Dear Markus

Here are the combined responses and changes made following the comments of the reviewers and editor. The corrected paper with track-changes marked is also included. We have tried to incorporate all of the substantive comments.

Bill Howcroft, Ian Cartwright, Uwe Morgenstern

Editor's comments (response in **blue**)

From my perspective, I would be glad if you could give special attention to the following points:

(1) in the objectives section it would be good to provide an explicit science question together with a research hypothesis. As it stands now, the objectives remain somewhat vague and unspecific.

We have rewritten the Objectives section (Section 1.3, lines 135-148) so that it is clearer what are objectives are and have framed them as a hypothesis. We have also indicted how the objectives inform research elsewhere.

(2) expand the discussion of your results to tie them more into the context of existing work, to better highlight the local and general relevance and to provide a more detailed overview of the limitations of this study.

We have significantly rewritten the Conclusions (Section 6, lines 550-599) so that they concentrate on the broader implications of the study with the more area-specific findings in the Discussion section. We have also reframed the first section of the Introduction (Section 1, lines 30-55) so that it too explains more of the general points and identifies some of the important gaps in our understanding.

(3) as noted by the reviewers, several parts of the results/discussion sections should actually go either into the study site section (catchment attributes - no matter if they were only derived for this study, only the results/discussion derived from these attributes need to go into the results/discussion sections) or into to methods section (e.g. analytical uncertainty).

We did much of the reorganisation, although it does result in the catchment attributes being presented before the methods (now lines 203-209). We also put much of the detail of how the MTTs were estimated including the ^{3}H input function and uncertainties in the Methods (Section 3.4). The discussion of the MTTs themselves and the uncertainties are in the Discussion (Sections 5.2 and 5.3). These are both interpretations of the data and it is also desirable that these sections follow each other and also Section 5.1.

(4) Much is, rightfully, made of aggregation errors. However, I would urgently encourage you to reflect and eventually re-think the concept of "true" MTTs. Of course, catchments do have true MTTs. However, in the presence of aggregation errors we can by no means meaningfully establish what this true MTT is. I understand your intention, but even by assessing the MTTS of smaller sub-catchments of a given catchment, these are not true MTTs, as smaller catchments are very likely also characterized by heterogeneity. What your approach does, it helps to quantify some incremental aggregation error between catchments at different scales, and which may (or may not) give some

idea into which direction the true MTT may converge. But it will never provide you a actual true MTT.

We agree and we have rephrased the sections accordingly (e.g., lines 316-325, 474-493). We have tried to be as honest as possible with our uncertainties (Sections 3.4 and 5.3). The sensitivity analysis incorporated a much greater range in ^{3}H input values than is commonly accounted for. Additionally, we have used varied the parameters in the LPMs more than in many studies. Aggregation is difficult to deal with as there is no simple way to assess it. The way that we approached it may be the worst case scenario as mixing of multiple waters produces apparent MTTs that are closer to the true value than does mixing of only a few end-members. Nevertheless the original wording was probably overoptimistic.

We have tried to put some values of the impact of the uncertainties on the MTTs (lines 494-499) and have also have illustrated while all uncertainties impact the absolute estimates of MTTs, the relative differences between MTTs within catchments at different flow conditions or within a given area are less impacted (lines 500-510). Overall, especially given that aggregation only makes the waters appear younger, the waters must be several years to decades old and the correlation of MTT with streamflow is also robust conclusion. We have stressed this in the paper (lines 550-554)

Reviewer Comments

Our previous responses to the reviewers' comments are below in **blue**, the resultant changes are in **green**

Response to Reviewer #1

- Generalizability

Line 158: "*It is expected that the results of this investigation will facilitate greater understanding of headwater streams not only within the Otway Ranges but in similar catchments worldwide*."

Line 626: "*This study demonstrates a new methodology for estimating groundwater recharge based upon 3H activities in river water.*"

I encourage the authors to reconsider why a potential reader from a different part of the world should read your manuscript? If as you propose in Line 158 your approach will facilitate greater understanding of headwater streams worldwide you should discuss which of your results are general and which are more specific to your landscape. Furthermore, I would reformulate your primary and secondary objective with a stronger focus on generalizability.

If your goal is to develop a novel methodology as written in Line 626 you should make that clear at the beginning of your manuscript, state a clear hypothesis and explain what is new compared to other approaches. However, if you prefer keeping your primary and secondary objectives as they are, which is perfectly valid, you should consider moving this paper to the "cutting-edge case studies", a relative new type of publication form in HESS.

We will revise the Objectives and Conclusions sections of the manuscript to more specifically address which of our conclusions are of relevance globally, and which apply specifically to the Otways region of Australia. Notwithstanding numerous studies over recent years, there is still not a complete understanding of the range of mean transit times (MTT) in headwater catchments nor what controls these. The realisation that MTTs in some Australian catchments are long (years to decades) is significant in their management, which is of local importance. That the MTTs are longer in these catchments than is perhaps commonly recorded elsewhere is important for understanding catchment behaviour more generally and we can emphasise this.

The paper has been restructured to emphasise the more general aspects and separate these from the area-specific conclusions. In particular, we have:

- Emphasised some of the current gaps in knowledge (such as the range of MTTs in headwater catchments globally, and the controls on MTTs) in the first paragraphs of the introduction (lines 30-55)
- Additionally, we have clarified the reasons that understanding MTTs is important (lines 30-48).
- Rewritten the conclusions (Section 6) so that it focusses on the broader outcomes of the study and better articulates how this study relates to our overall understanding of how headwater catchments behave.
- Rephrased the objectives (Section 1.3) so they address specific hypotheses and show how these relate to the broader understanding of the behaviour of headwater catchments.

Demonstrating a new method to estimate groundwater recharge was not a specific goal of this study. Nonetheless, through examination of groundwater volumes across different times of the year, we realised that our data could be used to estimate groundwater recharge to the regional aquifer. We are keen to retain this section as it is novel and potentially of interest to researchers elsewhere. However, it is a relatively minor part of the paper and requires testing in other regions. We chose to include this topic in our manuscript to demonstrate a broader use of MTT estimates. Given that, we consider that this paper constitutes a regular research paper. Further work in this field may be suitable for a “cutting-edge case study” type of publication.

In the end we took that recharge estimate out as it was overly speculative. For some of the catchments it was possible to estimate the turnover volumes of groundwater (which is more conventionally done) (Sections 3.5, 5.5)

- Model Selection

Line 117: “*As a consequence, LPMs must typically be assigned based upon knowledge of the geometry of the flow system and/or information from previous time-series studies.*”

It is interesting that you develop a perception of how you think the catchments are functioning to justify the basic assumptions of your general approach (see major comment 4) and upon which you chose your LPMs. However, in Line 464 you write that it is not possible to assess the most suitable LPM in your study which means that all chosen models are equally likely, doesn’t it? Though, just in the following lines you discuss which LPMs results are more or less realistic. To avoid confusion, I think you should clarify this in your manuscript and clearly state if you can constrain your model results or not.

We have explained this on lines 83-93

It is true that using the approach where MTTs are estimated from individual ^3H activities that one has to assume an appropriate LPM. The potential advantage of using ^3H as a tracer in the southern hemisphere is that it may be used in a similar way to other radioisotope tracers (e.g. ^{14}C or ^{36}Cl), whereby an age or mean transit time estimate can be derived from individual measurements. In turn, this permits estimation of MTTs at a range of flow conditions.

In the northern hemisphere, the use of ^{3}H as a tracer requires time series data collected over several years to estimate MTTs (due to the much larger bomb-pulse ^{3}H signal). Where samples are collected at similar flow conditions (e.g. summer low flows), this permits an independent assessment of the LPM via comparison of the measured and predicted ^{3}H activities. It is questionable, however, whether one could still apply the same time series approach in the southern hemisphere due to the diminution of the relic bomb-pulse ^{3}H activities. For example, the calculated decrease of ^{3}H activities for a water with a mean transit time of 10 years between 2016 and 2026, as predicted by the EPM and DM models used in the paper with the Melbourne ^{3}H record, is only 0.2 TU. Additionally, the time vs. ^{3}H trends produced by the LPMs in the southern hemisphere are similar within analytical uncertainty. Further, given the long MTTs in many southeast Australian catchments, it is not feasible to use other tracers (such as the stable isotopes) to better constrain the LPMs due to initial geochemical variations being attenuated when MTTs are more than a few years (e.g., Stewart et al., 2010, Hydrol. Process., 24, 1646-1659). We addressed these points on lines 96 to 118 of the paper and we will provide a few more details based on the above discussion to clarify our approach.

Our approach was to utilise different LPMs to bracket the estimates of MTTs. These LPMs have been used in many other studies, and where time-series ^{3}H data are available, they do reproduce the observed variation in ^{3}H activities. The LPMs are always a simplification; however, their geometries do agree with the likely form of the flow system and thus the approach is defensible. Estimating precise MTTs is difficult and the not knowing the best LPM to apply represents an uncertainty in these calculations (as we discuss in Section 6.3). However, the conclusions that the water in these streams has MTTs of several years to decades is independent of the LPM that is employed (as was emphasised throughout the paper).

Lines 93-100 & 110-114 explain this

Sections 5.2 and 5.3 now discuss this more explicitly.

As a general point, with the diminishing of the bomb pulse tritium signal, MTTs in the southern hemisphere are not overly sensitive to the models. Because of this, a change in the age distribution that occurs when different LPMs are used do not change the MTT dramatically. In the northern hemisphere, with significant bomb tritium still present, a change in the age distribution significantly changes the fraction of bomb-pulse tritium in the sample and therefore result in a different MTT. This was noted in the comparison of the MMTs from the different LPMs in section 6.2.

This is explained in Section 5.2 (lines 429-437) and Section 5.3 attempts to quantify the resultant uncertainties (lines 494-499).

- System Understanding

Overall, I found the discussion of your MTTs results a little short with respect to your system understanding. I recommend that you discuss in more detail, if and which of your calculated MTTs are realistic in your systems. For instance, are MTTs of 200 years and an annual groundwater recharge rate of 1 % in a headwater catchment of your geology realistic, if considering the hydraulic conductivity, the mean depth and average gradient of the groundwater bodies?

The 200 year value is an absolute maximum and is subject to considerable uncertainty (as we discuss in section 6.3). However, as outlined in the response to other comments below, the conclusion that mean transit times are years to decades is robust. The long mean transit times do imply slow recharge rates. There are only sparse measurements of hydraulic conductivities in these aquifers and, consequently, it is difficult to corroborate the recharge rates using the aquifer properties. The recharge rates are consistent with those generally proposed for eucalyptus forest areas in SE

Australia. For example, Allison & Hughes (1983. Journal of Hydrology 60, 157–173), Allison et al. (1990. Journal of Hydrology 119, 1-20), Herczeg et al. (2000. Marine and Freshwater Research 52, 41-52), and Cartwright et al. (2006, Journal of Hydrology 332, 69-92) estimate that recharge rates in areas dominated by native forest are at most a few mm per year and often less. These low recharge rates are due to the high transpiration rates in eucalypt dominated catchments. Further, because hydraulic conductivities are poorly known in most areas, it is important to find other means to estimate groundwater recharge. However, we recognise that there is potential for groundwater discharge from the catchment via deeper groundwater flow pathways. If this is the case, our estimates would underestimate the true recharge rate because the proposed method accounts only for the discharge at the stream gauge, not total discharge. We will discuss these uncertainties in more detail within the revised paper.

Additionally, the lack of significant near-river alluvial sediments may be a reason why the estimated MTTs are so long. The lack of near-river alluvial sediments precludes the possibility of significant bank storage and return flow contributing to total river discharge and, thus, probably influence the MTTs. We will also discuss this in the revised paper.

As discussed earlier, we removed the original recharge rate calculations from the paper. However, low recharge rates are consistent with previous studies (e.g. Allison et al., 1990) and are common in southeast Australia (lines 553-558).

We have also improved our discussion of the uncertainties in the revised paper (Section 5.3 and Section 6, lines 550-553) and have explained more clearly that despite the uncertainties in the MTT calculations, the observation that the ^{3}H activities are locally 10% of modern rainfall (and much less relative to the bomb-pulse rainfall) necessitates MTTs that are several decades (lines 500-503).

Furthermore, your calculated runoff coefficients vary from 8.6 % to 39 %. This is a pretty large spectrum, especially because the catchments are within the same climate and share a similar land-use (1.3 m of mean annual rainfall / forest cover 78 -95%). Do you have an explanation for this rather strong difference in the hydrological response? Are you seeing these clear differences in the hydrological behaviour also in your MTTs and what conclusions can be drawn from this? Do the basic assumptions you need to make to apply your approach (no significant dilution of groundwater inflow; see discussion point 4) also apply in the two catchments with a runoff coefficient of around 40 %?

The calculation of the runoff coefficient in a region with well measured rainfall and long streamflow records is relatively straightforward. What is less clear are the reasons for the variation. The high runoff coefficients for Upper Lardners, Lardners Gauge and James Access may be because these three rivers drain steeper catchments and are underlain almost entirely by low hydraulic conductivity Otway Group basement rocks.

This is now discussed on lines 342-347.

The runoff coefficients do correlate well with ^{3}H activities and the reason that we included them in this study is that they are useful in providing a first-order estimate of MTTs (in as much as they indicate whether the water is likely to be relatively young or old). The variation in the runoff coefficients is probably controlled by similar factors that control the variation in the MTTs. Catchments with low recharge rates may lose water to the atmosphere by evapotranspiration and consequently have both long MTTs and low runoff coefficients. However, as noted in Section 5.4, it is unclear whether and how catchment attributes such as slope, drainage density control the MTTs (and so by extension the runoff coefficients). A lack of a single catchment attribute controlling MTTs was also noted by Cartwright & Morgenstern (2015. Hydrol. Earth Syst. Sci., 20, 4757-4773) in the Ovens catchment of NE Victoria. In that catchment there was also a good correlation between the ^{3}H activities and the runoff coefficient. We will explain the importance of the correlation in terms of providing first order estimates of MTTs in the revised manuscript.

We have clarified the correlation of ^{3}H (and by extension MTTs) with the runoff coefficient (lines 524-529). We have explained that both are probably controlled by recharge and groundwater flow rates and that the correlation is probably useful in estimating broad differences in MTTs rather than more subtle differences. The general importance of using the runoff coefficient to estimate MTTs is also highlighted (lines 592-595)

As you treat all catchments in a similar fashion, why do you think your MTTs are so different in your catchments? Is it a result of the uncertainty in your models or are the catchments functioning differently and if so, could you identify catchment attributes which might be the reason for this dissimilarity? For instance, the Porcupine creek and the Yahoo share a similar runoff coefficient of 11.4 and 10.5. On the 20/03/2015, you took 3H samples in both catchments. If you calculate the specific discharge in both catchments, it shows that they are not too dissimilar with respect to their runoff generation at that given day. However, your MTTs differ in both catchments from a maximum of 80 years (DM 0.5) to a minimum of 2 years (EPM 3.0) and this is not the only day with such high differences.

The causes of the variations of the MTTs between the catchments remains an open question. In studies elsewhere, catchment attributes such as slope and drainage density were shown to correlate with MTTs. Given the multiple interacting processes that control the transmission of water through catchments (e.g., as discussed by McGuire and McDonnell, 2006; Hrachowitz et al., 2009; Stewart & Fahey, 2010), it is probably not surprising that no single catchment attribute controls mean transit times. Moreover, the lack of correlation confirms that multiple processes control water flux, and that these processes and their interaction are still poorly understood. Similar variations in MTTs between streams are also apparent in the Ovens Catchment (Cartwright & Morgenstern, 2015) and, in that case, the reasons are also not clear. While it is a negative outcome, it is worth us emphasising the lack of correlation between ^{3}H and the catchment attributes in the Discussion section, as it is important.

We have clarified the discussion around not being readily able to determine the controls on the MTTs both in this region and in general (Section 5.4, Section 6 lines 559-576). We have also highlighted this as one of the key gaps in regionalising MTTs in upper catchments (lines 145-147, 596-599).

Overall, given the large differences of your results, I encourage you to connect your research results much stronger with your system architecture and check if these results fit with the knowledge you have from these landscapes. Showing that two systems act differently can be relevant, however, identifying why they act differently is much more interesting for potential readers.

We agree that we can better integrate the results. As we discuss in response to later comments, while the calculation of MTTs has uncertainties (many of which we discuss in Section 6.3), the observation that the ^{3}H activities are far below those of modern rainfall means that the MTTs must be several years to decades. The reasons for the variations in MTTs are not clear, but making that observation is also of general importance.

As above, we have clarified and expanded the discussion the reasons that MTTs differ within these catchments and more generally (Section 5.4, Section 6 lines 559-576).

- The basic assumptions of your approach

Line 428: "*The flow system may therefore be viewed as a continuum that is dominated by older groundwater inflows at low flows and progressively shallower and younger stores of water (such as soil water or perched groundwater) that are mobilised during wetter periods.*"

Line 452: "Whether this reflects changes to the flow system or is due to uncertainties in the MTTs (discussed below) is not certain."

From McGuire and McDonnell (2006) which you cite in your manuscript: "*Most methods are based on early adaptations from the chemical engineering and groundwater fields (e.g., Danckwerts, 1953; Eriksson, 1958; Maloszewski and Zuber, 1982; Haas et al., 1997; Levenspiel, 1999) and may not apply in catchments where there are complex and important controlling processes like variable flow in space and time, spatially variable transmissivity, coupled vertical and lateral flow, immobile zones, and preferential flow, to name a few. These simplifications include one-dimensional transport, time-invariant transit time distributions, uniform recharge, linear and steady-state input and output relations, and contribution from the entire catchment area (Turner and Barnes, 1998).*"

We agree the LPM models are an approximation of real-world situations. Nevertheless, they are commonly used and have successfully predicted variation in tracer concentrations / activities in many catchments. It is generally not possible to constrain all the variations in hydraulic properties in a catchment and all modelling approaches contain some elements of generalisation.

The assumption regarding time-invariance is only correct where mean transit times are calculated from time series measurements (of ^{3}H or other tracers). Because ^{3}H is radioactive, it will yield a mean transit time regardless of whether the catchment is time invariant as long as the flow path geometry remains relatively constant. Further, there is no requirement that water from the entire catchment reaches the stream. The much-used exponential-piston flow model, for example, is applicable to catchments that have both confined and unconfined portions.

Regardless of the uncertainties in the LPM calculations, one can get a general idea of timescales from the ^{3}H activities. If a water with a ^{3}H activity of modern rainfall (~2.7 TU) were collected and isolated, it would take 30 to 40 years for the ^{3}H to decay to the lowest ^{3}H activities recorded in the streams (0.2 to 0.5 TU). Given that the ^{3}H activity of rainfall in the past 50 years was considerably higher, the timescales would be even longer. This is not a real calculation of water age or MTT; however it highlights that ^{3}H is an important qualitative or semi-quantitative tracer over and above its use in the calculations (i.e. waters with low ^{3}H activities are relatively old).

Some of this discussion is in the current version of the paper and we can expand on these points as they are important and perhaps not clear to a broad readership.

We have clarified these points in the revised paper.

- The discussion of uncertainties in MTTs is now more explicit (Sections 3.3 and 5.3)
- The section on LPMs and MTTs in the introduction has been revised to recognise the simplification of LPMs (however, it is still the case that they are probably a more viable alternative to estimating MTTs than alternative methods). Lines 57-65.
- The point that the low ^{3}H activities necessitates long MTTs despite any uncertainties in the calculations is also made explicitly (lines 500-502).

First of all, I would like to highlight that I am not an expert in isotope or tracer hydrology. I apologize for the following comments in advance. Nevertheless, I believe that the following questions, which came across my mind while reading your manuscript, could help readers apart from the tracer community, to better understand your approach.

Similar as it is the case in different unit hydrograph applications, your approach assumes a time invariant and linear input-output relationship of your tracers passing your catchments. However, it has been proven that catchment responses of different kinds are highly non-linear and time variant in several studies over the last 40 years. With respect to runoff predictions, it is nowadays widely accepted that concepts like the unit hydrograph will lead to unrealistic predictions on longer time scales. If we now consider your coarse sampling (3-6 observations in each catchment), the seemingly arbitrary choice of your LPMs and the corresponding parameters as well as the time frames you are working on (up to 233 year/sampling period 1.5 years), it comes to me as no surprise that your model results are so different and highlight how speculative they are.

As noted above, the use of ^{3}H does not assume time-invariance. Also, because the ^{3}H activities in the streams are much lower than those of rainfall, the conclusions that the MTTs must be years to decades are robust. We have been clear throughout the paper that there are considerable uncertainties in the calculated MTTs and have sought to address these where possible. However, the data allows an understanding of the broad mean transit times between and within the catchments, which was the main objective.

We have made our discussion on uncertainties clearer and emphasised what may be concluded with more certainty (the fact that MTTs must be years to decades and the relative differences between different flow conditions in the same catchment). We have also noted the point regarding time-invariance (lines 113-114).

Furthermore, in Line 428 you propose that the flow paths in your system are state dependent. You argue that you couldn't identify significant dilution of groundwater inflow by recent rainfall at the sampling time. However, you miss a detailed explanation how you came up with this fundamental conclusion. I believe, you need to have a rather good understanding of your systems to exclude that flow paths are interacting and especially when your system is switching between the two proposed states (groundwater or soil water dominated). If you have this knowledge why do you not use it to constrain your model results?

The conclusion comes from a variety of observations. Firstly, although we sampled throughout the year and at different flows, we avoided sampling immediately after heavy rainfall when new or event water may be important. Secondly, at the time of sampling, the major ion concentrations in the river do not suggest that there has been dilution between low salinity recent rainfall and older water from within the catchment. The observation that the ^{3}H activities appear to plateau at values that are less than those of rainfall also implies that, during our sampling, the rivers were not dominated by recent rainfall. Finally, during the sampling rounds, there was no overland flow observed in the catchments. Our conceptualisation is that the catchment contains several stores of water ranging from deeper groundwater to shallower soil water that progressively become more important as the catchment "wets up" during the winter months. The observation that the highest ^{3}H activities are similar to those recorded in soil / regolith water in this catchment is also consistent with that idea. Section 6.1 discusses this and we can add some of the above details to explain our reasons more fully.

We have revised Section 5.1 to clarify these points.

If I made some wrong conclusions here about the necessary assumptions you need to make (linearity (superposition principle) and time invariance (your filter shouldn't be time-varying on the scale you are working), I again apologize for these comments. Nevertheless, I suggest a much more comprehensive discussion of the assumptions you need to make to apply your approach in your systems and why you think they are valid on a time scale of decades.

The comments were valuable as many papers are written from a point of assuming a high level of background knowledge. Without turning the paper into a review article, we will broaden the explanation of these issues.

- Minor or technical comments

Line 27 "*The MTT of this ^{3}H activity is approximately ten years, which implies that changes within the catchments, including drought, deforestation, land use and/or bush fire, would not be realised within the streams for at least a decade.*"

Line 604 to 607: "*The reason for the unusually long MTTs is uncertain but could be related to very low aquifer recharge rates and/or high transpiration rates associated with eucalyptus forests (Allison et al., 1990). The long MTTs suggest that short-term events such as drought or bushfire may not impact the streams.*"

How can you exclude that the direct reaction of the stream flow to rainfall (rise of the hydrograph) is not influenced by the named land-use changes as you only analyzed your systems at times where they produced baseflow (following your definition). I would reformulate your statement and make clear what you mean with: "*The long MTTs suggest that short-term events such as drought or bushfire may not impact the streams.*"

That is what we meant to say and we will clarify this in the revised paper. The long MTTs mean that the base flows in the streams are buffered against short-term variations in rainfall (and indeed many of these streams continued to flow through the Millennium drought between 1996 and 2009) but that longer-term climate change will probably impact the catchments.

This is reworded in the Introduction (lines 32-38) and Conclusions (lines 577-584).

Line 431: I do not understand this sentence. Please rephrase.

This sentence notes that if the system does contain more than a single store of water (e.g. old baseflow and young event water), then the calculated MTT gives the minimum age of the baseflow component. We will rephrase it.

We have clarified this statement (lines 417-419).

Line 436: Are the catchments in New Zealand of which you chose one of the EPM ratios of 0.33 similar to the catchments you are working in? Have you chosen the EPM ratio of 3.0 as the minimum exponential flow (25 %) on basis of a catchment property or did you just randomly pick this value?

The catchments have a broadly similar geometry to those in New Zealand. The flow system here and in those examples comprises an unsaturated zone overlying the aquifers which is the basis for the choice of the EPM model (piston flow through the unsaturated zone followed by exponential flow through the aquifer). In recognition that we cannot constrain the suitable LMP, we utilised a range of values for the EPM ratio. An EPM ratio of 3 is a system with 75% piston flow. This may be too high in reality, but it does help limit the calculated range of MTTs.

We have reworded this section to indicate that the range of parameters that we have used is based on catchments elsewhere with similar geometries where time-series data are available (lines 421-425).

Line 443: July 2015 instead of 2014?

The date should be July 2014; we will correct this.

Corrected

Line 442 until 449: Belongs to the method section?

Respectfully, we disagree. Lines 442-449 discuss the MTT results, not how the MTTs were derived. Consequently, we will keep this in the discussion.

Line 455 until 464: Again method section?

As above, this paragraph does not discuss the methodology, but is a discussion of the MTT results. Again, we will keep in this section.

We have reorganised the paper to take into account the comments of both reviewers and editor. Specifically:

- All details of how the MTT calculations were made are now in the Methods as is the explanation of the uncertainties (Section 3.4)
- The actual MTT calculations are in the Discussion (Section 5.2).
- The details of the uncertainties are in the Discussion (Section 5.3).

This makes it clear what is background information (Methods) and what is interpretation (Discussion).

Line 561 until 585: I recommend to rework or remove this entire section. First of all, method, result and discussion parts are entirely mixed. Furthermore, the calculations seem to be widely speculative especially because your estimated MTTs are highly uncertain (see your subsection 6.3.1). I believe a potential reader understands that properly calculated MTTs can be used to estimate the groundwater recharge.

This was removed

As discussed earlier, this was an unintended but nevertheless important finding of this study. Estimating groundwater recharge is difficult and we have proposed a way to estimate it from the MTTs. Estimating groundwater recharge from groundwater MTTs is common; however, we are unaware of anyone attempting it from the MTTs of river water.

Section 6.6: Either you discuss this section in more detail with references to other studies and with a relation to the processes and potential hazards or you remove this section from your manuscript.

It is acknowledged that this section is a minor component of the study. Nonetheless, the data suggest that anthropogenic impacts to several of the streams have occurred and, for this reason alone, is worth mentioning. On a more global scale, these data demonstrate the usefulness of using ^{3}H in water quality studies, much in the way that Morgenstern and Daughney (2012) used ^{3}H activities to assess baseline groundwater quality in New Zealand. We will add more detail in the revised paper.

We integrated this material into the other sections. Specifically, the observations that nitrate correlates with ^{3}H and streamflow are discussed on lines 410-415 and the possible implications of this are on lines 580-585.

Response to Reviewer #2

The paper estimates mean transit times in 6 headwater catchments in southeast Australia using two methods and radioactive ^{3}H tracers. The study is very interesting and provides with an initial overview that stable isotope tracers cannot provide. However, I think the discussion could be more thorough and the structure of the paper be reorganized. Following I write my suggestions to improve this paper and hopefully the authors take them in the best way possible.

We are grateful for the suggestions.

General comments:

1. My first comment is a general concern since it was not mentioned anywhere in the document. Are all 3H activities used on the study normalized? If it was mentioned I missed it.

The ^{3}H activities are absolute values measured against the NIST standard. This is described by Morgenstern and Taylor (2009), which we cite in section 4.2, but we can add this detail to the paper.

This is noted on line 259 as is the definition of a TU

2. I mentioned that the results from this study are a good initial overview because the authors are ignoring the seasonal variation of tritium concentration in precipitation. In Varlam et al. (2016) and Tadros et al. (2014) is shown that seasonal variation is noticeable where autumn-winter precipitation has activities half or lower than spring season precipitation. From Tadros et al. (2014): ”Within the annual cycle, a clear maximum is observed in early spring between August and September and extends into summer, with the minimum concentration occurring in March/April.” The values ranging between 2.4 and 3.2TU are the annual average activities, but if the actual precipitation in March/April was at least half of the measured during those 78 days in July-September, the MTT would increase so much as it did. I understand that resources are not raining and analyzing samples for 3H are expensive, but this should be acknowledged as a flaw of the study and probably causing overestimation of MTT in March 2015.

It is true that ^{3}H activities in rainfall have seasonal variation. However, for waters with long mean transit times, this has little impact on the calculated MTTs unless recharge occurs dominantly during periods when rainfall either has high or low ^{3}H (see discussion in Morgenstern et al., 2010 doi: 10.5194/hess-14-2289-2010). The Otways have high rainfall distributed through the year and consequently there is not a distinct recharge season. The seasonal variation of ^{3}H activities in SE Australia is ~1 TU which is similar to the range of ^{3}H activities that we calculated MTTs for (2.4 to 3.2 TU) and so any potential impact of seasonal recharge is likely to be a similar order of magnitude or less. It does not alter the overall conclusions that MTTs are years to decades. We will include some discussion pertaining to this issue in our revised manuscript.

This has been explained in detail (lines 463-474).

3. The document lacks structure. Even when there are subtitles stating “Methodology”, “Study Area”, “Results” and “Discussion” there are results and methods in the discussion section, as well as study area information in the results section. I will point out in more detail in the specific comments.

We will ensure that the material in in the correct section. We address the reviewer’s specific comments that relate to the structure of the paper below. However, in many cases, we consider that we have the material in the correct sections. Specifically, our results section presented the data and the discussion section interpreted it (which is why the MTTs, catchment attributes, and uncertainties appear here). This is a common, albeit not universal, way of organising papers and is our preference. Perhaps the editor can comment as to their preferred structure for HESS.

As outlined above we significantly reordered the material to take into account the suggestions of both reviewers and the editor.

4. There are a lot of regressions were the only measurement for curve fit is the r2, using the p-value would also add information on the data that is correlated.

Agreed. P-values will be presented in the revised manuscript. While these are useful, they do not change the overall conclusions.

We have included the p-values throughout the paper

Specific comments:

1. P1 L18: 2.4 to 3.2 TU is the annual average value, not the real range of activities, big difference, which might explain partially the low values obtained on the stream water.

As discussed above, there is little evidence for a strong seasonal variation of recharge in this catchment. For catchments with long MTTs, the annual variation in the ^{3}H activities of rainfall has little impact where the MTTs are in excess of a few years. As also noted above, we will add a sentence or two to the discussion (Section 6.3) to explain this. The lowest ^{3}H values of <1 TU are much lower than any recorded in rainfall (either annual averages or seasonal measurements) and consequently, the water must be at least several years old.

We have discussed this in more detail in section 5.3 (lines 463-474).

2. Page 6 Line 148: “agricultural” I think the authors meant “agriculture” or rephrase.

Correct “agricultural” should be “agriculture”. This will be corrected in the revised manuscript.

This has been corrected

3. P7 L169: Is the forest cover in Table 1 only eucalyptus? If so, reference table 1.

No, the catchment percentages of forest cover presented in Table 1 include native eucalyptus as well as production forestry (much of which is eucalyptus). Here, we are providing a general description of the catchments but can add a few more words to clarify this.

We added this detail (lines 156-157).

4. P15 L390 to P16 L410: should be in “Study Area” section.

We disagree. Most of this material was derived as part of this study. While the WMIS website does provide estimates of catchment areas for most of the gauges, the values quoted here are from out GIS analyses. The other catchment attributes discussed here were calculated specifically for this project. The Study Area (section 3) summarises material from previous studies rather than results that arose from this study.

Following the suggestion of the editor, we moved this material to the study area section (lines 203-209).

5. P16 L411-412: What about the correlation with geology? As well as a multiple regression or a PCA?

Noted. There is likely a correlation between runoff coefficient and geology (and/or possibly slope) for three of the catchments (Larnders Gauge, Upper Lardners, and James Access), as these catchments are relatively steep compared to the other catchments and are underlain almost entirely by the low-permeability Otway Group basement rocks. The variation in MTTs is probably controlled by multiple factors and while multiple regression analysis could be carried out, the small number of data points and the large number of potential controlling catchment attributes make it difficult to derive a unique solution (the same holds for other approaches such as PCA).

We noted the lack of correlation with geology (lines 514-517). Given the small number of samples multiple correlations and PCS are not warranted.

6. P16 L413-Fig7: This is a good example where the p-value could give more information, it's easy to see there are two extreme points with higher runoff coefficient that create that "correlation", but if those two would not be there the slope of the correlation would be negative instead of positive.

Agreed that including P-values would be useful. However, as noted above, the conclusions do not change.

We have added the p-values throughout

7. P17 L433-P18 L475: This should be in Results, not Discussion (with few exceptions of a couple of sentences that were discussion).

In the paper we have made the distinction between Results (which reports what is measured) and Discussion (the interpretation of the data). Thus, the ^{3}H measurements are included in the results and the MTTs are included as discussion as these are the interpretation of the ^{3}H data. This is our preferred discussion, although we acknowledge that there is no standard way of doing this (either in HESS or other journals). Perhaps the editor can best advise which structure is the best fit for HESS.

For the reasons outlined above we have kept this in the Discussion.

8. P18 L 476: The authors could make a section called uncertainties in the "Methodology" section with a description of each of them so there is no need to explain them in the "Discussion" section.

Noted. We included a brief description of uncertainties in MTT determination in the Introduction section (Line 108) as they are part of the background to understanding MTTs. This follows a similar format to other papers of ours and other authors. However, we can move this material to the Methodology section.

We moved this material to the Methods section as indicated

9. P19 Eq 2: this equation goes in the "Methodology" section, not discussion. Additionally, the "d" is missing in the equation, which is correctly mentioned in the text afterwards.

This would be better in the Methodology section. The "d" is subscript and this will be corrected in the revised manuscript.

We moved this material to the Methods section as indicated and corrected the equation (Eq. 2).

10. P20 L515-520: As mentioned before, 2.45 TU is probably the high end of activity on the annual precipitation.

There appears to be some confusion here as the ^{3}H activity of 2.4 TU is on the **low**-end rather than the high-end of ^{3}H activities of rainfall for this area (line 239). We agree that there is uncertainty in the ^{3}H activity for modern rainfall. This is why we re-calculated MTTs using a range of ^{3}H activities (2.4 TU to 3.2 TU) which encompasses the range given by Tadros et al. (2014). As we noted in the response to the other reviewer, the assumed ^{3}H activities of modern rainfall make little difference to the calculated MTTs in waters with long MTTs such as these.

We have specified that the ^{3}H activities are weighted mean annual values and that the value of 2.8 TU represents the most likely value but that the possible range may be between 2.4 and 3.2 (lines 300-309).

11. P20 L523: If 2.45 TU is on the high end, for the March calculations the precipitation should be more on the 1-1.3 TU (being conservative).

We are unsure where the ^{3}H activities of 1-1.3 values come from. The measured ^{3}H activity (2.45 TU) in the single precipitation sample that we collected is the lowest recorded ^{3}H activity in rainfall for any area in Victoria, Australia (that we know of). The measured average annual ^{3}H activities (both from our studies and the IAEA datasets that Tadros et al., 2014 quote) are in the range 2.4 to 3.2 (lines 340 to 345). We will ensure that the ^{3}H activities in rainfall are clearly explained in the revised manuscript.

As discussed above, we have clarified the rainfall input (lines 300-309).

12. P20 L530-531: Yes unimportant for the surveys taken in September, partially for those in November and July, I don't think it was unimportant in March.

The March samples have the lowest ^{3}H activities, so the impact of the uncertainties in modern rainfall ^{3}H activities will have the lowest relative impact on the estimated MTTs. This sentence could usefully be expanded to explain the relative impacts more clearly.

We expanded the discussion of uncertainties (Section 5.3) and have also specified which impact the understanding of relative MTTs within and between the catchments (lines 494-510).

13. P21 L539-L544: This should be in the Results section.

We disagree as this is part of the interpretation of the results. We will, however, reword this section so that it better conveys the point that we are interpreting data not presenting new data (e.g., "Given that the analytical uncertainty of the ^{3}H are… …the resultant uncertainties in MTTs are…). The uncertainties were presented in the Methods (Section 4.2) and in Table 3 and we will refer to those sources here.

As well as the reordering of sections, we rephrased Section 5.3 to better convey that we are discussing the consequences of the results here rather than presenting new data.

14. P21 L547-549: I agree that the intermediate flow rates are important, maybe even the

This comment is incomplete, so we are not entirely sure of its meaning. It appears that you are agreeing with our conclusion that the greatest uncertainty in MTT estimates are for waters with intermediate ^{3}H activities. We believe that this is an important conclusion.

15. P21 Eq 3: This equation belongs to “Methodology”, not discussion.

Agreed, we will move it to the Methods section

We moved this as indicated (it is now Eq. 2).

16. P22 Eq 4 and 5: These should be in results.

Agreed, we will move them to the Methods section

This section was not included in the final version (as noted above).

17. P22 L569-582: This belongs to results.

This is a section that interprets the results, thus we consider that it is in the correct section

This section was not included in the final version (as noted above).

18. P22 L588-P23 L592: Discuss why there is no increase on the sulphate concentration in the Ten Mile Creek, are the anthropogenic activities different in this catchment than in the others?

We are unsure as to why there is no correlation between sulphate concentrations and discharge at Ten Mile Creek, when such correlations (including nitrate) do appear to exist at Upper Lardners and James Access. Sulphate concentrations are much higher at Ten Mile Creek than they are at Upper Lardners and James Access (Figure 6), which probably reflects the fact that Upper Lardners and the Gellibrand River at James Access are more pristine streams than Ten Mile Creek. Clearly, more data would help elucidate whether such correlations are real. We will touch upon this in greater detail within our revised text.

As noted above, we integrated this material into the other sections. Specifically, the observations that nitrate correlates with ^{3}H and streamflow in in section 4.3 (where other correlations with major ion geochemistry are discussed) and the possible implications of this are on lines 582-584.

Mean Transit Times in Headwater Catchments: Insights from the Otway Ranges, Australia

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Abstract

Understanding the timescales of water flow through catchments and the ~~origins~~sources of stream water at different flow conditions is critical for understanding catchment behaviour and managing water resources. ~~-~~Here, tritium (^{3}H) activities, major ion geochemistry and ~~discharge~~streamflow data were used in conjunction with Lumped Parameter Models (LPMs) to investigate mean transit times (MTTs) and the stores of water in six headwater catchments ~~of~~in the Otway Ranges ~~in~~of southeast Australia. ~~-~~^{3}H activities of stream water ranged from 0.20 to 2.14 TU, which are ~~far~~significantly lower than those of modern local rainfall (2.4 to 3.2 TU). ~~-~~The ^{3}H activities of the stream water are lowest during ~~the~~ low summer flows and increase with ~~stream discharge. Calculated MTTs~~increasing streamflow. The concentrations of most major ions vary ~~from approximately 7 to 234 years~~little with streamflow which, ~~in many cases, exceed those reported for river systems globally. The MTT estimates, however, are subject to a number of uncertainties, including, uncertainties in~~ together with the low ^{3}H activities, imply that there is no significant direct input of recent rainfall at the ~~most appropriate LPM to use, aggregation errors, and uncertainty~~streamflows sampled in this study. Instead, shallow younger water stores in the ~~modern and bomb pulse ^{3}H activity of rainfall. These uncertainties locally result in uncertainties in MTTs of several years; however, they do not change~~soils and regolith are most likely mobilised during the ~~overall conclusions that the water in these streams has MTTs of several years to decades. There is discharge threshold of approximately 10^4 m^3 day^{-1} in all catchments above which ^{3}H activities do not increase appreciably above ~2.0 TU. The MTT of this ^{3}H activity is approximately ten years, which implies that changes within the catchments, including drought, deforestation, land use and/or bush fire, would not be realised within the streams for at least a decade. A positive correlation exists between ^{3}H activities and nitrate and sulphate concentrations within several of the catchments, which suggests that anthropogenic activities have increasingly impacted water quality at these locations over time.~~wetter months.

MTTs vary from approximately 7 to 230 years. Despite uncertainties of several years in the MTTs that arise from having to assume an appropriate LPM, macroscopic mixing, and uncertainties in the ^{3}H activities of rainfall, the conclusion that they are years to decades is robust. Additionally, the relative

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differences in MTTs at different streamflows in the same catchment are estimated with more certainty. The MTTs in these and similar headwater catchments in southeast Australia are longer than in many catchments globally. These differences may reflect the relatively low rainfall and high evapotranspiration rates in southeast Australia compared with headwater catchments elsewhere.

The long MTTs imply that there is a long-lived store of water in these catchments that can sustain the streams over drought periods lasting several years. However, the catchments are likely to be vulnerable to decadal changes in landuse or climate. Additionally, there may be considerable delay in contaminants reaching the stream. An increase in nitrate and sulphate concentrations in several catchments at high streamflows may represent the input of contaminants through the shallow groundwater that contributes to streamflow during the wetter months. Poor correlations between ^{3}H activities and catchment area, drainage density, landuse, and average slope imply that the MTTs are not controlled by a single parameter but a variety of factors, including catchment geomorphology and the hydraulic properties of the soils and aquifers.

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1. **Introduction**

~~The~~Determining the timescales over which precipitation is transmitted from a recharge area through ~~an aquifer~~a catchment to where it discharges into rivers or ~~springs~~streams (the transit time) is important for understanding catchment behaviour and is of inherent interest to resource managers. ~~Changes~~Streams with long MTTs are connected to ~~the land use within a catchment, including~~ relatively large stores of water in the underlying aquifers (Maloszewski and Zuber, 1982; Morgenstern et al., 2010) that may sustain streamflow during droughts that last up to a few years. However, longer-term changes, such as deforestation~~ and/or~~, agricultural development~~ together with ~~, climate change, and/or landscape change following bushfires~~, drought, deforestation or contaminant loading, can~~ is likely to affect both the quality and the quantity of river flows. ~~Documenting the MTTs allows the timescales over which such changes may affect the streams to be assessed. In recent years, there has been considerable research addressing catchment transit times, for example as reviewed by McGuire and McDonnell (2006) and McDonnell et al. (2010). Much of this research has focussed on understanding transit times within upland (headwater) catchments (e.g. Mueller et al., 2013; Stockinger et al., 2014; Cartwright and Morgenstern, 2015, 2016a).~~

Headwater streams are important ~~for a variety of reasons:~~as they commonly support diverse ecosystems, provide ~~unique~~ recreational opportunities and~~,~~ in many catchments~~,~~ contribute a significant proportion of the total river ~~discharge~~flow (Freeman et al., 2007). Headwater streams also differ from lowland rivers in terms of their potential water inputs. Unlike lowland rivers, which typically receive groundwater inflows from regional ~~groundwater and~~ aquifers or near-river floodplain sediments, the ~~source(s)~~sources of water within headwater streams ~~is~~are far less well understood.

Headwater streams are commonly developed at elevations well above those of the regional water tables and/or ~~are seated upon~~occur on relatively impermeable bedrock. Yet~~,~~ such streams continue to flow~~,~~ even during prolonged dry periods. There are several potential water stores that could contribute to stream flow, including the soil zone, weathered or fractured basement rocks, and/or perched aquifers at the soil-bedrock interface.~~ The relative contribution of such stores to total~~

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~~stream flow has been examined for some decades now (e.g.~~ (e.g. Sklash and Farvolden, 1979; Kennedy et al., 1986; Swistock et al., 1989; Bazemore et al., 1994; Fenicia et al., 2006; ~~and Jensco and McGlynn, 2011). However, the transit times of such stores are less well understood. There are a growing number of estimates of transit times in headwater catchments that range from a few months (e.g.~~ Jensco and McGlynn, 2011). ~~Soulsby et al., 2000; Stewart and Fahey, 2010; Duvert et al., 2016) to several years (Atkinson, 2014; Cartwright and Morgenstern, 2015, 2016a). However, in many headwater catchments, the range of transit times is not well known, nor are the catchment attributes that control the transit times.~~

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~~1.1.~~ Estimates of MTTs in headwater catchments range from a few months to several decades (e.g. Soulsby et al., ~~**Estimation of Mean Transit Times (MTTs)**~~

~~MTTs can be estimated from numerical groundwater models. However, the hydraulic parameters used in such models are seldom known with great certainty and vary spatially, which can lead to unrealistic estimates of MTTs. More frequently, MTTs are estimated using geochemical tracers. These tracers include: stable (O, H) isotopes and major ion concentrations that vary seasonally in rainfall, radioactive isotopes (particularly ^{3}H) and atmospheric gases such as the chlorofluorocarbons (CFCs), SF$_6$, and ^{85}Kr, whose atmospheric concentrations have increased over recent decades (e.g. Cook and Bohlke, 2000; Morgenstern et al., 2010; Kirchner et al., 2010; Yang et al., 2011). Estimation of MTTs is commonly determined via~~2000; McGuire and McDonnell, 2006; Hrachowitz et al., 2009; McDonnell et al., 2010; Stewart and Fahey, 2010; Stewart et al., 2010; Mueller et al., 2013; Stockinger et al., 2014; Atkinson, 2014; Cartwright and Morgenstern, 2015, 2016a, 2016b; Duvert et al., 2016). However, in many regions globally the range of MTTs in headwater catchments is not well known. Additionally, it is not always clear why MTTs vary between different areas. This lack of knowledge limits our abilities to protect and manage headwater catchments.

1.1. Estimating Mean Transit Times (MTTs)

Groundwater follows a myriad of flow paths between the recharge areas to where it discharges into streams or rivers. Consequently, groundwater discharge does not have a discrete age but rather has

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a distribution of transit times. MTTs are commonly estimated using Lumped Parameter Models (LPMs) that describe the distribution of water with different ages or tracer concentrations in simplified aquifer geometries~~. With LPMs, the MTT at the time of sampling is evaluated by comparing the input history of a tracer in precipitation to the measured concentration of that tracer within a stream via the convolution integral~~ (Maloszewski and Zuber, 1982, 1996; Maloszewski et al., 1983; Cook and Bohlke, 2000; Maloszewski, 2000; Zuber et al., 2005). ~~).~~ LPMs represent a viable and commonly-used alternative to estimating MTTs using numerical groundwater models that rely upon hydraulic parameters that are seldom known with certainty and which vary spatially. However, the LPMs are only approximations of actual flow systems and the MTTs may be broad estimates rather than specific values.

The LPMs may be utilised with stable (O, H) isotopes or major ions if the concentrations vary seasonally in rainfall (e.g., Soulsby et al., 2000; McGuire and McDonnell, 2006; Tetzlaff et al., 2007, 2009, Hrachowitz et al., 2009, 2010; Kirchner et al., 2010). Determining MTTs from stable ~~isotopes~~isotope ratios or major ion concentrations relies on tracking the delay and dampening of ~~their~~the seasonal variations between precipitation and discharge. However, use of these tracers typically requires sub-weekly sampling over time periods equal to or exceeding that of the transit times (Timbe et al., 2015). In addition, these tracers become ineffective when transit times exceed 4 to 5 years as the initial variations in rainfall are progressively dampened to below ~~detection limits~~where they can be detected (Stewart et al., 2010).

Gaseous tracers (e.g. ^{3}He, chlorofluorocarbons, SF$_{6}$) are effective in determining residence times of groundwater ~~that is separated from the atmosphere~~ (Cook and Bohlke, 2000) but are difficult to apply to surface water due to gas exchange. ~~-~~With a half-life of 12.32 years, tritium (^{3}H) has been used to estimate MTTs of up to ~~about~~ 150 years (e.g. Morgenstern et al., 2010~~).~~ ; Stewart et al., 2010). Unlike other radioactive tracers (~~such as~~e.g., ^{14}C), ^{3}H is part of the water molecule and its activities are affected only by radioactive decay and dispersion and not by ~~water-rock interaction. Also, because~~geochemical or biogeochemical reactions in the soils or aquifers. Because ^{3}H activities are not affected by processes in the unsaturated zone, the MTTs~~ estimated using ^{3}H~~ reflect both recharge through the unsaturated zone and flow in the groundwater system.

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Utilisation of ^{3}H as a tracer ~~has been~~is facilitated by the fact that the ^{3}H activities of rainfall have been measured globally for several decades (~~IAEA~~International Atomic Energy Agency, 2016~~), including in southeast Australia (Tadros et al., 2014).~~). Due to atmospheric nuclear testing, ^{3}H activities ~~in~~of rainfall peaked during the 1950s and 1960s (the “bomb-pulse~~”), particularly~~”). The bomb-pulse ^{3}H activities in the ~~northern~~Southern Hemisphere ~~(Tadros et al., 2014). As a result, single ^{3}H activities of waters~~were much lower than in the Northern Hemisphere ~~yield non-unique MTTs, although MTTs may still be estimated using time series ^{3}H data. In the Southern Hemisphere, bomb-pulse ^{3}H activities~~ (Tadros et al., 2014) and have now largely declined to ~~levels~~ below ~~that~~those of modern rainfall ~~due to removal by precipitation and radioactive decay~~ (Morgenstern et al., 2010). ~~-~~As a consequence, ~~transit times~~MTTs can~~, in most cases, now~~ generally be determined from single ^{3}H measurements (Morgenstern et al., 2010; Morgenstern and Daughney, 2012) in an analogous manner to how other ~~isotopic tracers~~radioactive isotopes (e.g., ^{14}C or ^{36}Cl) are used in regional groundwater systems. This also allows MTTs at different streamflows to be estimated (Morgenstern et al., 2010; Duvert et al., 2016; Cartwright and Morgenstern, 2015, 2016a, 2016b).

~~Use of~~Using LPMs to ~~evaluate~~estimate MTTs ~~carries~~has a number of uncertainties~~, including deciding on which LPM to employ, aggregation error, the tracer input history, and analytical error. In the past, due to remnant bomb-pulse ^{3}H activities, the choice of LPM had a very large impact on the calculated MTTs. However, the gradual reduction of the bomb-pulse ^{3}H over time allowed the appropriateness of the LPM to be evaluated by time-series ^{3}H measurements (e.g., Maloszewski and Zuber, 1982;~~ .~~Zuber et al., 2005).~~ Due to the attenuation of the ^{3}H bomb-pulse in the ~~southern hemisphere, the calculated MTTs are now less sensitive to the choice of LPM employed. However, this also results in LPMs~~Southern Hemisphere, the suitability of the LPM can no longer ~~being able to~~ be evaluated by time-series ^{3}H measurements (Cartwright and Morgenstern, 2016a~~). As a consequence~~) as is still possible in the Northern Hemisphere (e.g. Blavoux et al., 2013). Hence, LPMs must~~ typically~~ be assigned based upon knowledge of the geometry of the flow system and/or information from previous time-series studies~~.~~ in similar catchments. While not being able to assess the form of the LPM results in uncertainties in

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the calculated MTTs, the MTTs are less sensitive to the choice of LPM than is the case in the Northern Hemisphere (e.g. Blavoux et al., 2013).

Rivers can receive water from numerous stores, including groundwater, tributaries, soil water, and perched aquifers, each of which may have different MTTs. ~~MTTs estimated using geochemical tracers in the aggregated~~ The mixing of water ~~tends to underestimate the actual MTT (i.e.~~from different flow systems potentially produces water samples with a residence time distribution that ~~which would be~~ does not correspond to those in the LPMs and calculated ~~using the weighted average of each store).~~ MTTs are lower than actual MTTs. This is known as the aggregation error (Kirchner, ~~2016a, b~~2016; Stewart et al., ~~2016~~2017) and it increases as the difference between the transit times of the individual end-members ~~also~~ increases. ~~However, for~~For transit times estimated from single ^{3}H activities, the aggregation error decreases with an increasing number of end-members as the mixing of numerous aliquots water with different transit times is similar to what is represented by the LPMs (Cartwright and Morgenstern, ~~2016b).~~2016a).

Despite the uncertainties in calculating MTTs, because the ^{3}H activities of the remnant bomb-pulse waters have largely decayed, Southern Hemisphere waters with low ^{3}H activities have longer MTTs than waters with high ^{3}H activities. This permits relative mean transit times to be readily assessed. Because ^{3}H is radioactive, there is no requirement for flow in the catchment to be time-invariant as long as the flow path geometry remains relatively constant.

1.2. Predicting Mean Transit Times

~~1.2.~~ Fundamentally, MTTs are a function of the recharge rate, length of groundwater flow paths, and rates of groundwater flow, and parameters that control those factors will control the MTTs. Large catchments may have some long groundwater flow paths and consequently have long MTTs (e.g. McGlynn et al., 2003; Hrachowitz et al., 2010). Catchments with higher drainage densities (i.e., higher total stream length per unit area) may contain numerous short groundwater flow paths and consequently have short MTTs (e.g. Hrachowitz et al., 2009). Large groundwater storage volumes will likely also result in long MTTs (e.g. Ma and Yamanaka, 2016).

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Groundwater flow is likely to be more rapid through steeper catchments due to the higher hydraulic gradients, resulting in shorter MTTs (e.g. McGuire et al., 2005). Forested catchments may have higher evapotranspiration and lower recharge rates than cleared catchments (Allison et al., 1990), and the degree of forest cover exerts a control on MTTs (e.g. Tetzlaff et al., ~~**Controls on Mean Transit Times**~~

~~A relatively large volume of work has been conducted to understand the catchment attributes that control MTTs. Being able to identify such controls is important as it would allow~~2007). The hydraulic conductivities of the bedrock and soils are also important in controlling the timescales of water movement through catchments (e.g. Tetzlaff et al., 2009; Hale and McDonnell, 2016).

Identify the controls on MTTs is important for understanding catchment functioning. It also potentially allows first order estimates of MTTs to be made in similar catchments for which detailed geochemical tracer data do not exist. ~~Previous studies have identified catchment size (e.g.~~ In some~~McGlynn et al., 2003; Hrachowitz et al., 2010), groundwater storage volumes (e.g. Ma and Yamanaka, 2016), topography (e.g. McGuire et al., 2005), bedrock permeability (e.g. Hale and McDonnell, 2016), drainage density (e.g. Hrachowitz et al., 2009), forest cover (e.g. Tetzlaff et al., 2007), and soils (e.g. Tetzlaff et al., 2009) as important controls. However, no single attribute has been shown to be the dominant control at all locations. In other~~ catchments, correlations between ^{3}H activities and major ion geochemistry or the runoff coefficient (the proportion of rainfall exported from the catchment by the stream) also allow first order estimates of MTTs to be made (Morgenstern et al., 2010; Cartwright and Morgenstern, 2015, 2016a).

~~2.~~1.3. Objectives

This study ~~focuses on six headwater catchments in~~evaluates the ~~Otway Ranges~~range of and controls on MTTs in headwater streams from the upper Gellibrand catchment of the Otway Ranges in southeast Australia. ~~Largely contained~~Specifically, we test the following hypotheses. Firstly that, in common with headwater catchments elsewhere in southeast Australia, the MTTs are several years to decades. Secondly, that the MTTs are most likely controlled by catchment attributes such as land cover, slope, or drainage density. Lastly, that shallower water stores within the catchment become

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progressively mobilised during higher rainfall periods contribute to streamflow at those times. We also use this study to evaluate whether there are geochemical proxies that could be used to make first order predictions of MTTs at times when no ^{3}H data is available. Documenting MTTs is critical to understanding and protecting headwater catchments and, while this study is based on a specific area, the results have relevance to catchments globally. There is not a complete understanding of the range of MTTs in headwater catchments, nor what controls these. Thus, these are important gaps in our understanding of headwater catchments.

2. Study Area

The Otway Ranges are located in southern Victoria, Australia, approximately 150 km southwest of Melbourne (Fig. 1). The region has a temperate climate, with average rainfall varying from approximately 1,000 mm yr^{-1} at Gellibrand and Forrest to approximately 1,600 mm yr^{-1} at Mount Sabine (Department of Environment, Land, Water and Planning, 2017) (Fig. 1) with the majority of rainfall occurring during the austral winter (July to September). Average potential evapotranspiration is 1,000 to 1,100 mm yr^{-1} and exceeds precipitation during the summer months (Bureau of Meteorology, 2016). The Otway Ranges occur within the Great Otway National Park, ~~the Otway Ranges~~ and hold ecological, cultural, historical and recreational significance. Much of the area is dominated by eucalyptus forest but also includes some commercial forestry, much of which is also eucalyptus.

The geology of the study area is described by Tickell et al. (1991). The basement comprises the Early Cretaceous Otway Group, which consists primarily of volcanogenic sandstone and mudstone with minor amounts of shale, siltstone, and coal. The Otway Group is considered to be a poor aquifer and crops out across most of the Lardners Creek and Gellibrand River Catchments, as well as within the higher elevation areas of the Yahoo Creek and Ten Mile Creek catchments (Fig. 1).

The Otway Group is uncomformably overlain by Tertiary sediments of the Eastern View Formation, Demons Bluff Formation, Clifton Formation and Gellibrand Marl. The Eastern View Formation is composed of three sand and gravel units that collectively form the Lower Tertiary Aquifer. These sediments crop out at various locations across the study area including at the Barongarook High (Fig.

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1), which is the primary recharge area for the aquifer (Stanley, 1991; Petrides and Cartwright, 2006). The Eastern View Formation is overlain by the Demons Bluff Formation, which is a calcareous silt having negligible permeability. The formation crops out sparsely within the study area, mainly along Yahoo and Ten Mile Creeks. Overlying this unit is the Clifton Formation, which is a limonitic sand and gravel aquifer. This unit crops out along Porcupine, Ten Mile, Yahoo and Love Creeks. The Clifton Formation is overlain by the Gellibrand Marl, which consists of approximately 200 to 300 m of calcareous silt. The Gellibrand Marl crops out extensively within the Love Creek and Porcupine Creek catchments and acts as a regional aquitard. Along Love Creek and parts of the Gellibrand River, the Tertiary units have been intruded by the Yaugher Volcanics, which consist primarily of basalt, tuff and volcanic breccia. Deposits of alluvium are present along most of the stream courses, particularly Porcupine Creek and Love Creek.

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Regional groundwater flows from the recharge area in the Barongarook High to the south and southwest (Leonard et al., 1981; Stanley, 1991; Atkinson et al., 2014). Additionally, localised recharge may occur elsewhere across the study area (Atkinson et al., 2014), particularly where the Eastern View Formation crops out. Regional groundwater discharges into the Gellibrand River, Love Creek, Porcupine Creek, Ten Mile Creek and Yahoo Creek (Hebblethwaite and James, 1990; Atkinson et al., 2013; Costelloe et al., 2015). In the higher elevations of the study area, including the upper reaches of Lardners Creek, the regional water table is likely to be below the base of the streambed (Costelloe et al., 2015). Based upon ^{14}C and ^{3}H activities, residence times of the regional groundwater are between 100 and 10,000 years (Petrides and Cartwright, 2012; Atkinson et al., 2014).

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~~In addition, these~~The Gellibrand River (Fig. 1) flows west-southwest for approximately 100 km from its highest point in the Otway Ranges before discharging into the Southern Ocean. This study focuses on six headwater catchments of the upper Gellibrand River: Lardners Creek, Love Creek, Porcupine Creek, Ten Mile Creek, Yahoo Creek and the Gellibrand River upstream of James Access (Fig. 1). The Lardners Creek catchment includes the whole catchment (Lardners Gauge) and a smaller upper subcatchment (Upper Lardners) (Fig. 1). Similarly, Love Creek includes the whole catchment (Love Creek Wonga) and a smaller portion of the upper catchment (Love Creek Kawarren). Porcupine Creek, Ten Mile Creek and Yahoo Creek are also tributaries to Love Creek. Love Creek and Lardners Creek

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flow into the Gellibrand River near Gellibrand (Fig. 1). These headwater streams contribute a significant portion of flow to the Gellibrand River, which ~~acts as a~~in turn provides water~~ source~~ for several towns, supports important aquatic and terrestrial fauna, and provides water for ~~agricultural.~~ agriculture. Current landuse in the upper Gellibrand catchment, including the cleared agricultural land which replaced the native eucalyptus forest, has been established for several decades. Despite their significance, the headwater catchments of the Otway Ranges face a number of threats, including urbanisation, further clearing of native vegetation, drought and bushfire, all of which have the potential to impact the quantity and quality of water within the streams.

~~The primary objective of this study is to determine the MTTs in these headwater streams to enable estimates of groundwater stores, lag times, controls on stream flow generation, and impact of land use on stream water quality. If the streams are to be protected, being able to answer this question is of utmost importance. Secondary objectives include: 1) assessing uncertainties in the MTTs, 2) evaluating potential water inputs into the streams, 3) assessing potential controls on MTTs, 3) investigating possible proxies for ^{3}H, and 4) appraising water quality impacts within the catchments. It is expected that the results of this investigation will facilitate greater understanding of headwater streams not only within the Otway Ranges but in similar catchments worldwide.~~

~~3.1. Study Area~~

~~The Otway Ranges are located in south-central Victoria, Australia, approximately 150 km southwest of Melbourne (Fig. 1). The region has a temperate climate, with average annual rainfall varying from approximately 1,000 mm at Gellibrand and Forrest to approximately 1,600 mm at Mount Sabine (Department of Environment, Land, Water and Planning (DELWP), 2017) (Fig. 1). The majority of rainfall occurs during the austral winter months (July to September) and, during summer months, potential evaporation exceeds precipitation (Bureau of Meteorology, 2016). The Otway Ranges are dominated by eucalyptus forest but include some production forestry.~~

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~~The Gellibrand River is one of the larger river systems draining the region. It flows west-southwest for approximately 100 km from its highest point in the Otway Ranges before discharging into the Southern Ocean near Princetown. This study focuses on six headwater sub-catchments of the Gellibrand River: Lardners Creek, Love Creek, Porcupine Creek, Ten Mile Creek, Yahoo Creek and the Gellibrand River upstream of James Access (Fig. 1). Porcupine Creek, Ten Mile Creek and Yahoo Creek are the main tributaries to Love Creek which, together with Lardners Creek, discharge into the Gellibrand River near Gellibrand (Fig. 1).~~

~~The geology of the study area has been discussed extensively by Tickell et al. (1991). The basement comprises the early-Cretaceous Otway Group, which consists primarily of volcanogenic sandstone and mudstone with minor amounts of shale, siltstone, and coal. The Otway Group is considered to be a poor aquifer and crops out across most of the Lardners Creek and Gellibrand River Catchments, as well as within the higher elevation areas of the Yahoo Creek and Ten Mile Creek catchments (Fig. 1).~~

~~The Otway Group is uncomformably overlain by a sequence of Tertiary sediments comprising the Eastern View Formation, the Demons Bluff Formation, the Clifton Formation and the Gellibrand Marl. The Eastern View Formation is composed of three sand and gravel units that collectively form the Lower Tertiary Aquifer. These sediments crop out at various locations across the study area including at the Barongarook High (Fig. 1), which is the primary recharge area for the aquifer (Stanley, 1991; Petrides and Cartwright, 2006).~~

~~The Eastern View Formation is overlain by the Demons Bluff Formation, which is a calcareous silt having negligible permeability. The formation crops out sparsely within the study area, mainly along Yahoo and Ten Mile Creeks. Overlying this unit is the Clifton Formation, which forms a minor aquifer and is comprised primarily of limonitic sand and gravel. This unit crops out along Porcupine, Ten Mile, Yahoo and Love Creeks. The Clifton Formation is overlain by the Gellibrand Marl, which consists of approximately 200 to 300 m of calcareous silt. The marl crops out extensively within the Love Creek and Porcupine Creek catchments and acts as a regional aquitard. Along Love Creek and parts of the Gellibrand River, the Tertiary units have been intruded~~

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~~by the Yaugher Volcanics, which consist primarily of basalt, tuff and volcanic breccia. Deposits of alluvium are present along most of the stream courses, particularly Porcupine Creek and Love Creek.~~

~~Regional groundwater flows from the recharge area in the Barongarook High to the south and southwest (Leonard et al., 1981; Stanley, 1991; SKM; 2012; Atkinson et al., 2014). Additional, localised recharge may occur elsewhere across the study area, particularly in those areas where the Eastern View Formation crops out. Regional groundwater discharges into the Gellibrand River, Love Creek, Porcupine Creek, Ten Mile Creek and Yahoo Creek (Hebblethwaite and James, 1990; SKM, 2012; Atkinson et al., 2013; Costelloe et al., 2015). In the higher elevations of the study area, including the upper reaches of Lardners Creek, the regional water table is likely to be below the base of the streambed (Costelloe et al., 2015). Based upon ^{14}C and ^{3}H activities, residence times of the regional groundwater are between 100 and 10,000 years (Petrides and Cartwright, 2012; Atkinson et al., 2014).~~

4. ~~Methodology~~

~~Water~~ The six catchments have areas ranging from 9.6 km^2 (Porcupine Creek) to 91.7 km^2 (Love Creek Wonga) (Table 1). Drainage densities are relatively similar and range from 8.7x10^{-4} m m^{-2} at Yahoo Creek to 1x10^{-3} m m^{-2} at Lardners Gauge and Upper Lardners (Table 1). Forest cover is lowest in the Love Creek Wonga (78%) and Love Creek Kawarren (82%) catchments. Forest cover in the other catchments is 88% in the Porcupine Creek and Ten Mile Creek catchments, 91 to 92% in the Lardners Gauge and Upper Lardners catchments, and 95% in the Gellibrand River and Yahoo Creek catchments. Average slopes range from 5.7° (Ten Mile Creek) to 11.3° (at James Access).

3. Methods

~~4.1.~~3.1. Sampling and streamflow

River water samples were collected from eight locations in the catchments (Fig. 1). ~~Two locations were sampled in the~~ Lardners Creek ~~Catchment:~~was sampled at an active gauging station ~~on~~ (Lardners ~~Creek (Lardners~~ Gauge) that is maintained by ~~DELWP~~the Department of Environment, Land, Water and Planning (DELWP) (Site ~~ID~~ 235210) and from the Lardners Creek East Branch (Upper Lardners), ~~located~~ approximately 3.5 km upstream from Lardners Gauge. Love Creek was sampled at

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~~two locations: at Kawarren (Love Creek ~~Kawarren~~), located~~ (Love Creek Kawarren), approximately 1 km upstream of ~~a ~~DELWP gauging station ~~(Site ID ~~235234~~)~~ and at the Wonga Road crossing (Love Creek Wonga), ~~which is located ~~approximately 4.5 km downstream of Kawarren. River water samples were collected from the Gellibrand River, Porcupine Creek, Ten Mile Creek and Yahoo Creek at the sites of former DELWP gauging stations (~~Site IDs~~Sites 235235, 235241, 235239 and 235240, respectively).

Streamflow at the time of sampling was determined for each of the eight locations with the exception of Upper Lardners, which is ungauged. Sub-daily streamflow is currently measured at Lardners Gauge (Site 235210) and at Love Creek (Site 235234) (Department of Environment, Land, Water and Planning, 2017) (Fig. 1). Streamflow at James Access on the Gellibrand River was estimated using a correlation ($R^2 = 0.97$, p-value = 10^{-8}) between streamflow at the former gauging station at this location and that at the existing Upper Gellibrand River gauging station (Site 235202), approximately 7 km upstream (Fig. 1). Likewise, streamflow at the Porcupine Creek, Ten Mile Creek and Yahoo Creek sampling sites was estimated using correlations ($R^2 = 0.95$; p-value = $10^{-6;}$ $R^2 = 0.77$, p-value = 10^{-195} and $R^2 = 0.84$, p-value =10^{-15}, respectively) between streamflow at the former gauging stations at these locations and the Love Creek gauging station.

River water samples were collected from each site in July 2014, September 2014, March 2015 and September 2015. An additional round of river water samples was collected from Lardners Gauge, Porcupine Creek, Ten Mile Creek and Love Creek Kawarren in November 2015. The water samples were collected from close to the centre of the streams using a polyethylene container fixed to an extendable pole. Additional data for ~~the Gellibrand River at ~~James Access is from Atkinson (2014).

A single precipitation sample was collected from Birnam in the Otway Ranges near Ten Mile Creek (Fig. 1) in September 2014 using a rainfall collector. The collector consisted of a polyethylene storage container equipped with a funnel positioned approximately 0.5 m above ground level. Prior to collection of the precipitation sample, the collector had been in the field for 78 days, during which time approximately 198 mm of rainfall was recorded at Forrest while 431 mm of rainfall was recorded at Mount Sabine (~~DELWP~~Department of Environment, Land, Water and Planning, 2017).

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~~4.2. **Discharge Determination**~~

~~Discharge at the time of sampling was determined for each of the eight locations with the exception of the Upper Lardners, which is ungauged. Discharge is monitored by DELWP at gauging stations located on Lardners Creek (Site ID 235210) and at Love Creek (Site ID 235234). At the Gellibrand River sampling site (James Access), discharge was estimated using a correlation ($R^2 = 0.97$) between discharge at the former gauging station at this location and that at the existing Upper Gellibrand River gauging station (Site ID 235202), located approximately 7 km upstream (Fig. 1). Likewise, discharge at the Porcupine Creek, Ten Mile Creek and Yahoo Creek sampling sites was estimated using correlations ($R^2 = 0.95$, $R^2 = 0.77$ and $R^2 = 0.84$, respectively) between discharge at the former gauging stations at these locations and that at the Love Creek gauging station.~~

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~~**Analytical Techniques**~~

3.2. ~~The EC~~Geochemical analyses

The electrical conductivity (EC) and pH of the river water and precipitation samples was measured in the field using a calibrated TPS® hand-held water quality meter and ~~probe~~probes. The EC measurements have a precision of 1 μS/cm. ~~The river water and precipitation samples were analysed for cations, anions and ^{3}H (Supplement).~~ Cation concentrations were measured at Monash University using a ~~ThermoFinnigan~~Thermo Fischer ICP-OES on samples that had ~~first~~ been filtered through 0.45 μm cellulose nitrate filters and acidified to a pH < 2 using double-distilled 16 M HNO_3. Anion concentrations were measured at Monash University on filtered, ~~un-acidified~~unacidified samples using a Metrohm ion chromatograph ~~(IC).~~. The precision of the cation and anion analyses, based upon replicate sample analysis, is ±2% while ~~the~~ accuracy~~,~~ based on analysis of certified water standards~~,~~ is ±5%. ~~Duplicate samples were prepared and analysed at~~HCO_3 concentrations were measured by colorimetric titration with H_2SO_4 using a ~~rate of approximately one per sampling event.~~ Hach digital titrator and reagents and are precise to ±5%. Total dissolved solids (TDS) concentrations were determined by summing the concentrations of cations and anions. Geochemical data is presented in the Supplement.

^{3}H analysis was conducted at the GNS Water Dating Laboratory in Lower Hutt, New Zealand. The samples were vacuum distilled and electrolytically enriched prior to analysis by liquid scintillation counting, as described by Morgenstern and Taylor (2009). ~~^{3}H activities are expressed in tritium units (TU) with a relative uncertainty of ± 2% and a quantification limit of 0.02 TU. Correlations between geochemical variables are discussed below. A reasonably strong correlation is viewed to exist if the correlation coefficient (R^2) is greater than 0.7~~Following further improvements the sensitivity is now further increased to a lower detection limit of 0.02 TU via tritium enrichment by a factor of 95, and reproducibility of tritium enrichment of 1% is achieved via deuterium-calibration for every sample. ^{3}H activities are expressed as absolute values in tritium units (TU) where 1 TU represents a $^3H/^1H$ ratio of 1x10^{-18}. The precision (1σ) is ~1.8% at 2 TU.

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3.3. Catchment Attributes

Catchment attributes (Table 1) were determined using ArcGIS 10.2 (ESRI, 2013) and datasets from (DataSearch Victoria, 2015). The Hydrology Modelling tools in ArcGIS were used to generate the stream network from a 20 m digital elevation model. A threshold catchment area of 50 Ha reproduces the observed perennial stream network of the area. Catchment areas upstream of each sampling site and drainage densities were determined using the watershed tool. Mean slopes were calculated using the Spatial Analysis tools. Vector-based landuse datasets were converted to raster formats and reclassified. Landuse was assigned as forest (native vegetation and plantations) and cleared land, which includes urban and agricultural regions. Runoff coefficients were calculated using streamflow data for each of the catchments (except Upper Lardners) for March 1986 to July 1990 (Department of Environment, Land, Water, and Planning, 2017), the only interval for which contiguous streamflow data are available for each catchment. The runoff coefficient calculations assumed a uniform average annual rainfall of 1.3 m for each catchment (Bureau of Meteorology, 2017). Correlations between catchment attributes and other parameters are considered to be strong where $R^2 \geq 0.7$

~~4.3.~~3.4. Calculating Mean Transit Times

~~Groundwater takes a myriad of flow paths between the recharge areas to where it discharges. Consequently, groundwater does not have a discrete transit time but instead has a distribution of transit times.~~ The ~~MTT may be estimated using LPMs. A number of commonly-used LPMs have been developed (e.g. Maloszewski and Zuber, 1982, 1992; Cook and Bohlke, 2000; Maloszewski, 2000; Zuber~~lumped parameter models implemented in the TracerLPM Excel workbook (Jurgens et al., ~~2005). In each of these models, the concentration of a tracer (e.g.~~2012) were used to estimate MTTs. The ^{3}H~~)~~ activity of water sampled from a stream ~~or bore~~ at time *t* ($C_0(t)$) is related to the input (C_i) of ~~that tracer at the recharge area~~^{3}H via the convolution integral:

~~$C_0(t) = \int_0^\infty C_i(t - T)\, g(T) e^{-\lambda T} dT$~~

$$C_0(t) = \int_0^\infty C_i(t - T) g(T) e^{-\lambda T} dT \qquad (1)$$

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where Ͳ is the transit time, t – Ͳ is the time that the groundwater entered the flow system, λ is the decay constant (0.0563 yr^{-1} for ^{3}H) and g(Ͳ) is the exit age distribution function, for which closed form analytical solutions have been derived (e.g. Maloszewski and Zuber, 1982; Maloszewski and Zuber, 1996; Kinzelbach et al., 2002). MTTs were estimated by matching the predicted ^{3}H activities from the LPMs to the observed ^{3}H activities of the samples.

As discussed earlier, the use of single ^{3}H activities to estimate MTTs requires that an LPM be assigned. ~~In this investigation,~~Here two LPMs were utilised: the Exponential Piston-Flow ~~Mode~~model (EPM) and the Dispersion ~~Model~~model (DM~~). These~~), which are among the most commonly ~~utilised~~used LPMs (McGuire and McDonnell, 2006~~) and are discussed briefly below.~~

; Stewart et al., 2010). The EPM describes flow in aquifers with ~~two segments of flow: a portion with an~~both exponential ~~age distribution,~~ and ~~a~~ piston-flow ~~portion. Conceptually, this model most closely applies to aquifers that are unconfined in the recharge area (the exponential segment) and confined (the piston flow segment) at lower elevations, where there is little to no recharge. The Yahoo Creek, Ten Mile Creek, Love Creek and Porcupine Creek Catchments can potentially~~portions. This model may be ~~described by this model, as recharge to the Lower Tertiary Aquifer occurs in the higher elevations of the catchments, but is limited in lower elevation areas by the presence of the Gellibrand Marl and/or the Demons Bluff Formation. The EPM has also been~~ applied to unconfined aquifers~~, as~~ where recharge through the unsaturated zone resembles piston flow ~~while~~and flow within the aquifer resembles exponential flow (~~e.g. Cook and Bohlke, 2000;~~ Morgenstern et al., ~~2010; Cartwright and Morgenstern, 2015; Cartwright and Morgenstern, 2016a). Utilisation of the EPM requires defining a value for the~~2010). TracerLPM defines an EPM ratio, which represents the relative contribution of ~~the~~ exponential and piston flow ~~model components~~ (Jurgens et al., 2012). The EPM ratio is ~~defined as~~ 1/f - 1, where f is the proportion of aquifer volume exhibiting exponential flow.

The Dispersion Model (DM) is based on the one-dimensional advection-dispersion equation for a semi-infinite medium (Jurgens et al., 2012). While ~~the DM~~this model can be applied to a wide variety of aquifer configurations, conceptually it is probably less realistic than other LPMs. Nonetheless, it has

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been successfully used to predict tracer concentrations over time in a number of flow systems (e.g. Maloszewski, 2000). ~~-~~Utilisation of this model requires defining ~~the value of the~~a dispersion parameter, D_p ~~(~~, which represents the ratio of dispersion to advection~~), which is seldom known *a priori*.~~.

~~MTTs were estimated using TracerLPM (Jurgens et al., 2012) and a ^{3}H record for rainfall modified from~~The ^{3}H input is based on the ~~Melbourne rainfall record. Modern~~ ^{3}H activities of rainfall in Melbourne ~~(located~~, which is approximately 150 km from the study area (International Atomic Energy Agency, 2016; Tadros et al., 2014). ~~) has a ^{3}H activity of approximately 3.0 TU, while~~The data of Tadros et al. (2014) suggest that modern rainfall in the study area has ~~an expected~~ a slightly lower annual weighted average ^{3}H activity ~~of approximately~~ (~2.8 TU ~~(~~) than that in Melbourne, which has a ^{3}H activity ~3.0 TU. Hence, ~~Tadros et al., 2014). Thus, a ^{3}H value of~~ 2.8 TU was ~~utilised for~~ used as the average annual ^{3}H activity of modern (2010 to 2016) rainfall~~,~~ as well as for the years prior to the atmospheric nuclear tests (pre-1951). ^{3}H activities of 9 to 17 month rainfall samples from elsewhere in Victoria are between 2.70 and 2.99 TU (Atkinson, 2014; Cartwright and Morgenstern, 2015 and unpublished data), suggesting that the ^{3}H activity of 2.8 TU is reasonable. The ^{3}H activities for rainfall between 1950 and 2009 ~~were~~ are those of Melbourne rainfall decreased by 6.7% to account for the expected difference in ^{3}H activities ~~within~~in the ~~Otways~~rainfall between the Otway Ranges ~~relative to~~and Melbourne. ~~-MTTs were estimated by matching the predicted ^{3}H activities from the LPMs to the observed ^{3}H activities of the samples.~~

There are several uncertainties in the MTT calculations. The analytical uncertainty ranges between 0.02 and 0.04 TU (Supplement). Tadros et al. ~~**Determining**~~ (2014) proposed that average annual modern rainfall ^{3}H activities were 2.4 to 2.8 TU to the west of the study area and 2.8 to 3.2 TU to the east. To assess the effect of uncertainties in rainfall ^{3}H activities, MTTs were recalculated assuming that modern and pre-1950 rainfall had an average ^{3}H activity of either 2.4 TU or 3.2 TU with the ^{3}H activities of the intervening years adjusted proportionally.

The aggregation or macroscopic mixing of waters also introduces uncertainties (Kirchner, 2016; Stewart et al., 2017). Consider a stream fed by several tributaries. The expected MTT (MTT_e) can be calculated using the streamflow data, ^{3}H activities, and MTTs of each tributary via:

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$MTT_e = a\ MTT_1 + b\ MTT_2 + c\ MTT_3 + ...$ (2)

(Stewart et al., 2017). In Eq. (2), a, b, c, represent the fraction of total flow contributed by tributaries 1, 2, 3. If the aggregation is minimal, MTT_e will be similar to that estimated from the measured ^{3}H activity via the LPM. The successful application of Eq. (2) relies on the MTTs of the different tributaries being defined by their ^{3}H activities (which in itself may not be straightforward due to aggregation within those subcatchments). Nevertheless, it provides a broad estimate of the error due to macroscopic mixing that is otherwise difficult to assess.

3.5. Groundwater Volumes

The volume (V in m^3) of groundwater stored within an aquifer that interacts with the stream (sometimes referred to as the turnover volume) is related to the MTT by:

$V = Q * MTT$ (3),

where Q is streamflow ($m^3\ yr^{-1}$) (Maloszewski and Zuber, 1982; Morgenstern et al., 2010).

~~4.4.1.1. Catchment Attributes~~

~~Catchment attributes were determined using ArcGIS 10.2 (ESRI, 2013) in combination with ground surface elevation contours, bedrock geology, stream courses, and land use data (DataSearch Victoria, 2015). A 20 m digital elevation model (DEM) of the study area was constructed, from which catchment area, drainage density, and average topographic slope for each catchment were determined. In addition, runoff coefficients were calculated using discharge data for each of the catchments (except Upper Lardners) for the period of March 1986 to July 1990, the only interval for which contiguous discharge data are available for each catchment. In the runoff coefficient calculations, an average annual rainfall of 1.3 m was assumed for each catchment.~~

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~~5.~~4. Results

~~5.1. River Discharge~~

4.1. Streamflow

Streamflow was highest during July 2014 (Supplement), ranging from 8.6x10^3 m^3 day^{-1} at Ten Mile Creek to 255x10^3 m^3 day^{-1} at James Access. Discharge was lowest during March and November 2015, ranging from 0.1x10^3 m^3 day^{-1} at Ten Mile Creek to 8.8x10^3 m^3 day^{-1} at James Access. Figure 2 illustrates the ~~discharge conditions under which~~ streamflows for the sampling ~~occurred~~rounds relative to the flow duration curves for ~~each catchment except for Upper Lardners.~~ the catchments. Samples were generally collected between the 10th and 100th percentiles of ~~discharge. Figure 3 shows discharge at Lardners Gauge and Love Creek over the sampling period.~~ streamflow, which encompasses a wide range of flow conditions. Samples were collected during the recession periods after high ~~discharge~~flow events that follow rainfall or during ~~base flow~~baseflow conditions~~.~~ (Fig. 3). Overland flow was not observed during any of the sampling events~~.~~ and small ephemeral tributaries in the catchments were dry.

~~Discharge was highest during July 2014 (Supplement), ranging from 8.6 x 10^3 m^3 day^{-1} at Ten Mile Creek to 255.2 x 10^3 m^3 day^{-1} in the Gellibrand River at James Access. Discharge was lowest during March and November 2015, ranging from 0.1 x 10^3 m^3 day^{-1} at Ten Mile Creek to 8.8 x 10^3 m^3 day^{-1} at James Access.~~

Runoff coefficients range from 33% and 39% at Lardners Gauge and James Access, respectively, to between 9% and 12% at Porcupine Creek, Ten Mile Creek, Yahoo Creek Wonga and Love Creek Kawarren (Table 1). The higher runoff coefficients at Lardners Gauge and James Access relative to the other catchments may be due to the fact that these rivers drain steeper catchments and are underlain almost entirely by low hydraulic conductivity Otway Group basement rocks (Fig. 1).

~~5.2.~~4.2. Tritium Activities

The precipitation sample collected from near Ten Mile Creek in September 2014 had a tritium activity of 2.45 TU, which is near the low end of the predicted range (2.4 to 3.2 TU) of ^{3}H activities of modern

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rainfall for this area (Tadros et al., 2014). ~~-~~This ^{3}H activity is also below the values of 2.70 and 2.76 TU from 9 to 12 month ~~samples of~~ rainfall samples in the Melbourne area (Atkinson, 2014; Cartwright, unpublished data), and 2.85 to 2.99 TU ~~for~~from 9 to 17 month ~~samples for~~ rainfall samples in the Ovens River ~~Catchment~~catchment in northern Victoria (Cartwright and Morgenstern, 2015). The lower than expected ^{3}H activity from the Otway sample is probably due to the sample representing rainfall of only part of the year.

Tritium activities in the ~~river water samples~~rivers are ~~all~~uniformly lower than those of modern rainfall and ranged from 0.20 TU at Porcupine Creek in March 2015 to 2.14 TU at Yahoo Creek in July 2014 (~~Supplement).~~ Fig. 4). The maximum ^{3}H activity (2.14 TU) in the rivers is within the range of ^{3}H activities of 1.80 to 2.25 TU for soil pipe water in higher elevations in the Gellibrand Catchment (Atkinson, 2014). In general, ^{3}H activities were highest at high ~~stream flows~~streamflow (July 2014) and lowest at low ~~stream flows~~streamflow (March and November 2015). ~~The ^{3}H activities of Love Creek were relatively similar between the upstream and downstream sampling locations during each sampling event. At Lardners Creek, ^{3}H activities decreased downstream during the two highest discharges (July 2014 and September 2015) but increased downstream during lower discharges (March and November 2015).~~

~~The range of ^{3}H activities~~The ^{3}H activities of Love Creek at the upstream (Love Creek Kawarren) and downstream (Love Creek Wonga) locations in individual events varied by <0.1 TU. The ^{3}H activities in Lardners Creek between Upper Lardners and Lardners Gauge were slightly more variable (up to 0.17 TU). The range of ^{3}H activities between the events was most variable at Porcupine Creek (0.20 to 1.97 TU), followed by Yahoo Creek (0.43 to 2.14 TU), Love Creek Kawarren (0.48 to 1.91 TU), Love Creek Wonga (0.55 to 1.88 TU), Ten Mile Creek (0.44 to 1.74 TU), Upper Lardners (1.54 to 1.99 TU), ~~the Gellibrand River at~~ James Access (1.73 to 2.08 TU) and Lardners Gauge (1.64 to 1.97 TU) (Fig. 4). ~~Thus, while~~Overall, the highest ^{3}H ~~activity values~~activities were similar across all catchments~~,~~ but the lower ~~values~~^{3}H activities varied considerably. The ^{3}H activities increase with increasing streamflow up to approximately 10^{4} m^{3} day^{-1}, above which ^{3}H activities do not increase appreciably (Fig. 4). Despite differences in catchment size, slope, geology, and, landuse, there is a strong correlation between ^{3}H activities and streamflow across the catchments (^{3}H = 0.2613 ln (Q) + 0.8973; R^{2} = 0.75, p-value = 0.15).

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~~There is a reasonably good correlation (R^2 = 0.75) between ^{3}H activities and discharge (Q) for the catchments as a whole (Fig. 4), whereby ^{3}H = 0.2613 ln (Q) + 0.8973. The ^{3}H activities increase with increasing discharge (Fig. 4) up to a threshold of approximately 10^4 m^3 day^{-1}, above which ^{3}H activities do not increase appreciably above ~2.0 TU. The maximum ^{3}H activity (2.14 TU) in the rivers is less than both the predicted and measured ^{3}H activities of rainfall in southeast Australia. However, it is within the range of ^{3}H activities of 1.80 to 2.25 TU for soil pipe water in higher elevation areas of the Gellibrand River Catchment (Atkinson, 2014).~~

~~5.3.~~4.3. Major Ion Geochemistry

River water geochemistry is similar across all catchments and is dominated by Na, Cl and HCO$_3$~~.~~ (Supplement). TDS concentrations are generally less than 100 mg/L at Lardners Gauge, Upper Lardners and ~~the Gellibrand River at~~ James Access but typically exceed 200 mg/L in Love Creek Wonga, Love Creek Kawarren, Porcupine Creek, Ten Mile Creek and Yahoo Creek. TDS concentrations ~~generally~~ increase downstream ~~at~~in Lardners and Love Creeks and are inversely correlated with ~~discharge~~streamflow in all catchments.

At Love Creek, Ten Mile Creek, Yahoo Creek and Upper Lardners, there is no correlation between ^{3}H activities and EC, TDS or major ion concentrations (Fig. 5). However, at Porcupine Creek, there is a strong correlation (R^2 > 0.95, p-values < 0.01) between ^{3}H activities and EC, TDS, and all major ion concentrations with the exception of chloride, nitrate and sulphate. In addition, there is a ~~relatively~~ strong correlation (R^2 = 0.84, p-value = 0.002) between ^{3}H activities and TDS at Lardners Gauge (Fig. 5).

At Upper Lardners, ~~the Gellibrand River at~~ James Access and Ten Mile Creek, there is a strong correlation (R^2 > 0.90, p-value < 0.15) between nitrate concentration and ^{3}H activities (Fig. 6a). The range of nitrate concentrations (0.08 to 2.0 mg/L) were relatively similar during each sampling event across all catchments except for in July 2014, when nitrate concentrations exceeded 3 mg/L at Love Creek Kawarren and Love Creek Wonga. A similar correlation exists between sulphate concentrations and ^{3}H activities at ~~the Gellibrand River at~~ James Access and at Upper Lardners, but not at Ten Mile

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Creek (Fig 6b). However, sulphate concentrations at these locations are lower than they are in the other catchments.

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~~5.4. **Catchment Attributes**~~

~~Love Creek Wonga has the largest drainage area of the six catchments at approximately 91.7 km^2 (Table 1). This drainage area includes the Love Creek Kawarren, Yahoo Creek, Ten Mile Creek and Porcupine Creek sub-catchments, which have drainage areas of 74.4 km^2, 16.6 km^2, 9.6 km^2 and 33.6 km^2, respectively. Lardners Gauge has a drainage area of 51.6 km^2, which includes the Lardner Creek East Branch (Upper Lardners) sub-catchment with an area of approximately 20 km^2. The Gellibrand River Catchment upstream of James Access has the second largest drainage area of approximately 81.0 km^2.~~

~~Drainage densities within the six catchments are relatively similar and range from approximately 8.7 x 10^{-4} m m^{-2} at Yahoo Creek to 1 x 10^{-3} m m^{-2} at Lardners Gauge and Upper Lardners. Forest cover is lowest in the Love Creek Wonga and Love Creek Kawarren catchments, at approximately 78% and 82%, respectively. Within the remaining catchments, forest cover varies from 88% within the Porcupine Creek and Ten Mile Creek catchments, 91 to 92% in in the Lardners Creek catchments, and 95% in the Gellibrand River and Yahoo Creek catchments. Average slope is approximately 11° in the Lardners Gauge, Upper Lardners and Gellibrand River at James Access Catchments and 8.6° in the Yahoo Creek Catchment. Within the Ten Mile Creek, Porcupine Creek, Love Creek Kawarren and Love Creek Wonga catchments, average slope varies from 5.7 to 6.7°.~~

~~Based upon an average annual rainfall of approximately 1.3 m across all catchments, runoff coefficients range from 33% and 39% at Lardners Creek and the Gellibrand River at James Access, respectively, to 9% to 12% at Porcupine Creek, Ten Mile Creek, Yahoo Creek and Love Creek.~~

~~There are either weak or no correlations ($R^2 \leq 0.6$) between ^{3}H activities and catchment area, drainage density or forest cover (Table 2). However, there are strong positive correlations between ^{3}H activities and the runoff coefficient ($R^2 = 0.94$) (Fig. 7) and between~~

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~~^{3}H activities and average topographic slope (R^2 = 0.87), but only for samples collected during March 2015, when stream flow was generally lowest. However, these correlations are based upon only a small number of samples. Further, the results may be skewed by the data for Lardners Gauge and the Gellibrand River at James Access catchments, which have much higher runoff coefficients and slopes than the other catchments.~~

~~6.~~5. Discussion

The ~~discharge, tritium and~~ combination of streamflow, ^{3}H activities, major ion geochemistry~~ data, in combination with~~, and catchment attributes~~, allow an assessment of MTTs,~~ allows aspects of the behaviour of the upper Gellibrand catchments to be understood. This section addresses the changing stores of water in the catchments, the range and uncertainties ~~in the MTTs, groundwater recharge and water quality impacts~~of MTTs, and whether MTTs can be predicted from catchment attributes or geochemical data.

~~6.1.~~5.1. Sources of ~~Baseflow~~River Inflows

It is important to determine how the water stores that contribute to streamflow change between high and low flows. Groundwater inflows are most probably the dominant source of water during the summer months. However, at times of higher streamflow there may be mobilisation of younger shallower water stores (e.g., water from the soils or the regolith) as the catchment wets up (c.f. Hrachowitz et al., 2013; Cartwright and Morgenstern, 2015, 2016a) or mixing between baseflow and recent rainfall (c.f., Morgenstern et al., ~~Each of the~~ 2010). The river water samples ~~was~~were collected during baseflow conditions or during recession periods after high ~~discharge events. Furthermore, there are few systematic variations in~~ streamflows that follow rainfall (Fig. 3) when recent rainfall is less likely to directly contribute to streamflows. That the major ion geochemistry varies little with ~~stream discharge that would suggest~~streamflow also suggests that there is not significant dilution of groundwater inflows with recent rainfall during the sampling periods~~. The~~ (c.f. Sklash and Farvolden, 1979; Kennedy et al., 1986; Jensco and McGlynn, 2011; Cartwright and Morgenstern, 2015). Additionally the ^{3}H activities plateau at ~2.0 TU, which is significantly lower than those of modern rainfall (Fig. 4).

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Together, these observations suggest that there is no significant direct input of recent rainfall during the sampling periods. The flow system may ~~therefore~~is concluded to be~~ viewed as~~ a continuum that is dominated by older groundwater inflows at low flows ~~and~~while progressively shallower and younger stores of water (such as soil water or perched groundwater) ~~that~~ are mobilised during wetter periods. ~~If this is the case, the system may be modelled using a single LPM. If there were some dilution~~The observations that nitrate and sulphate concentrations in several of the catchments are higher at high streamflows (Fig. 6) may reflect the input of contaminants from recent agricultural activities to the streams. This observation agrees with the conceptualisation that shallower stores of water in the catchment, which are more likely to be impacted by ~~recent rainfall, this approach yields~~contamination, are mobilised during the ~~minimum MTT~~wetter periods of the ~~baseflow component.~~ year.

~~6.2.~~5.2. Mean Transit Times

If the conceptualisation of the flow system is correct, MTTs may be calculated using a single LPM. If there were some dilution by recent rainfall, using a single LPM yields the minimum MTT of the baseflow component (Morgenstern et al., 2010). MTTs in the headwaters catchments were estimated using the EPM and the DM. ~~Initially, an~~For the EPM~~ ratio~~, EPM ratios of 0.33 (75~~ % exponential flow) was utilised, as this value has been shown to be effective~~ % exponential flow), 1.0 (50% exponential flow) and 3.0 (25% exponential flow) were adopted. The EPM model accords with the expected geometry of flow in ~~modelling~~ the catchment (vertical recharge through the unsaturated zone followed by flow along flow paths of varying length), and EPM models with these EPM ratios have reproduced the ^{3}H time series in headwater catchments ~~of New Zealand (~~with similar geometries elsewhere (Maloszewski and Zuber, 1982; Morgenstern and Daughney, 2012~~, Morgenstern et al. 2010). To assess the effects of adopting different LPMs, MTTs were also determined using the EPM with EPM ratios of 1.0 (50 % exponential flow) and 3.0 (25 % exponential flow) and~~ ; Blavoux et al., 2013; Morgenstern et al. 2010). For the DM~~ with~~, D_p values of 0.05 and 0.5~~. This range of Dp values applies to most~~ were adopted, which are appropriate for kilometre-scale flow systems ~~of this scale~~ (Zuber and Maloszewski, 2001; Gelhar et al., 1992). Utilisation of a variety of LPMs allows the impact of the assumed model on the MTTs to be assessed.

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Calculated MTTs ranged from approximately 7 years at Yahoo Creek in July ~~2015~~2014 to ~~234~~230 years at Porcupine Creek in March 2015 (Table 3). In general, the lowest ~~estimates of~~ MTTs were ~~derived using~~estimated from the EPM with an EPM ratio = 3.0 while the highest ~~estimates of~~ MTTs were ~~derived~~estimated using the DM with D_p = 0.5. ~~MTTs estimated with all models were relatively similar for ^{3}H activities greater than ~1.00 TU (Fig. 8). However, as ^{3}H activities decrease below this value, the relative difference between the estimates increases. At the lowest reported ^{3}H activity of 0.20 TU, the relative difference across the range of transit times is approximately 164 years (110%).~~

~~At Lardners Gauge, the Gellibrand River at James Access, Porcupine Creek and Love Creek, the samples collected at the highest flow rates have MTTs that are slightly higher (older) than that of the samples collected at the second highest discharge (Fig. 9). Whether this reflects changes to the flow system or is due to uncertainties in the MTTs (discussed below) is not certain.~~

~~In the individual catchments, MTTs for Lardners Gauge, Upper Lardners and the Gellibrand River at James Access were relatively similar and ranged from approximately 7 to 26 years. In contrast, MTTs for Porcupine Creek ranged from approximately 7 to 234 years, while those for Ten Mile Creek, Yahoo Creek and Love Creek ranged from approximately 13 to 149 years, 7 to 154 years and 10 to 141 years, respectively. In all catchments, the highest (oldest) MTTs are associated with the lowest discharge conditions (March 2015) while the lowest (youngest) MTTs are associated with higher discharge conditions (July 2014 and September 2015) (Fig. 9). The low discharge MTTs at Porcupine Creek, Ten Mile Creek, Yahoo Creek and Love Creek are considerably greater than the average MTT of 15 ± 22 years for headwater catchments worldwide reported by Stewart et al. (2010).~~

~~The MTTs for a given water sample, particularly where ^{3}H activities are less than ~0.5 TU (Fig. 8) vary considerably. However, as discussed earlier, it is not possible to assess the most suitable LPM. The EPM with an EPM ratio of 3.0 and the DM with a D_p value of 0.05 simulate groundwater having a large component of piston flow and, for this reason, are most likely less realistic representations of the flow systems. In contrast, MTTs derived using the EPM with~~

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~~an EPM ratio of 0.33 and the DM with a D_p value of 0.5 are relatively similar across the full range of ^{3}H activities. The EPM with an EPM ratio of 1.0 produces transit time estimates that fall approximately midway between the other four models.~~ Because of the remnant bomb pulse ^{3}H, a few samples with ^{3}H activities between 1.2 to 1.7 TU yield MTTs that are non-unique for models with high piston flow components (i.e., the EPM with EPM ratio = 3.0 and the DM with D_p = 0.05; Table 3, Fig. ~~8).~~8). The choice of the LPM has little impact on MTTs for ^{3}H activities greater than 1 TU (Fig. 8). However, as ^{3}H activities decrease, the relative difference between the MTTs from the different LPMs increases. At the lowest ^{3}H activity of 0.20 TU, the difference between the MTT estimates is approximately 164 years.

MTTs for Lardners Gauge, Upper Lardners and James Access were similar, and are between 7 and 26 years. In contrast, MTTs for Porcupine Creek ranged from approximately 7 to 230 years, while those for Ten Mile Creek, Yahoo Creek, Love Creek Wonga, and Love Creek Kawarren ranged from approximately 13 to 150, 7 to 15, and 10 to 140 years, respectively. In all catchments, the longest MTTs are recorded at the lowest streamflows (March 2015) while the shortest MTTs occur at the highest streamflows (July 2014 and September 2015) (Fig. 9). At Lardners Gauge, James Access, Porcupine Creek and Love Creek, the samples collected at the highest flow rates have MTTs that are slightly longer than that of the samples collected at the second highest streamflow (Fig. 9). Whether this reflects changes to the flow system or is due to uncertainties in the MTT estimates is not certain.

~~6.3.~~5.3. Uncertainties in ~~the~~ MTT Estimates

~~A number of uncertainties exist within the MTT estimates: a) potential aggregation error, b) uncertainty in the ^{3}H activity of rainfall, and c) analytical uncertainty in the laboratory-derived ^{3}H activities. Each of these uncertainties are discussed below.~~

~~6.3.1. Aggregation Error~~

~~Aggregation of water with different MTTs introduces uncertainty in the calculation of MTTs (Kirchner, 2016a, b; Stewart et al., 2016). In general, MTTs calculated from the aggregated water underestimate the MTT that would be calculated from the weighted average of the end-members. Quantifying this potential error is not straightforward, however, as the~~

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~~number of inputs (including tributaries) contributing to total stream flow at a given sampling location is generally unknown, as are the transit times of these inputs. Stewart et al. (2016) indicate that aggregation error becomes significant when MTTs determined using ^{3}H and simple LPMs exceed approximately 6 to 12 years. As most of the MTTs derived in this study are several decades (or longer), it is possible that the calculated MTTs underestimate the true MTTs.~~

~~To evaluate this potential error, true MTTs were estimated for Love Creek Kawarren using the discharge~~The uncertainties in the MTTs arising from the analytical uncertainties (Supplement) range from ±0.9 years for the sample with the highest ^{3}H activity to ±10 years for the sample with the lowest ^{3}H activity. These equate to relative uncertainties of ~±10%. Having to assume an LPM reflects a major uncertainty for calculating the MTTs, especially for waters with ^{3}H activities <1 TU (Fig. 8). For a water with a ^{3}H activity of 2 TU, the uncertainty in MTTs is ±1.2 years (±13%), while for waters with ^{3}H activities of 1 TU and 0.5 TU they are ±5 years (±8%) and ±31 years (±30%), respectively. The EPM with an EPM ratio of 3.0 and the DM with a D_p value of 0.05 have a large component of piston flow and are possibly less realistic representations of the flow systems; however, the differences between the MTTs estimated using the other LPMs are still considerable.

The influence of uncertainties in the ^{3}H input was assessed by varying the ^{3}H activities to encompass the spatial variability described by Tadros et al. (2014) as discussed in Section 2.4. These calculations used the EPM with an EPM ratio of 1.0 but the effect is similar in the other models. The relative difference between MTTs is generally highest when ^{3}H activities exceed 1 TU (Fig. 10). For ^{3}H activities of 2 TU, the uncertainty in MTTs is ±5 years (±54%), while for waters with ^{3}H activities of 1 TU and 0.5 TU they are ±10 years (±15%) and ±5 years (±5%), respectively.

^{3}H activities in rainfall can vary seasonally. If recharge has a strong seasonality, its ^{3}H activities may be different from those of annual rainfall. Rainfall in the Otway Ranges is distributed throughout the year and it is likely that some recharge occurs throughout the year. Less recharge probably occurs during summer due to some rainfall being lost to evapotranspiration. However, as is the case elsewhere in the Southern Hemisphere (Morgenstern et al., 2010), the ^{3}H activities in summer rainfall are closely similar to the average annual ^{3}H activities (Tadros et al., 2014; International Atomic Energy Agency,

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2017). The observation that the ^{3}H activities of summer (December to February) rainfall at Mount Buffalo in northeast Victoria were similar (2.86 TU) to those of two annual rainfall samples (2.99 and 2.85 TU) support this assertion (Cartwright and Morgenstern, 2015). With such a seasonal distribution of ^{3}H activities, the uncertainties in MTTs resulting from using the average annual ^{3}H activities are less than those that arise from the general uncertainty in the ^{3}H input function.

The impact of macroscopic mixing was estimated using Eq. (3) and the streamflow data~~, ^{3}H activities,~~ and MTTs for Porcupine, Ten Mile and Yahoo Creeks~~, whose confluence is located a short distance~~ that flow into Love Creek upstream of ~~the~~ Love Creek Kawarren ~~sampling point. These were then compared with the MTT calculated from the measured ^{3}H activities at that site.~~(Fig. 1). The analysis used the EPM with an EPM ratio of 1.0 (Table 3), but again similar results were obtained with the other LPMs. ~~Inputs from~~Based on the streamflow data, these three streams contribute 77 to 82% of total stream flow at Love Creek Kawarren~~.~~ (Table 3). The remaining portion of flow ~~is~~in Love Creek is assumed to be contributed by undefined inputs ~~that may include both~~such as groundwater inflow and inputs from smaller tributaries. ~~True MTTs~~It was assumed that there was one unidentified input, the ^{3}H activity of which was estimated by the difference between the weighted ^{3}H activities of Porcupine, Ten Mile and Yahoo Creeks and the ^{3}H activity at Love Creek Kawarren ~~were calculated~~. The MTT of this input was determined from the ^{3}H activity using the ~~relationship (modified after Stewart et al. (2016)):~~EPM.

~~MTT$_{LK}$ (true) = a * MTT$_{PC}$ + b * MTT$_{TC}$ + c * MTT$_{YC}$ + MTT$_{UI}$ (2)~~

~~Where a, b, c and d represent the fraction of total flow contributed by Porcupine Creek (PC), Ten Mile Creek (TC), Yahoo Creek (YC) and the undefined inputs (UI), and MTT$_{PC}$, MTT$_{TC}$, MTT$_{YC}$ and MTT$_{UI}$ are the MTTs for these inputs. MTT$_{UI}$ was determined from the calculated ^{3}H activity of the undefined inputs, which was estimated through ^{3}H mass balance and the same LPM.~~

~~During~~ March 2015, the ~~sample~~estimated MTT calculated using the LPM at Love Creek Kawarren ~~over-estimated the true MTT by approximately by approximately~~ was higher than MTT$_e$ calculated using Eq. (3) by 3.7 years or 4% (Table 4). ~~–~~At ~~all~~ other times, ~~sample MTTs~~

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~~underestimated true MTTs by approximately~~ the differences were 3.9 to 7.4 years (18 to 37%). ~~If the system aggregated~~These calculations may not truly address aggregation as there may be more than one unidentified additional store of water and there may be aggregation within the individual subcatchments (which impacts their estimated MTTs). Nevertheless, they do indicate that the potential uncertainties in MTTs due to aggregation are potentially several years (as discussed by Stewart et al., 2017). For waters with similar ^{3}H activities, Cartwright and Morgenstern (2016a) estimated that the aggregation error may be up to 20% where two waters with MTTs of 10 and 50 years or 1 and 5 years mixed but noted that this error became progressively lower if more stores of water with a similar range of ^{3}~~H activities, the aggregation error is likely to be less (Cartwright and Morgenstern, 2016a). While the aggregation error introduces uncertainties, it does not alter the conclusion that the~~ MTTs ~~are years to decades.~~mixed.

~~6.3.2. ^{3}H activity of Rainfall~~

~~There is obviously some uncertainty in the rainfall ^{3}H activities and~~If the uncertainties are uncorrelated, the overall uncertainty is given by the square root of the sum of the squares of the individual uncertainties. Assuming that uncertainties due to analytical uncertainties and aggregation are uniformly 10% and 20%, respectively, and the uncertainties from the range of LPMs and the ^{3}H input of rainfall are as discussed above. For a water with a ^{3}H activity of 2 TU, the overall uncertainty in MTTs are approximately ±60% (±5.4 years), whereas for waters with ^{3}H activities of 1 TU and 0.5 TU they are ±28% (±17 years) and ±38% (±35 years), respectively.

While these uncertainties are considerable, the observation that the ^{3}H activities of the streams are locally 10% of those of modern rainfall (and far less than the rainfall ^{3}H activities at the peak of the bomb-pulse) necessitates that the MTTs must be several decades. Because the aggregation error, which is probably the most difficult to assess, results in MTTs being underestimated (Kirchner et al., 2016; Stewart et al., 2017) some MTTs may be longer than calculated. Relative differences in MTTs between and within catchments may be estimated with more certainty. Because the ~~Tadros et al. (2014) proposed that modern rainfall ^{3}H activities were 2.4 to 2.8 TU to the west of the study area and 2.8 to 3.2 TU to the east. The single rainfall sample from near Ten Mile Creek in September 2014 had a ^{3}H~~

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~~activity of 2.45 TU, which is near the low end of the range. However, this sample was collected over a period of only 78 days and may therefore not be representative of annual rainfall. To assess the effect of uncertainties in rainfall ^{3}H activities, MTTs were recalculated assuming that modern and pre-1950 rainfall had a ^{3}H activity of either 2.4 TU or 3.2 TU with the ^{3}H activities of the intervening years adjusted proportionally. Again, this used the EPM with an EPM ratio of 1.0 but the effect is similar in the other models.~~

~~The relative difference between MTTs calculated from the three rainfall records is generally highest (up to 140%) when ^{3}H activities are greater than ~1 TU but decreases with decreasing ^{3}H activities (Fig. 10). However, the high relative differences in MTTs at ^{3}H activities greater than 1 TU is, in part, offset by low absolute differences. For ^{3}H activities less than ~0.6 TU, the variation in the rainfall input results in less than 4% difference in MTTs. These results indicate that uncertainties in the rainfall ^{3}H activities are relatively unimportant for waters with very low or very high ^{3}H activities.~~

~~The~~ catchments are located in a relatively small ~~geographic area and, for this reason,~~area, the ^{3}H inputs are likely ~~receive rainfall from the same weather systems.~~to be closely similar. Thus, uncertainties in the ^{3}H ~~inputs~~input are thus less likely to ~~be the closely similar~~ impact the comparison of MTTs between catchments. Additionally, as the geometry of the flow system in each catchment~~. If this is the case, uncertainties in the rainfall ^{3}H activities may result in uncertainties in the absolute MTT estimates but will have~~ is unlikely to vary substantially at different streamflows, not being able to assess the suitability of the LPM has less impact on the relative differences in MTTs at different ~~times~~streamflows in the same catchment~~, or between catchments.~~

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~~6.3.3. Analytical Uncertainty~~

~~The ^{3}H activities have a laboratory analytical uncertainty ranging from ± 0.02 to 0.04 TU. The ± 0.04 TU uncertainty for the sample with the highest ^{3}H activity (2.14 TU) results in a maximum uncertainty in the MTT of ± 0.9 years, depending on the LPM utilised. Likewise, the ± 0.02 TU uncertainty for the sample having the lowest ^{3}H activity (0.20 TU) results in a maximum uncertainty in the MTT of ± 10 years. Relative to aggregation error and uncertainty in the rainfall record, analytical uncertainty is relatively minor in significance.~~

~~In summary, the MTTs presented in Table 3 are subject to several uncertainties, including uncertainties about the most appropriate LPM to use, the aggregation error, uncertainty in rainfall ^{3}H inputs, and analytical error. Uncertainties in the LPM and the aggregation error are probably most significant, especially at intermediate flow rates, when ^{3}H activities within the streams are most variable.~~

~~6.4. Variability in MTTs at Porcupine Creek~~

5.4. ~~Between~~Predicting Mean Transit Times

There are weak ($R^2 \leq 0.7$) or no correlations between ^{3}H activities and catchment area, drainage density or forest cover (Table 2). There is a strong correlation between ^{3}H activities and average slope ($R^2 = 0.87$, p-value 0.01) during March 2015, when streamflow was lowest but not at other times. The variability of MTTs from James Access, Lardners Gauge, and Upper Lardners (which occur on the Otway Group: Fig. 1) and from Porcupine Creek, Yahoo Creek, Love Creek, and Ten Mile Creek (which have similar lithologies in their catchments: Fig. 1) indicates the MTTs are not simply related to the geology. A combination of the catchment properties together with the hydraulic properties of the soils and aquifers or evapotranspiration rates that are probably spatially variable and which are difficult to estimate likely controls the MTTs. The observation that relationship between ^{3}H activities and streamflow in all the catchments are similar (Fig. 4) suggests that the MTTs at high flows reflect the

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inflow of water from the shallower water stores which will be largely independent of the catchment attributes.

There is a strong positive correlations between ^{3}H activities and the runoff coefficient ($R^2 = 0.94$, p-value = 0.27) (Fig. 7). This may be due to both the runoff coefficient and MTTs being controlled by the rates of recharge and groundwater flow. The Lardners Gauge and James Access sites have much higher runoff coefficients than the other catchments, and the correlation with ^{3}H activities may reflect the difference between the two groups of catchments. If this is the case, the runoff coefficient may be useful in determining gross rather than subtle differences in MTTs.

EC and streamflow were measured on a monthly basis at the gauging station on Porcupine Creek (Site 235241) between January 1990 and January 1994~~, DELWP measured EC and discharge on a monthly basis at the former gauging station (Site ID 235241) on Porcupine Creek. These data, in combination with a strong correlation ($R^2 = 0.96$)~~ (Department of Environment, Land, Water and Planning, 2017). A strong correlation between MTTs and EC at this location~~, given by~~ (MTT = $1.362e^{0.0061*EC}$~~ allow a first order estimation of MTTs within the stream~~: $R^2 = 0.96$, p-value = 10^{-8}) allows MTTs at this site to be estimated over this four year period (Fig. 11). The estimated MTTs range from ~~approximately~~ 3 to 50 years ~~and exhibit a seasonal pattern whereby~~with the ~~highest~~longest MTTs ~~generally correspond~~corresponding to low~~,~~ summer flows and the ~~lowest~~shortest MTTs ~~correspond to~~during high~~,~~ winter flows. Although based upon a limited number of samples, these results demonstrate the high variability of transit times within the catchment and the value of finding ~~proxy analytes~~proxies for ^{3}H.

~~6.5.~~5.5. Groundwater ~~**Recharge at the Barongarook High**~~volumes

~~The volume of groundwater (V) stored within an aquifer can be estimated from the relationship:~~

~~$V = Q_R * MTT_R$ (3)~~

~~where Q_R represents river discharge and MTT_R is the MTT of the river water (~~The volume of water in the aquifers that contributes to the streamflow may be estimated from Eq. (3). Both the Lardners Gauge and the Love Creek Wonga catchments have active streamflow monitoring, and the

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calculations are carried out for these catchments. Using the relationships between MTT and streamflow (Fig. 9) and streamflow data for 2014 and 2015 (Department of Environment, Land, Water, and Planning, 2017), the average MTT for the two catchments is estimated as 29.7 years (Love Creek Wonga) and 10.8 years (Lardners Gauge). For the average annual streamflow over those two years, the turnover volumes are 2.6x10^5 m^3 (Love Creek Wonga) and 4.5x10^5 m^3 (Lardners Gauge). These volumes are small relative to the likely volumes of water stored in the catchments. For the catchment areas (Table 1) and a porosity of 0.1 to 0.3, which is appropriate for most soils and aquifers, this volume of water could be stored in a layer that is 0.01 to 0.1 m thick.

6. Summary and Conclusions

The calculated MTTs in the six headwater catchments in the Upper Gellibrand catchment of Otway Ranges vary from approximately 7 to 230 years, verifying one of the hypotheses. While there are significant uncertainties in the MTT estimates, the conclusion that they range from years to several decades and are longer at low streamflows is robust. Similar MTTs are recorded in other catchments in southeast Australia (e.g., Cartwright and Morgenstern, 2015, 2016a, 2016b). Especially at low streamflows, the MTTs are far longer than in most headwater catchments worldwide (e.g., Stewart et al., 2010) and are some of the longest yet recorded. The average MTT of 15±22 years calculated by Stewart et al. (2010) was for MTTs based on ^{3}H activities, which makes it directly comparable with MTTs from the south Australian catchments.

Understanding the reasons for the difference in MTTs between catchments in important for understanding catchment behaviour. The catchments in southeast Australia have similar dimensions, slopes, and stream densities to those elsewhere making it unlikely that the differences in MTTs result from catchment geomorphology. The Gellibrand catchments have only thin near-river alluvial sediments thus diminishing the likelihood of bank storage and return flows of young waters during the recession from the high streamflows. However, many headwater catchments globally lack extensive alluvial sediments. The hydraulic properties of the soils and aquifers may also result in slow recharge rates and long MTTs. These are very poorly known and it is difficult to assess their influence.

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Due to the high transpiration rates of eucalyptus forests, recharge rates in Australian catchments are generally lower than elsewhere globally (Allison et al., 1990). However, the observation that there is no correlation between the percentage of forest cover and MTTs in the upper Gellibrand catchments where land clearing occurred several decades ago is problematic for proposing this as a simple control. Despite being in the more temperate region of southeast Australia, the average rainfall in the Otway Ranges of 1,000 to 1,600 mm yr^{-1} is modest compared with upland areas in many parts of the world and the average evapotranspiration rate of 1,000 to 1,100 mm yr^{-1} includes a sizeable component of evaporation (which is more prevalent on the cleared land) (Bureau of Meteorology, 2016). The long MTTs in the catchments from southeast Australia may, therefore, reflect the low rainfall and high evaporation and/or transpiration rates that limit recharge.

The long MTTs are significant for understanding and managing the catchments. Firstly, there are likely to be long-lived stores of water in these catchments that can sustain the streams during droughts that last up to a few years, although longer-term changes (such as land use change or climate change) may eventually affect the streamflows. The long MTTs also imply that any contaminants in groundwater are likely to be released into the streams over years to decades (c.f. Morgenstern and Daughney, 2012). The locally higher nitrate and sulphate concentrations at high streamflows may reflect the input of contaminants from recent agricultural activities to the streams via the younger groundwater that is mobilised at those times.

Even at baseflow conditions, it was not possible to simply predict the MTTs across the catchments from catchment attributes or the geochemistry, although local correlations exist (this refutes one of the hypotheses). The MTTs are most likely controlled by a combination of catchment attributes and also soil properties, hydraulic conductivities, and evapotranspiration rates. This is in keeping with the observation that previous studies have identified correlations between a range of parameters and MTTs (i.e. no single attribute appears to provide the dominant control on MTTs across different regions). Characterising hydraulic properties and evapotranspiration rates on a catchment-wide scale is difficult, which limits the ability to predict MTTs. The runoff coefficient that is a reasonable indicator of MTTs elsewhere in southeast Australia (Cartwright and Morgenstern, 2015) was the best predictor

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of MTTs. This may reflect the fact that both the runoff coefficient and MTTs are controlled by recharge and groundwater flow rates.

This study illustrates that, while broad ranges of MTTs may be estimated using ^3H, precise determination of MTTs is difficult. Additionally, it highlights the challenge in understanding the reasons for the long MTTs in the Australian catchments compared with headwater catchments elsewhere. The potential controls on MTTs is catchments are numerous and more studies in catchments with different climate, landuse, geomorphology, and geology are needed if the desire to be able to predict catchment behaviour regionally or globally is to be realised.

~~Morgenstern et al., 2010). The relationship between~~ ~~MTT_R~~ ~~and~~ ~~Q_R~~ ~~at Ten Mile and Yahoo Creeks is defined by the best fit correlation between the two parameters (Fig. 9):~~

~~MTT = 86.77 *~~ ~~$e^{-2E-04\,Q}$~~ ~~(R^2 = 0.99, Ten Mile Creek) (4)~~

~~MTT = 4847 *~~ ~~$Q^{-0.64}$~~ ~~(R^2 = 0.98, Yahoo Creek) (5)~~

~~Using the above relationships and river discharge at the time of sampling, the volume of groundwater stored within the Ten Mile Catchment was approximately 5,500~~ ~~m^3~~ ~~in March 2015 and 42,000~~ ~~m^3~~ ~~in July 2014. Likewise, at Yahoo Creek, groundwater volumes varied from approximately 15,300~~ ~~m^3~~ ~~in March 2015 to 65,800~~ ~~m^3~~ ~~in July 2014. If it assumed that the difference between these values represents the average volume of water recharged to the aquifer in a year, then groundwater recharge can be estimated from average annual rainfall (approximately 1.3~~ ~~$m\ year^{-1}$~~~~) and the size of the recharge area. If groundwater within the two catchments is recharged entirely through the Eastern View Formation, which has outcrop areas of approximately 3,467,400~~ ~~m^2~~ ~~and 2,588,900~~ ~~m^2~~ ~~respectively, groundwater recharge is approximately 0.8 % (11~~ ~~$mm\ year^{-1}$~~~~) in the Ten Mile Creek and 1.5 % (20~~ ~~$mm\ year^{-1}$~~~~) in the Yahoo Creek catchments~~

~~The above calculations were based on the MTTs from the EPM with an EPM ratio of 1.0. If an EPM ratio of 3.0 is utilised, the same recharge rates are obtained. Using the DM and a~~ ~~D_P~~ ~~value of 0.5 leads to recharge estimates of 1.3 % and 1.4 %. These recharge estimates are considerably less than those estimated by Leonard et al. (1981) at 17 %, Witebsky et al.~~

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~~(1992) at 8 %, and Teng (1996) at 9 %. However, they are comparable to those derived for other parts of southeast Australia (e.g. Cook et al., 1994; Cartwright et al., 2007). This exercise demonstrates the potential for using MTTs to estimate groundwater recharge.~~

~~6.6. **Impacts to River Water Quality**~~

~~Nitrate concentrations increase with a corresponding increase in ^{3}H activities at Upper Lardners, the Gellibrand River at James Access and Ten Mile Creek. A similar increase in sulphate concentrations is apparent at the Gellibrand River at James Access and at Upper Lardners. These trends suggest increasing impacts to river water quality as a result of anthropogenic activities within the catchments upstream of the sampling points.~~

~~7. Conclusions~~

~~MTTs in the six headwater catchments in the Otway Ranges vary from approximately 7 to 234 years. There are a number of uncertainties in these MTT estimates. Some, such as the uncertainty in the rainfall ^{3}H, impact all of the catchments as a whole and will thus not result in major uncertainties in relative MTTs between catchments or within a single catchment at different flow conditions. Likewise, uncertainty in the most suitable LPM will affect the comparison of MTTs between catchments but not within the same catchment at different flow conditions. Aggregation error is of a similar magnitude to many of the other uncertainties and is more difficult to assess. Despite these uncertainties, that the MTTs are several years to decades remains a robust conclusion. This would place them amongst the oldest of any yet estimated globally.~~

~~The reason for the unusually long MTTs is uncertain but could be related to very low aquifer recharge rates and/or high transpiration rates associated with eucalyptus forests (Allison et al., 1990). The long MTTs suggest that short-term events such as drought or bushfire may not impact the streams. However, longer-term changes within the catchments, such as land use change, climate change or contaminant loading, may affect the streams but not for many years. An example of this is increasing nitrate and sulphate concentrations within~~

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~~several of the catchments, which implies increasing impacts to river water quality as a result of anthropogenic activities.~~

~~There is a strong correlation between ^{3}H activities and EC, major ion concentrations, and/or TDS at Porcupine Creek and between ^{3}H activities and TDS at Lardners Gauge. These relationships allow a first order estimate of ^{3}H activity and, therefore, MTTs at either of these two locations using a single water quality measurement. More broadly, ^{3}H activities within any catchment can be estimated using a simple ^{3}H-discharge relationship, which is characterised by a discharge threshold of approximately 10^{4} m^{3} day^{-1}. Despite differences in geology, catchment size, land use, drainage density, runoff, and slope, this ^{3}H-discharge relationship implies that the headwater streams in the Otway Ranges behave in a relatively uniform fashion. This further implies that the dominating control affecting the variability in ^{3}H activities is the relative contribution of groundwater and soil water, rather than physical catchment attributes.~~

~~The ^{3}H activities of the river water samples, in combination with a correlation between MTTs and river discharge, suggest that recharge to the regional aquifer is within the range of 0.8 to 1.5%. These values are lower than estimates provided by previous researchers but are in line with recharge estimates made in other parts of southeast Australia. This study demonstrates a new methodology for estimating groundwater recharge based upon ^{3}H activities in river water.~~

Data Availability

All geochemistry data utilised in this study are contained in the Supplement. ~~River discharge~~Streamflow data and historic EC data for Porcupine Creek are publicly available from the Victorian State Government, Department of Environment, Land, Water & Planning (DELWP), Water Measurement Information System (http://data.water.vic.gov.au/monitoring.htm~~)/~~).

Author Contributions

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William Howcroft undertook the sampling program and oversaw the analysis of the geochemical parameters and the MTT calculations. Uwe Morgenstern was responsible for the ^3H analysis. The manuscript was prepared by William Howcroft, Ian Cartwright and Uwe Morgenstern.

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Figure Captions

Fig. 1. Map of study area showing catchments, sampling locations and bedrock geology. Inset map shows location of study area in Australia. Source: DataSearch Victoria (2015). LG = Lardners Gauge, UL = Upper Lardners, JA = Gellibrand River at James Access, PC = Porcupine Creek, TC = Ten Mile Creek, YC = Yahoo Creek, LK = Love Creek Kawarren, and LW = Love Creek Wonga. Current or discontinued gauging stations exist at all sites except for Upper Lardiners.

Fig. 2. Streamflows at which samples were collected relative to flow duration curves for Lardners Gauge (**2a**), Gellibrand River at James Access (**2b**) – additional data (black circles) from Atkinson (2014), Porcupine Creek (**2c**), Ten Mile Creek (**2d**), Yahoo Creek (**2e**) and Love Creek (**2f**) Streamflow data from Department of Environment, Land, Water and Planning (2017).

Fig. 3. Hydrographs for Lardners Gauge (**3a**) and Love Creek (**3b**) together with the timing of sample collection. Data from Department of Environment, Land, Water and Planning (2017).

Fig. 4. ^{3}H activities of stream water as a function of streamflow for all catchments except Upper Lardners which is ungauged. ^{3}H data from Supplement, streamflow data from Department of Environment, Land, Water and Planning (2017) or calculated as discussed in the text. Shaded boxes show predicted range of rainfall ^{3}H activities from Tadros et al. (2014) and soil waters from Atkinson (2014).

Fig. 5. ^{3}H activities as a function of TDS for all catchments (data from Supplement). Strong inverse correlations between ^{3}H activities and TDS exist for Lardiners Gauge and Porcupine Creek.

Fig. 6. ^{3}H activities as function of nitrate concentrations (**6a**) and sulphate concentrations (**6b**). Data from Supplement. Strong ($R^2 > 0.7$) correlations indicated.

Fig. 7. ^{3}H activities vs. runoff coefficients for the March 2015 samples (data from Table 1 and Supplement). Although a strong correlation ($R^2 = 0.94$) exists, it may be a result of the grouping of the samples.

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Fig. 8. Estimated MTTs vs. ^{3}H activities in the stream waters calculated using the Exponential Piston Flow Model (EPM) with EPM ratios of 0.33, 1.0 and 3.0 and the Dispersion Model (DM) with Dp values of 0.05 and 0.5. Data from Supplement and Table 3.

Fig. 9. MTTs calculated using the EPM model with an EPM ratio of 1.0 (Table 3) as a function of streamflow (Q) for Lardners Gauge (**9a**), Gellibrand River at James Access (**9b**) - black circles are data from Atkinson (2014), Porcupine Creek (**9c**), Ten Mile Creek (**9d**), Yahoo Creek (**9e**), and Love Creek (**9f**) - blue circles are Love Creek Kawarren and red circles Love Creek Wonga. Curves are exponential trend lines. Streamflow data from Department of Environment, Land, Water and Planning (2017) or calculated as discussed in the text

Fig. 10. Impact of varying rainfall ^{3}H inputs on MTTs calculated using the EPM model with an EPM ratio of 1.0. The three rainfall inputs modern and pre bomb-pulse ^{3}H activities of 2.4, 2.8, and 3.2 TU and the ^{3}H activity of the bomb-pulse rainfall was varied by a similar proportion as discussed in the text.

Fig. 11: Variation in MTT as a function of streamflow at Porcupine Creek for January 1990 to January 1994 calculated using the relationship between EC and ^{3}H activity (Supplement) and monthly EC data from the Department of Environment, Land, Water and Planning (2017). Streamflow data also from Department of Environment, Land, Water and Planning (2017).

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