- 1 Using Radon to understand parafluvial flows and the changing locations of groundwater
- 2 inflows in the Avon River, SE Australia

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Abstract

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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. This study utilises ²²²Rn activities and Cl concentrations in the Avon River, southeast Australia to determine the distributions of groundwater inflows and to understand the importance of parafluvial flow on the ²²²Rn budget. The distribution of ²²²Rn activities and Cl concentrations implies that the Avon River contains alternating gaining and losing reaches. The location of groundwater inflows changed as a result of major floods in 2011 to 2013 that caused significant movement of the floodplain sediments. The floodplain of the Avon River comprises unconsolidated coarse-grained sediments with numerous point bars and sediment banks through which significant parafluvial flow is likely. The ²²²Rn activities in the Avon River, which are locally up to 3690 Bq m⁻³, result from a combination of groundwater inflows and the input of water from the parafluvial zone that has high ²²²Rn activities due to the ²²²Rn emanations from the alluvial sediments. If the high ²²²Rn activities were ascribed solely to groundwater inflows, the calculated net groundwater inflows exceed the measured increase in streamflow along the river by up to 490% at low streamflows. Uncertainties in the ²²²Rn activities of groundwater, the gas transfer coefficient, and the degree of hyporheic exchange cannot explain a discrepancy of this magnitude. The proposed model of parafluvial flow envisages that water enters the alluvial in reaches where the river is losing and subsequently re-enters the river in the gaining reaches with flow paths of tens to hundreds of metres. 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

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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 riverine 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 also result in the double allocation of water resources (i.e., 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 assessing the potential impacts of climate or landuse changes on river systems. In many catchments globally there are insufficient groundwater bores to understand the exchange between rivers and groundwater on anything other than a regional scale. In these cases geochemical tracers provide an alternative tool to understand groundwater-river interaction. Providing that groundwater and surface water have significantly different geochemistry, changes in the geochemistry of the river may be used to map the distribution of and quantify groundwater inflows (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, 2014; Cook, 2013; Bourke et al., 2014a,b). Geochemical tracers only quantify groundwater inflows, and while they are commonly used to determine the distribution of gaining and losing reaches, they do not quantify the magnitude of any groundwater outflows. River water also interacts with the sediments beneath and adjacent to the streams in the hyporheic and parafluvial zones. The hyporheic zone comprises the sediments of the stream bed and sides

through which the river water flows due to irregularities in the stream bed, and hyporheic flow generally occurs on the centimetre to tens of centimetre scale (Boulton et al., 1998). In rivers that have coarse-grained unconsolidated sediments on their floodplain, metre to hundreds of metre scale parafluvial flow may also occur (Holmes et al., 1994; Edwardson et al., 2003; Cartwright et al., 2014; Bourke et al., 2014a; Briody et al., 2016). By contrast with hyporheic exchange that occurs along the entire river, water enters the parafluvial zone in river reaches that are losing and then renters the river where it is gaining, augmenting the groundwater inflows. Both hyporheic exchange and parafluvial flow may impact the geochemistry of the rivers (Boulton et al., 1998; Edwardson et al., 2003; Cook et al., 2006; Cartwright et al., 2014; Bourke et al., 2014a; Briody et al., 2016) and must be taken into account when using geochemical tracers to determine groundwater inflows to rivers.

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 (Cecil and Green, 2000). Because ²²⁶Ra activities in minerals in the aquifer matrix are several orders of magnitude higher than those 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 viable tracer of groundwater inflows in catchments where the groundwater has similar major ion concentrations and/or 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 where the inflows are not quantified.

The successful application of ²²²Rn to determine groundwater inflows, however, requires careful consideration of several processes and uncertainties. ²²²Rn activities in groundwater may be spatially

or temporally heterogeneous (Cook et al., 2006; Mullinger et al., 2007; Unland et al., 2013; Yu et al.,

2013; Cartwright et al., 2011; Atkinson et al., 2015). Additionally, while it is well established that the rate of ²²²Rn degassing increases with increasing river turbulence and decreasing river depth, it is difficult to reliably quantify the rate of degassing (Genereux and Hemond, 1992; Mullinger et al., 2007; Cook, 2013; Cartwright et al., 2014). Finally, in rivers that run through coarse alluvial sediments, water from the hyporheic or parafluvial zones may provide a source of ²²²Rn additional to groundwater inflow (Cook et al., 2006; Cartwright et al., 2014, Bourke et al., 2014a). 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 calculations (Cook et al., 2003, 2006; Mullinger et al., 2007, 2009; Cartwright et al., 2011, 2014; McCallum et al., 2012; Unland et al., 2013). Carrying out studies at baseflow conditions when most of the water contributing to the streams is from groundwater inflows allows for a comparison between the caluclated groundwater inflows and the observed increase in streamflows, which in turn provides for a test of the parameters used in the ²²²Rn mass balance (Cartwright et al., 2014).

1.2. Objectives

This paper examines groundwater-river interaction in the Avon River, southeast Australia, primarily using ²²²Rn as a tracer. 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). There has been little attempt, however, to quantify groundwater inflows or determine their distribution, and there are insufficient groundwater monitoring bores in the catchment to understand the relationship of groundwater to the river using hydraulic data. Understanding groundwater-river interaction is required to protect and manage the Avon River, especially in assessing the potential impacts of increased groundwater or surface water use.

The paper has two specific aims. Firstly, we use data from a 6 year period to examine whether periodic major flooding events, which alter the geometry of the Avon River floodplain, change the locations of groundwater inflows. Understanding whether the locations of groundwater inflows change following

major flood events, and whether we can monitor those changes, is important to understanding groundwater-river interactions. Secondly, we assess the impacts of parafluvial exchange on the ²²²Rn budget. The Avon River floodplain comprises coarse-grained unconsolidated alluvial sediments with gravel banks, point bars, and pool and riffle sections that likely host parafluvial flows. Rivers with similar coarse-grained sediments on their floodplains are common at mountain fronts and parafluvial flow is likely to be an important process in these settings. Despite parafluvial inflows being a potential important contributor of ²²²Rn budget to rivers, few studies have explicitly considered this process in the ²²²Rn mass balance (e.g., Bourke et al., 2014a; Cartwright et al., 2014). Thus, the results of this study will help improve the general utility of ²²²Rn as a tracer of groundwater inflows into rivers.

2. The Avon Catchment

The Avon River is an unregulated river in the Gippsland Basin of southeast Australia (Fig. 1) that has a total catchment area of ~1830 km² (Cochrane et al., 1991; Department of Environment and Primary Industries, 2015). It 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 highland areas represent ~30% of the Avon catchment and are dominated by temperate native eucalyptus forest, whereas the majority of the plains representing ~70% of the catchment have been cleared for agriculture, which includes 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. 1). 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 10-7 and 10-5 m sec-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 sec⁻¹ (Brumley et al., 1981; Walker and Mollica, 1990). Average rainfall within the Avon catchment ranges from ~1.5 m yr⁻¹ in the highlands to ~0.9 m yr⁻¹ on the plains with most precipitation occurring 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 continuously at three sites (The Channel, Stratford, and Chinns Bridge: Fig. 1). Total annual streamflow at Stratford between 1977 and 2014 was between 1.3×10^7 and 9.0×10^8 m³ yr⁻¹ (median = 3.0×10^8 m³ yr⁻¹) and varied 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. 2). Streamflow generally increases downstream at all times, except at very low flows when streamflow decreases between Stratford and Chinns Bridge. Valencia Creek and Freestone Creek are the main tributaries; both have streamflow measurements (Department of Environment and Primary Industries, 2015) and enter the Avon in the upper reaches of the studied section (Fig. 1). The Avon River has incised through the Haunted Hill 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 long) and narrow (typically <5 m) faster-flowing riffle sections that commonly have steep longitudinal gradients.

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The floodplain of the Avon River between Browns (0.0 km) and Redbank (41.3 km) (Fig. 1) comprises numerous gravel banks and point bars of coarse-gained immature unconsolidated sediments with clasts of up to 50 cm in diameter. In regions where the river is incised, there are seeps of water at the base of the slope and permanent patches of water-tolerant vegetation. The alluvial sediments on the floodplain are sparsely vegetated and the geometry of the floodplain changes markedly following major flood events, such as those in 2011, 2012, and 2013 (Fig. 2). These changes include the downstream migration of pools (often by several tens of metres), scouring of the alluvial sediments, and changes to the location of the sediment banks. Downstream of Redbank, the Avon River occupies an incised channel with banks of finer-grained (clay to sand sized) sediments. The banks and floodplain are more vegetated and do not change markedly during the flood event.

Groundwater flows from the Victorian Alps to the coast (Hofmann and Cartwright, 2013: Fig. 1). Use of water from the Avon River and its tributaries for irrigation is up to $8x10^6$ m³ yr⁻¹ (~2.6% of the annual median streamflow at Stratford); however, there is a prohibition on river water use when the streamflow at Stratford is <10⁴ m³ day⁻¹ (Gippsland Water, 2012).

15 3. Methods

3.1. Sampling

Sampling took place between February 2009 and February 2015 in six campaigns at a variety of streamflows (Fig. 2a). These sampling campaigns were both before and after four major flood events that occurred between 2011 and 2013 and which caused the redistribution of the position of pools and sediment banks in the river. Each sampling campaign involved sampling the river sites (Table A1, Fig. 1) over a two to three day period, with the February 2015 sampling campaign involving additional sites to the others. Distances are measured relative to the first sampling site at Browns (0.0 km) (Fig. 1). Streamflow is measured at three permanent gauging stations: the Channel, which is close to the first sampling site at Browns; Stratford; and Chinns Bridge (Department of Environment and Primary Industries, 2015: Fig. 1). Streamflow was relatively constant during the sampling periods (the variation

in streamflow at Stratford over each sampling period was <5%). 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. 1) 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. Water was also 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.

3.2. Analytical techniques

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Analytical techniques were similar to those in other studies (e.g. Unland et al., 2013; Yu et al., 2013; Cartwright et al., 2014). Cations (Tables A1, A2) were analysed on samples that had been filtered through 0.45µm cellulose nitrate filters and acidified to pH <2 using a ThermoFinnigan quadropole ICP-MS at Monash University. Anions (Tables A1, A2) 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 A2) and surface water (Table A1) 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 Bequerels per m³ of water (Bq m⁻³). 0.5 L of sample was collected by bottom-filling a glass flask and ²²²Rn was subsequently degassed for 5 minutes into a closed air loop of known volume. Counting times were 2 hours for surface water and 20 minutes for groundwater. Typical relative precision based on repeat sample measurements in this and other studies (e.g., Cartwright et al., 2011, 2014) is <3% at 10,000 Bq m⁻³ and \sim 10% at 100 Bq m⁻³. Forty four samples of river bed sediments from sites along the Avon River were collected in March, 2014 and February 2015. ²²²Rn emanation rates (γ) from these were determined by sealing a known dry weight of sediment in airtight containers with water and allowing 222Rn to accumulate (Lamontagne and Cook, 2007). Following 4-5 weeks incubation, by which time the rate of ²²²Rn

- 1 production and decay will have reached steady state, 20 to 40 ml of pore water was extracted and
- 2 analysed for ²²²Rn activities using the same method as above but with counting times of 6 to 12 hours.
- 3 γ (Table 2) was calculated from ²²²Rn produced per unit mass of sediment E_m , sediment density ρ_s , and
- 4 porosity ϕ by:

6 (parameters summarised in Table 1).

7 3.3. Radon mass balance

8 Assuming that the atmosphere contains negligible radon, the change in ²²²Rn activities along a river is:

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$$Q\frac{dc_r}{dx} = I(c_{gw} - c_r) + wEc_r + F_h + F_p - kdwc_r - \lambda dwc_r$$
 (2)

10 (modifed from Mullinger et al., 2007; Cartwright et al., 2011; and Cook, 2013). In Eq. (2): Q is streamflow; c_r and c_{qw} are the ²²²Rn activities in the river and groundwater, respectively; I is the 11 12 groundwater flux per unit length of river; E is the evaporation rate; x is distance along the river; w is river width; d is river depth; F_h and F_p are the inputs of 222 Rn resulting from exchange with the 13 14 hyporheic zone and inflows of parafluvial waters, respectively; k is the gas-transfer coefficient; and λ 15 is the decay constant (Table 1). A similar mass balance also applies to major ion concentrations. Since 16 the concentration of a conservative tracer such as CI is controlled only by groundwater inflows and 17 evaporation, only the first two terms on the right-hand-side of Eq. (2) are relevant. If the river is gaining 18 throughout and solely fed by groundwater the increase in streamflow downstream is:

$$19 \qquad \frac{dQ}{dx} = I - Ew \tag{3}.$$

- The 222 Rn activity in the hyporheic zone waters (c_h) is governed by the 222 Rn activity of the water
- 21 flowing into the hyporheic zone (c_{in}), the ²²²Rn emanation rate γ , and the residence time t_h :

$$1 c_h = \left(\frac{\gamma}{\lambda} - c_{in}\right) \left(1 - e^{-\lambda t_h}\right) + c_{in} (4)$$

(Hoehn et al., 1992; Hoehn and Cirpka, 2006) (Fig. 3a). An identical expression relates the ²²²Rn activity in the parafluvial zone waters (c_p) to the residence time of that water in the parafluvial zone (t_p) . c_h increases with t_h until secular equilibrium is approached at which point, $c_h = \gamma/\lambda$. In a losing or neutral (i.e. neither gaining nor 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 ²²²Rn activities. Cartwright et al. (2014) discussed using the concentration of a conservative ion such as CI to estimate the degree of mixing within the alluvial sediments to estimate cin. Assuming that all the water entering the hyporheic zone subsequently re-enters the river, the 222 Rn flux from the hyporheic zone (F_h) is:

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$$F_h = \frac{\gamma A_h \phi}{1 + \lambda t_h} - \frac{\lambda A_h \phi}{1 + \lambda t_h} c_{in}$$
 (5),

where A_h is the cross-sectional area of the hyporheic zone (Lamontagne and Cook, 2007). Equation (5) 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 provides 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 (5) may also be used to calculate c_p from t_p and γ (e.g., Cartwright et al., 2014). However, where parafluvial flow involves long flow paths through alluvial sediments, an alternative conceptualisation is to consider the flux of ²²²Rn 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 that case, F_p is given by a similar expression to that which accounts for the input of ²²²Rn due to groundwater inflows:

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$$F_p = I_p (c_p - c_r)$$
 (6),

where I_p is the flux of water from the parafluvial zone per unit length of the river. The minimum I_p required to produce a given F_p is achieved when c_p approaches steady state (Fig. 3b), which requires

- 1 t_h to be at least several days (c_p is ~95% of the steady state activity after 16 days: Fig. 3a). If t_h is less
- than the time required to achieve steady state, c_p is lower, and a higher I_p is required to achieve the
- 3 same F_p . The volume of sediments with which the water has interacted during flow through the
- 4 parafluvial zone (V_p in m³ per m length of river) is governed by I_p , t_p and ϕ . If the flow paths through
- 5 the parafluvial zone are regular, V_p will be the cross-sectional area of the parafluvial zone through
- 6 which the water from the river flows (A_p) :

$$7 V_p = A_p = \frac{t_p I_p}{\phi} (7)$$

- 8 (Bourke et al., 2014a). For the same input parameters, Eqs (5) and (6) yield closely similar estimates
- 9 of F_p (Bourke et al., 2014a) and the least well-known parameters are in both cases A_p and t_p .
- 10 There are several approaches that may be used to estimate the rate of ²²²Rn degassing from rivers.
- 11 Firstly, as degassing involves diffusion of ²²²Rn through the boundary layer at the river surface, the
- stagnant film model yields a gas transfer velocity as D/z (which is closely related to k), where z is the
- thickness of the boundary layer at the water surface (Ellins et al., 1990; Stellato et al., 2008). z and by
- extension D/z can be calculated from differences in river ²²²Rn concentrations in losing reaches. The
- gas transfer coefficient k may be estimated in a similar way from the change in 222 Rn activities in losing
- reaches (e.g., Cartwright et al., 2011; Cook 2013) or even in gaining reaches if groundwater inflows
- 17 have been estimated using other tracers, numerical models, streamflow measurements, and/or
- streambed temperature profiles (Cook et al., 2003; Cartwright et al., 2014; Cartwright and Gilfedder,
- 19 2015). Determining k or z by comparing calculated and measured ²²²Rn activities requires that the
- 20 ²²²Rn contributed from the hyporheic or parafluvial zones is quantified, and that there are no inflows
- of water from tributaries that may increase or decrease 222 Rn activities. Since k values are typically
- 22 calculated from these methods for a few specific well-understood river reaches, it is possible that they
- are not valid for all river reaches.

- 1 It is also possible to measure k directly by using introduced gas tracers such as SF₆ (Cook et al., 2003;
- 2 Cook et al., 2006; McCallum et al., 2012; Bourke et al., 2014a), which has the advantage of estimating
- 3 *k* for the river being studied. However, such measurements are generally made along small reaches of
- 4 a river that may not be representative of the river as a whole. Additionally, if the experiments were
- 5 made at specific flow conditions, the gas transfer coefficients may or may not be applicable to
- 6 sampling campaigns made at different flow conditions.
- 7 There are several empirical relationships that estimate k from river velocities (ν) and depths. The
- 8 commonly used O'Connor and Dobbins (1958) and Negulescu and Rojanski (1969) gas transfer
- 9 equations as modified for ²²²Rn are:

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$$k = 9.301x10^{-3} \left(\frac{v^{0.5}}{d^{1.5}} \right)$$
 (8)

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

- 12 (Mullinger et al., 2007). As discussed by Genereux and Hemond (1992), however, there are numerous
- formulations that can yield very different estimates of k for the same flow conditions and some
- independent assessment of k (for example by matching the predicted and observed decline in 222 Rn
- activities in losing reaches) is needed.

16 **4. Results**

17 4.1. Streamflow

- 18 Between January 2000 and February 2015 streamflow at Stratford varied between 500 and 1.38x10⁸
- 19 m³ day⁻¹ (Department of Environment and Primary Industries, 2015). Despite this period including
- 20 years with well below average rainfall, for example 2006 when rainfall was ~50% of the long-term
- 21 average (Bureau of Meteorology, 2015), there were no periods of zero streamflow. Mean daily
- streamflows at Stratford during the sampling rounds ranged from 10,670 m³ day⁻¹ to 88,800 m³ day⁻¹
- 23 (Table A1, Fig. 2a) which represent streamflow percentiles of 39.5 to 89.9 (Fig. 2b). In February 2015,

- 1 which is the sampling round discussed in most detail below, the mean daily streamflow was 12,510
- 2 m³ day⁻¹ at The Channel, 23,090 m³ day⁻¹ at Stratford, and 25,780 m³ day⁻¹ at Chinns Bridge. Inflows
- from Valencia Creek and Freestone Creek in February 2015 were 2410 m³ day-1 and 600 m³ day-1, 3
- 4 respectively (Department of Environment and Primary Industries, 2015).

4.2. River Geochemistry

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Figure 4a shows the ²²²Rn activities of the Avon River for the six sampling campaigns. There are several 6 7 distinct zones of elevated ²²²Rn activities, notably at Wombat Flat (4.8 km) where ²²²Rn activities are up to 2040 Bq m⁻³ and between Bushy Park and Schools Lane (16.3 to 25.3 km) where ²²²Rn activities 8 are up to 3690 Bq m⁻³. Zones of lower ²²²Rn activities in the upper reaches occur at Smyths Road (8.1 9 10 km) and in the reaches between Stewarts Lane and Stratford (30.1 to 35.1 km). The downstream river 11 reaches between Knobs Reserve and Chinns Bridge (37.8 to 49.7 km) also have relatively low ²²²Rn activities that generally decline downstream. The position of the highest ²²²Rn activities changed in 12 the periods prior to and post the 2011 to 2013 floods. In March 2014 and February 2015 the highest 13 ²²²Rn activities were at Bushy Park (16.3 km), whereas this site had relatively low ²²²Rn activities in 14 February 2009 and April 2010 when the highest ²²²Rn activities were at Pearces Lane (20.0 km). The 15 16 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 17 were recorded during the periods of highest flow (September 2010 and July 2014). 18 19 EC values and CI concentrations generally increase downstream from 54 to 131 μS cm⁻¹ and 4 to 10 mg l⁻¹ at Browns (0.0 km) to as high as 934 μS cm⁻¹ and 98 mg l⁻¹ at Chinns Bridge (49.7 km) (Table A1, 20 Fig. 4b). CI concentrations at low streamflows in March 2014 were generally higher (up to 98 mg l⁻¹) 21 than in the other sampling campaigns, while CI concentrations were <20 mg I⁻¹ during the highest 22 23 streamflows in September 2010. A marked increase in EC values and Cl concentrations occurs downstream of Smyths Road (8.1 km) in the reaches where ²²²Rn activities are highest at low

- streamflows. The concentrations of other major ions (e.g., Na) increase downstream in a similar
- 2 manner (Table A1).

4.3. Groundwater Geochemistry

Groundwater from the near-river bores at Pearces Lane and Stratford has ²²²Rn activities that vary from 480 to 28,980 Bq m⁻³ (Table A2). There is some variation in ²²²Rn activities in individual bores between the sampling rounds with relative standard deviations between 6 and 34%. The mean value of all groundwater ²²²Rn activities (n = 26) is 12,890 Bq m⁻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 ²²²Rn activities is 13,830 Bq m⁻³ (n = 24) with a standard error of 1273 Bq m⁻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 Bq m⁻³. EC values of groundwater from the bores at Pearces Land and Stratford are between 100 and 680 μS cm⁻¹ and Cl concentrations range from 46 to 147 mg l⁻¹ with a mean value of 79±34 mg l⁻¹ (n = 16) (Table A2). If Bore 5 at Pearces Lane is again excluded the mean Cl concentration is 87±28 mg 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 elsewhere in the Avon valley and neighbouring catchments (Department of Environment and Primary Industries, 2015).

4.4. Geochemistry of water from the alluvial gravels

EC values of water within the gravels further than 1 to 2 m from the edge of the river are between 120 and 550 μ S cm⁻¹ (n = 52) (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 of these samples were between 7000 and 28,000 Bq m⁻³ (n = 21) (Fig. 5a), which are also significantly higher than the ²²²Rn

- 1 activities in the adjacent river. As discussed below, these data are interpreted as indicating that the
- 2 gravels contain a mixture of groundwater and parafluvial water.

3 4.5. ²²²Rn Emanation Rates

4 ²²²Rn emanation rates were determined via Eq. (1). The matrix density was assigned as 2700 kg m⁻³, 5 which is appropriate for sediments rich in quartz ($\rho = 2650 \text{ kg m}^{-3}$), and a porosity of 0.4 was used, 6 which is appropriate for unconsolidated poorly-sorted riverine sediments (Freeze and Cherry, 1979). 7 γ values range from 288 to 4950 Bg m⁻³ with a mean value of 2308±1197 Bg m⁻³ (n = 44) and a standard 8 error of 183 Bq m⁻³. 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. The 9 relative variability in γ between the sediments is similar to that reported elsewhere (e.g., Bourke et 10 al., 2014a; Cartwright et al., 2014). 222Rn activities of water in equilibrium with the sediments are given 11 by γ/λ (Cecil and Green, 2000), and the mean γ/λ value is 12,751±6615 Bq m⁻³ with a standard error 12 of 1009 Bq m⁻³. These γ/λ values are not significantly different (p ~0.5) to the measured ²²²Rn activities 13

5. Discussion

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of the groundwater.

The following observations imply that overall the Avon is a gaining river: 1) even during periods of prolonged low rainfall the river continues to flow and streamflow commonly increases between The Channel and Chinns Bridge gauges; 2) ²²²Rn activities are higher than those that could be maintained by hyporheic exchange alone (Cartwright et al., 2011; Cook 2013); 3) Cl 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 Cl concentrations are used to assess the location and magnitude of groundwater inflows.

5.1. Distribution of groundwater inflows

The February 2009, April, 2010, March 2014, and February 2015 sampling campaigns represent lower streamflows. Because the majority of water in the Avon River at these times is likely to be provided

by groundwater, the ²²²Rn activities from these sampling campaigns are most useful in understanding the distribution of groundwater inflows. The region between Smyths Road and Ridleys Lane (8.1 to 23.0 km) where ²²²Rn activities increase and remain high (Fig. 4a), especially at lower streamflows, and where there is a marked increase in Cl concentrations (Fig. 4b) is interpreted as receiving major groundwater inflows. This section of the Avon River is incised up to 4 m below the floodplain which likely produces steep hydraulic head gradients that result in groundwater discharge on the floodplain and into the river. There are also groundwater seeps and patches of perennial water tolerant vegetation at the edge of the floodplain in this area. The reaches between Browns and Wombat Flat (0.0 to 4.8 km) and Stewarts Lane and Stratford (30.1 to 35.1 km) are also characterised by high ²²²Rn activities and are again interpreted as receiving groundwater inflows. The reaches between Wombat Flat and Smyths Road (4.8 to 8.1 km), Ridleys Lane and Stewarts Lane (23.0 to 30.1 km), and Knobs Reserve and Chinns Bridge (37.8 to 49.7 km) where there is a gradual decline in ²²²Rn activities and little change in Cl concentrations (Fig. 4) are interpreted as either being losing or receiving minor groundwater inflows. The landscape is flatter and the river less incised in these areas which results in lower hydraulic gradients and consequently less groundwater inflows to the river. The difference in the location of the highest ²²²Rn activities between the sampling campaigns that were conducted before and after the major floods (i.e., pre 2011 vs. post 2013) indicates that the locations of groundwater inflows changed. The major floods changed the location of pools and sediment banks on the Avon River and caused scouring, which would change the relationship of the

5.2. Quantifying Groundwater Inflows

river to the groundwater.

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This section concentrates on modelling the ²²²Rn activities for the detailed February 2015 sampling campaign (Fig. 4a). It was considered that groundwater inflows, hyporheic exchange, and parafluvial flow all contributed ²²²Rn to the river. The groundwater ²²²Rn activity was assumed to be 13,000 Bq

1 m⁻³, which is consistent both with the measured ²²²Rn activities of groundwater (Table A2) and the 2 calculated ²²²Rn activities of water in equilibrium with the alluvial sediments. 3 The flux of 222 Rn from the hyporheic zone was estimated from Eq. (5) using the mean γ value of 2300 4 Bq m⁻³ day⁻¹ (Table 2), a porosity of 0.4 (which is appropriate for coarse-grained unconsolidated sediments), and a value for c_{in} that is the ²²²Rn activity of the river in that reach. The residence time of 5 6 water within the hyporheic zone is likely to be short (Boulton et al., 1998; Tonina and Buffington, 2011; 7 Zarnetske et al., 2011; Cartwright et al., 2014), and $t_h = 0.1$ days is assumed here; for $t_h < 1$ day, F_h is 8 relatively insensitive to the actual residence times in the hyporheic zone (Lamontagne and Cook, 2007; 9 Cartwright et al., 2014). The width of the hyporheic zone has been assigned as the river width. The 10 thickness of the hyporheic zone is less well known; however, by analogy with rivers elsewhere, it is 11 likely to be a few centimetres thick (Boulton et al., 1998; Hester and Doyle, 2008; Tonina and 12 Buffington, 2011) and a value of 10 cm is initially adopted. 13 Parafluvial flow is conceived to occur on the tens of metres to kilometre scale and to represent water 14 that is lost from the river into the floodplain sediments that subsequently re-enters the river downstream. The Cl and ²²²Rn data from the water contained within the gravels (Fig. 5) are interpreted 15 16 as reflecting mixing of groundwater and parafluvial flows that will occur where the river is gaining. 17 This scenario requires that the river is locally losing. As discussed above, on the kilometre scale the 18 Avon River may contain losing reaches. Additionally, the reaches that are interpreted as being overall 19 gaining may contain smaller sections that are losing. In particular, the riffle sections commonly have 20 steep longitudinal gradients and may transition from losing at the upstream end to gaining at the 21 downstream end. Parafluvial flow is probably hosted mainly within the coarser-grained alluvial 22 sediments (although conceivably it could also include water that flows through the upper levels of the 23 aquifers underlying the alluvial sediments). By contrast with hyporheic exchange which occurs along 24 all reaches (whether gaining or losing), inflows from the parafluvial zone require upward head 25 gradients and only occur where the river is gaining. The parafluvial inflows will increase the ²²²Rn

activities in the river in a similar manner to inflowing groundwater. However, because it represents

- water that originated from the river, the inflows from the parafluvial zone do not increase the overall streamflow. If the parafluvial zone water is in secular equilibrium with the sediments, $c_p \sim 12,700$ Bq m⁻³ (Table 2). Average evaporation rates in southeast Australia in February to April are 3x10⁻³ to 5x10⁻³ m day⁻¹ (Bureau of Meteorology, 2015) and a value of 4x10⁻³ m day⁻¹ was adopted. Average river width and depth is 10 m and 0.5 m, respectively, upstream of Wombat Flat (0.0 to 4.8 km) and 20 m and 1 m, respectively, for the rest of the river The gas transfer coefficient was estimated from the decline in ²²²Rn activities between Ridleys Lane and Schools Lane (23.0 to 25.3 km) (Fig. 4a). This approach estimates the net $kdwc_r$ term and k was estimated as 0.3 day⁻¹ using the measured widths, depths, and ²²²Rn concentrations. This requires that this is a losing stretch of the river, so that there are no groundwater or parafluvial inflows. That Cl concentrations do not increase over this stretch of river (Fig. 4b) are consistent with it being losing. A k 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, 2014; Atkinson et al., 2013; Unland et al., 2013; Yu et al., 2014). However, as the Avon River is dominated by slow-flowing pools, degassing
 - Groundwater inflows were calculated from the 222 Rn activities by solving Eq. (2) using a finite difference approach in a spreadsheet with a distance step of 10 m (the use of smaller or larger distance steps 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. (3). The calculations estimated the values of I and I_p in each reach by matching the calculated and measured 222 Rn activities along the river with the additional constraint that the total groundwater inflows cannot exceed the net increase in streamflow between the Channel gauge and the gauges at Stratford and Chinns Bridge (Fig. 1). Since there are few streamflow measurements, the calculations assumed that the ratio of I to I_p was the same in all gaining reaches of the river.

rates are expected to be low.

1 Assuming that in the gaining reaches there are 50% parafluvial inflows and 50% groundwater inflows 2 allows both the ²²²Rn variations and the increase in streamflow to be accounted for (Fig. 6a). Calculated groundwater and parafluvial inflows are highest in the reaches between Smyths Road and 3 4 Pearces Lane (8.1 to 20.0 km) (Fig. 6b), which is the region where CI concentrations also increase 5 markedly (Fig. 4b). Assuming that the waters are in secular equilibrium with the sediments, the 6 combined inflows of groundwater and parafluvial water for this reach are up 2.5 m³ m⁻¹ day⁻¹ of which 7 groundwater inflows are ~1.26 m³ m⁻¹ day⁻¹. 8 There is no process in the parafluvial or hyporheic zones other than mixing with groundwater that 9 increases the CI concentrations of the through-flowing water. Thus the CI concentrations in the river 10 reflect only the groundwater inflows and in theory it would be possible to use CI to quantify these 11 (c.f., McCallum et al., 2012). However, the high variability of Cl concentrations in the groundwater and 12 the relatively small difference between groundwater and river CI concentrations results in large uncertainties. The change in CI concentrations (Fig. 6d) was calculated from the groundwater inflows 13 assuming that groundwater has a CI concentration of 85 mg l⁻¹. The calculated CI concentrations are 14 15 slightly higher than those observed, but if the CI concentration of the groundwater is allowed to vary 16 within the 95% confidence interval ($\pm 16 \text{ mg l}^{-1}$) the observed trend can be reproduced. 17 If residence times in the parafluvial zone are shorter than those required to attain secular equilibrium, c_p will be lower and the inflows from the parafluvial zone (I_p) required to produce a given flux of ²²²Rn 18 (F_p) increases (Fig. 3). For example, if $c_r = 2300$ Bg m⁻³, which is a typical value in many of the reaches 19 between Valencia to Bushy Park (10.9 to 16.3 km) and $c_p = 12,700 \text{ Bq m}^{-3}$, $(c_p - c_r) = 10,400 \text{ Bq m}^{-3}$. If I_p 20 = 1 m³ m⁻¹ day⁻¹, F_p = 10,400 Bg m⁻¹ day⁻¹ (Eq. 6). If γ = 2300 Bg m⁻³ day⁻¹, c_p is 2487, 4023 and 11,004 21 Bq m⁻³ where t_p is 0.1, 1, and 10 days, respectively. To produce a value of F_p of 10,400 Bq m⁻¹ day⁻¹ 22 23 requires $I_p \sim 58 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$ for $t_p = 0.1 \text{ days}$, $\sim 6.0 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$ for $t_p = 1 \text{ day}$, and $\sim 1.2 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$ for $t_p = 10$ days. For $t_p > 30$ days the system is close to secular equilibrium and c_p and l_p are near constant 24

(Fig. 3). The cross-sectional area of the parafluvial zone A_p required to accommodate these parafluvial

- flows with ϕ = 0.4 and t_p between 0.1 and 100 days is between 14 and 250 m² (Eq. 7). The floodplain
- 2 of the Avon River is tens of metres wide with sediment thicknesses of several metres and even the
- 3 higher estimates of the cross-sectional area are not unreasonable given the volume of gravels on the
- 4 floodplain.

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5.3. Uncertainties and sensitivity

The proposal that parafluvial flow is important in the Avon River is consistent with the local hydrogeology and allows both the ²²²Rn and net increase in streamflow to be reproduced. The conclusion that inflows of parafluvial zone waters only occur in the gaining reaches is justifiable as the conditions required for groundwater inflows (gaining river with steep hydraulic gradients and highhydraulic 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. Given the multiple parameters in Eq. (2) and their inherent uncertainties, however, consideration needs to be given to whether both the ²²²Rn activities and the increases in streamflow can be accounted for without parafluvial inflows being a significant source of ²²²Rn. Matching the ²²²Rn profile along the Avon River using the parameters discussed above but without input of ²²²Rn from parafluvial zone would require net groundwater inflows of 28,300 m³ day⁻¹. However, these inflows exceed the measured increase in streamflow between The Channel and Chinns Bridge of 15,500 m³ day⁻¹ by 180% (Fig. 7a). The February 2015 sampling round took place at the end of summer when the small ephemeral tributaries were dry and there was no overland flow; however, there were still flows from Valencia Creek and Freestone Creek of 1,410 m³ day⁻¹ and 200 m³ day⁻¹, respectively. If these were included, the discrepancy between the calculated and observed streamflow increases. The calculated CI concentrations are also higher than observed (Fig. 7d), although given the uncertainty in groundwater Cl concentrations, the discrepancy is not large. 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 degassing and decay, lower

groundwater inflows are required to replicate the observed ²²²Rn activities if the groundwater inflows occur immediately upstream of a sampling point (Cook, 2013). Even assigning the groundwater inflows in each reach to the 10 m section upstream of the measurement point still results in the calculated streamflow overestimating the measured streamflow (Fig. 7c). The predicted ²²²Rn activities in the river in this case are also not realistic (Fig. 7a). The evaporation term in Eq. (2) 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. The main parameter impacting calculated groundwater inflows is the ²²²Rn activity of groundwater (Cartwright et al., 2011; Cook, 2013). Allowing c_{gw} to vary within the 95% confidence interval of the groundwater ²²²Rn activities (±2600 Bq m⁻³) makes little difference to the discrepancy between the calculated and observed increase in streamflow (Fig. 7c). Increasing c_{gw} to 27,000 Bq m⁻³ allows both the ²²²Rn profile and the observed increase in streamflow between The Channel and Chinns Bridge to be reproduced without the requirement for the input of ²²²Rn from the parafluvial zone (Fig. 8). However, there is no known groundwater in the Avon catchment with such high ²²²Rn activities and these activities are far higher than would be in equilibrium with the alluvial sediments that comprise the near-river aquifer lithologies. Hence, it is considered not possible that groundwater ²²²Rn activities could be this high. There is uncertainty in the gas transfer coefficient. 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⁻¹ increases the calculated groundwater inflows, which increases the discrepancy between the observed and calculated increases in streamflow. k estimated from Eqs. (8) and (9) ranges between 0.1 and 0.3 day⁻¹. Using $k = 0.1 \text{ day}^{-1}$ produces net groundwater inflows that more closely match the observed increase in streamflow. However, adopting k = 0.1 day⁻¹ results in the calculated ²²²Rn activities in a number of reaches being overestimated (Fig. 8). This is because even assuming no groundwater inflows into those reaches, the loss of ²²²Rn by 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.

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While there are uncertainties in c_h , the main uncertainty in the contribution of hyporheic exchange to the 222 Rn budget is the dimensions of the hyporheic zone. Increasing F_h also reduces the calculated groundwater inflows. Using the same emanation 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. However, the higher values of F_h again result in a poor fit between predicted and observed ²²²Rn activities (Fig. 8). Because the error in λ is negligible and the evaporation term is much smaller than the other terms, it is generally possible to produce identical trends in ²²²Rn activities with different combinations of k and F_h (Cartwright et al., 2014). If F_h is calculated assuming a 50 cm thick hyporheic zone, adopting k = 0.6 day^{-1} reproduces the observed ²²²Rn activities. Similarly, if $k = 0.1 day^{-1}$ a match between the observed and the predicted ²²²Rn activities is achieved with no hyporheic exchange ($F_h = 0$). However, these combinations of parameters again result in estimated net groundwater inflows that exceed the measured increase in streamflow. There is an unknown error in the streamflow measurements, 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-hand-side of Eq. (2), which in turn reduces I. If widths were reduced by 50% (an unrealistic error), net groundwater inflows broadly match the increase in streamflow. However, this again results in ²²²Rn activities being overestimated in many reaches (Fig. 8). Increasing k to 0.65 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. Overall it is concluded that there are no combination of parameters that can reproduce both the observed ²²²Rn activities and streamflows without incorporating parafluvial flow. It would be possible to explain the observed ²²²Rn activities and streamflows if there were losing reaches in the Avon River through which significant volumes of river water were lost to the underlying

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aquifers and, unlike parafluvial flow, this water did not subsequently return to the river. For this scenario to be valid, approximately 50% of the groundwater inflows would have to be lost from the river in these losing reaches in February 2015. The reaches between 25 are 30 km are interpreted as losing. However, these reaches do not dry up even during prolonged drought (Gippsland Water, 2012), and all reaches of the river were flowing during the 2009 sampling campaign (which had the lowest streamflows). Also while streamflows were not measured, such a major reduction in streamflow over such a short distance would be apparent in the field. Likewise, significant pumping of water from the river would also reduce streamflows. 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.

5.4. Other sampling campaigns

The predicted distribution of groundwater inflows in February 2009, April, 2010, and March 2014 when streamflows were low to moderate are similar to those in February 2015 (Fig. 4). Due to the lower number of sampling points, it is difficult to calculate groundwater inflows with certainty. The net groundwater inflows calculated using the same parameters as above but ignoring parafluvial flows are between 15,900 and 21,700 m³ day⁻¹, respectively (Fig. 9), which are up to 490% of the measured increases in streamflow between The Channel and Chinns Bridge. Again propagating the likely uncertainties in the parameters through Eq. (2) cannot resolve this discrepancy, implying that the inflows of water from the parafluvial zone must be a significant part of the ²222Rn budget.

At the higher streamflows there will likely be significant inputs to the river from overland flow or interflow; hence, it is not possible to use the comparison between calculated groundwater inflows and the net increase in streamflow to independently test for the input of ²222Rn from the parafluvial zone. For example, without incorporating parafluvial flow, the net groundwater inflows 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 day⁻¹, F_h adjusted for the higher river widths are 32,100 m³ day⁻¹ (September
- 2 2010) and 44,600 m³ day⁻¹ (July 2014). These net groundwater inflows are lower than the measured
- 3 increases in streamflow between The Channel and Stratford or Chinns Bridge (Fig. 9). However, it is
- 4 likely that significant parafluvial flow occurs at those times and consequently that these values also
- 5 represent an overestimation the actual groundwater inflows.

6. Conclusions

- 7 The variation in ²²²Rn activities and Cl concentrations clearly define the reaches of the Avon River that
- 8 are gaining. The distribution of ²²²Rn activities also indicate that the location of groundwater inflows
- 9 changed after major floods that occurred between 2011 and 2013. This approach can be applied to
- 10 other rivers where flood events change the geometry of the floodplain sediments and where the
- 11 groundwater monitoring bore network is insufficient to define groundwater-river interaction.
- 12 The Avon River has coarse-grained unconsolidated gravels along its floodplain and it was concluded
- that parafluvial flow was a significant process in controlling the ²²²Rn activities of the river. However,
- 14 this proposition is difficult to definitively test or explore in more detail. The groundwater and
- parafluvial inflows have been assumed to occur in similar proportions in each reach, which may not
- 16 necessarily be the case. Parafluvial flow is likely to be important in rivers with coarse-grained alluvial
- 17 sediments on their floodplains, especially where there are locally alternating gaining and losing
- 18 reaches, and must be taken into account in ²²²Rn mass balance calculations. Unlike hyporheic
- 19 exchange, which occurs in all stretches, parafluvial inflows are likely to dominantly occur in gaining
- reaches augmenting the groundwater inflows.
- 21 Theoretically, a conservative tracer such as CI that is unaffected by parafluvial flow could be used to
- 22 separate groundwater inflows from parafluvial inflows. However, the relatively high variability of
- 23 groundwater Cl concentrations and the relative small difference between groundwater and river Cl
- concentrations make this impractical in the Avon Catchment. Nevertheless, this may be possible in
- 25 other river catchments and illustrates the advantage of using multiple geochemical tracers.

1 More generally, this study illustrates the importance of carrying out geochemical studies at low

streamflows 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. It would be possibly to interpret the changes to ²²²Rn activities

during the periods of higher streamflow as being solely due to groundwater inflows because the net

groundwater inflows are lower than the net increases in streamflow (Fig. 9). However, it is likely that

there is groundwater and parafluvial inflows at all times, in which case calculated groundwater inflows

8 will be overestimated.

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Author Contributions

10 Both authors were involved in field data collection and lab analyses. Ian Cartwright prepared the

11 manuscript with contributions from Harald Hofmann.

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References

21 Atkinson, A., Cartwright, I., Gilfedder, B., Hofmann, H., Unland, N., Cendón, D., and Chisari, R.: A multi-

tracer approach to quantifying groundwater inflows to an upland river; assessing the influence of

variable groundwater chemistry, Hydrol. Proc., 29, 1-12, 2015.

- 1 Barron, O., Silberstein, R., Ali, R., Donohue, R., McFarlane, D.J., Davies, P., Hodgson, G., Smart, N., and
- 2 Donn, M.: Climate change effects on water-dependent ecosystems in south-western Australia, J.
- 3 Hydrol., 434–435, 95-109, 2012.
- 4 Boulton, A.J., Findlay, S., Marmonier, P., Stanley, E.H., and Maurice Valett, H.: The functional
- 5 significance of the hyporheic zone in streams and rivers, Annu. Rev. Ecol. Syst., 29, 59-81, 1998.
- 6 Bourke, S.A., Cook, P.G., Shanafield, M., Dogramaci, S., and Clark, J.F.: Characterisation of hyporheic
- 7 exchange in a losing stream using radon-222, J. Hydrol., 519, 94-105, 2014a.
- 8 Bourke, S.A., Harrington, G.A., Cook, P.G., Post, V.E., and Dogramaci, S.: Carbon-14 in streams as a
- 9 tracer of discharging groundwater, J. Hydrol., 519, 117-130, 2014b.
- 10 Briody, A.C., Cardenas, M.B., Shuai, P., Knapper, P.S., and Bennett, P.C.: Groundwater flow, nutrient,
- and stable isotope dynamics in the parafluvial-hyporheic zone of the regulated Lower Colorado
- 12 River (Texas, USA) over the course of a small flood. Hydrgeol. J., DOI 10.1007/s10040-016-1365-3,
- 13 2016.
- 14 Brodie, R., Sundaram, B., Tottenham, R., Hostetler, S., and Ransley, T.: An overview of tools for
- assessing groundwater-surface water connectivity. Bureau of Rural Sciences, Canberra, Australia,
- 16 133p., 2007.
- Brumley, J.: An investigation of the groundwater resources of the Latrobe Valley, Victoria, Proc. Geol.
- Soc. Austr. Coal Group Symposium, 562-581, 1982.
- 19 Bureau of Meteorology: Commonwealth of Australia Bureau of Meteorology http://www.bom.gov.au,
- 20 2015, last accessed 30 June 2015
- 21 Burnett, W.C. and Dulaiova, H.: Radon as a tracer of submarine groundwater discharge into a boat
- basin in Donnalucata, Sicily, Cont. Shelf Res., 26, 862-873, 2006.
- 23 Cartwright, I. and Gilfedder, B.: Mapping and quantifying groundwater inflows to Deep Creek
- 24 (Maribyrnong catchment, SE Australia) using ²²²Rn, implications for protecting groundwater-
- dependant ecosystems, Appl. Geochem., 52, 118-129, 2015.
- 26 Cartwright, I., Hofmann, H., Gilfedder, B., and Smyth, B.: Understanding parafluvial exchange and
- degassing to better quantify groundwater inflows using ²²²Rn: The King River, southeast Australia,
- 28 Chem. Geol., 380, 48-60, 2014.

- 1 Cartwright, I., Hofmann, H., Sirianos, M.A., Weaver, T.R., and Simmons, C.T.: Geochemical and ²²²Rn
- 2 constraints on baseflow to the Murray River, Australia, and timescales for the decay of low-salinity
- 3 groundwater lenses, J. Hydrol., 405, 333-343, 2011.
- 4 Cecil, L.D. and Green, J.R.: Radon-222, in: Environmental tracers in subsurface hydrology, Cook, P.G.
- 5 and Herczeg, A.L. (Eds), Kluwer, Boston, USA, 175-194, 2000.
- 6 Cochrane, G.W., Quick, G.W., and Spencer-Jones, D: Introducing Victorian Geology. Geological Society
- of Australia, Victorian Division, Melbourne, Australia, 304p, 1991.
- 8 Cook, P.G.: Estimating groundwater discharge to rivers from river chemistry surveys, Hydrol. Proc., 27,
- 9 3694-3707, 2013.
- 10 Cook, P.G., Favreau, G., Dighton, J.C., and Tickell, S.: Determining natural groundwater influx to a
- tropical river using radon, chlorofluorocarbons and ionic environmental tracers, J. Hydrol., 277, 74-
- 12 88, 2003.
- 13 Cook, P.G., Lamontagne, S., Berhane, D., and Clarke, J.F.: Quantifying groundwater discharge to
- 14 Cockburn River, southeastern Australia, using dissolved gas tracers Rn-222 and SF6, Water Resour.
- 15 Res., 42, W10411, doi:10.1029/2006WR004921, 2006.
- 16 Department of Environment and Primary Industries: Victoria Department of Environment and Primary
- 17 Industries Water Monitoring, http://data.water.vic.gov.au/monitoring.htm, 2015, last accessed 10
- 18 July 2015.
- 19 Edwardson, K.J., Bowden, W.B., Dahm, C., and Morrice, J.: The hydraulic characteristics and
- 20 geochemistry of hyporheic and parafluvial zones in Arctic tundra streams, north slope, Alaska, Adv.
- 21 Water Resour., 26, 907-923, 2003.
- 22 Ellins, K.K., Roman-Mas, A., and Lee, R.: Using ²²²Rn to examine groundwater/surface discharge
- interaction in the Rio Grande de Manati, Puerto Rico. J. Hydrol., 115, 319-341, 1990.
- Freeze, R.A. and Cherry, J.A.: Groundwater. Prentice-Hall, New Jersey, USA, 604p., 1979...
- 25 Genereux, D.P. and Hemond, H.F.. Determination of gas exchange rate constants for a small stream
- on Walker Branch watershed, Tennessee . Water Resour. Res., 28, 2365-2374, 1992.
- 27 Gippsland Water: Water Supply Demand Strategy. www.gippswater.com.au/Portals/
- 28 Gippsland_Water_Final_WSDS.pdf, 2012, last accessed 30 May 2015.

- 1 Hester, E.T. and Doyle, M.W.: In-stream geomorphic structures as drivers of hyporheic exchange.
- Water Resour. Res., 44, W03417, doi:10.1029/2006WR005810, 2008.
- 3 Hoehn, E. and Cirpka, O.A.: Assessing residence times of hyporheic ground water in two alluvial flood
- 4 plains of the Southern Alps using water temperature and tracers, Hydrol. Earth Syst. Sc., 10, 553-
- 5 563, 2006.
- 6 Hoehn, E. and Von Gunten, H.R.: Radon in groundwater: a tool to assess infiltration from surface
- 7 waters to aquifers, Water Resour. Res., 25, 1795-1803, 1989.
- 8 Hoehn, E., Von Gunten, H.R., Stauffer, F., and Dracos, T.: Radon-222 as a groundwater tracer. A
- 9 laboratory study, Environ. Sci. Technol., 26, 734-738, 1992.
- 10 Hofmann, H. and Cartwright, I.: Using hydrogeochemistry to understand inter-aquifer mixing in the
- on-shore part of the Gippsland Basin, southeast Australia, Appl. Geochem., 33, 84-103, 2013...
- 12 Holmes, R.M., Fisher, S.G., and Grimm, N.B.: Parafluvial nitrogen dynamics in a desert stream
- 13 ecosystem, J. N. Am. Benthol. Soc., 13, 468-478, 1994.
- 14 Kløve, B., Ala-aho, P., Bertrand, G., Boukalova, Z., Ertürk, A., Goldscheider, N., Ilmonen, J., Karakaya,
- 15 N., Kupfersberger, H., Kværner, J., Lundberg, A., Mileusnić, M., Moszczynska, A., Muotka, T., Preda,
- 16 E., Rossi, P., Siergieiev, D., Šimek, J., Wachniew, P., Angheluta, V., and Widerlund, A.: Groundwater
- dependent ecosystems. Part I: Hydroecological status and trends, Environ. Sci. Pol., 14, 770-781,
- 18 2011.
- 19 Lamontagne, S., Cook, P.G.: Estimation of hyporheic water residence time in situ using ²²²Rn
- disequilibrium, Limnol. Oceanogr. Methods, 5, 407-416, 2007.
- 21 McCallum, J.L., Cook, P.G., Berhane, D., Rumpf, C., and McMahon, G.A.: Quantifying groundwater
- 22 flows to streams using differential flow gaugings and water chemistry, J. Hydrol., 416-417, 118-
- 23 132, 2012.
- Mullinger, N.J., Binley, A.M., Pates, J.M., and Crook, N.P.: Radon in Chalk streams: Spatial and temporal
- variation of groundwater sources in the Pang and Lambourn catchments, UK, J. Hydrol., 339, 172-
- 26 182, 2007.
- 27 Mullinger, N.J., Pates, J.M., Binley, A.M., and Crook, N.P.: Controls on the spatial and temporal
- variability of ²²²Rn in riparian groundwater in a lowland Chalk catchment, J. Hydrol., 376, 58-69,
- 29 2009.

- 1 Négrel, P., Casanova, J., and Aranyossy, J.-F.: Strontium isotope systematics used to decipher the origin
- of groundwaters sampled from granitoids: the Vienne Case (France), Chem. Geol., 177, 287-308,
- 3 2001.
- 4 Negulescu, M. and Rojanski, V.: Recent research to determine reaeration coefficients. Water Res., 3,
- 5 189-202, 1969.
- 6 O'Connor, D.J. and Dobbins, W.E.: Mechanisms of reaeration in natural streams, T. Am. Soc. Civ. Eng.,
- 7 123, 641-684, 1958.
- 8 Sophocleous, M.: Interactions between groundwater and surface water: the state of the science,
- 9 Hydrogeol. J., 10, 52-67, 2002.
- Stellato, L., Petrella, E., Terrasi, F., Belloni, P., Belli, M., Sansone, U., and Celico, F.: Some limitations in
- using ²²²Rn to assess river–groundwater interactions: the case of Castel di Sangro alluvial plain
- 12 (central Italy), Hydrogeol. J., 16, 701-712, 2008.
- 13 Tonina, D. and Buffington, J.M.: Effects of stream discharge, alluvial depth and bar amplitude on
- 14 hyporheic flow in pool-riffle channels, Water Resour. Res., 47, W08508,
- 15 doi:10.1029/2010WR009140, 2011.
- 16 Unland, N.P., Cartwright, I., Andersen, M.S., Rau, G.C., Reed, J., Gilfedder, B.S., Atkinson, A.P., and
- Hofmann, H.: Investigating the spatio-temporal variability in groundwater and surface water
- interactions: A multi-technique approach. Hydrol. Earth Syst. Sc., 17, 3437-3453, 2013...
- 19 Walker, G. and Mollica, F.: Review of the Groundwater Resources in the Southeast Region,
- Department of Water Resources Victoria, Report 54, Melbourne, Australia, 68p., 1990.
- 21 Winter, T.C.: Relation of streams, lakes, and wetlands to groundwater flow systems. Hydrogeol. J., 7,
- 22 28-45, 1999.
- 23 Yu, M., Cartwright, I., Braden, J., and de Bree, S.: Examining the spatial and temporal variation of
- groundwater inflows to a valley-to-floodplain river using ²²²Rn, geochemistry and river discharge:
- 25 the Ovens River, southeast Australia, Hydrol. Earth Syst. Sc., 17, 4907-4924, 2013.
- 26 Zarnetske, J.P., Haggerty, R., Wondzell, S.M., and Baker, M.A. Dynamics of nitrate production and
- 27 removal as a function of residence time in the hyporheic zone, J. Geophys. Res., 116, G01025,
- 28 doi:10.1029/2010JG001356, 2011.

1 Figure Captions

- 2 Figure 1. Summary geological and hydrogeological map of the Avon River catchment (Hofmann and
- 3 Cartwright, 2015; Department of Environment and Primary Industries, 2015). Arrows show general
- 4 direction of groundwater flow. Main sampling sites (in order of distance downstream are) are BR =
- 5 Browns, WF = Wombat Flat, SM = Smyths Road, VA = Valencia, BP = Bushy Park, PL = Pearces Lane, RL
- 6 = Ridleys Lane, SC = Schools Lane, ST = Stewarts Lane, SA = Stratford, KR = Knobs Reserve, RB =
- 7 Redbank, CB = Chinns Bridge. Unnamed sampling sites are the additional sites from February 2015
- 8 (Table A1).
- 9 Figure 2a. Variation in streamflow at Stratford (Fig. 1) between January 2009 and February 2015. The
- major floods (highlighted) caused significant changes to the geometry of the floodplain. 2b. Flow
- 11 frequency curve for Stratford for streamflows between January 2000 and March 2015 and the
- 12 percentiles of discharge in the sampling campaigns. Data from Department of Environment and
- 13 Primary Industries (2015).
- 14 **Figure 3a.** Variation in the ²²²Rn activity in the parafluvial or hyporheic zone (c_p or c_h) with residence
- time $(t_p \text{ or } t_h)$ and ²²²Rn emanation rate (γ) (Eq. 3). **3b**. Variation in the water flux from the parafluvial
- zone (I_p) with the flux of ²²²Rn from the parafluvial zone (F_p) and t_p (Eq. 5). In both cases $c_r = c_{in} = 1000$
- 17 Bq m⁻³.
- 18 Figure 4. Downstream variations in ²²²Rn activities (4a) and Cl concentrations (4b) for the six sampling
- 19 campaigns (Data from Table A1, 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
- 21 (Table A1). Site abbreviations as for Fig. 1.
- Figure 5a. Variations in ²²²Rn activities (5a) and EC values (5b) of water extracted from river bank
- 23 gravels. Shaded boxes show range of values in the groundwater (excluding Bore 5 at Pieces Lane) and
- the Avon River (Data from Tables A1 and A2).

- 1 Figure 6a. Calculated and observed ²²²Rn activities for February 2015 resulting from assigning 50% of
- 2 the calculated inflows as parafluvial flow. **6b.** Variation in groundwater and parafluvial inflows. **6c.**
- 3 Calculated streamflow resulting from the groundwater inflows (Eq. 2) vs. measured streamflow at
- 4 Stratford and Chinns Bridge. **6d.** Predicted vs. observed Cl concentrations. Shaded field is the range
- 5 resulting from varying groundwater Cl concentrations within the 95% confidence interval.
- 6 **Figure 7a.** Calculated vs. observed ²²²Rn activities in the Avon River for February 2015 assuming both
- 7 uniform groundwater inflow within each reach and the situation where groundwater inflow occurs
- 8 immediately upstream of the measurement point. Site abbreviations as for Fig. 2. 7b. Calculated
- 9 groundwater inflows (1) assuming uniform inflows within each reach. 7c. 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_{gw} within the 95% confidence interval. **7d.** Predicted vs. observed Cl concentrations. Shaded
 - field is the range resulting from varying groundwater Cl concentrations within the 95% confidence
- 15 interval.

- 16 **Figure 8.** Calculated and observed ²²²Rn activities for February 2015 resulting from varying individual
- parameters in Eq. (1). In all cases the new parameters result in significant overestimation of ²²²Rn
- activities in many reaches and are unlikely to be realistic. Site abbreviations as for Fig. 1.
- 19 Figure 9. Calculated streamflows resulting from groundwater inflows for the sampling rounds
- 20 excluding February 2015 estimated without parafluvial flow. Aside from the high flow periods
- 21 (September 2010 and July 2014) the calculated increase in streamflow exceeds the observed
- streamflow at Stratford and Chinns Bridge. Site abbreviations as for Fig. 1.

Table 1. Summary of parameters used in ²²²Rn mass balance

Symbol	Parameter	Units	Comments
Q	Streamflow	m³ day ⁻¹	
Ε	Evaporation	m day⁻¹	
X	Distance downstream	m	
W	Stream width	m	
d	Stream depth	m	
ν	Stream velocity	m day⁻¹	
Cgw, Cr, Ch, Cp	²²² Rn activities in groundwater, river, hyporheic zone, parafluvial zone	Bq m ⁻³	
Cin	²²² Rn activity of water entering the hyporheic or parafluvial zone	Bq m ⁻³	
k	Gas-transfer coefficient	day ⁻¹	
λ	Decay constant	0.181 day ⁻¹	
1	Groundwater inflows	m³ m ⁻¹ day ⁻¹	Eq. (2)
Fh	²²² Rn flux from hyporheic zone	Bq m ⁻¹ day ⁻¹	Eq. (5)
F_p	²²² Rn flux from parafluvial zone	Bq m ⁻¹ day ⁻¹	Eq. (6)
γ	²²² Rn emanation rate	Bq m ⁻³ day ⁻¹	Eq. (1)
Em	²²² Rn produced from sediments	Bq kg ⁻¹	
$ ho_{\!\scriptscriptstyle S}$	Sediment density	kg m ⁻³	
I_p	Inflows from parafluvial zone	m³ m ⁻¹ day ⁻¹	
t_h , t_p	Residence time in hyporheic or parafluvial zone	day	
ϕ	porosity		
V_p	Volume of sediments that parafluvial inflows interact with	m ³ m ⁻¹	
A_h , A_p	Cross-sectional area of the hyporheic or parafluvial zone	m²	$A_p = V_p$

Table 2. ²²²Rn emanation rates from floodplain sediments

Site ^a / Sample	E _m	γ	γ/λ
	Bq kg ⁻¹	Bq m ⁻³ day ⁻¹	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	18295
Wombat Flat 3	4.19	3075	16988
Wombat Flat 4	6.13	4492	24819
Valencia 1	3.95	2899	16016
Valencia 2	1.86	1362	7525
Degrees Lane 1	0.62	454	2500
Pearces Lane 1 Pearces Lane 2	0.62	454 2383	2506
Pearces Lane 2 Pearces Lane 3	3.25		13167
	1.41 2.63	1034	5722 10636
Pearces Lane 4		1925	
Pearces Lane 5	6.76	4952	27360
Pearces Lane 6	5.60	4107	22689
Pearces Lane 7	4.12	3018	16674
Pearces Lane 8	1.54	1127	6225
Stewarts Lane 1	3.41	2497	13797
Stewarts Lane 2	5.78	4239	23418
Stewarts Lane 3	3.08	2258	12475
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	18294
Stewarts Lane 8	4.58	3354	18530
Stewarts Lane 9	1.96	1434	7925
Stewarts Lane 10	5.09	3733	20622
Stewarts Lane 11	4.25	3119	17230
Stewarts Lane 12	3.68	2699	14910
Stewarts Lane 13	1.77	1294	7150
Stewarts Lane 14	2.89	2122	11723
Stratford 1	2.13	1563	8634
Stratford 2	0.66	482	2663
Stratford 3	3.01	2206	12190
Stratford 4	3.77	2762	15259
Stratford 5	0.39	288	1591
Stratford 6	1.24	911	5032
Stratford 7	2.00	1469	8117

σ		1197	6615
Mean		2308	12751
Strationa 15	0.01	431	3202
Stratford 13	0.81	491	3282
Stratford 12	3.13	2293	12667
Stratford 11	4.55	3334	18419
Stratford 10	1.01	738	4077
Stratford 9	0.91	668	3692
Stratford 8	2.71	1985	10965

a: sites on Fig. 2

















