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Using geochemical tracers to distinguish groundwater and parafluvial inflows in rivers (the Avon Catchment, SE Australia)

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Understanding the location and magnitude of groundwater inflows to rivers is important for the protection of riverine ecosystems and the management of connected groundwater and surface water systems. Downstream trends in ²²²Rn activities and CI concentrations in the Avon River, southeast Australia, implies that it contains alternating gaining and losing reaches. ²²²Rn activities of up to 3690 Bq m⁻³ imply that inflows are locally substantial (up to 3.1 m³ m⁻¹ day⁻¹). However, if it assumed that these inflows are solely from groundwater, the net groundwater inflows during low-flow periods exceed the measured increase in streamflow along the Avon River by up to 490%. Uncertainties in the ²²²Rn activities of groundwater, the gas transfer coefficient, and the degree of hyporheic exchange cannot explain this discrepancy. It is proposed that a significant volume of the total calculated inflows into the Avon River represents water that exfiltrates from the river, flows through parafluvial sediments, and subsequently re-enters the river in the gaining reaches. This returning parafluvial flow has high ²²²Rn activities due to ²²²Rn emanations from the alluvial sediments. The riffle sections of the Avon River commonly have steep longitudinal gradients and may transition from losing at their upstream end to gaining at the downstream end and parafluvial flow through the sediment banks on meanders and point bars may also occur. Parafluvial flow is likely to be important in rivers with coarse-grained alluvial sediments on their floodplains and failure to quantify the input of ²²²Rn from parafluvial flow will result in overestimating groundwater inflows to rivers.

1 Introduction

Quantifying groundwater inflows to streams and rivers is critical to understanding hydrogeological systems, protecting riverine ecosystems, and managing water resources (e.g., Winter, 1999; Sophocleous, 2002; Brodie et al., 2007). Groundwater inflows may form the majority of water in gaining rivers during periods of low streamflow, and river-

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ine ecosystems are commonly sustained by groundwater inflows at those times (Kløve et al., 2011; Barron et al., 2012; Cartwright and Gilfedder, 2015). Thus, understanding the distribution and magnitude of groundwater inflows is important for managing and protecting these commonly vulnerable ecosystems. Failure to understand groundwater contributions to rivers may result in double allocation of water resources (i.e., the surface water and groundwater allocations might represent the same water). Documenting the distribution and quantity of groundwater inflows to rivers is also required for flood forecasting, understanding the impacts of contaminants on rivers, and understanding the potential impacts of climate or landuse changes on river systems.

There are many methods to assess the distribution and magnitude of groundwater inflows to rivers (e.g., Brodie et al., 2007). Differential gauging may be used to determine both groundwater inflows and outflows. In many catchments, however, the distribution of river gauges is insufficient to allow anything other than a large-scale categorisation of gaining and losing behaviour. Numerical techniques based on river hydrographs (e.g., Nathan and McMahon, 1990; Askoy et al., 2009) provide a straightforward method of estimating baseflow. However, these are of limited use in highly-regulated rivers and may aggregate several stores of water including regional groundwater, bank return flows, and interflow into the baseflow component (McCallum et al., 2012; Cartwright et al., 2014a). Groundwater flow models and water balance models may also be used to calculate groundwater inflows but their applicability requires a detailed knowledge of hydrological parameters such as hydraulic conductivity, groundwater elevations, evapotranspiration rates, and runoff.

Providing that groundwater and surface water have different concentrations of a given geochemical component, changes in the geochemistry of the river may be used to estimate groundwater inflows into individual reaches (e.g., Cook, 2013). Tracers such as major ions, stable isotopes, radioactive isotopes, and chlorofluorocarbons have been used to quantify groundwater inflows to rivers (e.g., Ellins et al., 1990; Genereux and Hemond, 1992; Négrel et al., 2001; Stellato et al., 2008; Cartwright et al., 2011; Cook, 2013; Bourke et al., 2014a, b). Geochemical tracers only quantify

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5 1.1 ²²²Rn as a tracer of groundwater inflows

²²²Rn, which is an intermediate isotope in the ²³⁸U to ²⁰⁶Pb decay series, is an important tracer for quantifying groundwater inflows to rivers. ²²²Rn has a half-life of 3.8 days and the activity of ²²²Rn reaches secular equilibrium with its parent isotope ²²⁶Ra over 3 to 4 weeks (approximately 5 half-lives) (Cecil and Green, 2000). Because the concentrations of Ra in minerals are several orders of magnitude higher than dissolved Ra concentrations in surface water, groundwater ²²²Rn activities are commonly two or three orders of magnitude higher than those of surface water (Cecil and Green, 2000). This makes ²²²Rn a valuable tracer in catchments where the groundwater has similar major ion concentrations and stable isotope ratios to the river water. As ²²²Rn activities in rivers decline downstream from regions of groundwater inflow due to radioactive decay and degassing to the atmosphere (Ellins et al., 1990; Genereux and Hemond, 1992), ²²²Rn is also useful in determining locations of groundwater inflow, even if the inflows are not quantified.

Assuming that the atmosphere contains negligible radon, the change in ²²²Rn activities along a river may be estimated by mass balance:

$$Q\frac{\mathrm{d}c_{\mathrm{r}}}{\mathrm{d}x} = I\left(c_{\mathrm{gw}} - c_{\mathrm{r}}\right) + wEc_{\mathrm{r}} + F_{\mathrm{h}} + F_{\mathrm{p}} - kdwc_{\mathrm{r}} - \lambda dwc_{\mathrm{r}} \tag{1}$$

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zone and the broader parafluvial zone, respectively; k is the gas-transfer coefficient (day^{-1}) ; and λ is the decay constant $(0.181\,day^{-1})$ (Table 1). Unlike most applications of 222 Rn mass balance, F_h and F_p have been separated which allows both small-scale (hyporheic) and larger-scale (parafluvial) flow through the near-river sediments to be considered (c.f., Bourke et al., 2014a). Equation (1) can also be used to estimate the changes in composition of major ion tracers. Since the concentration of a conservative tracer such as CI is controlled only by groundwater inflows and evaporation, only the first two terms on the right-hand-side of Eq. (1) are relevant. If the river is solely fed by groundwater the increase in streamflow downstream is:

$$_{10} \quad \frac{dQ}{dx} = I - Ew. \tag{2}$$

Uncertainties in the parameters in Eq. (1) can result in significant uncertainties in calculated groundwater inflows. The evaporation term is generally much smaller than the other terms in Eq. (1), and thus uncertainties in evaporation rates have little impact on the calculations (Cartwright et al., 2011; Cook, 2013). By contrast, heterogeneous 222 Rn activities in groundwater may result in substantial uncertainties in calculated groundwater inflows (Cook et al., 2006; Mullinger et al., 2007; Unland et al., 2013; Yu et al., 2013; Cartwright et al., 2011; Atkinson et al., 2015); groundwater CI concentrations are also commonly heterogeneous (Cook, 2013; Cartwright, 2014b) and this leads to similar uncertainties when using CI to estimate groundwater inflows. Additionally, while it is well established that the rate of Rn degassing increases with increasing river turbulence and decreasing river depth, different empirical formulations that predict k from river widths, depths, and velocities can yield disparate k values (Genereux and Hemond, 1992), which can also lead to significant uncertainties in calculated groundwater inflows.

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In most rivers, there is likely to be a contribution of ²²²Rn resulting from 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; Edwardson et al., 2003). Both hyporheic and parafluvial flow represent water that exfiltrates the river and subsequently re-infiltrates at some point downstream. The ²²²Rn activity in water flowing through the hyporheic and parafluvial zones increases over time due to ²²²Rn emanation from the sediments and the contribution of ²²²Rn from these sources needs to be estimated when calculating groundwater inflows (Cook, 2013). While the contribution of ²²²Rn derived from the hyporheic zone is commonly considered, few studies have considered the contribution of ²²²Rn from the parafluvial zone (c.f. Bourke et al., 2014a; Cartwright et al., 2014b); however, in rivers with coarsegrained sediments on the floodplain the input of ²²²Rn from the parafluvial zone may be significant.

The 222 Rn activity in the hyporheic or parafluvial zone waters ($c_{\rm h}$ or $c_{\rm p}$) is governed by the 222 Rn activity of the water flowing into the hyporheic zone ($c_{\rm in}$), the 222 Rn emanation rate (γ in Bq m $^{-3}$ day $^{-1}$), and the residence time ($t_{\rm h}$ or $t_{\rm p}$, in days):

$$c_{h} = \left(\frac{\gamma}{\lambda} - c_{in}\right) \left(1 - e^{-\lambda t_{h}}\right) + c_{in} \tag{3}$$

(Hoehn et al., 1992; Hoehn and Cirpka, 2006) (Fig. 1a). $c_{\rm h}$ increases with residence time $(t_{\rm h})$ and at steady state (i.e., as $t_{\rm h}$ in Eq. 3 \rightarrow ∞), $c_{\rm h} = \gamma/\lambda$. In a losing or neutral (i.e. neither gaining nor losing) river $c_{\rm in} = c_{\rm r}$. 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 and Cartwright et al. (2014b) discussed using the concentration of a conservative ion such as CI to estimate the degree of mixing within the alluvial sediments to estimate $c_{\rm in}$. Assuming that all the water entering the hyporheic zone

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subsequently re-enters the river, the 222 Rn flux from the hyporheic zone (F_h) is given by:

$$F_{\rm h} = \frac{\gamma A_{\rm h} \phi}{1 + \lambda t_{\rm h}} - \frac{\lambda A_{\rm h} \phi}{1 + \lambda t_{\rm h}} c_{\rm in},\tag{4}$$

where A_h is the cross-sectional area of the hyporheic zone and ϕ is the porosity (Lamontagne and Cook, 2007). Equation (4) treats the hyporheic zone as a homogeneous region adjacent to the river in which river water resides for a certain period of time and then re-enters the river. While recognising that this is an oversimplification, it does provide a means of calculating the changes in 222 Rn in the hyporheic zone from estimates of emanation rates and the dimensions of the hyporheic zone.

Equation (4) can also be applied to parafluvial exchange (e.g., Cartwright et al., 2014b). However, where parafluvial flow involves long flow paths through alluvial sediments, an alternative conceptualisation is to consider the flux of 222 Rn into the river at the end of discrete flow paths through the parafluvial zone (Hoehn and Von Gunten, 1989; Hoehn and Cirpka, 2006; Bourke et al., 2014a). In this case, F_p may be defined in a similar way to the groundwater inflow term in Eq. (1):

$$F_{\rm p} = I_{\rm p}(c_{\rm p} - c_{\rm r}),\tag{5}$$

where $c_{\rm p}$ is the activity of $^{222}{\rm Rn}$ in the parafluvial zone waters and $I_{\rm p}$ is the volume of water derived from the parafluvial zone per unit length of river (m³ m⁻¹ day⁻¹). $c_{\rm p}$ is given by Eq. (3) and the minimum value of $I_{\rm p}$ required to produce a given $F_{\rm p}$ is achieved when $c_{\rm p}$ approaches steady state (Fig. 1b), which requires $t_{\rm h}$ to be at least several days ($c_{\rm p}$ is ~ 95 % of the steady state activity after 16 days: Fig. 1a). The volume of sediments with which the water has interacted during flow through the parafluvial zone ($V_{\rm p}$ in m³ m⁻¹ length of river) is governed by $I_{\rm p}$, $t_{\rm p}$ and ϕ . If the distribution of flow paths through the parafluvial zone is regular $V_{\rm p}$ will be the cross-sectional area of the

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parafluvial zone through which the water from the river flows (A_n) :

$$V_{\rm p} = A_{\rm p} = \frac{t_{\rm p}I_{\rm p}}{\phi} \tag{6}$$

(Bourke et al., 2014a). For the same input parameters, Eqs. (4) and (5) yield closely similar estimates of F_h or F_p (as noted by Bourke et al., 2014a) and the least well-known parameters are the same in both cases ($A_{\rm h}$ or $A_{\rm p}$ and $t_{\rm h}$ or $t_{\rm p}$). The choice between the two approaches largely depends on how the system is conceptualised; in the case of hyporheic exchange it is more convenient to consider the exchange between the hyporheic zone and the river whereas parafluvial flow may be better conceptualised as discrete flow paths.

The successful application of ²²²Rn to determine groundwater inflows thus requires careful consideration of several processes. While this complicates the application of ²²²Rn as a tracer, it also allows characterisation of processes such as degassing and hyporheic or parafluvial flow that may be difficult to achieve using other chemical tracers. As has been outlined in several studies, comparison of the calculated groundwater inflows from ²²²Rn with those made from other geochemical tracers or with streamflow measurements is a crucial test of the veracity of the calculations (Cook et al., 2003; Mullinger et al., 2007, 2009; Cartwright et al., 2011, 2014b; Unland et al., 2013).

Objectives

This paper examines groundwater-river interaction in the Avon River, southeast Australia, primarily using ²²²Rn. Specifically, we test the following hypothesis: (1) that largescale parafluvial flow is an important contributor of ²²²Rn to the river; and (2) that major flooding events which alter the geometry of the floodplain result in changing locations of groundwater inflows. Being able to separate groundwater and parafluvial inflows and understand how the locations of groundwater inflows change following major flood events are important to studies of groundwater-river interactions elsewhere.

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Local geology and hydrogeology

The Avon River occurs within the Gippsland Basin in southeast Australia (Fig. 2) and has a total catchment area of ~ 1830 km² (Cochrane et al., 1991; Department of Environment and Primary Industries, 2015). The Avon River is unregulated and drains the southern slopes of the Victorian Alps (maximum elevation in the catchment is 1634 m), and discharges into Lake Wellington, which is a coastal saline lake connected to the Southern Ocean. The upper slopes represent ~ 30 % of the Avon catchment and are dominated by temperate native eucalyptus forest while the majority of the plains representing ~ 70 % of the catchment have been cleared for agriculture, including dairying, sheep grazing, and vegetable production. The estimated population of the Avon catchment is ~ 4000 with Stratford being the largest town (population ~ 2000).

The highlands of the Victorian Alps comprise indurated Palaeozoic and Mesozoic igneous rocks and metasediments that only host groundwater flow in fractures or in near-surface weathered zones (Walker and Mollica, 1990; Cochrane et al., 1991). These rocks form the basement to the Tertiary and Quaternary sediments of the Gippsland Basin (Fig. 2). The shallowest regional aquifer within the Avon Catchment is the Pliocene to Pleistocene Haunted Hill Formation which comprises up to 40 m of interbedded alluvial sands and clays that have hydraulic conductivities between 10^{-/} and 10⁻⁵ m s⁻¹ (Brumley et al., 1981; Walker and Mollica, 1990). Quaternary sediments that consist of coarse-grained sand and gravels interbedded with finer-grained silts occur mainly within the river valleys and have hydraulic conductivities of 10⁻⁵ and 10⁻² m s⁻¹ (Brumley et al., 1981; Walker and Mollica, 1990).

Average rainfall within the Avon catchment ranges from $\sim 1.5\,\mathrm{m\,yr}^{-1}$ in the highlands to $\sim 0.9 \,\mathrm{m\,yr}^{-1}$ on the plains and most precipitation occurs in the austral winter (June to September) (Bureau of Meteorology, 2015). The Avon River displays strong seasonal flows with ~ 80 % of annual streamflow occurring during winter (Department of Environment and Primary Industries, 2015). This study focusses on the reaches of the Avon River located on the plains formed by the Gippsland Basin sediments that are

upstream of tidal influence. Streamflow is measured at three sites (The Channel, Stratford, and Chinns Bridge: Fig. 2). Total annual streamflow at Stratford (Fig. 2) between 1977 and 2014 ranged from 1.3×10^7 to 9.02×10^8 m³ yr⁻¹ (median = 3.04×10^8 m³ yr⁻¹) and varies with total annual rainfall (Department of Environment and Primary Industries, 2015). The Avon River only ceases to flow during the summers of severe drought years (e.g., 1983) and experiences periodic floods during high rainfall periods (Fig. 3). Streamflow generally increases downstream at all flow conditions, except at very low flows when streamflow decreases between Stratford and Chinns Bridge. Valencia Creek and Freestone Creek are the main tributaries; both are have streamflow measurements and both enter the Avon in the upper reaches of the studied section.

The Avon River has incised through the Haunted Hill Formation and Quaternary sediments to create terraces that are up to 30 m high with a lower floodplain that is up to 500 m wide. Where it crosses the sedimentary plains, the Avon River comprises a sequence of slow-flowing pools that are typically 10 to 30 m wide, up to 2 m deep at low flows, and up to 2 km long. These pools are connected by shorter (typically 10's to 100's m) and narrow (typically < 5 m) faster-flowing riffle sections that commonly have steep longitudinal gradients.

The floodplain of the Avon River comprises numerous gravel banks and point bars of coarse-gained unconsolidated sediments with clasts of up to 50 cm in diameter. The coarse-grained alluvial sediments are most common between Browns and Redbank (Fig. 2); downstream of Redbank, the Avon River occupies an incised channel with steep banks of finer-grained (clay to sand sized) sediments. The alluvial sediments are sparsely vegetated and the geometry of the floodplain changes markedly following flood events, such as those in 2011, 2012, and 2013 (Fig. 3). These changes include the downstream migration of pools (often by several tens of metres), scouring of the alluvial sediments, and changes to the size and location of the sediment banks. In regions where the river is incised, there are seeps of water at the base of the slope and permanent patches of vegetation such as Juncus that commonly colonise waterlogged soils in Australia.

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Groundwater flows from the Victorian Alps to the coast (Hofmann and Cartwright, 2013: Fig. 2); however, there are few groundwater monitoring bores in the Avon catchment that can be used to construct detailed groundwater flow paths or define the relationship of the Avon River to the groundwater. Both the Avon River and shallow groundwater from the Haunted Hills and Quaternary aguifers are used for irrigation and stock use, and groundwater is also used for water supply for Briagolong (population ~ 950) (Gippsland Water, 2012). Current annual groundwater allocations are 1.4 × 10⁷ m³, although these are not fully utilised at present (Gippsland Water, 2012). Use of water from the Avon River and its tributaries for irrigation is up to $8 \times 10^6 \,\mathrm{m}^3 \,\mathrm{yr}^{-1}$ ($\sim 2.6 \,\%$ of the annual median streamflow at Stratford); however, this is seasonally adjusted with a prohibition on river water use when the streamflow at Stratford is $< 10^4 \,\mathrm{m}^3 \,\mathrm{day}^{-1}$.

The incised nature of the Avon River and the fact that it rarely ceases to flow has led to an assumption that it receives significant groundwater inflows (Gippsland Water, 2012); however, there has been little attempt to quantify the inflows or determine their distribution. Such information is required to protect and manage the Avon River, especially in assessing the potential impacts of increased groundwater or surface water use.

Methods

Sampling took place between February 2009 and February 2015 in six sampling campaigns. The sampling periods (Fig. 3a) include periods of very low streamflow (February 2009 and March 2014), periods of low to moderate streamflow (April 2010 and February 2015), and periods of higher streamflow (September 2010, and July 2014). Major flood events between 2011 and 2013 caused redistribution of the pools and sediment banks in the river. Each sampling campaign involved sampling the river sites (Table S1, Fig. 2) over a two to three day period, with the February 2015 sampling campaign involving additional sites to the others. Streamflow data is from gauging stations at The Channel, Stratford, and Chinns Bridge on the Avon River and from gauges

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on the Valencia Creek and Freestone Creek tributaries (Department of Environment and Primary Industries, 2015: Fig. 2). Distances are measured relative to the first site at Browns (Fig. 1)

Sampling and analytical techniques were similar to those in other studies (e.g. Un-5 land et al., 2013; Yu et al., 2013; Cartwright et al., 2014b). River samples were collected from 0.5-1 m below the river surface using a manual collector mounted on a pole. Groundwater was sampled from bores installed on the river bank and floodplain at Stratford and Pearces Lane (Fig. 2) that have 1 to 3 m long screens. Water was extracted using an impeller pump set at the screened interval and at least 3 bore volumes of water were purged before sampling. Cations (Tables S1 and S2) were analysed on samples that had been filtered through 0.45 µm cellulose nitrate filters and acidified to pH < 2 using a ThermoFinnigan quadropole ICP-MS at Monash University. Anions (Tables S1 and S2) were analysed on filtered unacidified samples using a Metrohm ion chromatograph at Monash University. The precision of major ion concentrations based on replicate analyses is 2–5 %. A suite of anions and cations were measured; however, only CI and Na are discussed in this study. ²²²Rn activities in groundwater (Table S2) and surface water (Table S1) were determined using a portable radon-in-air monitor (RAD-7, Durridge Co.) following methods described by (Burnett and Dulaiova, 2006) and are expressed in Bq m⁻³. 0.5 L of sample was collected by bottom-filling a glass flask and ²²²Rn was subsequently degassed for 5 min into a closed air loop of known volume. Counting times were 1-2 h for surface water and 20 min for groundwater. Typical relative precision is < 3% at $10\,000\,\mathrm{Bg\,m^{-3}}$ and $\sim 10\,\%$ at $100\,\mathrm{Bg\,m^{-3}}$.

Forty four samples of river bed sediments from sites along the Avon River were collected in order to determine ^{222}Rn production in the hyporheic and parafluvial zones and to further constrain groundwater ^{222}Rn activities. ^{222}Rn emanation rates (γ in $\text{Bq}\,\text{m}^{-3}\,\text{day}^{-1}$) (Table 2) were calculated by sealing a known dry weight of sediment in airtight containers with water and allowing ^{222}Rn to accumulate. Following 4–5 weeks incubation, by which time the rate of ^{222}Rn production and decay will have reached steady state, 20 to 40 mL of pore water was extracted and analysed for ^{222}Rn activities

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using the same method as above but with counting times of 6 to 12 h. γ is related to the ²²²Rn produced per unit mass of sediment $E_{\rm m}$ (Bq kg⁻¹ of sediment), sediment density $\rho_{\rm S}$ (kg m⁻³), and porosity ϕ by

$$\gamma = \frac{E_{\rm m}(1-\phi)\rho_{\rm s}\lambda}{\phi} \tag{7}$$

5 (Lamontagne and Cook, 2007).

4 Results

4.1 Streamflow

Streamflow at Stratford between January 2000 and February 2015 varied between 500 and $1.38 \times 10^8 \, \text{m}^3 \, \text{day}^{-1}$ (Department of Environment and Primary Industries, 2015). Despite this period including years with well below average rainfall, for example 2006 when rainfall was ~ 50 % of the long-term average (Bureau of Meteorology, 2015), there are no periods of zero streamflow. Mean daily streamflow at Stratford during the two low flow sampling rounds were 10 700 m³ day $^{-1}$ in February 2009 and 16 200 m³ day $^{-1}$ in March 2014 (Fig. 3a); these represent the 89.9 and 83.0 percentiles of streamflow, respectively (Fig. 3b). Mean daily streamflow in the other sampling rounds were 23 100 m³ day $^{-1}$ in February 2015, 24 300 m³ day $^{-1}$ in April 2010, 84 100 m³ day $^{-1}$ in July 2014, and 88 800 m³ day $^{-1}$ in September 2010, which represent the 73.5, 74.9, 41.0, and 39.5 percentiles of streamflow, respectively (Fig. 3b). In February 2015, which is the sampling round discussed in most detail below, the mean daily streamflow was 12 500 m³ day $^{-1}$ at The Channel and 25 800 m³ day $^{-1}$ at Chinns Bridge with inflows of 2410 m³ day $^{-1}$ from Valencia Creek and 600 m³ day $^{-1}$ from Freestone Creek (Department of Environment and Primary Industries, 2015).

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River geochemistry

Figure 4a shows the ²²²Rn variation in the Avon River for the six sampling campaigns. There are several distinct zones of elevated ²²²Rn activities notably at Wombat Flat (222Rn activities up to 2040 Bq m⁻³) and between Bushy Park and School Lane (222Rn activities up to 3690 Bq m⁻³). ²²²Rn activities are lowest in the upper reaches at Smyths Road (222Rn activities between 226 and 460 Bg m⁻³) and in the reaches between Stewarts Lane and Stratford (222Rn activities generally between 270 and 800 Bqm⁻³). The downstream river reaches between Knobs Reserve and Chinns Bridge also have low ²²²Rn activities of between 74 and 750 Bg m⁻³ that generally decline downstream. The position of the highest ²²²Rn activities in March 2014 and February 2015 (i.e., post the 2011 to 2013 floods) was at Bushy Park, while this site had relatively low ²²²Rn activities in February 2009 and April 2010 (highest ²²²Rn activities in these sampling periods was at Pearces Lane). The distribution of ²²²Rn activities in the detailed sampling campaign in February 2015 is similar to that at other periods of low to moderate streamflow (e.g. March 2014). The lowest overall ²²²Rn activities were recorded during the two highest flow sampling campaigns (September 2010 and July 2014).

CI concentrations generally increase downstream from 4 to 10 mg L⁻¹ at Browns to as high as 98 mg L⁻¹ at Chinns Bridge (Fig. 4b). Cl concentrations in March 2014 were generally higher (up to 98 mg L⁻¹) than in the other sampling campaigns, while CI concentrations during the highest streamflows in September 2010 are < 20 mg L⁻¹. A major increase in CI concentrations occurs downstream of Smyths Road in the reaches over which ²²²Rn activities are highest at low streamflow. The concentrations of other major ions (e.g., Na) increase downstream in a similar manner (Table S1).

Groundwater geochemistry

Groundwater from the near-river bores at Pearces Lane and Stratford has ²²²Rn activities that vary from 480 to 28 980 Bg m⁻³ (Table S2). There is some variation in ²²²Rn activities in individual bores between the sampling rounds with relative standard deviations varying from 6 to 34 %. The mean value of all groundwater 222 Rn activities (n = 26) is 12 890 Bqm $^{-3}$. Bore 5 at Pearces Lane is immediately adjacent to the Avon River and possibly samples water from the parafluvial zone rather than groundwater. Excluding data from that bore, the mean value of 222 Rn activities is 13 830 Bqm $^{-3}$ (n = 24) with a standard error of 1273 Bqm $^{-3}$ and a 95 % confidence interval (calculated using the Descriptive Statistics tool in Excel 2010 which assumes that the data follows a t-distribution) of 2634 Bqm $^{-3}$. EC values of groundwater from the bores at Pearces Land and Stratford are between 100 and $680 \,\mu\text{S cm}^{-1}$ and Cl concentrations range from 46 to 147 mg L $^{-1}$ with a mean value of $79 \pm 34 \,\text{mg} \,\text{L}^{-1}$ (n = 16) (Table S2). If Bore 5 at Pearces Lane is again excluded the mean Cl concentration is $87 \pm 28 \,\text{mg} \,\text{L}^{-1}$ (n = 14) with a standard error of 8 mg L $^{-1}$ and a 95 % confidence interval of 16 mg L $^{-1}$. These Cl concentrations are typical of groundwater in elsewhere in the Avon and neighbouring catchments (Department of Environment and Primary Industries, 2015).

4.4 ²²²Rn emanation rates

Emanation rates were determined via Eq. (7) from 222 Rn activities of extracted pore water that had equilibrated with the sediments. The matrix density was assigned as 2700 kg m $^{-3}$, which is appropriate for sediments rich in quartz (ρ = 2650 kg m $^{-3}$), and a porosity of 0.4 was used, which is appropriate for unconsolidated poorly-sorted riverine sediments (Freeze and Cherry, 1979). γ values range from 288 to 4950 Bq m $^{-3}$ with a mean value of 2308 ± 1197 Bq m $^{-3}$ (n = 44) and a standard error of 183 Bq m $^{-3}$. 222 Rn activities of water that is in equilibrium with the sediments are given by γ/λ , and the mean γ/λ values are 12751 ± 6615 Bq m $^{-3}$ with a standard error of 1009 Bq m $^{-3}$. These γ/λ values are not significantly different (ρ ~ 0.5) to the measured 222 Rn activities in groundwater. The mean emanation rates for sediments from the different sites vary between 1484 and 3461 Bq m $^{-3}$; however there is no systematic variation with position in the catchment.

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Water was extracted from the alluvial gravels at a number of locations along the Avon River during low flow periods either from open holes or from piezometers driven 1-2 m below the surface of the gravels (in total 52 samples were collected). EC values of water within the gravels further than 1 to 2 m from the edge of river edge are in the range 120 to 550 µS cm⁻¹ (Fig. 5b); these EC values are higher than those of the adjacent river water but similar to those of the groundwater. Only water extracted from within 1 to 2 m from the river had EC values similar to the river and in some cases the EC of water from the gravels within a few centimetres of the river edge was higher than the adjacent river. ²²²Rn activities measured in 21 of these samples were between 7000 and 28 000 Bg m⁻³ (Fig. 5a), which are also significantly higher than the ²²²Rn activities in the adjacent river.

Discussion

The following observations imply that overall the Avon is a gaining river: (1) even during periods of low rainfall the river continues to flow and streamflow commonly increases between The Channel and Chinns Bridge, (2) ²²²Rn activities are higher than those that could be maintained by hyporheic exchange alone (Cartwright et al., 2011; Cook, 2013), (3) CI concentrations increase downstream; and (4) there are seeps of water (presumed to be groundwater) at the base of steep slopes at the edge of the floodplain. In the following section the ²²²Rn activities and CI concentrations will be used to assess the location and magnitude of groundwater inflows.

Distribution of groundwater inflows

The region between Smyths Road and Ridleys Lane where ²²²Rn activities increase and remain high (Fig. 4a), especially at lower streamflows, and where there is a marked increase in CI concentrations (Fig. 4b) is interpreted as receiving major groundwater

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inflows. This section of the Avon River is incised up to 4 m below the floodplain which likely produces steep hydraulic head gradients that will result in groundwater discharge on the floodplain and into the river. There are also groundwater seeps and patches of perennial wetland vegetation at the edge of the floodplain in this area. In February 2009 and April 2010, the highest ²²²Rn activities were recorded at Pearces Lane, while in March 2014 and February 2015 the highest ²²²Rn activities were recorded at Bushy Park (Fig. 4a). The major floods between 2011 and 2013 (Fig. 3a) changed the location of pools and sediment banks on the Avon River probably also changed the locations of groundwater inflow. The reaches between Browns and Wombat Flat and Stewarts Lane and Stratford are also characterised by increasing ²²²Rn activities and are again interpreted as recording groundwater inflows.

The reaches between Wombat Flat and Smyths Road, Ridleys Lane and Stewarts Lane, and Knobs Reserve and Chinns Bridge show gradual decline in ²²²Rn activities and little change in Cl concentrations (Fig. 4); these are interpreted as either losing or receiving minor groundwater inflows. In these areas, the landscape is flatter and the river less incised which likely results in lower hydraulic gradients and consequently less groundwater discharge to the river.

5.2 Quantifying groundwater inflows

Groundwater inflows were calculated from the 222 Rn activities for February 2015 (Table S1) by solving Eq. (1) using a finite difference approach in a spreadsheet with a spatial discretisation of 10 m; comparisons for subsets of the data indicate that finer-scale discretisation does not significantly change the results. The streamflow at The Channel gauge was used as the initial streamflow and Q was increased after each distance step via Eq. (2). The calculations specified all parameters except I and matched the observed I activity at the sampling sites by varying I in each reach (Fig. 6a).

For the initial set of calculations, the groundwater ²²²Rn activity was assumed to be 13 000 Bq m⁻³, which is consistent both with the measured ²²²Rn activities in the bores and the calculated ²²²Rn activities of water in equilibrium with the alluvial sediments.

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Average evaporation rates in southeast Australia in February to April are 3 x 10⁻³ to 5×10^{-3} m day⁻¹ (Bureau of Meteorology, 2015) and a value of 4×10^{-3} m day⁻¹ was adopted. Average river width and depth is 10 and 0.5 m, respectively, upstream of Wombat Flat and 20 and 1 m, respectively, for the rest of the river. The gas transfer coefficient was estimated as 0.3 day⁻¹ from the decline in ²²²Rn activities between Ridleys Lane and Schools Road (Fig. 4a). Assuming that this is a losing stretch of the river, which is consistent with the lack of increase in CI concentrations (Fig. 4b), the decrease in ²²²Rn activities will be mainly due to degassing with a small contribution from decay. A value of 0.3 day⁻¹ is at the lower end of estimates of Rn gas transfer coefficients (Genereux and Hemond, 1992; Cook et al., 2003, 2006; Cartwright et al., 2011, 2014b; Atkinson et al., 2013; Unland et al., 2013; Yu et al., 2014). However, as the Avon River is dominated by slow-flowing pools, degassing rates are expected to be low.

As there are few processes that increase the EC of water in the hyporheic or parafluvial zones, the water in the river gravels within 1 to 2 m of the river that has high EC values (Fig. 5b) represents groundwater or a mixture of river water and groundwater. Consequently, the width of the hyporheic zone has been assigned as the river width. The thickness of the hyporheic zone is less well known; however, by analogy with rivers elsewhere, it is likely to be a few centimetres thick (Boulton et al., 1998; Hester and Dovle, 2008; Tonina and Buffington, 2011) and a value of 10 cm is initially adopted. The flux of 222 Rn from the hyporheic zone was estimated from Eq. (3) using the mean γ value of 2300 Bq m⁻³ day⁻¹ (Table 2), a porosity of 0.4 (which is appropriate for coarsegrained unconsolidated sediments), and a value for $c_{\rm in}$ that is the ²²²Rn activity of the river in that reach. The residence time of water within the parafluvial zone is likely to be short (Boulton et al., 1998; Tonina and Buffington, 2011; Zarnetske et al., 2011; Cartwright et al., 2014b), and $t_h = 0.1$ days is assumed here; for $t_h < 1$ day, F_h is relatively insensitive to the actual residence times in the hyporheic zone (Lamontagne and Cook, 2007; Cartwright et al., 2014b)

Using these parameters it is possible to model the observed ²²²Rn activities in the Avon River in February 2015 (Fig. 6a). Calculated groundwater inflows are up to 2.7 m³ m⁻¹ day⁻¹ in the reaches between Smyths Road and Pearces Lane (Fig. 6b), which is the region where Cl concentrations also increase markedly (Fig. 4b). Most reaches of the river between Ridleys Lane and Stewarts Lane and downstream of Knobs Reserve have negligible groundwater inflows or are losing (Fig. 6b). The change in Cl concentrations (Fig. 6d) was calculated from the groundwater inflows assuming that groundwater has a Cl concentration of 85 mg L⁻¹. The calculated Cl concentrations are higher than observed, although the calculations do predict the marked increase in Cl concentrations between Smyths Road and Ridleys Lane. Given the uncertainty in groundwater Cl concentrations, the discrepancy between observed and predicted Cl concentrations is not large. If the Cl concentration of the groundwater is allowed to vary by the 95 % confidence interval (±16 mg L⁻¹) the observed trend can be reproduced (Fig. 6d).

The calculated net groundwater inflow of 28 300 m³ day⁻¹, however, exceeds the measured increase in streamflow between The Channel and Chinns Bridge of 15 500 m³ day⁻¹ by 180 % (Fig. 6c). Much of the mismatch between observed and calculated streamflow results from the high estimates of groundwater inflows upstream of Stratford (Fig. 6b). The net increase in streamflow between The Channel and Stratford in February 2015 is 10 500 m³ day⁻¹, whereas the predicted net groundwater inflow from the ²²²²Rn mass balance was 26 700 m³ day⁻¹. While the February 2015 sampling round represents the end of summer when the small ephemeral tributaries were dry and there was no overland flow, there were still flows from Valencia Creek and Freestone Creek of 1410 and 200 m³ day⁻¹, respectively. If these were included, the mismatch between calculated and observed streamflow increases.

Although constrained by fewer data points, the predicted distribution of groundwater inflows in the other low to moderate flow sampling campaigns calculated using the same parameters as for February 2015 are similar (Fig. 7a). In February 2009, April 2010, and March 2014, groundwater inflows are up to 3.1 m³ m⁻¹ day⁻¹ and the

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calculated net groundwater inflows are 21 700, 15 900, and 21 300 m³ day⁻¹, respectively (Fig. 7b). These calculated groundwater inflows are up to 490 % of the measured increases in streamflow between The Channel and Chinns Bridge. Groundwater inflows were also calculated for the higher streamflow sampling rounds (September 2010 and July 2014) using widths of 15 m upstream of Wombat Flat and 25 m elsewhere, depths of 1.25 upstream of Wombat Flat and 1.6 m elsewhere, $k = 0.3 \,\mathrm{day}^{-1}$, and $F_{\rm h}$ adjusted for the higher river widths. The estimated groundwater inflows are lower (up to 1.3 m³ m⁻¹ day⁻¹) than at the periods of low streamflow and the net groundwater discharges of 32 100 m³ day⁻¹ in September 2010 and 44 600 m³ day⁻¹ in July 2014 are lower than the measured increases in streamflow between The Channel and Stratford or Chinns Bridge (Fig. 7b).

5.3 Uncertainties and variability

The impacts of uncertainties need in the parameters in Eq. (1) on the calculated groundwater inflows need to be considered. This will be done in reference to the February 2015 sampling round but the same considerations apply to the other sampling rounds. The evaporation term in Eq. (1) is one to two orders of magnitude lower than most of the other terms and errors in the assumed evaporation rate have little influence on the calculations (Cook, 2013). The main factor impacting calculated groundwater inflows in reaches where groundwater inflows are high is the assumed ²²²Rn activity of groundwater (Cartwright et al., 2011; Cook, 2013). Allowing c_{qw} to vary by ±2600 Bg m⁻³, which represents the 95% confidence interval of the groundwater ²²²Rn activities, makes little difference to the discrepancy between the calculated and observed increase in streamflow (Fig. 6c). Increasing c_{ow} to $\sim 27\,000\,\text{Bg}\,\text{m}^{-3}$ would reproduces the observed increase in streamflow along the Avon (Fig. 8). However, there is no known groundwater in the Avon catchment with such high 222Rn activities, and an activity of 27 000 Bq m⁻³ is far higher than would be in equilibrium with the alluvial

In common with most studies, the calculations assumed that the groundwater inflows are uniform along a particular reach. However, because ²²²Rn is lost from rivers by 5 degassing and decay, lower groundwater inflows are required to replicate the observed ²²²Rn activities if the groundwater inflows are predominantly just upstream of each sampling point (Cook, 2013). However, even assigning all groundwater inflows in an individual reach to the 10 m section upstream of the measurement point still results in the calculated groundwater inflows that slightly overestimate the measured streamflow in the Avon River (Fig. 6c). The trends in ²²²Rn activities in the river in this case are also not realistic (Fig. 6a).

There is uncertainty in the gas transfer coefficient. The value of k was estimated assuming that the Avon River contains losing reaches; if those reaches were actually gaining then this methodology underestimates k. However, increasing k from 0.3 day produces higher groundwater inflows, which increases the mismatch between the observed and calculated increases in streamflow. k may also be estimated from river velocities and depths. The O'Connor and Dobbins (1958) and Negulescu and Rojanski (1969) gas transfer equations as modified for ²²²Rn are:

$$k = 9.301 \times 10^{-3} \left(\frac{v^{0.5}}{d^{1.5}} \right)$$

$$k = 4.87 \times 10^{-4} \left(\frac{v}{d} \right)^{0.85}$$
(8)

$$k = 4.87 \times 10^{-4} \left(\frac{V}{d}\right)^{0.85} \tag{9}$$

(Mullinger et al., 2007), where ν is velocity (m day⁻¹). k estimated from Eqs. (8) and (9) ranges between 0.1 and 0.3 day⁻¹. Using a value of k of 0.1 day⁻¹ produces net groundwater inflows that more closely match the observed increase in streamflow between The Channel and Chinns Bridge (Fig. 8). However, adopting $k = 0.1 \,\text{day}^{-1}$ results in the calculated ²²²Rn activities in a number of reaches being overestimated (Fig. 9). This is because even assuming no groundwater inflows into these reaches,

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the loss of ²²²Rn due to degassing is insufficient to explain the observed decrease in ²²²Rn. Such a poor correspondence between predicted and observed ²²²Rn activities implies problems with the adopted variables.

Increasing F_h will also reduce calculated groundwater inflows. Using the same ema-5 nation rates, residence times, and porosities but assigning a thickness of the hyporheic zone of 50 cm, increases F_h and produces groundwater inflows that broadly match the increase in streamflow between The Channel and Chinns Bridge (Fig. 8). However, the higher values of F_h again result in the calculated ²²²Rn activities in many of the reaches being higher than observed (Fig. 9). As with the lower k values, the poor fit between predicted and observed ²²²Rn activities implies problems with the choice of variables.

Because the error in λ is insignificant and the evaporation term is much smaller than the other terms, it is generally possible to produce observed trends in ²²²Rn activities with different combinations of k and F_h (Cartwright et al., 2014b). If F_h is calculated assuming a 50 cm thick hyporheic zone, adopting a k value of 0.6 day⁻¹ reproduces the observed 222 Rn activities. Similarly, if a k value of 0.1 day $^{-1}$ was used, a match between the observed and the predicted ²²²Rn activities is achieved by ignoring hyporheic exchange. However, these combinations of parameters again result in estimated net groundwater inflows that exceeding the measured increase in streamflow (Fig. 8).

There is likely to be some error in river gauging, but it is unlikely to be sufficient to explain the gross overestimation of groundwater inflows. Uncertainties in the assumed river widths and depths will also impact the calculations. Specifically, reducing the width or depth decreases the magnitude of the last two terms on the right-handside of Eq. (1), which in turn reduces /. If widths were reduced by 50 % (an unrealistic error), net groundwater inflows broadly match the increase in streamflow (Fig. 8). However, this again results in ²²²Rn activities being overestimated in many reaches (Fig. 9). Increasing k to $0.65 \,\mathrm{day}^{-1}$ would allow the ²²²Rn activities to be predicted using these lower widths but again results in the estimated net groundwater inflow exceeding the measured increase in streamflow (Fig. 8).

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5.4 Incorporating larger-scale parafluvial flow

It is clear that it is difficult to find combinations of parameters that allow the observed variation in ²²²Rn activities to be replicated without the calculated net groundwater inflows exceeding the measured increase in streamflow. The proposed resolution is that there is large-scale (100's of m to km) parafluvial flow through the alluvial sediments on the Avon River floodplain that contributes to the river in the gaining reaches. This flow represents water that exfiltrates from and then reinfiltrates into the river which is likely hosted mainly within the coarse-grained alluvial sediments (although conceivably it could also include water that flows through the upper levels of the aquifers underlying the alluvial sediments). By contrast with hyporheic flow which contributes ²²²Rn to the river along all reaches (whether gaining or losing), the inflows from the parafluvial zone will only contribute ²²²Rn where the river is gaining.

If residence times of water in the parafluvial zone are sufficiently long, then the inflowing parafluvial water will have high $^{222}{\rm Rn}$ activities and increase the $^{222}{\rm Rn}$ activities in the river. However, because it represents exfiltrated river water, the inflows from the parafluvial zone do not add to the overall streamflow. Figure 10 shows the results of assuming that 50 % of the water inflowing in each reach in February 2015 represents parafluvial zone waters that has a residence time in the parafluvial zone sufficiently long for secular equilibrium to be attained. The maximum value of $I_{\rm p}$ is 1.26 m $^{\rm 3}$ m $^{\rm -1}$ day $^{\rm -1}$ (Fig. 10b) in the Valencia to Bushy Park reaches where $c_{\rm r}$ is 2500 to 2900 Bq m $^{\rm -3}$. If the parafluvial zone water is in secular equilibrium with the sediments, $c_{\rm p}\sim$ 12 700 Bq m $^{\rm -3}$ (Table 2), $c_{\rm p}-c_{\rm r}\sim$ 10 000 Bq m $^{\rm -3}$, and $F_{\rm p}\sim$ 12 600 Bq m $^{\rm -1}$ day $^{\rm -1}$ (Eq. 5). For $t_{\rm p}$ in excess of \sim 30 days the system is close to secular equilibrium and $c_{\rm p}$ and $I_{\rm p}$ are near constant and independent of $t_{\rm p}$ (Fig. 1). If residence times in the parafluvial zone are shorter than those required to attain secular equilibrium, $c_{\rm p}$ will be lower and the inflows from the parafluvial zone ($I_{\rm p}$) required to produce a given flux of $^{\rm 222}{\rm Rn}$ ($F_{\rm p}$) increases (Fig. 1). For example, if $\gamma=2300$ Bq m $^{\rm -3}$ day $^{\rm -1}$, $c_{\rm r}=2700$ Bq m $^{\rm -3}$, and $t_{\rm p}=0.1$ days,

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 $c_{\rm p}$ = 2718 Bqm⁻³ and if $F_{\rm p}$ = 12600 Bqm⁻¹ day⁻¹, $I_{\rm p}$ ~ 70 m³ m⁻¹ day⁻¹. The cross-sectional area of the parafluvial zone $A_{\rm p}$ required to accommodate these parafluvial flows with ϕ = 0.4 and $t_{\rm p}$ between 0.1 and 100 days is between 17 and 350 m² (Eq. 6). The floodplain of the Avon River is tens of metres wide with sediment thicknesses of several metres and even the higher estimates of the cross-sectional area are not unreasonable. For the lower estimates of $A_{\rm p}$, the flow could be accommodated in a zone < 1 m thick beneath the river.

The proposal that inflows of parafluvial zone waters augment groundwater inflows is justifiable hydrologically as the conditions required for high groundwater inflows (gaining river with steep hydraulic gradients and high-hydraulic conductivity sediments) will likely drive the return of parafluvial waters to the river. By contrast losing reaches are likely to be where the water enters the parafluvial sediments. The riffle sections of the Avon River commonly have steep longitudinal gradients and may transition from losing at their upstream end to gaining at the downstream end. Additionally, there may be parafluvial flow through the numerous sediment banks and point bars.

By allowing the parafluvial input to vary between reaches, both the variations in ²²²Rn activities (Fig. 10a) and calculated increases in streamflow are predicted (Fig. 10c). There is no process in the parafluvial zone that increases the CI concentrations of the through-flowing water. Thus the CI concentrations in the river reflect only the groundwater inflows. The calculated CI concentrations are again higher than observed (Fig. 10d) but not substantially when the uncertainty in groundwater CI concentrations is taken into account.

While the model of parafluvial zone flow explains the geochemical observations and is consistent with the hydrogeology of the Avon River, it is difficult to test. The inflows from the parafluvial zone have been assumed to vary proportionally to the groundwater inflows, which may not necessarily be the case. Nevertheless, this conceptualisation utilises a process that is known to occur in river valleys to explain observations that are otherwise difficult to resolve. An alternative explanation for the discrepancy between the calculated inflows and the increase in streamflow is that significant pumping of water

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from the river occurred. While the surface water is licenced for use, streamflow during February 2009 and March 2014 was below the minimum levels where that is permitted and the streamflows in April 2010 and February 2015 were such that use would be restricted; hence, large-scale pumping of river water at those times is unlikely.

Similar parafluvial flow is likely to occur under higher flow conditions. Thus, while the calculated groundwater inflows during the higher streamflow periods are plausible in as much as they are less than the observed increase in streamflow, if significant parafluvial flow occurs at those times they too will overestimate the actual groundwater inflows.

6 Conclusions

While this study has illustrated some of the difficulties in using geochemical tracers to understand groundwater inflows to rivers, it also shows that additional information may be gleaned from such tracers. The patterns of variation of ²²²Rn activities and Cl concentrations clearly define the reaches of the Avon River that are gaining. Differences in the location of the zones of high groundwater inflows between the 2009 to 2010 and 2014 to 2015 sampling rounds reflect changes to the river and its floodplain following major floods and demonstrate the utility of ²²²Rn in determining changes to the location of groundwater inflows in dynamic environments. The difficulty in calculating groundwater inflows using ²²²Rn activities indicates that there were processes that were not initially correctly accounted for; in this case it was concluded that significant large-scale parafluvial flow occurs. Parafluvial flow is likely to be important in rivers with coarse-grained alluvial sediments on their floodplains, especially where there are alternating gaining and losing reaches, and must be taken into account in ²²²Rn mass balance calculations. Unlike hyporheic exchange, which occurs in all stretches, parafluvial inflows are likely to dominantly occur in gaining reaches augmenting the groundwater inflows.

Theoretically, a conservative tracer such as CI that is unaffected by parafluvial flow could be used to determine the groundwater inflows. However, the relatively high vari-

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ability of groundwater CI concentrations and the relative small difference between groundwater and river CI concentrations reduce its utility in quantifying groundwater inflows in this instance. Nevertheless using multiple tracers is potentially valuable and may indicate the importance of other water stores that can feed rivers such as bank return flows or parafluvial flow (c.f. Cook et al., 2003; Mullinger et al., 2007; Cartwright et al., 2011, 2014b; Unland et al., 2013).

The Avon study also illustrates the importance of carrying out geochemical studies at baseflow conditions where the majority of inflows into the river are likely to be from groundwater. While this might appear redundant in terms of determining the water balance, it does provide for a test of assumptions and parameterisation. In the Avon River, the calculated groundwater inflows for the higher flow sampling rounds (Fig. 7) are plausible in as much as they are lower than the net increases in streamflow; however, the results from the low flow sampling rounds make it likely that the ²²²Rn activities reflect both groundwater inflows and parafluvial exchange. Without the data from the baseflow sampling rounds this would not have been evident and possible erroneous estimated of groundwater inflows may have been made.

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Table 1. Summary of parameters used in calculations.

Symbol	Parameter	Units	Comments
Q	Streamflow	m ³ day ⁻¹	
E	Evaporation	m day ⁻¹	
X	Distance downstream	m	
W	Stream width	m	
d	Stream depth	m	
V	Strem velocity		
$c_{\text{gw}}, c_{\text{r}}, c_{\text{h}}, c_{\text{p}}$	²²² Rn activities in groundwater, river, hyporheic zone, parafluvial zone	$Bq m^{-3}$	
k	Degassing constant	day ⁻¹	
λ	Decay constant	0.181 day ⁻¹	
1	Groundwater inflows	$m^3 m^{-1} day^{-1}$	Eq. (1)
F_{h}	²²² Rn flux from hyporheic zone	$Bq m^{-1} day^{-1}$	Eq. (4)
$F_{\rm p}$	²²² Rn flux from parafluvial zone	$Bq m^{-1} day^{-1}$	Eq. (5)
γ	²²² Rn emanation rate	$Bq m^{-3} day^{-1}$	Eq. (7)
F _n F _p γ E _m	²²² Rn produced from sediments	Bq kg ⁻¹	
$ ho_{s}$	Sediment density	kg m ⁻³	
	Inflows from parafluvial zone	$m^3 m^{-1} day^{-1}$	
$t_{\rm p}$ $t_{\rm h}, t_{\rm p}$ ϕ	Residence time in hyporheic or parafluvial zone	day	
$\ddot{\phi}$	porosity		
$V_{\rm p}$	Volume of sediments that parafluvial inflows interact with	${\rm m}^{3}{\rm m}^{-1}$	
$A_{\rm h}$, $A_{\rm p}$	Cross-sectional area of the hyporheic or parafluvial zone	m^2	$A_{\rm p} = V_{\rm p}$

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Table 2. ²²²Rn emanation rates from floodplain sediments.

Site ^a /Sample	E _m Bq kg ⁻¹	γ Bg m ⁻³ day ⁻¹	γ/λ Bq m ⁻³
Chinns Bridge 1	2.01	1473	8138
Chinns Bridge 2	4.02	2949	16293
Wombat Flat 1	4.04	2964	16376
Wombat Flat 2	4.52	3311	18 295
Wombat Flat 3	4.19	3075	16 988
Wombat Flat 4	6.13	4492	24819
Valencia 1	3.95	2899	16016
Valencia 2	1.86	1362	7525
Pearces Lane 1	0.62	454	2506
Pearces Lane 2 Pearces Lane 3	3.25 1.41	2383 1034	13 167 5722
Pearces Lane 3 Pearces Lane 4	2.63	1925	10 636
	2.63 6.76		27360
Pearces Lane 5 Pearces Lane 6	5.60	4952 4107	22 689
Pearces Lane 7	4.12	3018	16 674
Pearces Lane 8	1.54	1127	6225
Stewarts Lane 1	3.41	2497	13 797
Stewarts Lane 2	5.78	4239	23 4 1 8
Stewarts Lane 3	3.08	2258	12 475
Stewarts Lane 4	2.88	2110	11656
Stewarts Lane 5	4.63	3391	18732
Stewarts Lane 6	3.64	2669	14745
Stewarts Lane 7	4.52	3311	18 294
Stewarts Lane 8	4.58	3354	18 530
Stewarts Lane 9	1.96	1434	7925
Stewarts Lane 10	5.09	3733	20 622
Stewarts Lane 11	4.25	3119	17 230
Stewarts Lane 12	3.68	2699	14910
Stewarts Lane 13	1.77	1294	7150
Stewarts Lane 14	2.89	2122	11723
Stratford 1 Stratford 2	2.13 0.66	1563 482	8634
Stratford 3	3.01	2206	2663 12 190
Stratford 4	3.01	2762	15 259
Stratford 5	0.39	2762	15259
Stratford 6	1.24	911	5032
Stratford 7	2.00	1469	8117
Stratford 8	2.71	1985	10 965
Stratford 9	0.91	668	3692
Stratford 10	1.01	738	4077
Stratford 11	4.55	3334	18419
Stratford 12	3.13	2293	12 667
Stratford 13	0.81	491	3282
Mean		2308	12751
σ		1197	6615

^a Sites on Fig. 2.

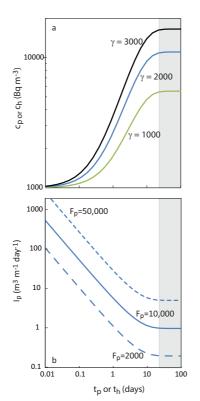


Figure 1. (a) Variation in the ²²²Rn activity in the parafluvial or hyporheic zone ($c_{\rm p}$ or $c_{\rm h}$) with residence time ($t_{\rm p}$ or $t_{\rm h}$) and ²²²Rn emanation rate (γ) (Eq. 3). **(b)** Variation in the water flux from the parafluvial zone ($I_{\rm p}$) with the flux of ²²²Rn from the parafluvial zone ($F_{\rm p}$) and $F_{\rm p}$ 0 (Eq. 5). In both cases $F_{\rm in}$ 1 = 1000 Bq m⁻³.

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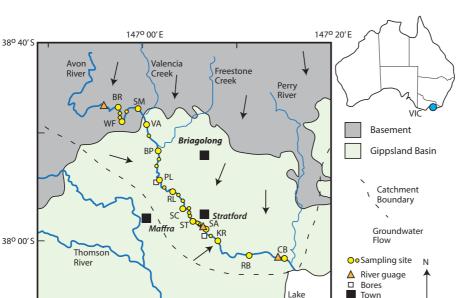
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Figure 2. Summary geological and hydrogeological map of the Avon River catchment (Hofmann and Cartwright, 2015; Department of Environment and Primary Industries, 2015). Main sampling sites are BP = Bushy Park, BR = Browns, CB = Chinns Bridge, PL = Pearces Lane, RL = Ridleys Lane, RB = Redbank, SA = Stratford, SC = Schools Lane, ST = Stewarts Lane, SM = Smyths Road, VA = Valencia, WF = Wombat Flat. Unnamed sampling sites are the additional sites from February 2015 (Table S1).

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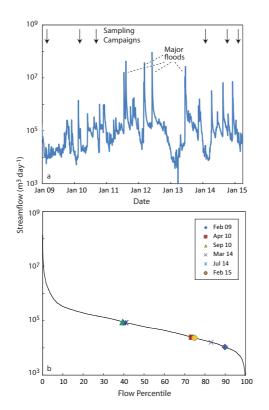


Figure 3. (a) Variation in streamflow at Stratford (Fig. 1) between January 2009 and March 2015. The major floods (highlighted) caused significant changes to the geometry of the floodplain. (b) Flow frequency curve for Stratford for streamflows between January 2000 and March 2015 and the percentiles of discharge in the sampling campaigns. Data from Department of Environment and Primary Industries (2015).

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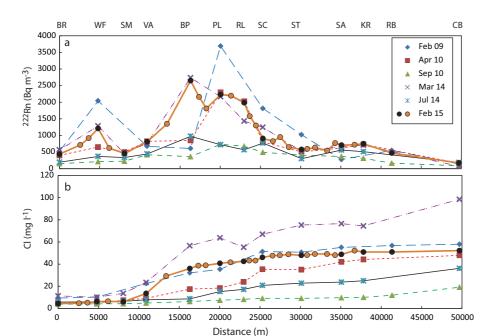


Figure 4. Downstream variations in ²²²Rn activities **(a)** and Cl concentrations **(b)** for the six sampling campaigns (Data from Table S1, abbreviations as for Fig. 2). Closed symbols for February 2015 are from the main sites, open symbols are from the additional sites specific to that sampling campaign (Table S1). Site abbreviations as for Fig. 2.

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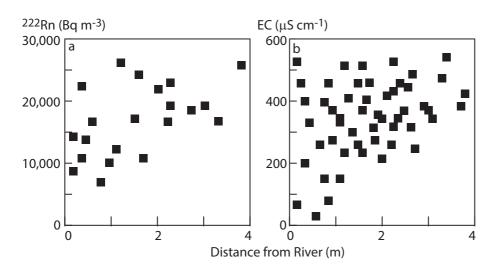


Figure 5. (a) Variations in ²²²Rn activities **(a)** and EC values **(b)** of water extracted from river bank gravels.

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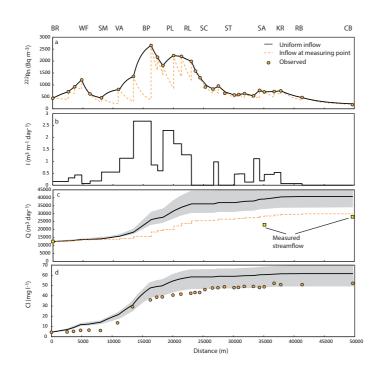


Figure 6. (a) Calculated vs. observed ²²²Rn activities in the Avon River for February 2015 assuming both uniform groundwater inflow within each reach and the situation where groundwater inflow occurs immediately upstream of the measurement point. Site abbreviations as for Fig. 2. (b) Calculated groundwater inflows (/) assuming uniform inflows within each reach. (c) Calculated increase in streamflow from groundwater inflows (Eq. 2). Both uniform groundwater inflow within each reach and the situation where groundwater enters the river immediately upstream of the measurement point overestimate the measured streamflow. Shaded area is the range of streamflow resulting from varying c_{aw} within the 95% confidence interval. (d) Predicted vs. observed CI concentrations. Shaded field is the range resulting from varying groundwater CI concentrations within the 95 % confidence interval.

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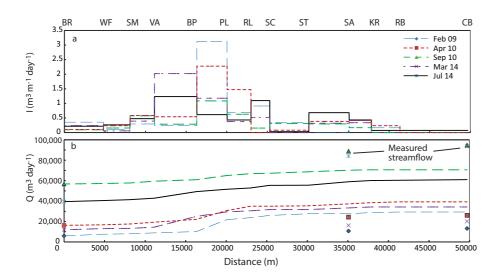


Figure 7. Calculated groundwater inflows (a) and increases in streamflow due to groundwater inflows (b) for the sampling rounds excluding February 2015. Aside from the high flow periods (September 2010 and July 2014) the calculated increase in streamflow exceeds the observed streamflow at Stratford and Chinns Bridge. Site abbreviations as for Fig. 2.



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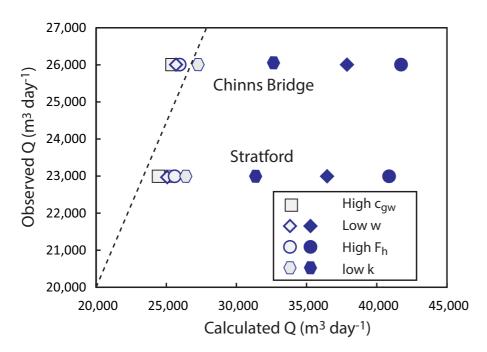


Figure 8. Calculated vs. observed streamflow at Stratford and Chinns Bridge for February 2015 resulting from varying parameters in Eq. (1). High c_{aw} is for a groundwater $^{222}\mathrm{Rn}$ activity of 27000 Bg m⁻³; low k is where k is reduced to 0.1 day⁻¹, high F_h increase the thickness of the parafluvial zone to 0.5 m, and low w reduces the width of the Avon River by 50 %. Open symbols represent the result of these changes in isolation (see also Fig. 9); closed symbols represent the results of varying other parameters so that the predicted and observed 222Rn activities match.

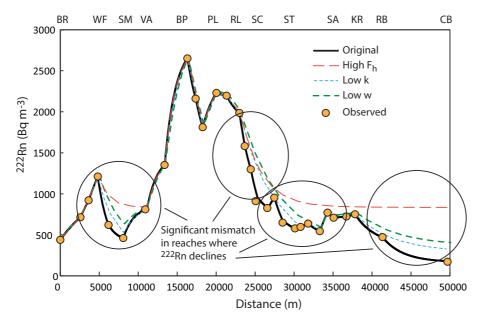


Figure 9. Calculated and observed 222 Rn activities for February 2015 resulting from varying individual parameters in Eq. (1) in isolation. Original is the predicted variation in 222 Rn activities from Fig. 5, low k is where k is reduced to $0.1 \, \mathrm{day}^{-1}$, high F_{h} increase the thickness of the parafluvial zone to $0.5 \, \mathrm{m}$, and low w reduces the width of the Avon River by 50 %. In all cases the new parameters result in significant overestimation of 222 Rn activities in many reaches and are unlikely to be realistic. Site abbreviations as for Fig. 2.

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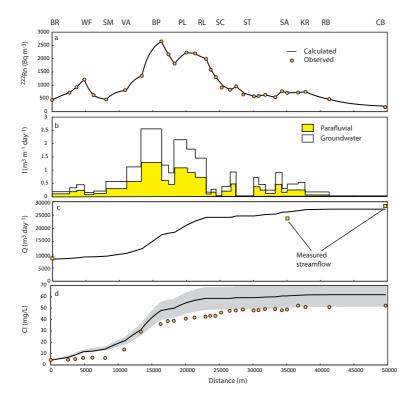


Figure 10. (a) Calculated and observed ²²²Rn activities for February 2015 resulting from assigning 50% of the calculated inflows as parafluvial flow. **(b)** Variation in groundwater and parafluvial inflows. **(c)** Calculated streamflow resulting from the groundwater inflows (Eq. 2) vs. measured streamflow at Stratford and Chinns Bridge. **(d)** Predicted vs. observed Cl concentrations. Shaded field is the range resulting from varying groundwater Cl concentrations within the 95% confidence interval.

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