly in areas where the subsurface stratigraphy is complex and so hydraulic approaches will be subject to large uncertainty. Radon differs from other environmental tracers in that it allows the groundwater inflow rate to be determined without the need to accurately quantify other components of the water budget. Because of its short residence time, it provides information on groundwater inflow rates over a relatively short period of time prior to measurement. Future studies should further assess the importance of diffusive input of radon from underlying lake sediments, and compare estimated inflow rates with those obtained from independent methods.

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