

We thank the reviewers for their helpful comments. As requested by the Associate Editor, we have made the conclusions to the paper more specific and discussed how our study here is important for a general understanding of processes (e.g. lines 818-850). Our responses are below (in blue), and line numbers refer to the track-changes version of the paper. We also reorganised various sections of the paper by

- Moving the objectives earlier in the Introduction
- Moving the sections on mean residence times and recharge rates into the Discussion (as these now include more discussion that interprets the results)
- Moving the more general aspects of the discussion to the Conclusions

### **Reviewer 1**

How does this study inform the sustainable management of groundwater (from the opening line of the abstract)? The description of the study area does not mention groundwater use lower in the catchment, or reference to estimate of sustainable yields on a larger scale. A context of groundwater management issues in the region is not provided.

The study took place in two catchments instrumented to study the effects of land-use changes on groundwater and surface water systems and resources. The aims of our study were a better understanding of the uncertainties and limitations of commonly-applied recharge methods using these well-instrumented catchments as examples. As we explained in the Introduction (lines 75-92), estimating recharge is important to understanding the hydrogeology of semi-arid regions and this study has relevance to other regions where these techniques are used. In particular, as discussed below, the WTF method remains widely used but is prone to large uncertainties.

Groundwater is not extensively used in the catchments studied, and our comments on sustainability were a more general recognition that groundwater can be a vital resource in semi-arid areas. We have removed these points from the Abstract, Introduction and Conclusions.

In terms of the specific need to understand recharge in these catchments, defining whether recharge rates changed with land-use changes is important (especially the potential impacts on waterlogging and streamflow caused by changing water table elevations). We clarified these aims in the Introduction (lines 121-129) and have better reflected the outcomes of this study in the Conclusions (lines 818-830).

How do the authors reconcile their view of the importance of recharge estimates with the ‘water budget myth’? A related myth that sustainable development of groundwater resources can be defined by groundwater residence times has recently been highlighted by Ferguson et al. 2020, citing classic papers on the water budget myth.

This is beyond what we can easily discuss in this paper. We agree that defining sustainable yields is difficult (and these may be a flawed concept); however, understanding recharge is a critical part of assessing the impacts of groundwater use (e.g., Gleeson et al., 2012, Nature,

11295). We have removed the references to sustainability from the introduction and conclusions as it is not something that is specifically discussed.

The use of “residence times” also differs between this paper and Ferguson et al. (2020). We use it to refer to individual samples rather than the average age of water in the aquifer (sometimes called the turnover time). We avoid using the term “age” for specific samples as it is not a valid concept for most groundwater and gives a misleading impression (Suckow et al., 2014, *Applied Geochemistry*, 50, 222–230).

Specific suggestions are provided below.

1) The objectives of the study were to examine uncertainties in varying methods of estimating recharge. However, there is no discussion of how the method comparison is similar or distinct from other recharge studies in semi-arid areas. Have other studies also found the WTF method overestimates recharge for example?

This was mentioned in the original version of the paper (Section 5) but we have moved this to the Conclusions (lines 797-830) to give it more prominence. We have made reference to previous studies (Cartwright et al., 2007, *Journal of Hydrology*, 332, 69-92; Crosbie et al., 2010, *Hydrology and Earth System Sciences*, 14, 2023-2038; Dean et al., 2015, *Hydrology and Earth System Sciences*, 19, 1107–1123; Perveen, 2016, <http://hdl.handle.net/1959.9/560005>) that show that the WTF method generally overestimates recharge rates, mainly due to specific yields being overestimated (for reasons discussed in Section 5.2.1).

2) Comparing methods for recharge rates is interesting, but the authors argue (Line 481 in the previous version) that it is ‘fundamentally important to assess the impacts of land clearing’. Why?

Understanding the recharge changes is important for understanding the rise of the water table and consequent impacts of salinization of soils and rivers. We have indicated that in the Introduction (lines 80-90) and discussed it more explicitly in the Discussion (Section 5.3, lines 740-797) and the Conclusions (lines 840-850).

Inevitably, understanding the impacts of land clearing necessitates using different methods to understand pre- and post-land clearing recharge. Pre-land clearing recharge is commonly estimated using the CMB or longer-lived radioisotopes (e.g.,  $^{14}\text{C}$ ). Modern recharge may be estimated using the WTF method or  $^3\text{H}$ . However, this assumes that the rates from those different methods are all broadly correct (or at least comparable), which is probably not the case (as we discussed in the Conclusions).

3) Section 5.1 (in the previous version) on the impacts of reforestation only considers the TRR method, which surprisingly does not find a significant difference in recharge between pasture and forest. Other evidence indicates the forest is using more water, so the study appears to demonstrate the limitations of recharge estimation methods?

The area was partially replanted in the past ~20 years. We discussed that the WTF method overestimates recharge rates and CMB method yields long-term recharge rates.  $^3\text{H}$  activities and the TRR method should be applicable to understanding the initial land-use changes, and

the recharge rates from this technique is higher (as is discussed in Section 5.3). There are two reasons why the TRR method may not detect changed to recharge rates following reforestation. Firstly, recharge estimates based on groundwater geochemistry are probably averaged over areas of a few 10's to 100's m<sup>2</sup> (Scanlon et al., 2002, Hydrogeology Journal, 10, 18-39). Commonly, the bores in plantation forests are in cleared areas that are larger than this, meaning that the recharge rates may not be representative of the whole plantation (we discussed this on lines 824-830). Secondly, the TRR method averages recharge rates over several years to decades, and so it may not yet detect the latest reforestation. We discussed this in Section 5.3 (lines 790-796).

4) How do the authors recommend these results inform groundwater modelling? Line 495 (in the previous version).

Popular groundwater models, such as MODFLOW, use recharge rates as a boundary condition at the water table. Isotope methods give estimates of recharge rates across large areas over a relatively long period of times dating back hundreds of years. Because the WTF method estimates recharge rates at smaller spatial scales for the years when data are available, it is often considered to be more appropriate to constrain the models. However, the large discrepancies in the values of recharge rates from different techniques and the overestimated recharge rates with the WTF method pose questions on the quantification of boundary conditions for groundwater models. We have noted that care must be used in assigning recharge rates as boundary conditions numerical models (lines 844-850).

As also suggested in the Conclusions (lines 831-838), the use of integrated surface and subsurface hydrogeologic models might overcome this issue and provide an additional tool for the estimation of recharge rates to support experimental analyses.

5) Both WTF and TRR rely on estimating the effective porosity (or effective specific yield). Mean porosity was previously reported as 0.15 and 0.1 respectively for the pasture and forested catchments but is unclear how this was determined, and how sensitive the WTF and TRR methods are to the range of possible values.

The values of porosity were taken from the previous study in this area (Adelana et al., 2014, Hydrological Processes, 29, 1630-1643). The effective porosity from 0.03 to 0.1 seems reasonable given the nature of the aquifer materials (e.g. Morris and Johnson, 1967, U.S. Geological Survey Water-Supply Paper 1839-D, 42p). The uncertainties in the recharge estimates from TRR and WTF using the values of effective porosity and effective specific yield are discussed explicitly in Sections 5.2.2 and 5.2.3.

Line 385 (in the previous version) states  $S_y$  is 'not well known' which is an understatement, as the parameter is highly uncertain. There is also a possibility of semi-confined conditions to develop at very shallow depths, and that hydraulic loading could account for part of the water level response to rainfall.

We agree that specific yield is highly uncertain, as was discussed in the paper. Given that it is rarely measured and is not an invariant property, it is not surprising that there are errors in the WTF method. This is discussed extensively in the paper (Sections 1.2, lines 232-239, and

5.2.2, lines 687-693). As discussed in response to the other reviewers, we have reduced the discussion of specific yield in the Introduction (Section 1.2, lines 232-239). This material is well covered by previous studies (e.g., Gillham, 1984; Sophocleous, 1985; Healy and Cook, 2002; Crosbie et al., 2019); however, it is still the case that most recharge studies that use the WTF method make the oversimplifying assumption that the specific yield is spatially and temporally uniform (which is the point that we were trying to get across)

If semi-confined conditions developed near the surface, one would expect rapid increases in the levels of WT following large rainfall events. We do not see the fast response to rainfall events, and the changes in levels of WT are seasonal. We discussed this in Section 5.2.2 (lines 675-677).

6) CMB method is most reliant on the assumption of the long-term rate of CI delivery and can only be applied in catchments with negligible runoff and sedimentary CI inputs. How are the results sensitive to 8% runoff measurement from the catchments?

Like most semi-arid catchments, our study sites have minor surface water outflows. Not accounting for the export of CI in surface water can lead to recharge rates being overestimated using the CMB method (discussed in Section 5.2.1, lines 619-625). This has a little overall effect on the conclusions as the recharge rates estimated from the CMB method are still lower than from the other two methods.

Moreover, the stream discharge has probably increased due to the initial land clearing as is commonly the case throughout southeast Australia (Alison, 1990, *Journal of Hydrology*, 119, 1-20) and the streamflow prior to land clearing would have most probably been much lower. Additionally, the stream water is saline, and much of the CI exported by the streams is probably derived from shallow groundwater discharge; not from direct surface runoff. That component of CI does not need to be corrected for in the CMB calculations. We discussed these issues in Section 5.2.1 (lines 619-623).

7) The limitations of lumped parameter models (LPMs) should be discussed, as the dimensionless ratios assumed to vary over a very wide range (e.g. 0.05 to 1). Are the estimated residence times linearly related to these lumped parameters? Also, can it be clarified why the PEM and DM lumped parameter models were applied and not the exponential-piston flow model?

We discussed the limitations briefly in Section 5.1 (lines 577-587). The lumped parameter models are used for two purposes: to estimate residence times using  $^{14}\text{C}$  and to examine mixing using  $^3\text{H}$  and  $^{14}\text{C}$ . In terms of mixing, similar  $^3\text{H}$  vs.  $^{14}\text{C}$  trends are apparent using lumped parameter models and other models that predict the concentrations of the radioisotopes (e.g., the renewal rate calculations; Leduc et al., 2000, *Earth and Planetary Science*, 330, 355-361; Le Gal La Salle et al., 2001, *Journal of Hydrology*, 254, 145-156). The aims were here to broadly constrain the MRTs of the groundwater (i.e., to demonstrate that much of it is old). Other lumped parameter models (e.g., the EPM or the Gamma model) could have been used but yield MRTs that are similar to the ones that we reported (Section 3.4, lines 457-459). As we noted on lines 578-580 use of lumped parameter models are considerably preferable to

using the decay equation that assumes piston flow and ignores variations in the  $^{14}\text{C}$  of the atmosphere.

8) Clarify Line 295 (now lines 509-511), regarding Cl/Br ratios 'and do not indicate that Cl is predominantly derived from rainfall and concentrated by evapotranspiration'.

We corrected the sentence (lines 507-511).

9) Schematic cross-sections could help explain the relationship between regional vs. riparian groundwater. An additional map that shows the regional catchment context of the catchment divides for groundwater vs. surface water would also be helpful, as the current mapping provides very large scale and small-scale maps.

We added cross-sections to better show the context and the hydrogeology (Figure 2 in the revised manuscript). The catchments are at the top of a major surface water divide, and the groundwater divide will likely correspond to the surface water divide.

10) Mean residence times, estimated from both  $^3\text{H}$  and  $^{14}\text{C}$ , were ~4K in pasture and ~24K in the forest. Yet the forest was planted the only ~20 years ago, after ~160 years of pasture. The CMB method suggests chloride accumulation over ~10K years of rainfall inputs, to account for relatively high salinities. These differential time scales should be discussed further.

This is an important point that we now discussed in Section 5.3 (lines 790-796). The time taken for the  $^3\text{H}$  activities to achieve steady-state is  $\sim 1/R_N$  (i.e., the average residence time of the water in the well-mixed zone at the top of the aquifer). The initial land clearing should be evident in the TRR estimates, but the later revegetation may not be. Hence, in the cleared catchment, we would expect that WTF and TRR estimates agreed, but in the revegetated catchment, we may still be in the lag period where the  $^3\text{H}$  activities are showing transient behaviour between different recharge rates. This may also explain the observations in Comment 3.

#### **Reviewer 2:**

How do the results impact water management for the study area and for the region? Regarding the uncertainty in the estimated recharge rates and spatial and temporal variability, it is not obvious to me how sustainable water resource management can set up. Perhaps the authors have some thoughts about this problem and might provide some suggestions.

Our aim in this study is a better understanding of recharge rates in this area and assessing the uncertainties and limitations of commonly-applied recharge methods in general. As we have now made clear in the Introduction (lines 75-93) and the Conclusions (lines 839-846), estimating recharge is important to understand the impact of land-use change in semi-arid regions (especially the potential impacts on waterlogging and streamflow caused by changing water table elevations). As such, this study has relevance to other semi-arid regions.

Groundwater is not extensively used in the catchments studied, and our comments on sustainability were a more general recognition that groundwater can be a vital resource in semi-arid areas. As noted above, in response to Reviewer #1, we have removed that

discussion. In terms of the specific need to understand recharge, defining whether recharge rates changed with land-use changes is important for a complete understanding of the catchment water balance, and thus, land and water management.

If one of the objectives of this study is to assess and compare uncertainty in the methods, then this has to be more elaborated and systematically compared. In addition, these results should be compared to similar studies.

This was in the paper, but we have moved this discussion to the Conclusions to make it more prominent (lines 805-823). We compare our results with other studies in semi-arid areas (e.g., Dean et al., 2015, *Hydrology and Earth System Sciences*, 19, 1107–1123; Perveen, 2016, <http://hdl.handle.net/1959.9/560005>); Cartwright et al., 2007, *Journal of Hydrology*, 332, 69-92; Crosbie et al., 2010, *Hydrology and Earth System Sciences*, 14, 2023-2038). These studies also show that the WTF method yields higher recharge rates.

I miss a conceptual model which describe the processes. That can be a schematic figure or a cross-section describing the different flow systems and geochemical signatures.

We added cross-sections (Figure 2 of the revised manuscript), which will help understand the hydrogeology of the area and the processes.

Some further comments and suggestions are provided below.

Introduction: Personally, I believe that the study objectives should be clearly communicated in 1. Introduction. I found it a bit confusing to get information about the different methods before knowing the target of the study.

Although they normally appear at the end of the Introduction, we have moved the objectives to earlier in the Introduction (lines 121-129) before where we discussed the different methods to estimate recharge rates.

Line 48. Not only in semi-arid areas recharge varies in space and time. Also in humid areas, recharge can be considerable spatially and temporally different (see, for example, Moeck et al., 2020 and Mohan et al., 2018, among many others)

This is certainly the case. We have noted that estimating recharge rates, in general, is difficult (lines 103-105) and referred to the Moeck et al. (2020) study here and elsewhere.

Line 50: You could add Darcy methods, soil moisture methods, heat tracers, baseflow separation techniques, empirical relationships, etc. for completeness of the provided list (see for instance Healy, 2010, Walker et al., 2019).

We mentioned these other methods for completeness (lines 106-114).

Section 1.1.1: When residence times are around ~25000 years, how likely is that all Cl is originating from rainfall only and the impact of runoff can be neglected. This is more of a question rather than a critic. You already indicate based Cl/Br ratios that evapotranspiration rather than halite dissolution is the main process in controlling groundwater salinity but would be the error in estimated recharge rates if a small amount of Cl is not only originating from precipitation?

This method does assume that all the Cl is derived from rainfall. These are upland catchments with intermittent stream systems, located at the top of a major regional catchment divide; therefore, the catchments do not receive any Cl input from sources other than rainfall. In the study area, the Cl/Br ratios, and the lack of halite in the soils and bedrock make this the case (regardless of the residence time of the waters), as discussed in Section 4.2 (lines 509-514). In general, it is also the case for other semi-arid areas in southeast Australia where recharge rates have been calculated (e.g., Cartwright et al., 2007, *Journal of Hydrology*, 332, 69-92), and similar recharge rates could be estimated using Br rather than Cl. However, it is something that needs to be tested whenever this method is used.

Line 85: In the study area with an actual ET of ~600 mm/a, to what depth can ET impact be observed. I am asking because I am not sure if the observation wells 3008 (depth 1.3, pasture) and 3657 (depth 2.5 m, forest with deeper root zones) can be reliably used by applying the water table fluctuation method, although I have to note that the estimated rates seem to be in the same range as for the other observation points.

The recharge rates using the WTF method were calculated for those bores because they show a clear seasonal variation (Section 3.3, lines 407-411). The effect of ET on water tables is likely small during winter when radiation and temperatures are lower, and rainfall is larger (as we discussed on lines 684-686). Additionally, the bores in the plantation are installed in cleared areas near the stream where trees are not planted to create a buffer zone and limit the effect of the plantation on streamflow, which may also limit the impact of evapotranspiration.

Line 295-297: Maybe I misunderstood something here, but did you not indicate that all Cl is delivered by rainfall (e.g. Line 351; now Lines 618-619). Please check the statement and maybe reformulate the second part of the sentence.

We corrected the sentence (lines 505-507)

Line 333: Not clear. Please explain why it is not possible.

It is because it would require an initial  $a^{14}\text{C}$  that is not possible. All the samples with measurable  $^3\text{H}$  that lie on the  $^3\text{H}$  vs.  $^{14}\text{C}$  covariance curves will be less than 200 years old. Over that time span, there has been negligible decay of  $^{14}\text{C}$ , and the initial  $a^{14}\text{C}$  of the sample can be calculated by mass balance (it is the measured  $a^{14}\text{C}/q$ ). Consider a sample with a measured  $a^{14}\text{C}$  of 95 pMC; if there were 10% contribution of DIC from  $^{14}\text{C}$ -free calcite dissolution ( $q = 0.9$ ), it would imply an initial  $^{14}\text{C}$  activity of 106 pMC (which is plausible for water recharged during the bomb-pulse period). However, if we were to propose that  $q = 0.7$ , then the initial  $a^{14}\text{C}$  would need to be 136 pMC. This exceeds the highest  $a^{14}\text{C}$  recorded in soil  $\text{CO}_2$  of ~120 pMC (Jenkinson et al., 1992, *Soil Biology and Biochemistry*, 24, 295-308; Kuc et al., 2004, *Geochronometria*, 23, 45-50; Tipping et al., 2010, *Geoderma*, 155, 10-18) and so is implausible. We have explained this in Section 5.1 (lines 571-576).

Line 392: Just from Fig. 4 it is not possible to identify the samples. Perhaps you can better highlight these samples in Fig. 4 or provide the link to Table 1.

We highlighted the samples that show the mixing in Figure 5 (previously Fig. 4) and also referred to Table 1 in the and text (lines 695-697) where this information is also shown.

Apart from that, I am wondering that location 3663 do not show mixing with older groundwater, even though it is one of the deepest locations (~25m) and based on the drawn picture with the stratification I was expecting that older groundwater exists.

While that may be expected, the observations from the  $^{14}\text{C}$  and  $^3\text{H}$  imply that little mixing has occurred at this locality. Although bore 3663 is deep, this is largely due to the topography, rather than depth below the water table (the screen is approx. 10-15 m below the water table). Location 3663 is in the regional recharge area, and the groundwater in this area is expected to be relatively young. The groundwater flow here is likely to be downwards with little mixing with older laterally flowing groundwater. The cross-section (Fig. 2b) helps to illustrate this.

The decline in the groundwater level for 3663 is uniformly over the monitoring period, which is in contrast to the wells 3657 and 3669. But for all these, the TRR is applied and no mixing is assumed. Could you please elaborate more on these differences?

Bore 3663 is the only one that is actually in the forest. The other two (and most others) are in cleared areas between the stands of trees. Since the water levels probably respond to recharge over areas of a few  $\text{m}^2$  (Scanlon et al., 2002, *Hydrogeology Journal*, 10, 18-39), the difference probably reflects the more limited recharge in the forest areas (this is now discussed in Section 5.3, lines 781-810). Some recharge will occur at all the sites since the aquifers are unconfined.

Line 491: Yes, I absolutely agree, and this is an important point. The question of which arises from the results is how we can set up sustainable water resource management than when we have such a spatial and temporal variability as well as uncertainty on ~500 ha. Perhaps the authors have some thoughts about this problem and might provide some suggestions.

As explained above the study is less concerned with sustainable water use than the impacts of a rising water table on salinization of soils and streams. We have discussed the implications more specifically on lines 788-830 and 839-980.

Line 495: Although I agree that physical-based models are useful tools, the models will likely not represent in every detail the recharge processes because of the lack of observations although a relatively high density of data exists and information about the subsurface heterogeneity are missing (in both, x, y and z-direction). Thus, calibration is required, which will also lead to uncertainty. For me, this part sounds like we always should use physical-based models, and then we get the right recharge rates which are obviously not true, even though the models are very powerful. Please reformulate.

We modified this discussion not to give the impression that models can be used to estimate recharge rates (lines 832-838). This was not the intention of this paragraph, which rather meant to stress the uncertainties associated with the use of measured recharge rates as boundary conditions for groundwater models. Integrated models with several simplifying assumptions and without accounting for the large spatial soil heterogeneity, can be used to support experimental studies by providing an additional estimate of recharge rates and suggest reasonable upper limits of recharge rates.



Figure 1: If available, it would be useful to add the precipitation on top of the graphics (second y-axis).

This relates to Figure 2. We added the rainfall to this figure (now Fig. 2a).

Also, why does well 3658 show an increase?

Bore 3658 shows very little response over the monitoring period (head levels vary by <1.4 m). The bore monitors groundwater at ~16 m in the lower part of the catchment and probably does not record the short-term recharge.

### Reviewer 3:

1. There is some confusion in the paper about the specific yield. Specific yield is, as the authors quote, “the volume of water that an unconfined aquifer releases from storage per unit surface area of aquifer per unit decline in the water table”. This is the water that drains from the aquifer under the influence of gravity as the water table falls. As the water table falls, some water remains in the smaller pore spaces and as rims and menisci around grains. This definition does not “ignore the moisture in the unsaturated zone held in and above the capillary fringe”. The moisture in the unsaturated zone does not drain significantly. The capillary fringe is saturated (not unsaturated) and moves downward with the water table at the same rate. Therefore, the reason that the WTF method of estimating recharge gives unrealistically large values in this study is not the result of “the presence of moisture in the unsaturated zone and capillary fringe .. reduc(ing) the effective values of  $S_y$ ”.

We agree with the points raised in this comment. As discussed in response to the other reviewers, we shortened our discussion of specific yield in the introduction (Section 1.2, lines 232-239). This material is well covered by previous studies (e.g., Gillham, 1984; Sophocleous, 1985; Healy and Cook, 2002; Crosbie et al., 2019) that we cite, and our longish discussion of that material was confusing. It remains the case though that most recharge studies that use the WTF method make the oversimplifying assumption that the specific yield is spatially and temporally uniform. Indeed, many assume that the specific yield is close to the effective porosity. We have clarified these points throughout the paper (Sections 1.2 and 5.2.2).

2. There is also some confusion about porosity and effective porosity. Effective porosity is the porosity through which fluid can flow and is almost always less than total porosity. Effective porosity is generally similar to, or slightly less than, specific yield, as shown by the comparison of  $S_y$  values from pumping tests and  $n_e$  values from Darcy’s Law ( $n_e = Ki/v$ ). In this study, values of  $S_y$ ,  $n$  and  $n_e$  are used interchangeably, and the authors need to correct this or at least explain why they used these values. Previous studies found  $S_y$  values of 0.03 to 0.1 and mean porosity of 0.1-0.15; in this paper values of  $S_y$  of 0.03 to 0.1 were used for the WTF method, whereas for the mass balance calculation (line 365 in the previous version) and the TRR method,  $n$  values of 0.03 to 0.1 were used. The latter is likely to be too low and will make the TRR numbers calculated also too low.

We agree with the reviewer and have clarified this in the paper (lines 232-238). As discussed above, the assumption that specific yield is similar to the effective porosity is one that is

commonly made for the WTF calculations but one that is probably incorrect (as is also discussed by Gillham, 1984; Sophocleous, 1985; Healy and Cook, 2002; Crosbie et al., 2019).

The values of porosity used in this study are those from Adelana et al. (2014) and are similar to typical values for these aquifer materials. We discussed the uncertainties in the TRR recharge estimates arising from these values in Section 5.2.3; however, the uncertainties arising from having to estimate  $b$  and the input function of  $^3\text{H}$  are greater (lines 716-740). Overall, the TRR recharge rates are still considerably higher than those from the CMB method but lower than the WTF estimates.

3. Note that “if the soil becomes fully saturated due to the rise of the capillary fringe”, the top of the capillary fringe is at the ground surface and therefore no recharge can occur. Small recharge events cannot “produce significant and rapid increases in the head”. This has no effect on the amount of water that can drain from the aquifer when the water table drops, so  $S_y$  does not become “close to 0”.

This discussion was taken from Gillham (1984). As we have noted above, we have simplified this in Section (1.2) and just noted that the common assumption made in the WTF method overestimates the specific yield and consequently recharge rates.

4. Another puzzling aspect is the definition of  $b$  for calculating TRR;  $b$  is “the thickness of the upper part of the aquifer system that receives annual recharge”. Is this the part of the aquifer that is subject to water table fluctuations, i.e. is  $b$  equal to the maximum fluctuation? If this is the case, why not use this value? In this paper,  $b$  is estimated from chemical stratification of regional groundwater (p. 18: now p. 19-20). But if the groundwater is stratified, then this could be because the upper part is not recharging the lower part, i.e. there are two separate aquifers. Alternatively, the chemical stratification could reflect the difference in recharge since the clearing of native vegetation in the area. In either case, the use of chemical stratification to estimate  $b$  is unjustified, and the presence of chemical stratification has implications for the CMB calculations; there should be separate calculations for the upper and lower groundwater.

We defined  $b$  using observations of chemical stratification (lines 702-705). The value of 1 to 5 m is the distance at the top of the aquifer over which the groundwater chemistry is relatively uniform. These values of  $b$  are similar to those proposed elsewhere (Le Gal La Salle et al., 2001; Cartwright et al., 2007). We discussed the uncertainties in the  $b$  values in Sections 5.2.3 (lines 736-738) and 5.3 (lines 716-741); increasing the depth of the zone of active mixing at the top of the aquifer to 10 m (which is unlikely given the observations of the depths over which the geochemistry varies) would increase the recharge rates, but the significant differences between CMB, WTF and TRR recharge rates remain. This method of estimating  $b$  is the same as in the previous studies (including those of Leduc et al., 2000 and Le Gal La Salle et al., 2001) from which we derived the method.

There is no evidence of two discretely separate aquifers; the bore logs do not indicate any major low  $K$  layers, and the groundwater generally contains measurable  $^3\text{H}$  at depths of up to 29 m (Table S1). This precludes the presence of a deeper groundwater system that is not isolated from the shallower part of the aquifers. The joint use of  $^3\text{H}$  and  $^{14}\text{C}$  activities allows

macroscopic mixing of old and recently recharged groundwater in the aquifers to be tested (Fig. 5 and discussion on lines 554-559), and adds confidence to the TRR calculations. Many aquifers show some chemical stratification without consisting of discretely separated flow systems.

In terms of the CMB technique, if recent recharge rates have changed due to land clearing, the Cl concentrations in the upper part of the aquifer may be lower (this might be expected mainly in the pasture catchment). However, the Cl concentrations of the shallow and deeper groundwater overlap and there is no correlation between Cl and  $^3\text{H}$  (we have added Figures 4b & 4c to show this). This probably reflects the timescale of Cl delivery. Because Cl in saline groundwater may take several thousand years to accumulate, recent changes in evapotranspiration (which reduce the Cl concentrations) are not yet visible in the groundwater. Thus, the CMB recharge rates represent the average of those over the last several hundred to thousands of years. This has also been noted elsewhere in SE Australia (Crosbie et al., 2010; Cartwright et al., 2007; Dean et al., 2015). We have noted this in Section 5.3 (lines 748-751)

5. The forest bores show relatively small seasonal fluctuations compared to pasture bores, and some show no fluctuations at all (Fig 2: now Figs 3b, 3c). Yet the WTF recharge values for the forest are the same as those for the pasture (Fig 6: now Fig 7). This seems very unlikely and requires explanation.

In the forest, the annual variations of the head in bores 3656, 3657 & 3669 are up to 3 m, which is similar to many of those in the pasture catchment (Figs. 3b, 3c). These are the bores that yield high WTF recharge rates. As we discussed in Section 5.3 (lines 788-790), the trees cover ~62% of the forest catchment, and many of the bores are in cleared areas between the stands of trees (Fig. 1a). So, the recharge rates may not be representative of the forest as a whole and are similar to the cleared areas in the pasture.

6. The WTF values calculated are not just unlikely and higher than expected, they are impossible. Recharge of this magnitude would imply that the vegetation was not extracting significant levels of water, and the consistent drop in the water table beneath the forest shows that this is not the case.

This is what we concluded in the paper (Sections 5.2.2, lines 666-670 and the Conclusions, lines 804-807). The WTF method with the common assumption of a consistent specific yield remains widely used as we discussed. We also explored other reasons why the WTF method yields high recharge rates (e.g., focussed recharge and the subsequent evaporation of water from the water table – lines 681-685). While these are also issues, the inaccurate assumptions around the specific yield are probably more serious.

7. The authors note that “there has been a rise in the water table caused by the increased recharge, and in some cases increased drainage in the streams”; what is the evidence for this in the study area? This topic has been much discussed in the Australian groundwater literature, and needs more discussion and explanation, with the comparison with other areas in SE Australia.

The Gatum area is one of many in SE Australia that was identified as being impacted by dryland salinity due to land clearing and rising water tables (Clark and Harvey, 2008: Dryland salinity in Victoria in 2007, Department of Primary Industries Report). The area has common saline discharge to streams and local salt scalds. The bore monitoring and streamflow network were set up in the pasture catchment in this area on that basis. During the Millennium Drought in the first decade of the century, the water table levels dropped considerably and the emphasis on soil salinity diminished. The focus of water management in the area switched from salinity to water availability and the effect of land use on the water balance of these catchments. Accordingly, monitoring in the forest was set up to assess the subsequent impact of the tree plantation on the groundwater and surface water. We added these details to the study area (lines 327-335) and emphasised in the Conclusions (lines 840-980) that it is typical of many similar areas in SE Australia and elsewhere.

8. Rainfall was sampled for tritium content. The sampling method needs to be briefly described and the results given in Table S1 (not a single average value).

We briefly described this Section 3.1 (lines 371-373). The rainfall sample is an aggregate (i.e. successive samples were collected and mixed into a single sample), not an average.

9. The aquifer is described as “silty clay to coarse-grained sediments” and as comprising “inter-layered clays and silts”. Silts are not coarse-grained and the porosity values (0.1-0.15) suggest sandy sediments. The authors need to resolve this.

The aquifer materials are mainly silt-sized to coarse-grained weathered ignimbrites with minor discontinuous clay layers. These are described by Brouwer & Fitzpatrick (2002) and Adelen et al. (2014) who also report aquifer properties such as porosity. We have clarified this in Section 2 (lines 296-314).

10. There are a few small grammatical/spelling errors: lines 107, 263, 295-296, 342, 358, 429 (now lines 241, 463-466, 509-511, 588, 625, 775).

We have corrected the grammatical and spelling errors on these lines.

11. Fig 2 (now Figs 3b, 3c) would be better plotted as depth bgs.

Because we use Figures 3b, 3c to discuss heads in the catchment and this figure links to the head values in Figure 1b, we prefer to leave this as it is. The individual bore hydrographs are also more easily seen on this version of the figure.

Using multiple methods to **investigate the effects of land-use changes on** groundwater recharge in a semi-arid area

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## Abstract

Understanding the applicability and uncertainties of methods for documenting recharge rates in semi-arid areas is important for assessing the successive effects of land-use changes and understanding groundwater systems. This study focuses on estimating groundwater recharge rates and understanding the impacts of land-use changes on recharge rates in a semi-arid area in southeast Australia. Two adjacent catchments were cleared ~180 years ago following European settlement and a eucalypt plantation forest was subsequently established ~20 years ago in one of the catchments. Chloride mass balance yields recharge rates of 0.2 to 61.6 mm yr<sup>-1</sup> (typically up to 11.2 mm yr<sup>-1</sup>). The lower of these values probably represent recharge rates prior to land clearing, whereas the higher likely reflects recharge rates following initial land clearing. The low pre-land clearing recharge rates are consistent with the presence of old groundwater (residence times up to 24,700 years) and the moderate to low hydraulic conductivities (0.31 to 0.002 m day<sup>-1</sup>) of the aquifers. Recharge rates estimated from tritium activities and water table fluctuations reflect those following the initial land clearing. Recharge rates estimated using water table fluctuations (15 to 500 mm yr<sup>-1</sup>) are significantly higher than those estimated using tritium renewal rates (0.01 to 89 mm yr<sup>-1</sup>; typically <14.0 mm yr<sup>-1</sup>) and approach the long-term average annual rainfall (~640 mm yr<sup>-1</sup>). These recharge rates are unrealistic given the estimated evapotranspiration rates of 500 to 600 mm yr<sup>-1</sup> and the preservation of old groundwater in the catchments. It is likely that uncertainties in the specific yield results in the water table fluctuation method significantly overestimating recharge rates and that, despite the land-use changes, the present-day recharge rates are relatively modest. These results are ultimately important for the assessing the impacts of land-use changes and management of groundwater resources in semi-arid regions in Australia and elsewhere.

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# 1. Introduction

Groundwater is a critical resource for meeting the expanding urban, industrial and agricultural water requirements, especially in semi-arid areas that lack abundant surface water resources

(de Vries and Simmers, 2002; Siebert et al., 2010). ~~Groundwater also makes a significant contribution to the streamflow of rivers in semi-arid areas. Land-use changes may modify groundwater recharge rates, which thus affect groundwater systems as well as groundwater resources (Foley et al., 2005; Lerner and Harris, 2009; Owuor et al., 2016). In many semi-arid regions, there has been the conversion of native forests to agricultural land (Foley et al., 2005). Deep-rooted trees generally return more water to the atmosphere via transpiration than shallow-rooted crops and grasses (Hewlett and Hibbert, 1967; Bosch and Hewlett, 1982; Fohrer et al., 2001). In southeast Australia, the reduction in evapotranspiration following land clearing has commonly resulted in a net increase in recharge and a rise of the regional water tables. In turn, this has resulted in waterlogging and salinization of cleared lands and increased stream salinity (Allison et al., 1990). Eucalyptus tree plantations were subsequently initiated partially to reduce groundwater recharge and thus prevent the rise of regional water tables (Gee et al., 1992; Benyon et al., 2006). In order to assess the impacts of successive land-use changes on the groundwater and surface water systems, estimates of recharge are required. Estimation of recharge rates is also important for groundwater modelling, because recharge represents the water flux used as a boundary condition at the water table.~~

Recharge is the water that infiltrates through the unsaturated zone to the water table and thus increases the volume of water stored in the saturated zone (Lerner et al., 1990; Healy and Cook, 2002; Scanlon et al., 2002; ~~Moeck et al., 2020~~). A distinction between gross and net recharge may also be made (Crosbie et al., 2005). The total amount of water that reaches the water table is the gross recharge, while net recharge accounts for the subsequent removal of water from the saturated zone by evapotranspiration. In areas with shallow water tables and deep-rooted

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80 vegetation, this subsequent water loss can be considerable. Estimating groundwater recharge rates, in general, is not straightforward (Lerner et al., 1990; Healy, 2010; Moeck et al., 2020) and recharge rates potentially vary in space and time (Sibanda et al., 2009).

85 Several techniques may be used to estimate groundwater recharge, including Darcy's Law, measuring water infiltration using lysimeters installed in the unsaturated zone, measuring and modelling soil moisture contents, use of heat flow calculations, catchment water budgets, remote sensing, numerical models, water table fluctuations, chemical (chloride) mass balance calculations, and/or the concentrations of radioisotopes such as <sup>3</sup>H (tritium), <sup>14</sup>C (carbon), <sup>36</sup>Cl (chloride) or other time-sensitive tracers (e.g., chlorofluorocarbons) in groundwater (Scanlon et al., 2002, 2006; Healy, 2010; Doble and Crosbie, 2017; Cartwright et al., 2017; Moeck et al., 2020; Gelsinari et al., 2020).

90 Different techniques estimate recharge over different spatial-temporal scales, and they may thus yield different results (Scanlon et al., 2002). Because each technique has different uncertainties and limitations, it is recommended that multiple methods are used to constrain recharge (Healy and Cook, 2002; Sophocleous, 2004; Scanlon et al., 2006). Understanding the broader hydrogeology also helps to understand recharge. For example, areas where recharge rates are high, should contain high proportions of young groundwater. Additionally, recharge rates are likely to be low if evapotranspiration rates approach rainfall totals.

100 This study estimates recharge rates using Cl mass balance, water table fluctuations, and <sup>3</sup>H renewal rate methods in a semi-arid area that has undergone successive land-use changes. We evaluate the applicability and uncertainties of these commonly applied methods to determine the changes in recharge rates caused by these successive land-use changes. While based on a specific area, the results of this study, in particular the comparison of the estimates of recent recharge rates, will be applicable to similar semi-arid areas in southeast Australia and elsewhere. Specifically, predicting the impacts of changes to land-use on recharge rates is

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1.1 Quantifying groundwater recharge¶  
Estimating groundwater recharge rates is not straightforward (Lerner et al., 1990) and recharge rates in semi-arid areas

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required to understand and manage waterlogging and salinization of soils and streams. A brief description of the assumptions and limitations of these techniques is provided below.

### 1.1 Cl mass balance

The Cl mass balance (CMB) approach yields average regional net recharge rates (Bazuhair and Wood, 1996; Scanlon, 2000; Scanlon et al., 2002). The assumptions of this method are that all Cl in groundwater originates from rainfall and that Cl exported in surface runoff is negligible or well known. Under these conditions, the net groundwater recharge ( $R_{net}$  in  $\text{mm yr}^{-1}$ ) is estimated from:

$$R_{net} = P \frac{Cl_p}{Cl_{gw}} \quad (1)$$

(Eriksson and Khunakasem, 1969) where  $P$  is mean annual precipitation ( $\text{mm yr}^{-1}$ ),  $Cl_p$  is the weighted mean Cl concentration in precipitation ( $\text{mg L}^{-1}$ ), and  $Cl_{gw}$  is Cl concentration in groundwater ( $\text{mg L}^{-1}$ ). The CMB method estimates net recharge rates averaged over the time that the Cl contained within the groundwater is delivered; this may be several years to millennia. Uncertainties in the CMB method are mainly the long-term rate of Cl delivery and the assumptions that runoff has remained negligible over time.

### 1.2 Water table fluctuations

Water table fluctuations may be used to estimate gross recharge over the time period for which groundwater elevation data are available. Because bore hydrograph data are abundant, this probably is the most common method of estimating modern recharge rates. The water table fluctuation (WTF) method strictly requires the water table to be located within the screened interval of the bore; however, it can be used in bores screened within a few metres of the water table (Healy and Cook, 2002). The method assumes that: evapotranspiration from the water table has not occurred; the rise in the water table is solely due to recharge following rainfall events; groundwater elevations are not influenced by pumping; and the water table falls in the absence of recharge.  $R_{gross}$  is calculated from

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$$R_{gross} = S_y \frac{\Delta h}{\Delta t} \quad (2)$$

185 where  $S_y$  is the specific yield (dimensionless) of the aquifer, and  $\Delta h/\Delta t$  is the variation in the hydraulic head over the recharge event ( $\text{mm yr}^{-1}$  where there is an annual recharge event).

Despite its simplicity, there are several potential uncertainties in the WTF method.  $S_y$  is not commonly measured, and most studies rely on typical values based on aquifer materials. More importantly, the retention of moisture in the unsaturated zone between recharge events reduces  $S_y$  and results in  $S_y$  being spatially and temporally variable (Gillham, 1984; Sophocleous, 1985; Healy and Cook, 2002; Crosbie et al., 2019). However, many recharge studies assume that  $S_y$  is constant and close to the effective porosity. This may result in this method significantly overestimating recharge rates (Gillham, 1984; Sophocleous, 1985; Crosbie et al., 2019). Other processes may also affect head measurements. These include entrapment of air during rapid recharge events (the Lisse effect) and the impacts of barometric pressure changes and ocean or Earth tides, especially when the head is measured using sealed pressure transducers (Crosbie et al., 2005). The estimation of the recession curve of the groundwater hydrograph used to calculate  $\Delta h$  in Eq. (2) also involves some judgement.

### 1.3 $^3\text{H}$ renewal rate

200 The  $^3\text{H}$  renewal rate (TRR) method envisages that recharge mixes with pre-existing groundwater at the top of the aquifer. The renewal rate ( $R_n$ ) represents the proportion of new water added in each recharge cycle with an equivalent amount displaced lower into the groundwater system. If there is an annual cycle of groundwater recharge, the  $^3\text{H}$  activity of groundwater in year  $i$  ( $^3H_{gw_i}$ ) is related to  $R_n$  by

$$^3H_{gw_i} = (1-R_n)^3 H_{gw_{i-1}} e^{-\lambda_t} + R_n ^3 H_{p_i} \quad (3)$$

205 (Leduc et al., 2000; Le Gal La Salle et al., 2001; Favreau et al., 2002) where  $\lambda_t$  is the radioactive decay constant for  $^3\text{H}$  (0.0563 yr<sup>-1</sup>), and  $^3H_{p_i}$  is the average  $^3\text{H}$  activity of rainfall in year  $i$  (in

**Deleted:** Additionally,  $S_y$  is commonly viewed as a constant aquifer property ("the volume of water that an unconfined aquifer releases from storage per unit surface area of aquifer per unit decline in the water table": Freeze and Cherry, 1979). However, that ignores the moisture in the unsaturated zone held in and above the capillary fringe (Gillham, 1984; Crosbie et al., 2005, 2019), which results in  $S_y$  varying with depth and over time (Childs, 1960). The presence of this soil moisture reduces the volume of water needed to saturate the aquifer matrix, thus reducing the effective  $S_y$ . If the soil becomes fully saturated due to the rise of the capillary fringe, it has an effective  $S_y$  close to 0, and small recharge events can produce significant and rapid increases in the head (Gillham, 1984). This can be an issue in areas of fine-grained soils where the capillary fringe may be several metres thick. Not considering the potential reduction in the effective  $S_y$  in areas of shallow water tables leads to recharge being overestimated by the WTF method. Other processes may also affect head measurements, such as

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230 Tritium Units, TU where 1 TU corresponds to  ${}^3\text{H}/{}^1\text{H} = 1 \times 10^{-18}$ ). The application of the TRR method requires the  ${}^3\text{H}$  input function over the past few decades to be known. The  ${}^3\text{H}$  activities of southern hemisphere groundwater recharged during the 1950s and 1960s atmospheric tests were several orders of magnitude lower than northern hemisphere groundwater (Morgenstern et al., 2010; Tadros et al., 2014). These  ${}^3\text{H}$  activities have now decayed and are lower than those of present-day rainfall, which results in individual  ${}^3\text{H}$  activities yielding a single  $R_n$  estimate (Cartwright et al., 2007, 2017), which is not yet the case in the northern hemisphere (Le Gal La Salle et al., 2001).

Groundwater recharge rates are related to  $R_n$  by

$$R_{net} = R_n b n \quad (4)$$

240 where  $b$  is the thickness of the upper part of the aquifer system that receives annual recharge and  $n$  is the effective porosity. Uncertainties in the TRR estimates include uncertainties in the  ${}^3\text{H}$  input function and having to estimate  $b$  and  $n$ , which may be variable and not well defined. The recharge rates are net estimates averaged over the residence time of the groundwater in the upper part of the aquifer, which ~~in an ideal system is~~  $R_n^{-1}$ .

## 2. Study area

245 Gatum is situated in western Victoria, southeast Australia (Fig. 1a). The native eucalyptus forests in this region were originally cleared for grazing following European settlement ~180 years ago (Lewis, 1985) and then partially replaced by eucalyptus plantation in the last ~20 years (Adelana et al., 2014). Gatum lies in the regional recharge area of the Glenelg River Basin to the south of the drainage divide between the Glenelg and Wannon Rivers, and surface water drains to the Wannon River via the Dundas River (Dresel et al., 2012). The area is predominantly composed of ~~fine- to coarse-grained weathered~~ Early Devonian ignimbrites containing abundant large locally derived clasts near their base (Cayley and Taylor, 1997). Post-Permian weathering has produced a deeply weathered saprolitic clay-rich regolith and

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**Deleted: 1.2 Objectives¶**

This study estimates recharge rates using the

**Moved up [6]:** CI mass balance, water table fluctuations, and  ${}^3\text{H}$  renewal rate methods in a semi-arid area that has undergone successive land-use changes. We evaluate the applicability and uncertainties of these commonly applied methods to determine the changes in recharge rates caused by these successive land-use changes. While based on a specific area, the results of this study, in particular the comparison of the estimates of recent recharge rates, will be applicable to similar semi-arid areas

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ferruginous laterite duricrust (Brouwer and Fitzpatrick, 2002). Some of the drainage areas contain Quaternary alluvium and colluvium (Adelana et al., 2014).

The study area consists of two catchments with contrasting land-use, one catchment is predominately dryland pasture used for sheep grazing, and the other is mostly occupied by plantation *Eucalyptus globulus* forestry. The pasture catchment is around 151 ha and is typical of the cleared land in this region. It is covered by perennial grasses with about 3 % remnant eucalyptus trees. The forest catchment is around 338 ha and comprises approximately 62 % plantation forest, established in 2005, and 38 % grassland (Adelana et al., 2014). The elevations of the pasture and forest catchments range from 236 to 261 m and 237 to 265 m AHD (Australian Height Datum), respectively (Fig. 2). The two catchments were subdivided into the upper slope, mid-slope and lower slope, based on the elevation of the study area; the drainage zones are in the riparian zones of the small streams (Dresel et al., 2018). The catchments are drained by two small intermittent streams (Banool and McGill; Fig. 1a) that export ~8 % of annual rainfall (Adelana et al., 2014; Dresel et al., 2018).

The regional groundwater is not extensively used in this area. However, the study area is one of many in southeast Australia that was identified as being impacted by dryland salinity due to land clearing and rising water tables (Clark and Harvey, 2008). During the Millennium Drought in the first decade of the century, the water tables dropped considerably and the emphasis on dryland salinity diminished. The focus of water management in this area switched from salinity to water sustainability and the effect of land-use changes on the water balance of this area (Dresel et al., 2012). In addition to the regional groundwater system, shallow (1 to 4 m deep) perched groundwater exists in the riparian zones (Brouwer and Fitzpatrick, 2002; Adelana et al., 2014).

The climate is characterized by cool, wet winters and hot, dry summers. From 1884 to 2018, the average annual rainfall at Cavendish (Station 089009) ~19 km south of Gatum was ~640

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295 mm (Bureau of Meteorology, 2020), with most rainfall in the austral winter between May and  
October, (Fig. 3a). Average annual actual evapotranspiration across the two catchments  
between 2011 and 2016 was estimated at about 580 mm (Dresel et al., 2018). The mean  
concentrations of Cl in rainfall range from 2.2 mg L<sup>-1</sup> at Cavendish (Hutton and Leslie, 1958)  
to 4.4 mg L<sup>-1</sup> at Hamilton (~34 km south of Gatum; Bormann, 2004; Dean et al., 2014). Similar  
300 Cl concentrations are recorded in rainfall across much of southeast Australia (Blackburn and  
McLeod, 1983; Crosbie et al., 2012).

### 3. Methods and Materials

#### 3.1 Water sampling

305 There are 19 monitoring bores at different landscape positions sampling the regional  
groundwater in the pasture and forest catchments (Fig. 1a) with sample depths ranging from  
1.3 to 29.7 m (Supplementary Table S1). Hydraulic heads have been measured since 2010 at  
four hourly intervals using In Situ Aquatroll or Campbell CS450 WL pressure loggers corrected  
for barometric pressure variations using In Situ Barotroll loggers. Occasional spikes (generally  
resulting from the logger being removed from the bores) were removed. Twelve shallow  
310 piezometers (~1 m deep with ~10 cm wide screens at their base) were installed in 2018 near  
the monitoring bores in the drainage zones and the lower slopes of the pasture and forest  
catchments (Fig. 1a). These piezometers intercept the riparian groundwater that in places is  
perched above the regional groundwater. Regional groundwater was sampled from the bores  
(n = 24) and riparian groundwater from shallow piezometers (n = 24) between May and  
315 November 2018. The groundwater samples were collected from the screened interval using a  
submersible pump or bailer following the removal of at least three bore volumes of  
groundwater or removing all water and allowing it to recover. Following sampling, hydraulic  
conductivities (K<sub>s</sub>: m day<sup>-1</sup>) were determined from the rate of recovery of the groundwater  
levels measured at 3-minute intervals using an In Situ Aquatroll pressure logger (Hvorslev,

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1951). A one-year aggregated rainwater sample was collected in a narrow-mouthed container with an open funnel. The sample was periodically removed from the container and aggregated into a single sample.

### 3.2 Analytical techniques

Geochemical data are presented in Table S1. Electrical conductivity (EC) was measured in the field using a calibrated hand-held TPS WP-81 multimeter and probe. Groundwater samples were collected in high-density polyethylene bottles and stored at ~4°C prior to analysis. Alkalinity (HCO<sub>3</sub><sup>-</sup>) concentrations were measured within 12 hours of sampling by titration. Major ion concentrations were measured at Monash University. Cation concentrations were determined on filtered (0.45 µm cellulose nitrate filters) water samples that were acidified to pH <2 with double distilled 16 N HNO<sub>3</sub> using ICP-OES (Thermo Scientific iCAP 7000). Concentrations of anions were determined on unacidified filtered water samples by ion chromatography (Thermo Scientific Dionex ICS-1100). Based on replicate analyses, the precision of cation and anion concentrations are ±2 %; from the analysis of certified standards, accuracy is estimated at ±5 %. Total dissolved solids (TDS) concentrations are the sum of the cation and anion concentrations.

<sup>3</sup>H and <sup>14</sup>C activities were measured at the Institute of Geological and Nuclear Sciences (GNS) in New Zealand. Samples for <sup>3</sup>H activities were measured by liquid scintillation in Quantulus ultra-low-level counters following vacuum distillation and electrolytic enrichment as described by Morgenstern and Taylor (2009). The quantification limits are 0.02 TU and the relative uncertainties are typically ±2 % (Table S1). <sup>14</sup>C activities were measured by AMS following Stewart et al. (2004). Dissolved inorganic carbon (DIC) was converted to CO<sub>2</sub> by acidification with H<sub>3</sub>PO<sub>4</sub> in a closed evacuated environment. The CO<sub>2</sub> was purified cryogenically and converted to graphite. <sup>14</sup>C activities are normalised using the δ<sup>13</sup>C values and expressed as percent modern carbon (pMC), where the <sup>14</sup>C activity of modern carbon is 95 % of the <sup>14</sup>C

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355 activity of the NBS oxalic acid standard in 1950. Uncertainties are between 0.27 and 0.35 pMC  
(Table S1).

### 3.3 Recharge calculations

Recharge rates were estimated using the methods discussed in sections 1.1-1.3. Net recharge  
rate estimates from the CMB (Eq. 1) utilised present-day average rainfall amounts (~640 mm)  
360 and Cl concentrations of 2.2 to 4.4 mg L<sup>-1</sup> together with the measured Cl concentrations of  
groundwater (Table S1). Gross recharge rates were estimated using the WTF method (Eq. 2)  
from the hydrographs of bores that display seasonal variations in water levels (Figs. 3b, 3c).

There is a single pronounced annual increase in the hydraulic head following winter rainfall,  
and  $\Delta h$  was estimated as the difference between the highest head value and the extrapolated  
365 antecedent recession curve, (Healy and Cook, 2002). The effect of evapotranspiration on the  
magnitude of the hydraulic heads is assumed to be low, especially during winter when radiation  
and temperature are lower.  $S_y$  was assumed to be close to  $n$  (0.03 to 0.1; Adelana et al. 2014;  
Dean et al., 2015), which will be the case if the unsaturated zone dries up between recharge  
events (Sophocleous, 1985). The TRR calculations (Eq. 3) used the <sup>3</sup>H activities in Melbourne  
370 rainfall as the input function (Tadros et al., 2014). The annual average <sup>3</sup>H activity of present-  
day rainfall in both Melbourne and Gatum is ~2.8 TU (Tadros et al., 2014; Table S1) and the  
rainfall prior to the atmospheric nuclear tests was assumed to have had the same <sup>3</sup>H activity as  
present-day rainfall.  $n = 0.03$  to  $0.1$  was again used and estimates of  $b$  are discussed below.

### 3.4 Mean residence times

375 Mean residence times (MRTs) and the covariance of <sup>3</sup>H and <sup>14</sup>C activities in groundwater were  
estimated via lumped parameter models (LPMs: Zuber and Maloszewski, 2001; Jurgen et al.,  
2012). LPMs relate the <sup>14</sup>C activity of water at time  $t$  ( $C_{out}$ ) to the input of <sup>14</sup>C of recharge over  
time ( $C_{in}$ ) via the convolution integral

$$C_{out}(t) = \int_0^{\infty} q C_{in}(t - \tau_m) e^{-\lambda_c \tau_m} g(\tau_m) d\tau_m \quad (5)$$

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Moved up [7]: The annual average <sup>3</sup>H activity of present-day rainfall in both Melbourne and Gatum is ~2.8 TU (Tadros et al., 2014; Table S1

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(Zuber and Maloszewski, 2001; Jurgens et al., 2012) where  $q$  is the fraction of DIC derived from  
395 rainfall or the soil zone,  $(t - \tau_m)$  is the age of the water,  $\tau_m$  is the mean residence time,  $\lambda_c$  is the  
decay constant for  $^{14}\text{C}$  ( $1.21 \times 10^{-4} \text{ yr}^{-1}$ ), and  $g(\tau_m)$  is the system response function that  
describes the distribution of residence times in the aquifer (described in detail by Maloszewski  
and Zuber, 1982; Zuber and Maloszewski, 2001; Jurgens et al., 2012).  $^3\text{H}$  activities may be  
calculated from the input of  $^3\text{H}$  over time in a similar way. Unlike  $^{14}\text{C}$ ,  $^3\text{H}$  activities are not  
400 changed by reactions between the groundwater and the aquifer matrix; hence the  $q$  term is  
omitted.

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There are several commonly used LPMs. The partial exponential model (PEM) may be applied  
aquifers where only the deeper groundwater flow paths are sampled. The dimensionless PEM  
ratio defines the ratio of the unsampled to sampled depths of the aquifer (Jurgens et al., 2012).  
405 This study used PEM ratios of 0.05 to 0.5 that cover the ratios of the sample to unsampled  
portions of the aquifers at Gatun. The dispersion model (DM) is derived from the one-  
dimensional advection-dispersion transport equation and is applicable to a broad range of flow  
systems (Maloszewski and Zuber, 1982; Zuber and Maloszewski, 2001; Jurgens et al., 2012).  
The dimensionless dispersion parameter (DP) in this model describes the relative contributions  
410 of dispersion and advection. For flow systems of a few hundreds of metres to a few kilometres,  
DP values are likely to be in the range of 0.05 to 1.0 (Zuber and Maloszewski, 2001). Other  
commonly applied LPMs, such as the exponential-piston flow or gamma model, produce  
similar estimates of residence times (Jurgens et al., 2012; Howcroft et al., 2017). The long-  
term variations of atmospheric  $^{14}\text{C}$  concentrations in the southern hemisphere (Hua and  
415 Barbetti, 2004; McCormac et al., 2004) were used as the  $^{14}\text{C}$  input function, and the  $^3\text{H}$   
activities in rainfall for Melbourne (Tadros et al., 2014) were used as the  $^3\text{H}$  input function.

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## 4. Results

### 4.1 Hydraulic heads and properties

425 The hydraulic heads in regional groundwater from both pasture and forest catchments decrease  
from the upper to lower slopes implying that the regional groundwater flows southwards (Fig.  
1b). In the pasture, the hydraulic heads in groundwater from all bores generally gradually  
increase over several weeks to months following the onset of winter rainfall (Fig. 3b). The  
increase in hydraulic heads was higher in 2016, which was a year of higher than average rainfall  
430 (~800 mm; Bureau of Meteorology, 2020). This was especially evident at bore 63 (Fig. 3b). In  
the forest, groundwater heads from bores in the upper (3663 and 3665) and mid (3668) slopes  
decline uniformly over the monitoring period, and the groundwater head from bore 3658 near  
the drainage zones does not show seasonal variations (Fig. 3c). However, fluctuations of heads  
from three bores near the drainage zones (3669) and lower slopes (3656 and 3657) show  
435 seasonal variations similar to that of the groundwater in the pasture, (Figs. 3b, 3c).  
Values of  $K_s$  range from 0.06 to 0.31 m day<sup>-1</sup> in the pasture (Fig. 2a) and from 0.002 to 0.18 m  
day<sup>-1</sup> in the forest catchments, (Fig. 2b). The aquifers in the upper and lower slopes of pasture  
catchment have the highest  $K_s$  values of ~0.31 m day<sup>-1</sup>, whereas  $K_s$  values of the aquifers in  
the forest are lowest on the lower slopes (Fig. 2, Table S1). The aquifers contain rocks from  
440 the same stratigraphic unit, and the heterogeneous hydraulic properties probably reflect the  
degree of weathering, cementation, and clay contents.

### 4.2 Major ions

TDS concentrations of regional groundwater range from 282 to 7850 mg L<sup>-1</sup> in the pasture  
catchment and 1190 to 7070 mg L<sup>-1</sup> in the forest catchment (Table S1); the lowest salinity  
445 regional groundwater is from the upper slope of the pasture catchment. The TDS concentrations  
of the shallow riparian groundwater (≤1 m depth) are between 3890 and 8180 mg L<sup>-1</sup> in the  
pasture and from 169 to 13600 mg L<sup>-1</sup> in the forest (Table S1). The regional and riparian

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groundwater from both catchments has similar geochemistry. Na constitutes up to 67 % of the total cations on a molar basis, and Cl accounts for up to 91 % of total anions on a molar basis.

455 Cl concentrations range between 45.2 and 8140 mg L<sup>-1</sup>, which significantly exceed the mean concentrations of Cl in local rainfall (2.2 to 4.4 mg L<sup>-1</sup>: Hutton and Leslie, 1958; Bormann, 2004; Dean et al., 2014). Molar Cl/Br ratios are between 180 and 884 with most between 450 and 830 (Fig. 4a), which spans those of seawater and coastal rainfall (~650: Davies et al., 1998, 2001). Cl/Br ratios are significantly lower than those that would result from halite dissolution (10<sup>4</sup> to 10<sup>5</sup>: Kloppmann et al., 2001; Cartwright et al., 2004, 2006) and do not increase with increasing Cl concentrations. These observations indicate that, as is the case throughout southeast Australia (e.g., Herczeg et al., 2001; Cartwright et al., 2006), Cl is predominantly derived from rainfall and concentrated by evapotranspiration. There is also no halite reported in the aquifers in this region. Cl concentrations of the shallow and deeper groundwater overlap (Fig. 4b) and there is no correlation between Cl and <sup>3</sup>H (Fig. 4c). Ca and HCO<sub>3</sub> concentrations are uncorrelated (Fig. 4d) indicating that the dissolution of calcite is not a major process influencing groundwater geochemistry.

### 4.3 Radioisotopes

470 <sup>3</sup>H activities of the regional groundwater are up to 1.48 TU (Table S1, Fig. 5). These are lower than the average annual <sup>3</sup>H activities of present-day rainfall in this region of ~2.8 TU (Tadros et al., 2014; Table S1). The highest <sup>3</sup>H activities (>1 TU) are from the regional groundwater in the upper slopes (15.5 m depth) and the drainage zone (~1.3 m depth) of the pasture catchment and between 15.8 and 28.8 m depths in the forest catchment (Table S1). Regional groundwater from ≥28 m depth in the lower slopes of the pasture catchment and the drainage zone of the forest catchment locally have below detection (<0.02 TU) <sup>3</sup>H activities (Table S1). The <sup>3</sup>H activities of the shallow riparian groundwater in the pasture vary from 0.26 to 0.79 TU with the highest activities from the lower slopes (Table S1, Fig. 5). The riparian groundwater in the

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forest catchment has  $^3\text{H}$  activities ranging from 2.01 to 4.10 TU (Table S1, Fig. 5), which are locally higher than the annual average  $^3\text{H}$  activity of present-day rainfall (~2.8 TU). These high  $^3\text{H}$  activities probably reflect seasonal recharge by winter rainfall that in southeast Australia has higher  $^3\text{H}$  activities than the annual average (Tadros et al., 2014).

The  $^{14}\text{C}$  activities in regional groundwater from the pasture and forest catchments range from 70.7 to 104 (pMC) and from 29.5 to 101 (pMC), respectively (Table S1, Fig. 5). The highest  $^{14}\text{C}$  activities (>100 pMC) are from the groundwater in the upper slopes of the pasture catchment and the lower zones of the forest catchment that also has high  $^3\text{H}$  activities (Table S1). The lowest  $^{14}\text{C}$  activities are from groundwater at 18 to 28.4 m depths in the mid-slope and drainage lines of the forest catchment (Table S1).  $^{14}\text{C}$  activities of the shallow riparian groundwater are 85.5 to 102 pMC, with higher activities (>100 pMC) in the drainage zones of the forest catchment (Table S1, Fig. 5).

## 5 Discussion

The combined groundwater elevation and geochemical data allow residence times, mixing, and recharge rates at Gatun to be interpreted.

### 5.1. Mean residence times and mixing

The  $^3\text{H}$  and  $^{14}\text{C}$  activities help understand water mixing within the aquifers (Le Gal La Salle et al., 2001; Cartwright et al., 2006, 2013) and the mean residence times. The predicted  $^3\text{H}$  vs.  $^{14}\text{C}$  activities (Fig. 5) were calculated for all DIC being introduced by recharge ( $q = 1$ ) and for 10% contribution of  $^{14}\text{C}$ -free DIC from the aquifer matrix ( $q = 0.9$ ). Mixing between older (low  $^3\text{H}$  and low  $^{14}\text{C}$ ) and recently-recharged groundwater (high  $^3\text{H}$  and high  $^{14}\text{C}$ ) results in groundwater samples that plot to the left of the decay trends in Fig. 5. It is difficult to calculate MRTs for these mixed waters; however, it is possible to estimate MRTs from the  $^{14}\text{C}$  activities for groundwater lying close to the predicted decay trends. The aquifers are dominated by siliceous rocks, and the major ion geochemistry implies little calcite dissolution. Similar values of  $q$

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were estimated for groundwater from other siliceous aquifers, in southeast Australia (Cartwright et al., 2010, 2012; Atkinson et al., 2014; Raiber et al., 2015; Howcroft et al., 2017) and elsewhere (Vogel, 1970; Clark and Fritz, 1997). Much lower  $q$  values are precluded as samples

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cannot lie to the right of the  $^3\text{H}$  vs.  $^{14}\text{C}$  curves (Cartwright et al., 2006, 2013, 2017). This is because samples that are not a mixture of old and young groundwater, containing measurable  $^3\text{H}$  will be less than 200 years old. Over that time span, there has been negligible decay of  $^{14}\text{C}$ , and the initial  $a^{14}\text{C}$  of the sample is  $a^{14}\text{C}/q$  (Clark and Fritz, 1997). If there were greater than 10% contribution of DIC from  $^{14}\text{C}$ -free calcite dissolution, the estimated initial  $a^{14}\text{C}$  would exceed the highest  $a^{14}\text{C}$  recorded in soil  $\text{CO}_2$  of  $\sim 120$  pMC.

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The calculated MRTs are up to 3,930 years in the pasture and up to 24,700 years in the forest (Table 1, Fig. 6). While using LPMs is preferable to using a simple decay equation that assumes piston flow and ignores variations in the  $^{14}\text{C}$  input function, there are uncertainties in the calculated MRTs. The different LPMs have different residence time distributions, and so yield different MRT estimates. Additionally, there are uncertainties in  $q$  and the input function of  $^{14}\text{C}$ . Previous studies (e.g., Atkinson et al., 2014; Howcroft et al., 2017) estimated overall uncertainties in MRTs were up to 25%. While these are considerable, much of the regional groundwater undoubtedly have residence times of several thousands of years and were recharged prior to land clearing. These long residence times are consistent with the locally clay-rich nature of the aquifers and the moderate to low hydraulic conductivities.

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## 5.2 Recharge rates

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### 5.2.1 Cl mass balance

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Recharge rates calculated from the CMB method (Eq. 1) using total rainfall of  $\sim 640$  mm  $\text{yr}^{-1}$  and Cl concentrations of 2.2 to 4.4 mg  $\text{L}^{-1}$  are similar between the pasture (0.3 to 61.6 mm  $\text{yr}^{-1}$ ) and forest (0.2 to 58.8 mm  $\text{yr}^{-1}$ ) catchments (Figs 2, 7a). The typical recharge rates for most of the regional groundwater are from 0.3 to 2.5 mm  $\text{yr}^{-1}$  in the pasture and 0.2 to 11.2 mm  $\text{yr}^{-1}$  in

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the forest (Figs. 2, 7a). The Cl/Br ratios imply that dissolution of halite is negligible, and all the Cl is delivered by rainfall. Whether the rate of Cl delivery has been constant over long time periods is more difficult to assess; however, the rainfall Cl concentrations are typical of inland rainfall, and southeast Australia does not record major climate fluctuations such as glaciations or monsoons (Davies and Crosbie, 2018).

The CMB technique also assumes that the export of Cl by surface runoff is negligible. The streams at Gatam currently discharge ~8 % of local rainfall, and much of the Cl that they export represents groundwater discharging into the stream (Adelana et al., 2014). This component of Cl does not impact the CMB recharge rate calculations. If some direct export of Cl has occurred, the recharge estimates would be slightly lower than estimated above. However, because the initial land-clearing has most likely increased streamflow in this region (Dresel et al., 2018), streamflows and the export of Cl would have historically been lower than the present day.

Because Cl in groundwater accumulates over hundreds to thousands of years (Scanlon et al., 2002, 2006), the CMB method generally yields longer-term recharge rates; these largely reflect pre-land clearing recharge in Australia (Alison and Hughes, 1978; Cartwright et al., 2007; Dean et al., 2015; Perveen, 2016). This conclusion is consistent with the long <sup>14</sup>C residence times of much of the deeper regional groundwater at Gatam. The higher recharge rates (25.3 to 61.6 mm yr<sup>-1</sup>) are from regional groundwater in the upper slopes of the pasture (bore 63) and shallow riparian groundwater in the drainage zones (piezometer FD2) and lower slopes (piezometer FB1) of the forest (Figs. 2, 7a). The groundwater at these sites has high <sup>3</sup>H and <sup>14</sup>C activities, and the recharge rates from the CMB technique are likely to represent recent recharge.

### 5.2.2 Water table fluctuations

The recharge rates were calculated using the WTF method (Eq. 2) from the bore hydrographs, which show seasonal head variations assuming  $S_y = 0.03$  to  $0.1$ . The estimated recharge rates range from 15 to 500 mm yr<sup>-1</sup> (2 to 78 % of rainfall) in the pasture and 30 to 400 mm yr<sup>-1</sup> (5 to

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Deleted: This can be demonstrated by mass balance (Cartwright et al., 2007). A 10 to 20 m thickness of aquifer with a unit area of 1 m<sup>2</sup> and porosity 0.03 to 0.1 contains 300 to 2000 L of water. Cl concentrations in the groundwater at Gatam range from 45.2 to 8140 mg L<sup>-1</sup> which equates to 1.4 x 10<sup>4</sup> to 1.6 x 10<sup>7</sup> mg Cl in that section of the aquifer. Annual rainfall of ~640 mm with Cl concentrations of 2.2 to 4.4 mg L<sup>-1</sup> would deliver 1410 to 2820 mg Cl per m<sup>2</sup> each year. Thus, it takes up to 11,500 years to deliver the Cl contained in that section of the aquifer.

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63 % of rainfall) in the forest (Figs. 2, 7b). As with the CMB estimates, the recharge rates are generally high at the upper slopes of the pasture catchment (Figs. 2, 7b). However, the highest recharge rates from the WTF method are unlikely given that evapotranspiration rates in this region approach the rainfall rates (Dean et al., 2016; Dresel et al., 2018; Azarnivand et al., 2020). The lower recharge rates estimated from the WTF method appear more reasonable but are still larger than most recharge rates estimated from the TRR method. The observation that much of the older saline groundwater has not been flushed from the catchments also implies that present-day recharge rates cannot be this high.

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The WTF method requires the hydrograph recession curves to be estimated. There are significant steep and straight recession curves in the bore hydrographs (Figs. 3b, 3c) that can lead to errors in recharge estimates. The WTF method may overestimate recharge due to air entrapped during recharge (the Lisse effect: Crosbie et al., 2005). However, this occurs during rapid recharge, which is not observed in the Gatam area. Dean et al. (2015) suggested that the high recharge rates estimated from the WTF method in the adjacent Mirranatwa catchments might reflect focussed recharge from streams. This is not the case at Gatam as high WTF recharge rates are recorded at all landscape positions and the streams only export ~8% of rainfall (Adelana et al., 2014). Because the WTF estimates gross recharge and geochemical methods estimate net recharge, there may be differences if the water is removed from the water table by evapotranspiration, especially in spring after the water tables reach their seasonal peak. The plantation forest plausibly has high evapotranspiration rates (Benyon et al., 2006; Dean et al., 2015; Dresel et al., 2018); however, this explanation is unlikely in the pasture where water tables are locally several metres below land surface, and there is not deep-rooted vegetation. It is most likely that the unrealistically high recharge rates estimated from the WTF method reflect an overestimation of  $S_y$  due to the presence of remnant moisture in the unsaturated zone between recharge events (Gillham, 1984; Sophocleous, 1985; Crosbie et al., 2005, 2019).

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While this is not unexpected, it is difficult to determine realistic values of  $S_y$  to improve these estimates.

### 5.2.3 $^3\text{H}$ renewal rate

650 The recharge rates for bores and shallow piezometers were estimated using the  $^3\text{H}$  activities and the TRR method (Eqs. 3, 4). These recharge rates were calculated for those groundwater samples which do not show the mixing of recent and older groundwater (Fig. 5, Table 1).

Regional groundwater from nested bores commonly has different TDS contents, EC values,  $^3\text{H}$  and  $^{14}\text{C}$  concentrations (Table S1), indicating that the groundwater is stratified. Much of the  
655 deeper groundwater has low  $^3\text{H}$  and  $^{14}\text{C}$  activities implying that it is not recently recharged.

Based on these differences in geochemistry, (Table S1),  $b$  is estimated as being between 1 and 5 m in the regional groundwater.  $b$  values for the shallow riparian groundwater are estimated as 1 to 2 m, which is the approximate thickness of the shallow perched aquifers (Brouwer and Fitzpatrick, 2002). The estimated  $n$  values of 0.03 to 0.1 (Adelana et al., 2014) were used for  
660 these calculations.

Recharge rates from the regional groundwater are 0.5 to 14.0 mm yr<sup>-1</sup> in the pasture and 0.01 to 59.5 mm yr<sup>-1</sup> in the forest catchment with most in the range of 0.01 to 0.6 mm yr<sup>-1</sup> (Figs. 2, 7c). The higher recharge rates were from the upslopes of the pasture (14.0 mm yr<sup>-1</sup>) and lower slopes of the forest (59.5 mm yr<sup>-1</sup>). The recharge rates in the riparian groundwater are from  
665 0.05 to 0.5 mm yr<sup>-1</sup> in the pasture and 13.3 to 89.0 mm yr<sup>-1</sup> in the forest (Figs. 2, 7c).

The average annual  $^3\text{H}$  activity in present-day rainfall at Gatum (~2.8 TU) is within the predicted range of the  $^3\text{H}$  activities in present-day Melbourne rainfall ( $3.0 \pm 0.2$  TU), implying that the Melbourne  $^3\text{H}$  input function is appropriate to use for this area. Assuming uncertainty in the  $^3\text{H}$  input function of 5 to 10% (which is similar to the present-day variability of  $^3\text{H}$   
670 activities reported by Tadros et al., 2014) results in <5% uncertainties in recharge estimates. The variation resulting from analytical uncertainties are lower than this. Recharge rates are

**Deleted:** Fig. 2) that can lead to errors in recharge estimates. The values of  $S_y$  are not well known, which also results in uncertainties in the recharge estimates. Also, as discussed above, the presence of moisture in the unsaturated zone and capillary fringe may reduce the effective values of  $S_y$  leading to recharge rates being overestimated. These uncertainties are discussed further below. Overall, the recharge rates estimated by this method are higher than those estimated using CMB and reflect present-day recharge rates. ¶ 4.

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most sensitive to the  $b$  values, which are not explicitly known and may be variable. However,  $b$  is unlikely to be  $>5$  m based on the observed degree of chemical stratification. The recharge rates are again generally higher than those calculated using the CMB, which reflects the effects of the initial land clearing. However, despite both reflecting post-land clearing recharge, they are significantly lower than those estimated using the WTF.

### 5.3. Predicting the effect of land-use changes

In large regions of southeast Australia (including the study area), understanding whether and by how much recharge increased following the initial land clearing is important in predicting the impact of a rising water table in causing salinization of the soils and streams. For areas where plantation forests have been established, it is important to assess any subsequent impact of those plantations on recharge.

As expected, the recharge estimates from the CMB method are generally lower than those from the WTF and TRR methods and largely reflect those prior to the initial replacement of native eucalyptus vegetation by pasture. Although both methods determine present-day recharge rates (Scanlon et al., 2002, 2006), those estimated using the WTF method are significantly higher than the TRR estimates (Fig. 8). Having to estimate  $b$  represents a major uncertainty in the TRR calculations; however,  $b$  would have to be up to 50 m to achieve agreement between the recharge estimates from these two methods. This is unlikely given the observations that major ion geochemistry,  $^3\text{H}$  and  $^{14}\text{C}$  activities of groundwater vary over vertical scales of a few metres (Table S1), implying that the groundwater is compartmentalised on those scales. It is also unlikely that  $b$  could be so large given the heterogeneous nature of the aquifers and the presence of clay layers. It is most likely that the WTF method systematically overestimates recharge due to issues in estimating  $S_y$ .

The recharge estimates from the TRR method differ little between the pasture and the forest; this is unexpected given that the establishment of plantation forests aimed to reduce the

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740 recharge rates. The evapotranspiration rates in the forest are also higher than in the pasture  
(Adelena et al., 2014; Dresel et al., 2018) and water levels are declining in some areas of the  
forest with no corresponding decline in the pasture (Figs. 3b, 3c), suggesting higher water use  
by the trees. The plantation covers ~62% of the forest catchment, and many of the bores are in  
cleared areas between the stands of trees (Fig. 1a). Thus, the recharge rates may not be  
745 representative of the forest as a whole. Additionally, the TRR averages recharge rates over the  
timespan of the residence times of the aliquots of water contained in the water sample  
(Maloszewski and Zuber, 1982; Cartwright et al., 2017). If the zone at the top of the aquifer  
approximates a well-mixed reservoir, the timespan is  $1/R_T$  (Leduc et al., 2000; Favreau et al.,  
2002).  $R_T$  values at Gatum are  $3 \times 10^{-4}$  to  $4 \times 10^{-1}$ , implying that recharge rates are averaged over  
750 decades to centuries. Thus, the recharge rates in the forest catchment may reflect those from  
both before and following the recent reforestation.

## 6. Conclusions

As has been discussed elsewhere (Scanlon et al., 2002; Healy, 2010; Crosbie et al., 2010, 2019;  
Cartwright et al., 2017; Moeck et al., 2020), estimating recharge rates can be difficult and a  
755 range of techniques together with other data (such as estimates of residence times) is required  
to produce reliable results. By necessity, estimating pre- and post-land clearing recharge rates  
requires different methods. Both the CMB and WTF methods use data that is readily available  
(or is relatively low cost to attain). The uncertainties in the CMB estimates are relatively  
straightforward to address, and this represents a viable method of estimating historic recharge  
760 rates; however, the commonly-used WTF method may not be able to be applied in a  
straightforward manner to estimate modern recharge rates. Relatively high WTF recharge rates  
(up to 161 and 366 mm yr<sup>-1</sup>) were also calculated in adjacent catchments with similar land-use  
(Dean et al., 2015; Perveen, 2016). <sup>3</sup>H activities in groundwater from those catchments are  
similar to those in the same region, implying that recharge estimates based on the TRR method

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would again be significantly lower. Cartwright et al. (2007) and Crosbie et al. (2010) also reported that the recharge estimates from the TRR method and other geochemical tracers in semi-arid catchments elsewhere in Australia are lower than those from the WTF method.

780 Additionally, the recharge rates are likely to be spatially variable across both catchments, and even with a relatively high density of data such as at Gatum, it is difficult to estimate typical or area-integrated values. In the case of understanding recharge rates in the plantation forest, the necessity that bores are in cleared areas (between the stands of trees) also makes it questionable whether the recharge rates are representative. Finally, all the geochemical techniques integrate recharge rate estimates over years to centuries and are thus ineffective at 785 determining changes over shorter timescales than this.

Detailed soil moisture measurements that would improve  $S_v$  estimates and geochemical tracers, such as  $^3\text{H}$ , may not always be available. Integrated surface and subsurface hydrogeologic models, which simulate coupled groundwater, surface water, and soil water fluxes, might provide additional tools to estimate recharge rates that could be used to support the field and 790 geochemical data (Scudeler et al., 2016; Daneshmand et al., 2019). With the increasing availability of soil moisture, evapotranspiration, rainfall, streamflow and groundwater elevation data, catchment water balance models (e.g., Wada et al., 2010; Moeck et al., 2020) might also represent viable methods of estimating recharge, especially over large areas.

795 The results of this study inform the understanding of hydrogeological processes in this and similar semi-arid regions globally. The present-day recharge rates in the pasture, which is typical of cleared land in southeast Australia, are likely to be  $<10 \text{ mm yr}^{-1}$ . Despite these being significantly higher than the pre-land clearing recharge rates, they only result in the gradual replacement of the older saline water stored in these aquifers, (as is implied by the trends of Cl vs depth and  $^3\text{H}$  vs. Cl: Figs. 4b, 4c). Additionally, while there has been a rise in the water 800 table caused by increased recharge, and in some cases increased drainage in the streams, the

**Moved up [11]:** However, this occurs during rapid recharge, which is not observed in the Gatum area. Dean et al. (2015) suggested that the high recharge rates estimated from the WTF method in the adjacent Mirranatwa catchments might reflect focussed recharge from streams. This is not the case at Gatum as high WTF recharge rates are recorded at all landscape positions and the streams only export ~8% of rainfall (Adelana et al., 2014). Because the WTF estimates gross recharge and geochemical methods estimate net recharge, there may be differences if the water is removed from the water table by evapotranspiration

**Moved up [12]:** The plantation forest plausibly has high evapotranspiration rates (Benyon et al., 2006; Dean et al., 2015; Dresel et al., 2018); however, this explanation is unlikely in the pasture where water tables are locally several metres below land surface, and there is not deep-rooted vegetation. ¶

**Moved up [13]:** Crosbie et al., 2005, 2019).

**Deleted:** This conclusion is consistent with the soils being fine-grained and thus

**Deleted:** retain moisture between recharge events. While it is difficult to test this possibility, it is clear that the estimates from the WTF method are higher than expected, given the evapotranspiration rates and the preservation of old groundwater in these catchments. The recharge rates estimated from the TRR method are still subject to uncertainty (especially in determining  $b$ ) but are probably a more reasonable estimate. ¶

**Moved up [14]:** is unexpected given that the establishment

**Moved up [15]:** higher water use by the trees. The

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**Moved up [16]:** Thus, the recharge rates in the forest

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magnitude of these changes will be limited by the modest recharge rates. The results also indicate that care must be used in assigning recharge rates as boundary conditions numerical models.

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*Author contribution:* Shovon Barua and Ian Cartwright conducted the sampling assisted by P. Evan Dresel and Edoardo Daly. Shovon Barua carried out the analytical work conducted at Monash University. P. Evan Dresel and Edoardo Daly manage the field site and provided pre-existing data. All authors were involved in writing the manuscript.

*Competing interests:* The authors declare that they have no conflict of interest.

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*Data Availability:* All analytical data is presented in the Supplement. Groundwater head data are from Dresel et al. (2018).

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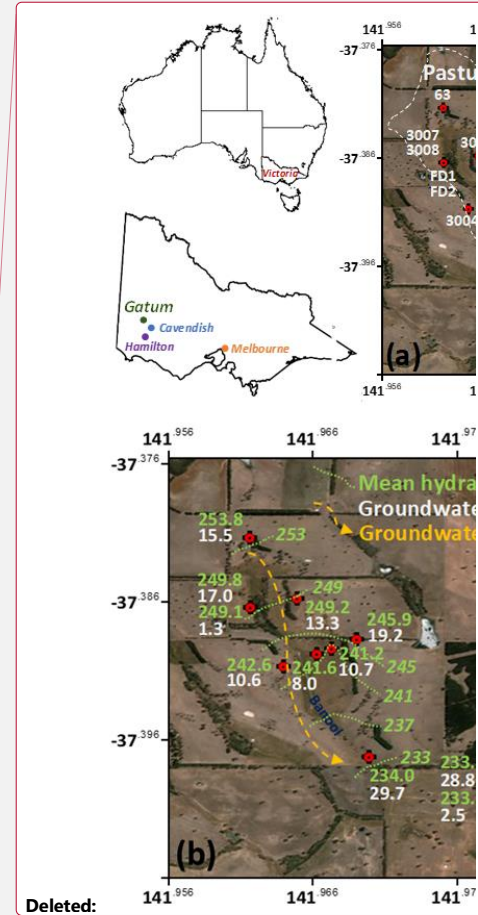
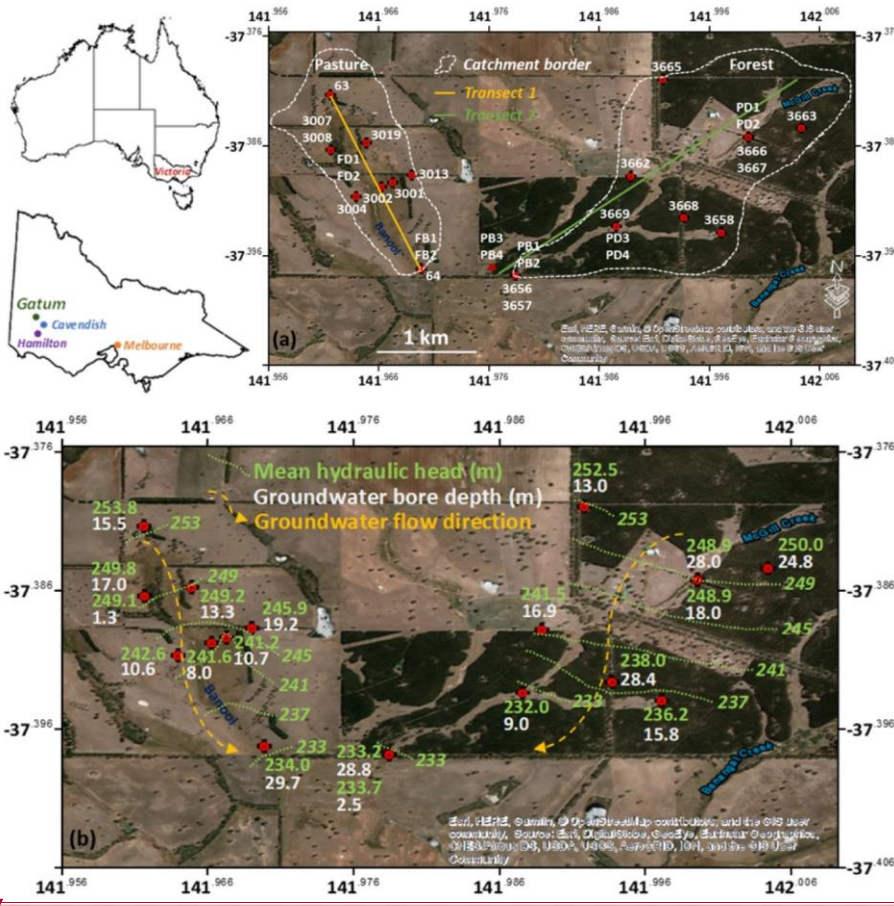
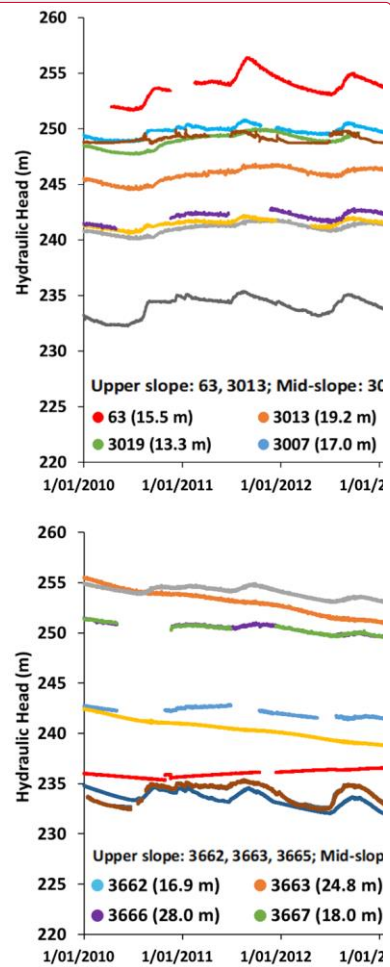
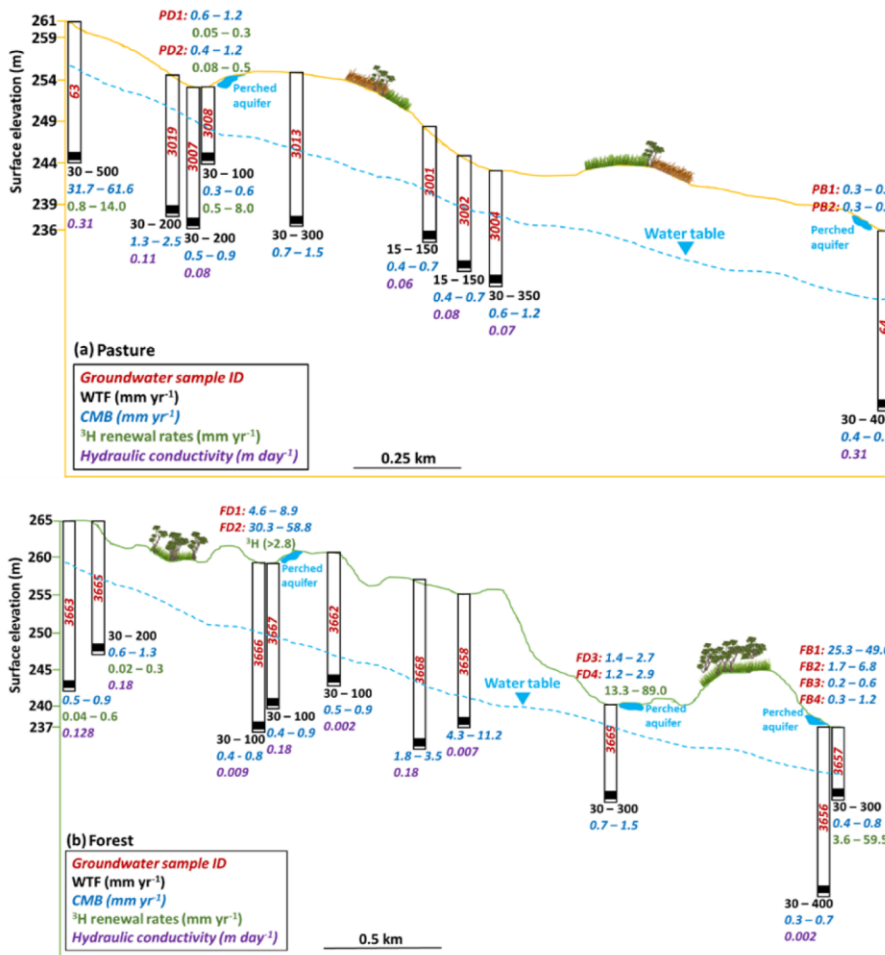


Figure 1: (a) Map of the Gatam pasture and forest catchments with the locations of groundwater bores (3007 & 3008, 3666 & 3667, and 3656 & 3657 are nested bores); shallow piezometers are at PD (pasture drainage zone), PB (pasture lower slope), FD (forest drainage zone), and FB (forest lower slope). The catchment boundaries for the streams are from Dresel et al. (2018). (b) Mean hydraulic heads of groundwater from 2010 to 2017 except for 3008 (from 2010 to 2015) and 3658 (from 2010 to 2016) with sample depths and flow directions. Background ArcGIS®10.5 image (Esri, HERE, Garmin, ©OpenStreetMap contributors and the GIS User Community, Source: Esri, DigitalGlobe, GeoEye, Earthstar Geographics, CNESAirbus DS, 1315 USDA, USGS, AeroGRID, IGN, and the GIS User Community).



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Figure 2: Simplified cross sections of (a) pasture and (b) forest catchments showing variability of groundwater recharge estimated via WTF, CMB, <sup>3</sup>H methods and variable hydraulic conductivity of the aquifer lithologies. Transects are on Fig. 1a. PD and FD represent the shallow groundwater in the pasture drainage and forest drainage areas, respectively. Data from Table 1.

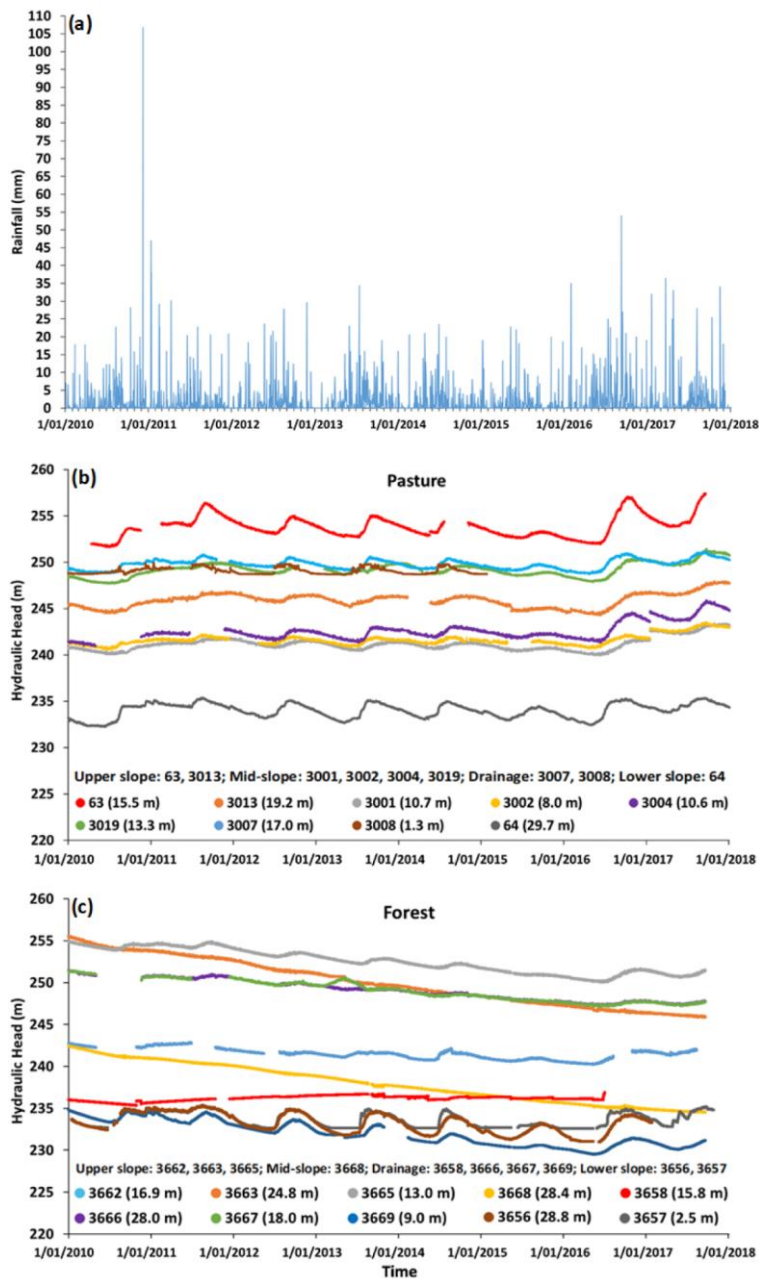


Figure 3: (a) Daily rainfall at Datum. Variation in groundwater heads from bores in (b) pasture and (c) forest (Dresel et al., 2018). The legend shows the sample depths (in parentheses) and landscape positions.

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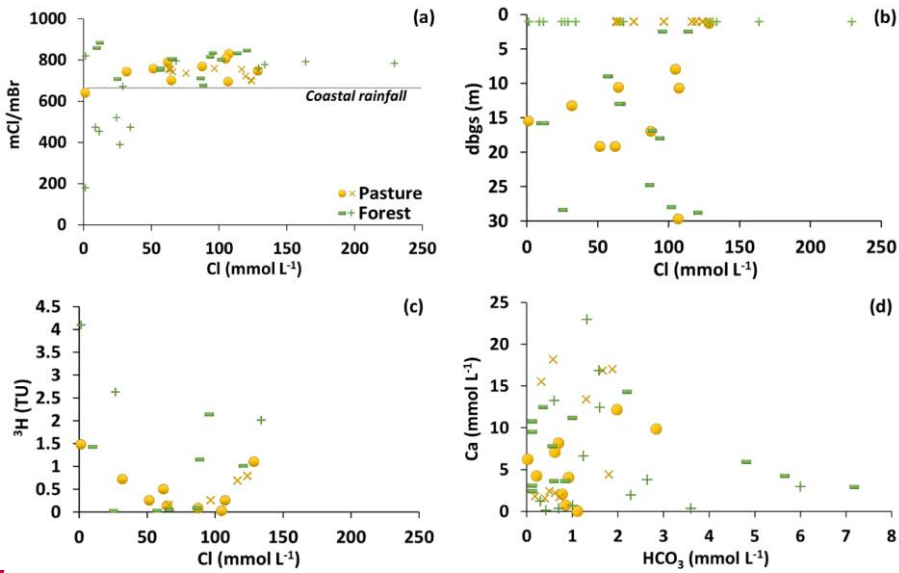
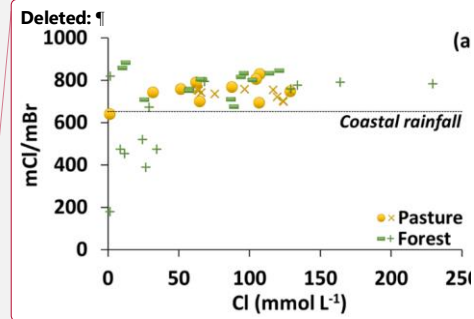
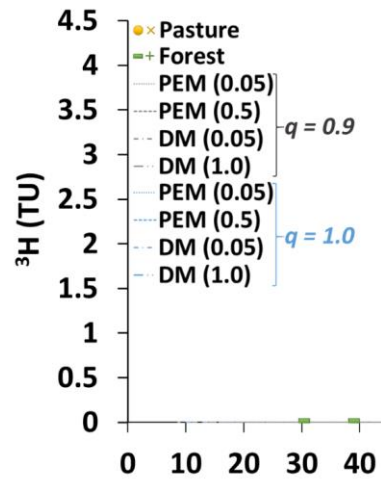


Figure 4: (a) Variation of molar Cl/Br ratios with molar concentrations of  $\text{Cl}_w$ . (b) Molar Cl concentrations across depth below ground surface (dbgs: m). (c)  $^3\text{H}$  (TU) vs. molar Cl concentrations. (d) Molar Ca vs.  $\text{HCO}_3$  concentrations. Cross and plus symbols are for the shallow riparian groundwater and other symbols are for the regional groundwater.



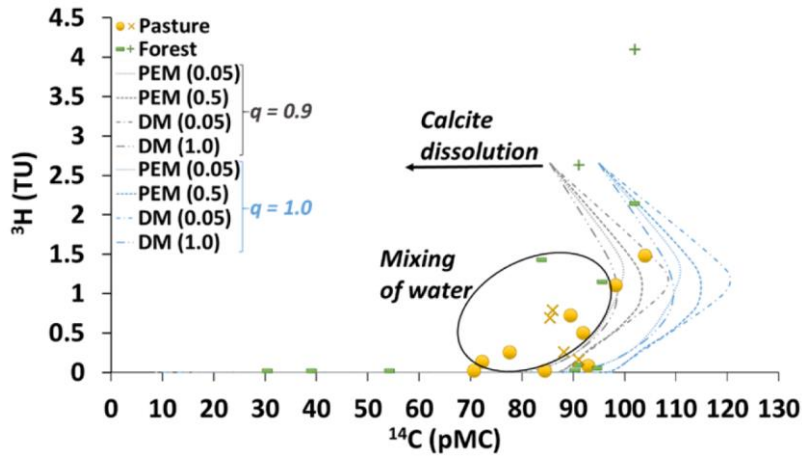
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Figure 5: Activities of  $^3\text{H}$  (TU) and  $^{14}\text{C}$  (pMC) in the pasture and forest groundwater. PEM = partial exponential model (PEM ratio in brackets) and DM = dispersion model (DP parameter in brackets). Cross and plus symbols are for the shallow riparian groundwater and other symbols are for the regional groundwater. The single high  $^3\text{H}$  activity possibly reflects recharge by winter rainfall. Samples lying to the left of the covariance curves probably record mixing between younger and older groundwater (see text for discussion).

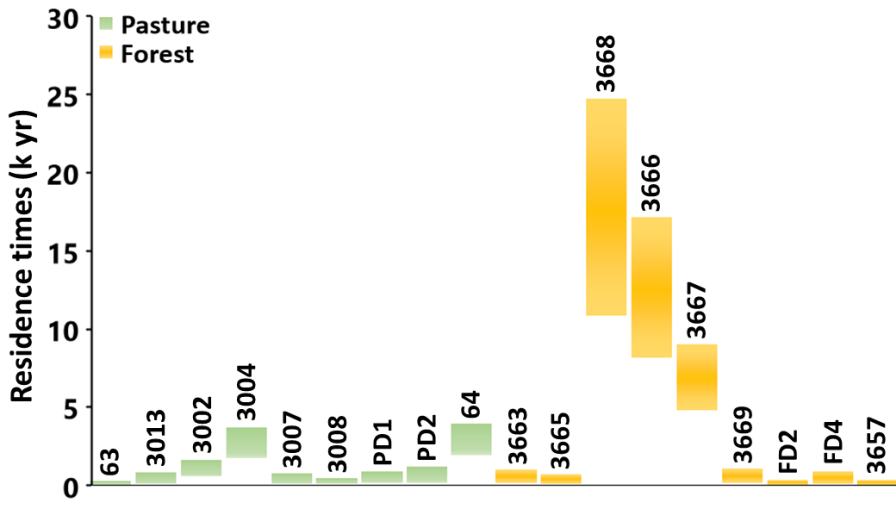
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1365 Figure 6: Ranges of groundwater residence times in k yr estimated using different LPMs. The numbers above the box represent sample IDs. PD and FD represent the shallow groundwater in the pasture drainage and forest drainage areas, respectively. Data from Table 1.

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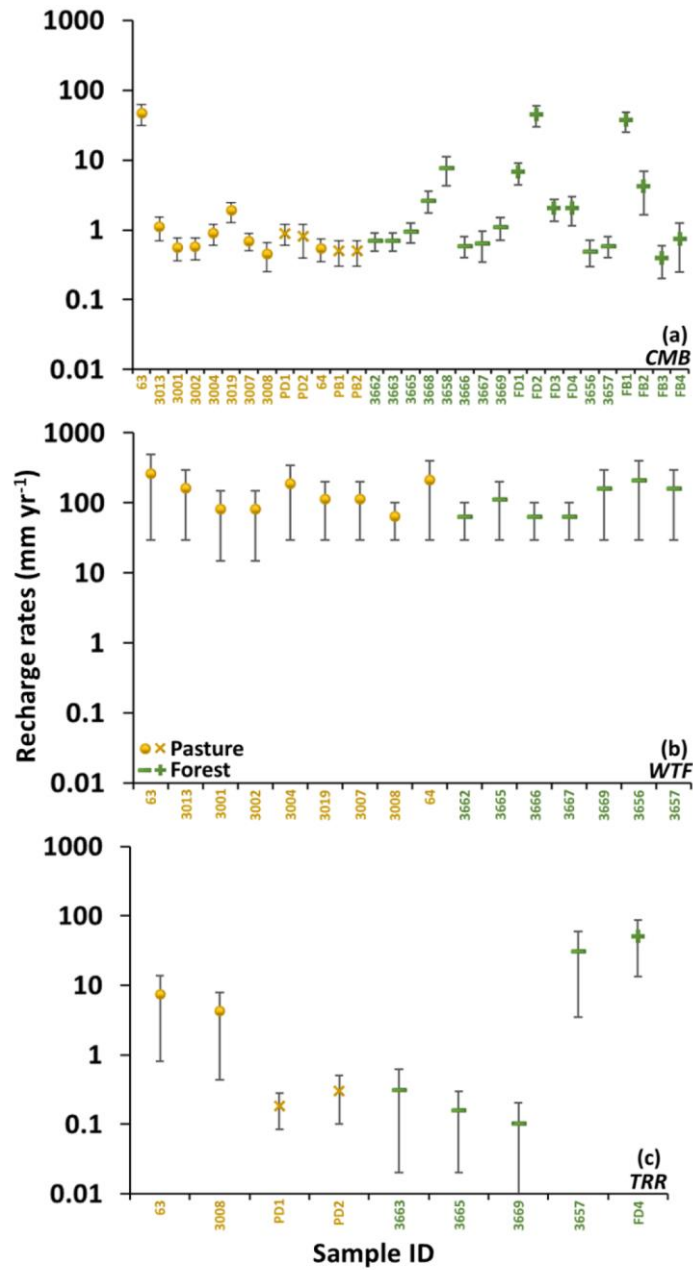
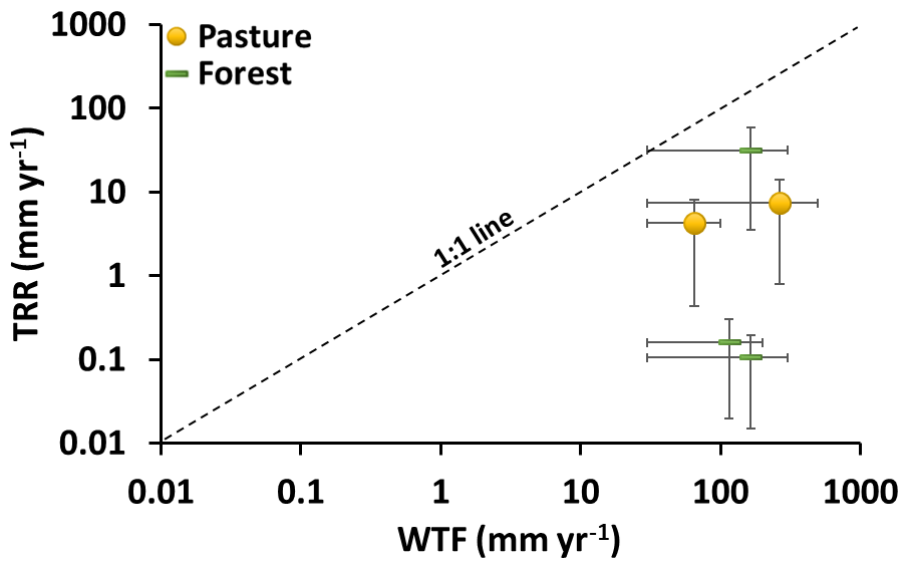


Figure 7: Recharge rates in  $\text{mm yr}^{-1}$  estimated from (a) CMB, (b) WTF and (c) TRR. PD and FD are for the shallow groundwater in the pasture drainage and forest drainage areas, respectively. Bars indicate the ranges of recharge rates from Table 1.

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1380 Figure 8: Comparison between recharge rates for the regional groundwater estimated from WTF and TRR. Bars represent the ranges of calculated recharge values from Table 1.

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Table 1: Groundwater recharge rates and estimated residence times of groundwater.

Sample	Sample depth (m)	Landscape position	Recharge rates (mm yr <sup>-1</sup> )			Groundwater residence times (yr)							
			WTF	CMB	TRR	PEM (0.05)		PEM (0.5)		DM (0.05)		DM (1.0)	
						q = 0.9	q = 1.0	q = 0.9	q = 1.0	q = 0.9	q = 1.0	q = 0.9	q = 1.0
<b>Pasture Catchment</b>													
63	15.5	Upper	30-500	31.7-61.6	0.8-14.0		180	60	150	70	80		270
3013	19.2	Upper	30-300	0.7-1.5		210	780	140	690	90	680	270	780
3001	10.7	Mid	15-150	0.4-0.7									
3002	8	Mid	15-150	0.4-0.7		660	1470	540	1380	540	1290	650	1620
3004	10.6	Mid	30-350	0.6-1.2		2010	3200	1860	2910	1710	2730	2220	3650
3019	13.3	Mid	30-200	1.3-2.5									
3007	17	Drainage	30-200	0.5-0.9		190	720	160	600	90	600	270	720
3008	1.3	Drainage	30-100	0.3-0.6	0.5-8.0	70	390	110	200	80	120	90	420
PD1	1	Drainage		0.6-1.2	0.05-0.3	240	860	170	750	110	740	320	870
PD2	1	Drainage		0.4-1.2	0.08-0.5	390	1080	200	1020	120	960	420	1170
64	29.7	Lower	30-400	0.4-0.7		2240	3470	2070	3150	1920	2960	2510	3930
PB1	1	Lower		0.3-0.7									
PB2	1	Lower		0.3-0.7									
<b>Forest Catchment</b>													
3662	16.9	Upper	30-100	0.5-0.9									
3663	24.8	Upper		0.5-0.9	0.04-0.6	320	960	180	870	110	830	360	990
3665	13	Upper	30-200	0.6-1.3	0.02-0.3	170	660	150	540	90	560	250	660
3668	28.4	Mid		1.8-3.5		17000	19600	13100	14700	10800	11900	21400	24700
3658	15.8	Drainage		4.3-11.2									
3666	28	Drainage	30-100	0.4-0.8		11500	13600	9480	10900	8160	9230	14300	17100
3667	18	Drainage	30-100	0.4-0.9		5850	7440	5160	6450	4780	5870	6930	9000
3669	9	Drainage	30-300	0.7-1.5	0.01-0.2	330	990	180	930	110	870	380	1020
FD1	1	Drainage		4.6-8.9									
FD2	1	Drainage		30.3-58.8	<sup>3</sup> H (>2.8)		210	70	170	80	90		300
FD3	1	Drainage		1.4-2.7									
FD4	1	Drainage		1.2-2.9	13.3-89.0	260	860	170	750	90	740	320	870
3656	28.8	Lower	30-400	0.3-0.7									
3657	2.5	Lower	30-300	0.4-0.8	3.6-59.5		300	90	170	80	110		330
FB1	1	Lower		25.3-49.0									
FB2	1	Lower		1.7-6.8									
FB3	1	Lower		0.2-0.6									
FB4	1	Lower		0.3-1.2									

Landscape positions: Upper, Mid, and Lower slopes as discussed in text. Sample depth is the middle of the screened interval. The recharge rates from WTF method were calculated for bore hydrographs that show seasonal variations in hydraulic head. The recharge rates with TRR were calculated assuming *b* was 1 to 5 m (bores) and 1 to 2 m (shallow piezometers). The groundwater samples that do not show mixing of young and old groundwater, were calculated for recharge rates from TRR and residence times. Groundwater residence times not calculate for those samples which exceed the upper limit of <sup>14</sup>C concentrations in lumped parameter models.

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