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Understanding parafluvial exchange and degassing to better quantify groundwater inflows using ^{222}Rn: The King River, southeast Australia

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ABSTRACT

^{222}Rn is an important tracer for quantifying groundwater inflows to rivers, especially where groundwater and surface water have similar major ion and stable isotope geochemistry. Uncertainties in the ^{222}Rn mass balance arise, however, from not accurately estimating the degree of degassing of ^{222}Rn to the atmosphere and the extent to which interaction within the parafluvial zone provides an additional source of ^{222}Rn. This study estimates both ^{222}Rn production in the parafluvial zone and degassing along a 75 km stretch of the King River, Australia, in order to more precisely quantify groundwater inflows. The contribution of ^{222}Rn from the parafluvial zone (F_p) was estimated using ^{222}Rn emanation rates from near-river sediments and assessment of the residence time of water in the parafluvial zone from ^{222}Rn activities and Cl concentrations in water the alluvial sediments. Values of F_p range from 40,400 Bq/m/day in the upper King to 8500 Bq/m/day in the lower King, corresponding to differences in the mineralogy and the volume of the parafluvial zone. The gas transfer coefficient (k) was estimated by matching groundwater inflows to observed increases in river discharge during a period of low discharge; k decreases from 25 day^{-1} in the upper King to 3 day^{-1} in the lower King. These k values are higher than those from most empirical formulations, probably due to the extensive pool and riffle sections that promote degassing. The King River is gaining in its upper and middle sections but several of the lower reaches are losing. Groundwater inflows on a reach scale are as high as 10 m^3/m/day and cumulative inflows along the 75 km stretch are up to 19,000 m^3/day. Groundwater inflows increase proportional to total flow reflecting the response of both groundwater and surface water systems to rainfall. Uncertainties in the calculated groundwater inflows are reduced by independently estimating k from the discharge data; however, calculated inflows are up to 40% higher if parafluvial flow were not taken into account.

1. Introduction

Documenting groundwater–river interaction is critical to understanding hydrological processes, which in turn informs protection and management of groundwater and surface water resources (Winter, 1995; Sophocleous, 2002; Brodie et al., 2007; Winter, 2007). Groundwater extraction adjacent to gaining rivers may reduce river flow, especially during periods of low rainfall, which may detrimentally impact riverine ecosystems. Underestimating the groundwater inflows to rivers may result in water resources being doubly allocated (i.e., river water and groundwater allocations might partially represent the same water). Understanding the relative contribution of groundwater to river discharge is also important for assessing potential impacts of climate change, flood forecasting, and understanding the impact of contaminants on rivers. While estimating the fluxes of groundwater to gaining rivers is commonly attempted, it is not always straightforward (Sophocleous, 2002; Brodie et al., 2007).

Providing that groundwater and surface water have different concentrations of a geochemical component, changes in concentration of that component in the river may be used to define the distribution of gaining and losing river reaches and to quantify groundwater inflows in gaining reaches (Brodie et al., 2007; Cook, 2012). Potential tracers include major ions, stable isotopes, radiogenic isotopes, and chlorofluorocarbons (Ellins et al., 1990; Genereux and Hemond, 1992; Genereux and Pringle, 1997; Cook et al., 2003; Negrel et al., 2003; Lamontagne et al., 2005; Negrel and Petelet-Giraud, 2005; Cook et al., 2006; Lamontagne and Cook, 2007; Mullinger et al., 2007; Stellato et al., 2008; Mullinger et al., 2009; Cartwright et al., 2011; Cook, 2012; Unland et al., 2013; Yu et al., 2014).

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1.1. Radon as a tracer

^{222}Rn, which is part of the ^{238}U to ^{206}Pb decay series, is commonly used for quantifying groundwater inflows to rivers. ^{222}Rn has a half-life of 3.8 days and the activity of ^{222}Rn reaches secular equilibrium with its parent isotope ^{226}Ra over a few weeks (Cecil and Green, 2000). Because the concentration of Ra in minerals is several orders of magnitude higher than dissolved Ra concentrations in surface water, groundwater ^{222}Rn activities are commonly two or three orders of magnitude higher than those of surface water (Ellins et al., 1990; Hoehn et al., 1992; Cecil and Green, 2000; Cook, 2012). This makes ^{222}Rn a useful tracer in catchments where groundwater and surface water have similar major ion concentrations or stable isotope ratios. ^{222}Rn activities in rivers decline downstream from regions of groundwater inflow due to radioactive decay and degassing to the atmosphere.

Assuming that the ^{222}Rn activity in water in equilibrium with the atmosphere is negligible, groundwater fluxes (I in m^3/m/day) may be calculated from:

$$I = \frac{\left(Q\dfrac{dc_r}{dx} - wEc_r - F_p + kdwc_r + \lambda dwc_r\right)}{\left(c_{gw} - c_r\right)} \tag{1}$$

(Cook et al., 2003, 2006; Mullinger et al., 2007; Cartwright et al., 2011; Cook, 2012) where Q is the river discharge (m^3/day), c_{gw} and c_r are the ^{222}Rn activities of groundwater and river water respectively (Bq/m^3), x is the distance along the river (m), d is the river depth (m), w is the river width (m), E is the evaporation rate (m/day), F_p is the flux of ^{222}Rn derived from interaction of the river water with the hyporheic or broader parafluvial zone (Bq/m/day), k is the gas transfer coefficient (day^{-1}), and λ is the decay constant (0.181 day^{-1}: Cecil and Green, 2000); parameters are summarised in Table 1. Eq. 1 accounts the increase ^{222}Rn activities of river water by groundwater inflows, addition from the parafluvial zone, and evaporation (which increases the concentration of all solutes including Rn) and the loss of ^{222}Rn by degassing to the atmosphere and radioactive decay.

While ^{222}Rn has been used in numerous studies to determine groundwater inflows to rivers, several uncertainties remain. ^{222}Rn activities of groundwater in some catchments are poorly defined or heterogeneous (Mullinger et al., 2009; Cartwright et al., 2011; Unland et al., 2013). Where the ^{222}Rn activities of groundwater are well characterised, the gas transfer coefficient and the flux of ^{222}Rn from the hyporheic and parafluvial zones are the least well characterised parameters (Ellins et al., 1990; Genereux and Hemond, 1992; Cook et al., 2003, 2006; Mullinger et al., 2007, 2009). Evaporative concentration is generally insignificant compared with the other terms.

1.2. Degassing

The rate of Rn degassing increases with increasing river turbulence and decreasing river depth. While there are several empirical formulations that predict k from river widths, depths, and velocities, different formulations yield different k values (Ellins et al., 1990; Genereux and Hemond, 1992; Mullinger et al., 2007, 2009; Cartwright et al., 2011; Wallin et al., 2011). Additionally, measured gas transfer coefficients for CO_2, propane, and O_2 of >100 day^{-1} in first-order upland streams may be significantly higher than those predicted from empirical formulations due to turbulent flow (Genereux and Hemond, 1992; Wallin et al., 2011). While it is possible to measure gas transfer coefficients directly using introduced gas tracers such as propane or SF_6 (Genereux and Hemond, 1992; Wallin et al., 2011), such measurements have generally been made along small reaches of a river that may not be representative of the river as a whole (e.g., if the reach did not include rapids). It is also possible to estimate k by comparing the observed variations in ^{222}Rn concentrations with those predicted from various formulations of k (Mullinger et al., 2009; Cartwright et al., 2011); this is most easily accomplished in rivers that are losing or where groundwater inflows are otherwise known.

1.3. Flux of ^{222}Rn from hyporheic and parafluvial zones

^{222}Rn can also be introduced into rivers by emanation from alluvial sediments into water flowing through the hyporheic zone (the zone immediately below the river bed through which water flows driven by irregularities in the river bed) and the broader parafluvial zone, which includes features such as point bars and gravel banks (Boulton et al., 1998; Fernald et al., 2001; Wörman et al., 2002; Edwardson et al., 2003). It is important to quantify this source of ^{222}Rn in order not to overestimate groundwater inflows (Cook et al., 2006). At steady state the ^{222}Rn activity of water in the alluvial sediments is γ/λ where γ is the radon emanation rate (Bq/m^3/day) (Cecil and Green, 2000). The ^{222}Rn activity in the parafluvial zone (c_p) is a function of the ^{222}Rn activity of the water flowing into the parafluvial zone (c_{in}), the equilibrium ^{222}Rn activity, and the residence time in the parafluvial zone (t_p, in days):

$$c_p = \left(\frac{\gamma}{\lambda} - c_{in}\right)\left(1 - e^{-\lambda t_p}\right) + c_{in} \tag{2}$$

(modified from Hoehn et al., 1992). For t_p less than few days, the numerical solution of Lamontagne and Cook (2007, their Eq. 4) yields near identical t_p vs. c_p relationships. In a losing river $c_{in} = c_r$. In a gaining river water derived from the river will mix in the alluvial sediments with upwelling regional groundwater that has high ^{222}Rn activities. It

Table 1
Summary of parameters.

Symbol	Parameter	Units	Comments
c_r, c_{gw}, c_p	^{222}Rn activities in river, groundwater, parafluvial zone	Bq/m^3	
c_{in}	Initial ^{222}Rn activity in water entering the parafluvial zone	Bq/m^3	
Cl_r, Cl_{gw}, Cl_p	Cl concentrations in river, groundwater, parafluvial zone	mg/L	
x	Distance along river	m	From site 1
w	River width	m	Upper: 10–15, Middle: 15–20, Lower: 10–20
d	River depth	m	Upper: 0.5–1.5, Middle: 0.5–2, Lower: 0.75–2.5
A_p	Cross-sectional area of parafluvial zone	m^2	Upper: 7.5, Middle: 10, Lower: 5
Q	River discharge	m^3/day	Measured at sites 1, 14, 24, 27
E	Evaporation rate	m/day	
k	^{222}Rn gas transfer coefficient	day^{-1}	Calculated from March 2013 observations
λ	^{222}Rn decay constant	day^{-1}	0.181
F_p	Flux of ^{222}Rn from parafluvial zone	Bq/m/day	Eq. (4)
γ	^{222}Rn emanation rate	Bq/m^3/day	
I	Groundwater inflows	m^3/m/day	Eq. (1)
t_p	Residence time in parafluvial zone	day	
ϕ	Porosity		0.4