Summary of Revisions

We carried out most of the changes that we proposed in our response to the reviewers. We summarise these below in **green** linked to the original comments and have also included the marked manuscript. Overall we have taken the vast majority of the comments on board in revising the paper, especially in the request to reorder the material in the paper (i.e. collating the equations and presenting the mass Rn balance with the parafluvial inflows included first).

Response to comments on "Using geochemical tracers to distinguish groundwater and parafluvial inflows in rivers (the Avon Catchment, SE Australia)" by Cartwright and Hofmann.

We thank the two reviewers for their constructive comments on the paper and consider that we can use these constructively to improve the clarity of the paper. Our responses to the specific comments are outlined below (in **blue**, references are those in the original paper or comments except where indicated).

Reviewer #1

This reviewer is thanked for a very comprehensive and detailed review. Part of the substantive comments raised by the reviewer relate to the paper organisation. Overall the reviewer seems to be requesting that the paper is written in the format that they would have used. While the suggested format is logical, it should be noted that other papers in the literature (including in HESS) follow a variety of formats and the second reviewer was happy with the structure of the paper.

Some of the suggested reorganisation, specifically aggregating the equations into a separate section, is reasonably straightforward. Since we do not develop the equations, having them in a single place would probably make the introduction and discussion easier to follow as these sections are currently rather long.

We presented the parafluvial flow as a conclusion that arose from not otherwise being able to reconcile the Rn and streamflow data. The reason that we chose that approach is that it reflects the way that the study developed. The alternative is (as suggested by this reviewer) to start off from the standpoint that parafluvial flow is likely in in this environment, incorporate it into the Rn mass balance from the outset, and then to show that the Rn mass balance is less viable without it. This follows the logic that parafluvial flow is a known process but it has been little-studied in terms of its impact on the Rn mass balance, which is similar to the approach used in other studies (e.g., the discussion of hyporheic flow by Cook et al., 2006). We can certainly recast the paper in this format if it better explains the importance of parafluvial flow, in which case Fig. 10 would become the primary figure that presented the initial results with parafluvial flow and a modified version of Fig. 6 could be used to illustrate the situation with no parafluvial flow.

We restructured the paper as requested by the reviewer. Specifically, we collated the material relating to the Rn mass balance in one section at the end of the methods (Section 3.2) and included more review material on determining the degassing coefficient. We also

reordered the discussion section as suggested so that it presents the Rn mass balance with parafluvial flow at the outset (Section 5.2) and then looks at the issues if that is not included.

The paper has 2 objectives, of which the second is arguably the most novel, but the least well treated within the manuscript. Perhaps part of the issue is that the objectives are loosely related rather than following linearly one from the other.

The second objective (that major flooding events which alter the geometry of the floodplain result in changing locations of groundwater inflows) was part of the original motivation of the study.

However, the question of the extent of parafluvial flow is also important and probably more generally relevant. Most groundwater-surface water studies that have utilized Rn have not explicitly dealt with the impact of parafluvial flow on the Rn activities in the river. To our knowledge only the study by Bourke et al. (2014) has attempted to separate smaller scale hyporheic from larger-scale parafluvial flow in these types of studies, although others (such as Cartwright et al., 2014) essentially combined the effects parafluvial and hyporheic flow. Parafluvial flow is likely to be important in rivers with coarse-grained alluvial sediments such as the Avon and thus is a process that must be taken into account when utilising Rn.

The paper is probably the first to use Rn to understand the changes to groundwater surface-water interaction resulting from changes to the streambed and provides us with a methodology to understand those changes. Additionally, it will be one of only a few to directly address the impacts of parafluvial flow on the Rn mass balance and that too is important. Finally, comparatively few studies have attempted to carry out Rn mass balances at baseflow conditions when the water in the stream will largely be provided by groundwater inflows. While this seems somewhat redundant as the groundwater contribution can be measured by differential gauging, it turns out to be important assessing whether Rn provides a reasonable estimate of groundwater inflows or whether there are problems in the (many) assumptions in the Rn mass balance. Where studies are carried out only at higher flows where the water in the stream comprises groundwater and surface runoff, Rn may imply groundwater inflows that seem reasonable but which are difficult to test. While we have made this point before (Cartwright et al., 2014), it is certainly worth emphasising in this paper as it improves the application of Rn to understanding groundwater-surface water interaction.

We have made the emphasis of the paper clear throughout. The objectives section (1.2) now more clearly outlines the two objectives in the paper. The influence of parafluvial flow on the Rn mass balance is probably the most generally applicable of the objectives and that is reflected in the paper. The changing locations of groundwater inflows is important and novel but may not be something that is as generally applicable. Nevertheless, it provides a good example of how Rn can be used to examine whether this has occurred.

Significant parafluvial fluxes have previously been found in other streams with coarse sediments (eg. Holmes et al 1994, Goosef et al 2003, Bourke et al 2014). Further clarification around the novelty of this work should be provided.

We referenced the Bourke et al. (2014) study which in terms of reported parafluvial flows is probably the most similar to the Avon (in terms of stream characteristics and the implied scale of the parafluvial flow), albeit in a losing stream. That study also used Rn as a tracer and so the approach and results are comparable and address the point made above that this is a process that we need to consider in applying Rn in these types of environments. The Dry Valleys study of Goosef et al. (2003) envisages hyporheic / parafluvial exchange on a smaller scale (e.g. their Fig. 3 shows it to extend a few 10's of cm to possibly a metre or so from the stream edge). The scale of parafluvial flow envisaged by Holmes et al. (1994) is more comparable to that in the Avon and we thank the reviewer for bringing it to our attention. We again emphasise the point that the paper was not trying to prove that parafluvial flows occur, but that it needs to be accounted for in utilising Rn (the former is well established, but the latter is not).

We have made this point more clear throughout the paper. Section 1.2 makes it explicit that while it is well known that parafluvial flow occurs, there has hitherto been little discussion as to the impact on the Rn mass balance. We have also included the additional references.

Is this perhaps the first estimate of the influence of parafluvial fluxes on radon mass balance in gaining stream (or alternating gainging/losing)?

To our knowledge that is the case and actually it is one of only a few studies to explicitly discuss the impacts of parafluvial flow on Rn activities. Rivers with broad coarse-grained alluvial floodplains which contain features such as point bars and pool and riffle section are relatively common at mountain fronts and it is in these that parafluvial flow is likely to be most important. However, the use of Rn mass balance is probably influenced by the work on lowland rivers with finer-grained bank and bed sediments and incised water courses where parafluvial flow may be more limited. A parallel may be drawn here with hyporheic flow; while hyporheic exchange had been documented for many years, its impact on Rn activities was largely ignored until some studies explicitly addressed it (e.g., Cook et al., 2006).

As discussed above, we have made this point more clearly in the Introduction to the paper and also revisited it in the Conclusions.

The group of comments addressed above deal with the reasons for the study and how it was framed. We thought that we had addressed these objectives in the paper, but we can be more explicit at the outset by emphasising them in the aims and more clearly focussing on them throughout the discussion.

The inference of spatial variation in groundwater inflows over time is an interesting application of this method (longitudinal radon mass balance), but it is unclear if this approach is valid using data measured under different flow regimes, some of which were non-baseflow conditions.

The conclusion that the spatial pattern of groundwater inflows has changed is robust. High Rn activities in rivers almost invariably correlates to zones of high groundwater inflows (see Cook et al., 2013 and references therein). While we agree that estimating the groundwater inflows at different flow conditions is more difficult (as we discuss in the paper), understanding where groundwater inflows occur is simpler. Additionally the February 2009 and February 2015 sampling rounds were both at baseflow conditions,

and these occurred before and after the floods which rearranged the floodplain. This was noted but we will make the latter point more clearly in the revised paper.

This point was clarified in Section 5.1.

Further support for the validity of the steady state assumption implicit in the method should be provided.

The reviewer is correct that the steady-state assumption is implicit in these calculations. This is mentioned in Cook (2013), although it is very rarely discussed in Rn studies. In terms of the Avon study, the flows did not change significantly during the sampling rounds (i.e. we did not sample during times of rapidly increasing or decreasing river flow) which implies that the assumption of steady state is reasonable. For reference, the variation in the flows at Stratford during day of the sampling and the couple of days either side (which would account for the time taken for water to transit the river) the rounds are <5%. We will add these details to the revised paper.

We added these details to the discussion on sampling (Section 3.2)

Major comments:

It is more common to simultaneously fit the water, radon and solute mass balances, rather than fitting them individually as was done in this paper. Simultaneous fitting of multiple tracers reduces the uncertainty in the groundwater inflow estimate (McCallum et al. 2012). The approach taken in this manuscript should be justified, and possibly reconsidered.

There are several ways in which the water mass balance has been addressed in the literature. Mullinger et al. (2007, 2009) and Cartwright et al. (2011, 2014) amongst others calculated groundwater inflows from the Rn data using Eq. (1) rearranged to make I the subject. This approach yields the same results as forward modelling if it is assumed that the inflows are uniform in the reach. Frei & Gilfedder (2015, Water Resources Research, 51, 6776-6786) use a PEST approach based on the radon data alone (the Finiflux program). While it is correct that multiple tracers can be used simultaneously, in this case the errors that arise from the use of major ions such as Cl are large due to the variability of Cl concentrations in the groundwater and the relatively small difference in Cl concentrations between the groundwater and river water. We discuss that the Cl concentrations only broadly constrain groundwater inflows in the text. There are only three points where the streamflow is measured on the Avon, and while that provides an indication of the overall groundwater inflows, it cannot be used to constrain the reach-by-reach inflows. The use of streamflow in the calculations also implicitly assumes that gaining reaches do not contain any smaller losing sections, which (as discussed below) is not likely to be the case in the Avon and possibly elsewhere. From a pragmatic viewpoint fitting the fluxes to the Rn data alone is readily achievable in Excel and allows the effect of varying parameters in the Rn mass balance (such as k) to be readily assessed. The methodology is similar to that of Cook et al. (2006) or Cartwright et al. (2014) where Rn (& SF_6) was used to calculate the groundwater inflows and then these were used to construct predicted trends in EC or Cl. We will explain this more in the revised paper.

We included these details in Section 5.2 and noted that for the reasons discussed above, it was not practicable to use either Cl or streamflow to simultaneously determine groundwater inflows.

The first of the two objectives is to test the hypothesis that "large scale parafluvial flow is an important contributor of 222Rn to the river". It was then somewhat surprising that the authors didn't introduce a parafluvial flow component to their analysis until section 5.4 of the discussion. Given that this is stated as one of the main objectives, I suggest that the simulation of stream radon concentrations should be presented as a function of varying amounts of parafluvial flux. This would allow the author to demonstrate that a fit with zero parafluvial flux is not plausible while keeping the focus on the stated objectives.

The structure of the paper reflected the development of our understanding of the system. Our realisation that parafluvial flow was an important contributor to the Rn mass balance came when we examined the data and found it difficult (impossible) to reconcile the Rn data without inclusion of the parafluvial flows. Thus, we envisaged the parafluvial flow as a conclusion to the study and wrote it in that format (i.e., the paper builds to that point). The alternative as suggested by the reviewer that we would expect parafluvial flow and this need to understand its impact on the Rn mass balance is also valid. As discussed above we would be happy to reorganise the paper in that way if it results in a clearer explanation of the importance of parafluvial flow.

As outlined above, the paper was restructured as suggested by the reviewer.

Further consideration should also be given to the effects of "losing" reaches on the water balance. The study river is said to contain "alternating gaining and losing reaches". Could not accounting for water loss along losing reaches result in the discrepancy observed between simulated and measured streamflow? The authors acknowledge this on p9208L4, but do not appear to discuss it further. The influence of these losing sections on the water, chloride and radon balances should be quantified and discussed to fully justify the estimate of parafluvial flow.

It is true that a river with a large number of net-losing reaches might account for the discrepancies between the calculated and observed streamflows. However, in the case of the Avon, it is unlikely to be the only explanation. Between Wombat Flat and Stratford (the first two gauges), only the reaches around 25 to 30 km are net losing (ie it is not the majority of reaches in the stream). To account for the discrepancies in flows, the losses in the reaches at 25 to 30 km would have to be a significant portion of the groundwater inflows in the preceding reaches. There is no indication from field observations that this is the case. While we did not measure streamflows, a reduction in streamflow of 50% or more over such a short distance would be readily observable. Additionally, there are no indications from reports from state agencies or anecdotal evidence from local landowners that these or other reaches in the river dry up even during prolonged drought, and all reaches of the river were flowing during the 2009 sampling campaign (which had the lowest flows).

Our interpretation that parafluvial flows are important is consistent with the nature of the Avon River (coarse-grained alluvials and numerous gravel banks and point bars on the floodplains). There also pool and riffle sections, and many of the riffles have steep longitudinal gradients that are likely to result in river water outflows at their upper

section and inflows in the lower sections. Thus it is likely that the net gaining reaches have some sections which lose water that then reinfiltrates the river.

It is a common but unstated assumption in papers using geochemistry that individual reaches designated as gaining are gaining throughout. This is apparent in the numerous Rn papers where groundwater inflows have been calculated – the net increase in streamflow is calculated from the values of I in Eq. (1) and the reach lengths. If streamflow is to be used in the fitting of data (comment above), this is a necessary assumption. In reality many streams may contain reaches that are dominantly gaining but locally losing, so all calculated inflows must be maxima.

The distinction between parafluvial flow and a river that loses water into underlying aquifer systems is a matter of detail. Both scenarios involve water loss from portions of the stream which then flows through the underlying and adjacent sediments before returning to the river. The impact on Rn will be the same (i.e. Rn activities will increase along the flowpaths between the outflow and the inflow points). Given that the coarse-grained sediments on the floodplain are several metres thick and the scale of parafluvial flow is likely on the tens to hundreds of metre scale (by analogy with other studies), we think that it is more likely that much of the stream water interacts with the alluvial sediments rather than penetrating into the upper section of the regional aquifers.

In our treatment of parafluvial flow, we have assigned a portion of the inflows as being these returning waters which reduces the discrepancy in streamflows and the calculations in Fig. 10 include the impacts on Rn and Cl.

We can better explain these aspects in the paper as our current discussion may not capture all of these points. In particular the point that calculated increases in streamflow must be maximum estimates as it is difficult to account for small losing sections in an otherwise gaining reach is one that has general applicability and it would be well worth mentioning. What would be useful in this explanation is to add a schematic figure to illustrate how we conceptualise the parafluvial flow.

We have incorporated most of this discussion in the paper (Sections 5.1-5.3) and ensured that our conceptualisation of the system is clear. Given, the multiplicity of processes, the Rn mass balance will always have considerable uncertainties and we have tried to be honest about these. In reality many papers that have used geochemical tracers to study groundwater inflows gloss over a number of these issues (for example by assuming uniform gaining behaviour or not carrying out studies at baseflow conditions when reality checks can be carried out); that we have tried to address many of these issues makes the conclusions of the study look less certain but it does highlight many general principles and potential pitfalls which are of general importance. We decided not to add the figure as the paper was becoming overly long.

The treatment of chloride in the parafluvial zone requires further justification. It appears that the Cl- in the parafluvial zone is assumed to remain constant at the concentration from the river at the point of exfiltration. However, given that EC readings at distances of 1-2m from the river were consistent with groundwater concentrations (section 4.5), it seems likely that after mixing with this water, the Cl- concentration in parafluvial water may be more similar to groundwater than the river.

Our interpretation of the Cl data is that it represents a mixture of water that is derived from the river mixed with regional groundwater. Given that the Avon is a gaining system, mixing of water from these two sources in the gravels is likely.

We have added a sentence to explain this to Sections 4.4 and 5.2.

The second of the two main objectives is to test the hypothesis that "major flooding events which alter the geometry of the floodplain result in changing locations of groundwater inflows". However, in reading the remainder of the manuscript, this point does not stand out as a major part of the paper. This is an interesting point, arguably the most novel idea in the paper, and should be further addressed throughout the results, discussion and conclusion. Satellite imagery or mapping of the geomorphic changes along the river channel may be helpful. As major question that arises is what are the hydrogeological conditions that have allowed for this change in the location of groundwater discharge zones. Are there particularly lithologies that are more susceptible to erosion and movement?

As noted above, we considered that this was an important point of the paper but were more focussed on discussing parafluvial flows as the impacts of these on the Rn mass balance has hitherto been little considered. However, we can certainly highlight it further. As to the specific points

- **There is insufficient detailed imagery to show the changes to the floodplain which occur on the tens to hundreds of metre scale.**
- **The floodplain sediments are unconsolidated and there is little vegetation on the floodplain that can stabilise the point bars and gravels. During large floods, the gravels migrate along the river which alters the geometry and position of the floodplain landforms.**

We have given more emphasis to the changes in the floodplain sediments following the major floods throughout the paper.

One apparent shortcoming of the work is that the authors compare groundwater inflows at multiple times with differing streamflows to address this objective. However, a conclusion of both this work and previous studies seems to be that the method works poorly except at low-flow (baseflow), which appears to undermine this approach.

This is not what we meant to imply. Our point (which we also discussed above) was that undertaking studies at baseflow conditions allows a degree of verification of the parameters (as the net groundwater inflows should match the measured increase in streamflow, given that groundwater is likely to represent the only / main source of water at those times). This is valuable as it allows checking of the parameters in the Rn mass balance. At higher flows, as long as the calculated groundwater inflows are less than the measured increase in streamflow, the results are plausible but there are less cross-checks. While that will always be the case, demonstrating that the adopted parameters produce acceptable results under baseflow conditions gives some confidence to the calculations at higher flows. We will clarify this in the revised version.

We have made these points more clearly in Sections 1.2, 5.4 and in the Conclusions.

Was the river at steady state during the non baseflow sampling campaigns as required by the method (Cook 2013)? Further discussion and justification of this approach for estimating groundwater inflows under non-baseflow conditions is required.

Yes it was and we will quote the variation in flows around the sampling times (<5% variation at Stratford). This is an important point but one that is hardly ever discussed in geochemistry papers (either by our group or others) and it would be well worth noting.

We added this detail to Section 3.1

The introduction is quite long and would benefit from significant editing. The authors may consider implementing a theory section that contains the theoretical background and all equations, separate to the introduction. This would allow for the scope and objectives of the paper to be more clearly presented to the reader in the first instance and remove the need for sub-headings within the introduction.

Throughout the manuscript it seems that information is not presented in the appropriate section. Results are presented in discussion, equations in discussion, and methods in results and discussion. These will be outlined in more detail in minor comments.

Some of this is a question of preference and papers on this topic in general have ordered the material in a variety of ways. The papers by our group and also by others (e.g. Mullinger et al., 2007, 2009) have introduced equations in the sections where they were utilised with the main equations in the introduction. It is also not uncommon to have equations presented in the methodology. Other papers (e.g., Cook 2006) have a Theory section following the Introduction. Given that we do not develop new equations here, there is no reason not to group them together and this would shorten the Introduction and Discussion sections, which are already long.

As discussed above we aggregated this material into Section 3.3

Minor comments:

9) Consider changing units to Bq/L instead of Bq/m3 as this removes the need for large concentration values (10000 becomes 10).

Using Bq/m^3 is logical from the point of view of the dimensions of the terms in Eq. 1. The terms in Eq. 1 have units of Bq/m/day which for the hyporheic or parafluvial flux is relatively easy to envisage. Using Bq/L, these terms become Bq/L.m^2/day which is not as elegant or as easy to envisage (although the calculations are the same). Since m^3 is an SI unit, we would propose to keep it as is.

Not changed for the reasons listed

10) Consider changing the title to something more specific - as written it is quite general and doesn't suggest anything novel.

We consider that the title reflects the study but will modify it to include reference to the changing loci of groundwater inflows and replace geochemistry with Rn.

We changed the title as indicated.

11) The authors may wish to reconsider abbreviations such as ~for approximately, and i.e., e.g. or c.f. within references.

Changing ~ to c. is relatively straightforward. We are not clear on the problems with i.e. etc but will follow HESS house style.

We have ensured that the manuscript follows HESS house style.

12) P9207 L11, more references required for methods to assess gw inflow to rivers.

13) P9207 L114. Specify baseflow separation rather than "numerical techniques"

14) P9207 L114. Are the authors referring to the type of water balance models used in this paper? Clarification required.

We did not want to turn this part of the introduction into a review. Considering the length of the paper, we propose to omit these sections (P9207 L10-22) as they are too brief to add much of importance and they detract from the discussion of geochemical tracers (P9207 L23 et seq.)

As indicated we removed this section as to expand it would have made the paper overly long and this review aspect probably wasn't needed.

15) P9209 L20 Other methods of estimating k should also be mentioned; k can be directly measured using an artificial tracer release while the authors use an observed decrease to estimate k.

We do discuss other methods of estimating k later in the paper (the empirical equations that relate k to velocity and width) and compare them with the approach that we used to estimate k (which is similar to that of Mullinger 2007, 2009 and some of our other studies such as Cartwright et al., 2011, 2014). The degassing coefficient is a difficult parameter to constrain with confidence and we have done far more to understand k than in many other Rn papers, which is appropriate given its importance. k can be measured directly using artificial tracers; however, this is sometimes only attempted at a specific flow condition or in a small portion of the river, and that leads to questions about how representative the values are. We will add a comment regarding the issues around k and note there are other methods. In the revised paper, if the equations are grouped into a single section then that discussion would logically belong there.

We added this discussion to Section 3.3 where the Rn mass balance is discussed.

16) P9210 L7 Use of exfiltrate and infiltrate somewhat confusing, given that "infiltration" is commonly used to refer to water percolating into the subsurface.

We agree that the terminology is not totally clear. We will use "water outflows" and "water inflows" to remove the ambiguities.

We changed the terminology throughout as indicated.

17) P9210 L21 suggest: increases with increasing residence time until secular equilibrium is reached.

Will reword as suggested (makes it clearer).

Changed as indicated

18) P9211, 9212. The difference between numerical approaches for hyproheic zone and parafluvial zone is fundamentally because in the hyporheic zone it is reasonable to assume it is well mixed with one concentration, whereas in the parafluvial zone, with longer flow paths, this assumption may not be valid. This should be clarified.

We can add those details, although in many ways it is a matter of scale. The hyporheic zone probably has gradients in Rn activities, especially if there is mixing between high Rn groundwater at the base of the zone or variable length flow paths; however those would be difficult to resolve these during sampling whereas conceivably sampling within the parafluvial zone would be able to resolve the differences in Rn along the flow paths.

We clarified this material (Section 3.3)

19) P9213. Section 2 contains information other than local geology and hydrogeology. Consider renaming as Site Description.

We will change this to something more suitable (e.g., Study Area or Site Description).

We changed this to "The Avon Catchment"

20) P9214 L1 Clarify that streamflow was measured at fixed gauging stations, rather than using velocity meter.

We will add these details. Using a velocity meter to estimate streamflow is feasible in some circumstances but in wide shallow rivers with irregular beds (such as the Avon) it would be difficult to get reliable results.

We clarified this (Section 3.1)

21) P9214 Some discussion of whether the characteristics of the site described in section 2 make it a unique study site, or one that is representative of a large number of river catchments would be helpful.

This is a good suggestion. The Avon is similar to many streams that occur at mountain fronts both in Australia (e.g. many of the streams draining the Australian Alps) and elsewhere (e.g., New Zealand) and so the results of this study will be generally applicable. We will mention this in the introduction and echo it in the conclusions.

We added this material to Section 1.2 and also the Conclusions

22) P9215 L1-11 Not required, consider deleting.

We can shorten this section, but some of these details are needed. The comments regarding the paucity of monitoring bores is required to address comments by Reviewer #2 (below) and the prohibition of river water use is made use of in the discussion where it is noted that sampling occurred during periods when there was little water abstraction from the river.

The section was shortened whilst retaining the details that we made use of later.

23) P9215 L12-17 Consider moving to introduction.

We agree that it would sit better in the introduction where the reasons for carrying out the study are explained.

This was moved to section 1.2.

24) P9216 L22 Reference?

The precisions are ones that we have determined in-house by repeated measurement over a short period (a couple of days) of water samples with a range of Rn activities on our RAD-7 meters. We will add those details to the paper.

This was clarified

25) P9217 Eqn7: Suggest presenting all equations in one section.

As discussed above given that we do not develop the equations as part of the study, we could easily do this. It would also help facilitate the discussion of estimating k (comment 15).

As noted above we did this.

26) P9217 Streamflow results description is confusing, suggest a table. These data are important context for the comparison of data that the paper purports to undertake and subsequent conclusions.

We agree that this paragraph is very dense and difficult to wade through and that a table would present the data better.

We added this material to Table A1 and shortened the text.

27) P9218 Chloride concentrations are reported for the river and groundwater but the alluvium, while EC is reported for the groundwater and alluvium but not the river. At least one of either EC or chloride should be reported for all three end-members.

We can readily report EC for all the end members. We do have some Cl data for the alluvial waters and given the good correlation ($r^2 \sim 0.97$) between EC and Cl in the waters as a whole, we can also report Cl in the alluvial waters; although for some of these, it would obviously be a calculated value.

We now report EC for all the end-members (Section 4 and Tables A1 & A2).

28) P9219-20 Suggest swapping order of S4.4 and S4.5

We agree that would make more sense as section 4.5 describes water geochemistry that is more akin to the data in sections 4.3 and 4.2.

We swapped these sections as indicated.

29) P9220 L18 Chloride increase could also relate to evaporation along river.

Evaporation would probably occur relatively uniformly along the river, which would produce a steady increase in Cl even in reaches that were losing, whereas the observed pattern has discrete zones of increasing Cl that correlate with the zones of high Rn. This makes it more likely that the vast majority of the Cl increase is due to groundwater inflows. The reaches interpreted as losing have little or no increase in Cl concentrations, which would not be the case if significant evaporation had occurred. At the evaporation rate of $5x10^{-3}$ m^{-1} that we quote in the study, the increase in Cl concentration over a 10 km reach due to evaporation (calculated by rearranging Eq (1) and using the measured discharge values and widths) is <0.1 mg/L.

We also have stable isotope data (not reported) and most of the river samples lie close to the local meteoric water line rather than defining distinct evaporation trends. Since a relatively modest degree of open-surface evaporation produces a displacement in stable isotope ratios (10% evaporation ~1‰ change in δ^2H), this also points to relatively minor evaporation.

We can readily discuss the first points, but are reluctant to add the stable isotope data as the paper is already quite long and the data does not inform about other processes.

We didn’t change this as evaporation is not that significant in this catchment.

30) P9220 L23 Specify river distance that you’re referring to here.

We can add the distances to the text here and elsewhere. The names make for easier reading but are probably not as informative.

We added the distances.

31) P9221 L19-28, P9222 Suggest moving to methods.

See comments above regarding aggregation of equations.

As discussed above, all this material is now in Section 3.3.

32) P9222 L7 Chloride concentrations along a losing reach will still increase due to evaporation

That is true and the observation that there are regions where the Cl does not increase is consistent with the points made above that evaporation is not that important in increasing the Cl concentrations.
We have not changed this as evaporation has very little impact on either Rn or Cl.

33) P9222 L14 Specifically, mixing is the only mechanism that will increase the EC of water in the hyporheic or parafluvial zones

As was pointed out to us in a review of a previous paper (Cartwright et al., 2014), evapotranspiration can occur from river gravels. However, on the timescales that are involved in parafluvial flow or hyporheic exchange, ET is a minor process and mixing is the main player. We will clarify this sentence.

We clarified the sentence as indicated.

34) P9222 L22 What is the variance on this mean? And therefore the associated uncertainty?

This was discussed in Section 4.4. The standard error is ~180 Bq/m^3/day (or ~8%). In section 5.3, we did not propagate this error as even using the 95% confidence interval of ~16%, the impact of this error is small compared with the assumptions around estimating the dimensions of the hyporheic zone (which in most systems is only broadly constrained). We will note this in the revised version.

We noted this in Section 5.3.

35) P9222 L24 I think you mean hyporheic here, not parafluvial

Yes, should be hyporheic.

We changed this as indicated.

36) P9223 Estimates quantities of groundwater inflow should be reported in the results section.

We disagree with this comment as this is an interpretation of the data and as such belongs in the discussion.

We retained this in the discussion section.

37) P9224 Heading 5.3 Not sure what you mean by variability here?

It is clearer if we just call this "uncertainties and sensitivity"

We changed this as indicated.

38) P9225 L15 The gas transfer term also includes w and d, is it possible that your k is underestimated but these other parameters are underestimated?

It is correct that there is a combination of terms in the gas transfer term. The approach that we used, which was to match the observed decrease in Rn in the losing reaches, estimates the whole kdwc term and then k is derived from the d, w, and c estimates / measurements. If we assign different values to w or d, then our k value will be different but the kdwc term remains the same (which is what is important for the Rn mass balance). We will clarify this in the text.
We clarified this as indicated (Section 5.2).

39) P9225 Consider moving eqns 8 and 9 to theory or methods sections.

See comments above.

We aggregated this material.

40) P9226, Fig 8&9. Adjusting these parameters individually does not account for the fact that there are multiple parameters in a given term, ie gas transfer contains both k and w.

That is true, although as discussed above what we estimated in this case was the net kdwc term. What we tried to show in this section is that there are limits to how the parameters can be varied independently. So in a losing reach, there will be combinations of Fh and k (or strictly kwdc) that will produce the observed Rn profiles. This is important as parameterisation of Eq (1) is difficult, but showing that there is not freedom to change all the parameters independently of each other helps with reducing the overall uncertainty on the calculations. This is a little-reported point in Rn papers and we will clarify this in the revised version.

We clarified this discussion (Section 5.3)

41) P9229 L11 What are the difficulties? Which of these were known prior to this study and which are new based on this study?

This was not a very specific or informative start of the conclusions. What we were trying to convey was that the Rn mass balance could not be achieved without considering parafluvial flow.

We rewrote this part of the Conclusions.

42) P9229 L26, P230 L6. Suggest this belongs in introduction.

We agree that as a statement it reads like introductory material and a similar statement appears at the end of section 1.2. We can remove it from this section.

Material was removed.

43) Fig 1b What is Cr in this calculation?

C_r is the same as C_{in} (i.e. 1000 Bq/m^3). This is noted in the text and we can add it to the figure caption for clarity.

We added this to the caption.

44) Fig2 What are the arrows on the map?

They are the generalised directions of groundwater flow. We will add this to the legend.

We added this to the caption.

45) Fig 4 Suggest adding streamflow

We can add this as a small third panel or as a dataset with a second Y-axis on the lower panel.

We decided not to do this as the streamflow data appear on Figs 7 and 9.

46) Fig 5 Suggest adding distributions of Rn and EC in river and groundwater to demonstrate presence/absence of distinct end-members.

Agreed that this would be useful and we can add the endmembers.

We added the groundwater compositions to this Figure.

47) Fig 8 Suggest this fig not required.

If the paper is reformatted then this Figure will probably disappear.

We removed this Figure

48) Fig 10 Radon fit identical to Fig 6, consider that panel may not be required.

Since the Rn fit is that produced by this specific calculation it is better to retain it.

This was retained.

49) Ensure font sizes are adequate and consistent across all tables and figs

We will check the figures

Reviewer #2

We also thank this reviewer for their helpful comments and provide the following responses.

The experimental design focuses mainly on the use of radon-222 as a hydrogeologic tracer of groundwater and/or parafluvial inflows. The main critical point of the applied approach is the definition/measurement of an average groundwater value for radon and major ions, especially chloride. In the study, the authors have measured radon specific activities and major ions concentrations in 8 boreholes, finding big discrepancies among values likely due to the sampling in the riverbank. It would have been very important for such kind of studies to enlarge the sampling network to boreholes surely unaffected by river water and also to reconstruct the morphology of the water table to identify gaining river reaches.

We chose to install bores close to the river to sample the groundwater that directly interacts with the river. Utilising regional bores for Rn data is commonly done; however, since Rn activities in groundwater are a function of the mineralogy of the aquifer it is always questionable how representative the Rn activities are when measured in a bore several km distant from the river. Only the closest bore at Pierces Lane is actively

exchanging with the stream. The geochemical variation in the other bores both temporally and spatially is similar to that observed elsewhere in bores further from the stream (e.g., Yu et al., 2013; Atkinson et al., 2015). Given the scale of the catchment it would be implausible to install a whole network of bores to map the water table (desirable though that would be).

We have noted that the bore network is insufficient to study groundwater interactions in detail (Section 1.2) and made it clear that most bores sample the groundwater rather than the water from the gravels (Section 4.3).

The abstract should include also a brief description of the applied methodological approach.

We will add a sentence on methodology to the abstract.

We added this sentence as indicated.

The description of the methodology is clear and thorough and well evidences the critical points. The discussion of the shifting inflow reaches (paragraph 5.1) has to be improved and reorganized, since the reader has poor and fragmented information about that.

As with our responses above, we agree that the changes to the loci of groundwater inflows over time should be better emphasised as it is a key result of the study.

We reworded Section 1.2 to make the aims more clear.

Specific Comments (on the uploaded supplement)

Page 9212 L16. Veracity vs. Accuracy

Plausibility is probably a better term

We reworded this sentence to make it more clear (Section 1)

Page 9212 L20. Hypothesis vs. Hypotheses

Agreed, should be hypotheses (plural)

Changed as indicated.

Page 9214 L9-10. It would be better writing: "both are instrumented with discharge gauges" or similar. Nevertheless, it seems that non data about the tributaries' discharge have been reported.

Clarification of the streamflow data (that it is from established gauges) was also requested by Reviewer #1 and we will add that. The streamflow data from Valencia and Freestone Creeks is mentioned in Section 5.2 but probably should have been also be reported in Section 4.1.

We noted that the streamflow measurements are from fixed gauges (Section 3.1) and also added the tributary discharge data to Section 4.1.

Page 9216 L1. No mention in the text on discharge data from tributaries. In Figure 2 also the river gauges on the tributaries are not reported. This information is useless if incomplete as it is.

We agree that we should report the data in this section and we can add the gauge locations to Fig. 2.

We added the tributary data to Section 4.3 and show the location of the gauges on Fig. 1.

P9216 L19. The expression of radon specific activities in Bq m^-3 are quite common, nevertheless this way to present data can confound the reader since it is not very clear if the cubic meters are referred to a volume of water or air. Commonly radon is reported as Bq L^-1. The authors are requested to put a conversion expression between Bq m^-3 and Bq L^-1 (i.e., 1 Bq m^-3 = tot Bq L^-1).

We had not considered the possible ambiguity. As discussed in the responses to Reviewer #1, the choice of units gives the terms in Eq. (1) more useful dimensions. We will note the equivalence and specify that it is m^3 of water.

We specifies that it was per m^3 of water in Section 3.2.

Section 4.3.
We will correct the table numbering to make it consistent.

We corrected the Table numbering

Section 4.3

Units of Rn activity discussed above

As discussed above, Bq/m^3 was retained as the unit

Section 5.1. This paragraph could be better organized in order to discuss also the temporal shifts of the gw inflows, which are only mentioned in the text (lines 7-9, page 9221).

As with our responses to Reviewer #1, we agree that the changes to the loci of groundwater inflows over time should be better emphasised as it is a key result of the study. We will better explain this point here.

We have placed more emphasis on the changing locations of groundwater inflows (Section 5.1)

P9221 L10-11. It would be desirable to explain why the authors have not carried out a water table surface reconstruction, using the agricultural wells in the floodplain, besides those sampled.

Unfortunately, this was not possible. Groundwater levels in these wells were generally (but not always) measured during their construction, but not thereafter and so it is not possible to construct the water table at any given time. There is a water table elevation map for the region but it is constructed from a digital elevation model and estimates of

depth to water that were made with numerous assumptions and little verification. This combination of data indicates the general direction of groundwater flow but is not suited for more detailed analysis and could not be used to determine the distribution of gaining vs. losing reaches. We will note the limitation in the revised manuscript and emphasise that in many river systems there is not sufficient hydraulic data to understand the details of groundwater-surface water interaction making geochemical tracers a more viable option.

We noted that it was not possible to determine groundwater-surface water relationships from the head data (Section 1.2).

P9221 L21-22. Please, explain better this sentence.

We meant that using a smaller distance step in the calculations did not change the results. We will clarify this in the revised paper.

We clarified the sentence (Section 5.2).

P9224 L13. Delete "need"

Agreed, this is a typo.

We changed this as indicated

P9226 L11. Insignificant vs negligible

Agreed, negligible is a better term

We changed this as indicated (Section 5.3).

P9229 L5-9. The clarity of this period needs to be improved.

The point that we were trying to convey here is that we can produce plausible estimates of groundwater inflows at the higher streamflow conditions even if we ignore parafluvial flows but the analysis of Rn at low streamflow conditions makes it likely that there is parafluvial flow at all times. This also goes back to the point that we made above that carrying out studies at baseflow conditions is important in testing the parameterisation. We will clarify this section.

This material was clarified (Section 5.4).

P9229 L18-21. This sentence is in contrast with the statement of the first working hypothesis. In other words, from this sentence the reader understands that in a natural process of trial and error the authors came to the conclusion that parafluvial flow was occurring, while from the reading of the working hypothesis the parafluvial flow seems already foreseen. There is a subtle mismatch between the two sentences. If the authors agree with this suggestion, they could adjust the text accordingly.

This comment is similar to those of Reviewer #1 as to how we framed the paper. In reality, the process was one of trial and error (or realisation). As discussed above, the paper

would probably be clearer if we followed the format of stating that parafluvial flow is likely and then going on to understand it's impact on Rn. This would also agree better with how the hypotheses are expressed.

As discussed above, we reformatted the paper.

P9229 L27. As pointed out in the general comments, the choice of the groundwater samples has not been addressed properly due to the great measured spatial and temporal variability likely caused by the sampling of river bank water.

As noted above, the logic behind the location of the groundwater bores is that we wanted to sample the water that directly interacts with the stream. The Cl concentrations of the regional groundwater that is sporadically reported from the bores on the floodplain are similar to those in our bores. Importantly, the spatial variability of Cl concentrations are similar and this restricts the use of Cl in the mass balance equations. This is true in most of the catchments in SE Australia in which we have worked so is an important general point. We will note this in the results section.

We noted that the Cl concentrations in the bores were similar to those elsewhere in this catchment and adjacent catchments. We also noted that aside from the one bore at Pierces Lane, the bores sampled the groundwater not water from the gravels (Section 4.3). The ^{222}Rn activities for the groundwater is actually very similar to that which is predicted from the emanation rates, which is further evidence that we sampled groundwater not water from the parafluvial zone,

P9230 L7-9. This consideration is meaningful. It would be desirable that in the discussion session a major emphasis is given to the comparison of the results during baseflow and high flow conditions, also to better introduce and discuss the second working hypothesis that changing flow conditions alter the location of groundwater inflows.

We agree that the importance of the baseflow sampling needs to be emphasized throughout as it is an important point. The second hypothesis isn't that the changing flow conditions alter the location of groundwater inflows but that major floods rearrange the sediments and landforms on the floodplain and that this results in the changes over time. We will ensure that this is clear in the revised manuscript.

We have clarified this material throughout the paper.

Table 1.

Will correct typo in "Stream" and add units for velocity

Corrected as indicated.

Table 2. In the table the sampling date has not been reported. The discrepancies found in the sediment emanation rate could be due to the sampling carried out in different times at the same location (e.g. before and after a flood which could move the sediments, as the authors have reported).

All the sediments were sampled post the flood (which we will clarify). We are not sure that these are discrepancies but rather the natural variability in emanation rates; the variability is similar to that in other studies (e.g. Bourke et al., 2014; Cartwright et al., 2014) which we will also mention.

We clarified the time of sampling (Section 3.2) and also mentioned that the variability that we see is similar to that in other studies (Section 4.5).

Fig. 2. For the sake of clarity and to help the reader, the names of the sampling sites could be reported following the sequence along the river, starting from Browns (BR) and ending to Chinns Bridge (CB), instead of being reported in alphabetical order.

We agree that this would probably be more useful.

We changed this as indicated.

Fig. 3. It would be desirable to clearly evidence the sampling campaigns on the streamflow diagram (e.g., tracing a line intersecting the discharge curve for each sampling campaign) to give immediately the information on the flow regime at the time of sampling.

We agree that this would be a better illustration of the sampling times.

We changed this as indicated.

Moreover, the fact that the major floods changed the geometry of the floodplain could be deleted from the figure caption and kept in the text only.

We consider that it is useful to have this in the caption so the reader can relate this to the Figure.

We kept this for the reasons indicated.

Fig. 8. From the text it not clear if the predicted and observed 222Rn activities match also in the case of the isolated change of parameters.

This statement is incorrect in the figure caption. It is the streamflows that match and the Rn activities that are not predicted correctly. We will ensure that this is correctly expressed in text and figures.

As discussed above, this Figure was removed.

Fig. 10. The range of variation (95% confidence interval) of the calculate streamflow should be reported on the plot (as a shaded field in the d) plot).

We can add the confidence intervals to the streamflow on the Figures that are discussed in Section 5.

We elected not to do this as the Figure became unreadable.

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

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Abstract

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 ^{222}Rn 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 ^{222}Rn budget. The distribution of ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn activities due to the ^{222}Rn emanations from the alluvial sediments. If the high ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn from parafluvial flow will result in overestimating groundwater inflows to rivers.

1. Introduction

Quantifying groundwater inflows to streams and rivers is critical to understanding hydrogeological systems, protecting riverine ecosystems, and managing water resources (e.g., Winter, 1999; Sophocleous, 2002; Brodie et al., 2007). Groundwater inflows may form the majority of water in gaining rivers during periods of low streamflow, and 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. ^{222}Rn as a tracer of groundwater inflows

^{222}Rn, which is an intermediate isotope in the ^{238}U to ^{206}Pb decay series, is an important tracer for quantifying groundwater inflows to rivers. ^{222}Rn has a half-life of 3.8 days and the activity of ^{222}Rn reaches secular equilibrium with its parent isotope ^{226}Ra over 3 to 4 weeks (Cecil and Green, 2000). Because ^{226}Ra activities in minerals in the aquifer matrix are several orders of magnitude higher than those in surface water, groundwater ^{222}Rn activities are commonly two or three orders of magnitude higher than those of surface water (Cecil and Green, 2000). This makes ^{222}Rn 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 ^{222}Rn 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), ^{222}Rn is also useful in determining locations of groundwater inflow, even if where the inflows are not quantified.

The successful application of ^{222}Rn to determine groundwater inflows, however, requires careful consideration of several processes and uncertainties. ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn mass balance (Cartwright et al., 2014).

1.2. Objectives

This paper examines groundwater-river interaction in the Avon River, southeast Australia, primarily using ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn budget to rivers, few studies have explicitly considered this process in the ^{222}Rn 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 ^{222}Rn 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 km^2 (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 1.3x10^{7} and 9.0x10^{8} m^3 yr^{-1} (median = 3.0x10^{8} m^3 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.

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 m^3 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 $<10^4$ m^3 day^{-1} (Gippsland Water, 2012).

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

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 Cl and Na are discussed in this study. ^{222}Rn 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 m^3 of water (Bq m^{-3}). 0.5 L of sample was collected by bottom-filling a glass flask and ^{222}Rn 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 ~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. ^{222}Rn emanation rates (γ) from these were determined by sealing a known dry weight of sediment in airtight containers with water and allowing ^{222}Rn to accumulate (Lamontagne and Cook, 2007). Following 4-5 weeks incubation, by which time the rate of ^{222}Rn production and decay will have reached steady state, 20 to 40 ml of pore water was extracted and analysed for ^{222}Rn activities using the same method as above but with counting times of 6 to 12 hours. γ (Table 2) was calculated from ^{222}Rn produced per unit mass of sediment E_m, sediment density ρ_s, and porosity ϕ by:

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

(parameters summarised in Table 1).

3.3. Radon mass balance

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

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

(modifed from Mullinger et al., 2007; Cartwright et al., 2011; and Cook, 2013). In Eq. (2): Q is streamflow; c_r and c_{gw} are the ^{222}Rn activities in the river and groundwater, respectively; I is the 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 hyporheic zone and inflows of parafluvial waters, respectively; k is the gas-transfer coefficient; and λ is the decay constant (Table 1). A similar mass balance also applies to major ion concentrations. Since the concentration of a conservative tracer such as Cl is controlled only by groundwater inflows and evaporation, only the first two terms on the right-hand-side of Eq. (2) are relevant. If the river is gaining throughout and solely fed by groundwater the increase in streamflow downstream is:

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

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

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

(Hoehn et al., 1992; Hoehn and Cirpka, 2006) (Fig. 3a). An identical expression relates the ^{222}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}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}Rn flux from the hyporheic zone (F_h) is:

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

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 ^{222}Rn into the river at the end of discrete flow paths through the parafluvial zone (Hoehn and Von Gunten, 1989; Hoehn and Cirpka, 2006; Bourke et al., 2014a). In that case, F_p is given by a similar expression to that which accounts for the input of ^{222}Rn due to groundwater inflows:

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

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 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 same F_p. The volume of sediments with which the water has interacted during flow through the parafluvial zone (V_p in m^3 per m length of river) is governed by I_p, t_p and ϕ. If the flow paths through the parafluvial zone are regular, V_p will be the cross-sectional area of the parafluvial zone through which the water from the river flows (A_p):

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

(Bourke et al., 2014a). For the same input parameters, Eqs (5) and (6) yield closely similar estimates of F_p (Bourke et al., 2014a) and the least well-known parameters are in both cases A_p and t_p.

There are several approaches that may be used to estimate the rate of ^{222}Rn degassing from rivers. Firstly, as degassing involves diffusion of ^{222}Rn 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 ^{222}Rn 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 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, 2015). Determining k or z by comparing calculated and measured ^{222}Rn activities requires that the ^{222}Rn 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 calculated from these methods for a few specific well-understood river reaches, it is possible that they are not valid for all river reaches.

It is also possible to measure *k* directly by using introduced gas tracers such as SF_6 (Cook et al., 2003; Cook et al., 2006; McCallum et al., 2012; Bourke et al., 2014a), which has the advantage of estimating *k* for the river being studied. However, such measurements are generally made along small reaches of a river that may not be representative of the river as a whole. Additionally, if the experiments were made at specific flow conditions, the gas transfer coefficients may or may not be applicable to sampling campaigns made at different flow conditions.

There are several empirical relationships that estimate *k* from river velocities (*v*) and depths. The commonly used O'Connor and Dobbins (1958) and Negulescu and Rojanski (1969) gas transfer equations as modified for ^{222}Rn are:

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

$$k = 4.87x10^{-4}\left(\frac{v}{d}\right)^{0.85} \qquad (9)$$

(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.

4. Results

4.1. *Streamflow*

Between January 2000 and February 2015 streamflow at Stratford varied between 500 and 1.38x10^8 m^3 day^{-1} (Department of Environment and Primary Industries, 2015). Despite this period including years with well below average rainfall, for example 2006 when rainfall was ~50% of the long-term average (Bureau of Meteorology, 2015), there were no periods of zero streamflow. Mean daily streamflows at Stratford during the sampling rounds ranged from 10,670 m^3 day^{-1} to 88,800 m^3 day^{-1} (Table A1, Fig. 2a) which represent streamflow percentiles of 39.5 to 89.9 (Fig. 2b). In February 2015, which is the sampling round discussed in most detail below, the mean daily streamflow was 12,510 m^3 day^{-1} at The Channel, 23,090 m^3 day^{-1} at Stratford, and 25,780 m^3 day^{-1} at Chinns Bridge. Inflows from Valencia Creek and Freestone Creek in February 2015 were 2410 m^3 day^{-1} and 600 m^3 day^{-1}, respectively (Department of Environment and Primary Industries, 2015).

4.2. River Geochemistry

Figure 4a shows the ^{222}Rn activities of the Avon River for the six sampling campaigns. There are several distinct zones of elevated ^{222}Rn activities, notably at Wombat Flat (4.8 km) where ^{222}Rn activities are up to 2040 Bq m^{-3} and between Bushy Park and Schools Lane (16.3 to 25.3 km) where ^{222}Rn activities are up to 3690 Bq m^{-3}. Zones of lower ^{222}Rn activities in the upper reaches occur at Smyths Road (8.1 km) and in the reaches between Stewarts Lane and Stratford (30.1 to 35.1 km). The downstream river reaches between Knobs Reserve and Chinns Bridge (37.8 to 49.7 km) also have relatively low ^{222}Rn activities that generally decline downstream. The position of the highest ^{222}Rn activities changed in the periods prior to and post the 2011 to 2013 floods. In March 2014 and February 2015 the highest ^{222}Rn activities were at Bushy Park (16.3 km), whereas this site had relatively low ^{222}Rn activities in February 2009 and April 2010 when the highest ^{222}Rn activities were at Pearces Lane (20.0 km). The distribution of ^{222}Rn 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 ^{222}Rn activities were recorded during the periods of highest flow (September 2010 and July 2014).

EC values and Cl 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, Fig. 4b). Cl concentrations at low streamflows in March 2014 were generally higher (up to 98 mg l^{-1}) than in the other sampling campaigns, while Cl concentrations were <20 mg l^{-1} during the highest 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 ^{222}Rn activities are highest at low streamflows. The concentrations of other major ions (e.g., Na) increase downstream in a similar manner (Table A1).

4.3. Groundwater Geochemistry

Groundwater from the near-river bores at Pearces Lane and Stratford has ^{222}Rn activities that vary from 480 to 28,980 Bq m^{-3} (Table A2). There is some variation in ^{222}Rn activities in individual bores between the sampling rounds with relative standard deviations between 6 and 34%. The mean value of all groundwater ^{222}Rn 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 ^{222}Rn 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 µS 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. ^{222}Rn activities of these samples were between 7000 and 28,000 Bq m^{-3} (n = 21) (Fig. 5a), which are also significantly higher than the ^{222}Rn activities in the adjacent river. As discussed below, these data are interpreted as indicating that the gravels contain a mixture of groundwater and parafluvial water.

4.5. ^{222}Rn Emanation Rates

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

5. Discussion

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) ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn 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 river to the groundwater.

5.2. Quantifying Groundwater Inflows

This section concentrates on modelling the ^{222}Rn activities for the detailed February 2015 sampling campaign (Fig. 4a). It was considered that groundwater inflows, hyporheic exchange, and parafluvial flow all contributed ^{222}Rn to the river. The groundwater ^{222}Rn activity was assumed to be 13,000 Bq m^{-3}, which is consistent both with the measured ^{222}Rn activities of groundwater (Table A2) and the calculated ^{222}Rn activities of water in equilibrium with the alluvial sediments.

The flux of ^{222}Rn from the hyporheic zone was estimated from Eq. (5) using the mean γ value of 2300 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 ^{222}Rn activity of the river in that reach. The residence time of water within the hyporheic zone is likely to be short (Boulton et al., 1998; Tonina and Buffington, 2011; 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 relatively insensitive to the actual residence times in the hyporheic zone (Lamontagne and Cook, 2007; Cartwright et al., 2014). The width of the hyporheic zone has been assigned as the river width. The thickness of the hyporheic zone is less well known; however, by analogy with rivers elsewhere, it is likely to be a few centimetres thick (Boulton et al., 1998; Hester and Doyle, 2008; Tonina and Buffington, 2011) and a value of 10 cm is initially adopted.

Parafluvial flow is conceived to occur on the tens of metres to kilometre scale and to represent water that is lost from the river into the floodplain sediments that subsequently re-enters the river downstream. The Cl and ^{222}Rn data from the water contained within the gravels (Fig. 5) are interpreted as reflecting mixing of groundwater and parafluvial flows that will occur where the river is gaining. This scenario requires that the river is locally losing. As discussed above, on the kilometre scale the Avon River may contain losing reaches. Additionally, the reaches that are interpreted as being overall gaining may contain smaller sections that are losing. In particular, the riffle sections commonly have steep longitudinal gradients and may transition from losing at the upstream end to gaining at the downstream end. Parafluvial flow is probably hosted mainly within the coarser-grained alluvial sediments (although conceivably it could also include water that flows through the upper levels of the aquifers underlying the alluvial sediments). By contrast with hyporheic exchange which occurs along all reaches (whether gaining or losing), inflows from the parafluvial zone require upward head gradients and only occur where the river is gaining. The parafluvial inflows will increase the ^{222}Rn 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 ~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 ^{222}Rn 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 ^{222}Rn 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 rates are expected to be low.

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.

Assuming that in the gaining reaches there are 50% parafluvial inflows and 50% groundwater inflows allows both the ^{222}Rn 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 Pearces Lane (8.1 to 20.0 km) (Fig. 6b), which is the region where Cl concentrations also increase markedly (Fig. 4b). Assuming that the waters are in secular equilibrium with the sediments, the combined inflows of groundwater and parafluvial water for this reach are up 2.5 m^3 m^{-1} day^{-1} of which groundwater inflows are ~1.26 m^3 m^{-1} day^{-1}.

There is no process in the parafluvial or hyporheic zones other than mixing with groundwater that increases the Cl concentrations of the through-flowing water. Thus the Cl concentrations in the river reflect only the groundwater inflows and in theory it would be possible to use Cl to quantify these (c.f., McCallum et al., 2012). However, the high variability of Cl concentrations in the groundwater and the relatively small difference between groundwater and river Cl concentrations results in large uncertainties. The change in Cl concentrations (Fig. 6d) was calculated from the groundwater inflows assuming that groundwater has a Cl concentration of 85 mg l^{-1}. The calculated Cl concentrations are slightly higher than those observed, but if the Cl concentration of the groundwater is allowed to vary within the 95% confidence interval (±16 mg l^{-1}) the observed trend can be reproduced.

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 ^{222}Rn (F_p) increases (Fig. 3). For example, if c_r = 2300 Bq m^{-3}, which is a typical value in many of the reaches between Valencia to Bushy Park (10.9 to 16.3 km) and c_p = 12,700 Bq m^{-3}, (c_p - c_r) = 10,400 Bq m^{-3}. If I_p = 1 m^3 m^{-1} day^{-1}, F_p = 10,400 Bq m^{-1} day^{-1} (Eq. 6). If γ = 2300 Bq m^{-3} day^{-1}, c_p is 2487, 4023 and 11,004 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} requires I_p ~58 m^3 m^{-1} day^{-1} for t_p = 0.1 days, ~6.0 m^3 m^{-1} day^{-1} for t_p = 1 day, and ~1.2 m^3 m^{-1} day^{-1} for 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 (Fig. 3). The cross-sectional area of the parafluvial zone A_p required to accommodate these parafluvial flows with $\phi = 0.4$ and t_p between 0.1 and 100 days is between 14 and 250 m^2 (Eq. 7). The floodplain of the Avon River is tens of metres wide with sediment thicknesses of several metres and even the higher estimates of the cross-sectional area are not unreasonable given the volume of gravels on the floodplain.

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 ^{222}Rn 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 high-hydraulic conductivity sediments) will likely drive the return of parafluvial waters to the river. By contrast losing reaches are likely to be where the water enters the parafluvial sediments. Given the multiple parameters in Eq. (2) and their inherent uncertainties, however, consideration needs to be given to whether both the ^{222}Rn activities and the increases in streamflow can be accounted for without parafluvial inflows being a significant source of ^{222}Rn.

Matching the ^{222}Rn profile along the Avon River using the parameters discussed above but without input of ^{222}Rn from parafluvial zone would require net groundwater inflows of 28,300 m^3 day^{-1}. However, these inflows exceed the measured increase in streamflow between The Channel and Chinns Bridge of 15,500 m^3 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 m^3 day^{-1} and 200 m^3 day^{-1}, respectively. If these were included, the discrepancy between the calculated and observed streamflow increases. The calculated Cl 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 ^{222}Rn is lost from rivers by degassing and decay, lower

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groundwater inflows are required to replicate the observed ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn activity of groundwater (Cartwright et al., 2011; Cook, 2013). Allowing c_{gw} to vary within the 95% confidence interval of the groundwater ^{222}Rn 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 ^{222}Rn profile and the observed increase in streamflow between The Channel and Chinns Bridge to be reproduced without the requirement for the input of ^{222}Rn from the parafluvial zone (Fig. 8). However, there is no known groundwater in the Avon catchment with such high ^{222}Rn 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 ^{222}Rn 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 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 ^{222}Rn activities in a number of reaches being overestimated (Fig. 8). This is because even assuming no groundwater inflows into those reaches, the loss of ^{222}Rn by degassing is insufficient to explain the observed decrease in ^{222}Rn. Such a poor correspondence between predicted and observed ^{222}Rn activities implies problems with the adopted variables.

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 ^{222}Rn 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 ^{222}Rn 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 ^{222}Rn activities. Similarly, if k = 0.1 day^{-1} a match between the observed and the predicted ^{222}Rn 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 ^{222}Rn activities being overestimated in many reaches (Fig. 8). Increasing k to 0.65 day^{-1} would allow the ^{222}Rn 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 ^{222}Rn activities and streamflows without incorporating parafluvial flow.

It would be possible to explain the observed ^{222}Rn activities and streamflows if there were losing reaches in the Avon River through which significant volumes of river water were lost to the underlying 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 m^3 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 ^{222}Rn 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 ^{222}Rn 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 m^3 day^{-1} (September 2010) and 44,600 m^3 day^{-1} (July 2014). These net groundwater inflows are lower than the measured increases in streamflow between The Channel and Stratford or Chinns Bridge (Fig. 9). However, it is likely that significant parafluvial flow occurs at those times and consequently that these values also represent an overestimation the actual groundwater inflows.

6. Conclusions

The variation in ^{222}Rn activities and Cl concentrations clearly define the reaches of the Avon River that are gaining. The distribution of ^{222}Rn activities also indicate that the location of groundwater inflows changed after major floods that occurred between 2011 and 2013. This approach can be applied to other rivers where flood events change the geometry of the floodplain sediments and where the groundwater monitoring bore network is insufficient to define groundwater-river interaction.

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 ^{222}Rn activities of the river. However, 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 necessarily be the case. Parafluvial flow is likely to be important in rivers with coarse-grained alluvial sediments on their floodplains, especially where there are locally alternating gaining and losing reaches, and must be taken into account in ^{222}Rn mass balance calculations. Unlike hyporheic exchange, which occurs in all stretches, parafluvial inflows are likely to dominantly occur in gaining reaches augmenting the groundwater inflows.

Theoretically, a conservative tracer such as Cl that is unaffected by parafluvial flow could be used to separate groundwater inflows from parafluvial inflows. However, the relatively high variability of 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 other river catchments and illustrates the advantage of using multiple geochemical tracers.

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 ^{222}Rn 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 will be overestimated.

Author Contributions

Both authors were involved in field data collection and lab analyses. Ian Cartwright prepared the manuscript with contributions from Harald Hofmann.

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Figure 1. Summary geological and hydrogeological map of the Avon River catchment (Hofmann and Cartwright, 2015; Department of Environment and Primary Industries, 2015). Arrows show general direction of groundwater flow. Main sampling sites (in order of distance downstream are) are BR = Browns, WF = Wombat Flat, SM = Smyths Road, VA = Valencia, BP = Bushy Park, PL = Pearces Lane, RL = Ridleys Lane, SC = Schools Lane, ST = Stewarts Lane, SA = Stratford, KR = Knobs Reserve, RB = Redbank, CB = Chinns Bridge. Unnamed sampling sites are the additional sites from February 2015 (Table A1).

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 frequency curve for Stratford for streamflows between January 2000 and March 2015 and the percentiles of discharge in the sampling campaigns. Data from Department of Environment and Primary Industries (2015).

Figure 3a. Variation in the ^{222}Rn activity in the parafluvial or hyporheic zone (c_p or c_h) with residence time (t_p or t_h) and ^{222}Rn emanation rate (γ) (Eq. 3). **3b.** Variation in the water flux from the parafluvial zone (I_p) with the flux of ^{222}Rn from the parafluvial zone (F_p) and t_p (Eq. 5). In both cases $c_r = c_{in} = 1000$ Bq m^{-3}.

Figure 4. Downstream variations in ^{222}Rn activities (**4a**) and Cl concentrations (**4b**) for the six sampling 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 (Table A1). Site abbreviations as for Fig. 1.

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

Figure 6a. Calculated and observed ^{222}Rn activities for February 2015 resulting from assigning 50% of the calculated inflows as parafluvial flow. **6b.** Variation in groundwater and parafluvial inflows. **6c.** Calculated streamflow resulting from the groundwater inflows (Eq. 2) vs. measured streamflow at Stratford and Chinns Bridge. **6d.** Predicted vs. observed Cl concentrations. Shaded field is the range resulting from varying groundwater Cl concentrations within the 95% confidence interval.

Figure 7a. Calculated vs. observed ^{222}Rn activities in the Avon River for February 2015 assuming both uniform groundwater inflow within each reach and the situation where groundwater inflow occurs immediately upstream of the measurement point. Site abbreviations as for Fig. 2. **7b.** Calculated groundwater inflows (*I*) 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 interval.

Figure 8. Calculated and observed ^{222}Rn activities for February 2015 resulting from varying individual parameters in Eq. (1). In all cases the new parameters result in significant overestimation of ^{222}Rn activities in many reaches and are unlikely to be realistic. Site abbreviations as for Fig. 1.

Figure 9. Calculated streamflows resulting from groundwater inflows for the sampling rounds excluding February 2015 estimated without parafluvial flow. Aside from the high flow periods (September 2010 and July 2014) the calculated increase in streamflow exceeds the observed streamflow at Stratford and Chinns Bridge. Site abbreviations as for Fig. 1.