

## Reviewer 1 comments (response in red)

### General Comments

This is an interesting study that helps inform our understanding of recharge, groundwater flow, and spring origins. However, it needs attention before it is suitable for publication.

Firstly, the paper needs better focus. Some sections (e.g. the discussion of geochemical processes) do not seem to inform the overall story and are a distraction. The description of the study area is long and some of the information does not seem to be relevant. There are also descriptive sections in the discussion and a tendency to repeat information. A more succinct tightly-written paper would be easier to follow and have more impact.

More importantly, in several places the discussion of processes is not convincing (inc. the geochemical processes, groundwater residence times, and lapse rates). The explanations tend to be long and are sometimes not consistent or are overly speculative. The central point that the springs are recharged at altitude and the water follow different flowpaths appears reasonable, but this sometimes is lost.

Finally, the Conclusions are mainly parochial . Some idea of the broader significance of this study or comparisons with similar environments would give the paper a broader appeal.

I hope that these comments are useful.

We thank for the reviewer for the thoughtful feedback and suggestions for making the manuscript more concise. Most of the comments can be addressed during manuscript revisions. Detailed responses follow below. We apologize for the inconsistent verb tense throughout. Some changes have already been incorporated into the manuscript, though the revision has not been invited by HESS.

### Specific Comments

#### *Abstract*

The abstract gives a good idea of the major conclusions of the paper. However, it needs more focus on the results and conclusions rather than the aims. You should report a

few key values in the Abstract as qualitative descriptors (“stable”, “higher” etc) do not convey much specific information.

The review makes good suggestions we can address in a revised manuscript.

### *Introduction*

The introduction is clear and well structured. The first paragraph (lines 40-55) would benefit from a few extra details if these are available, specifically:

- Are the impacts on groundwater resources quantified? **Groundwater resources in this region are relatively poorly quantified. There are substantial knowledge gaps with respect to hydrogeology in the region and our paper attempts to address a subset of these knowledge gaps.**
- Likewise are there estimates for how much the recharge may decline? **No, this is difficult without 1) some baseline assessment on how much recharge occurs historically in this region, 2) some assessment on the sources of recharge (provided in our paper), and 3) some information on how regional climate change will impact the source of recharge (e.g., changes in rainfall, changes in high-elevation snowfall, loss of alpine glacial cover). These data are lacking.**

This would help with understanding the context of this study.

Lines 57-65. Provide some references for this material. **References added.**

Lines 73-79. This is only true if there is no other mechanism for exporting Cl. In some saline lakes Cl is lost via salt deposits being eroded by the wind (deflation). Is that the case here? If the lakes do recharge the local groundwater system, I would expect there to be some evidence of that (shallow high salinity groundwater around the lakes) – any evidence of that? **The reviewer's point is noted and we can add wind erosion as a potential mechanism to remove salts from the salar. The studies that we cite in this introductory section have not come to that conclusion though. With respect to high-salinity shallow groundwater in the salar, wells are sparse in this area and we do not have access to wells used by mining companies. To the best of our knowledge there are no biological surveys indicating the presence of basin fringe halophytic plants that can be used to mark the spatial extent of shallow saline groundwater. We will be sure to discuss the salinity in the wells we did sample near the salar in this study.**

### *Study Area*

This is comprehensive, but in places the descriptions are lengthy.

- There is some repetition with the introduction (eg lines 119-129). **The information provided in this section is specific to our study area and highlights the importance of La Bedoya spring and the intensity of agricultural and mining activities in this region. We condensed the material to these main points.**
- Some of the details of the geological history (lines 137-148) seem superfluous. **There are only 2 sentences that reference the geologic history, but we can trim slightly.**
- The climate description (Section 2.3) could also be more succinct – the important details are a bit lost in the narrative. **We can cut the description of recent trends which are not particularly relevant to this study and condense that idea in the introduction motivation.**
- Again with Section 2.4, how much of this detail is really necessary

**We can focus on describing the specific low and high permeability geologic units and the faults located in the region. We can describe the salar but save the geochemical description of the lacustrine deposits for the discussion when they are most relevant to the reader.**

The lengthy descriptions mean that it is not always clear what the important points are. Try to give more focus to what is important for the study rather than trying to cover everything. **We believe specific comments related to this theme are addressed below.**

The order is also not intuitive – Sections 2.2 and 2.4 both deal in part with the Geology and Geomorphology. It would be better to merge these two (or at least make them sequential) and to present the material in a large- to smaller scale order. So the descriptions of the large-scale geology (lines 200-207) would be better presented before the description of the basin (Section 2.2).

**Sections 2.2 and 2.4 can be reduced in length and reorganized as suggested in a revised manuscript.**

Figures 2 & 3 can be merged – they are maps of the same area. That way it would be easier to see the relationships between the samples and the faults. Also, it is not currently clear from Fig. 2 what is sampled (springs, surface water etc) without referring to the Table – make the symbol for each different.

We can make these suggested figure changes in a revised manuscript.

### *Methods*

Here also there are some diversions (eg Lines 261-265) that are distracting and do not belong in the methods. If this information is important, then it belongs in the study area section.

The additional descriptions of sampling locations were moved to the study area section.

Lines 270-272. Really it is only SpC that is measured (This is not correct. It is only the electrical conductivity that is measured. Specific conductivity and TDS are calculated from the electrical conductivity), the TDS is just calculated from the SpC using an assumed conversion. If you have a full suite of ions (which it looks like you do), you can calculate TDS as the sum of the ions. If you stick with the TDS estimated from SpC, you need to note that and provide the conversion. We can provide the conversion factor.

Section 3.2. A few more details on the Tritium procedure (enrichment and equipment) needed

The tritium analyses were not performed in house, but we will add additional details from the analytical lab.

Section 3.3. Need to report precision for the major ions

This was added.

### *Results*

As explained below, this section loses focus in places and it is not always clear what the important points are. You need to be clearer as to what information is important for this study and concentrate on that. The figures need improvements -duplicating the figures just to show the two Salinas samples is excessive. It is clear that these are evaporated surface waters and you do not say much more about them than that. In that case just omit them from the figures and note that in the caption.,

H isotope ratios should have no decimal places to be consistent with the quoted precision (O is fine with one decimal place)

We can make these suggested changes in a revised manuscript.

Lines 314-317. Report the low 3H activities as bd without the +/- (which have no meaning for 3H below the lower detection limit). If the precision is 0.1 TU (Section 3.2) then the +/- of 0.04 for BED is overoptimistic. Given that this is very close to the lower detection limit, how sure are you that this is real. Given that you sampled the springs at their surface outlets, could it just be a small amount of modern water (eg recent rainfall) mixed in with the spring water?

We state in the manuscript that water samples at or below the detection limit are tritium dead. However, we can make a note stating that the samples had 3H values that were below the detection limit and we can remove the lab-reported numeric values. In any case, we think that it is sufficient to call these samples tritium dead.

Figure 4. The two diagrams are a little confusing as they present the same data. If you feel that you need 4b to show the detail, you could make 4a in inset with just the Salinas surface water on it. Also need to reference the LMWL in the caption.

We can make Fig 4a an inset in Fig 4b and reference the LMWL in the caption.

Figure 5. Again, you do not need both figures – you can just use Fig. 5b and explain that you have omitted the Salinas waters. It would be useful to have Fig. 4 & 5 as a single figure as it would show the relevant stable isotope data in one place.

We can take the reviewer's suggestion, remove Fig 5a and combine current panels 4b and 5b into one figure.

### Section 4.3

This section lacks focus. What is important here (the processes or the differences in chemistry). The discussion of the processes using the Gibbs Diagram is not very convincing and could be done better. however, consider whether that is important. If the important point is that the waters have different geochemistry and so follow different flow paths, then just show that (the Piper and/or a couple of bivariate plots like Fig. 10, and a brief description would suffice). The discussion of processes is not very clear and may not be necessary.

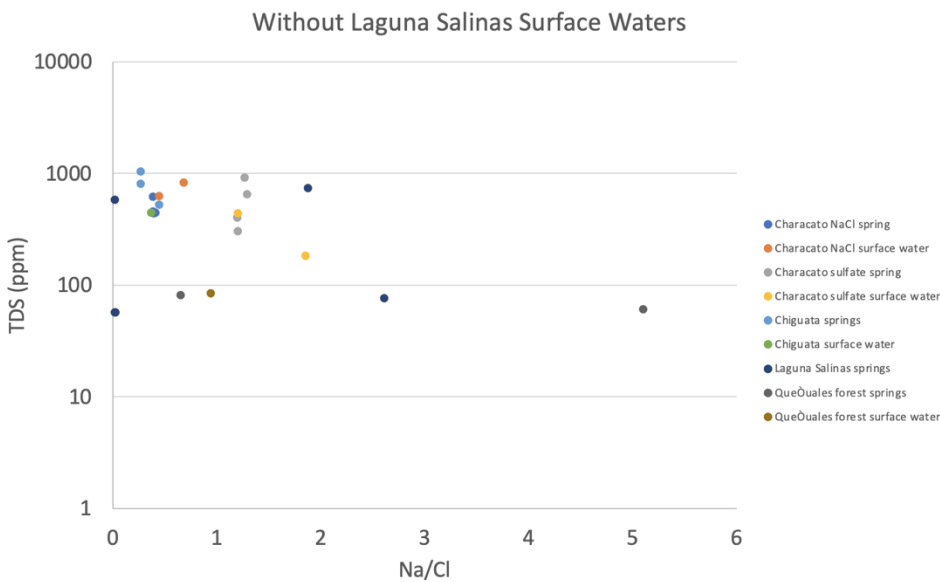
The Gibbs Diagram in particular is not very informative in determining processes and you have the data to do that more rigorously. Waters dominated by evapotranspiration have Cl/Br and Na/Cl ratios close to those of rainfall (which you can probably estimate). Extensive rock weathering produces high cation/Cl ratios while halite dissolution produces very high Cl/Br ratios. A few bivariate plots (eg Na/Cl vs. TDS and Cl/Br vs. TDS) would show that much better than the approach that you are currently using. There is extensive literature on this (eg numerous groundwater papers by Mike Edmunds).

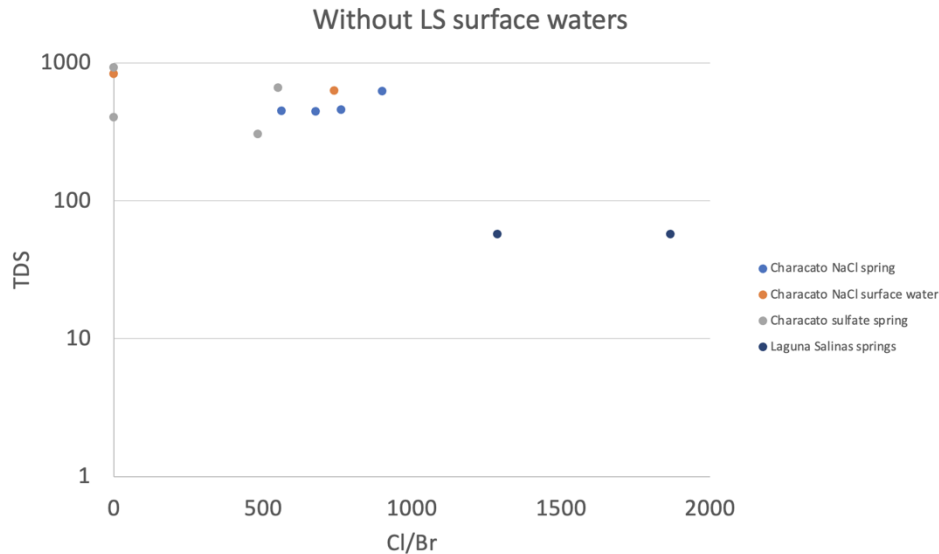
The Gibbs diagrams were used to show that flowpath groups fall within the expected processes within the diagram (eg. Salar waters within evaporation and bedrock springs within rock weathering). We also reference the paper by Marandi and Shand (2018) which uses an example from Edmunds et al. (1987) to show that groundwaters which plot in the evaporative enrichment region of the Gibbs diagram may not have experienced dissolution of evaporites. In the case of Edmunds et al. (1987), the groundwaters evolved toward this Na-Cl type water due, in part, to mixing with saline connate groundwater. Thus, an argument can be made against the Gibbs diagram that false evaporation processes can arise in geologic settings where marine deposits and saline groundwater are present. In our study area there are no ancient marine deposits that would conflate our interpretation of the salar waters as originating from evaporative processes. The Gibbs diagram clearly shows that the low-elevation spring samples which appear to be receiving some mixture of high-elevation high-salinity recharge fall along a proposed mixing line with this subspace. This does not appear as clearly in the plots suggested by reviewers (provided below). The Cl/Br plots are also problematic because few samples had detectable levels of Br and so this data set is much smaller than that shown in the Na/Cl and Gibbs diagram.

In addition, Marandi and Shand (2018) state in their critique of Gibbs Diagrams that, "It remains possible that recharging waters cross the water table with a chemical composition plotting in the upper right corner of the Gibbs Diagram, perhaps particularly in regions where land uses have caused salinization of soils or other artificial impacts to near surface mineralogy." This is the exact condition which we propose is happening in the connection between saline recharge from the salar and the Chiguata springs. Saline recharge would plot in the evaporative enrichment field where the Laguna Salinas surface waters plot. This saline recharge then mixes with non-saline groundwater flowpaths in the mountain block that were recharged by high-elevation snow and/or rain. Thus, Chiguata springs plot between the fields for evaporative enrichment and precipitation dominance. In comparison, springs such as the Quenales Forest springs and Pichu Pichu springs, which are supported by high-elevation recharge from snowmelt and/or rain, plot near the precipitation dominance

field for anions and rock-weathering for cations. Their placement in these fields make physical sense.

We will restructure and streamline this section to make these points clear. We are not convinced that adding other bivariate plots will necessarily change this story. For example, we plotted Cl/Br mass ratios and these essentially show the same result. Saline recharge has high Cl/Br and relatively low TDS because high-elevation springs we sampled flow into the salar and the salar refills during the rainy season with dilute runoff from surrounding mountains. As this saline recharge mixes with other non-saline groundwater flowpaths, the Cl/Br ratios tend to decrease while the TDS increases due to rock-weathering reactions. This interpretation is not different from that inferred from the Gibbs Diagram.





Lines 386-394. I presume that this also shows up in the other parameters? I'm not sure that you have enough data to do a PCA or cluster analysis but you should make the point with the other parameters.

We do not think it's appropriate to use a cluster analysis on this dataset. Figures 8 and 10 supports this interpretation as well. We can edit the discussion to summarize all evidence of mixing multiple groundwater flow paths in the discussion.

### Discussion

The Discussion covers a range of topics but there are a number of potential inconsistencies and unclear explanations. This is the most important part of the paper, so more clarity would help. I also suggest that you add a couple of sentences to the start of the Discussion as a guide to what you will be dealing with.

This is a good suggestion that we plan to incorporate.

Sections 5.1.1 & 2.

The discussion regarding the lapse rate (lines 415-434) needs to be clearer. While it is true that springs can have stable isotope ratios that vary with altitude that is mainly the case where they are recharged close to where they discharge. In the case of your springs, you make the case (Section 5.1.2) that they are recharged at high altitudes. That interpretation is reasonable. However, the way that this discussion is presented is to set up the idea that the springs should vary with altitude (line 416) and then point



out that that is not the case and then interpret the data in terms of recharge altitude in Section 5.1.2.

We can edit the framing to make it easier on the reader.

Much of Section 5.1.1 (the correlation with altitude etc) contains observations that should be part of Section 4 – some of it is in there already and it is just repeated here.

We do not think any new results are presented in section 5.1.1 other than 1 sentence reminding the reader that the isotopic composition of rain at the lower elevation matches the spring water at the higher elevation. We can cut that. The rest of this section provides discussion and context for the observations reported in Section 4.

The magnitude of the lapse rate. If the springs are recharged at higher altitudes then you can't use them to estimate the lapse rate. In that case, your lapse rate should be based on the surface water samples.

We agree with the reviewer's point that springs recharged at higher elevations will be more isotopically depleted than precipitation at the same elevation as the spring emergence unless the flowpaths are very short or storage is small. The curious thing in this case is that the springs at a HIGHER elevation are isotopically similar to precipitation at a LOWER elevation. Since we do not claim that lower elevation rain is recharging higher elevation springs (against gravity), we can conclude that precipitation at the higher elevation spring recharge area is similar to the lower elevation precipitation we observed.

This led us to consider the inferred precipitation isotope lapse rate in this section of the mountain, ~0 permil per 1.4 km. If the precipitation at the elevation of the Quenuales forest were much more depleted than Characato elevation, there is no way to explain the isotopic composition of the mid-elevation Quenuales forest springs. This is probably an unusual situation that confused the reviewer. Also note that we are discussing lapse rates across 1.4 km of elevation change in one section of the mountain, not following dominant wind patterns. When considering the entire mountain, the highest elevation snow samples are much more depleted than the lower elevation. We will include the lapse rate calculated over the entire elevation we have data for in the revision and highlight the anomaly between the Characato and Quenuales elevations within that larger gradient. We also have additional data from the 2020 rainy season that we present in one of the following comments. More importantly, we will edit this section to make the main focus on the recharge elevations inferred from the isotopic data and not an analysis of the isotopic lapse rate itself.

However, those data may not be suitable, specifically:

- Rainfall sampling is referenced to an unpublished study. It is not clear how many samples this represents and the duration of the rainfall record. Given the likely variability of rainfall isotope values, you ideally would have a multi-year weighted average value, but is this the case?

This is true. Precipitation collection was initiated for this project and included daily rain collections during the 2019 rainy season (effectively 1 year because there is only trace rain in other months). In a revision, we can now also include additional data generated after this manuscript submission that shows 2020 precipitation collected at both Characato and Chiguata were isotopically similar to each other, but also that they were less negative than the 2019 precipitation collected at Characato.

Characato 2019 (-9.12,-63.51); Chiguata 2020 (-6.3,-38.1) ; Characato 2020 (-5.5, -31)

- Snow is probably mainly winter precipitation and is difficult to use with samples that represent long-term averages.

We are unclear what the reviewer's point is here. The snow on Pichu Pichu occurs during the summer. Summer is the only season with measurable precipitation with rain at lower elevations and snow at higher elevations. Seasonal bias is not a complication in this system.

- The surface water samples seem to be partially fed by spring water (Section 5.2). If those springs were recharged at high altitudes then using river water to calculate lapse rate is possibly not valid as it is not capturing only rainfall at the altitude where you sample it.

We did not use the surface waters to calculate the lapse rate.

- A similar concern would apply to any rivers that flow from high to low altitudes and thus mix rainfall from a variety of altitudes

Agreed. See earlier comments.

- Even if the rivers are mainly fed by local rainfall, their stable isotope values are likely to vary seasonally (as you discuss in Section 5.2) and so again are difficult to use in this way.

Agreed. See earlier comments.

I'm not convinced that you can determine the lapse rate with the data that you have. If you are going to include this discussion, it needs to be more convincing. Otherwise you may be able to estimate it using other studies?

The main goal is to estimate the recharge elevation of the lower-elevation Characato springs. These lower elevation springs are isotopically similar to the mid-elevation Quenalaes springs. We interpret the recharge zone of the low elevation springs as the same elevation of the Quenalaes spring recharge. This is the most important observation. The discussion of the inferred precipitation isotopic lapse rate may have created a distraction for the reader.

Regardless, the biases that the reviewer mentions above are avoided in this specific case. If we consider Pichu Pichu snow, there is a substantial isotopic gradient over the region. The comments in section 5.2 are specific to the 1.4 km between Characato and the Quenuales forest on one directional slope. We agree with the reviewer that interpreting lapse rates from surface waters to determine recharge elevation would be problematic. That is not what we did. There are very few precipitation isotopic measurements in this region. In a revision, we could also include additional data generated after this manuscript submission that shows 2020 precipitation collected at both Characato and Chiguata were isotopically similar, but also that they were less negative than the 2019 precipitation. This could be used to show that the Characato precipitation is variable year to year, but that it could be more enriched compared to the Characato spring observations which do not vary and we infer are recharged from higher mid-elevations.

Characato 2019 (-9.12,-63.51); Chiguata 2020 (-6.3,-38.1) ; Characato 2020 (-5.5, -31)

Spring recharge elevation. This seems broadly correct; however, it becomes more doubtful if there are palaeowaters in the basin (lines 463-465). Discharge of paleowaters into surface water bodies also complicates the lapse rate calculations. Is there anyway to test this idea? While you do not have radiocarbon data are there examples of palaeowaters in analogous settings or examples of nearby springs for which there are better residence time calculations?

We do not have evidence of paleowater discharge from this basin, but since the region has a glacial history, we cannot absolutely rule it out. We thought it best to mention this possibility, but can remove this mention if the reviewer thinks it's unnecessary.

Section 5.3 also needs attention.

- It is not clear where the 3H activity of “young aquifers” of 2.9 TU comes from

This came from recharge-weighted well-mixed aquifer mixing models informed using time series from Albero & Panarello (1981) - this includes a compilation of time-series from South America. High-elevation snow was measured in this study and it had a tritium activity of 2.2 TU. A tritium activity of 2.9 TU is not beyond the realm of possibilities based on our measured data. In fact, we commonly find that

tritium activities decrease with decreasing elevation (with inferred increasing flowpath length, degree of mixing, and storage). Manciatì et al (2021) show a mean precipitation activity of 3.0 TU near Quito, Ecuador which is admittedly north of our study site, but not disparate from our measurement on Misti Volcano. In any case, we can restate that we think high-elevation mountain-block aquifers are “relatively young” based on our tritium activity for Misti and cite recent research in the western U.S. T

- The definition of fossil water as being >60 years is largely a northern hemisphere viewpoint as the higher  $^3\text{H}$  bomb pulse waters are still detectable in groundwater. This is not the case in the southern hemisphere where the bomb-pulse tritium has decayed back to natural levels (e.g., Morgenstern, U., Stewart, M.K., Stenger, R., 2010. Dating of streamwater using tritium in a post nuclear bomb pulse world: Continuous variation of mean transit time with streamflow. *Hydrology and Earth System Sciences*, 14, 2289-2301; Tadros, C.V., Hughes, C.E., Crawford, J., Hollins, S.E., Chisari, R., 2014. Tritium in Australian precipitation: A 50 year record. *Journal of Hydrology*, 513, 262-273).

We provide data from Moran et al. (2019) where they define “fossil water” as being greater than 60 years. Please note our parentheses in this statement. We do not call groundwater in this manuscript “fossil groundwater” because we do not necessarily think that fossil groundwater is as young as 60 years. Tritium-dead groundwaters are not unique to the southern hemisphere; this is increasingly a problem in the northern hemisphere. “Tritium dead” is admittedly an ambiguous term, as we state in the manuscript. Rather than use tritium data from Australia, which is well documented, we wanted to place our tritium data in the context of the tritium breakthrough in South America. We specifically state in the manuscript that, “The atmospheric breakthrough of  $^3\text{H}$  associated with nuclear weapons testing in the 1950s and 1960s for South America peaked at approximately 60 TU in 1965 (Albero & Panarello, 1981).”

- The residence time of 300 years seems arbitrary. Presumably it is based on mixing at the top of the aquifer but you have a fractured flow system that is likely to behave very differently.

This is based on recharge-weighted well-mixed aquifer models that were informed using time series from Albero & Panarello (1981) and some back-of-the-envelope calculations of specific discharge. The mixing models have high uncertainty because these models are sensitive to the fraction of recharge that is used to weight the tritium annually. We think that these springs likely have

residence times falling between the ranges for tritium and radiocarbon. The flow calculations are uncertain because the region is data poor and we do not have well-constrained hydraulic parameters. Therefore, we will simply state that the groundwaters are older than 60 years until additional data become available.

As you have only three tritium measurements (all of which are close to or below detection) and you do not have a good idea of the rainfall values (2.5 to 10 TU is a large range), there is little quantitative that you can say here and this section is not that informative. I'd just make a case for the water being at least a few decades old in Section 5.4.

We also provide tritium data on modern snowfall collected at high elevations on Misti Volcano and in rainfall at low elevations. Although our samples are limited in number (funding was not sufficient to do a broader scale sampling campaign), modern precipitation has tritium activities ranging from 1.4 to 2.2 TU. This range is much smaller than the other data which we provide for context only. We will state explicitly that this data was provided for context only to avoid confusion. In any case, we would expect "young aquifers" at elevations greater than the elevation of the salar to have measurable tritium. The springs emerging above the salar are tritium dead. We can conservatively interpret those observations as at least several decades old.

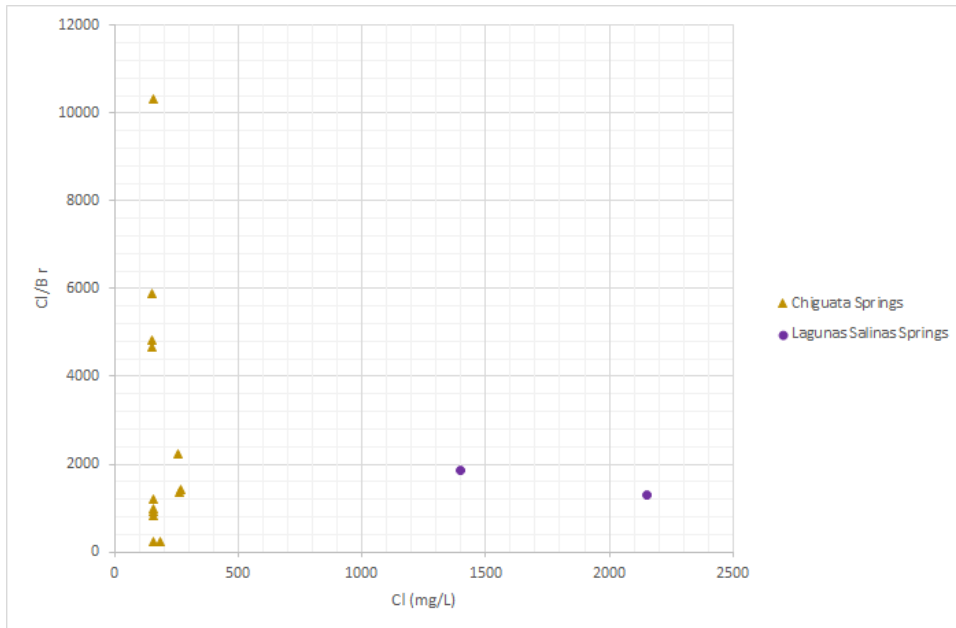
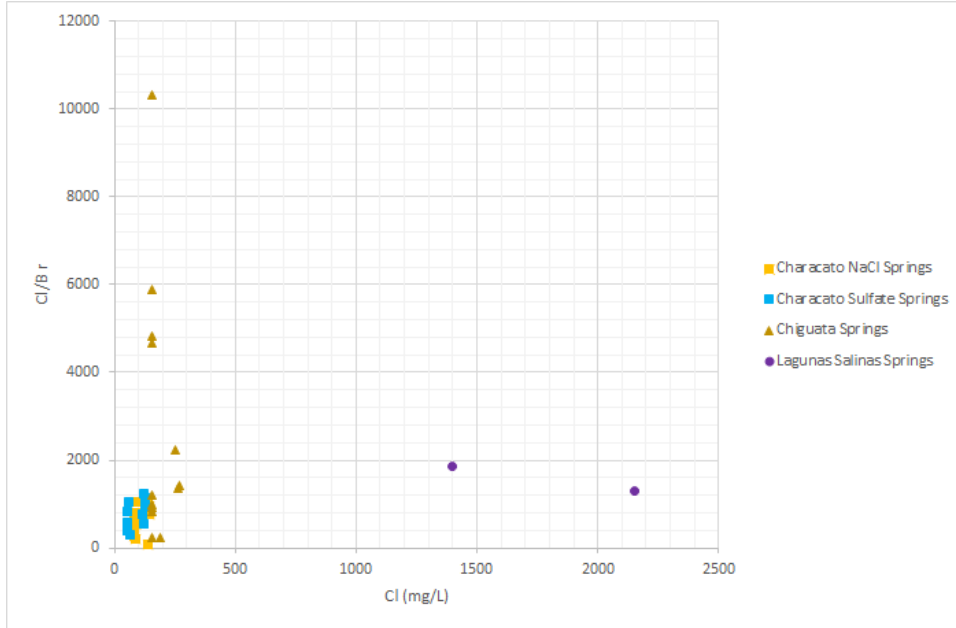
#### Section 5.4

Lines 517-520. I'm not sure that I'd expect Cl to increase along flow paths. To do so implies that Cl needs to be added from the rock matrix as evapotranspiration is a surface process. That will only occur if there is halite in the rocks (which is not that common). This concept does appear in many textbooks but the supposed process is never really explained.

Cl concentrations should only be invariable along flow paths if: 1) piston-flow processes are invoked and groundwater doesn't mix with other flowpaths, or 2) if Cl is not removed or added by dilution or biological processes. We propose that mixing occurs at this large spatial scale within a fractured mountain block. We can cite Bresciani et al. 2018 for this.

Lines 517-525. You look to have measured Br. Cl/Br ratios will readily determine whether you have halite dissolution (Cartwright, I., Weaver, T.R., Fifield, L.K., 2006. Cl/Br ratios and environmental isotopes as indicators of recharge variability and groundwater flow: An example from the southeast Murray Basin, Australia. *Chemical Geology* 231, 38-56). You may not need to speculate here.

Br was not detected in all samples, but below is a plot of samples with Br measurements from Chiguata and the springs feeding into Lagunas Salinas. The plot below is consistent with halite dissolution influence in the Chiguata springs. The Lagunas Salinas springs are sampled before they interact with the salar water.



Section 5.5.

The first paragraph (lines 569-574) repeats the previous section and is not needed. OK

Figure 11 only needs one panel as you can show all three flow paths without confusion.

We can merge the information in this figure in the revision.

### *Conclusions*

Again, there is some repetition here. Instead of repeating the specific findings, which you cover in Section 5, try to outline the general points. However, you should explain the general importance of the study or compare it with similar studies elsewhere. This will make the paper appeal to a wider readership.

We can shorten the summary information and focus on the broader impacts.

Two general points we can emphasize are:

1) Results from our study show evidence of this high elevation closed basin salar is hydrologically connected to lower elevation springs. We place this in the context of other salar studies in the Andes and conclude that not all salars are evaporation pans, but as other studies have found, some leakage through salars recharges regional groundwater.

2) This study provides perspective on the long-term stability of groundwater in this arid region. The tritium data as well as repeat discharge and stable isotope measurements supports long-flow paths with residence times of water being at least a few decades old if not longer. In the face of climatic warming these data suggest that regional groundwater may provide a stable water resource over the next few decades.

### References:

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Bresciani, E., Cranswick, R. H., Banks, E. W., Batlle-Aguilar, J., Cook, P. G., and Batelaan, O.: Using hydraulic head, chloride and electrical conductivity data to distinguish between mountain-front and mountain-block recharge to basin aquifers, *Hydrol. Earth Syst. Sci.*, 22, 1629–1648, <https://doi.org/10.5194/hess-22-1629-2018>, 2018.

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Manciati, C., Taupin, J. D., Patris, N., Leduc, C., and Casiot, C.: Diverging Water Ages Inferred From Hydrodynamics, Hydrochemical and Isotopic Tracers in a Tropical Andean Volcano-Sedimentary Confined Aquifer System, *Front. Water*, 3, 597641, <https://doi.org/10.3389/frwa.2021.597641>, 2021.

Marandi, A. and Shand, P.: Groundwater chemistry and the Gibbs Diagram, *Applied Geochemistry*, 97, 209–212, <https://doi.org/10.1016/j.apgeochem.2018.07.009>, 2018.

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