

General Comments: This manuscript has potential to be a very interesting article and could have a big impact on our current understanding of the interactions between alluvial aquifers, regional aquifers, and surface-water systems. The topic should have broad interest to other research on alluvial aquifers and broad applicability to global alluvial groundwater systems. The combined geochemical tracer and multiple isotopic tracer approach is a potentially robust way to sort out these interactions. However, the manuscript suffers from poor organization and lack of specificity in the methods and interpretations. This prevents a recommendation to accept the article as it is currently written. Instead, the recommendation is to accept after major revisions including the organization and technical detail of the article. In addition, it was very difficult to read the manuscript due to awkward syntax and lack of focus (see Specific Comments).

We thank the reviewer for their time in reviewing our manuscript closely. We have attempted to address all concerns below, particularly the major concern of the organisation and technical detail of the article and the syntax and focus. The manuscript has been restructured and this has made the manuscript more focused on the key messages.

The major findings/contributions of this manuscript hinge on the authors' interpretations of the isotopic and geochemical data. In general, these sections need much more clarification than is provided and in some cases, additional data mining may be necessary (although I think the latter is the smaller of the issues). For example, a considerable emphasis is placed on 3H concentrations. However, almost all of the 3H concentrations are close to background or are 3H dead. This has implications for the mixing model and it has implications for the residence time estimates. It's not unusual to find 3H dead waters which have slightly elevated 36Cl/Cl ratios. 3H, even from the bomb-pulse, is decaying faster than 36Cl even in the presence of mixing with recent recharge and sometimes 36Cl does a better job of sorting out mixing than 3H (especially for waters that are 100 years or older). However, the real strength that the authors have is the ability to sort out young and old fractions (relative terminology) using 14C pmc. In the absence of carbonate exchange and mixing in the aquifer (another topic which needs additional clarification) the 14C pmc value is very useful in sorting out these endmembers as the authors attempt to do in Figures 5 and 6. In any case, the authors need to provide additional clarification on how they sorted out the mixing processes that affect these tracers.

Regarding data mining, this manuscript and the authors' interpretations would be strengthened by providing the 3H, 14C, and 36Cl/Cl of modern precipitation. Are these data available? If so, cite them. This would enhance the mixing model which is used to determine the fractions of GAB water in the LNA. This calculation also needs additional clarification and the authors should present the equation and discuss sources/estimates of uncertainty in this calculation.

These are some interesting observations that we have endeavoured to address here and in the re-organised manuscript. Firstly, we would like to stress a couple of points specific to our data and the southern hemisphere that we will emphasise in the revised manuscript (lines 422-425). The quantification limit for 3H in our lab is 0.04 TU, so only 16 out of 50 samples are either at the detection limit or below it. This low detection limit has been driven by low 3H concentrations in the southern hemisphere. At the peak of the bomb pulse, weighted averages in Australia were around 60 TU (compared to, for example, ~ 6000 TU in Ottawa). In Australia, tritium in rainfall has been at natural background levels for some time. Values from samples in

the Namoi River, collected at the time of our groundwater sampling (and provided in the dataset: ~2.3 TU) are in good agreement with rainfall data provided in Tadros et al. 2014.

Regarding ^{14}C , whilst not generally sampled in rain or surface samples, it is almost at natural/background content, as seen in the Namoi River samples (~102 pmc; also provided in the dataset). However, shallow groundwater, recharged in recent years, still has a slight bomb pulse component in some boreholes (values of around 107 pmc).

Regarding $^{36}\text{Cl}/\text{Cl}$, no data in rainfall exist in the region to our knowledge. Namoi River samples at two different times (summer and winter 2016) have values of ~416 ($\times 10^{-15}$), while tributaries draining the igneous rock terrains have higher ratios than modern river water. However, most shallow groundwater also shows the mixing with deeper $^{36}\text{Cl}/\text{Cl}$ sources and therefore has lower $^{36}\text{Cl}/\text{Cl}$ ratios than modern surface waters.

We have added a 'Geochemical calculations' section to the methods (section 3.3) that details the method used for the calculations in old Section 4 (now section 5 in the rearranged manuscript). In terms of sources of uncertainty in this calculation, mixing and other processes (such as evapotranspiration and irrigation returns) can overprint the simple Cl mixing. However, we draw our interpretations from our Cl mixing results, alongside the other tracers that we used. For example, we consider samples with $[\text{Cl}] < 31 \text{ mg/L}$, ^{3}H activities above our detection limit and/or $^{14}\text{C} > 90 \text{ pmc}$ as originating from 100% modern (< 70 a) flood recharge (eg 36001-1&2, 25329-1, 30345-1, 25332-1, 25327-1&2). Samples recovered from generally deeper piezometers along the B-B' section in figure 2 show higher $[\text{Cl}]$ that coincide with ^{3}H below or very close to detection limits, ^{14}C contents generally < 5 pmc and, most importantly, $^{36}\text{Cl}/\text{Cl}$ below 58 ($\times 10^{-15}$).

Specific Comments:

Line 56: This paragraph lacks focus. The authors bring in agriculture and it's not clear how this is connected to the bigger issue.

This paragraph has been edited to try to bring it into focus. Agriculture is no longer mentioned here, just groundwater abstraction for irrigation, stock and domestic water supplies (lines 57-58).

Line 64: What is meant by the international export market?

This part of the introduction has been removed.

Lines 65-78: What is the focus here? Aquifers in general or alluvial aquifers?

Alluvial aquifers are the focus. This has been clarified in text at line 62.

Line 81: It's not just the half-life that is important, the tracer systematics including mixing and processes that affect their interpretations are needed.

We have acknowledged that each tracer undergoes processes specific to it that can often affect the interpretation of it and therefore these tracers can provide insights into the groundwater, but for only a given window of time and in favourable conditions (lines 80-81).

Lines 96-105: The manuscript needs a focused statement about the current knowledge

gaps (this should be developed in the Introduction more succinctly) and what is new and novel about this research in addressing those gaps. The manuscript would benefit from a clear hypothesis statement or statement of science questions. This, in my opinion, would help focus the entire manuscript and the authors should return to this statement in the opening paragraph of the conclusions.

The introduction has been edited and aspects of it cut. The last paragraph (lines 97-114) has been edited to highlight that we are for the first time (to our knowledge) combining multiple geochemical tracers to assess artesian discharge to an alluvial aquifer and that we show a need to combine this with water balance models of alluvial systems. This is reiterated in the conclusions.

Lines 208-213: This sentence is difficult to understand, yet this paragraph is critical in identifying the knowledge gaps.

This sentence has been changed to “..there are multiple plausible solutions for all water balance models...” at lines 198-199

Line 237: This is a good place to re-state or reiterate what is missing by identifying the knowledge gaps and how your research addresses those gaps.

We have moved a section of the introduction to here (lines 226-230), stating that there is an over-reliance on water balance models, which are not constrained by hydrochemistry and that this study shows that the conceptual insights from geochemistry provide a new set of constraints not considered in previous hydrogeological models.

Lines 292-293: Please clarify the statement on NH₄ concentrations.

A sentence has been added to clarify the statement on NH₄ concentration (lines 333-335). We explain that the NH₄ concentration skewed the charge balance because it was not considered in the initial calculations. This is now in section 3.3 – geochemical calculations.

Line 336: Does it make more sense to separate the Results and Discussion. This may help streamline and better organize the manuscript.

The results and discussion have now been split and the discussion streamlined.

Lines 343-362: This needs to be better organized, perhaps start with description of how GAB works hydrologically, then describe the ratios and their implications for the LNA. Suggest breaking this paragraph into 2 new, concise paragraphs.

The original lines 343-362 have been changed because of the restructure. Some of the original text is now in section 5.1, line 449, but the organisation has been changed considerably.

Lines 375-376: Be assertive here. This does suggest...rather than may suggest.

We have changed to ‘This suggests...’ (line 520).

Lines 376-378: Needs clarification.

This has been split up because of the split in results and discussion. Section 4.2 details the ranges of the isotopic values and that they lie on an evaporation line whereas lines 443-448 now explain the connection between evaporated surface water and shallow groundwater.

Lines 380-381: Please provide sources of F⁻ and how that relates to the point you are making.

The source of F⁻ is the weathering of silicate minerals in the region (noted now at lines 460-461). This relates to our overall message because the covariation of Na and F (from the weathering of silicate minerals) show the same mixing trend in the alluvium between shallow groundwater and GAB samples that other parameters do (such as Na-HCO₃).

Lines 416-431: Please clarify. Most 3H values are close to background or are dead with respect to 3H. This complicates your interpretation, but you seem to pull it back in focus with the figure. Suggest picking specific sites and describe what the data is telling you.

Please refer to our previous comments above regarding the activity of 3H in the southern hemisphere.

Lines 432-434: Prevalence of 3H?? Again, the 3H values are almost all very low or 3H dead. Consider rewording and clarifying this statement. Also, are there recent 3H values for precipitation?

“Prevalence of 3H” has been changed to “the activities of 3H above the detection limit” (line 492) so as to be more precise with our wording. Please refer to our previous comment regarding 3H activities in Aus and values for precipitation.

Line 434: 3H and 14C and not entirely consistent are they? This needs clarification. Why are they inconsistent?

Figure 5 (now Figure 6 in revised manuscript) illustrates why the 3H and 14C in the groundwater are inconsistent. Normally we would expect to see a very sharp decline in 3H activities as 14C (pmc) decreases. However, we see a mixing trend that shows that some groundwater samples still have measurable 3H despite having a 14C content of less than 50 pmc. We have clarified this in text at lines 492-503, better explaining the figure.

Lines 432-445: Please clarify and provide a more concise discussion on mixing effects.

This section (now lines 492-503) has been clarified with respect to the 3H (see above comment). It has also been shortened as a result of the streamlining of tangential information in the manuscript (mention of calcrete removed).

Line 458: What about 36Cl/Cl of modern precipitation? High 36Cl/Cl can be indicative of mixing of bomb-pulse with recent (low 36Cl/Cl) recharge. But, low 36Cl/Cl can also imply very old groundwater (your case). Can you clarify this uncertainty with modern precipitation?

Please refer to our comments above concerning 36Cl/Cl values of modern precipitation. In regards to lingering 36Cl/Cl derived from the bomb pulse: Cl has a very low residence time in the atmosphere (2 days) so most of the bomb derived 36Cl would be gone.

Line 487 (Figure 7): Please cite Phillips (2000) Chapter 10.

This has been cited (line 535) and added to the reference list.

Lines 510-523: Please provide the equation used to calculate the mixing proportions. Can you also provide estimates of uncertainty in these calculations? Are there other solutes or solute ratios (Cl/Br for example) which may be more suitable for these calculations? It's not clear how this was estimated.

Mixing proportions were calculated in NetPath and were treated as a mixing problem. We have added a 'geochemical calculations' section to the methods that explains this (section 3.3). Please refer to our comment above regarding mixing proportions. Additionally, we believe that the Cl ion was the most appropriate because of its conservative nature (line 556) and in our study area the end member water sources have distinct Cl signatures that facilitate its use as a tracer.

Line 535: Can you provide plots showing the spatial map of appropriate chemical concentrations?

Figure 2 has now been adapted as below. We show a spatial map of the 36Cl data, which is crucial for understanding the discussion regarding GAB discharge into the alluvium at lines 433-439.

