

There were a few minor corrections on the final version of this paper that we have attended to.

The anonymous reviewer made the following minor comments and our responses are as indicated in **blue** (page and line numbers are the same as in the comments)

1. Radon does not need to be capitalised in the title. **“Radon” has been changed to “radon”**
2. Page 1, Line 14: insert word “... inflows would exceed ...” **Missing word “would” inserted**
3. P1, L18: Missing word “... alluvial (what?) in reaches ...”. Alluvial sediments? **Missing word “sediments” inserted**
4. P2, L18: “... distribution and quantity of groundwater ...” not “distribution of and quantity groundwater ...” **We changed this to “map and quantify groundwater inflows” to make it less ambiguous**
5. P3, L22: “... even where ...” not “... even if where ...” **Changed to “if where”**
6. P4, L11: “calculated” not “caluclated” **Corrected**

Additionally we have added our first names to the title page (Page 1, line 4) and thanked Luisa Stellato and the other anonymous referee in the acknowledgements (Page 25 lines 22-23).

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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1 **Abstract**

2 Understanding the location and magnitude of groundwater inflows to rivers is important for the  
3 protection of riverine ecosystems and the management of connected groundwater and surface water  
4 systems. This study utilises <sup>222</sup>Rn activities and Cl concentrations in the Avon River, southeast Australia  
5 to determine the distributions of groundwater inflows and to understand the importance of  
6 parafluvial flow on the <sup>222</sup>Rn budget. The distribution of <sup>222</sup>Rn activities and Cl concentrations implies  
7 that the Avon River contains alternating gaining and losing reaches. The location of groundwater  
8 inflows changed as a result of major floods in 2011 to 2013 that caused significant movement of the  
9 floodplain sediments. The floodplain of the Avon River comprises unconsolidated coarse-grained  
10 sediments with numerous point bars and sediment banks through which significant parafluvial flow is  
11 likely. The <sup>222</sup>Rn activities in the Avon River, which are locally up to 3690 Bq m<sup>-3</sup>, result from a  
12 combination of groundwater inflows and the input of water from the parafluvial zone that has high  
13 <sup>222</sup>Rn activities due to the <sup>222</sup>Rn emanations from the alluvial sediments. If the high <sup>222</sup>Rn activities  
14 were ascribed solely to groundwater inflows, the calculated net groundwater inflows would exceed  
15 the measured increase in streamflow along the river by up to 490% at low streamflows. Uncertainties  
16 in the <sup>222</sup>Rn activities of groundwater, the gas transfer coefficient, and the degree of hyporheic  
17 exchange cannot explain a discrepancy of this magnitude. The proposed model of parafluvial flow  
18 envisages that water enters the alluvial sediments in reaches where the river is losing and  
19 subsequently re-enters the river in the gaining reaches with flow paths of tens to hundreds of metres.  
20 Parafluvial flow is likely to be important in rivers with coarse-grained alluvial sediments on their  
21 floodplains and failure to quantify the input of <sup>222</sup>Rn from parafluvial flow will result in overestimating  
22 groundwater inflows to rivers.

1 **1. Introduction**

2 Quantifying groundwater inflows to streams and rivers is critical to understanding hydrogeological  
3 systems, protecting riverine ecosystems, and managing water resources (e.g., Winter, 1999;  
4 Sophocleous, 2002; Brodie et al., 2007). Groundwater inflows may form the majority of water in  
5 gaining rivers during periods of low streamflow, and riverine ecosystems are commonly sustained by  
6 groundwater inflows at those times (Kløve et al., 2011; Barron et al., 2012; Cartwright and Gilfedder,  
7 2015). Thus, understanding the distribution and magnitude of groundwater inflows is important for  
8 managing and protecting these commonly vulnerable ecosystems. Failure to understand groundwater  
9 contributions to rivers may also result in the double allocation of water resources (i.e., surface water  
10 and groundwater allocations might represent the same water). Documenting the distribution and  
11 quantity of groundwater inflows to rivers is also required for flood forecasting, understanding the  
12 impacts of contaminants on rivers, and assessing the potential impacts of climate or landuse changes  
13 on river systems.

14 In many catchments globally there are insufficient groundwater bores to understand the exchange  
15 between rivers and groundwater on anything other than a regional scale. In these cases geochemical  
16 tracers provide an alternative tool to understand groundwater-river interaction. Providing that  
17 groundwater and surface water have significantly different geochemistry, changes in the  
18 geochemistry of the river may be used to map and quantify groundwater inflows (e.g., Cook, 2013).

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19 Tracers such as major ions, stable isotopes, radioactive isotopes, and chlorofluorocarbons have been  
20 used to quantify groundwater inflows to rivers (e.g., Ellins et al., 1990; Genereux and Hemond, 1992;  
21 Négrel et al., 2001; Stellato et al., 2008; Cartwright et al., 2011, 2014; Cook, 2013; Bourke et al.,  
22 2014a,b). Geochemical tracers only quantify groundwater inflows, and while they are commonly used  
23 to determine the distribution of gaining and losing reaches, they do not quantify the magnitude of any  
24 groundwater outflows.

25 River water also interacts with the sediments beneath and adjacent to the streams in the hyporheic  
26 and parafluvial zones. The hyporheic zone comprises the sediments of the stream bed and sides

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

#### 11 **1.1. $^{222}\text{Rn}$ as a tracer of groundwater inflows**

12  $^{222}\text{Rn}$ , which is an intermediate isotope in the  $^{238}\text{U}$  to  $^{206}\text{Pb}$  decay series, is an important tracer for  
13 quantifying groundwater inflows to rivers.  $^{222}\text{Rn}$  has a half-life of 3.8 days and the activity of  $^{222}\text{Rn}$   
14 reaches secular equilibrium with its parent isotope  $^{226}\text{Ra}$  over 3 to 4 weeks (Cecil and Green, 2000).  
15 Because  $^{226}\text{Ra}$  activities in minerals in the aquifer matrix are several orders of magnitude higher than  
16 those in surface water, groundwater  $^{222}\text{Rn}$  activities are commonly two or three orders of magnitude  
17 higher than those of surface water (Cecil and Green, 2000). This makes  $^{222}\text{Rn}$  a viable tracer of  
18 groundwater inflows in catchments where the groundwater has similar major ion concentrations  
19 and/or stable isotope ratios to the river water. As  $^{222}\text{Rn}$  activities in rivers decline downstream from  
20 regions of groundwater inflow due to radioactive decay and degassing to the atmosphere (Ellins et al.,  
21 1990; Geneux and Hemond, 1992),  $^{222}\text{Rn}$  is also useful in determining locations of groundwater  
22 inflow, even if the inflows are not quantified.

23 The successful application of  $^{222}\text{Rn}$  to determine groundwater inflows, however, requires careful  
24 consideration of several processes and uncertainties.  $^{222}\text{Rn}$  activities in groundwater may be spatially  
25 or temporally heterogeneous (Cook et al., 2006; Mullinger et al., 2007; Unland et al., 2013; Yu et al.,

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1 2013; Cartwright et al., 2011; Atkinson et al., 2015). Additionally, while it is well established that the  
2 rate of  $^{222}\text{Rn}$  degassing increases with increasing river turbulence and decreasing river depth, it is  
3 difficult to reliably quantify the rate of degassing (Genereux and Hemond, 1992; Mullinger et al., 2007;  
4 Cook, 2013; Cartwright et al., 2014). Finally, in rivers that run through coarse alluvial sediments, water  
5 from the hyporheic or parafluvial zones may provide a source of  $^{222}\text{Rn}$  additional to groundwater  
6 inflow (Cook et al., 2006; Cartwright et al., 2014, Bourke et al., 2014a). As has been outlined in several  
7 studies, comparison of the calculated groundwater inflows from  $^{222}\text{Rn}$  with those made from other  
8 geochemical tracers or with streamflow measurements is a crucial test of the calculations (Cook et al.,  
9 2003, 2006; Mullinger et al., 2007, 2009; Cartwright et al., 2011, 2014; McCallum et al., 2012; Unland  
10 et al., 2013). Carrying out studies at baseflow conditions when most of the water contributing to the  
11 streams is from groundwater inflows allows for a comparison between the calculated groundwater  
12 inflows and the observed increase in streamflows, which in turn provides for a test of the parameters  
13 used in the  $^{222}\text{Rn}$  mass balance (Cartwright et al., 2014).

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## 14 **1.2. Objectives**

15 This paper examines groundwater-river interaction in the Avon River, southeast Australia, primarily  
16 using  $^{222}\text{Rn}$  as a tracer. The incised nature of the Avon River and the fact that it rarely ceases to flow  
17 has led to an assumption that it receives significant groundwater inflows (Gippsland Water, 2012).  
18 There has been little attempt, however, to quantify groundwater inflows or determine their  
19 distribution, and there are insufficient groundwater monitoring bores in the catchment to understand  
20 the relationship of groundwater to the river using hydraulic data. Understanding groundwater-river  
21 interaction is required to protect and manage the Avon River, especially in assessing the potential  
22 impacts of increased groundwater or surface water use.

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

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

## 10 **2. The Avon Catchment**

11 The Avon River is an unregulated river in the Gippsland Basin of southeast Australia (Fig. 1) that has a  
12 total catchment area of  $\sim 1830 \text{ km}^2$  (Cochrane et al., 1991; Department of Environment and Primary  
13 Industries, 2015). It drains the southern slopes of the Victorian Alps (maximum elevation in the  
14 catchment is 1634 m) and discharges into Lake Wellington, which is a coastal saline lake connected to  
15 the Southern Ocean. The highland areas represent  $\sim 30\%$  of the Avon catchment and are dominated  
16 by temperate native eucalyptus forest, whereas the majority of the plains representing  $\sim 70\%$  of the  
17 catchment have been cleared for agriculture, which includes dairying, sheep grazing, and vegetable  
18 production. The estimated population of the Avon catchment is  $\sim 4000$  with Stratford being the largest  
19 town (population  $\sim 2000$ ).

20 The highlands of the Victorian Alps comprise indurated Palaeozoic and Mesozoic igneous rocks and  
21 metasediments that only host groundwater flow in fractures or in near-surface weathered zones  
22 (Walker and Mollica, 1990; Cochrane et al., 1991). These rocks form the basement to the Tertiary and  
23 Quaternary sediments of the Gippsland Basin (Fig. 1). The shallowest regional aquifer within the Avon  
24 Catchment is the Pliocene to Pleistocene Haunted Hill Formation which comprises up to 40 m of  
25 interbedded alluvial sands and clays that have hydraulic conductivities between  $10^{-7}$  and  $10^{-5} \text{ m sec}^{-1}$

1 (Brumley et al., 1981; Walker and Mollica, 1990). Quaternary sediments that consist of coarse-grained  
2 sand and gravels interbedded with finer-grained silts occur mainly within the river valleys and have  
3 hydraulic conductivities of  $10^{-5}$  and  $10^{-2}$  m sec<sup>-1</sup> (Brumley et al., 1981; Walker and Mollica, 1990).

4 Average rainfall within the Avon catchment ranges from  $\sim 1.5$  m yr<sup>-1</sup> in the highlands to  $\sim 0.9$  m yr<sup>-1</sup> on  
5 the plains with most precipitation occurring in the austral winter (June to September) (Bureau of  
6 Meteorology, 2015). The Avon River displays strong seasonal flows with  $\sim 80\%$  of annual streamflow  
7 occurring during winter (Department of Environment and Primary Industries, 2015). This study  
8 focusses on the reaches of the Avon River located on the plains formed by the Gippsland Basin  
9 sediments that are upstream of tidal influence. Streamflow is measured continuously at three sites  
10 (The Channel, Stratford, and Chinns Bridge: Fig. 1). Total annual streamflow at Stratford between 1977  
11 and 2014 was between  $1.3 \times 10^7$  and  $9.0 \times 10^8$  m<sup>3</sup> yr<sup>-1</sup> (median =  $3.0 \times 10^8$  m<sup>3</sup> yr<sup>-1</sup>) and varied with total  
12 annual rainfall (Department of Environment and Primary Industries, 2015). The Avon River only ceases  
13 to flow during the summers of severe drought years (e.g., 1983) and experiences periodic floods  
14 during high rainfall periods (Fig. 2). Streamflow generally increases downstream at all times, except at  
15 very low flows when streamflow decreases between Stratford and Chinns Bridge. Valencia Creek and  
16 Freestone Creek are the main tributaries; both have streamflow measurements (Department of  
17 Environment and Primary Industries, 2015) and enter the Avon in the upper reaches of the studied  
18 section (Fig. 1).

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



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

11 Groundwater flows from the Victorian Alps to the coast (Hofmann and Cartwright, 2013: Fig. 1). Use  
12 of water from the Avon River and its tributaries for irrigation is up to  $8 \times 10^6 \text{ m}^3 \text{ yr}^{-1}$  ( $\sim 2.6\%$  of the annual  
13 median streamflow at Stratford); however, there is a prohibition on river water use when the  
14 streamflow at Stratford is  $< 10^4 \text{ m}^3 \text{ day}^{-1}$  (Gippsland Water, 2012).

### 15 **3. Methods**

#### 16 **3.1. Sampling**

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

1 in streamflow at Stratford over each sampling period was <5%). River samples were collected from  
2 0.5-1 m below the river surface using a manual collector mounted on a pole. Groundwater was  
3 sampled from bores installed on the river bank and floodplain at Stratford and Pearces Lane (Fig. 1)  
4 that have 1 to 3 m long screens. Water was extracted using an impeller pump set at the screened  
5 interval and at least 3 bore volumes of water were purged before sampling. Water was also extracted  
6 from the alluvial gravels at a number of locations along the Avon River during low flow periods either  
7 from open holes or from piezometers driven 1-2 m below the surface of the gravels.

### 8 **3.2. Analytical techniques**

9 Analytical techniques were similar to those in other studies (e.g. Unland et al., 2013; Yu et al., 2013;  
10 Cartwright et al., 2014). Cations (Tables A1, A2) were analysed on samples that had been filtered  
11 through 0.45µm cellulose nitrate filters and acidified to pH <2 using a ThermoFinnigan quadropole  
12 ICP-MS at Monash University. Anions (Tables A1, A2) were analysed on filtered unacidified samples  
13 using a Metrohm ion chromatograph at Monash University. The precision of major ion concentrations  
14 based on replicate analyses is 2-5%. A suite of anions and cations were measured; however, only Cl  
15 and Na are discussed in this study. <sup>222</sup>Rn activities in groundwater (Table A2) and surface water (Table  
16 A1) were determined using a portable radon-in-air monitor (RAD-7, DurrIDGE Co.) following methods  
17 described by (Burnett and Dulaiova, 2006) and are expressed in Bequerels per m<sup>3</sup> of water (Bq m<sup>-3</sup>).  
18 0.5 L of sample was collected by bottom-filling a glass flask and <sup>222</sup>Rn was subsequently degassed for  
19 5 minutes into a closed air loop of known volume. Counting times were 2 hours for surface water and  
20 20 minutes for groundwater. Typical relative precision based on repeat sample measurements in this  
21 and other studies (e.g., Cartwright et al., 2011, 2014) is <3% at 10,000 Bq m<sup>-3</sup> and ~10% at 100 Bq m<sup>-3</sup>.  
22 Forty four samples of river bed sediments from sites along the Avon River were collected in March,  
23 2014 and February 2015. <sup>222</sup>Rn emanation rates ( $\lambda$ ) from these were determined by sealing a known  
24 dry weight of sediment in airtight containers with water and allowing <sup>222</sup>Rn to accumulate  
25 (Lamontagne and Cook, 2007). Following 4-5 weeks incubation, by which time the rate of <sup>222</sup>Rn

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

$$5 \quad \gamma = \frac{E_m(1-\phi)\rho_s\lambda}{\phi} \quad (1)$$

6 (parameters summarised in Table 1).

### 7 **3.3. Radon mass balance**

8 Assuming that the atmosphere contains negligible radon, the change in  $^{222}\text{Rn}$  activities along a river is:

$$9 \quad Q \frac{dc_r}{dx} = I(c_{gw} - c_r) + wEc_r + F_h + F_p - kdw c_r - \lambda dw c_r \quad (2)$$

10 (modified from Mullinger et al., 2007; Cartwright et al., 2011; and Cook, 2013). In Eq. (2):  $Q$  is  
 11 streamflow;  $c_r$  and  $c_{gw}$  are the  $^{222}\text{Rn}$  activities in the river and groundwater, respectively;  $I$  is the  
 12 groundwater flux per unit length of river;  $E$  is the evaporation rate;  $x$  is distance along the river;  $w$  is  
 13 river width;  $d$  is river depth;  $F_h$  and  $F_p$  are the inputs of  $^{222}\text{Rn}$  resulting from exchange with the  
 14 hyporheic zone and inflows of parafluvial waters, respectively;  $k$  is the gas-transfer coefficient; and  $\lambda$   
 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 Cl 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:

$$19 \quad \frac{dQ}{dx} = I - Ew \quad (3).$$

20 The  $^{222}\text{Rn}$  activity in the hyporheic zone waters ( $c_h$ ) is governed by the  $^{222}\text{Rn}$  activity of the water  
 21 flowing into the hyporheic zone ( $c_{in}$ ), the  $^{222}\text{Rn}$  emanation rate  $\gamma$ , and the residence time  $t_h$ :

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

(Hoehn et al., 1992; Hoehn and Cirpka, 2006) (Fig. 3a). An identical expression relates the  $^{222}\text{Rn}$  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  $^{222}\text{Rn}$  activities. Cartwright et al. (2014) discussed using the concentration of a conservative ion such as Cl to estimate the degree of mixing within the alluvial sediments to estimate  $c_{in}$ . Assuming that all the water entering the hyporheic zone subsequently re-enters the river, the  $^{222}\text{Rn}$  flux from the hyporheic zone ( $F_h$ ) is:

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

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  $^{222}\text{Rn}$  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  $\gamma$  (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  $^{222}\text{Rn}$  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 that case,  $F_p$  is given by a similar expression to that which accounts for the input of  $^{222}\text{Rn}$  due to groundwater inflows:

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

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  
2 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  $m^3$  per m length of river) is governed by  $I_p$ ,  $t_p$  and  $\phi$ . 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 ( $A_p$ ):

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

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  $^{222}\text{Rn}$  degassing from rivers.  
11 Firstly, as degassing involves diffusion of  $^{222}\text{Rn}$  through the boundary layer at the river surface, the  
12 stagnant film model yields a gas transfer velocity as  $D/z$  (which is closely related to  $k$ ), where  $z$  is the  
13 thickness of the boundary layer at the water surface (Ellins et al., 1990; Stellato et al., 2008).  $z$  and by  
14 extension  $D/z$  can be calculated from differences in river  $^{222}\text{Rn}$  concentrations in losing reaches. The  
15 gas transfer coefficient  $k$  may be estimated in a similar way from the change in  $^{222}\text{Rn}$  activities in losing  
16 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  
18 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  $^{222}\text{Rn}$  activities requires that the  
20  $^{222}\text{Rn}$  contributed from the hyporheic or parafluvial zones is quantified, and that there are no inflows  
21 of water from tributaries that may increase or decrease  $^{222}\text{Rn}$  activities. Since  $k$  values are typically  
22 calculated from these methods for a few specific well-understood river reaches, it is possible that they  
23 are not valid for all river reaches.

1 It is also possible to measure  $k$  directly by using introduced gas tracers such as SF<sub>6</sub> (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 ( $v$ ) and depths. The  
8 commonly used O'Connor and Dobbins (1958) and Negulescu and Rojanski (1969) gas transfer  
9 equations as modified for <sup>222</sup>Rn are:

$$10 \quad k = 9.301 \times 10^{-3} \left( \frac{v^{0.5}}{d^{1.5}} \right) \quad (8)$$

$$11 \quad k = 4.87 \times 10^{-4} \left( \frac{v}{d} \right)^{0.85} \quad (9)$$

12 (Mullinger et al., 2007). As discussed by Genereux and Hemond (1992), however, there are numerous  
13 formulations that can yield very different estimates of  $k$  for the same flow conditions and some  
14 independent assessment of  $k$  (for example by matching the predicted and observed decline in <sup>222</sup>Rn  
15 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  $1.38 \times 10^8$   
19  $\text{m}^3 \text{ day}^{-1}$  (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  
22 streamflows at Stratford during the sampling rounds ranged from  $10,670 \text{ m}^3 \text{ day}^{-1}$  to  $88,800 \text{ m}^3 \text{ day}^{-1}$   
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 m<sup>3</sup> day<sup>-1</sup> at The Channel, 23,090 m<sup>3</sup> day<sup>-1</sup> at Stratford, and 25,780 m<sup>3</sup> day<sup>-1</sup> at Chinns Bridge. Inflows  
3 from Valencia Creek and Freestone Creek in February 2015 were 2410 m<sup>3</sup> day<sup>-1</sup> and 600 m<sup>3</sup> day<sup>-1</sup>,  
4 respectively (Department of Environment and Primary Industries, 2015).

#### 5 **4.2. River Geochemistry**

6 Figure 4a shows the <sup>222</sup>Rn activities of the Avon River for the six sampling campaigns. There are several  
7 distinct zones of elevated <sup>222</sup>Rn activities, notably at Wombat Flat (4.8 km) where <sup>222</sup>Rn activities are  
8 up to 2040 Bq m<sup>-3</sup> and between Bushy Park and Schools Lane (16.3 to 25.3 km) where <sup>222</sup>Rn activities  
9 are up to 3690 Bq m<sup>-3</sup>. Zones of lower <sup>222</sup>Rn activities in the upper reaches occur at Smyths Road (8.1  
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 <sup>222</sup>Rn  
12 activities that generally decline downstream. The position of the highest <sup>222</sup>Rn activities changed in  
13 the periods prior to and post the 2011 to 2013 floods. In March 2014 and February 2015 the highest  
14 <sup>222</sup>Rn activities were at Bushy Park (16.3 km), whereas this site had relatively low <sup>222</sup>Rn activities in  
15 February 2009 and April 2010 when the highest <sup>222</sup>Rn activities were at Pearces Lane (20.0 km). The  
16 distribution of <sup>222</sup>Rn activities in the detailed sampling campaign in February 2015 is similar to that at  
17 other periods of low to moderate streamflow (e.g. March 2014). The lowest overall <sup>222</sup>Rn activities  
18 were recorded during the periods of highest flow (September 2010 and July 2014).

19 EC values and Cl concentrations generally increase downstream from 54 to 131 μS cm<sup>-1</sup> and 4 to 10  
20 mg l<sup>-1</sup> at Browns (0.0 km) to as high as 934 μS cm<sup>-1</sup> and 98 mg l<sup>-1</sup> at Chinns Bridge (49.7 km) (Table A1,  
21 Fig. 4b). Cl concentrations at low streamflows in March 2014 were generally higher (up to 98 mg l<sup>-1</sup>)  
22 than in the other sampling campaigns, while Cl concentrations were <20 mg l<sup>-1</sup> during the highest  
23 streamflows in September 2010. A marked increase in EC values and Cl concentrations occurs  
24 downstream of Smyths Road (8.1 km) in the reaches where <sup>222</sup>Rn activities are highest at low

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

### 3 **4.3. Groundwater Geochemistry**

4 Groundwater from the near-river bores at Pearces Lane and Stratford has  $^{222}\text{Rn}$  activities that vary  
5 from 480 to 28,980  $\text{Bq m}^{-3}$  (Table A2). There is some variation in  $^{222}\text{Rn}$  activities in individual bores  
6 between the sampling rounds with relative standard deviations between 6 and 34%. The mean value  
7 of all groundwater  $^{222}\text{Rn}$  activities ( $n = 26$ ) is 12,890  $\text{Bq m}^{-3}$ . Bore 5 at Pearces Lane is immediately  
8 adjacent to the Avon River and possibly samples water from the parafluvial zone rather than  
9 groundwater. Excluding data from that bore, the mean value of  $^{222}\text{Rn}$  activities is 13,830  $\text{Bq m}^{-3}$  ( $n =$   
10 24) with a standard error of 1273  $\text{Bq m}^{-3}$  and a 95% confidence interval (calculated using the  
11 Descriptive Statistics tool in Excel 2010 which assumes that the data follows a t-distribution) of 2634  
12  $\text{Bq m}^{-3}$ . EC values of groundwater from the bores at Pearces Land and Stratford are between 100 and  
13 680  $\mu\text{S cm}^{-1}$  and Cl concentrations range from 46 to 147  $\text{mg l}^{-1}$  with a mean value of  $79 \pm 34 \text{ mg l}^{-1}$  ( $n =$   
14 16) (Table A2). If Bore 5 at Pearces Lane is again excluded the mean Cl concentration is  $87 \pm 28 \text{ mg l}^{-1}$   
15 ( $n = 14$ ) with a standard error of 8  $\text{mg l}^{-1}$  and a 95% confidence interval of 16  $\text{mg l}^{-1}$ . These Cl  
16 concentrations are typical of groundwater elsewhere in the Avon valley and neighbouring catchments  
17 (Department of Environment and Primary Industries, 2015).

### 18 **4.4. Geochemistry of water from the alluvial gravels**

19 EC values of water within the gravels further than 1 to 2 m from the edge of the river are between 120  
20 and 550  $\mu\text{S cm}^{-1}$  ( $n = 52$ ) (Fig. 5b); these EC values are higher than those of the adjacent river water  
21 but similar to those of the groundwater. Only water extracted from within 1 to 2 m from the river had  
22 EC values similar to the river and in some cases the EC of water from the gravels within a few  
23 centimetres of the river edge was higher than the adjacent river.  $^{222}\text{Rn}$  activities of these samples were  
24 between 7000 and 28,000  $\text{Bq m}^{-3}$  ( $n = 21$ ) (Fig. 5a), which are also significantly higher than the  $^{222}\text{Rn}$



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. <sup>222</sup>Rn Emanation Rates**

4 <sup>222</sup>Rn emanation rates were determined via Eq. (1). The matrix density was assigned as 2700 kg m<sup>-3</sup>,  
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  $\gamma$  values range from 288 to 4950 Bq m<sup>-3</sup> with a mean value of 2308±1197 Bq m<sup>-3</sup> (n = 44) and a standard  
8 error of 183 Bq m<sup>-3</sup>. The mean emanation rates for sediments from the different sites vary between  
9 1484 and 3461 Bq m<sup>-3</sup>; however, there is no systematic variation with position in the catchment. The  
10 relative variability in  $\gamma$  between the sediments is similar to that reported elsewhere (e.g., Bourke et  
11 al., 2014a; Cartwright et al., 2014). <sup>222</sup>Rn activities of water in equilibrium with the sediments are given  
12 by  $\gamma/\lambda$  (Cecil and Green, 2000), and the mean  $\gamma/\lambda$  value is 12,751±6615 Bq m<sup>-3</sup> with a standard error  
13 of 1009 Bq m<sup>-3</sup>. These  $\gamma/\lambda$  values are not significantly different (p ~0.5) to the measured <sup>222</sup>Rn activities  
14 of the groundwater.

### 15 **5. Discussion**

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

#### 23 **5.1. Distribution of groundwater inflows**

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

1 by groundwater, the  $^{222}\text{Rn}$  activities from these sampling campaigns are most useful in understanding  
2 the distribution of groundwater inflows. The region between Smyths Road and Ridleys Lane (8.1 to  
3 23.0 km) where  $^{222}\text{Rn}$  activities increase and remain high (Fig. 4a), especially at lower streamflows, and  
4 where there is a marked increase in Cl concentrations (Fig. 4b) is interpreted as receiving major  
5 groundwater inflows. This section of the Avon River is incised up to 4 m below the floodplain which  
6 likely produces steep hydraulic head gradients that result in groundwater discharge on the floodplain  
7 and into the river. There are also groundwater seeps and patches of perennial water tolerant  
8 vegetation at the edge of the floodplain in this area. The reaches between Browns and Wombat Flat  
9 (0.0 to 4.8 km) and Stewarts Lane and Stratford (30.1 to 35.1 km) are also characterised by high  $^{222}\text{Rn}$   
10 activities and are again interpreted as receiving groundwater inflows.

11 The reaches between Wombat Flat and Smyths Road (4.8 to 8.1 km), Ridleys Lane and Stewarts Lane  
12 (23.0 to 30.1 km), and Knobs Reserve and Chinns Bridge (37.8 to 49.7 km) where there is a gradual  
13 decline in  $^{222}\text{Rn}$  activities and little change in Cl concentrations (Fig. 4) are interpreted as either being  
14 losing or receiving minor groundwater inflows. The landscape is flatter and the river less incised in  
15 these areas which results in lower hydraulic gradients and consequently less groundwater inflows to  
16 the river.

17 The difference in the location of the highest  $^{222}\text{Rn}$  activities between the sampling campaigns that  
18 were conducted before and after the major floods (i.e., pre 2011 vs. post 2013) indicates that the  
19 locations of groundwater inflows changed. The major floods changed the location of pools and  
20 sediment banks on the Avon River and caused scouring, which would change the relationship of the  
21 river to the groundwater.

## 22 **5.2. Quantifying Groundwater Inflows**

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

1  $\text{m}^{-3}$ , which is consistent both with the measured  $^{222}\text{Rn}$  activities of groundwater (Table A2) and the  
2 calculated  $^{222}\text{Rn}$  activities of water in equilibrium with the alluvial sediments.

3 The flux of  $^{222}\text{Rn}$  from the hyporheic zone was estimated from Eq. (5) using the mean  $\gamma$  value of 2300  
4  $\text{Bq m}^{-3} \text{ day}^{-1}$  (Table 2), a porosity of 0.4 (which is appropriate for coarse-grained unconsolidated  
5 sediments), and a value for  $C_m$  that is the  $^{222}\text{Rn}$  activity of the river in that reach. The residence time of  
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  
15 downstream. The Cl and  $^{222}\text{Rn}$  data from the water contained within the gravels (Fig. 5) are interpreted  
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  $^{222}\text{Rn}$   
26 activities in the river in a similar manner to inflowing groundwater. However, because it represents

1 water that originated from the river, the inflows from the parafluvial zone do not increase the overall  
2 streamflow. If the parafluvial zone water is in secular equilibrium with the sediments,  $c_p \sim 12,700 \text{ Bq}$   
3  $\text{m}^{-3}$  (Table 2).

4 Average evaporation rates in southeast Australia in February to April are  $3 \times 10^{-3}$  to  $5 \times 10^{-3} \text{ m day}^{-1}$   
5 (Bureau of Meteorology, 2015) and a value of  $4 \times 10^{-3} \text{ m day}^{-1}$  was adopted. Average river width and  
6 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,  
7 respectively, for the rest of the river

8 The gas transfer coefficient was estimated from the decline in  $^{222}\text{Rn}$  activities between Ridleys Lane  
9 and Schools Lane (23.0 to 25.3 km) (Fig. 4a). This approach estimates the net  $kdwc$  term and  $k$  was  
10 estimated as  $0.3 \text{ day}^{-1}$  using the measured widths, depths, and  $^{222}\text{Rn}$  concentrations. This requires that  
11 this is a losing stretch of the river, so that there are no groundwater or parafluvial inflows. That Cl  
12 concentrations do not increase over this stretch of river (Fig. 4b) are consistent with it being losing. A  
13  $k$  value of  $0.3 \text{ day}^{-1}$  is at the lower end of estimates of Rn gas transfer coefficients (Genereux and  
14 Hemond, 1992; Cook et al., 2003, 2006; Cartwright et al., 2011, 2014; Atkinson et al., 2013; Unland et  
15 al., 2013; Yu et al., 2014). However, as the Avon River is dominated by slow-flowing pools, degassing  
16 rates are expected to be low.

17 Groundwater inflows were calculated from the  $^{222}\text{Rn}$  activities by solving Eq. (2) using a finite  
18 difference approach in a spreadsheet with a distance step of 10 m (the use of smaller or larger distance  
19 steps does not significantly change the results). The streamflow at The Channel gauge was used as the  
20 initial streamflow and  $Q$  was increased after each distance step via Eq. (3). The calculations estimated  
21 the values of  $I$  and  $I_p$  in each reach by matching the calculated and measured  $^{222}\text{Rn}$  activities along the  
22 river with the additional constraint that the total groundwater inflows cannot exceed the net increase  
23 in streamflow between the Channel gauge and the gauges at Stratford and Chinns Bridge (Fig. 1). Since  
24 there are few streamflow measurements, the calculations assumed that the ratio of  $I$  to  $I_p$  was the  
25 same in all gaining reaches of the river.

1 Assuming that in the gaining reaches there are 50% parafluvial inflows and 50% groundwater inflows  
2 allows both the  $^{222}\text{Rn}$  variations and the increase in streamflow to be accounted for (Fig. 6a).  
3 Calculated groundwater and parafluvial inflows are highest in the reaches between Smyths Road and  
4 Pearces Lane (8.1 to 20.0 km) (Fig. 6b), which is the region where Cl 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 to  $2.5 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$  of which  
7 groundwater inflows are  $\sim 1.26 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$ .

8 There is no process in the parafluvial or hyporheic zones other than mixing with groundwater that  
9 increases the Cl concentrations of the through-flowing water. Thus the Cl concentrations in the river  
10 reflect only the groundwater inflows and in theory it would be possible to use Cl 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 Cl concentrations results in large  
13 uncertainties. The change in Cl concentrations (Fig. 6d) was calculated from the groundwater inflows  
14 assuming that groundwater has a Cl concentration of  $85 \text{ mg l}^{-1}$ . The calculated Cl concentrations are  
15 slightly higher than those observed, but if the Cl 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,  
18  $c_p$  will be lower and the inflows from the parafluvial zone ( $I_p$ ) required to produce a given flux of  $^{222}\text{Rn}$   
19 ( $F_p$ ) increases (Fig. 3). For example, if  $c_r = 2300 \text{ Bq m}^{-3}$ , which is a typical value in many of the reaches  
20 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  $I_p$   
21  $= 1 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$ ,  $F_p = 10,400 \text{ Bq m}^{-1} \text{ day}^{-1}$  (Eq. 6). If  $\gamma = 2300 \text{ Bq m}^{-3} \text{ day}^{-1}$ ,  $c_p$  is 2487, 4023 and 11,004  
22  $\text{Bq m}^{-3}$  where  $t_p$  is 0.1, 1, and 10 days, respectively. To produce a value of  $F_p$  of  $10,400 \text{ Bq m}^{-1} \text{ day}^{-1}$   
23 requires  $I_p \sim 58 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$  for  $t_p = 0.1$  days,  $\sim 6.0 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$  for  $t_p = 1$  day, and  $\sim 1.2 \text{ m}^3 \text{ m}^{-1} \text{ day}^{-1}$  for  
24  $t_p = 10$  days. For  $t_p > 30$  days the system is close to secular equilibrium and  $c_p$  and  $I_p$  are near constant  
25 (Fig. 3). The cross-sectional area of the parafluvial zone  $A_p$  required to accommodate these parafluvial

1 flows with  $\phi = 0.4$  and  $t_p$  between 0.1 and 100 days is between 14 and 250 m<sup>2</sup> (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.

### 5 **5.3. Uncertainties and sensitivity**

6 The proposal that parafluvial flow is important in the Avon River is consistent with the local  
7 hydrogeology and allows both the <sup>222</sup>Rn and net increase in streamflow to be reproduced. The  
8 conclusion that inflows of parafluvial zone waters only occur in the gaining reaches is justifiable as the  
9 conditions required for groundwater inflows (gaining river with steep hydraulic gradients and high-  
10 hydraulic conductivity sediments) will likely drive the return of parafluvial waters to the river. By  
11 contrast losing reaches are likely to be where the water enters the parafluvial sediments. Given the  
12 multiple parameters in Eq. (2) and their inherent uncertainties, however, consideration needs to be  
13 given to whether both the <sup>222</sup>Rn activities and the increases in streamflow can be accounted for  
14 without parafluvial inflows being a significant source of <sup>222</sup>Rn.

15 Matching the <sup>222</sup>Rn profile along the Avon River using the parameters discussed above but without  
16 input of <sup>222</sup>Rn from parafluvial zone would require net groundwater inflows of 28,300 m<sup>3</sup> day<sup>-1</sup>.  
17 However, these inflows exceed the measured increase in streamflow between The Channel and  
18 Chinns Bridge of 15,500 m<sup>3</sup> day<sup>-1</sup> by 180% (Fig. 7a). The February 2015 sampling round took place at  
19 the end of summer when the small ephemeral tributaries were dry and there was no overland flow;  
20 however, there were still flows from Valencia Creek and Freestone Creek of 1,410 m<sup>3</sup> day<sup>-1</sup> and 200 m<sup>3</sup>  
21 day<sup>-1</sup>, respectively. If these were included, the discrepancy between the calculated and observed  
22 streamflow increases. The calculated Cl concentrations are also higher than observed (Fig. 7d),  
23 although given the uncertainty in groundwater Cl concentrations, the discrepancy is not large.

24 In common with most studies, the calculations assumed that the groundwater inflows are uniform  
25 along a particular reach. However, because <sup>222</sup>Rn is lost from rivers by degassing and decay, lower

1 groundwater inflows are required to replicate the observed  $^{222}\text{Rn}$  activities if the groundwater inflows  
2 occur immediately upstream of a sampling point (Cook, 2013). Even assigning the groundwater inflows  
3 in each reach to the 10 m section upstream of the measurement point still results in the calculated  
4 streamflow overestimating the measured streamflow (Fig. 7c). The predicted  $^{222}\text{Rn}$  activities in the  
5 river in this case are also not realistic (Fig. 7a).

6 The evaporation term in Eq. (2) is one to two orders of magnitude lower than most of the other terms  
7 and errors in the assumed evaporation rate have little influence on the calculations. The main  
8 parameter impacting calculated groundwater inflows is the  $^{222}\text{Rn}$  activity of groundwater (Cartwright  
9 et al., 2011; Cook, 2013). Allowing  $c_{\text{gw}}$  to vary within the 95% confidence interval of the groundwater  
10  $^{222}\text{Rn}$  activities ( $\pm 2600 \text{ Bq m}^{-3}$ ) makes little difference to the discrepancy between the calculated and  
11 observed increase in streamflow (Fig. 7c). Increasing  $c_{\text{gw}}$  to  $27,000 \text{ Bq m}^{-3}$  allows both the  $^{222}\text{Rn}$  profile  
12 and the observed increase in streamflow between The Channel and Chinns Bridge to be reproduced  
13 without the requirement for the input of  $^{222}\text{Rn}$  from the parafluvial zone (Fig. 8). However, there is no  
14 known groundwater in the Avon catchment with such high  $^{222}\text{Rn}$  activities and these activities are far  
15 higher than would be in equilibrium with the alluvial sediments that comprise the near-river aquifer  
16 lithologies. Hence, it is considered not possible that groundwater  $^{222}\text{Rn}$  activities could be this high.

17 There is uncertainty in the gas transfer coefficient.  $k$  was estimated assuming that the Avon River  
18 contains losing reaches; if those reaches were actually gaining then this methodology underestimates  
19  $k$ . However, increasing  $k$  from  $0.3 \text{ day}^{-1}$  increases the calculated groundwater inflows, which increases  
20 the discrepancy between the observed and calculated increases in streamflow.  $k$  estimated from Eqs  
21 (8) and (9) ranges between  $0.1$  and  $0.3 \text{ day}^{-1}$ . Using  $k = 0.1 \text{ day}^{-1}$  produces net groundwater inflows  
22 that more closely match the observed increase in streamflow. However, adopting  $k = 0.1 \text{ day}^{-1}$  results  
23 in the calculated  $^{222}\text{Rn}$  activities in a number of reaches being overestimated (Fig. 8). This is because  
24 even assuming no groundwater inflows into those reaches, the loss of  $^{222}\text{Rn}$  by degassing is insufficient  
25 to explain the observed decrease in  $^{222}\text{Rn}$ . Such a poor correspondence between predicted and  
26 observed  $^{222}\text{Rn}$  activities implies problems with the adopted variables.

1 While there are uncertainties in  $c_h$ , the main uncertainty in the contribution of hyporheic exchange to  
2 the  $^{222}\text{Rn}$  budget is the dimensions of the hyporheic zone. Increasing  $F_h$  also reduces the calculated  
3 groundwater inflows. Using the same emanation rates, residence times, and porosities but assigning  
4 a thickness of the hyporheic zone of 50 cm, increases  $F_h$  and produces groundwater inflows that  
5 broadly match the increase in streamflow. However, the higher values of  $F_h$  again result in a poor fit  
6 between predicted and observed  $^{222}\text{Rn}$  activities (Fig. 8).

7 Because the error in  $\lambda$  is negligible and the evaporation term is much smaller than the other terms, it  
8 is generally possible to produce identical trends in  $^{222}\text{Rn}$  activities with different combinations of  $k$  and  
9  $F_h$  (Cartwright et al., 2014). If  $F_h$  is calculated assuming a 50 cm thick hyporheic zone, adopting  $k = 0.6$   
10  $\text{day}^{-1}$  reproduces the observed  $^{222}\text{Rn}$  activities. Similarly, if  $k = 0.1 \text{ day}^{-1}$  a match between the observed  
11 and the predicted  $^{222}\text{Rn}$  activities is achieved with no hyporheic exchange ( $F_h = 0$ ). However, these  
12 combinations of parameters again result in estimated net groundwater inflows that exceed the  
13 measured increase in streamflow.

14 There is an unknown error in the streamflow measurements, but it is unlikely to be sufficient to explain  
15 the gross overestimation of groundwater inflows. Uncertainties in the assumed river widths and  
16 depths will also impact the calculations. Specifically, reducing the width or depth decreases the  
17 magnitude of the last two terms on the right-hand-side of Eq. (2), which in turn reduces  $I$ . If widths  
18 were reduced by 50% (an unrealistic error), net groundwater inflows broadly match the increase in  
19 streamflow. However, this again results in  $^{222}\text{Rn}$  activities being overestimated in many reaches (Fig.  
20 8). Increasing  $k$  to  $0.65 \text{ day}^{-1}$  would allow the  $^{222}\text{Rn}$  activities to be predicted using these lower widths  
21 but again results in the estimated net groundwater inflow exceeding the measured increase in  
22 streamflow. Overall it is concluded that there are no combination of parameters that can reproduce  
23 both the observed  $^{222}\text{Rn}$  activities and streamflows without incorporating parafluvial flow.

24 It would be possible to explain the observed  $^{222}\text{Rn}$  activities and streamflows if there were losing  
25 reaches in the Avon River through which significant volumes of river water were lost to the underlying



1 aquifers and, unlike parafluvial flow, this water did not subsequently return to the river. For this  
2 scenario to be valid, approximately 50% of the groundwater inflows would have to be lost from the  
3 river in these losing reaches in February 2015. The reaches between 25 and 30 km are interpreted as  
4 losing. However, these reaches do not dry up even during prolonged drought (Gippsland Water, 2012),  
5 and all reaches of the river were flowing during the 2009 sampling campaign (which had the lowest  
6 streamflows). Also while streamflows were not measured, such a major reduction in streamflow over  
7 such a short distance would be apparent in the field. Likewise, significant pumping of water from the  
8 river would also reduce streamflows. While the surface water is licenced for use, streamflow during  
9 February 2009 and March 2014 was below the minimum levels where that is permitted and the  
10 streamflows in April 2010 and February 2015 were such that use would be restricted; hence, large-  
11 scale pumping of river water at those times is unlikely.

#### 12 **5.4. Other sampling campaigns**

13 The predicted distribution of groundwater inflows in February 2009, April, 2010, and March 2014  
14 when streamflows were low to moderate are similar to those in February 2015 (Fig. 4). Due to the  
15 lower number of sampling points, it is difficult to calculate groundwater inflows with certainty. The  
16 net groundwater inflows calculated using the same parameters as above but ignoring parafluvial flows  
17 are between 15,900 and 21,700 m<sup>3</sup> day<sup>-1</sup>, respectively (Fig. 9), which are up to 490% of the measured  
18 increases in streamflow between The Channel and Chinns Bridge. Again propagating the likely  
19 uncertainties in the parameters through Eq. (2) cannot resolve this discrepancy, implying that the  
20 inflows of water from the parafluvial zone must be a significant part of the <sup>222</sup>Rn budget.

21 At the higher streamflows there will likely be significant inputs to the river from overland flow or  
22 interflow; hence, it is not possible to use the comparison between calculated groundwater inflows  
23 and the net increase in streamflow to independently test for the input of <sup>222</sup>Rn from the parafluvial  
24 zone. For example, without incorporating parafluvial flow, the net groundwater inflows using widths  
25 of 15 m upstream of Wombat Flat and 25 m elsewhere, depths of 1.25 m upstream of Wombat Flat and

1 1.6 m elsewhere,  $k = 0.3 \text{ day}^{-1}$ ,  $F_h$  adjusted for the higher river widths are  $32,100 \text{ m}^3 \text{ day}^{-1}$  (September  
2 2010) and  $44,600 \text{ m}^3 \text{ day}^{-1}$  (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 **6. Conclusions**

7 The variation in  $^{222}\text{Rn}$  activities and Cl concentrations clearly define the reaches of the Avon River that  
8 are gaining. The distribution of  $^{222}\text{Rn}$  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  
13 that parafluvial flow was a significant process in controlling the  $^{222}\text{Rn}$  activities of the river. However,  
14 this proposition is difficult to definitively test or explore in more detail. The groundwater and  
15 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  $^{222}\text{Rn}$  mass balance calculations. Unlike hyporheic  
19 exchange, which occurs in all stretches, parafluvial inflows are likely to dominantly occur in gaining  
20 reaches augmenting the groundwater inflows.

21 Theoretically, a conservative tracer such as Cl 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  
24 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  
2 streamflows where the majority of inflows into the river are likely to be from groundwater. While this  
3 might appear redundant in terms of determining the water balance, it does provide for a test of  
4 assumptions and parameterisation. It would be possibly to interpret the changes to  $^{222}\text{Rn}$  activities  
5 during the periods of higher streamflow as being solely due to groundwater inflows because the net  
6 groundwater inflows are lower than the net increases in streamflow (Fig. 9). However, it is likely that  
7 there is groundwater and parafluvial inflows at all times, in which case calculated groundwater inflows  
8 will be overestimated.

#### 9 **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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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 = Pearce 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  
10 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  $^{222}\text{Rn}$  activity in the parafluvial or hyporheic zone ( $c_p$  or  $c_h$ ) with residence  
15 time ( $t_p$  or  $t_h$ ) and  $^{222}\text{Rn}$  emanation rate ( $\gamma$ ) (Eq. 3). **3b.** Variation in the water flux from the parafluvial  
16 zone ( $I_p$ ) with the flux of  $^{222}\text{Rn}$  from the parafluvial zone ( $F_p$ ) and  $t_p$  (Eq. 5). In both cases  $c_r = c_{in} = 1000$   
17  $\text{Bq m}^{-3}$ .

18 **Figure 4.** Downstream variations in  $^{222}\text{Rn}$  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  
20 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.

22 **Figure 5a.** Variations in  $^{222}\text{Rn}$  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  
24 the Avon River (Data from Tables A1 and A2).



1 **Figure 6a.** Calculated and observed  $^{222}\text{Rn}$  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  $^{222}\text{Rn}$  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 ( $I$ ) assuming uniform inflows within each reach. **7c.** Calculated increase in  
10 streamflow from groundwater inflows (Eq. 2). Both uniform groundwater inflow within each reach  
11 and the situation where groundwater enters the river immediately upstream of the measurement  
12 point overestimate the measured streamflow. Shaded area is the range of streamflow resulting from  
13 varying  $c_{gw}$  within the 95% confidence interval. **7d.** Predicted vs. observed Cl concentrations. Shaded  
14 field is the range resulting from varying groundwater Cl concentrations within the 95% confidence  
15 interval.

16 **Figure 8.** Calculated and observed  $^{222}\text{Rn}$  activities for February 2015 resulting from varying individual  
17 parameters in Eq. (1). In all cases the new parameters result in significant overestimation of  $^{222}\text{Rn}$   
18 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  
22 streamflow at Stratford and Chinns Bridge. Site abbreviations as for Fig. 1.