Dear Editor,

First, we thank the reviewers for their review of our manuscript. Their comments have helped us improve the quality of our manuscript. Please find attached our response to the reviewers' comments on our manuscript, 'A post-wildfire response in cave dripwater chemistry' by Nagra et al.

We have provided a detailed reply to all questions and comments raised by the three reviewers, as requested. We will provide a revised version should the decision be made to accept the manuscript.

If you have any further questions we will be happy to answer them. Thanks again!

Kind regards,

For the authors,

Gurinder Nagra

Reviewer Comments

Reviewer #1

The authors present an excellent multi-year cave monitoring study that uniquely provides insight into the response of cave dripwater isotope and geochemical compositions to POST-fire vegetation dynamics.

We thank the reviewer for these kind remarks.

Overall, the manuscript does not present a rigorous, integrated argument. Many of the interpretations come across as speculative without rigorous constraint or consideration of alternative hypotheses.

We appreciate the reviewers' comments, and agree to address them in our revised manuscript. We will go through the manuscript again to further integrate our argument and assure the manuscript reads more smoothly. And to apply more rigor to our argument, we will consider alternate hypotheses, and place more clear constraints on the claims we make. We constrain our claims by making a more quantitative comparison to differences in solute concentrations as a result of natural heterogeneity as suggested by Reviewer # 2.

Furthermore, the use of groundwater and cave dripwater monitoring in another cave used as a pre-fire baseline is not compelling nor necessary, as the authors should be focusing on the response of dripwater to the recovery of the ecosystem following the fire, not to the fire itself. That is, the authors argue that a progressive decrease elemental concentrations reflects a gradual decrease in transpiration due to tree death - however, a fire would result in a dramatic,

instantaneous decrease in transpiration not a prolonged, multi-year response. It is recommended that the authors switch their perspective from "response to the fire" to "response to the recovery from the fire".

We appreciate and acknowledge these comments. Regarding the use of data from another site as a pre-fire baseline, reviewer 2 and 3 recommend putting our site into context. We will adjust the wording in our revised text to reflect the 'response to the recovery from the fire'.

L72 – is there a way to quantify "intense wildfire" and put it into context of the range of wild fires in the region or historical time interval? This seems pertinent to understanding the magnitude of the event when thinking about its implications for interpreting past events from cave deposits.

The wildfire burnt 1200ha and was of a high enough intensity to calcine and cause fracturing of the limestone at the cave's entrance. We will add these details in the revised draft.

L193 – What is 3 years based on? Another study? A best guess?

The three years is an informed maximum water residence time based on the depth of the cave, the response time observed at a greater depth at Golgotha cave (Mahmud et al., 2016) and the isotope variability. We will make this clearer in the revised text.

L201-205 – It seems that the authors tuned parameters of a forward model to best match modeled and observed drip hydrology. What observational window/interval was used? 2005-2011? I assume that the model was tuned to observations at Yonderup, not Golgotha, correct? Is it possible that multiple parameterization schemes could result in similar comparison between observations and data?

We used the observational time window of 2005 to 2011. However, since our seepage reservoir requires a minimum 10-month residence time for year-round flow, we add monthly average rainfall data from 2003 to 2005 as a 'warm up' period for the model to avoid edge effects. But only the observation time window between 2005 and 2011 is shown. Yes, the model was tuned to observations at the studied cave site (Yonderup). We will make this clearer in the revised text.

The input parameters for the model are few (rainfall isotopic composition and the bedrock flow thresholds). And using numerous flow scenarios we show the full range of possibilities. We find that no hydro-climatic scenarios can explain our observed $\delta^{18}O$. Thus the model vs data offset suggests another factor is affecting $\delta^{18}O$. We attribute the higher $\delta^{18}O$ values in our data to increased evaporation conditions post-fire. We will re-write this section in our revised manuscript in and add this detail to make this point clearer.

L240 – Does a bivariate plot of δH vs $\delta^{18}O$ support evaporative enrichment of these dripwaters? I.e., trend off the local meteoric water line with lower slope?

Yes. Firstly, we would like to note evaporative enrichment that occurs in a high humidity environment (> 95% relative humidity) will occur with a similar slope to the local meteoric water line (LMWL), and this has been observed in cave environments (e.g. Cuthbert et al 2014). Cuthbert et al (2014) also characterized the scenarios which would result in offset from the LMWL and we have

based our interpretation on these. In our case, the bivariate plot (Fig.5) shows that the least squares regression (LSR) for cave dripwater falls within the standard error (± 0.45‰) of the slope for the LMWL (weighted LSR), but with drip water isotopic composition shifted towards higher $\delta^{18}O$ and $\delta^{2}H$ on the LMWL. This falls under a *type 1* scenario suggested by Cuthbert et al., (2014). In the *type 1* scenario drip $\delta^{18}O$ and $\delta^{2}H$ do not deviate from the LMWL but are relatively enriched. The *type 1 scenario* suggested by Cuthbert et al., supports our case for near-surface evaporation occurring in a humid near-surface environment (> 95% relative humidity).

L270 – What does "it" refer to?

Here 'it' refers to observed drip water composition. We have made the appropriate changes in text.

L329-331, 338-340 – A fire is an abrupt event that likely decimated vegetation instantaneously. How can this be reconciled with a gradual trend of increasing dilution? If the fire resulted in a shut-down of transpiration, a flushing of transpiration-concentrated poor water might be expected followed by relatively dilute concentrations until vegetation reestablished and transpiration lead began to concentration waters again. This might explain a gradual increase in solute concentrations (as seen at Site 2a and noted in L340-342), but not decrease. However, increasing solute concentrations with increased transpiration assumes that vegetative nutrient uptake is reliable relative to concentration due to transpiration.

Yes, a fire would lead to a dramatic reduction in transpiration above a site and we do agree that at Site 2a we see a vegetation recovery response post-fire. However, at site 1a a transpiration-reduction response is not necessarily reflected immediately after the fire. This can be due to a number of reasons; firstly, it simply takes time for water to reach the cave as the soil moisture deficit would have to be overcome. Second, many of the solutes would be released from the ash, so their concentrations depend on the dissolution of the elements from the ash over time. It is likely the decrease in concentrations are reflecting both a return of element concentrations to values without the influence of the tuart tree and the diminishing leaching of elements from the ash as suggested by reviewer 2. We will make the appropriate changes to shift our argument and add this explanation to our discussion in the revised draft.

L332 – Is Golgotha Cave further inland, and therefore have less aerosol Cl deposition?

Both caves are ~5 km from the coast and we have added this detail to the text. So, based on location alone, both are likely to have a similar amount of aerosol Cl deposition. However, variations in vegetation density between the coastline and the site could also influence Cl compositions to some extent. We add this to the revised text so the reader is aware.

L346 – How would an increase in surface evaporation induce PCP?

A higher rate of surface evaporation creates longer water-rock interaction times which is ideal for PCP. This is further detailed in Fairchild et al. 2000, which is referenced in the text. We will re-write this to further clarify the appropriate references in the text.

L349 – Secession of microbial and root respiration should be abrupt and coincident with the fire, not a gradual signal.

We agree and have deleted this.

L364-386 – This discussion seems highly speculative with very limited constraints on the proposed interpretation.

We argue that at Site 1a, elevated concentrations of SO_4 and K are being maintained by the abundance of above-ground biomass ash – sourced from the tuart tree. While, Cl and other solutes such as Mg, Sr and Ca at Site 1a, are reflecting dilution or a decrease in leaching of these elements post-fire, as suggested by reviewer #2 (see comment L333 – 334). Grove et al., 1986 found post-fire soils in this southwest Australia region to contain 23% more S and 16% more K than pre-fire soils, up to 1 year after the fire, as a result of the ash deposition of the overlying biomass. Given the abundance of biomass above our site (the tuart tree), we attribute the high SO_4 and K concentrations in post-fire soils from the burnt tree to be maintaining high SO_4 and K concentrations in dripwater. However, we acknowledge the reviewer's comments and will re-write this section and reduce the amount of speculation by quantifying the difference between SO_4 and K concentrations in comparison to other solutes at our sites, as suggested by reviewer #2 (see comment Lines 430 – 431).

L397-398 – How does model dripwater $\delta^{18}O$ agree with observed $\delta^{18}O$? They look to be substantially offset. Additionally, it is not clear how simulation of dripwater $\delta^{18}O$ supports the interpretation that tree death gradually reduced transpiration resulting in gradual decrease in solute concentrations.

Yes, we agree with Reviewer 1 that the observed dripwater $\delta^{18}O$ and modelled $\delta^{18}O$ are distinctly offset after the fire. This offset is underpinning our argument: that there was increased evaporation after the fire due to the lack of shading (see reply to reviewer 1, regarding L201-205). We will make this clearer in the text at L258-269. We agree that the statement in L397-398 is inconsistent with this interpretation and thank the reviewer for pointing this out. We will delete this statement in a revised manuscript.

L399 – A bit of context would be helpful – does this apply to a single tree? A stand of trees? At this (fire-affected) site? In this region?

This is specific to a full grown tuart tree that is native to the coastal dune systems of southwest Australia. Drake et al., (2011) tested the transpiration potential of seedlings and full grown trees from Yalgorup Nationl Park in different seasons to determine the pressure gradient created by their roots. We will add this detail in the revised text.

L400 – it is not clear what "this potential" refers to, nor is it clear what gradient (i.e., from where to where) is being referred to. . .. How does a hydraulic gradient maintain high Cl concentrations?

Our use of 'this potential' refers to the 'potential energy' or ability of tuart tree to extract water (by transpiration). This generates a pressure gradient between the soil and the tuart tree. The gradient transports both water and nutrients towards the tree, but not all are taken up and as a consequence are left behind in the soil solution, in particular salts like Cl. We will re-write and further clarify this in the revised manuscript.

L408-410 – It is not clear how a fire in Feb 2005 results in a sharp increase in evaporation in the Mar 2007.

As mentioned in the text, since the cumulative water balance (CWB) is positive we don't yet observe the effect of increased evaporation. Once CWB becomes negative (P < ET) we see $\delta^{18}O$ rise sharply. We believe the decrease in shading post-fire coupled with the P < ET condition was enough to drive this sharp increase in $\delta^{18}O$ and eventually exhaust the reservoir feeding the drip.

L419 – $\delta^{18}O$ reflecting more evaporation but lower solute concentrations tracking less concentration (more dilution) due to less (evap)transpiration does not make the most compelling argument.

Forward modelling of dripwater $\delta^{18}O$ compared with the observations provides firm evidence that the relatively higher observed $\delta^{18}O$ at both sites can only be explained by increased evaporation (see reply to reviewer 1, regarding L201-205). With this constraint, the simplest explanation for the declining trend in most of the solutes is a decrease in tree water use at Site 1a and removal from the surface and subsurface (leaching) post-fire (suggested by reviewer #2; see comment L333 – 334). The dominating effect of tree water-use on dripwater solute concentrations, post-fire is reported in Treble et al., (in press - accepted 8/4/16) which, using a mass-balance approach, quantifies the role transpiration from regrowth on solute concentrations at Golgotha Cave. We will rewrite this explanation in the revised text section to make it clearer for the reader.

L243-276 – It is not clear how modeling $\delta^{18}O$ contributes to this study. The results are not well integrated into the interpretations, and it is not clear why a model tuned to the drip hydrology does so poorly in accounting for observed dripwater $\delta^{18}O$. Is this meant to support the interpretation that evaporation of infiltrating water occurs somewhere between the surface and drip site, and that evaporation might also play a role in dictating solute compositions? If so, this does not come across clearly.

And

L453 – It is not clear how/why the modeled $\delta^{18}O$ represents a control (no fire) scenario.

By forward modelling $\delta^{18}O$ we aim to predict the composition of $\delta^{18}O$ in dripwater using rainfall isotopic composition and bedrock flow parameters. The model represents hydro-climatic influences only. It simulates the probable drip $\delta^{18}O$ based on rainfall $\delta^{18}O$ as an input and parameters representing the physical hydrology, as outlined in the m/s. It therefore acts as a control scenario in which only hydro-climatic influences are modifying drip $\delta^{18}O$. Hence, the fact that our drip $\delta^{18}O$ are higher than that simulated, supports our argument that a post-fire increase in surface/near-surface evaporation is driving drip $\delta^{18}O$. See reply to Reviewer 1, L201-205. For further clarity, we will re-edit the modelling text with these comments in mind.

L436 – use of groundwater and nearby cave dripwater as pre-fire conditions is not compelling, nor necessary. The argument that the recovery of an ecosystem after a disturbance is potentially reflected in cave dripwater is compelling on its own.

Reviewer #2 suggests it is necessary to compare with regional sites. In our revisions we will try to reconcile comments by both reviewers.

L441-444 – The case supporting these statements is not compelling. L446-447 – How has the vegetation forcing been delineated from the climate (i.e., CWB) signal?

See reply above for L453 with regards to $\delta^{18}O$. Our interpretation that a response to post-fire conditions is the dominating response is further supported by our solute data. The fact that most of our solutes display contrasting behaviour at our two sites demonstrates that highly localised factors, not climate, are dominating. Cl is a conservative ion and hence is primarily driven by dilution/evaporation. We interpret Cl's decline post-fire at Site 1a to reflect dilution of the water store that the dead tree previously exploited, and a decrease in leaching of post-fire as suggested by reviewer #2 (see comment L333 – 334). In contrast, Cl increases at Site 2a, sympathetically with $\delta^{18}O$, consistent with an increased evaporative demand on shallow water stores driven by post-fire reduction in shading and reduced albedo. We will clarify our concluding remarks to strengthen our arguments in a revised manuscript.

L448 – Increasing K and SO4 trends are not obvious from Fig. 3, and why would the degree of leaching increase with time from fire? It might be expected that there would be a pulse of K and SO4 following the fire, then leaching would decline after the initial pulse.

At Site 1a, K and SO_4 do not decrease like other solutes due to dilution and removal from the surface and subsurface through leaching, as above. The fact that K and SO_4 appear to be unaffected suggests that their flux has increased, counteracting the dilution and source decline, observed from other solutes. K and SO_4, have been found to have higher concentrations, in post-fire soils (Grove et al., 1986), up to one-year post-fire, as result of over-lying biomass ash deposition. Thus our interpretation for the sustained, high concentrations of K and SO_4 is that ash from the burnt tuart tree has increased the K and SO_4 flux. Further the difference in dissolution rates could also explain the rate at which these solutes are leached into drip water in comparison to others, as mentioned in our reply to Reviewer #1 comment L329 – 331, 338 – 340.

At Site 2a, we see that SO_4 and K follow the concentration trend driven by the dominant forcing, evaporation, at this site (see previous comment) and do not show an ash signal. This due to the site having less biomass available to be converted to soluble ash, and thus it is a less dominant forcing. We will make sure that we quantify the differences between K and SO_4 vs. other solutes at our sites (see reviewer #2 comment L 430 – L431) and outline and possible scenarios for these differences in our revised manuscript.

L449 – This is vague and not all that helpful of a conclusion.

And

L450 – No evidence to support this was presented in this manuscript so it is a bit odd to present as a conclusion.

Thank you for pointing out the weaknesses in the conclusions. We will re-write the conclusion in a more quantitative fashion summarizing the findings of our study.

Reviewer #2

This manuscript presents a high quality cave monitoring study from 2005-2011. The authors collected a suite of analyses to better understand cave and climate processes, and ultimately how these signals are incorporated into speleothems.

Thank you for your kind comments.

Plot the raw rainfall δ^{18}O time series in Figure 3c along with the forward model dripwater δ^{18}O. It will be interesting to see how the model alters the above-ground signal.

We were very fortunate to be able to access these data for the purpose of performing the forward model calculations. These rainfall δ^{18}O data are unpublished data created for the IAEA/GNIP program by ANSTO non-coauthors who are acknowledged in the Acknowledgements. As a separate paper on these data is currently in preparation, the owners of these data, understandably have declined to have these data published here in the time series format that is being requested. However, we do have permission to provide Reviewer 2 with a version of Figure 3 that does contain these data to satisfy their query.

Why is the modelled dripwater δ^{18}O so much smoother than the dripwater data? Is the temporal resolution of the rainfall collection too low? Or is the rainfall data being smoothed too much by the model?

The rainfall data is monthly, as stated in the manuscript, so it is not the temporal resolution of the rainfall data. The model output had been smoothed to reflect a typical stalagmite sampling resolution. This smoothing has now been removed.

Lines 262-264: The slope calculations are subject to serious edge effects. For example, the modelled dripwater δ^{18}O has an inflection point early in the record in 2006. This is not observed in the dripwater data. Perhaps you could use a bootstrap to calculate the error on the slope, but given the high density of points in blue curve of Figure 3c leaving out one or two or even three points probably will not change the slope too much. But it is this inflection point early in the modelled dripwater record – that is not in the actual data that is causing the very different slopes. The trend over both look very similar from 2007-2011. Also, the slope at for site1a should be compared with the slope of the modelled δ18O over the same time period: ~2005-2007. Therefore, I'm not convinced that evaporation is driving such a large difference in the dripwater δ^{18}O.

Given the slope calculation are prone to edge effects, we will remove the slope calculations. We believe that the 1-2‰ offset between modelled δ^{18}O and observed δ^{18}O is compelling enough to show δ^{18}O is being driven by increased evaporation. This range in isotopic enrichment is consistent with isotopic enrichment due to evaporation in caves from semi-arid regions (Rutlidge et al., Markowska et al., 2014). Further the shallow depth of this cave (4 m) as well as the reduced shading and change in albedo following the fire, makes it even more prone to evaporative effects.

Plot the Mg/Ca and Sr/Ca time series. I cannot do the calculation in my head using data from Figure 3e, f, and g. Do they co-vary in time? Or is the ln(Mg/Ca) vs. ln(Sr/Ca) relationship driven by changes that are not coeval?

Yes, Mg/Ca and Sr/Ca time series co-vary in time we will add this as a figure in the revised manuscript. This coupled with the ln(Sr/Ca) vs ln(Mg/Ca) slopes (Fig. 6) and the diagnostic range (a slope± 0.88) given by Sinclair et al., (2011) support our case for PCP. We will add these details in the revised manuscript.

Mark on Figure 3 when the fire occurred

We will add this in our next draft.

Line 331: how do pre-fire Cl values compare between caves?

Unfortunately, we do not have pre-fire Cl values for the studied cave site (Yonderup).

Lines 333-334: without values from before the fire, it's hard to discern exactly what is the cave response to the fire. Could it not be that at Site 1a there was a spike of Cl, Mg, Ca, and Sr after the fire due to dissolving ash (Lines 357-358)? Then the downward trend would be the slow removal of those from the surface and sub-surface. Without data from before the fire to establish a baseline, most of the arguments about what the fire did are too speculative and unsupported.

It is possible that at Site 1a there may have been a prior peak in Cl, Mg, Ca and Sr. However, any peak would have been a rapid increase prior to monitoring as they are highly soluble and CWB was positive. Solutes would also be affected by a direct concentration effect created as the soil and vadose zone water was heated by the fire. But as suggested by the reviewer, the trends we see here, post-fire, in these solutes, could be reflecting both the slow removal of these nutrients from the surface and subsurface, and reflecting a reduction in tree water-use. We will add this other possible cause for the decline of solutes at Site 1a to our argument in the revised text.

Lines 430-431: The differences between Yonderup and Golgotha should be quantified. Listing many values in the table does not support the differences quoted in the text. There are 10 differences to calculate 2 sites at one cave, 5 sites at the other. From the population of 10 differences, one may then calculate the median, mean and standard deviation. Then it will be clear 1) how much the drip chemistry differs and 2) how much variability exists just heterogeneous environments, which means to say *not* fire-related.

We will calculate the difference suggested to further quantify our interpretation and pre-empt speculation. We think a better way to describe the natural heterogeneity is by the observed differences between sites at each cave (e.g. Site 1A vs Site 2A). As this will show how each site responds over time.

Reviewer #3

Monitoring studies such as this provide insight into karst and speleothem processes and as such are valuable data sets for the scientific community. Understanding the response of karst systems to fire is potentially a great asset for paleoclimate interpretations, as it may be a new way to track

past limitations in water availability. Application of a forward model to aid in the interpretation of the monitoring data is also a valuable aspect of this study.

We thank the reviewer for these kind comments.

However, the manuscript lacks rigor in the presentation and interpretation of the results as well as in the overall presentation. For example, the abstract provides no results that would support any of the conclusions.

We will add more details from our finding in the abstract. This includes specifics on how we utilized the forward model to demonstrate that drip $\delta^{18}O$ was 1-2 per mil higher than predicted by hydro-climatic processes supporting enhanced evaporation post-fire, further supported by the Cl data; that the distinct spatial and temporal differences between site $\delta^{18}O$ and solutes rules out that both sites could be climatically controlled. We will also detail the potential role of the death of the tree in explaining the post-fire observations at dripwater site 1a. Further we will also detail the use of ash-derived SO_4 and K that are leached in dripwater.

The application to speleothem studies discussion in the Conclusions should be its own section and come before the Conclusions. To be useful to other researchers, it would be helpful to provide exposition of the subtleties that would be involved in such applications. For example, 1) how many different proxies would be needed to delineate fire influence, given that $\delta^{18}O$ in speleothems is affected by many processes, including in-cave processes not related to climate or vegetation disturbance?; 2) How would a researcher delineate between drip sites impacted by fire but without a tree in the recharge zone for the drip, vs. climate processes, vs. in-cave processes?

We thank the reviewer for this constructive comment and will add an 'application to speleothems' section discussing the information that can be used out of our study to find a proxy for fire. We will bolster this section by discussing recently published studies from Golgotha Cave (Treble et al 2015 and Treble et al GCA – accepted) that further strengthen the interpretation presented here.

We will specifically address the suggestions that the reviewer has made above. We believe a multi-proxy approach that uses a suite of soil and bedrock sourced elements and $\delta^{18}O$ should be used as proxies to analyse a fire signal.

We recommend multivariate statistical analysis should be used to separate climate/seasonal forcing from a soil/ vegetative forcing. Here, we would expect the local soil/vegetative forcing to preserve the impact of a fire on stalagmite composition, in trace elements like S, K and P. All of which have been found to increase in abundance in post-fire soils, due to burnt biomass-ash. Further, given fire can increase discharge by reducing tree water-use, it is also worth looking at colloid associated metals, such as Al, Fe and Cu which could show immediate spikes after the timing of the fire.

We will also discuss the importance of site selection i.e. the depth of the cave, the overlying vegetation and the climate setting, as not all sites will be affected by fire. Our study helps to place constraints on the search for a paleo-fire signal in caves and also informs other monitoring studies of this nature.

Line 28. How is the analysis unique? What specifically is unique about this analysis?

AND

75. see comment on abstract re 'unique'; 'analysis' is used twice in this sentence.

The analysis is one of the first monitoring studies conducted in a post-fire regime that seeks to identify the nutrient dynamics and effects of wildfire on dripwater composition. We will make this point clearer in the revised copy.

29-31. Run-on and awkward sentence.

We will re-write this sentence.

33. This is the most significant claim of the study. Explain how the δ^{18}O, chemistry, indicate and support this claim. There is nothing in the abstract that provides any hint of what the results of the study are.

AND

35. How so? What are the results that indicate this?

AND

36. 'here we open a new avenue for speleothem science': Without answers to the above questions, this statement is not supported.

These points will be covered by the modifications suggested in the first reply to Reviewer 3.

46. 'local environmental factors' such as. . .?

Evaporation, transpiration and leaching from biomass-sourced ash. This will be inserted in this sentence for clarification in the revised copy.

48-49. Monitoring studies have also focused on controls on calcite growth, the role of CO2, respiration, and other factors and processes (Wong et al. 2011, GCA; Breecker et al. 2012, GCA)

We will add these references in the revised copy.

51. AET – define acronyms upon first use. AET is not defined until figure 3

AND

137. ANSTO: define acronyms on first use.

All acronyms will be explained upon first use in the revised manuscript.

52. Is this 'the exception' or the rule? There are many more monitoring studies in water limited regions, including those cited above

Will remove 'the exception'.

62. Wong reference – this study is not an example of vegetation loss due to fire.

We will more clearly state this study is an example of vegetation loss not vegetation loss due to fire.

74. give cave name and location

We will add the cave name (Golgotha Cave) and location 36.10 deg S, 115.05 deg E to the text and location figure of the revised manuscript.

85-6. Give length of time comprised by temp record; give geologic age of Tamala Limestone. 113. 'heterogeneous' in what way?

We will add the length of time from which we obtained the averages for the temp and rainfall records. We will give the age (Quaternary) of the Tamala limestone. Further we will also add that the soil is heterogeneous in thickness and spatial coverage above our sites.

114-5. make into two sentences

We will separate this into two sentences.

123-5. state the length of the collection interval.

The length of the collection interval varied over time between 2 – 4 times a month we will detail this in the new revised draft. Bi-monthly is the best way to describe it.

129, 132. State location of instruments. Is the ICP-AES a spectroscope or spectrometer? List it as such.

Our cation concentrations were measured using a Thermo-Fischer inductively coupled plasma-atomic emission spectrometer (ICP-AES) at the Australian Nuclear Science and Technology Organization facility in Lucas Heights. We will add this to our revised draft.

145. Was this drip size verified for the site? Why refer to an experimental study if you have both drip speed and volume/time?

When drips were fast enough to be measured we recorded the time intervals between the drips. When the drip was too slow we left a bottle and measured the volume of the discharge over a particular period of time. Thus in order to represent the data in common units we needed to use this drip volume in order to convert all our discharge data into volume data. The calculations have been provided in the supplementary info excel sheet. We will clarify this in the new draft.

151-4. What does 'this' refer to? RMC? P-AET? This is a run-on sentence and hard to follow.

Here 'this' refers to the residual mass curve (RMC). We will detail this in the revised draft.

157 what is 'FWE'? 160-161. define terms upon first use, which is much earlier than here

FWE is the acronym for AET in the Australian Water Availability Project data sheets. We have clarified in this sentence that this is the parameter that we used since there are multiple parameters available to estimate evaporation terms from this dataset.

170. How far away? Yanchep is not on the map FIG 1.

Yonderup Cave which is shown on the map is in Yanchep NP. We will make this clearer in text.

178-9. Only one year different from the study's fire. Was the burn less intense? Less destructive? How did it effect the lower and middle understory growth?

The prescribed burn at Golgotha Cave was much less intense and much more spatially heterogeneous than the fire that is reported in our study. We will make this clearer in the text and refer to the manuscript currently under review for the impact of this fire on Golgotha Cave.

Run-on sentence 188. the 'latter' what? Several different things are developed in the previous sentence.

We use the term 'latter' to refer to seepage and fracture thresholds, we will make this clearer in the text.

192-4. confusing sentence, try restating as: 'Storage time for water that enters the seepage reservoir is modeled as a Gaussian distribution. This time is set as a maximum age of 3 years to reflect the shallow depth of our cave system. The model allows for the mean and standard deviation to be specified for these functions.' Furthermore, what is this based on? It's more the nature of the flowpath through the vadose zone than it is the thickness of the vadose zone that will determine storage time. It seems that this value is adjusted later based on observations at the cave, but need to have a basis for this starting point.

We will readjust the sentence as suggested. These sites have been found to be predominantly controlled by seepage or matrix flow due to the calcarenite type lithology at our sites (Mahmud et al., 2015). Further, capillary barriers have been found to affect hydrology at these sites which delay downward movement of water. With this knowledge and the shallow depth of the cave we set a 3-year maximum limit.

198. What is 'a karst store'?

By 'karst store' we were referring to water storage in the overlying bedrock. We will make this clearer in our next section

217. Why 'soil water availability' here, and 'water availability' above?

We will change 'soil water availability' to water availability to make it more consistent.

224. 'increases' should be past tense

We will change to 'increased'.

236-9. Run-on sentence 239. Add (DFJ) for northern hemisphere readers

We will add 'DJF' after 'summer' and 'JJA' after 'winter' for northern hemisphere readers and divide the sentence with punctuation.

240-1. when first defining 'thresholds' also define it in context of 'seepage thresholds'

We will re-write our definition of thresholds by defining 'thresholds' in the context of seepage and fracture thresholds.

243. Unclear. Try rewording: 'We attempted to model dripwater $\delta^{18}O$ that matched the measured drip water values based on using the rainfall isotopic data set as our input?'

We will reword this sentence, as suggested, to make it clearer.

244. Is 30 mm rainfall? State this. Could other variables in the model cause the shutoff?

Yes, 30 mm is the P – AET threshold. Given P – AET is the only input in the model and the only thing limiting the water from entering the seepage reservoir is the threshold. Thus it is unlikely that other variable in the model would cause the shutoff.

247-251. Run-on. 253. is this the mean or the weighted mean?

It is just the mean. We will make this clearer and fix the Run-on sentence.

265. What makes a given model 'meaningful'?

First, meaningful models referred to modeled scenarios that maintain full year round drip flow to match observed drip flow. This eliminated models with a seepage threshold of 40 mm or greater. And given not all rainfall enters the seepage reservoir, some threshold must exist, so we set a minimum seepage threshold of 10 mm. Second, models that reflect the minimum residence time (10 months) to maintain full year round flow at our t our sites were chosen. Under these bedrock constraints we tested all possible scenarios from predominantly fracture flow (10 – 15 mm) to all seepage flow (10 – 1000 mm). We found that fracture dominated flow showed more variability than seepage dominated flow. However despite the variability, none of the models that we ran fit the observed $\delta^{18}O$ at both sites. It is clear, $\delta^{18}O$ at both sites is offset from modelled hydro-climatic $\delta^{18}O$ We will provide brief clarification on this in the revised manuscript.

273-4. try rewording to: 'This is similar to a type 1 scenario defined by Cuthbert...'

We will reword this sentence as suggested.

288. Interpretation, not Results, belongs in Discussion 291-6. difficult to follow, rewrite. 300. Arguments should be in Discussion, not Results.

We will move L288 and L297-301 to the Discussion.

316. For sure evaporation will affect chloride in water in the same way dilution and mixing will, not potentially.

We will change 'and potentially' to 'as well as'.

348. This figure shows no diagnostic model, even though the caption states that both sites fall within the model, the figure doesn't show it.

We will clarify this sentence by replacing 'shown by the diagnostic model' with 'as evidenced by the agreement of the ln(Sr/Ca) vs ln(Mg/Ca) slopes in our data with the diagnostic range (a correlation co-efficient ± 0.88) given by Sinclair et al., (2011)'.

358. Unclear. If 3 increases Ca in dripwater, why would dripwater not reflect the increase? Is the signal from 1 and 2 so large as to make 3 background? Or is it simply sequence of events? Run-on sentence.

Here we were aiming to put forward a possible number of scenarios as it is hard to constrain. It is likely 1 and 2 are more important than 3 which we will clarify in the revised manuscript.

389. Reference figure earlier in text when describing site and processes.

We will reference this figure earlier in the text as suggested.

450-2, and 455. This is new information, more appropriate to include it prior to Conclusions

We will move this information to the Discussion.

456-7. Further, that a fire signal may be much more subtle in a speleothem if the fire impacted drip sites without trees above them.

Thank you. We will add this point to our Discussion.

458. Growth rate not covered in text, this is more new information that is more appropriate to include prior to conclusions. See general comment above about.

We will move this information to the Discussion.

Fig. 1. Where is Yanchep National Park located? Where does the inset sit on the map of Australia?

Yonderup Cave is situated within Yanchep National Park. We will make this clearer in the figure in our revised manuscript

Fig. 2. The same data are presented in Figs. 2 and 6. Only one of these is needed. If the authors are going to employ the Sinclair graphical model, then it would be Fig. 6. However, the discussion of Fig. 6 in the text and caption claim to show that the Sinclair PCP model holds and can account for the trends, yet there is no text or addition to the figure that supports this claim. In Fig. 2, state that the values plotted are for drip water. The interpretation given that both (each) site has an independent flow path is not explained. What specifically indicates this? Different starting points? Distinct slopes?

We will replace Figure 2 with Figure 6 as it is diagnostic for PCP as suggested. We will quote the regression equations for both the normal and log-transformed data. We will clarify in the text that the difference between regressions calculated for each site (using either method) indicate independent drip paths as suggested by the reviewer. We will also add a ln Mg/Ca and ln Sr/Ca time series to make our case for PCP more clear.

Fig. 3B. It's difficult to see Site 2a measured values. Since 'Est Dis', 'Meas Dis' used, also label sites as 'Calc' for clarity.

We will make this adjustment to Fig 3.

641. concentrations appear to be quadruple, not 'double' that of Site 2a.

The concentrations are x4 for K and x2 to x3 for SO4. We will make this adjustment in the text.

643-4. differentiation of temporal trends between 1a and 2a: I disagree with trying to make a difference here, as the data do not support this. Both sites show nearly the same slope of increase and the trends are obscured by gaps in the time series.

We will delete this sentence.

Fig. 4. Explain which thresholds. Why would the 10-75 mm threshold have a lower response than the 10-15 mm model? Why not present time series for the proposed fire-sensitive ions such as SO4 and P? Show where the fire event occurred in relation to the time series.

We thank the reviewer for picking up on this. We found that one of our input rainfall isotopic data points for the date for 07/2009 was + 5.16 instead of -5.16, this was offsetting the model for 15 – 100, 10-15, 40 – 100 and 10 – 1000 mm thresholds which had the (+) instead of the (–) value while the 10 – 75mm had the correct negative value. We have also eliminated the smoothing in the model as suggested by reviewer #2. This figure is provided below. We will also add the time series of SO_4 and K along with an indicator for when the fire occurred to put the time series into context as requested.

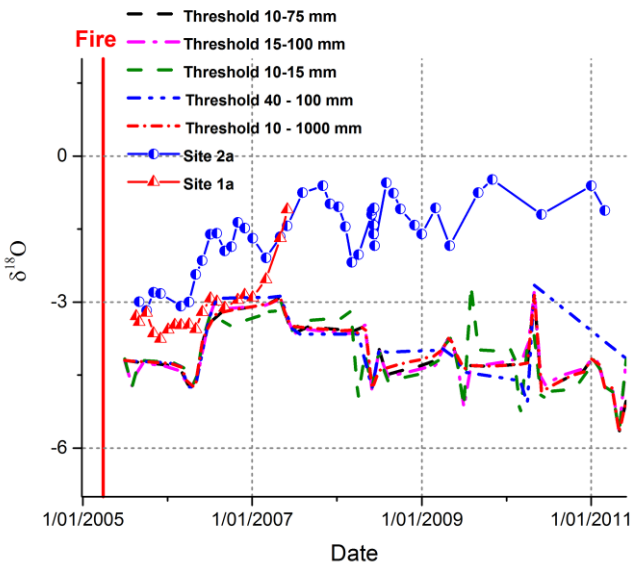

Fig. 5. Move Mean rainfall to below rainfall. Use color other than red since red is for cave drip info. Is the mean rainfall the mean or weighted mean?

538 We will move the mean rainfall to below the rainfall and use a color other than red.

539 **Fig. 6. Place both sites on same plot with different symbols, in order to help the reader directly**
540 **compare them. See comments above on Fig. 2. Presentation of time series for these element**
541 **ratios would aid in their interpretation and how the processes proposed to account for the**
542 **variation change with seasons, etc.**

543 We will add Mg/Ca and Sr/Ca time series to aid in the interpretation of processes affecting these
544 trace elements such as PCP.

545

546

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579

A post-wildfire response in cave dripwater chemistry

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Abstract

Surface disturbances above a cave have the potential to impact cave dripwater discharge, isotopic composition and solute concentrations, which may subsequently be recorded in the stalagmites forming from these dripwaters. One such disturbance is wildfire, however the effects of wildfire on cave chemistry and hydrology remains poorly understood. Using dripwater data monitored at two sites in a shallow cave, beneath a forest, in southwest Australia, we provide one of the first cave monitoring studies, conducted in a post-fire regime, which seeks to identify the effects of wildfire, and post-fire vegetation dynamics, on dripwater $\delta^{18}O$ composition and solute concentrations. We compare our post-wildfire $\delta^{18}O$ data with predicted dripwater $\delta^{18}O$ using a forward model based on measured hydro-climatic influences alone. This will help to delineate hydro-climatic and fire-related influences on $\delta^{18}O$. Further we also compare our data with both data from Golgotha Cave - which is in a similar environment but was not influenced by this particular fire - as well as regional groundwater chemistry, in an attempt to determine the extent to which wildfire affects dripwater chemistry. We find in our forested, shallow cave, $\delta^{18}O$ is higher after the fire relative to modelled $\delta^{18}O$. We attribute this to increased evaporation due to reduced albedo and canopy cover. While the solute response post-fire varied between the two drip sites: at Site 1a, which had a large tree above it but was lost in the fire, we see a response reflecting both a reduction in tree-water use and a removal of nutrients (Cl, Mg, Sr and Ca) from the surface and subsurface. Solutes such as SO_4 and K maintain a high concentration, which we believe is due to the abundance of above ground ash. At Site 2a, which was covered by lower-middle storey vegetation, we see a solute response reflecting evaporative concentration of all studied ions (Cl, Ca, Mg, Sr, SO_4 and K) similar to the trend in $\delta^{18}O$ for this drip site. We open a new avenue for speleothem science in fire-prone regions, focusing on the geochemical records of speleothems as potential paleo-fire archives.

Comment [GN1]: An additional perspective as suggested by reviewer 2 (comment about incorporating vegetation recovery to the story)

Comment [GN2]: We detail how this study is unique in response to Reviewer 3's comment L28 and L75

Comment [GN3]: We add a clear description of the role of the forward model early in the paper (and throughout) for the reader as reviewer 1's comments suggested its role needed to be made clearer.

Comment [GN4]: Detailed outline of results added in the abstract as suggested by reviewer 3 (Comment L33 to L36). Further some changes in the writing of the manuscript have been made to improve the writing (suggested by Reviewers 1 and 3)

1 Introduction

Caves are observatories, that preserve invaluable geochemical archives of past-climates; in the form of speleothems (stalagmites, stalactites and flowstones). The existing paradigm in speleothem science has largely focused on establishing paleoclimate proxies in stalagmites (e.g. McDermott et al., 2001; Treble et al., 2008; Woodhead et al., 2010). While these proxies are useful for reconstructing paleoclimates, their interpretations may hold a predisposed bias towards using these proxies as indicators of paleoclimate only.

To avoid this bias, we need to consider the sensitivity of these proxies to the effects of local environmental factors like in our case, fire. This is especially important as incorporating this perspective may not only be used to correct the climate proxy interpretation, but also yield novel information about paleo-environments. Paleo-environmental proxies are verified by conducting process-based in-cave monitoring studies. However, in-cave monitoring has predominantly focused on understanding the extent to which dripwater $\delta^{18}O$ (Lachniet, 2009), dripwater solute concentrations (Fairchild and Treble, 2009), speleothem calcite growth (Wong et al., 2011) and cave CO_2 processes (Breecker et al., 2012), are affected by climate. Further, such studies have largely been restricted to mid to high latitude climate regions where precipitation (P) is larger than actual evapotranspiration (AET), and climate is likely to be a major control on dripwater composition.

In water-limited regions, dripwater chemistry is influenced to a greater extent by environmental factors such as evaporation (E) (Pape et al., 2010; Cuthbert et al., 2014; Rutlidge et al., 2014) and transpiration (T), (Tremaine and Froelich, 2013; Treble et al., 2016). Wildfires, common in water-limited regions, are agents of change than can dramatically alter evaporation and transpiration rates by destroying vegetation. The potential impacts of vegetation loss from fire are both short-term and long-term. The short-term

Comment [G5]: Addition of an example of a local environmental factor. Response to reviewer 3 (comment L46)

Comment [GN6]: References added in response to reviewer 3 comment L 48 to 49)

Comment [GN7]: Added more references to demonstrate geographical evidence and broad range of literature that has been read. (Reviewer 3 comment L48-49)

Comment [GN8]: All acronyms have been stated on first use. In response to reviewer 3's comments

impacts could include: (1) an increase in evaporation rates due to changes in albedo and/or lack of shading (Silberstein et al., 2013); (2) a reduction in transpiration from reduced tree water use; (3) a reduction in soil microbial and root CO_2 production (Coleborn et al., 2016); (4) a decrease in cave CO_2 due to the destruction of vegetation (Wong et al., 2010), which can influence in-cave prior calcite precipitation (PCP); (5) the addition of plant ash to the soil profile, increasing concentrations of Ca, K, Mg, and S (Grove et al., 1986; Yusiharni and Gilkes, 2012a); and (6) altered infiltration patterns (González-Pelayo et al., 2010). While long-term impacts include: (1) the spatial redistribution of nutrients (Abbott and Burrows, 2003); (2) regrowth impacts on water balance and nutrient flux (Treble et al., 2016); and (3) a reduction in total soil CO_2 due to the destruction of CO_2 sequestering microbial communities and plant roots, both significant sources of soil CO_2 (Coleborn et al., 2016). Despite the fact that wildfires regularly affect water-limited regions, their impacts on $\delta^{18}O$ and solute concentrations in cave dripwater have not been reported.

We analyse the composition of cave dripwater over five years (August 2005 - March 2011) of cave monitoring in Yonderup Cave, a shallow cave system, in southwest Australia. Our monitoring followed an intense wildfire in February 2005 that burnt 1200 ha of Yanchep National Park. The fire was hot enough to calcine and fracture the limestone observed at the caves entrance (Supplementary Fig. 1). We compare our monitoring data to the regional groundwater geochemistry and published monitoring data (Treble et al. 2015) from Golgotha Cave in southwest Australia (lat. 36.10° S, long. 115.05° E). Our analysis provides one of the first analyses of the response of dripwater $\delta^{18}O$ and solute concentrations to post-wildfire conditions in shallow caves located in the tree rooting zone.

Comment [pT9]: Added a more detailed description of the study (suggested by reviewers 3 L63).

Comment [GN10]: Cave name added in response to R3 comment L74

Comment [GN11]: A clearer description of the intensity of the wildfire has been added as suggested by Reviewer 1 (Comment L 72)

2 Site description

Our study was conducted in Yonderup Cave in Yanchep National Park (lat 31.5475^{o} S, long 115.6908^{o} E), 20 km north of Perth, southwestern Australia (Fig. 1A). This region has a Mediterranean climate characterised by dry hot summers and cold wet winters with a 25-year (1990 to 2015) average annual surface temperature of 15.1^{o}C and rainfall of 664 mm with 85% of rainfall falling between May and October. Yonderup Cave is located in the young Quaternary Tamala Limestone Formation, a porous, partially lithified calcareous coastal dune sand. This karst process is said to be "syngenetic" with karstification occurring simultaneously with lithification of the host rock (Jennings, 1964; Fairchild and Baker, 2012).

Comment [G12]: Time of temp record given (R3 comment L85)

Yonderup Cave is situated in a tuart forest *(Eucalyptus gomphocephala)*, with mature tuart trees 30 m high, and an understory of shrubs and trees standing 5-10 m high, Sheoak trees (*Allocasuarina fraseriana)* approx. 5-15 m high, and Balga trees (*Banksia attenuata*, *Banksia menziesii*, *Banksia grandis*, *Allocasuarina fraseriana*, *Xanthorrhoea pressii*). Tree roots are exposed in the cave, both in the roof (fine roots), and cave floor (thick tap roots). In February 2005, the area above the cave was burnt in an intense wildfire (Fig. 1B, Department of Parks and Wildlife, pers. comm., 2015), substantially modifying vegetation above the cave including the death of mature trees and complete removal of canopy and understorey.

Over the period of August 2005 to March 2011, two drip sites in Yonderup Cave (Site 1a and Site 2a), were monitored for their chemical and hydrological variations. These two sites are 22.8 m apart (Site 2a east of Site 1a, ~ 1 m slope towards the East), located at similar depths below the surface (~ 4 m) within the same chamber (~ 7 m height) and partially separated by a large boulder fall-in. We use an existing cave survey to determine the location of each cave drip site relative to the ground surface (Fig. 1C). A soil depth survey was conducted within 5 m of each site (Supp. Table 3), along with visual vegetation/ground surface observations

699 post-fire. Soil depths were measured every meter with a dynamic soil penetrometer in north,
700 south, east and west directions and averaged soil depth above each site were calculated.

701 Site 1a, 30 m from the cave entrance, has a drip source within a large cluster of soda-straw
702 stalactites known as the 'Wheatfield' (Supp. Figure 2B). This circular feature is
703 approximately 1 m across and as it appears in an otherwise very sparsely decorated part of the
704 ceiling, suggesting that it represents a focused flow path into the cave. The land surface
705 above this site is flat with 70% coverage by shallow soil (average 124 mm thickness) and the
706 remaining surface is exposed bedrock (approx. 30%). A tuart tree, located directly above Site
707 1a, burnt and collapsed during the 2005 wildfire which resulted in the entire removal of
708 canopy cover above Site 1a. No other trees are close enough to provide shade on the surface
709 above Site 1a.

710 In contrast, Site 2a situated 50 m from the cave entrance is in a highly decorated part of the
711 cave known as the 'Cathedral' characterised by large icicle shaped stalactites. Above Site 2a,
712 the soil cover is thicker (200 mm) and more homogenous with no bedrock exposure, and no
713 trees directly above, however there is a partial canopy cover from adjacent trees ~15 m away.

714

715 ## 3 Data collection

716 Cave dripwater was collected from 1L high-density polyethylene (HDPE) collection vessels
717 at the two sites Site 1a and Site 2a between August 2005 and March 2011 (~5.5 years) at
718 approximately bi-monthly intervals. The water was separated into three aliquots: two aliquots
719 were filtered with 0.45 µm mixed-cellulose filters into two 50 ml polypropylene bottles for
720 major and minor ion determination; the third was stored with zero-headspace in a 12 ml
721 amber glass bottle for stable isotopes. All aliquots were refrigerated below 5°C until analysis.
722 Anion concentrations (Cl and SO_4) were determined using a Dionex DX-600 ion

Comment [GN13]: 'heterogenous' removed in response to (R3 comment L113). We already demonstrate that the soil is heterogeneous by explaining that only 70% of the surface is covered by soil.

Comment [GN14]: Length of collection interval stated. (R3 comment L123-5)

chromatograph with self-regenerating suppressor on one aliquot. The second aliquot was acidified to 2% HNO_3 in the collection bottle and used for cation concentrations (Ca, K, Mg, Na, Si and Sr) using a Thermo Fisher inductively coupled plasma-atomic emission spectrometer (ICP-AES) ICAP7600 at the Australian Nuclear Science and Technology Organisation (ANSTO) facility. An internal standard with concentrations approximating the cave waters was included in each cation batch to check for between-run reproducibility.

Dripwaters collected between August 2005 and May 2008 were analysed for $\delta^{18}O$ using Isotope-Ratio Mass Spectrometry (IRMS) at the Australian National University (see Treble et al., 2013 for method). The remaining dripwaters were analysed for $\delta^{18}O$ and $\delta^{2}H$ at ANSTO using the Cavity Ring Down Spectroscopy (CRDS) method. Additionally, as there was sufficient remaining water in the stored aliquots analysed by IRMS for Site 2a, these were also re-analysed using CRDS to obtain a complete time series for $\delta^{2}H$. After Jan 2007 dripwater volume at Site 1a became insufficient to collect all three aliquots. Collections of aliquots were prioritised in the following order: 1) stable isotopes; 2) cations and; 3) anions.

At each cave visit for dripwater sampling, drip rates were manually recorded using a stopwatch and the level of water accumulated in the bottles was recorded to the nearest 100 ml. Weekly discharge was estimated using a drip volume of 0.2 ml per drip (Collister and Mattey, 2008). When timing drip intervals became impractical, only the bottle level was recorded. Thus in order to represent the data in common units we needed to use the Collister and Mattey, 2008 drip volume in order to convert all our discharge data into volume data. We use both sets of measurements from the overlapping period to convert volume to discharge for when direct measurements for drip interval using the stop watch were lacking. The calculations are provided in the supplementary info as excel sheets.

Comment [GN15]: Spectrometer defined response to reviewer 3 L 129

Comment [G16]: ANSTO acronym defined (reviewer 3, Comment L145)

Comment [GN17]: Location of instrumental analysis has been specified in response to Reviewer 3 comment L129,132

Comment [GN18]: Drip volume use justified in response to R3 comment L145

To distinguish dry and wet periods we applied a residual mass curve (RMC), (Hurst, 1951) to monthly P - AET data. The RMC is the cumulative sum of the monthly anomaly calculated from the 22 year mean and used to generate a time series of cumulative potential water surplus or deficit starting from Jan 2000, highlighting trends in above average or below average P – AET, we refer to this calculation as cumulative water balance (CWB) throughout the rest of this paper.

Unpublished monthly $\delta^{18}O$ and $\delta^{2}H$ rainfall data (2005 – 2011) from Perth were obtained from ANSTO. We used modelled regional precipitation (P) and actual evapotranspiration (AET/ F_{WE} is the sum of soil evaporation and transpiration by vegetation based on Priestly-Taylor equations) from the Australian Water Availability Project (AWAP) (Raupach et al., 2009; Raupach et al., 2011) with monthly parameters, to determine P – AET. AWAP precipitation (P), actual evapotranspiration (AET/ F_{WE}) and rainfall $\delta^{18}O$ data were then used as input to the forward model (detailed in the next section) to predict cave dripwater $\delta^{18}O$ composition under various hydro-climatic scenarios. Predictions are based solely on P - AET data which are then compared to the dripwater observations.

Monthly rainfall $\delta^{18}O$ and $\delta^{2}H$ compositions were amount weighted and fitted with a linear regression (Hughes and Crawford, 2012) and compared to the long-term groundwater mean obtained from Turner and Thorpe (2001) and the cave dripwater to determine whether evaporation has affected cave dripwater isotopic composition (see section 4).

Post-fire solute and $\delta^{18}O$ data from Yonderup Cave dripwater are also compared to other relevant published data. These include, long term Perth rainfall $\delta^{18}O$ from Turner and Thorpe (2001), and local Yanchep rainfall solute data from Hingston and Gailitis, (1976), and published dripwater data from Golgotha Cave, located 300 km south of Yanchep. Golgotha Cave has been monitored since 2005 (Treble et al. 2013; 2015; 2016; Mahmud et al 2015).

Comment [GN19]: Defined. In response to Reviewer 3 comment L151-4

Comment [GN20]: FWE defined, in response to reviewer 3 comment L157

Comment [GN21]: FWE, defined in response to reviewer 3 comment L157

The climate at Golgotha Cave is also Mediterranean, but receives annual mean rainfall of approx. 795 mm, which is 23% higher than Yanchep. Both caves are located within the Tamala Limestone Formation, however the caves vary in depths: Golgotha Cave is significantly deeper than Yonderup approx. 30 - 35 m. Golgotha Cave is covered by a more extensive forest of mixed marri/karri (*Eucalyptus calophylla / Eucalyptus diversicolor*) trees and this site has not experienced an intense wildfire since 1992 and no prescribed burns since 2006. However, the prescribed burn at Golgotha Cave was much less intense and it was more controlled than the fire that is reported in our study.

Comment [GN22]: We detail that the Golgotha burn was much less intense than the wildfire noted in this study. Response to Reviewer 3 comment L178-179

4 Forward model

We use the forward model employed by Baker et al., (2010). This model uses monthly rainfall $\delta^{18}O$, monthly (P – AET), from 2003 to 2011 (we use 2003 to 2005 data as a 'warm up' period to avoid edge effects), and adjustable bedrock flow thresholds for seepage flow and fracture flow to predict dripwater $\delta^{18}O$ based on hydro-climatic influences. Seepage flow and fracture flow thresholds are hydrological P - AET thresholds that are required for infiltrating water to enter seepage or fracture reservoirs (for further details see Baker and Bradley, 2010). The Tamala Limestone retains high primary porosity thus seepage flow is likely to be dominant whilst fracture flow is less dominant and only likely to be activated during high infiltration (Treble et al., 2013; Mahmud et al., 2015).

Comment [GN23]: Addition of observational window in response to Reviewer 1 comment L201-L205.

Further we also re-wrote section 4 to make it clearer to the reader the role of the forward model in this study. (in response to reviewer 1 comment L453 and Reviewer 1 comment 243-272 and other general comments from the other reviewers and the editor.

Storage tiem for the water that enters the seepage reservoir is modelled as a Gaussian distribution. A maximum residence time of 3 years is set; this reflects the dominating seepage or matrix flow type at our site, the shallow depth (4 m) and the potential for capillary barrier effects to impact hydrology in this region (Mahmud et al., 2015). Further a minimum residence time of 10 months is required, to maintain the observed year round discharge at

Comment [GN24]: Changed – in response to Reviewer 1 comment L193

both sites. The model allows for the mean and standard deviation to be specified for these functions. Being conservative we specify the minimum residence time of 10 ± 2 months. In contrast, the fracture-fed flow is instantaneously passed through the system (i.e. with a travel time of less than one month). In the model we can adjust the (P – AET) thresholds required for flow into the seepage reservoir and the threshold required for it to spill into the fracture flow. The seepage and fracture-fed components are mixed in the overlying bedrock reservoir, before predicting dripwater $\delta^{18}O$ composition. By request the authors can supply the forward model as a spread sheet.

We tested a full range of seepage and fracture possibilities. This suite of model runs helps to place constraints on $\delta^{18}O$ variability that can be explained by hydro-climatic variability alone. We compare these scenarios to the observed dripwater $\delta^{18}O$ at our sites, to assist in our interpretation of the post-fire dripwater $\delta^{18}O$ response.

Comment [GN25]: Re-written in response to Reviewer 3 comment L192-4

Comment [GN26]: Changed from Karst store to overlying bedrock reservoir. Response to reviewer 3 comment L198

5 Results

A time series of monthly P – AET, cumulative water balance (CWB), discharge, dripwater $\delta^{18}O$, and ion concentrations for Sites 1a and 2a from August 2005 – March 2011 are shown in Figure 2.

5.1. Water balance

Firstly, we observe a distinct seasonality in the water availability (P – AET) (Fig. 2A), where winter months generate an excess (P > AET), while summer months generate a deficit (P < AET). Further, CWB shows three distinct trends throughout the monitoring period: 1) a decline over the period of January 2006 to June 2006, consistent with very low excess in P - AET; 2) an overall rise from June 2006 to February 2010; 3) a decrease in P - AET from February 2010 to September 2010. Site 1a and 2a display moderate and similar discharge rates, at the start of the monitoring period that continue until July 2006; Site 1a an average of 90 ml ± 21 per week and Site 2a an average of 92 ml ± 23 per week. This coincides with infiltration indicated by positive CWB (Fig. 2B). In July 2006, Site 1a dramatically increased discharge five-fold to 468 ml/week on one cave visit, but had decreased to 55 ml ± 3 ml per week on the subsequent visit two weeks later and was completely dry, three months later. This site has not re-activated since (Department Parks and Wildlife, pers. comm.). Site 2a shows much less variation in discharge overall, but contains smooth long-term trends. Two periods of higher discharge are observed in August 2005 to May 2006 (average 92 ml ± 23 ml per week) and April 2008 to February 2009 (average 93 ml ± 29 ml per week), both coinciding with positive trends in CWB.

Comment [GN27]: Changed to water availability to make the term consistent in response to Reviewer 3 comment L217.

Comment [GN28]: Changed from 'increases' to 'increased'. (reviewer 3 comment L224)

5.2 Water isotopes

Dripwater $\delta^{18}O$ from Site 1a (Fig. 2C) shows no seasonal pattern but we see a steady increase of 1‰ to January 2007, then a further steeper rise of 1.5‰ in June 2007, after which the drip ceases. Dripwater $\delta^{18}O$ from Site 2a presents an overall increasing trend rising from -3‰ to +0.7‰ over the monitoring period with a 6-month quasi-seasonal signal (approx. 2‰ range) that peaks in cooler months (June to October) generally coinciding with months when infiltration from rainfall occurs. We hypothesise that the P < AET environment in drier summer months isotopically enriches soil water, but this only arrives at the cave when seepage thresholds are exceeded in periods of P > AET (winter months).

We forward modelled our rainfall isotopic data in order to predict drip-water $\delta^{18}O$ under various hydro-climatic scenarios (Fig 3). Our sensitivity analyses of hydrological residence times and thresholds showed that seepage residence times, less than 10 months resulted in the seasonal cessation of dripwater, which is not observed at our sites. Therefore, a minimum seepage residence time is required to match our observations. Further, seepage threshold values greater than 40 mm (P – AET) also resulted in the cessation of our drip site. Thus seepage threshold must be below 40 mm (P – AET) to match our observation. Next we varied the fracture threshold between 15 mm and 1000 mm, the wide range reflecting our uncertainty over this parameter. However, we know that seepage flow is dominant at these sites (Mahmud et al., 2015). This suggests two things, first, the seepage threshold is low, second, the threshold required for water to 'overflow' from the seepage reservoir to fracture reservoir must be significantly higher than the seepage threshold. We note that scenarios with a lower fracture threshold (10 – 15 mm) show high variability in comparison to sites with a seepage dominated flow and no fracture flow (10 – 1000 mm). Based on the variable

Comment [GN29]: We have removed the slope calculations and p-tests in this section in response to reviewer 2's comment L262-264.

Comment [GN30]: Re-written and added month for Northern Hemisphere readers. Response to reviewer 3 comment 239.

Comment [GN31]: Re-written in continued response to reviewer 1 comment L243-L276 and Reviewer 3 comment L243

Comment [GN32]: We have defined the thresholds required for seepage and fracture flow (P-AET), in response to Reviewer 3 comment L244).

854 morphology of stalactites and stalagmites at our sites we interpret discharge to be a
855 combination of seepage and fracture flow, but with seepage clearly dominating. Hence we
856 chose the 15 – 100 mm scenario to represent the hydrology at our cave site (Fig. 3). Our
857 forward-modelled dripwater $\delta^{18}O$ mean is -4.1‰, slightly less than the mean of Perth rainfall
858 (-3.1‰). The time series of modelled dripwater $\delta^{18}O$ (Fig. 2C) starts and remains at ~ -4.2‰
859 until February 2006 where it dips slightly before rising sharply to -3‰ where it remains
860 steady until February 2007. Here it begins a step-wise decline; declining from February to
861 March 2007 by 0.5‰ and remaining stable again until February 2008. It then shows a further
862 step-decline in March 2008 to -4.5‰, where it remains at approximately this value, albeit
863 with a few small variations on timescales of months, until the end of the monitoring period.

864 In all meaningful modelled scenarios i.e. ones that have full year flow and test the full range
865 of hydrological variability, estimated dripwater $\delta^{18}O$ cannot replicate the higher observed
866 dripwater $\delta^{18}O$ which are +1‰ to +3‰ higher compared to modelled (Fig. 3). This clearly
867 suggests another factor is affecting dripwater $\delta^{18}O$ composition: likely near-surface
868 evaporation.

869

870 To investigate an evaporation effect, we plot cave dripwater along the local meteoric water
871 line (LMWL, weighted LSR) to test for isotopic enrichment (Fig. 4). Figure 4 shows that
872 while the least squares regression (LSR) for cave dripwater falls within the standard error (±
873 0.45‰) of the slope for the local meteoric water line (LMWL, weighted LSR), drip water
874 isotopic composition is concentrated towards heavier $\delta^{18}O$ and $\delta^{2}H$. These results are
875 consistent with evaporation in a high humidity environment as has been observed in semi-
876 arid cave environments elsewhere (e.g. Cuthbert et al., 2014). Adopting Cuthbert's
877 classification, our data falls under a *type 1* scenario reported in Cuthbert et al., (2014). In the

Comment [GN33]: We have defined what comprises a 'meaningful model' in response to reviewer 3 comment L265.

Comment [GN34]: Re-worded as suggested by reviewer 3 comment L 273-274.

type 1 scenario, $\delta^{18}O$ and $\delta^{2}H$ do not deviate from the LMWL but are shifted along the LMWL towards higher values, as is the case with our data (Fig. 4). This means that our data are similarly impacted by evaporation occurring in a high humidity environment.

Comment [GN35]: Re-written to more clearly state that our bi-variate plot of d18O vs d13C supports our case for evaporation at our sites. Response to reviewer 1 comment L240

5.3 Water Solutes

There are significant differences in solute concentrations and trends between the two sites (Fig. 2E, 2F, 2G and 2H). Solute concentrations are typically higher at Site 1a versus Site 2a and they demonstrate opposite trends post-fire. At Site 1a, Cl, Ca, Mg and Sr decline overall, although this trend is step-wise for Ca, and reverses for Cl ~ 6 months before the drip ceases. The trends in these solutes at Site 1a are inconsistent with the declining CWB during this period (Fig 2A), as we would expect a drying trend reflected through the evaporative concentration of solutes. In contrast at Site 2a, Cl and other solute concentrations show a direct relationship to CWB (i.e. increasing solute concentration with decreasing CWB from 2006 until mid-2008 followed by decreasing solute concentrations with increasing CWB.

Comment [G36]: We have moved the interpretation of tree roots affecting the drip water chemistry for the discussion to Line 349-351. (in response to R3 comment 288)

Trends in SO_4 and K are more subtle than for other solutes: at Site 1a, K shows a slight decline from the beginning of the monitoring until early 2007 and then has a small rise prior to drips ceasing. Although harder to judge in the shorter SO_4 time series, SO_4 also shows a small rise before drips cease, similar to K. Trends in K and SO_4 for Site 2a are more subtle, although they both increase slightly over time. K and SO_4 concentrations are, two to three times higher at Site 1a versus Site 2a and are considerably higher than those recorded at Golgotha Cave (Table 1). We also note that initial Cl and other solute concentrations at Site 1a are twice that at Site 2a.

6 Discussion

6.1 Post-fire hydrology

Discharge at Site 1a is inconsistent with CWB: discharge rose as rainfall fell below the long-term mean (P < AET) (Fig. 2A and 2B), suggesting that Site 1a received a localised increase in discharge despite the declining input from rainfall. In contrast, discharge at Site 2a is more closely related to the cumulative water balance (CWB), with higher discharge coinciding with periods of higher water surplus and lower discharge with lower water surplus.

Chloride is a chemically conservative and highly soluble solute (Graedel and Keene, 1996), and its concentrations in dripwater will therefore reflect concentration/dilution effects (Tooth and Fairchild, 2003; Tremaine and Froelich, 2013). Chloride concentrations at Site 2a increase during the period of declining CWB (2006 to mid-2008) suggesting that evaporation is concentrating Cl. Rising $\delta^{18}O$ and other solutes over this period are also consistent with increased evaporation. From mid-2008 onwards, when CWB is positive (P > AET), Cl decreases, consistent with an increase in infiltration and thus dilution (Fig. 2E).

Comment [G37]: Re-arranged sentences such that interpretation followed observations and observations made more conclusive/definitive in response to reviewer 3 comment L316

At Site 1a higher discharge also coincides with falling Cl concentrations also suggesting dilution (Fig. 2B, 2D). However, we note this coincides with a highly negative CWB i.e. drier than normal conditions. This suggests in this case, a non-climatic driver has influenced infiltration. We propose that a reduction in localised transpiration, following the 2005 fire, may be driving this. Deeply-rooted trees within the area have been reported to produce high Cl concentrations in the unsaturated zone (Turner et al., 1987). Site 1a had a tuart tree directly above it and tree roots are visible above Site 1a in the cave, but not at Site 2a. The proximity of the tree to Site 1a is the most likely explanation for the higher solute concentrations here (Treble et al., 2016). The death of the tree in the 2005 fires would remove the previous transpiration demand and hence result in effective dilution of the solutes during

infiltration, as observed. However, this reduction in transpiration would have been abrupt but we observe a response lasting 1.5 years after the fire. This could be due to a number of reasons; firstly, the minimum residence time is 10 months (for a year of continuous discharge) so a delay in the response is to be expected. Second, this occurred during a period in which the soil moisture deficit would have been larger than average, so a larger volume of cumulative infiltration would be needed to overcome this deficit- and move the more dilute solute into the cave.

It is also possible the decrease in concentrations reflect the diminishing element concentrations after an immediate flush of the more soluble ash-derived material (i.e. the tail of a solute pulse). However, post-fire, highly soluble solutes like Cl, will still reflect dilution due to increased discharge. So, it is likely that we are seeing a decline in these elements due to a combination of the removal of these nutrients from the surface and subsurface, and dilution.

In the broader context we look at the differences in Cl at Yonderup Cave vs Golgotha Cave. Both caves are ~5 km from the coastline, so they likely have a similar amount of Cl aerosol deposition. Yet, post-fire Cl values at Yonderup Cave, are more than double the Cl concentrations from Golgotha Cave (Table 1). Further, within cave variability in Cl concentrations at Yonderup Cave are twice that at Golgotha Cave (Table 2). While variations in vegetation density (along with wildfire history) may have some role to play in the difference in mean dripwater Cl at each location, the higher within cave variability at Yonderup Cave (Table 2), suggests a post-fire setting increases variability in dripwater chemistry. The impact of this is discussed further and a conceptual model (Fig. 6) devised later in section 6.3.

Comment [GN38]: Paragraph added for further clarification of our interpretation in response to the comment by Reviewer 1 L329-331, 338-340.

Comment [GN39]: We add the other possible cause for the decline in solutes at Site 1a in our interpretation here and throughout the manuscript. The removal of solutes from the surface and subsurface post-fire. (response to reviewer 2 comment L333-334).

Comment [G40]: Figure 6 introduced earlier in response to Reviewer 3 L389

Comment [GN41]: Distance between Yonderup and the coast and Golgotha and their possible effects on Cl added in response to reviewer 1 comment L332.

950

951 **6.2 Post-fire carbonate chemistry**

952 Similar to Cl, concentrations of carbonate metals (Mg, Ca and Sr) at Site 1a also decrease;
953 this reflects solutes being diluted due to reduced tree water-use. However, we note that at Site
954 1a, Ca, for example, declines twice as much (in concentration, ~75 %) in comparison to Cl
955 (~30%). Thus for Ca, another mechanism along with dilution is required to explain its non-
956 linear step-like decline (Fig. 2G).

957 There are a number of mechanisms that could influence post-fire Ca concentrations. First, we
958 consider increased near-surface evaporation inducing prior calcite precipitation (PCP).
959 Increased evaporation, can saturate solutes relative to calcite in karstic waters and promote
960 degassing. Further, evaporation will slow the flow increase water-rock interaction times in
961 the remaining water. Both of these conditions are ideal for PCP. Both our sites show evidence
962 of PCP: the ln(Sr/Ca) vs ln(Mg/Ca) slopes in our data agree with the diagnostic range for
963 PCP (a slope of + or - 0.88; cf. Fig. 5B) (Sinclair et al., 2011). Expressed as a time series
964 (Fig. 5A), we see ln(Sr/Ca) and ln(Mg/Ca) increase simultaneously with $\delta^{18}O$ and Cl (Fig. 2),
965 suggesting that evaporation is indeed the common driving mechanism and is inducing PCP at
966 Site 1a. For further information on PCP processes we recommend the reader to Fairchild et
967 al., 2000; Sinclair 2011 and Treble et al., 2015.

968 A second mechanism influencing post-fire Ca concentrations may be the addition of plant ash
969 (Yusiharni and Gilkes, 2012a) and highly soluble CaO (produced by the burning of exposed
970 surface rock to the fire; Yusiharni and Gilkes, 2012b). Further, it is possible that the Ca
971 decline may also reflect a decrease in Ca being leached post-fire that may have followed an
972 earlier spike in Ca concentrations from the above. The extent to which each process is
973 affecting the Ca concentration is difficult to assess in our data, especially since our

Comment [GN42]: Instead of saying that our figure provides a diagnostic model we have said that our figure shows our data 'falls within the diagnostic range' as was initially intended. Response to reviewer 3 comment L348. Further we have also re-written the Ca section in response to R3 comment 358

Comment [GN43]: We add further description on how increased surface evaporation could induce PCP, and direct the reader to further references in response to reviewer 1 comment L346.

Comment [GN44]: We have integrated a response to reviewer 3's Comment L358 and Reviewer 2's other possible explanation for the decline in solutes to help explain the decline in Ca.

monitoring did not commence until 6 months after the fire, and processes such as PCP and the addition of Ca from plant-ash are difficult to constrain.

We now consider other carbonate metals: Mg and Sr, at Site 1a to further constrain our interpretation. We find Mg and Sr decline by ~30% (in terms of relative concentration). Thus it is likely that the same process affecting Cl is also affecting Mg and Sr, that is, dilution, a decline in leaching of biomass-sourced ash, or some combination of both. Ca concentrations decline by a relatively larger amount (75%) suggesting that additional processes are specifically affecting Ca. The rise in Mg/Ca at Site 1a strongly suggests that the remaining portion of the Ca decline may be attributed to PCP (Fig. 5A).

At Site 2a, a rising trend, reflects the concentration of solutes due to a rise in post-fire evapotranspiration, evidenced by increasing $\delta^{18}O$ and Cl, evaporation of near-surface water stores (Fig. 2F, 2G and 2H respectively), and possibly to some extent, an increase in transpiration from vegetation recovery for Cl (Treble et al., 2016). Additionally, Ca concentrations also show a quasi-seasonal response, interpreted from the Mg/Ca time series to be driven by PCP, possibly due to seasonal P – AET (supported by similar seasonality in dripwater $\delta^{18}O$; Fig. 2C) although in-cave PCP could also be contributing (Treble et al., 2015).

Sulphate and K post-fire at Site 1a are abnormally high in concentration, approximately three times higher in comparison to Site 2a (Figs. 2H, 2I; Tables 1, Table 2). This is counterintuitive to the initial dilution signal (owing to a decrease in tree water-use) interpreted for the other solutes. While at Site 2a, SO_4 and K increase similar to other solutes, consistent with evaporative concentration (from post-fire conditions) and an increase in transpiration from (vegetation recovery). These observations suggest that there was an

Comment [G45]: Reworded

Comment [GN46]: Table 2 (suggested by Reviewer 2) helps quantify the differences between SO4 and K between the two sites, pre-empting speculation. Of how the two Sites response post-fire. Helping satisfy Reviewer 1 comment L364-386. And Reviewer 2 comment 374-377 .

increase in the availability of SO_4 and K after the fire at Site 1a despite a decrease in tree-water use here (Fig. 2H and 2I).

Comment [G47]: Reduced speculation. Response to reviewer 1 comment L364 – L386

We note the majority of aboveground SO_4 is predominantly stored within the lower to middle storey of the forest (O'Connell and Grove, 1996), and post-fire soils contain 23% more S and 16% more K than pre-fire soils due to biomass-sourced ash deposition (Grove et al., 1986). So SO_4 and K concentrations at each site may respond differently since the amount of available SO_4 and K above each site is influenced by the amount of biomass burnt above the site. Further, the dissolution rates of ash minerals containing these elements could also affect the rate at which these nutrients are leached from the surface and subsurface and subsequently their concentrations in dripwaters. We propose that the large amount of biomass burnt above Site 1a – the tuart tree - is responsible for the much higher concentrations of SO_4 and K at Site 1a dripwater relative to Site 2a. We also propose that the increase in near-surface evaporation from 2007 onwards drives even higher concentrations of SO_4 and K (Fig 2H and 2I). Site 2a, which has much less pre-fire biomass, has much lower SO_4 and K concentrations, consistent with our argument. Here, these solutes show a steady increasing trend over the monitoring period. This is consistent with increased evapotranspiration post-fire, which is also evident in the other solutes.

Now, we compare our Yonderup Cave results to those of Golgotha Cave to put SO_4 and K concentrations into context. Golgotha Cave, last experienced a wildfire in 1992 and a controlled low-temperature prescribed burn in 2006, while Yonderup experienced a high intensity burn in 2005. Firstly, we see five and ten times higher within cave differences in SO_4 and K (respectively) at Yonderup Cave than we do at Golgotha Cave (Table 2). Also, concentrations at Yonderup Cave are up to four and three times higher in SO_4 and K

(respectively) at Yonderup Cave than at Golgotha Cave (Table 1). These results coupled with the difference between Cl composition at Yonderup vs. Golgotha presents a clear case for more variability in burnt sites in comparison to unburnt sites.

6.3 A multi-proxy fire signal in dripwater

Here we propose post-fire scenarios for both sites at Yonderup Cave (refer to conceptual model Fig. 6) that account for the altered dripwater chemistry that is observed post-wildfire. A straight-forward relationship between cumulative water balance (CWB), discharge and Cl concentrations at Site 2a, suggests increased concentration of solutes (Ca, Mg, Sr, K and SO_4) in response to an increase in near-surface evaporation (Fig. 2C and 2D).

In contrast solutes such as Cl, Mg, Sr, Ca at Site 1a show a declining trend. We have argued that this declining trend in these solutes is due to two underlying processes. First, a decrease in tree water-use (transpiration) due to the death of the tuart tree in the wildfire. Tuart trees are deeply-rooted, having adapted their root systems to access water from both the surface and subsurface. These roots have been found to generate potential energy between the tree and the soil to extract water and nutrients. Specifically, mature tuart trees generate a pressure gradient ranging from -0.86 ± 0.11 MPa (summer) to -0.35 ± 0.02 MPa (winter) (Drake et al., 2011). So the death of the tuart tree in the wildfire had a significant local effect on hydrology at Site 1a resulting in an increase in discharge and dilution of solutes (Cl, Mg, Sr and Ca). Our study suggests that the consequent reduction in transpiration may not be immediately detected in dripwater, owing to transit time through the limestone and the requirement for overcoming a soil moisture deficit during drier than average climatic conditions. Hydrological effects such as 'capillary barriers' can also slow down vertical transport of infiltrating waters at our site (Mahmud et al., 2015).

Comment [GN48]: Context of Tuart tree transpiration added in response to reviewer 1 comment L399 and L400

Second, the decline in solutes at Site 1a could be the result of a gradual return to pre-fire concentrations following a pulse of increased input of solutes from ash (e.g. Site 1a scenario). A contribution of SO_4 and K from burnt biomass may explain their relatively high concentrations.

Further, isotopic composition at Site 1a show that $\delta^{18}O$ is offset from modelled hydro-climatic $\delta^{18}O$ by ~+1‰ suggesting increased near-surface evaporation post-fire which we attribute to the reduction of shading from the tuart tree post-fire - even in P > AET periods (Fig. 2C). And post-2007, when P < AET conditions arrive, $\delta^{18}O$ rises even higher and discharge declines. Eventually, due to the persistent duration of P < AET conditions Site 1a ceased dripping owing to eventual depletion of the near-surface reservoir feeding this drip.

Site 2a also shows higher $\delta^{18}O$, during the post-fire period, which we attribute to an increase in near-surface evaporation as a result of low albedo and reduced vegetation cover. The case for evaporation at this site is supported by the rise in Cl and other solutes. The possibility of an immediate spike in solutes from ash and a long-term decline from leaching, as was discussed for Site 1a, is limited here, as there was less biomass available to burn above Site 2a. Therefore, we interpret increased evaporation and transpiration from regrowth post-fire to be the dominant forcing at this site, similar to the findings of Treble et al. (2016).

We propose that differences in surface vegetation above sites can influence site specific drip chemistry. For example, we interpret Site 1a was influenced by a reduction in transpiration after the fire, due to the forcing biomass above the site; which may also have been a source of post-fire ash at this site. At Site 2a the response to the fire was primarily an increase in near-surface evaporation owing to changes in surface albedo. This variability within the cave response at Yonderup Cave is significant, so too is the comparison between Yonderup Cave drip chemistry and the drip chemistry at Golgotha Cave. The latter more generally highlights

Comment [G49]: We have reduced speculation. This sentence along with adjustments in section 6.2 to answer the comment L448 by Reviewer 1.

Comment [GN50]: Added, in response to Reviewer 1 comment L397, showing that observed d18O and modelled do not agree. Further we demonstrate how the climate forcing (P > ET) inhibits a full evaporation post-fire to take effect.

Comment [GN51]: Re-structured sentence to respond to Reviewer 1 comment L408 to 410. Further with the re-structuring of this section and appropriate clarification we believe we have a more compelling argument – answering Reviewer 1 comment L419

that an intense wildfire has variable, but multi-year effects on dripwater composition in shallow caves.

From this we propose that post-fire condition persist up-to 5 – 10 years' post-fire, affecting dripwater $\delta^{18}O$ and solute concentrations. We would expect a full recovery, of $\delta^{18}O$ and solute concentrations back to pre-fire levels within 10 - 20 years as a result of revegetation growth (Treble et al., 2016) and re-establishment of vegetation cover and pre-fire albedo.

7 Application for a speleothem paleo-fire signal

Our post-wildfire dripwater response from $\delta^{18}O$ was a 2‰ increase above that predicted by a hydro-climatic model, and measured regional groundwater and Golgotha Cave $\delta^{18}O$ data. If this signal is preserved at equilibrium in speleothems this is equivalent to some of the largest interpreted climatic changes seen in the Quaternary record. This highlights the significance of the findings in our study, which suggests a fire signal could in fact be misinterpreted as climate variability. Furthermore, the impact on Ca dissolution from the limestone bedrock could have a significant effect by decreasing on speleothem growth rate.

However, before attributing $\delta^{18}O$ and growth rate abnormalities to fire, we must remember there are a number of processes that effect speleothem $\delta^{18}O$. Thus it is important that a multi-proxy approach, which uses isotopic composition as well as a suite of trace elements (sourced from both soil and bedrock), is used to separate fire from other forcings such as climate and other local factors. Further, our study was conducted in a shallow cave environment, where perhaps the overlying vegetation can exert a more dominant forcing on dripwater hydrology and chemistry relative to deeper caves. Deeper caves have more complex hydrology; this involves mixing with other flow paths, which are possibly not fire affected. This may result in the smoothing of the fire signal; making it harder to isolate (McDonald and Drysdale,

Comment [GN52]: We have added a table of differences between Golgotha and Yonderup to quantify our interpretation and pre-empt speculation. As suggested by reviewer 2 comment (L430-431)

Comment [GN53]: Introduced earlier in the text in response to reviewer 3 comment L450-4452 and 455

Comment [GN54]: We add this section in response to Reviewer 3's general comments and comments L450-458. This section helps to move hypothetical implication for future studies of this kind from the conclusion, addressing general comments from Reviewer 1 and 3 on the conclusion. And in the process adding my conclusiveness to our conclusion.

Comment [GN55]: Possible effect of Ca on Growth rate introduced in text prior to conclusion as suggested by reviewer 3 comment L458.

2007). Further, a fire signal in cave dripwater and stalagmites~~,~~ may be much subtler in grassland environment in comparison to a forested environment as changes in the biomass would be smaller and vegetation recovery presumable faster (Coleborn et al., 2016). We recommend searching for fire signals in shallow cave environments in the tree rooting zone in forested areas.

One further approach that may help to differentiate fire and climate signals in a stalagmite would be to use multivariate statistical techniques such as principal component analyses (PCA). Using this technique, we would expect one component to reflect a bedrock/hydro-climatic signal and another to preserve a local soil/vegetative forcing. The soil/vegetative component could preserve the impact of a fire on stalagmite composition, in trace elements like S, and K. Further, it is also possible an immediate spike in solutes (Mg, Ca, Cl, Sr, S, K and P) from post-fire ash may be preserved in stalagmites and colloid associated metals, such as Al, Fe and Cu from an increase in discharge post-fire. Future studies of this kind will open a new avenue in speleothem research; speleothems as archives of paleo-fire.

Comment [GN56]: Suggesting added, in response to reviewer 3, comment L456-457.

8 Conclusions

We isolate a post-wildfire response by comparing a recently burnt cave monitoring site with forward modelled $\delta^{18}O$, which predicts $\delta^{18}O$ based on hydro-climatic factors, and nearby cave monitoring and groundwater data. We provide a novel analysis of the multi-year impacts wildfire has on cave dripwater. Our analysis shows a strong hydrologic relationship between surface environments and shallow caves that are located within the tree rooting zone. This finding is especially important in water-limited environments (P < ET) as the overlying vegetation can exert controls on the cave hydrogeochemical environment.

A post-wildfire dripwater response is clearest in $\delta^{18}O$ and Cl due to their sensitivity to variation in near-surface evaporation (both $\delta^{18}O$ and Cl) and transpiration (Cl). Cl is a conservative ion and hence is driven mainly by by dilution/evaporation. Cl declines post-fire at Site 1a which we interpret as dilution of the water store that the dead tree previously exploited. In contrast, Cl increases at Site 2a, sympathetically with $\delta^{18}O$, consistent with an increased evaporative demand on shallow water stores driven by post-fire reduction in shading and reduced albedo. SO_4 and K are also important as at sites with abundant biomass they can be leached at high concentrations as they are made more abundant in post-fire soils due to the ash generated from a fire. Other solutes such as Mg, Sr and Ca support the dominant local forcing at the site post-fire and can be extremely powerful when using a multi-proxy approach.

We propose a conceptual model for a multi-year post-wildfire cave dripwater response in forested water-limited regions. This involves a 5 - 10 yr response of: 1) higher $\delta^{18}O$ and Cl in cave dripwater due to increased evaporation and decreased shading after the wildfire; 2) increased K and SO_4 due to the leaching of biomass-sourced ash, particularly in areas with large biomass; and 3) increased variability in Mg, Sr and Ca due to changes in evaporation,

Comment [GN57]: The addition of section 7 and refinements in our conclusion help to make our conclusion more compelling and supportive. In response to reviewer 1 comment L449.

Comment [GN58]: By clarifying our model and rewriting our argument in response to all the reviewers comments we have a compelling case for our conclusion (response to Reviewer 1 comment L441-444 and L446-447

transpiration and water-rock interactions post-fire. We may expect a recovery within 10-20 years after the wildfire and a restore to pre-fire isotopic and trace element concentrations as a result of increased bio-productivity from forest regrowth and a re-establishment of canopy cover.

Acknowledgements

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Tables and Figures

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Table 1. A summary table comparing the hydrogeochemistry of, (A) shallow Yonderup Cave drip sites burnt in a 2005 wildfire to; (B) deeper Golgotha Cave sites burnt in a 1992 wildfire and a 2006 prescribed burn; (C) unpublished Perth rainfall data during the monitoring period (ANSTO); (D) rainfall isotopic composition (Turner and Thorpe, 2001); (E) groundwater isotopic composition (Turner and Thorpe, 2001); (F) Yanchep rainfall solute composition (Hingston and Gailitis, 1976).

Location		Discharge (ml/day)	Ca (mmol/L)	Mg (mmol/L)	Sr (mmol/L)	Cl (mmol/L)	SO_4^{2+} (mmol/L)	K (mmol/L)	$\delta^{18}O$ (per mil)	n
(A) Yonderup										
Site 1A	Median	12.2	1.2	0.46	0.01	8.1	0.25	0.15	-3.2	27
	SD	2.6	0.6	0.13	0.2×10^{-2}	0.6	0.02	0.03	1.23	
Site 2A	Median	6.1	1.42	0.37	0.01	5.91	0.14	0.05	-1.48	49
	SD	4.8	0.2	0.04	0.5×10^{-3}	1.75	0.05	0.8×10^{-2}	0.84	
(B) Golgotha										
Site 1A	Median	63	1.2	0.3	0.2×10^{-2}	3.35	8.0×10^{-2}	0.02	-4.1	85
	SD	4.7	0.22	0.03	0.183×10^{-3}	0.18	0.4×10^{-2}	0.3×10^{-2}	0.3	
Site 1B	Median	40	1.1	0.3	0.2×10^{-2}	3.41	8.0×10^{-2}	2.3×10^{-2}	-3.9	82
	SD	2	0.25	0.03	2.39×10^{-4}	0.25	0.5×10^{-2}	0.5×10^{-2}	0.1	
Site 2A	Median	47	1.1	0.32	0.2×10^{-2}	2.76	0.13	3.8×10^{-2}	-3.9	77
	SD	10	0.25	0.02	0.1×10^{-3}	0.22	0.8×10^{-2}	0.5×10^{-2}	0.1	
Site 2B	Median	67	1.22	0.33	0.2×10^{-2}	4.49	9.8×10^{-2}	3.8×10^{-2}		84
	SD	85	0.27	0.03	0.2×10^{-3}	0.24	0.6×10^{-2}	0.7×10^{-2}		
Site 2E	Median	524	1.9	0.31	0.1×10^{-3}	4.11	0.085	3.8×10^{-2}		51
	SD	45	0.2	0.02	0.2×10^{-3}	0.17	0.4×10^{-2}	0.4×10^{-2}		
(C) ANSTO Rainfall	*Mean*								-3.1	64
(D) CSIRO long-term rainfall (Perth)	*Mean*								-3.85	165
(E) Regional groundwater (Perth)	*Mean*								-4.68	43
(F) Yanchep Rainfall Solutes	*Mean*		0.008			0.01	0.1×10^{-2}	0.4×10^{-2}		

Table 2. Summary of differences in mean concentration of solutes and isotopic composition of solutes among sites at Yonderup and Golgotha caves. We see that Cl, SO_4, K and $\delta^{18}O$ values, at both sites are distinctly different. Specifically, the solutes have higher concentrations and $\delta^{18}O$ is higher at Yonderup Cave in comparison to Golgotha Cave.

Comment [GN59]: Table of differences added in response to reviewer 3 comment L430-431.

Site Differences								
	Discharge (ml/day)	*Ca (mmol/L)*	*Mg (mmol/L)*	*Sr (mmol/L)*	*Cl (mmol/L)*	*SO_4 (mmol/L)*	*K (mmol/L)*	*$\delta^{18}O$ (mmol/L)*
Yonderup site differences in mean								
Site 1a and Site 2a	6.1	0.22	0.09	0.02	2.19	0.11	0.1	1.72
Golgotha site differences in mean								
Site 1a and Site 1b	*23*	0.100	0.000	0.000	0.060	0.000	0.003	0.200
Site 1a and Site 2a	*16*	0.100	0.020	0.000	0.290	0.050	0.018	0.200
Site 1a and Site 2b	*22*	0.020	0.030	0.000	1.140	0.018	0.018	--
Site 1a and Site 2e	*461*	0.700	0.010	0.019	0.760	0.005	0.018	--
Site 1b and Site 2a	*7*	0.000	0.020	0.000	0.650	0.050	0.013	0.000
Site 1b and Site 2b	*27*	0.120	0.030	0.000	1.080	0.018	0.013	--
Site 1b and Site 2e	*484*	0.800	0.010	0.019	0.700	0.050	0.013	--
Site 2a and Site 2b	*20*	0.120	0.010	0.000	1.730	0.032	0.000	--
Site 2a and Site 2e	*477*	0.800	0.020	0.019	1.350	0.045	0.000	--
Site 2b and Site 2e	*457*	0.680	0.010	0.019	0.380	0.013	0.000	--
Average Gol. difference	199.400	0.344	0.016	0.008	0.814	0.028	0.010	0.133
St. dev of Gol. difference	220.907	0.331	0.009	0.009	0.488	0.019	0.005	0.200

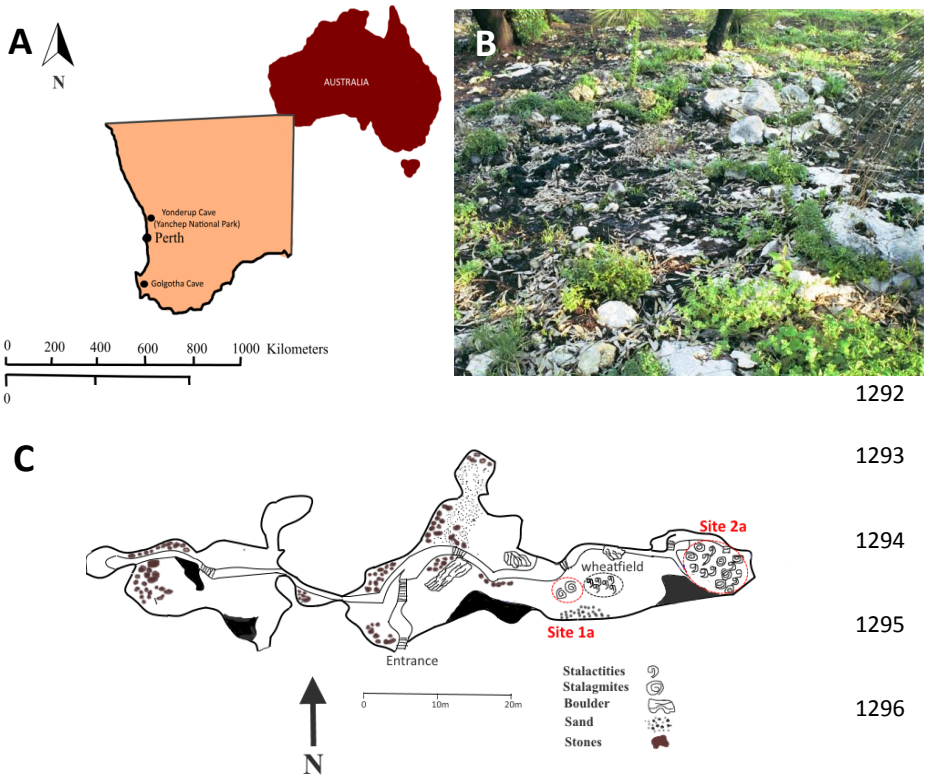


Figure 1. Geographical location of our study site (A), a post fire photograph of the area (B) taken in August 2005 a photo of recovering shrubs and grass post wildfire, and (C) a map of Yonderup cave to scale originally surveyed by Watts and Henley, (1973).

Comment [GN60]: Yanchep national park noted on the map as suggested by Reviewer 3 comment Fig. 1

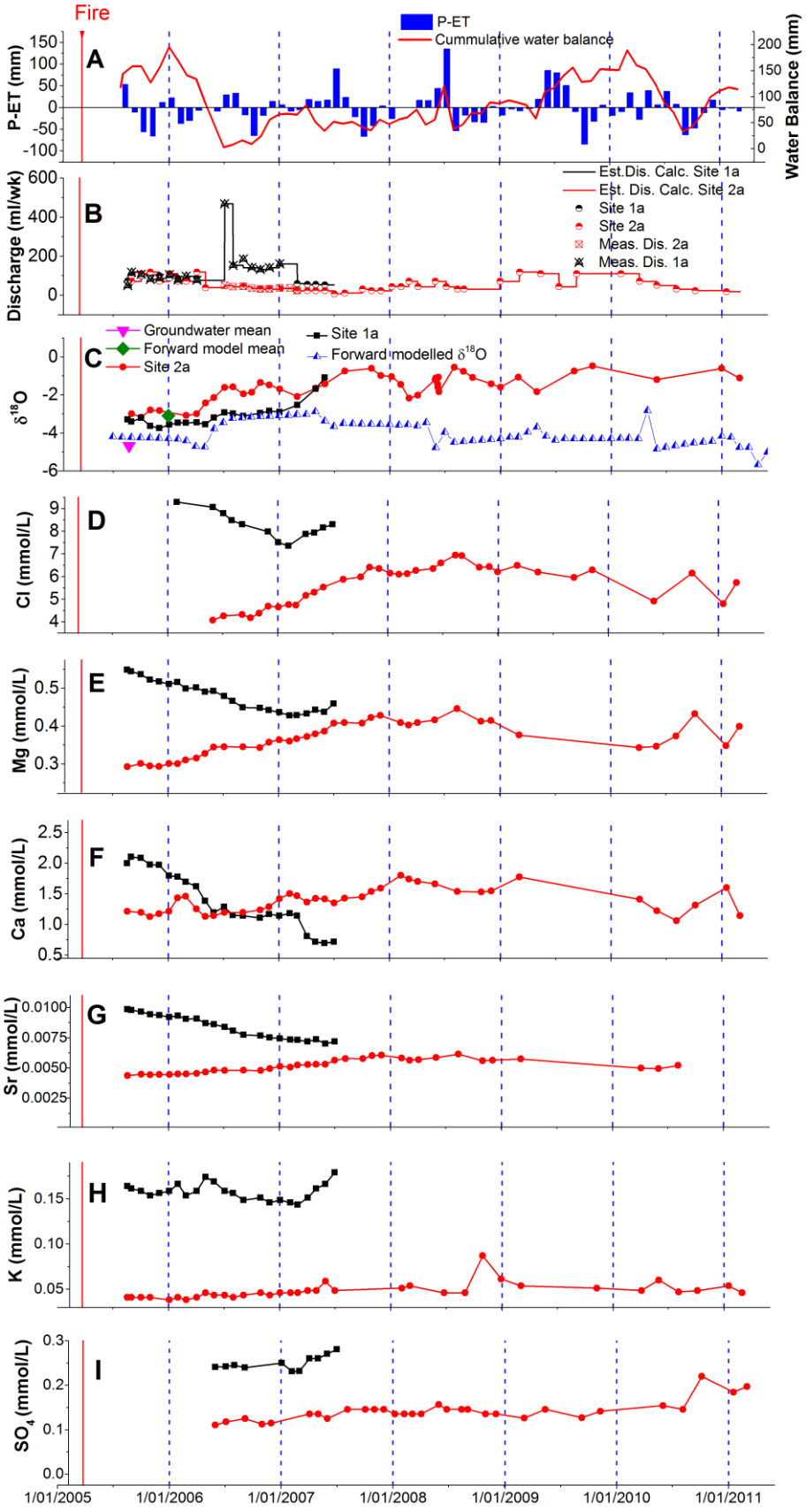

Fire
P-ET
Cummulative water balance
A
P-ET (mm)
Water Balance (mm)
B
Discharge (ml/wk)
Est.Dis. Calc. Site 1a
Est. Dis. Calc. Site 2a
Site 1a
Site 2a
Meas. Dis. 2a
Meas. Dis. 1a
C
Groundwater mean
Forward model mean
Site 2a
Site 1a
Forward modelled δ18O
δ18O
D
Cl (mmol/L)
E
Mg (mmol/L)
F
Ca (mmol/L)
G
Sr (mmol/L)
H
K (mmol/L)
I
SO4 (mmol/L)
1/01/2005
1/01/2006
1/01/2007
1/01/2008
1/01/2009
1/01/2010
1/01/2011

Figure 2. Presents a post-fire time series of data from Site 1a and Site 2a. Note: Site 1a dries up in June 2007. (A) Precipitation – actual evapotranspiration (P – AET), shows seasonal variations of excess (above threshold) and deficiency (below threshold) on a monthly scale overlayed with cumulative surface water balance. (B) Discharge is given in ml/week. Actual measured discharge data is given in blue (Site 1a) and Black (Site 2a) while estimated data is given in (red) this is then inferred to give measured discharge. Site 1a shows a spike in measured discharge in August 2006 and a consequent decrease until the site is dry, while Site 2a shows little variability in discharge throughout the monitoring period. (C) Shows observed $\delta^{18}O$ composition of cave dripwater from Site 1a and Site 2a with the forward modelled $\delta^{18}O$ (red) and mean modelled $\delta^{18}O$ (orange) and long-term groundwater $\delta^{18}O$ mean (pink). (D) Cl declines at Site 1a until Feb where it shows a slight increase until the drip becomes dry, while Site 2a shows a steady increase until in July 2007 where it stabilizes for the remainder of the monitoring period. (E) Post-fire response shows a decline in Mg at Site 1a until dry and a steady increase at Site 2a until in Dec 2007 where it remains stable. (F) Site 1a shows step wise decline in Ca at Site 1a until dry, while at Site 2a a very gradual increase until June 2007 is seen while the remainder of the monitoring period remains steady. (G) Response for Sr shows Site 1a declining and Site 2a peaking in Dec 2007; an identical response to Mg and at both sites. (H) K post-fire at Site 1a shows high concentrations, triple that of Site 2a but stable, while at Site 2a shows a slight increase over time. (I) SO_4 both Site 1a and Site 2a show a slight increasing trend over time. But Site 1a has more than double the initial absolute concentration in comparison to Site 2a, similar to other solutes.

Comment [GN61]: We have added the time of the fire to figure three. As suggested by Reviewer 2 and 3.

Comment [GN62]: Labels on figure adjusted in response to reviewer 3 comments figure 3B

Comment [GN63]: Changed to triple by Reviewer 3 comment L641

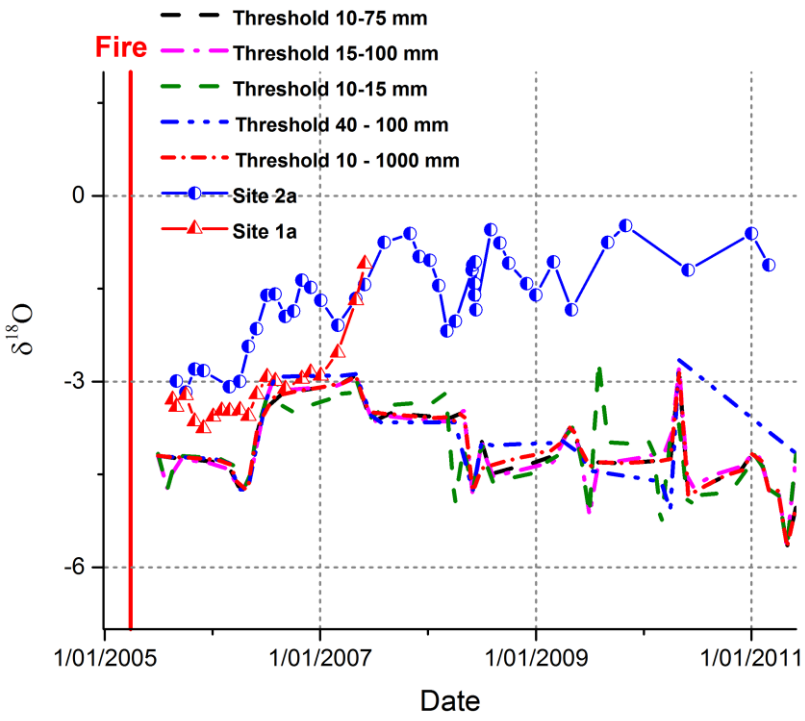


Figure 3. Modelled dripwater $\delta^{18}O$ outputs under varying thresholds in our forward model (model from Baker et al., 2010) which accounts for climatic and various epikarst threshold values that control isotopic values. Given no output matches observed dripwater composition we can infer that a localised factor has influenced isotopic compositions

Comment [G64]: Updated Figure 3 as per response

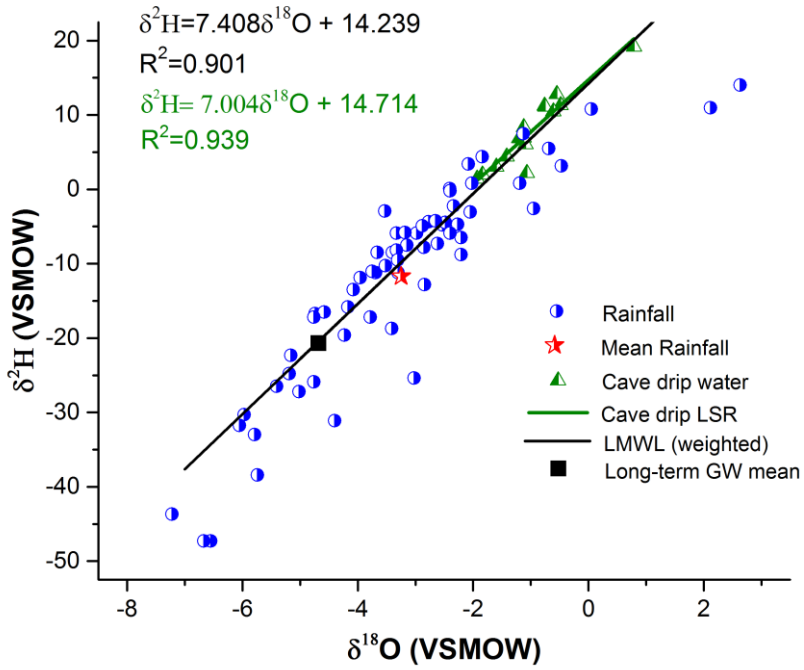


Figure 4. Shows compositions of δ^2H against $\delta^{18}O$ of our rainfall data (blue) during the monitoring period (ANSTO, unpublished), cave dripwater (red), long-term local groundwater mean (black) (0 – 10 ka, n=43, Southern Perth Basin from Turner and Thorpe, (2001)), and rainfall mean (red). A least squares regression (LSR) is plotted for cave dripwater (red line) and falls close to the local meteoric water line (LMWL, black) which is calculated using a weighted least squares regression (WLSR) using Hughes and Crawford, (2011).

Comment [GN65]: Moved Mean rainfall to below rainfall and changed colour for cave drip data. In response to Reviewer 3

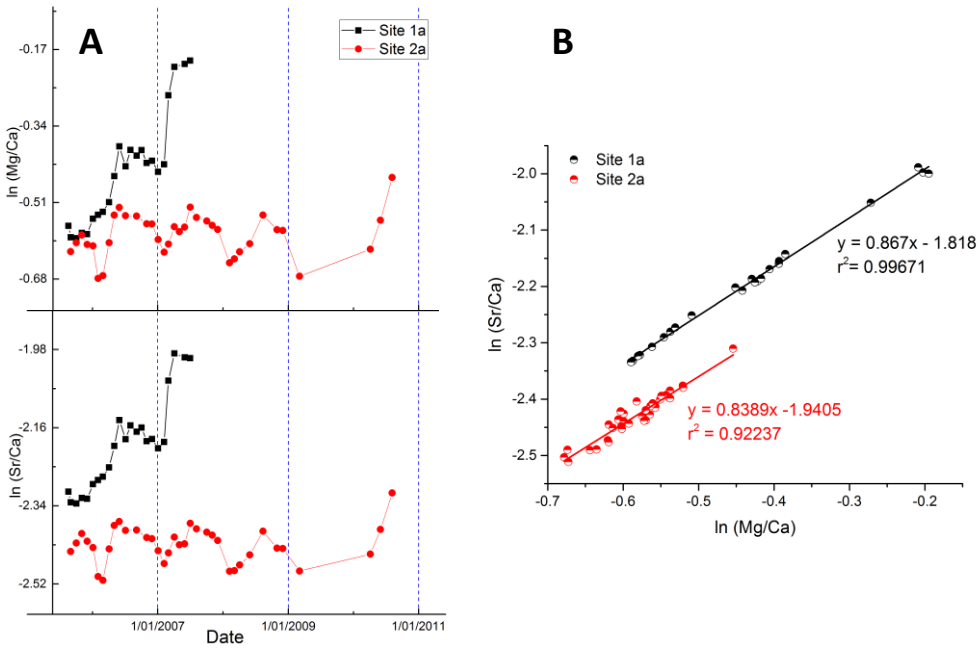
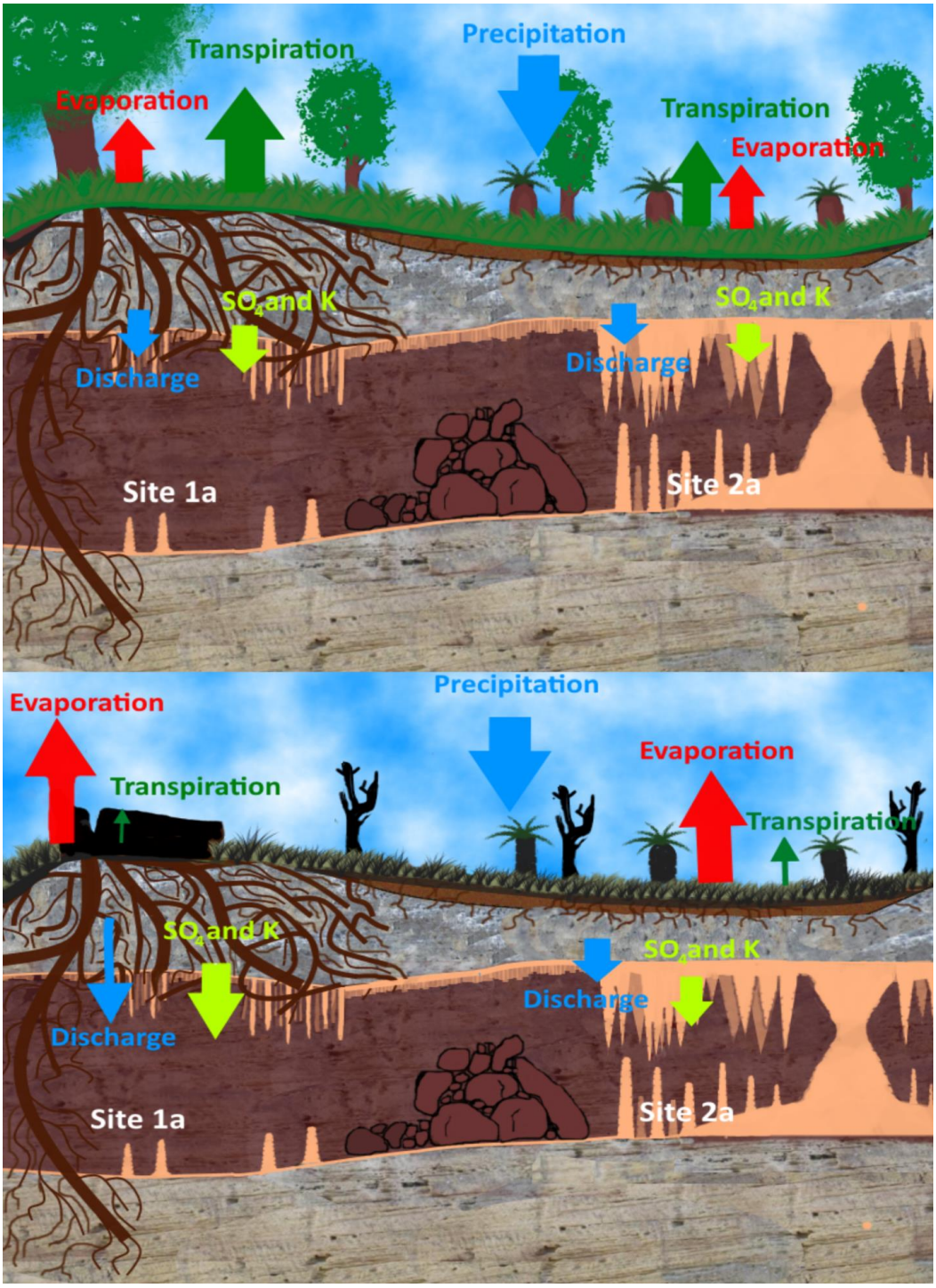


Figure 5. (A) A time series of ln(Sr/Ca) vs. ln(Mg/Ca). Site 1a shows a clear enrichment in Mg/Ca and Sr/Ca, or an increase in PCP, post-2007, driven by evaporation. Site 2a on the other hand shows quasi-seasonal variational in ln(Mg/Ca) and ln(Sr/Ca), suggesting here PCP is likely dominated by seasonality. (B) Shows that at both sites, ln(Mg/Ca) vs. ln(Sr/Ca) falls within the diagnostic range of PCP (a slope of + or - 0.88) suggesting PCP is occurring.

Comment [GN66]: We have added a Mg/Ca and Sr/Ca time series showing that thet covary over time. And at site 1a it increased post-fire. In response to evaporation as suggested in Section 6.2 (in response to reviewer 2 comment Figure 5)


Transpiration
Evaporation
Precipitation
Transpiration
Evaporation
SO_4 and K
Discharge
SO_4 and K
Discharge
Site 1a
Site 2a
Evaporation
Precipitation
Evaporation
Transpiration
Transpiration
SO_4 and K
SO_4 and K
Discharge
Discharge
Site 1a
Site 2a

1374

Figure 6) Evaporation (red) increases post-fire at both sites due to a reduction in albedo and vegetation cover while precipitation (blue) remains the same and initial transpiration (green) decreases, but recovers over time. Site 2a shows higher $\delta^{18}O$ and an increase in concentrations of solutes including SO_4 and K (lime) due to evaporation and slow increase in transpiration due to vegetation recovery, with cumulative water balance (CWB) remaining the same. While Site 1a shows, higher $\delta^{18}O$ in response to increased evaporation and a decline in solute concentrations in response to increased discharge and a decrease in transpiration and removal of nutrients from the surface and subsurface. However, since SO_4 and K are from biomass-sourced ash, the tuart tree above this site acts as a source of increased SO_4 and K. Discharge increased immediately (blue). But the drip became inactive one year after the fire due to an increase in evaporation, which outweighed the reduction in transpiration (green), leading to depletion of the near-surface reservoir feeding Site 1a and an in active drip site.