Authors Response to Reviews of 'Oxidation of sulphides and rapid weathering in recent landslides'

We are grateful for three thorough and helpful reviews, which have helped improve the manuscript
significantly. In the following responses, the original comments are kept in full, and our responses and actions are italicised.

Anonymous Referee #1

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- 10 In their manuscript entitled "Oxidation of sulfides and rapid weathering in recent landslides", Emberson et al report data on the solute geochemistry and size of landslide deposits in the mountains of Taiwan. Exploiting the fact that a majority of the exposed landslides were triggered during a single storm event, the authors test the links between landsliding and chemical weathering independent of time. While the authors do not find a relationship between the size of landslide
- deposits and the solute concentrations they generate, they do find that rivers in catchments with more landslides tend to have the higher solute concentrations.
 Overall, I think that this manuscript is timely and appropriate for publication in Esurf. However, there are a variety of criticisms that I would like to see addressed. In general, I found many instances where either key assumptions were not made clear or statements were not appropriately
- 20 backed up by published literature. I've tried to highlight these instances in my more detailed comments below.

Line 34 This is hardly an exhaustive list of weathering-erosion models. Additionally, the Dixon

- and Von Blanckenburg paper does not present a new model of the coupling between weathering and erosion. I'd recommend that the authors cite some of the key papers in this field (included below). Additionally, the authors state say that existing weathering-erosion models do not account for the effects of landsliding. While this broadly true, the issue was previously addressed by Gabet (2007; EPSL), who argued that landsliding did not affect the coupling between weathering and erosion
- 30 predicted by existing models. Since it seems as if the authors are trying to argue the opposite point, it'd be worth discussing the conclusions of Gabet (2007).

Gabet, E. J. (2007). A theoretical model coupling chemical weathering and physical erosion in landslide-dominated landscapes. Earth and Planetary Science Letters, 264(1-2), 259–265.

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Ferrier, K. L., & Kirchner, J. W. (2008). Effects of physical erosion on chemical denudation rates: A numerical modeling study of soil-mantled hillslopes. Earth and Planetary Science Letters, 272(3-4), 591–599. <u>http://doi.org/10.1016/j.epsl.2008.05.024</u>

40 Lebedeva, M. I., Fletcher, R. C., & Brantley, S. L. (2010). A mathematical model for steady-state regolith production at constant erosion rate. Earth Surface Processes and Landforms, 35(5), 508–524. http://doi.org/10.1002/esp.1954

West, A. J. (2012). Thickness of the chemical weathering zone and implications for erosional and
climatic drivers of weathering and for carbon-cycle feedbacks. Geology, 40(9), 811–814.
http://doi.org/10.1130/G33041.1

Li, D. D., Jacobson, A. D., & McInerney, D. J. (2014). A reactive-transport model for examining tectonic and climatic controls on chemical weathering and atmospheric CO2 consumption in granitic regolith. Chemical Geology, 365, 30–42. <u>http://doi.org/10.1016/j.chemgeo.2013.11.028</u>

The reviewer correctly points out that the list of references in not exhaustive. We have added a discussion of the Gabet paper, as that has been an important precursor for both this work and our previous work in the WSA. We have also cited the other papers in the introduction to increase the completeness of our citation.

Line 45 You provide no citations for mineralogical abundances at the different sites. Is this something you measured yourself? If not, you should provide the appropriate citations.

60 The missing references (Das et al. 2012 for Taiwan, Chamberlain et al. 2005, Koons and Craw 1991 for the WSA) have been added.

Line 80 You make the distinction between landslides receiving water directly from rainfall or from runoff into the crest of the landslide. However, in all figures and tables, it is not made clear which type of landslide each sample corresponds to. I think this distinction would be very helpful

since it is reasonable to expect runoff to have higher initial solute concentrations then rainfall.

The reviewer raises an important point that runoff is likely to have more concentrated solute contents than rainfall. However, we would suggest that the input of solutes along a flow path is not

70 additive; it is subject to saturation with respect to the weathering minerals. Both rainfall and runoff may be undersaturated with respect to the minerals weathering, but could both become saturated within landslides (we point to evidence of supersaturation with respect to calcite in all seepage samples). We have no constraints on the chemistry of runoff into the landslides from which we have sampled seepage. Moreover, the resolution of available DEMs does not always permit a robust

75 estimation of the drainage area upslope of the landslide crest. For these reasons, we have chosen not to provide detailed information about this issue. Despite this, we have altered the text to resolve confusion.

Lines 91-92: "This water has circulated through the landslides and derives from rainfall, and in some cases is likely supplemented by runoff into crests of landslides."

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Line 98 What is thick? Is there anyway to put an approximate scale on soil depths in this system?

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The original use of the word 'thick' is subjective, and we have changed it. Lee and Ho (2009) found soils in the mountains of Taiwan > 1m thick only in the bottom of valleys. We note that in these steep landscapes the soil thickness in slopes on short (10s metres) length scales varies widely even in more stable catchments.

Lines 111-113: "Soils are generally only >1m in local concavities in the landscape (Lee and Ho 2009), but contiguous, dense vegetation on hillslopes that have not been dissected by landsliding suggests sufficient mobile regolith or soil is present to support flora."

Line 105 These statements seem into imply that all deep groundwater is geothermal in origin,

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which I do not think is true (e.g., I don't think the groundwaters in Calmels et al 2011 EPSL are geothermal). Additionally, the authors state that samples are corrected for geothermal inputs but never describe how this is done.

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The reviewer is completely correct. We agree that geothermal and deep groundwaters are not identical. We have better constraint on geothermal waters in this system, having sampled them. In the first version of the text we described a correction, but what we meant was that we checked the

100 non geothermal samples for elevated Lithium contents; if they are >10umol/l then we would exclude them, as this implied >1% geothermal input (based on the measured lithium concentration in the hot spring sample). None of the samples fit this description though, so none were excluded. We have altered the text to remove mention of correction, and instead stress that the geothermal sample is used to qualitatively check for input from this source to either sulphate or other elements 105 in the seepage samples.

See lines 123-125: "We compare sampled seepage with our own measurements of geothermal water as well as with the composition of deep groundwater reported in the literature to qualitatively assess the importance of deeper-sourced water in landslide outflow."

110 ### Line 108 It'd be interesting to know what percentage of all landslides in the catchment were visited during your fieldwork. This would help readers assess whether or not the observation that half of the visited landslides were dry is likely to be representative of the entire system.

We mapped 576 landslides in the Taimali catchment, so we visited about 10% of these. We have included this detail in the text –

Lines 129: "despite the dry conditions more than half of around 50 deep-seated landslides we visited had persistent seepage" & 204: "We mapped 576 landslides extant in the Taimali"

Line 141 The authors attempt a correction for atmospheric deposition using chloride concentrations and element/chloride ratios. While this is reasonable, it should be mentioned that 120 geothermal waters, which the authors acknowledge are likely solute sources, are extremely enriched in chloride such that a small geothermal input would have a disproportionate effect on chloride concentrations and preclude the use of their proposed chloride mixing equation to correct for atmospheric deposition. Furthermore, while the element/chloride ratios of rainfall are often assumed

- to be close to those of seawater, it is widely observed that ratios such as SO4/Cl are enriched in 125 rainfall relative to seawater by a factor of 2 (Andreae et al. 1990 JGR). This is true even for rain that falls directly over the ocean (e.g., Stallard and Edmond 1981 JGR) and likely arises from marine biogenic aerosols. So, while the authors try and defend their choice of using seawater ratios, I am skeptical that it is a good assumption for all of the major elements.
- Considering the overall point of the manuscript, I do no think it is that important that the authors 130 correct their data for atmospheric inputs. By selecting a single rainfall end-member, the authors already assume that the chemistry of rainfall does not vary spatially across their catchment. So instead of assuming that 1) all chloride comes from rain and 2) that rainwaters = diluted seawater, which are both probably wrong, they can just compare their data with the assumption that the
- contribution from atmospheric deposition is not the key source of variability. Alternatively, the 135 authors could utilize a more rigorous >2 end-member mixing approach (too simultaneously account for geothermal inputs), but that seems beyond the scope of this manuscript.

We thank the reviewer for this valuable comment. The fact that the correction using measured

- 140 rainfall values [e.g. Wai et al. 2008, Ezoe et al. 2002, Cheng and You 2010] gives negative concentrations of some elements suggests that deep groundwater derived Chloride might also be important (even 1% would significantly alter any correction) and therefore we have followed the suggestion of the reviewer and removed our earlier correction. The dryness of the sampling period means that this might stand as a source of error for some readers but we hope that the updated text
- 145 and argument will convince readers that the cyclic correction is likely smaller than the effect of landsliding that we investigate. We have added & altered text to reflect this choice. Lines 171-187: "3.2. Cyclic Input
- Solute input from atmospheric sources can be significant in rivers and streams, and is routinely corrected for in many weathering studies. Previous work on the average chemistry of rainfall in different parts of Taiwan, has found that it is
 highly dependent on location (e.g. higher pollutant content near the densely populated west coast (Wai et al. 2008) and intensity of rainfall (typhoon rainfall boasts the highest solute concentrations in some cases (Cheng and You 2010)). Using any of these measurements to correct our samples based on the assumption that all present chloride derives from cyclic input (following e.g. Calmels et al. 2011) resulted in clearly erroneous estimates of the proportion of cyclic salts
- in the water samples (e.g., >100% of dissolved K⁺ and SO₄²⁻ from cyclic sources). Notably, chloride is a minor
 component in all samples, with a median value of 32 µmol/l (minimum 14.6, maximum 175 µmol/l). This is a similar range of values to rainwater measured in other coastal environments (Galloway et al. 1982; Nichol, Harvey, and Boyd 1997), and the overall correction of TDS is at most 10% even with the most concentrated rainfall we have found in published literature (spot samples measured by Calmels et al. (2011) in the Taroko gorge). We note that deep groundwater may also introduce chloride, which could lead to overestimates of cyclic input. Moreover, during the dry
- 160 sampling conditions, excess evaporation could lead to larger proportions of cyclic solutes. In view of the minor amounts of chloride in our samples and because of the caveats mentioned above, we elect not to correct our samples for cyclic input. In the discussion, we assess whether this simplifying assumption is valid."

Line 173 The authors are missing a citation here. Li et al. (2014; G3) also addressed the issuesassociated with the automatic mapping of landslides.

We have added this reference; the Marc and Hovius (2015) paper explicitly addresses the effect of amalgamation, but this is also part of the Li et al. (2014) work.

- 170 ### Line 191 While it is true that the oxidation state of Si is +4 in natural waters, I don't think this qualifies it as a cation. For example, most dissolved sulfur also has a positive oxidation state (+6), but the authors correctly label it as an anion because it is speciated as sulfate in most natural waters. Typically, Si is speciated as silicic acid in natural waters and, because the pKa of silicic acid is high, it is fully protonated at the pH values observed in most rivers. So, maybe it is best to call Si a neutral species.

Both reviewer 1 and reviewer 3 have correctly pointed out our mis-description of Silicon as a Cation. We have changed this throughout the manuscript.

- 180 ### Line 200 The authors state that solute concentrations in their Taiwanese streams are high relative to global rivers, but provide neither an average value nor a citation for the typical concentrations of solutes in global rivers. Maybe the using values from the compilation of Gaillardet et al. (1999 Chem. Geo.) would be useful here.
- 185 The reviewer correctly points out that this claim is unsupported. We include both the suggested citation as well as West et al (2005) who measured mountain streams.

Line 217 (and elsewhere) What is "weathering efficiency"? My interpretation would be that

weathering efficiency is the same as what other authors call "weathering intensity", which is the

190 mass of rock solubilized by weathering divided by the mass of rock exposed by uplift. If this is what the authors mean by weathering efficiency, then I do not understand how differences in solute concentrations between streams and landslide seepage provide a useful constraint on it. Steams presumably average over different timescales and have different mass fluxes compared to landslide seepage, which could both obscure inferences about the proportion of rock solubilized. If the authors mean something else, they should perhaps define this term in the text.

The reviewer is correct that we have used weathering efficiency in the same vein as weathering intensity, and we acknowledge that our lack of solute flux measurements make this an unsubstantiated point. We have corrected this throughout, removing any use of the term efficiency.

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Line 224 The authors argue that the effects of landslide area not evident in their data by reporting R2 values. First, what model are the authors using to describe this relationship? Using an incorrect model could lead to a poor R2 despite there being an underlying relationship. Overall, this would by much easier to assess if the authors showed us a scatter plot of landslide area and TDS.

205 After making one myself, its pretty clear that there is not an obvious correlation. Additionally, I am a bit confused how the authors get negative R2 values. Unless the authors are calculating some sort of adjusted R2 and just not telling us, they shouldn't be able to get negative values.

We have added in a figure (fig 2) showing the relationship between the mapped landslide size and

210 the TDS in seepage from the deposit. In terms of the 'model' – we have clarified our phrasing to refer directly to the correlation coefficient between area and TDS and volume and TDS. Lines 308-310: "In the Taimali catchment, there is no strong correlation between landslide size and TDS; R² values for correlation between TDS and landslide area (Figure 2), and TDS and landslide volume are 0.11 and 0.09 respectively (p = 0.62, p = 0.70, respectively)."

215 We have corrected the R2 values.We have also included a discussion of how volumes were calculated.

Lines 221-225: "From the mapped landslide areas, we also estimate the volumes of individual slides, using global area-volume relationships (Larsen et al. 2010). We lack direct measurements of the depth to which the landslides we sampled have scoured. Errors in the mapped landslide area are assumed to be <20%, which are propogated into the

220 total volume estimated, as well as the catchment-wide volume estimates. This follows the approach described in (Emberson et al. 2015), which allows direct comparison of the estimated landslide volumes in Taiwan and the WSA (Figure 4)."

225 ### Line 226

The authors report a correlation between sulfate and TDS and argue that this is evidence for the effects of sulfide mineral oxidation. Such a correlation could arise for any number of reasons and is observed globally (e.g., the data of Gaillardet et al.1999 Chem. Geo. show this relationship). In general, solute concentrations are all negatively correlated with discharge, so variations in

230 discharge alone could produce a correlation between sulfate and TDS. Similarly, solute concentrations should scale with water/rock interaction time, which could also give you a correlation between TDS and sulfate. Dilution by rainfall, evaporative concentration, and end-member mixing could all also give you a relationship between sulfate and TDS. And finally, as the

authors acknowledge, the dissolution of evaporites could also be responsible for this correlation.

- 235 To me, the fact that the catchment is underlain by marine shales is a strong argument for sulfate being sourced from sulfide mineral oxidation. Personally, I do not see the additional insight gained from reporting a correlation between TDS and sulfate concentrations, which is expected (and observed) in most river systems independent of sulfide mineral oxidation. Personally, I would find a scatter plot comparing elemental ratios much more convincing if the authors are
- 240 trying to make the point that sulfide mineral oxidation coupled to carbonate dissolution is the dominant chemical reaction occurring within the landslide deposits. For example, SO4 / (Na+K+Ca+Mg) versus Ca / (Na+K+Ca+Mg) would show what proportion of the cationic and anionic budgets are sourced from the weathering products of coupled sulfide mineral oxidation and carbonate dissolution.
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The reviewer is correct to point out that correlation between these two variables could result from effects other than greater landslide input. We have replaced the Ca vs SO4 figure with two plots of Ca/(Na+K+Ca+Mg) vs SO4/(Na+K+Ca+Mg) and Na/(Na+K+Ca+Mg) vs SO4/(Na+K+Ca+Mg), which demonstrate that the increase in Calcium with respect to increasing Sulphate is not an

- 250 artefact of dilution or mixing. The fall in Sodium further reflects this. Looking at the original TDS-SO4 plot in detail, the slope of the relationship is close to 2:1, which could well represent either dilution or mixing of sources. As such, we remove this figure, and have rephrased the text to emphasise the importance of the chemical ratios.
- Lines 318-325: "However, the ratio of calcium to the sum of other cations (sodium, potassium, magnesium and calcium) is correlated with the ratio of dissolved sulphate to the same cations (Figure 3A; $R^2 = 0.64$, p < 0.005), which suggests a change in weathering regime. A decrease in the fraction of sodium to other cations also correlates with the increasing fraction of sulphate (Figure 3B, $R^2 = 0.63$, p < 0.005)."
- ### Line 235 The authors attribute the fact that solute concentrations are independent of landslide
 area to a heterogenous distribution of pyrite. Since landslide areas and volumes vary by 3 to 4 orders of magnitude respectively, does this imply that bedrock pyrite concentrations also vary by 3 to 4 orders of magnitude? Alternatively, could it be that bedrock pyrite concentrations are more or less the same, but water fluxes co-varies with landslide area so that water/rock ratios are approximately constant? Or, what if sulfide mineral oxidation is limited by the diffusion of oxygen into the landslides? In this scenario, the lack of correlation between landslide area and sulfate concentration arises from the fact that pyrite in deep landslide deposits is essentially inaccessible to weathering. I suppose that this is what the authors mean by "the degree of its exposure for oxidation", but it might be useful to develop this idea further.
- We agree with the reviewer. It is highly unlikely that the distribution of sulphides varies over several orders of magnitude. The reviewer helps develop an idea which we briefly explored in Emberson et al. NatGeo 2016, that the weathering boost may be limited in very large (deep landslides). In very large Taiwanese landslides, oxidation may be limited; although we stress that size is unlikely to be the only controlling factor on the diffusion of oxygen into deposits. The internal hydrology is likely to be just as key, but we have no constraint on this.

Lines 337-340: "The area and volume of landslides varies over 3 and 4 orders of magnitude respectively; it is unlikely that the distribution of sulphides in the rock mass varies as much over short distance. Instead we suggest that the absence of correlation between size and solute concentration is likely indicative of a limit to diffusion of oxygen in landslides, either linked to the size or the unconstrained internal hydrology."

Line 241 The say that landslides "remove the supply limit" on weathering. Conceptually, this does not make sense to me. To me, supply-limited weathering refers to conditions where all weatherable-material exposed by uplift is dissolved before being exported by erosion. This contrasts with kinetically-limited weathering, where un-weathered material is exported by erosion due to

material residence times being shorter than the time required for complete reaction. Ferrier et al. (2016 G3) have a good discussion about these definitions. Based on these definitions, landslide weathering can be either supply-limited or kinetically-limited depending on how quickly landslide deposits are transported to the river channel and exported from the catchment. Since the authors do not constrain the residence time of landslides deposits with their study catchment, I am not sure
how they are able to discuss supply- versus kinetically-limited weathering in landslide deposits.

The reviewer correctly points out that true 'supply limited weathering' would be the situation where all material is weathered before erosive export can occur. However, for the most reactive minerals, the amount of time for which they must be stored in the weathering zone to be completely dissolved is much less than the least reactive minerals. In a fresh landslide deposit, none of the minerals are

- 295 is much less than the least reactive minerals. In a fresh landslide deposit, none of the minerals are depleted if they have been sourced from below the oxidation fronts; this means that weathering in the deposits is (at least initially) not limited by the absence of one or other mineral phase. However, our use of the term 'supply limit' invokes connotations of a more general, rather than mineralspecific, model, which was not our intention. We have removed the previous discussion of 'supply
- 300 *limits' to address this.*

Line 246 Silicon, and to a lesser extent Ca, are not conservative in natural waters (e.g., Garrels and Mackenzie 1967, Jacobson et al. 2002 GCA, and many more), so I do not necessarily see how

- 305 Si/Ca can be used as a reliable proxy for the proportion of cations sourced from silicate weathering. Furthermore, all of the reported data have elevated Na to Si ratios relative to common silicate minerals (e.g., albite), which is consistent with Si loss due to secondary silicate mineral precipitation (i.e. non-conservative Si behavior).
- 310 We observed significant precipitation of secondary calcite in the field in association with seepage from deposits, so we are well aware that Calcium is certainly not conservative. A full evaluation of this is well beyond the scope of this paper (in fact it forms part of a much longer manuscript we are currently preparing that investigates the CO_2 effect of landsliding), but the reviewer is correct in noting this. We don't have evidence of non-conservative Si behaviour other than the elevated ratios,
- but to account for this we have edited our discussion to focus on the Calcium:Sodium ratios rather than Silicon. As such, we use the ratio of Sodium to total Cations.
 Lines 362-365: "the lower proportion of other purely silicate derived cations and silicic acid in landslide seepage (e.g. Na⁺:Ca²⁺ ratio always <0.5, compared to the silicate mineral value of 2.85 (Calmels et al. 2011)), as well as the markedly different behaviour of sodium and calcium (Figure 3A and 3B) supports the interpretation that weathering in the sampled landslides is dominated by the dissolution of carbonates."

We have removed the cyclic correction at the suggestion of the reviewer, which could lead to erroneous sodium in the measurements, but sodium in seepage is uncorrelated with chloride, suggesting this is not the case.

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Line 255 While I think I agree with the authors, I find the wording in this section unclear. If I understand correctly, the authors are trying to argue that sulfate is sourced predominantly from in-

situ sulfide mineral oxidation as opposed to geothermal inputs. If one assumes that their single sample of geothermal water is representative of all geothermal inputs, then seepage Li

- 330 concentrations can be used to place an upper bound on potential sulfate contributions from geothermal waters. Assuming all Li is sourced from geothermal waters, the observed Li enrichments in some of the seepage samples would be insufficient to account for a bulk of the sulfate budget due to the low SO4/Li ratio measured is in the single geothermal sample. So, to me, Li concentrations not "preclude" geothermal inputs, but instead constrain their contributions to the sulfate budget.
- 335 sulfate budget.

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The reviewer is correct. Unfortunately we have not been able to sample geothermal water in other locations in the Taimali catchment, so have to rely on the single sample we obtained. We have changed the text to address the comment – the geothermal sample constrains the input of geothermally sourced sulphate to the overall sulphate budget, rather than the overall geothermal input.

Line 368-379: "Previous studies in Taiwan (Calmels et al. 2011) have shown that Sulphate is a key component in deep groundwater (sampled >100m below the surface), but clearly not all pyrite is oxidised at these depths. Groundwater circulating in the bedrock topography is unlikely to be the source of sulphate in seepage. Such water circulating in the bedrock topography is unlikely to be the source of sulphate in seepage. Such water circulating in the bedrock topography is unlikely to be the source of sulphate in seepage.

- 345 would not systematically surface at landslide sites, given the highly variable morphologic characteristics (size, scar/deposit ratio, position with respect to ridge crest and valley floor, and proximity to major faults) of individual landslides. The same argument applies to geothermal outflow. Moreover, hot spring waters sampled in the Taimali catchment have extremely high Lithium concentrations, 1610 μmol/l, and the lack of elevated Li⁺ concentrations in the landslide seepage samples suggests the elevated sulphate in seepage derives from in-situ weathering. Other studies
- 350 have found high concentrations of silicate-derived cations in deeper groundwater (Calmels et al. 2011); we do not observe any strong increase in the proportion of cations exclusively sourced from silicate minerals (particularly sodium figure 3B) and sulphate in landslide seepage."

Line 258 I suppose this comment is similar to the one above. If one assumes that potential groundwater inputs would be characterized by high concentrations of both sodium and sulfate, then the limited co-variation between sodium and sulfate concentrations would suggest that variations in sulfate concentrations are not due to mixing with groundwater. While this seems like a reasonable argument, it'd be better supported with some scatterplots showing actual mixing relationships based on elemental ratios (not concentrations). The authors could even use literature data to better 360 constrain some of the end-members.

360 constrain some of the end-members.

We have taken the suggestion of the reviewer and introduced a scatterplot of SO4/(Ca+Mg+K+Na)vs Na/(Ca+Mg+K+Na) to demonstrate the negative correlation between the two in seepage samples.

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Line 270 Does it matter that you are comparing solute concentrations from catchments with different surface areas?

We suggest that since all of the streams are saturated with respect to calcite (likely the key
dissolving phase in the coupled sulphuric acid-carbonate weathering) the difference in size of catchment is likely of secondary importance. We also find no correlation between the catchment area and the concentrations of any of the dissolved species.

Line 276 If sulfide oxidation is limited by O2 diffusion, the weathering of silicates could be de-

375 coupled from the oxidation of sulfides. In this case, a correlation between Na and sulfate would not be expected. And, as stated above, variations in Si concentrations are strongly influenced by secondary mineral precipitation which affects neither Na nor sulfate.

We acknowledge that the inclusion of Silicon in this analysis made assumptions about the
conservative nature of Silicon in these waters, and have removed it. A decoupling of sulphide
oxidation from silicate weathering may be possible, but we suggest that this requires invoking both
oxygenated and deoxygenated (or at least without sulphate) hydrological pathways. This may well
be the case, with landslides providing an oxygenation window for sulphides in catchments, and
silicate weathering occurring elsewhere. We have changed the text to avoid confusion about what

- 385 we mean. Lines 379-383: "Extremely elevated Na^+ and Mg^{2+} concentrations at some sites are not clearly associated with any other measured parameter, or with each other, and we have no ready explanation for these isolated observations at present. Since these high concentrations are not linked to dissolved sulphate, they may be linked to flowpaths with limited oxygen where sulphate oxidation cannot proceed, but we do not have a constraint on this."
- 390 This is also helped by defining the section in question as a results section, as suggested by reviewer 2 (see below).

Lines 149-153: "3. Methods In this section, we report the analytical techniques used to obtain the data in section 4 summarise these results. In later discussion, we compare measurements of both chemical concentrations and ratios with physical data about landslides and catchments derived from remote sensing. In general, we focus on qualitative links between these parameters, using statistical tests (R^2 coefficient of determination, and p-value to test significance of relationships) to support suggestions that parameters are connected."

Line 305 The authors state that the maximum solute concentrations observed in the landslides are 15 to 20 mM. While I don't disagree with this, I am not sure how this observation alone

- 400 provides insight into the "saturation" concentration of landslide weathering. While there probably is a maximum possible concentration, it need not be the highest concentration observed in this dataset. Also, saturation might be a poor word choice in this context since it has thermodynamic implications.
- We have rephrased this to remove saturation. The reviewer correctly points out that even if we haven't found higher concentrations, they may still exist.
 Lines 431-435: "The peak TDS values we measured in seepage are between 15000-20000µmol/l. Other locations may have greater concentrations, if local weathering has greater oxidation of sulphides, either due to more efficient distribution or greater diffusion of oxygen at depth in the landslide deposit. Such hypothetical, very high
 concentrations, in excess of what we have recorded, would serve as an upper limit to solute concentrations in the entire

Line 319 The authors state that the amount of landsliding in each catchment is not correlated with the catchment slope. Where does this observation from from?

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catchment."

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ArcGIS analysis of ASTER GDEM V2 (post Morakot) data provided the modal slope of each of the subcatchments. We have included this data in the table and included a description of the analysis in the text. Lines 226-227: "In addition, we have quantified the area, modal slope, and the mean and range in elevation of each catchment in an ArcGIS environment, analysing ASTER GDEM V2 data provided by NASA. These data are reported in table 2."

^{###} Line 331 Lee et al. (2015 Env. Sci. Poll. R.) also report concentration-discharge relationships for Taiwanese rivers.

425 *We were not aware of this paper, and we thank the reviewer for the suggestion. It has been included in the text.*

Line 336 Tipper et al (2006 GCA) report solute concentration-discharge relationship for Himalayan rivers and Torres et al. (2015 GCA) specifically link mountain catchments with "chemostasis" in the Andes/Amazon.

We have included these two references. We thank the reviewer for the suggestions.

Line 345 Again, I am not sure how a correlation between TDS and sulfate can be used to argue
that solutes in stream water are sourced predominantly from landslides. This correlation is not
unique to landslides and could arise from many different processes.

The reviewer is correct that in isolation this correlation can arise for other reasons. However, in these rivers that are saturated with respect to calcite, the effect of dilution is limited (especially at the dry conditions we sampled). This is supported by the comparison with sodium concentrations (Fig 5B). Sodium in streams does increase to some extent with chloride, suggesting a cyclic input. However, this is not correlated with the increasing sulphate concentrations. Figure 4C, showing increasing dissolved SO₄²⁻ with increasing volume of landslides, supports our point that streams are mirroring the landslides. In these rivers the variability in TDS is not readily explained by catchment size, or other topographic variables. We feel it is important to point out that the sulphide oxidation

not only affects the ratios of elements but also the overall concentrations.

Line 335 The statement "supersaturated everywhere with respect to Ca" doesn't make sense. Waters cannot be supersaturated with respect to an element. They can, however, be supersaturated with respect to a mineral phase.

We incorrectly referred to Calcium here. It has been changed to Calcite here and elsewhere.

Line 359 The authors define the term "non-equilibrium weathering" to describe landslide
weathering because "the depletion rates of the various minerals involved are likely to be very different." I have a few issues with this statement. First, the fact that weathering occurs in the first place is because primary minerals are out of equilibrium with respect to surface conditions. So, by definition, all weathering is non-equilibrium. Second, chemical weathering in any setting will involve minerals that react at different rates. That said, I agree that landslide weathering is somehow
"different". To me, the important difference is that landslides rapidly transport weatherable material from depth to the surface. In contrast, "hillslope" weathering involves the much slower transport of material from depth. Rapid transport of material from depth allows minerals that typically have deep weathering fronts (e.g., pyrite) to be exposed to weathering at the surface.

465 Our use of the term 'non-equilibrium' was perhaps overeager. We have altered the text to remove this phrase and replace it with a point emphasising that the weathering of all minerals occurs in 3 dimensions in deposits rather than at specific reaction fronts at depth in stable slopes. Line 547-549: "The concept of a reaction front for weathering is less applicable to these settings, as the distribution of reactions will be controlled by the localisation of detrital sulphides and the flow paths providing oxygenated fluid."

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Line 393 There definitely needs to be some citations for the links between weathering and atmospheric pCO2. Many of the papers cited elsewhere in the text should also be cited here. However, the authors could also consider citing:

- 475 Lerman, A., Wu, L., & Mackenzie, F. T. (2007). CO2 and H2SO4 consumption in weathering and material transport to the ocean, and their role in the global carbon balance. Marine Chemistry, 106(1-2), 326–350. http://doi.org/10.1016/j.marchem.2006.04.004
- Torres, M. A., West, A. J., Clark, K. E., Paris, G., Bouchez, J., Ponton C., Fakirs S.J., Galy, V.,
 Adkins, J. F. (2016). The acid and alkalinity budgets of weathering in the Andes-Amazon system: Insights into the erosional control of global biogeochemical cycles, Earth Planet. Sci. Lett. http://doi.org/10.1016/j.epsl.2016.06.012

We thank the reviewer for the extra citations; the Torres et al. 2016 paper came out while this
manuscript was in review. We have incorporated these and the Galy and France-Lanord (1999)
reference used elsewhere in the paper. We mistakenly assumed that the pCO₂ link to weathering
would be clear to the general reader and not need citation.

Line 400 The authors mention a "feedback between drawdown of CO2 and erosion." Since
erosion rates are not necessarily modulated by atmospheric pCO2, I do not think this can be
accurately called a feedback. Additionally, the authors should probably cite some of Maureen
Raymo's work on this topic.

We agree with the reviewer; this is not a feedback (at least not a direct one). We adjusted the text,
and have also cited the work of Raymo.
Line 590-592: "As erosion rates increase and mass-wasting through bedrock landsliding begins to dominate

erosional budgets, the erosional impact on CO₂ and climate, achieved through sustained weathering of silicates (Berner and Kothavala 2001; Brady 1991, Raymo and Ruddiman 1992), will weaken."

500 ### Line 404 Should probably include the relevant citations for the links between sulfide mineral oxidation and erosion rates.

We have added in citations for Calmels et al (2007) as well as Torres et al. 2016, to address this.

- 505 ### Line 417 The authors state that the area affected by Typhoon Morakot is now a CO2 source but provide no evidence for this. The affect of weathering on atmospheric pCO2 depends on the precise balance between acid producing and acid consuming reactions (e.g., Torres et al. 2016 EPSL), which was not appropriately addressed in this manuscript.
- 510 This paragraph was discussed by all three reviewers. It has been extensively reworked to reflect these comments; the statement 'the area affected... is now a CO2 source' is unsubstantiated within this dataset. As mentioned above, we are currently finalising a manuscript dealing with this, and there was mistakenly some overlap between the trains of thought that went into the two separate pieces of work. As such, we now discuss the importance of quantifying substrate weathered and the
- 515 acids at work to determine the CO2 output, but only suggest that it is possible that the area is now a source of CO2.

Lines 622-629: "As a result, the area most affected by erosion due typhoon Morakot could be a net source of CO_2 to the atmosphere. The weathering boost is likely to end before the next major meteorological perturbation of southern Taiwan, so that the weathering of carbonates and silicates with Carbonic acid must be taken into account when considering the longer-term effects of erosion in South Taiwan. Future work to quantify the net sequestration or release of CO_2 would require a closer evaluation of the ratios of acid at work, as well as correction for the extensive secondary

525 *###* Line 448 It might be worth citing some of the other work on carbonate versus silicate weathering in New Zealand. For example,

Jacobson, A. D., Blum, J. D., Chamberlain, C. P., Craw, D., & Koons, P. O. (2003). Climatic and tectonic controls on chemical weathering in the New Zealand Southern Alps. Geochimica et Cosmochimica Acta, 67(1), 29–46.

Moore, J., Jacobson, A. D., Holmden, C., & Craw, D. (2013). Tracking the relationship between mountain uplift, Silicate weathering, And long-term CO2 consumption with Ca isotopes: Southern Alps, New Zealand. Chemical Geology, 341, 110–127.

535 http://doi.org/10.1016/j.chemgeo.2013.01.005

precipitation evident in the catchment."

The reviewer correctly notes the lack of citation here. We have incorporated the first of the suggested citations.

540 ### Line 455 Again, stochastic landsliding effects were previously incorporated into a weatheringerosion model by Gabet (2007 EPSL). This work should be mentioned and discussed here.

As with the introduction, we have incorporated the Gabet (2007) paper into the discussion.

- 545 ### Line 464 Again, the link between erosion and weathering the authors describe is not necessarily a feedback. The rates of tectonic uplift do not necessarily depend upon pCO2, so changes in weathering would not necessarily feedback on erosion rates. While I am aware that there are some possible weathering-erosion feedbacks (e.g., through rock strength and/or glaciation), I do no think that the authors are referring to these in this manuscript.
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Here again we agree with the reviewer in general that this is not a direct feedback – we have clarified to incorporate the effect of climate on tropical storm intensity, which is what we were alluding to.

Lines 675-679: "In this light, we draw attention to the possibility that disproportionate dissolution of highly labile mineral phases such as pyrite or carbonate as a result of landslide driven weathering may reduce the strength of the link between erosion and atmospheric CO_2 drawdown via silicate weathering in fast eroding settings, which in turn would modulate any feedback on erosion through changes in tropical storm intensity (e.g. Knutson et al. 2010). The role of active mountain belts in the global Carbon cycle remains ambiguous."

560

Table 1. Was DIC measured directly? If so, how? If this column is actually charge balance, then it should be labeled as such. Charge balance does not necessarily equal DIC because of species like carbonic acid and organic bases. Also, it seems curious that sample TWS15-91 has no anions other than Cl and SO4 yet has a pH of 8.19. I tried to re-produce this pH with the reported solute

565 concentrations using PHREEQC and couldn't get it to work. That said, I did not try very hard.

Anyways, it might be worth looking into that sample a bit more to make sure there are not any analytical issues.

DIC was assumed to be just HCO3- (at these pH conditions we feel this is a fair assumption) which
was calculated by charge balance. This is mentioned at the end of the methods section but we have clarified this and included this in the Table as well.

Having looked in more detail at the sample TWS15-91, there was indeed a problem with the analysis. I (RE) missed the fact that this sample was over the calibrated range in Calcium on the initial OES run, and as such it was not rerun at a higher dilution. We have excluded it from the final analysis and manuscript.

We thanks Reviewer 1 for an extremely thorough and insightful review.

580 Anonymous Referee #2

Received and published: 5 July 2016 Emberson et al., Oxidation of sulphides and rapid weathering in recent landslides. Earth Surface Dynamics.

Determining how erosion and weathering are related to one another in tectonically active mountains

- 585 is important for understanding how mountain uplift influences geochemical cycling. Here Emberson et al. present geochemical data from surface waters in Taiwan. They focus on the chemistry of stream waters as well as waters that have flowed through landslide debris, deposited by extensive hillslope failures associated with Typhoon Morakot. The pyrite and carbonate bearing marine sedimentary rocks in the study area offer a mineralogical contrast to a complimentary study the
- 590 group conducted in the Southern Alps of New Zealand, where the mineralogy of the bedrock differs. Emberson et al. conclude that sulfuric acid generated by pyrite oxidation drives the dissolution of carbonate rock within the fractured landslide mass and that, above a threshold minimum of landsliding, that total dissolved solids, Ca, and sulfate increase with increasing catchment landslide volume. These results provide new, process-level insight on the generation of
- 595 solutes in tectonically-active landscapes, and will be of broad interest and is an appropriate contribution to Earth Surface Dynamics. I believe the manuscript can be accepted for publication following revision to address questions regarding several aspects of the paper.

We thank the reviewer for taking the time to review our work. The comments have greatly aided its development.

Main comments: This manuscript lacks a distinct Results section, nor is the statistical treatment of the data clearly described in the Methods section, such that the reader does not know how the various dataset are analyzed with respect to one another until the statistics are presented as part of the Discussion. A revised manuscript without an explicit Results section could work, but if so, additional description of the Methods are needed to better link the different sections of the paper

and prepare the reader for the Discussion.

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615

We acknowledge that the methods and results sections are not clearly defined in the earlier version.
610 We have retitled our sections to better separate the results from discussion. We have noted how we have structured the results and discussion sections in the methods section.

Lines 146-153: "3. Methods In this section, we report the analytical techniques used to obtain the data in section 4 summarise these results. In later discussion, we compare measurements of both chemical concentrations and ratios with physical data about landslides and catchments derived from remote sensing. In general, we focus on qualitative links between these parameters, using statistical tests (R^2 coefficient of determination, and p-value to test significance of relationships) to support suggestions that parameters are connected."

Line 318-319. It is worthwhile to present data that demonstrate that the magnitude of landsliding (and TDS) does not scale with topographic metrics such as slope or relief. The reason being is that erosion rates (and hence coupled weathering rates) scale with catchment topography. Hence it might be expected that, even in the absence of any landsliding, the range of TDS values could be explained by topographic variation of the watersheds. For the arguments presented in the paper, it is important to show this is not the case.

625 The reviewer correctly signals that the topographic aspects of the catchments in question are missing, and we have fixed this by adding the modal slope and the relief of each of the catchments

(Table 2). The TDS values in the streams do not vary with any of these factors, and while we have not added a figure to demonstrate this lack of correlation, we discuss it in the text. Lines 454-456: "Moreover, neither modal slope nor the range of elevation in a catchment is correlated with the measured TDS, suggesting these factors do not strongly control the weathering in this setting."

Line 406-414. It would be worthwhile to elaborate and make the logic very clear here, as this paragraph is laying out a conceptual framework, based on the observations, for understanding the role landslides play in the weathering budget of mountains. In particular, more explanation of the role of sediment retention time would be useful. The instantaneous weathering flux from the 635 landslide debris should decay as a function of time, whereas the cumulative flux of weathering products from landslide debris will increase with time. Hence there should be more explanation to support the claim that long sediment residence times would have a limited effect on CO2 budgets-if this indeed is the case, which seems unlikely. The return interval of landslide-triggering events should also play a role, as this ultimately sets the volume per time of landslide debris subjected to 640 sulfuric acid weathering. A conceptual diagram showing the roles of residence time and return interval would be a useful approach to organize these thoughts in this paragraph. Additionally, there has been at least some work on 14C dating of alluvial deposits in southern Taiwan, presumably which record valley alluviation due to landsliding in some cases. Although these studies may not be 645 in the same catchment as was investigated here, these studies could be drawn upon to estimate to a

first order the length of time landslide derived sediment is likely to persist in the watershed.

Hsieh, Meng-Long, and Shyh-Jeng Chyi. "Late Quaternary mass-wasting records and formation of fan terraces in the Chen-yeo-lan and Lao-nung catchments, central southern Taiwan." Quaternary Science Reviews 29.11 (2010): 1399-1418.

Hsieh, Meng-Long, and Peter LK Knuepfer. "Synchroneity and morphology of Holocene river terraces in the southern Western Foothills, Taiwan: A guide to interpreting and correlating erosional river terraces across growing anticlines." GSA Special Paper (2002): 55-74.

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This is a really important point and one we want to address in detail. Material that is retained in the catchment will have a cumulative effect on weathering – as pointed out by the reviewer. However, if the labile minerals are depleted quickly, they will no longer impact the cumulative weathering after their depletion time. Silicate weathering can still occur in these deposits, and

660 therefore affect CO2, but the key physical effect of landsliding is to excavate the reactive phases. It is this landslide-driven effect that we were initially referring to, and we have tried to clarify our argument.

The terraces that are discussed in the two cited papers are sites we have visited; in an earlier version of this manuscript we had hoped to include the chemistry of seepage from these terraces as

665 a long-lived version of the landslides, but the lithological differences between the terraces and landslides in the Taimali make a direct comparison difficult and the structure of the argument unwieldy.

As such, we have rewritten this paragraph to better summarise the key points. We hope that it is now clearer. The storage time of the material in catchments should have been included in the first place and we have incorporated both of the references to fix this.

Lines 564-579: "A second time scale of relevance is that of the physical removal of landslide debris from the catchment. If debris remains in a catchment after depletion of exposed pyrite, then its weathering will revert to mineral

reactions with Carbonic Acid. If the time to depletion is greater than the time required for removal of the debris, then the labile phases it contains will remain unweathered and will be removed through sediment transport processes. In the

- case of Taiwan, some samples of river bed material show Sulphur content of up to 1.07% (Hilton et al. 2014), 675 suggesting that weathering in the landscape does not purge all sulphides brought to the surface by rock mass exhumation. In the opposite case the weathering from a given landslide will return to a constant background rate shared with other colluvium and soil covered parts of the landscape. How this scales up to the catchment scale depends on the return time of large drivers of mass-wasting, such as typhoons and earthquakes, and the subsequent advection of
- 680 debris into the fluvial network and the transport capacity of this network. A disparity between short term production of sediment on hillslopes and the capacity for onward transport can lead to up to millenial residence times of eventproduced sediment in mountain landscapes (Blöthe and Korup 2013). In the Taimali catchment, many headwater streams were still clogged with sediment 6 years after typhoon Morakot, and large colluvial fans had formed below several landslide-dominated tributaries. Elsewhere in Taiwan, mountain valleys contain large colluvial terraces, which

have formed hundreds to thousands of years ago (Hsieh and Chyi 2010). This implies that time scales of pyrite 685 depletion and of debris removal must both be considered in further explorations of the links between erosion and weathering in Taiwan."

- 690 Lines 415-420. Here it would be useful to point the way forward in terms of what data are needed to determine the sign of the CO2 budget in a rapidly eroding mountain range. There are already elements of this in the paragraph, but a thorough explanation of what suite of measurements are needed would be a forward looking way to wrap up the manuscript. Additionally, with the understandably limited data that are available, is there at least a signal of what process is
- 695 dominating the CO2 budget, sulfuric acid weathering of carbonate, carbonic acid weathering of silicates, or organic carbon burial? For example, back of the envelope calculations assuming regional-scale runoff values to infer fluxes for this study, and comparison with previous work on organic carbon export in Taiwan may be useful, even if imprecise. Even if a very first order budget cannot be calculated, due to lack of constraints in the values for the various parts of the budget,
- 700 indicating that this is the case is a useful thing to do, as it points to the need for a different strategy in approaching erosion-CO2 links in mountains. On a similar note, the paper begins by asking a question regarding the general influence of landslides on weathering and roles lithology and climate may play (Line 42-44). In concluding the Discussion it would be worthwhile to return to the specific parts of that question and assess the state of knowledge, as informed by this work, and prior
- 705 publications.

715

As mentioned above, we have tried to rework this paragraph to remove the unsubstantiated claims and to explain which data we would need to discuss the CO2 impact quantitatively. We have also used the rewrite to actually answer the question that the reviewer correctly points out that we left

710 unanswered in the earlier version.

> Lines 622-629: "As a result, the area most affected by erosion due typhoon Morakot could be a net source of CO_2 to the atmosphere. The weathering boost is likely to end before the next major meteorological perturbation of southern Taiwan, so that the weathering of carbonates and silicates with Carbonic acid must be taken into account when considering the longer-term effects of erosion in South Taiwan. Future work to quantify the net sequestration or release of CO_2 would require a closer evaluation of the ratios of acid at work, as well as correction for the extensive secondary

precipitation evident in the catchment."

Line 427-429 (also line 241). There is a contradiction in this statement, in that the suggestion is that weathering is kinetically-limited, so long as the involved minerals are 'abundantly exposed'. The 720 'abundant exposure' is suggestive of supply limited weathering, and one interpretation of figure 3 (excluding the smaller landslide volumes in Taiwan), is that the supply of material by landsliding is setting the concentrations of dissolved species and likely also the weathering flux. My suggestion is

- to be very explicit here regarding use of the terms kinetic and supply limited weathering. Pyrite oxidation depends on kinetics (as do all reactions), but when the numbers of minerals involved in 725 such reactions are increased, likely by orders of magnitude due to fracturing of the rock mass during landslide runout, then the supply of minerals plays a very large role in setting the concentrations of various dissolved species measured in the water.
- 730 *Reviewer 2 echoes reviewer 1 in questioning our use of the terms kinetic and supply limited* weathering. As mentioned in response to reviewer 1, we have rewritten the relevant sections to remove all confusion. Both reviewers have correctly understood our message, but more clarity is required for ease of understanding, and we have addressed this.
- See comments above and lines 634-641: "Under these conditions, weathering is limited by the kinetics of the 735 reactions of the most labile mineral phases, but as long as these phases are abundant in the rockmass the rate of weathering is vastly increased. We have found that these labile phases may be minority constituents, driving the weathering in fresh landslide deposits away from the slower process affecting the surface materials elsewhere in the landscape over longer time scales."
- Other comments: Line 14. Insert 'landslide' before seepage 740

Added 'landslide'

Line 18 and elsewhere. Two significant digits on the R2 values is probably sufficient.

745

Removed extraneous digits.

Line 34. Several additional citations are warranted here, including Gabet and Mudd (2009), Ferrier and Kirchner (2008) and West, (2012). Also, Dixon and von Blanckenburg do discuss the role of landsliding.

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See notes above in response to reviewer 1; added in extra citations.

Line 38 and elsewhere. Many citations do not follow a uniform format, with author initials 755 appearing in some cases. Some editing the entries in the reference management software/database that was used here is needed.

We thank the reviewer for their attention to detail! We have corrected this.

760 Line 46. 'local' is ambiguous, perhaps replace with 'landslide'

Local is indeed subjective. Changed it to 'landslide' accordingly.

Line 56. 'importantly' can be omitted in this case and elsewhere in the manuscript. Alternatively, switch the order of 'importantly' and 'propagate' 765

Changed phrase to remove importantly.

Line 79-81. Perhaps break this into two sentences or put parentheses around: either

770 directly from rainfall. crest of landslides.

Sentence removed and changed in response to comment from reviewer 1.

Line 90-91. This statement ought to have a citation to support it.

775

Added citation (Hartmann and Moosdorf 2012).

Line 98. While I have not been to the field site, observations elsewhere in Taiwan would suggest, based on the continuity of vegetation coverage, that a soil (or mobile regolith) mantle, while not necessarily laterally continuous, must cover a large fraction of the bedrock on hillslopes.

We have added a citation in response to the comment of reviewer 1. Reviewer 2 is correct that the vegetation probably indicates more persistent soil than we had initially written. In the field we have struggled to get a good idea of what lies beneath the forest because of its density. Thus, we remove the unsubstantiated assertion.

Lines 111-113: "Soils are generally only >1m in local concavities in the landscape (Lee and Ho 2009), but contiguous, dense vegetation on hillslopes that have not been dissected by landsliding suggests sufficient mobile regolith or soil is present to support flora."

790

785

Line 105. Perhaps change 'in view of this' to 'in light of this'

Changed to 'in light of this'.

795 Line 115. The term 'channel fill' has an ambiguous meaning in this sentence. Does it have the same meaning as 'alluvium'? Some clarification is needed.

Reviewer 2 correctly points out the ambiguity. We meant alluvium, and have changed it as such.

800 Line 165. 'landslides' should be singular

We respectfully disagree here, as we mapped many landslides.

Line 169. Perhaps edit 'as prescribed by' to 'as described in'

805

Changed to 'described in'.

Line 173. A G-cubed paper by Gen Li and colleagues also provides an important example of the effects of amalgamating landslide areas. Li, Gen, et al. "Seismic mountain building: Landslides
associated with the 2008 Wenchuan earthquake in the context of a generalized model for earthquake volume balance." Geochemistry, Geophysics, Geosystems 15.4 (2014): 833-844.

Thanks to reviewers 1 and 2 here for this suggestion. We have included this reference.

815 Line 197. Edit so that 'sit at' is removed

We decided that this sentence was extraneous and it has been removed.

Line 201. The statement that TDS values are high with respect to global concentrations needs a 820 citation.

Added citations in response to comments from reviewer 1 and also this comment. (Gaillardet et al. 1999, West et al. 2005)

825 Line 220-221. Whether deeper landslides will expose proportionally more calcite to weathering relative to shallow landslides will depend on the depth of the calcite weathering front. Whereas much calcite is likely lost from the soil, the soils are likely thin (<1m?) and a small proportion of the total landslide thickness. Given the lithology there may well be considerable calcite in the nearsurface environment in the study sites. Hence the lack of a correlation between TDS and landslide 830 size may not be altogether unexpected.

This is another good argument for the lack of correlation between size and the seepage chemical concentration. We have adjusted our argument to incorporate this point.

Lines 299-305: "In the WSA, TDS concentrations in landslide seepage are strongly impacted by the dissolution of 835 trace calcite, the abundance of which is likely to be greater at depth (Brantley et al. 2013). The depth of a landslide scales with its area (Larsen et al. 2010), implying that the amount of calcite as a proportion of the total landslide volume correlates with the landslide size. This correlation will be limited to the range of landslides where a significant proportion of excavated material is from above the depth to which calcite is depleted. By contrast, in the largest landslides, near-surface materials, poor in calcite, form only a small proportion of the deposit, and a correlation with

840 size my no longer hold."

> Line 223. If reduced hydraulic conductivity at the base of landslide debris, relative to debris that is stratigraphically higher, has been documented this statement needs a citation. Otherwise some clarification is needed.

845

Here we have changed the text to be more exact; larger slides may have reduced diffusion and advection of fluids and oxygen to the deeper parts due to the non-linear area-volume scaling. We cite our previous work as a reference for the linearity of the landslide area to water input as in that work we also calculated how the area of landslides scales with the area that drains into them.

850 Lines 305-307: "Moreover, large landslides may also be subject to reduced diffusion of fluids and oxygen to the base of the deposits; the amount of water entering the slide scales with the area of the slide (Emberson et al. 2015), but the volume increases at a greater rate (Larsen et al. 2010)."

Line 224. Replace the slashes with 'per'.

855

Are correlations reportable as e.g. 'TDS per landslide volume'? We have changed the phrasing to ' R^2 values for correlation between TDS and landslide area, and TDS and landslide volume...'

Line 236. To link this with the statistics above, replace 'size' with 'area or volume'

860

This sentence has been removed, but we have used 'area and volume' in the latest version in accordance with this comment.

Line 241. Avoid starting a sentence with 'This'. Instead explicitly indicate something to the effect of: 'Generating landslide debris from unweathered bedrock removes the supply limit. ..'

We removed this sentence to respond to a comment from reviewer 1.

Line 242. Replace 'and surrounds' with some explicit definition of what is meant by surrounds. The other sites in southern Taiwan that were sampled?

Corrected to 'other sampled sites in Southern Taiwan'

Line 253. Omit importantly. Unless there is evidence the groundwater is 'circulating', simply refer to it as 'deep groundwater'

Removed 'importantly' and 'circulating'.

Line 271. Replace 'tops' with 'exceeds'

880

Replaced.

Line 302. Rainfall is not dependent on area, but runoff is. Clarify as needed.

885 Sentence removed to address comment from reviewer 1. However, we agree with reviewer 2 here, our initial statement was not accurate.

Line 316. '...and these may be a source of variability. . .' It is unclear what 'these' refers to in this sentence and this sentence doesn't really follow from the previous one or set up the following one well. Some editing is needed here.

Edited the sentence – replaced 'these' with 'these factors, such as local slope or hillslope length,...'

Line 321. 'importantly' can be omitted.

895

Removed 'importantly'

Line 329. First two words have unclear meaning.

900 Changed 'Since' to 'As', but we are unclear about what is unclear?

Line 334. Replace 'in keeping' with consistent

Changed to 'consistent'.

905

Line 336. Problems with these references. e.g., should be Lyons et al.

Thanks to the reviewer for pointing this out. Changed accordingly throughout the manuscript.

910 Line 348. A 'likely' should be inserted between 'exhumation' and 'not'. There are no data on the distribution of sulfide minerals within the rock, so you can only hypothesize that this variability also controls the variability in seepage concentrations at individual landslides.

Added 'likely'.

915

Line 357-358. Suggest editing to: '... and the associated generation of sulfuric acid.'

Adjusted to the suggested phrasing.

920 Line 360. Different than what? Please complete this thought.

Sentence removed in response to comment from reviewer 1.

Line 361. Is there a better term than 'minority' minerals – reactive, highly soluble, labile? Choose one and use it consistently throughout. For example, there are likely many phases present in low abundance that do not contribute to non-equilibrium weathering.

The reviewer is correct to point out that minority is an overly general term. We have replaced it where relevant. We have used the words 'reactive' or 'labile' to be as specific as possible. Where the
930 word minority is used it is now explicitly to point out that these reactive phases are often a low proportion of the lithology.

Line 368. Some sort of punctuation to separate the sample numbers and the values of the measurements would be helpful.

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Added punctuation.

Line 376. The statement regarding weathering by carbonic acid dominating in other parts of the landscape should be qualified. It is likely the case, but data are not discussed that support this. Can the data from the rivers be used to support this inference?

Removed statement '[reactions which] dominate elsewhere in the landscape.'. If weathering doesn't happen via sulphuric acid, carbonic acid is likely the key agent of weathering (e.g. Torres et al. 2016, Galy and France-Lanord 1999). However, we have no support for the statement that this dominates elsewhere in the landscape.

Line 378. It is not clear what 'restricted' means in this context. Please clarify.

Replaced 'restricted' with 'some samples of river bed material' – we had meant that we only have one reference for the sulphur content of bed material.

Line 381. There is a comma that can be omitted.

Removed.

955

Line 403. Elaborate on '...a more central role,...' It will be useful to be explicit regarding what the central role refers to.

Altered to 'Weathering of carbonates will form a greater portion of solute budgets'

960

Line 406. Suggest replacing the 'As' with 'when'

Replaced as with when.

965 Line 426. Again, unclear if the waters in the landslide debris indeed circulate.

Replaced 'persistently circulating' with 'infiltrating'

Line 430. Omit 'in the first instance' or replace with another term, such as foremost.

970

Replaced 'in the first instance' with 'primarily'

Line 434. Replace 'units' with 'rocks' and replace 'This' with a more explicit description of the process.

975

Replaced 'units' with 'rocks'. Expanded 'This' to 'This oxidation'.

Line 436. Meaning of 'subsumed' is unclear in this context. Some editing is needed.

980 Changed phrase to 'while the weathering of silicate minerals is not elevated'.

Line 339. Omit 'first' or replace with another term than does not imply a temporal relationship.

Replaced 'first' with 'better correlated'

985

Line 454. 'mineralogical' instead of 'lithological'?

Changed 'minor lithological' to 'reactive mineral'

990 Line 457. Gabet 2007, EPSL is a study that has examined the stochastic role of landslides in weathering.

Added Gabet citation in response to this comment and comment from reviewer 1.

995 Figure captions. 'Total dissolved load (TDS)' – the acronym should match the first letters of each word, e.g., total dissolved solids or TDL.

Replaced Dissolved 'load' with dissolved 'solids'

1000 Figure 1. It would be useful to have an inset map showing Taiwan with a box showing the area of

Figure 1a. This inset could go in the lower right corner of Fig. 1a.

Added inset figure.

1005 Figure 2. There should be at least two labels on the y-axis in each panel to easily allow the reader to see the data range.

Figure 2 has been altered to address comments from reviewer 1, but in doing so we also added the extra y-axis labels.

1010

We thank reviewer 2 for a thoughtful and detailed review.

Anonymous Referee #3

Received and published: 12 July 2016

- The authors argue that landslides place unweathered material at the land surface, exposing pyrite to oxidation which drives weathering. They discuss many implications. The paper was interesting and this phenomenon deserves attention because it is important. I have seen dissolved Fe emanating from beneath landslides in Puerto Rico (and soon thereafter precipitating), right after a landslide, so I know this happens and the central idea is not surprising. Pyrite oxidizes fast when exposed to oxygen and water and especially when bacteria are involved. So that is not that surprising. I think
- 1020 what the authors are trying to do that is interesting is quantify this effect by looking at different watersheds with different densities of landslides. This is an interesting idea. But I don't think they quite nailed down their story. I have a hard time even knowing exactly what they did show, and what this means exactly. I suggest that the authors work hard to probe their data more. With that effort, their results will be more impactful.

1025

We thank the reviewer for taking the time to put together a thorough and careful review of our manuscript. It is encouraging to hear of similar observations in field sites we have not visited! We have not intended to suggest that this process should come as a surprise. What is novel about this study is the empirical data from seepage and the demonstration of the effect on a large scale. Apart

1030 from our work on weathering in landslide deposits in New Zealand (Emberson et al. 2015) we are not aware of other work on this subject.

Part of the problem is that much of the phrasing is a bit vague and confusing. I got tired trying to figure out what they meant. Even the abstract has such phrasing. For example, this phrasing in the abstract is awkward and unclear and should be revised: "Bedrock landslides create conditions for weathering where all mineral phases in a lithology are initially unweathered within landslide deposits, and therefore the most labile phases dominate the weathering at the outset and during a transient period of depletion." I can't really read the abstract and get the main point. Given that I am not surprised about pyrite oxidation being accelerated by landslides. . .what is the big take-home
1040 message that they can show? One interesting aspect that is not explained clearly is why Taimali operates differently than WSA: shouldn't this basic difference be in the abstract?

We apologise for the complex phrasing in parts! Where possible, we have tried to address this, including the sentence in question in the abstract. The big difference between the WSA and the Taimali – the importance of pyrite – has been included.. We are encouraged by the fact that the

reviewer is not surprised by our conclusions; this means that our observations have wider significance. Line 20-23: "The predominance of coupled carbonate-sulphuric acid driven weathering is the key difference between these sites and previously studied landslides in New Zealand (Emberson et al. 2015), but in both settings increasing volumes of landslides drive greater overall solute concentrations in streams."

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I do think the authors should rework their paper before final publication. Here are some points of various importance to consider:

The key question in my mind is the depth to which landslides scour the landscape in comparison
 to the depth of the pyrite depletion zone and the carbonate depletion zone. The authors sort of
 mention this but not fully. Where are the reaction fronts for pyrite and carbonate in these rocks?
 And what is the depth to which landslides scour? Often pyrite is oxidized down to the water table.

The reviewer is exactly correct to pinpoint this relationship; how deep do landslides scour in
comparison to the depth of the pyrite and carbonate depletion zone? The answer is we don't have a good idea. The depth of landsliding does scale with area (Larsen et al. 2010) but this can vary over several orders of magnitude globally. We therefore have only very limited constraint on the depth to which the landslides we measure actually scour. The exception to this is that we only assess bedrock landslides, meaning they excavate at least below the saprolite-bedrock interface. However, in the

- 1065 mountains of Taiwan there exists limited data on the depth of soils, and we are not aware of any systematic study investigating the depth of pyrite or carbonate depletion. The lack of boreholes in the mountains of Taiwan or the Taimali in particular (Taiwan Water Resources Agency Hydrological Yearbook(s) 1990-2012) means we don't have a good sense of the depth of the water table either. The heterogeneity of flow paths in mountain belts (e.g. Andermann et al. 2012 Nature
- 1070 Geoscience, Calmels et al. 2011 EPSL) means that the depth might well vary widely. To address these points, we have expanded the discussion of the scour depth vs depletion depth in the initial discussion (where we reiterate the points above). See lines 241-249, and also lines 299-303.
- 1075 2. Please discuss the calculation of bicarbonate by charge balance. This would usually have a large or reasonably large error just because it is a small difference between larger numbers. But, for example, the calculation could also have error due to neglect of organic anions in the waters, or any other anions. What is the error in bicarbonate? (and it should not be labelled DIC, it should be labelled bicarbonate). I see that the measurements of sulfate and chloride are each +_10%...seems
- 1080 like the calculation of bicarbonate by difference might have big error bars. In that regard, there are most likely too many significant figures on the bicarbonate (and probably some of the other element concentrations?)
- The error in Bicarbonate is given in the table, but we have added it to the text. The calculated
 error the root mean square error of the uncertainties on the other elements is actually small compared to the value of 10% that we use, because the small analytical error on Calcium tends to dominate the overall error. We estimate a 10% error based on previous work (Galy and France-Lanord 1999 GCA) that demonstrated charge-balance estimates are generally within 10% of calculated alkalinity. Lines 165-166: "Bicarbonate (HCO₃) was calculated by charge balance, which has been shown to introduce errors of approximately 10% (Galy and France-Lanord 1999)."

-Naturally, alkalinity is not just HCO3-. We make the simplifying assumption that at the pH values measured, it is the only important Carbonate species (compared to an error of 10%) and that the alkalinity derived from other components is also negligible. Lines 166-168: "We make the simplifying assumption that the Dissolved inorganic carbon (DIC) is formed only of HCO_3^- , which is generally applicable at the range of pH values measured (Zeebe and Wolf-Gladrow 2001)."

3. The calculation of bicarbonate was then used to assess calcite supersaturation. I would think there is a big error in this calculated SI as well.

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- To our knowledge, PHREEQC does not automatically calculate the uncertainties on the saturation indices. We expect that manual calculation of every possible saturation index for the range of chemistry defined by all errors (i.e. each variation of errors for each chemical element) would require an extreme amount of time; we cite previous work (Moore et al. 2013, Chemical

- 1105 *Geology)* where saturation indices are also published without errors attached. Nevertheless, we acknowledge the problems with publishing data without errors attached; any guidance the reviewer could offer as to how to calculate these errors would be welcomed.
 - 4. I wouldn't capitalize element names (Calcium versus calcium).

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Corrected capitalisation where necessary.

5. Generally, I would not call Si "cationic load" although we do usually refer to it as a cation simply because we analyze it with other cations. But it is present as a neutral species unless the pH is very high. For this reason, Si does not contribute to cation balance.

The reviewer echoes reviewer 1 here, and we have adjusted both text and figures to address this issue. Silicon is no longer referred to as a cation, and we have removed it from figure 3 (replaced with sodium).

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6. With respect to Sr: what does this mean? "peaks at 30 umol/L

Replaced 'peaks at' with 'the maximum measured value of which is'

- 1125 7. The paper has a lot of assertions that are really undefended assumptions. Such assertions need to be defended more thoroughly: "the depth of a landslide scales with the area". . . is this always true for all locations? or was Larsen reporting about one locality more than another? I am also not sure about "the proportion of calcite available correlates with the size of the issuing landslide". I think this statement could be true but doesn't have to be true. Are there data corroborating this
- assumption? The authors later state that "the lack of correlation between landslide size and seepage concentration suggests that the distribution of pyrite is highly heterogeneous.." I am not sure I would say the pyrite distribution is heterogeneous but I would argue that in many places there could be a pyrite oxidation front like those identified in Chigira et al. 1990; 1991; Drake et al., 2009; Brantley et al., 2013. I suppose that the base of landsliding in any given area might be somehow
 correlated with the depth to pyrite, but I think it is also very possible that the depth of landsliding
- might not correlate with the depth to pyrite, but I think it is also very possible that the depth of landsliding

We refer back to the comments we made on the first point by reviewer 3; the Larsen dataset is global, but it does vary over several orders of magnitude. In response to both this comment and
earlier comments from reviewer 1 we have adjusted the argumentation in this section, as the reviewers correctly point out that the proportion of carbonate in the deposit will depend on more factors than just the size. Again see lines 241-249, and also lines 299-303.

8. The paper needs some mineral abundance data and element abundance data.

1145

In terms of mineral abundance data, we have incorporated data from Kao et al. (2004) suggesting 0.05-0.27% Sulphur in South Taiwan Sediment sections. Carbonate abundance is estimated to be between 0.28-0.66% (Hilton et al. 2014) although this does not sample the Taimali. Elemental abundance data have been measured by Selveraj and Chen (2006), but we have not included this situation measured by selverated abundance data in the measured by Selverated abundance data in the measured by Selverated abundance data measured by S

1150 *citation as we have not explicitly used elemental abundance data in the manuscript.*

9. The authors start using the phrase "Landslide weathering efficiency" without defining it. What exactly is this?

1155 *Our use of the term efficiency has caused problems. We have addressed this above and it has been removed to avoid confusion.*

10. Please explain why you are using Kendall's tau value?

- 1160 We initially used K-Tau values to estimate the degree of dependence of the variables; this does not make the same assumptions about linearity and Homoscedasticity as the R2 coefficient, but is (in our experience) not as widely used. However, in general the trends which we observe are linear and the assumption of homoscedasticity is qualitatively valid, so we have removed any discussion relating to K-Tau to avoid confusion.
- 1165

11. Line 243, acidity means the base neutralizing capacity of a water. Do the authors mean acidity or protons? (Pyrite is a source of protons.)

The reviewer is correct; acidity is the wrong term to use here. We have changed this to state: 'this
provides a source of sulphuric acid'. Elsewhere we have also changed 'acidity' to 'acid' as appropriate.

12. I suggest the authors include some sort of figure or more text to defend this statement: "the lower proportion of purely silicate derived cations in landslide seepage . . .supports the
1175 interpretation that weathering in the sampled landslides is dominated by the dissolution of carbonates".

The reviewer correctly points out that we have not substantiated this point. We cite previous work in Taiwan (Calmels et al. 2011) for the generally used ratio of calcium to sodium in silicate rock
(0.35), which is well below the ratio in the dissolved load (>>1) that we measure to support the importance of carbonate weathering in landslides. Line 363: "(seepage (e.g. Na⁺:Ca²⁺ ratio always <0.5, compared to the silicate mineral value of 2.85 (Calmels et al. 2011)),"

- 13. I don't understand this: ". . .a significant proportion of the sedimentary pyrite contained within
 the rock mass survives exhumation. .." I think in general, pyrite moves up and out of the system
 like any other mineral until it gets oxidized at some depth related to the rate of erosion and the
 amount of water. Is this is what is meant by survival (death by oxidation)? It is generally oxidized
 down to the water table unless it is a very pyrite rich rock.
- 1190 We broadly agree with the interpretation of the reviewer, but point out that much of the pyrite contained within solid rock mass is inaccessible to circulating groundwater. In general we expect that a fraction of the pyrite will be oxidised in deep groundwater (e.g. Calmels et al. 2011) but some survives to be exploited by landsliding. We have rephrased this sentence to reflect this. The depth at which groundwater was collected by Calmels and coauthors (100s metres below the surface) is not
- 1195 systematically excavated by the landslides we sample (if at all). Lines 368-369, and more importantly 612-614, where we state cite work showing that some pyrite is removed in river

sediment: "Shorter sediment residence times promote the export of unweathered sediment to the ocean and its burial in marine basins. This can result in a fraction of sulphides contained in the rock mass bypassing the weathering window (Hilton et al. 2014),"

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14. The authors argue that Na and Si do not vary with sulfate but Ca and K do vary with sulfate. They argue that this points to multiple pathways relevant for silicate weathering on a catchment scale. I don't see how an observation about landslide seepage can tell us about the other pathways: it can only tell us about the landslide contribution, not the other contributions. I think there are

- 1205 nested reaction fronts. . .deeper flowpaths will interact with parent lithology and shallower flowpaths will not. The authors are arguing that landslides bring some of the deeper material to the surface: landslides are egg beaters. What we need to know is, how deep do they scour relative to where the minerals are?
- 1210 Again the reviewer here stresses the need for information on how deep the landslides scour relative to the reaction fronts. We point out that in such a tectonically dissected mountain belt with extensive faulting and fracturing (e.g. Lin et al. 2008 Engineering Geology) there might well be fluid percolation that is not necessarily 'top down'. This may limit the extent to which 'nested' reaction fronts can persist on a catchment scale. Nevertheless, our phrasing has evidently led to confusion.
- 1215 When we say there are multiple pathways for silicate weathering then we mean that the landslides are not responsible for all of the silicate weathering, but they may have a role to play in some of it. In other words, there is a landslide pathway, and an indeterminate number of other pathways, which we haven't constrained. See paragraphs between lines 359-388.
- 1220 15. I think this is unsupported as written: "the relationship between landslide volume and stream solute concentration will also saturate above a hypothetical maximum concentration"

This sentence was also pointed out to be unsubstantiated by reviewer 1, and we have rephrased it to address both this comment and the comment of reviewer 1. Lines 431-435: "The peak TDS values we
measured in seepage are between 15000-20000µmol/l. Other locations may have greater concentrations, if local weathering has greater oxidation of sulphides, either due to more efficient distribution or greater diffusion of oxygen at depth in the landslide deposit. Such hypothetical, very high concentrations, in excess of what we have recorded, would serve as an upper limit to solute concentrations in the entire catchment."

1230

16. Waters cannot be supersaturated with respect to an element. . .only wrt a mineral (unless we are talking about native sulfur or gold or some such). See line 356. This needs to be fixed.

Thanks to reviewers 1 and 3 for picking up on this. It has been changed accordingly (to 'with respect to calcite'.) (line 270).

17. I don't understand this sentence at all! I think it should be deleted or re phrased. "We describe this style of weathering as non equilibrium weathering as the depletion rates of the various minerals involved are likely to be very different."

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This sentence has caused issue for the other reviewers (see above) and as a result has been removed and the discussion rephrased.

Lines 550-553: "Landslide weathering rates will remain elevated until the relevant reactive mineral(s) have been

depleted, after which weathering in landslides is likely to proceed much like in other parts of the landscape, driven by
 organic and carbonic acids. The initial abundance of trace sulphide and/or carbonate together with their exhaustion, over longer time scales, is likely to be an important control on the duration of rapid weathering in landslides."

18. What is important is not the "minority minerals" but "reactive minerals". Once the reactive minerals are removed, then the landslide material will weather like all the other weathered material
in the landscape (i.e. material above the reaction front for the reactive mineral). . .unless the landslide is grinding up the material and increasing surface area, which is another possible effect. I do think that pyrite is often a minor mineral but that is not what is important about it.

This echoes comments from the other reviewers; we have rephrased sections of the manuscript that refer to minority minerals to instead refer to reactive minerals.

19. Once pyrite oxidizes, weathering will revert from H2SO4 to carbonic plus organic acids.

Presuming the reviewer here means that we've not mentioned the organic acids, we have added
them into the discussion. Lines 550-552: "Landslide weathering rates will remain elevated until the relevant reactive mineral(s) have been depleted, after which weathering in landslides is likely to proceed much like in other parts of the landscape, driven by organic and carbonic acids."

20. The authors' use of "ambivalent" is incorrect: "the impact of landsliding on climate is 1265 ambivalent". I think the authors mean ambiguous.

Changed to 'ambiguous'.

21. I am not sure that the authors can defend the statement, "the area most affected by erosion due1270 [to] typhoon Morakot is now a net source of CO2"

As pointed out by the other reviewers, we have overstated our case here. We have rephrased the paragraph to only state substantiated claims or, where we are not certain, to state this uncertainty. See whole paragraph – lines 581-629.

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22. Conclusions: I don't understand this sentence: Five years after landsliding, carbonic acid is a weathering agent of lesser importance and the weathering of silicate mienrals is subsumed.

This sentence has been rephrased in response to this comment and a comment from reviewer 2.
(Subsumed is the wrong term here). Line 644-645: "Five years after landsliding, carbonic acid is a weathering agent of lesser importance, while the weathering of silicate minerals is not elevated."

23. The term "lithological phases" is not correct. A lithology is a lithology and a phase is a phase. . .not sure what a lithological phase is. . .a mineral?

1285

This sentence has been rephrased in response to this comment and a comment from reviewer 2. (Lines 656-657)

24. In figure 4, SiO2 is silica, Si is silicon.

Figure 4 has been changed to replace Si with sodium.

25. The way the figure on page 20 is plotted is confusing. The fonts on the y axis differ above and below the break. I don't really understand what the tick marks mean on the y axis, because of the break in the axis. Why plot the two figures together in this way?

We've altered this figure to remove any confusion over the overlap, as well as adding extra labels to the y-axis. To answer the question posed by the reviewer, we plotted these together originally to use space as efficiently as possible, but we've changed this to improve the clarity.

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Interactive comment on Earth Surf. Dynam. Discuss., doi:10.5194/esurf-2016-31, 2016.

We thank reviewer 3 for a review that reflects extensive field and lab experience, which has helped us improve our work.

Oxidation of sulphides and rapid weathering in recent landslides

Robert Emberson^{1,2}, Niels Hovius^{1,2}, Albert Galy³, Odin Marc^{1,2},

¹GFZ Deutsches Geoforschungszentrum, 14473 Potsdam, Germany

²Institute of Earth and Environmental Science, University of Potsdam, 14476 Potsdam, Germany

³CRPG-CNRS-UL, 54500 Nancy, France

Correspondence to: robert.emberson@gfz-potsdam.de

Abstract

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Linking together the processes of rapid physical erosion to the resultant chemical dissolution of rock is a crucial step in building an overall deterministic understanding of weathering in mountain belts. Landslides, which are the most volumetrically important geomorphic process at these high rates of erosion, can generate extremely high rates of very localised weathering. To elucidate how this process works we have taken advantage of uniquely intense landsliding, resulting from Typhoon Morakot, in the Taimali river and surrounds in Southern Taiwan. Combining detailed analysis of landslide seepage chemistry with estimates of catchment-by-catchment landslide volumes, we demonstrate that in

- 15 this setting the primary role of landslides is to introduce fresh, highly labile mineral phases into the surface weathering environment. There, rapid weathering is driven by the oxidation of pyrite and the resultant sulphuric acid-driven dissolution of primarily carbonate rock. The total dissolved load correlates well with dissolved sulphate – the chief product of this style of weathering – in both landslides and streams draining the area (R² = 0.841 and 0.929 respectively, p <0.001 in both cases), with solute chemistry in seepage from landslides and catchments affected by significant
- 20 landsliding governed by the same weathering reactions. The predominance of coupled carbonate-sulphuric acid driven weathering is the key difference between these sites and previously studied landslides in New Zealand (Emberson et al. 2015), but in both settings increasing volumes of landslides drive greater overall solute concentrations in streamsBedrock landslides create conditions for weathering where all mineral phase in a lithology are initially-unweathered within landslide deposits, and therefore the most labile phases dominate the weathering at the outset and-
- 25 during a transient period of depletion. This mode of dissolution can strongly alter the overall output of solutes fromeatchments and their contribution to global chemical cycles if landslide-derived material is retained in catchments forextended periods after mass wasting.

Bedrock landslides, by excavating deep below saprolite-rock interfaces, create conditions for weathering where all mineral phases in a lithology are initially unweathered within landslide deposits. As a result, the most labile phases

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<u>dominate the weathering immediately after mobilisation and during a transient period of depletion. This mode of</u>
 <u>dissolution can strongly alter the overall output of solutes from catchments and their contribution to global chemical</u>
 <u>cycles if landslide-derived material is retained in catchments for extended periods after mass wasting.</u>

35 **1. Introduction**

Bedrock landslides can produce favourable conditions for weathering by funnelling of runoff into deposits of freshlybroken rock mass with massive freeexposed mineral surface area and modest hydraulic conductivity. It has been shown in the Western Southern Alps (WSA) of New Zealand (Emberson et al. 2015) that this can give rise to strong gradients in dissolved solid concentrations in surface waters on hillslopes, with highest concentrations in seepage from landslides,

- 40 and that the rate of landsliding is an important control on the chemistry of the rivers draining the mountain belt. Localized landslide weathering, prone to the spatial and temporal variability of mass wasting, contrasts to common models in which weathering is controlled by steady and distributed erosion of a laterally continuous soil and regolith mantle (Dixon and von Blanckenburg 2012; Hilley et al. 2010; Ferrier et al. 2008; West 2012, Lebedeva et al. 2010, Li et al. 2014)). Such models anticipate that at high erosion rates weathering is kinetically limited, so that the link between
- material supply and weathering is progressively weakened and finally lost. Landsliding circumvents this by
 <u>exhumingmobilizing</u> relatively unweathered <u>bedrockmaterials</u> from deeper below the surface (<u>Gabet 2007</u>) and,
 crucially, fragmenting <u>the mobilized materials</u> <u>them</u>-so that weathering is locally optimized. Landsliding is the dominant
 erosion process in orogenic settings where rapid crustal shortening is paired with fast and persistent exhumation (<u>S. J.</u>
 Dadson et al. 2004; Hovius <u>et al. 1997</u>), <u>Stark, and Allen 1997</u>). We expect, therefore, that it may act as a first order
 control on dissolved solids export from active mountain belts. Here, we ask: does highly localised landslide weathering
- persist in different settings with variable lithology and climate <u>so that</u>, and can a more general case for the impact of landslides on weathering <u>can be made</u>, and if so, then what is <u>, including</u> the underlying mechanism by which weathering is boosted, <u>be made</u>?
- To address these questions, we have investigated chemical weathering in the rapidly eroding mountains of Taiwan at the landslidelocal and catchment scale. Our study site has a substrate of typical continental margin sediments, comprising calcareous sandstones and shales, and containing sulphide minerals (Das et al. 2012). – This is a marked contrast with the WSA, where the meta-sedimentary silicate rocks contain only limitedtrace amounts of carbonate and very little pyrite (Chamberlain et al. 2005, Koons and Craw 1991). – In this study, we exploit the synchronous occurrence of voluminous mass wasting in southern Taiwan due to exceptional rainfall associated with typhoon Morakot in 2009 to
- 60 eliminate the effects of progressive loss of soluble minerals, allowing a sharp focus on the causes of elevated weathering rates in recent landslides. Using samples of seepage from recent bedrock landslide deposits and water from mountain streams and rivers paired with catchment-wide estimates of landslide volume, we show that enhanced weathering in landslide deposits results primarily from the rapid reaction of the most labile minerals in the substrate, which are excavated from below the saprolite-bedrock interface by deep-seated slope failure. In the case of our 65 Taiwanese site, the oxidative weathering of highly reactive pyrite and the ensuing rapid sulphuric acid-driven
 - weathering of carbonate dominate the output from recent landslides and <u>are a first order control on propagate-importantly into</u> the stream chemistry.

2. Field Sites and sampling

- The mountains of Taiwan have erosion rates that are amongst the highest on Earth (between 4-6mm/yr (Willett et al. 2003; Fuller et al. 2003)), driven by rapid tectonic convergence at the East Asian margin and aided by tropical storm activity and earthquakes (S.-Dadson et al. 2003). Average annual mass-wasting rates are high (Hovius et al. 2000), but can be pushed to much higher rates by single, exceptional storms or seismic activity (Hovius et al. 2011; G.-W.-Lin et al. 2008; Marc et al. 2015). For example, in 2009 typhoon Morakot caused over 2.7m of rain in 48 hours (Chien and Kuo 2011) on the Southern part of the island, the highest storm totals ever recorded in Taiwan. The typhoon generated more than 22000 landslides (C.-W.-Lin et al. 2011; A.J-West et al. 2011), with an average density of 35000 m²/km², orders of magnitude more than under less exceptional meteorological conditions. This presents a rare opportunity to study an extensive population of recent, synchronous landslides, which is important because the rapid weathering in landslides might decay over a decadal time scale to levels indistinguishable from that on surrounding hillslopes
 - 2

- 80 (Emberson et al. 2015). Having a constant, known date of initiation for sampled landslides reduces any systematic changes to weathering that would be introduced by variable timing of the mass wasting. Moreover, having a total landslide surface area and volume dominated strongly by input from one single typhoon makes the solute load of rivers draining catchments with otherwise diverse landslide histories directly comparable.
- We sampled water chemistry from a variety of sources within the Taimali river catchment (Fig.1b) between February
 and March 2015. This catchment drains an area of 118km² in the East flank of the Taiwan Central Range, from the main divide to the Pacific Ocean. It had among the highest catchment landslide densities resulting from typhoon Morakot,
 andwith several subcatchments sustained catastrophic alluviation eatastrophically alluviated by debris. As a result, human settlement and activity in the catchment were abandoned inward of 7km from the coast, and at the time of
- sampling, anthropogenic influence was negligible. In addition to the Taimali main stream, we sampled stream water.
 from 19 subcatchments with areas between 1-10km², and geothermal spring water. We also sampled seepage from the deposits of 14 landslides. This water has circulated through the landslides and derives from rainfall, and in some cases is likely supplemented by runoff into crests of landslides seepage from the deposits of 14 landslides, representing water-draining through the landslides, either directly from rainfall or from runoff into the crests of landslides, stream water-from 19 subcatchments with areas between 1-10km², and geothermal spring water. We supplemented our landslide
- 95 seepage samples from the Taimali catchment with seepage from 8 recent landslides elsewhere in Southern Taiwan (Fig.1A), to assess landslide weathering over a wider range of locations.
 - Lithologically, the Taimali catchment is representative of the Tertiary sedimentary cover that forms a large proportion of the Taiwanese orogen (Central Geological Survey 2000); the slates, shales and sandstones of the Miocene Lushan and Eocene Pilushan formations contain important amounts of sedimentary and diagenetic carbonate and are pyrite bearing
- throughout (Das, et al. Chung, and You-2012). The sampled landslides outside the Taimali catchment are also rooted in the Pilushan formation, with the exception of two landslides in the Chenyoulan catchment in central West Taiwan, underlain by the Eo-Oligocene Paileng formation of slates and sandstones. Elsewhere in Taiwan the sedimentary carbonate content The lithologies of the sampled formations is between 0.28-0.66% (Hilton et al. 2014), and the sulphide abundance in the South of Taiwan has been estimated at 0.05-0.27% (Kao et al. 2004). The lithologies of these
- 105 <u>formations</u> are the dominant sedimentary rocks formed at continental margins with a clastic input, and they are found globally in active mountain belts (Hartmann and Moosdorf 2012).-
 - Sampled landslides vary in size between 7.6x10³-2.8x10⁶m², and their deposits cover a range of shapes. The largest landslides in the Taimali catchment initially blocked the main river channel and have since been incised, leaving massive fan terraces, tens of metres above the current river level. Intermediate landslides have built debris cones on
- 110 hillslopes and at the edges of valley floors, and some smaller landslides have filled debris chutes. All sampled landslides had mobilized bedrock in addition to the overlying regolith and soil mass and any present vegetation. Soils are generally only >1m in local concavities in the landscape (Lee and Ho 2009), but contiguous, dense vegetation on hillslopes that have not been dissected by landsliding suggests sufficient mobile regolith or soil is present to support flora. In the Taimali catchment and at other landslide sample sites, the modal topographic slope is around 26°, with many steeper
- 115 <u>hillslope segments.</u> In the Taimali catchment and at other landslide sample sites, the modal topographic slope is around 26°, with many steeper hillslope segments. This precludes the build up of a thick, laterally continuous soil and regolithmantle, and bedrock is present at or near the surface except where local colluvial fills have formed. Seepage was found at the base of many landslide deposits, immediately above the interface with underlying bedrock. The flow paths of this seepage could enable fluid interaction with exposed rock in the landslide scar as well as

- 120 fragmented rock mass in the deposit. We do not draw distinction between these, as the physical process of bedrock landsliding remains responsible for the exposure of the scar and the production of the deposit. Moreover, it is not possible to distinguish systematically between landslide scar and deposit from available satellite imagery. Landslide seepage may also incorporate deeper groundwater, which could exfiltrate through the scar. We compare sampled seepage with our own measurements of geothermal water as well as with the composition of deep groundwater reported
- 125 <u>in the literature to qualitatively assess the importance of deeper-sourced water in landslide outflow</u>In view of this, we eorrect our samples for any geothermal input.

Notably, during the early part of 2015, when we sampled, the South of Taiwan underwent one of the driest periods on record, with an estimated return time of 50-100 years (CWB (Central Weather Bureau) 2016). At this time, the sampled streams were at or close to a baseflow condition, but despite the dry conditions more than half of around 50 the deep-

130 seated landslides we visited had persistent seepage. <u>It Therefore, it seems probable that many landslides in Southern</u> Taiwan are never fully 'dry' in the current climate. Landslides in the Chenyoulan catchment were sampled in July and November of 2013, when more normal rainfall conditions prevailed.

Further, it is important to note that the sediment generated by landslides and debris flows does not reside exclusively on the hillslopes from where it was sourced. Significant storage of material over long timespans in larger river channels is

- 135 common, and is indeed substantial along the Taimali River, with <u>alluvial</u> channel fill occupying about 6% of the total catchment area. This channel fill may be an important weathering reservoir, and to investigate whether this stored sediment affects the observed river chemistry we have mapped the fill deposits along sampled streams. Since we sampled small seepages and 1st order tributaries, we lack good constraints on the fluxes of solutes. Discharge gauging is not available in the Taimali, as the hydrometric infrastructure on the river was destroyed by channel bed
- 140 aggradation after typhoon Morakot. Here, we report measured solute concentrations in landslide seepage and stream waters with unknown flow rates. These measurements can't be converted into solute fluxes without major assumptions, and weathering flux estimation is not the purpose of this study. Instead, we aim for a direct comparison of landslide seepage and river chemistry from a set of lithologically similar streams with identically timed mass wasting but varying populations of landslides.

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3. Methods

3. Analytical techniques

In this section, we report the analytical techniques used to obtain the data- in section 4 summarise these results. In later discussion, we compare measurements of both chemical concentrations and ratios with physical data about landslides and catchments derived from remote sensing. In general, we focus on qualitative links between these parameters, using statistical tests (R² coefficient of determination, and p-value to test significance of relationships) to support suggestions that parameters are connected.

155 3.1. Analytical Techniques

Samples were collected from source using an HDPE syringe, filtered on site through single-use 0.2µm PES filters into several HDPE bottles, thoroughly rinsed with filtered sample water. Samples for cation analysis were acidified using ultrapure HNO₃⁻. pH and temperature values were measured in the field at the time of sample collection. Analysis of cations was carried out with a Varian 720 ICP-OES, using SLRS-5 & USGS M212 as external standards,

and GFZ-RW1 as an internal standard and quality control (QC). QC samples were included for every 10 samples to account for drift; no systematic drift was found, with random variability less than 5%. Sample uncertainties were determined from calibration uncertainties, and were always lower than 10% (see Table 1 for element-specific uncertainties). Anion analysis was performed using a Dionex ICS-1100 Ion Chromatograph, using USGS standards M206 & M212 as external standard and QC. Uncertainties were always less than 10% for each of the major anions (Cl⁻⁻
 SO₄²⁻, and limited NO₃⁻). Bicarbonate (HCO₃⁻) was calculated by charge balance, which has been shown to introduce errors of approximately 10% (Galy and France-Lanord 1999). We make the simplifying assumption that the Dissolved inorganic carbon (DIC) is formed only of HCO₃⁻, which is generally applicable at the range of pH values measured. (Zeebe and Wolf-Gladrow 2001). The and the calcite saturation index of measured waters was determined using the USGS PHREEQC software package (Pankhurst and Appelo 2013)₂--

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3.2. Cyclic Input

3.1. Correction for cyclic input

Solute input from atmospheric sources can be significant in rivers and streams, and is routinely corrected for in many weathering studies. Previous work on the average chemistry of rainfall in different parts of Taiwan, has found that it is 175 highly dependent on location (e.g. higher pollutant content near the densely populated west coast (Wai et al. 2008) and intensity of rainfall (typhoon rainfall boasts the highest solute concentrations in some cases (Cheng and You 2010)). Using any of these measurements to correct our samples based on the assumption that all present chloride derives from cyclic input (following e.g. Calmels et al. 2011) resulted in clearly erroneous estimates of the proportion of cyclic salts in the water samples (e.g., >100% of dissolved K⁺ and SO₄²⁻ from cyclic sources). Notably, chloride is a minor 180 component in all samples, with a median value of 32 µmol/l (minimum 14.6, maximum 175 µmol/l). This is a similar range of values to rainwater measured in other coastal environments (Galloway et al. 1982; Nichol, Harvey, and Boyd 1997), and the overall correction of TDS is at most 10% even with the most concentrated rainfall we have found in published literature (spot samples measured by Calmels et al. (2011) in the Taroko gorge). We note that deep groundwater may also introduce chloride, which could lead to overestimates of cyclic input. Moreover, during the dry 185 sampling conditions, excess evaporation could lead to larger proportions of cyclic solutes. In view of the minor amounts of chloride in our samples and because of the caveats mentioned above, we elect not to correct our samples for cyclic input. In the discussion, we assess whether this simplifying assumption is valid. Solute input from atmospheric sources is small but significant in the Taimali and at most other locations where we have sampled landslide seepage. Several studies have measured the average rainfall chemistry in different parts of Taiwan, 190 finding that it is highly dependent on both location (e.g. higher pollutant content near the densely populated west coast-(Wai et al. 2008) and intensity of rainfall (typhoon rainfall boasts the highest solute concentrations in some cases-(Cheng and You 2010)). However, we found that using any of these published values for correction resulted in clearlyerroneous estimates of the proportion of cyclic salts in the water samples (e.g., >100% of dissolved K⁺ and SO₄²⁻ fromeyclic sources). Therefore, we have applied a correction to all samples based on ratios of dissolved species in seawater. 195 The Taimali catchment lies within 15km of the ocean, within the first orographic barrier, which supports the use of seawater ratios. Other sample sites are more widely spread but we prefer a single correction to a piecemeal, individual

treatment. Using the ratios of Cl⁻ to major cations and anions (Ca²⁺, K⁺, Mg²⁺ and Na⁺, and SO4²⁻) in seawater and assuming that all Cl⁻ present in samples results from cyclic contribution, we removed extrapolated cyclic cation and anion contributions according to the following equation:

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	$[X]_{eyelie} = [Cl^{-}]_{sample} \times ([X]_{rainfall}/[Cl^{-}]_{rainfall}) (1)$
	3.3. Mapping of landslides
	We mapped 576 landslides extant in the Taimali and surrounds prior to typhoon Morakot and those generated during
205	and after the event until 2015 from LandSat imagery (LS7 & LS8, resolution 30 and 15m respectively). Landslides
	generated during typhoon Morakot were also mapped from a mosaic of Formosat images taken within one month of the
	typhoon with a resolution of 2m. Landslides were identified manually, based on their geometry, topographic position
	and surface properties, as described in recent studies (Marc and Hovius 2015), and digitized on screen in an Arc-GIS
	environment; scars and deposits were mapped together as a clear distinction was not always possible from the available
210	imagery. Manual mapping prevents the inclusion of other active landscape elements with recent reflectivity changes, for
	example aggraded riverbeds, which were mapped separately. It also suppresses the possibility of amalgamation of
	multiple landslides into single mapped polygons, which blights automatic mapping (Marc and Hovius 2015, Li et al.
	2014). In our analysis we draw distinction between older landslides and those generated by typhoon Morakot, prompted
	by our previous work (Emberson et al. 2015) that has shown the chemical output from individual slides is likely to
215	significantly decay over a decadal timescale. It should be noted that the proportion of landslides predating typhoon
	Morakot is dwarfed by the mass wasting caused by this exceptional typhoon in much of the Taimali catchment. The
	proportion of the Taimali catchment occupied by landslides increased from 2% to 8% due to typhoon Morakot, while
	some subcatchments have upwards of 16% of their area occupied by typhoon-induced landslides. However, in a few
	subcatchments there is a larger proportion of older landslides, which allows for comparison with landscape units with
220	only negligible prior mass-wasting.
	From the mapped landslide areas, we also estimate the volumes of individual slides, using global area-volume
	relationships (Larsen et al. 2010). We lack direct measurements of the depth to which the landslides we sampled have
	scoured. Errors in the mapped landslide area are assumed to be <20%, which are propogated into the total volume_
	estimated, as well as the catchment-wide volume estimates. This follows the approach described in (Emberson et al.
225	2015), which allows direct comparison of the estimated landslide volumes in Taiwan and the WSA (Figure 4).
	In addition, we have quantified the area, modal slope, and the mean and range in elevation of each catchment in an
	ArcGIS environment, analysing ASTER GDEM V2 data provided by NASA. These data are reported in table 2.
	Where [X] is the measured concentration of any of the dissolved species. The assumption that all Cl ⁻ results from cyclic-
	input is conservative and justified as there are no evaporitie units reported in the sampled areas (Central Geological
230	Survey 2000; Ho 1975), and because the Taimali eatchment and most of the other landslide sites are essentially free
	from anthropogenic chemical input. Chloride is a minor component in all samples, with a median value of 32 µmol/l
	(minimum 14.6, maximum 175 µmol/l). This is a similar range of values to rainwater measured in other coastal-
	environments (Galloway et al. 1982; Nichol, Harvey, and Boyd 1997). Individual corrections for cyclic input are small;
	only Sodium has larger corrections as in some cases approximately 20% of the total molar concentration is derived from
235	eyclic sources, and the average correction is 10%.

3.2. Mapping of landslides

We mapped landslides extant in the Taimali and surrounds prior to typhoon Morakot and those generated during and after the event until 2015 from LandSat imagery (LS7 & LS8, resolution 30 and 15m respectively). Landslides-

generated during typhoon Morakot were also mapped from a mosaic of Formosat images taken within one month of the 240 typhoon with a resolution of 2m. Landslides were identified manually, based on their geometry, topographic positionand surface properties, as preseribed by recent studies (Mare and Hovius 2015), and digitized on screen in an Are-GISenvironment; sears and deposits were mapped together as a clear distinction was not always possible from the available imagery. Manual mapping prevents the inclusion of other active landscape elements with recent reflectivity changes, for 245 example aggraded riverbeds, which were mapped separately. It also suppresses the possibility of amalgamation of multiple landslides into single mapped polygons, which blights automatic mapping (Marc and Hovius 2015). In ouranalysis we draw distinction between older landslides and those generated by typhoon Morakot, especially in the eontext of previous work (Emberson et al. 2015) that has shown the chemical output from individual slides is likely tosignificantly decay over a decadal timescale. It should be noted that the proportion of landslides predating typhoon-250 Morakot is dwarfed by the mass wasting caused by this exceptional typhoon in much of the Taimali eatchment. The proportion of the Taimali eatchment occupied by landslides increased from 2% to 8% due to typhoon Morakot, while some subcatchments have upwards of 16% of their area occupied by typhoon-induced landslides. However, in a fewsubcatchments there is a larger proportion of older landslides, which allows for comparison with landscape units withonly negligible prior mass-wasting.

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4. Results: Landslide Seepage and Stream Chemistry

4. Measured water chemistry-

Concentrations of Total Dissolved Solids and major cations and anions in landslide seepage and stream water in the Taimali catchment and elsewhere in Taiwan are listed in Table 1.

TDS in seepage from sampled landslides in Taiwan ranges between $70807200 \mu mol/l$ to $2330023400 \mu mol/l$, while river TDS values lie between $42304000 \mu mol/l$ and $1270012600 \mu mol/l$. Thus, the measured total dissolved solids (TDS) in seepage from sampled landslides is consistently higher than in other surface water sources, with the exception of geothermal springs (up to c. $121000100150 \mu mol/l$)

- 265 Dissolved Calcium is the major cation in most samples, forming up to 90% (molar) of the cation load in some cases. In some seepage samples this is replaced by Magnesium, with Mg²⁺ as much as 70% of total cations. Sodium <u>makes and-Silicon make-</u>up much of the remainder of the cationic load, although <u>it only exceeds 15% molar in three seepage</u> <u>samples. Dissolved Silicon (as silicic acid in these pH conditions) never forms more than 10% of TDS Silicon never-</u> represents more than 10% molar, while Sodium only exceeds 15% in three seepage samples. All measured samples are
- supersaturated with respect to calcite (Table 1). The anion load is always more than 50% Bicarbonate; the remainder is formed of Sulphate, SO₄²⁻, which is always more than 10% and can be as much as 50%. Seepage from the two sampled landslides in the Chenyoulan catchment share the same overall relationships between individual elements as those-sampled in the Taimali catchment, and they both sit at the higher end of the range of concentrations. There is significant overlap between landslides sampled within the Taimali river and those in the immediate surrounds (Fig.1A); TWS15-68
 is the most notable, with relatively elevated SO₄²⁻ and TDS (4078 and 14932 µmol/l respectively). Seepage from the two sampled landslides in the Chenyoulan catchment share the same overall relationships between individual elements as those sampled landslides in the Chenyoulan catchment share the same overall relationships between individual elements as those sampled landslides in the Chenyoulan catchment share the same overall relationships between individual elements as those sampled in the Taimali catchment, although the measured concentrations are higher than in the majority of

Taimali seepage samples.

The concentrations of TDS in the streams of the Taimali catchment $-4230-12700 \mu mol/l -$ are systematically lower

than in landslide seepage, but high <u>compared with mean global rivers or mountain streams (e.g. Gaillardet et al. 1999,</u>
<u>West et al. 2005).</u> in a global riverine perspective, 4141-12200 µmol/l. Ca²⁺ is again the most significant cation, always forming more than 21% and as much as 37% of TDS; Na⁺₋₇ Si⁴⁺, and K⁺ are minor components, always less than 7%..., 6% and 1% of TDS, respectively. Mg²⁺ is quite variable; in most streams it only forms 1-3%, but it constitutes as much as 11% of the cationic load in three of the streams. Silicic acid averages 6% of TDS. In the anion load, HCO₃⁻ is often the most important component, forming between 20-50% of TDS. Sulphate is between 5 and 31% of TDS and exceeds HCO₃⁻ in six streams. We also measured trace strontium, the maximum measured value of which iswith peaks at 30 µmol/l. Lithium in the streams and main river is always below the detection limit of the OES (c.0.1 µmol/l).

290 5. Discussion

5.1. Landslide weathering

- Seepage from landslides in the Taimali catchment and scattered locations elsewhere in Taiwan has high TDS concentrations when compared with local streams. This supports the hypothesis that landslides are primary seats of 295 rapid weathering in this setting. We have found the same in the WSA of New Zealand (Emberson et al. 2015), and therefore the intense fragmentation of newly exposed rock mass, and the concentration and slow percolation of rain and runoff water in debris accumulated in topographic hollows are likely universal mechanisms promoting landslide weathering in steep mountain settings where distributed weathering in soils is kinetically limitedless efficient. In the WSA, TDS concentrations in landslide seepage are strongly impacted by the dissolution of trace calcite, the 300 abundance of which is likely to be greater at depth (e.g. Brantley et al. 2013). The depth of a landslide scales with its area (Larsen et al. 2010), implying that the amount of calcite as a proportion of the total landslide volume correlates with the landslide size. This correlation will be limited to the range of landslides where a significant proportion of excavated material is from above the depth to which calcite is depleted. By contrast, in the largest landslides, nearsurface materials, poor in calcite, form only a small proportion of the deposit, and a correlation with size my no longer 305 hold. Moreover, large landslides may also be subject to reduced diffusion of fluids and oxygen to the base of the deposits; the amount of water entering the slide scales with the area of the slide (Emberson et al. 2015), but the volume increases at a greater rate (Larsen et al. 2010). In the Taimali catchment, there is no strong correlation between landslide size and TDS; R² values for correlation between TDS and landslide area (Figure 2), and TDS and landslide volume are 0.11 and 0.09 respectively (p = 0.62, p =310 0.70, respectively). This suggests that here, other factors have an overriding effect on the solute concentration in seepage from landslides. Notably, we find a strong correlation between the TDS and dissolved Sulphate concentration in landslide seepage ($R^2 = 0.88$; p < 0.001). This is not unexpected WSA, TDS concentrations in landslide seepage arestrongly impacted by the dissolution of trace calcite which is likely to be more abundant at depth (Brantley et al. 2013). The depth of a landslide seales with the area (Larsen, Montgomery, and Korup 2010), implying that the proportion of 315 ealeite available correlate with the size of the issuing landslide. Where substrate mineralogy is relatively uniform, thissize effect may be due to the greater depth of the sears and deposits of larger landslides, the mobilization of relativelyless weathered rock mass and its more comprehensive fragmentation, and could arise from dilution or mixingthereduced hydraulie conductivity at the base of such landslides. However, the ratio of calcium to the sum of other cations (sodium, potassium, magnesium and calcium) is correlated with the ratio of dissolved sulphate to the same cations
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- 320 (Figure 3A; R^2 these effects are not evident in the Taimali catchment; R^2 values for TDS / landslide area and TDS / landslide volume are -0.025 and -0.009 respectively (p = 0.64, p<0.005), which suggests a change in weathering regime. A decrease in the fraction of sodium to other cations also correlates with the increasing fraction of sulphate (Figure 3B, 706, p = 0.772, respectively). This suggests that here, other factors have an overwhelming effect on landslide-weathering efficiency. Notably, we find strong correlations between the TDS and Ca²⁺ and dissolved Sulphate-
- 325 concentrations in landslide seepage (R² = 0.63, p<0.005841 and 0.891, respectively; Kendall's Tau value of 0.5238 and 0.6381, respectively; p<0.001 in both cases; Fig 2a and 2b). We attribute these changes this to the oxidation of pyrite in the mobilized and fragmented rock mass and an ensuing increase in weathering of carbonates driven by sulphuric acid. Although the sporadic presence of other sulphide minerals, or mineral sulphates, can't be excluded, this has not been documented in Southern Taiwan. Instead, the rapid oxidation of pyrite has previously been observed in the region and elevated TDS concentrations in Taiwan mountain rivers have been viewed in this light (Calmels et al. 2011; Das et al., Chung, and You-2012; Kao et al. 2004). Sulphide oxidation is so prevalent in the larger Kaoping basin (draining the South Western part of Taiwan) that the delivered sulphate is 1.2-1.6% of global fluxes, from a basin only ~0.003% of global drainage (Das et al. 2012), Chung, and You 2012), and these prior studies link this explicitly to elevated physical disintegration of rock. The lack of correlation between landslide size and seepage concentration suggests that the-
 335 distribution of pyrite is highly heterogeneous, or that the degree of its exposure for oxidation varies significantly-

between different sites.

- The area and volume of landslides varies over 3 and 4 orders of magnitude respectively; it is unlikely that the distribution of sulphides in the rock mass varies as much over short distance. Instead we suggest that the absence of correlation between size and solute concentration is likely indicative of a limit to advection and diffusion of oxygenated 340 water in landslides, either linked to the size or the unconstrained internal hydrology. The importance of pyrite in landslide weathering in Taiwan is disproportionate with its relatively minor abundance in the overall rockmass (Das et al. 2012; Kao et al. 2004). Instead, its prominent role is determined by its reactivity. By introducing significant quantities of fresh rock material into the surface weathering zone, landslides reset the proportions of individual minerals within this zone to that of the deeper substrate. This differs from weathering linked to soil production, as in many cases 345 the most reactive phases are depleted below the bedrock-regolith interface (e.g. Brantley et al. 2013, Drake et al. 2009). The importance of pyrite in landslide weathering in Taiwan is disproportionate with its relatively minor abundance in the overall rockmass (Das, Chung, and You 2012; Kao et al. 2004). Instead, its prominent role is determined by itsreactivity. By introducing significant quantities of fresh rock material into the surface weathering zone, landslides resetthe proportions of individual minerals within this zone to that of the deeper substrate. This removes the supply limit on 350 weathering for any phase, and allows rapid weathering of the most labile minerals present. In the Taimali and surrounds, the oxidation of pyrite is an important reaction in recent landslide deposits. It provides a potent source of acidity, which further accelerates the weathering of other minerals. The majority of the high TDS in landslide scepage is accomplished through carbonate weathering via either this sulphuric acid or carbonic acid. Calcium can also be sourced as a product of silicate weathering. However, the lower proportion of purely silicate derived cations in landslide seepage (e.g. Si:Ca²⁺ 355 ratio), as well as the markedly different behaviour of some silicate derived cations, with the exception of broadly
 - increasing K⁺ concentrations from e.20-300µmol/l over the measured range of SO₄²⁻, supports the interpretation that weathering in the sampled landslides is dominated by the dissolution of carbonates.

The majority of the high TDS in landslide seepage is accomplished through carbonate weathering via either the

- 360 sulphuric acid derived from pyrite oxidation or carbonic acid. Calcium can also be sourced as a product of silicate weathering. Whilst silicate-derived potassium does vary with sulphate concentrations, ranging from c.20-300µmol/l.
 over the measured range of SO_{4²⁻} the lower proportion of other purely silicate derived cations and silicic acid in.
 landslide seepage (e.g. Na⁺:Ca²⁺ ratio always <0.5, compared to the silicate mineral value of 2.85 (Calmels et al. 2011)), as well as the markedly different behaviour of sodium and calcium (Figure 3A and 3B) supports the interpretation that.
 365 weathering in the sampled landslides is dominated by the dissolution of carbonates.
- An implication of our observations is that in the Taimali catchment, a significant proportion of the sedimentary pyrite contained within the rock mass survives exhumation, and is only exposed by landslide fragmentation at the surface. Previous studies in Taiwan (Calmels et al. 2011) have shown that Sulphate is <u>a key component importantly present</u> in deep groundwater (sampled >100m below the surface)eirculating groundwater, but clearly not all pyrite is oxidised at
- 370 these depths. Groundwater circulating in the bedrock topography is unlikely to be the source of sulphate in seepage. Such it is unlikely that such water would not systematically surface at landslide sites, given the since individuallandslides have highly variable morphologic characteristics (size, scar/deposit ratio, position with respect to ridge crest and valley floor, and proximity to major faults) of individual landslides. The same argument applies to geothermal outflow. Moreover, hot spring waters sampled in the Taimali catchment have extremely high Lithium concentrations,
- 375 1610 μmol/l, and the lack of elevated Li⁺ concentrations in the landslide seepage samples suggests the elevated sulphate in seepage derives from in-situ weathering. precludes a deep geothermal source for this outflow. Other studies have found high concentrations of silicate_-derived cations in deeper groundwater (Calmels et al. 2011); we do not observe any strong increase in the proportionlink between the concentrations of cations exclusively sourced from silicate minerals (particularly sodium – figure 3BSodium, Silicon) and sulphateSulphate in landslide seepage. Extremely;-
- 380 extremely elevated Na⁺ and Mg²⁺ concentrations at some sites are not clearly associated with any other measured parameter, or with each other, and we have no ready explanation for these isolated observations at present. Since these high concentrations are not linked to dissolved sulphate, they may be linked to flowpaths with limited oxygen where sulphate oxidation cannot proceed, but we do not have a constraint on this.
- Overall, we suggest that the spatial heterogeneity of groundwater flow paths in mountain belts (Andermann et al. 2012,
 Calmels et al. 2011) and extensive tectonic fracturing (Molnar et al. 2007, Clarke and Burbank 2011; Menzies et al. 2014) will to some extent preclude the existence of well-defined depletion depths for reactive mineral phases as seen elsewhere (e.g. Brantley et al. 2013, Drake et al. 2009). As such, some reactive phases likely reach the near-surface where landslides can expose them to oxygenated fluid.

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5.2. Landslide weathering products in stream water

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with variable landslide activity, we have used the volumes of landslides calculated for each sampled subcatchment within the Taimali drainage area, normalised to catchment area and evaluated this against the solute load of the streams. For this purpose, we use landslide volume rather than surface area, because much of the weatherable mineral surface area is contained within the landslide debris, rather than at the landslide surface. In doing so, we assume that the hydraulic connectivity in the groundmass of the landslide is always high enough to wet the mineral surface area within

To directly compare the impact of landsliding on weathering in the steep mountains of Taiwan between landscape units

hydraulic connectivity in the groundmass of the landslide is always high enough to wet the mineral surface area within the entire landslide body. We find that TDS and Ca^{2+} increase with increased landslide volumes in the sampled subcatchments where the landslide volume density <u>exceedstops</u> approximately $10^4m^3/km^2$ of catchment area (Fig. <u>43</u>).

- 400 Above this 'threshold', we observe a doubling of TDS from c.6000 to more than 12000 μ mol/l and increases in the concentration of Ca²⁺ from c.1000 to nearly 5000 μ mol/l, as landslide densities increase up to c.10⁶ m³/km². There is an increase in the SO₄²⁻ over this same landslide density range, from c.300 to nearly 4000 μ mol/l, and as in the individual landslide seepage samples, there is a strong correlation of both stream TDS and dissolved Ca²⁺ concentrations with the dissolved Sulphate (R² = 0.929 and 0.9318, respectively; p<0.001 for both relationships). Dissolved Na^{+SiO}₂ (Fig.5A)
- 405 does 4b) and Na⁺ do not systematically vary with SO_4^{2-} (p = 0.7492, p = 0.56 respectively), although K⁺ does increase significantly, from 15-130µmol/l with Sulphate concentration (p <0.001). At lower specific landslide volumes, there is a larger degree of variability and below 10^4 m³/km², equivalent to a debris layer with a uniform thickness of about 1 cm over the catchment, the stream TDS and Ca²⁺ concentrations do not vary systematically with the landslide volume. The importance of landsliding in setting the chemical load of streams varies depending on which element is considered.
- 410 For example, landslides exert a strong control over dissolved Calcium and Potassium in streams, but have almost no impact on dissolved Sodium. At the catchment scale, solute concentrations remain primarily dependent on the additional acidity provided by oxidation of pyrite; coupled carbonate-sulphuric acid weathering increases the Ca^{2+} output, and with it of the total exported solute load. On the other hand, dissolved SiO₂ is unaffected by either increasing landslide volume or changes in SO₄²⁻. Notably, values of SiO₂ measured in the streams of the Taimali catchment can exceed those
- 415 measured in landslide seepage, suggesting that any landslide-induced impact on local silicate weathering is not significant enough to outweigh weathering elsewhere, either in soils on stable hillslopes or deeper in the subsurface. Longer flow paths of deep groundwater, reaching well below the levels affected by landsliding, are more likely to be enriched in slower dissolving silicate weathering products (Bickle et al. 2015; Calmels et al. 2011; Galy and France-Lanord 1999), and the relatively low fraction of sodium and silicic acid Silicon-in landslide seepage suggests a comparatively short water residence time in mass wasting deposits (Maher 2011). In view of this, we posit that the disconnection between stream-borne Sulphate and some silicate-weathering products (Na⁺, silicic acid SiO₂), juxtaposed with a link of Sulphate and Potassium, which is also only sourced from silicate, reflects the existence of multiple pathways relevant for silicate weathering on a catchment scale. More congruent weathering in deep groundwater likely complements the rapid weathering of carbonates and incongruent weathering of silicates (such as rapid K release
- fromby chlorite and biotite, (Malmstrom et al. 1996)) in mass wasting deposits.
 The sampled subcatchments have up to 16.6% of the total area occupied by landslide deposits and scars. However, the Taimali watershed contains some exceptionally large landslides, the outlines of which define tributary catchments in their own right. The largest landslide we sampled (TWS15-46) has a surface area of 2.7km². At these very large scales and where landsliding has affected such an important part of the topography, the distinction between landslide and
- 430 stream catchment becomes blurred, and solute concentrations from such catchments will also be subject to the same limits on TDS concentrations in the landslides themselves. The peak TDS values we measured in seepage are between 15000-20000µmol/l. Other locations may have greater concentrations, if local weathering has greater oxidation of sulphides, either due to more efficient distribution oxygenated water at depth in the landslide deposit. Such hypothetical, very high concentrations, in excess of what we have recorded, would serve as an upper limit to solute
- 435 concentrations in the entire catchment. saturation of the surface water chemical load is likely. This depends on the local distribution and exposure of sulphides and, since rainfall is dependent on area, also on the area:volume ratio of the landslides, which limits the amount of fluid for dissolution per unit landslide volume. Therefore, it is likely that the relationship between landslide volume and stream solute concentration will also saturate above a hypothetical-"maximum concentration" in seepage from landslides above a given size. In the landslides sampled in and around the

440 Taimali river, this maximum TDS appears to be around 15000-20000µmol/l, which we observe in landslides 50000m²and larger (although larger slides can remain below this point). Even where landsliding is less prominent, there is still significant variability in tributary stream dissolved load (Fig. <u>4</u>3). This is unlikely to be an artefact of the mapping; landslides below the detection limit (about 500 m²) are unlikely to mine significant quantities of bedrock. Instead, they primarily mobilize colluvial or soil material, which limits their 445 impact via the oxidation of sulphides. Meanwhile, other environmental factors that result in variability in TDS at higher landslide rates will also apply to the steep mountain landscape of southern Taiwan in the absence of landslides. This includes variable oxidation of pyrite in a tectonically fractured substrate, which will affect both deep and shallow groundwater fluxes, depending on where in the weathering zone the respective hydrological flow paths locate. Other studies have incorporated topographic factors, such as local slope or hillslope length, into the models of weathering 450 (Heimsath et al. 1997; Maher and Chamberlain 2014). These factors may also be a source of variability in subcatchments with low landslide rates. However, landsliding during typhoon Morakot was primarily controlled by the intensity and volume of rainfall (Lin et al. 2011), and the pattern of landsliding in the Taimali catchment is not strongly tied to topographic characteristics. Hence, the amount of mass wasting in individual sub-catchments is not strongly related to the local modal slope (see data in Table 2). Moreover, neither modal slope nor the range of elevation in a 455 catchment is correlated with the measured TDS, suggesting these factors do not strongly control the weathering in this setting. Other studies have incorporated topographic factors into the models of weathering (Heimsath et al. 1997; Maher and Chamberlain 2014), and these may also be a source of variability at low landslide rates. Since the intensity and volumeof rainfall during the typhoon were the primary control on the rate of landsliding during typhoon Morakot (C. W. Lin et-460 al. 2011), the pattern of landsliding in the Taimali catchment is not strongly tied to topographic characteristics. Hence, the amount of mass-wasting in individual sub-catchments is not strongly related to the local modal slope. Many subcatchments of the Taimali watershed contain significant amounts of alluvial material, which derives importantly from landslide debris delivered to streams. Alluviated streambeds could be a locus of enhanced weathering, but we do not observe any correlation between the measured stream chemistry and the proportion of the catchment

- filled with alluvial material in the six subcatchments with significant fill (>1% of total catchment area see Table 1. $R^2 = -0.365$; Kendall's Tau = -0.067; p = 0.48). However, due to the lack of rainfall prior to and during the sampling period we were unable to sample hyporheic water from within the alluvial streambeds. Presence of secondary precipitates in fill deposits suggests that at low flow these sedimentary bodies could act as sinks for solutes, rather than sources. Although the role of alluvial materials in the weathering budget of the Taimali catchment is not fully clear, our
- 470 observations suggest that it is not a first order control on the chemical output during the sampling interval.
 AsSince we did not measure the discharge in these small streams we can only report concentrations of solutes. Weatherdetermined dilution could affect the measured solute concentrations and cause <u>themit</u> to vary between catchments. However, observations from other parts of the Taiwanese mountains (Calmels et al. 2011, Lee et al. 2015)) suggest that dissolved Calcium <u>concentrations are only diluted and SiO₂-concentrations do not dilute</u> by more than approximately
- 475 35% <u>during peak typhoon</u> for the full range of discharge conditions in mountain rivers. Therefore, we expect that although the period when we sampled was relatively dry, our measurements are a fair first order representation of solute concentrations and ratios during the majority of the year. This is <u>consistent in keeping</u> with observations from other mountain belts, including the Southern Alps of New <u>Zealand</u>, the <u>Himalayas</u> Zeanland and the <u>AndesHimalayas</u>, where rivers <u>exhibit near-chemostatic behaviour</u> are quite close to chemostatic over a range of discharge conditions (Lyons <u>et</u>

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5.3. Landsliding and the weathering of labile minerals

Solute concentrations in landslide seepage and stream water in the Taimali catchment are significantly higher than in the rivers of the WSA by approximately 5-10 times (Fig.<u>4a</u>3a). Both locations share a steep, landslide-dominated mountain 485 physiognomy and very high rainfall rates, but Taiwan is warmer and has carbonate rich, pyrite bearing lithologies, whereas the WSA formations have only trace amounts of these minerals. We suggest that lithology and climate set a baseline for weathering in a given setting, above which landslides can control the weathering variability. This control is especially clearly expressed in the WSA, where the substrate for weathering is relatively homogeneous. The Taiwanese river chemistry has a larger spread with respect to landslide volumes, but the clear correlation between stream TDS and 490 SO_4^{2-} (Fig.5A, $R^2 = 0.93$, p < 0.0014) reflects the importance of the weathering occurring within the landslides in setting catchment scale river solute concentrations. It is unlikely that this correlation arises from dilution or mixing of sources. The lack of correlation between sodium and sulphate concentrations (Fig 5B, p = 0.74) rules out a strong influence of both dilution and evapotransporation, and likely precludes weathering pathways with elevated sulphate and silicate weathering such as deeper groundwater from contributing to the higher TDS. ehemistry. Heterogeneous distribution of 495 sulphides in the bedrock substrate of the Taimali catchment and its variable oxidation during exhumation likely not only affect the individual landslide seepage concentrations, but also the chemistry of streams throughout the catchment. This may explain why the total volume of landslides is a weaker control over stream TDS than in New Zealand. Reactions of the most labile mineral phases dominate the chemistry of landslide seepages. In the Taimali catchment and elsewhere in Taiwan, the fastest reaction is the oxidation of sulphide. In the case of the WSA, elevated landslide-500 seepage concentrations (Emberson et al. 2015) result from weathering of small amounts of highly labile carbonate, which become exposed through rock mass fragmentation. However, unlike the Taimali, the fragmentation of WSA bedroek does not introduce an extra source of acidity. The relative abundance of carbonate in Taiwan means fluids are supersaturated everywhere with respect to Calcium (Table 1). Therefore, the potential for landsliding to increase earbonate weathering due to rock mass fragmentation would be limited but for the exposure of pyrite and the generation 505 of sulphuric acid this entails. Thus, exposure of minority minerals due to rock mass fragmentation boosts weatheringrates in landslides, but the minerals and their roles vary between settings. We describe this style of weathering as nonequilibrium weathering, as the depletion rates of the various minerals involved are likely to be very different. Landslide weathering rates will remain elevated until the relevant minority mineral(s) have been depleted, after which weathering in landslides is likely to proceed much like in other parts of the landscape. The initial abundance of trace-510 sulphide and/or carbonate together with their exhaustion, over longer time seales, is likely to be an important control onthe duration of rapid weathering in landslides. However, in choosing the Taimali eatchment, where the vast majority of the mass wasting observed occurred very recently and all at once, we have also selected a location where the long-termimpact of rock mass fragmentation in landslides is difficult to observe. Nevertheless, some constraints may be gleanedfrom two subcatchments (TWS15-40 and TWS15-53) where the TDS and dissolved SO42-values are lower than others-515 with similarly high landslide incidence (TWS15-40 9220µmol/l, TWS15-53 7617µmol/l compared to other streams with TDS greater than 12000µmol/l). These two subcatchments have large landslides that had been moving persistently for at least two decades prior to typhoon Morakot. Although these landslides were reactivated during the typhoon, they likelydisplaced materials that had already been significantly depleted of pyrite due to prior fragmentation and weathering. This is circumstantial evidence of a substantial loss of weathering efficiency by depletion of especially labile minority-

- 520 minerals on a decadal time scale in this instance, similar to previous findings in the WSA (Emberson et al. 2015). A second time scale of relevance is that of the physical removal of landslide debris from the catchment. If debrisremains in a catchment after depletion of exposed pyrite, then its weathering will revert to mineral reactions with-Carbonic Acid, which dominate elsewhere in the landscape. If the time to depletion is greater than the time required for removal of the debris, then the labile phases it contains will remain unweathered and will be removed through sediment-525 transport processes. In the case of Taiwan, restricted data on river bed material show Sulphur content of up to 1.07%-(Hilton et al. 2014), suggesting that weathering in the landscape does not purge all sulphides brought to the surface byrock mass exhumation. In the opposite case the weathering from a given landslide will return to a constant backgroundrate shared with other, colluvium and soil covered parts of the landscape. How this seales up to the catchment sealedepends on the return time of large drivers of mass-wasting, such as typhoons and earthquakes, and the subsequent 530 advection of debris into the fluvial network and the transport capacity of this network. A disparity between short termproduction of sediment on hillslopes and the capacity for onward transport can lead to up to millenial residence times of event-produced sediment in mountain landscapes (Blöthe and Korup 2013). In the Taimali catchment, many headwater streams were still clogged with sediment 6 years after typhoon Morakot, and large colluvial fans had formed belowseveral landslide-dominated tributaries. Elsewhere in Taiwan, mountain valleys contain large colluvial terraces, which-535 have formed hundreds to thousands of years ago (Hsieh and Chyi 2010). This implies that time seales of pyrite depletion and of debris removal must both be considered in further explorations of the links between erosion and weathering in Taiwan.
- Reactions of the most labile mineral phases dominate the chemistry of landslide seepage. In the Taimali catchment and
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 generation of sulphuric acid. Thus, exposure of reactive minerals due to rock mass fragmentation boosts weathering.
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 is less applicable to these settings, as the distribution of reactions will be controlled by the localisation of detrital.
 sulphides and the flow paths providing oxygenated fluid.
- Landslide weathering rates will remain elevated until the relevant reactive mineral(s) have been depleted, after which weathering in landslides is likely to proceed much like in other parts of the landscape, driven by organic and carbonic acids. The initial abundance of trace sulphide and/or carbonate together with their exhaustion, over longer time scales, is likely to be an important control on the duration of rapid weathering in landslides. However, in choosing the Taimali catchment, where the vast majority of the mass wasting observed occurred very recently and all at once, we have also selected a location where the long-term impact of rock mass fragmentation in landslides is difficult to observe. Nevertheless, some constraints may be gleaned from two subcatchments (TWS15-40 and TWS15-53) where the TDS and dissolved SO₄²⁻ values are lower than others with similarly high landslide incidence (TWS15-40 9220µmol/l; TWS15-53 7617µmol/l compared to other streams with TDS greater than 12000µmol/l). These two subcatchments have large landslides that had been moving persistently for at least two decades prior to typhoon Morakot. Although.

- 560 these landslides were reactivated during the typhoon, they likely displaced materials that had already been significantly depleted of pyrite due to prior fragmentation and weathering. This is circumstantial evidence of a substantial loss of weathering intensity by depletion of especially labile minority minerals on a decadal time scale in this instance, similar to previous findings in the WSA (Emberson et al. 2015).
- A second time scale of relevance is that of the physical removal of landslide debris from the catchment. If debris 565 remains in a catchment after depletion of exposed pyrite, then its weathering will revert to mineral reactions with Carbonic Acid. If the time to depletion is greater than the time required for removal of the debris, then the labile phases it contains will remain unweathered and will be removed through sediment transport processes. In the case of Taiwan, some samples of river bed material show Sulphur content of up to 1.07% (Hilton et al. 2014), suggesting that weathering in the landscape does not purge all sulphides brought to the surface by rock mass exhumation. In the
- 570 opposite case the weathering from a given landslide will return to a constant background rate shared with other colluvium and soil covered parts of the landscape. How this scales up to the catchment scale depends on the return time of large drivers of mass-wasting, such as typhoons and earthquakes, and the subsequent advection of debris into the fluvial network and the transport capacity of this network. A disparity between short term production of sediment on hillslopes and the capacity for onward transport can lead to up to millenial residence times of event-produced sediment 575 in mountain landscapes (Blöthe and Korup 2013). In the Taimali catchment, many headwater streams were still clogged with sediment 6 years after typhoon Morakot, and large colluvial fans had formed below several landslide-dominated tributaries. Elsewhere in Taiwan, mountain valleys contain large colluvial terraces, which have formed hundreds to thousands of years ago (Hsieh and Chyi 2010). This implies that time scales of pyrite depletion and of debris removal
 - must both be considered in further explorations of the links between erosion and weathering in Taiwan.

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5.4. Impact on climate

The link between weathering and atmospheric pCO_2 is determined by the mineralogy of the weathered substrate, the composition of the weathering acid, and the residence time of material in the weathering zone (Torres et al. 2016, Galv and France-Lanord 1999, Lerman et al. 2007). - Therefore, the balance of silicate:carbonate weathering, the source of 585 acidity in mass-wasting deposits and the retention of these sediments in the landscape are all crucial to the role of landsliding in the Carbon cycle. As a result of this complexity, the impact of landsliding on climate is ambiguousambivalent. Weathering of silicates with Carbonic acid can lower the atmospheric pCO₂, but with Sulphuric acid it has no effect. Dissolution of carbonates has no net effect on atmospheric pCO2, if achieved with Carbonic acid, and it can increase pCO₂ if it is accomplished with Sulphuric acid (Calmels et al. 2007; Gaillardet et al. 1999; Torres, 590 West, and Li 2014). As erosion rates increase and mass-wasting through bedrock landsliding begins to dominate erosional budgets, the erosional impact on feedback between drawdown of CO₂ and climateerosion, achieved through sustained weathering of silicates (Berner and Kothavala 2001; Brady 1991, Raymo and Ruddiman 1992), will weaken. Weathering Then, weathering of carbonates will form a greater portion of solute budgetsassume a more central role, even where they are only present in small amounts, and in mountain belts where pyrite is ubiquitous, erosion-driven weathering may even become a source of CO2 (e.g. Calmels - If the retention time of sediment derived from masswasting is long compared to the decay time of the weathering boost provided by these highly labile minerals, then anyeffect of landsliding on the CO2 budget of a mountain belt may be limited. As the sediment retention time equals the weathering boost decay time, the effect of landsliding on the CO2 budget is maximized. Shorter sediment residencetimes promote the export of unweathered sediment to the ocean and its burial in marine basins. This can result in a

- 600 fraction of sulphides contained in the rock mass bypassing the weathering window (Hilton et al. 2007, Torres 2014),reducing the potential effect of their exhumation on atmospheric pCO₂ and returning the landscape to a state in whichdistributed weathering in soils and colluvial fills dominates. In addition to this inorganic complexity, the export andburial of organic carbon via landsliding also has an impact on the Carbon budget of a mountain belt (Hilton, Galy, and Hovius 2008; Hilton et al. 2016). 2011; West et al. 2011). Ultimately, quantification of the processes side by side is-
- 605 important to understanding of the role of mountain erosion in the Earth's Carbon cycle.
 The length of time over which sediment is stored in a catchment will determine the cumulative effect of this weathering. In Taiwan, storage times of greater than 50kyrs have been demonstrated for sedimentary terraces derived from masswasting (Hsieh et al. 2013, 2010). In such locations, where the retention time of sediment derived from mass-wasting is long compared to the decay time of the weathering boost provided by the highly labile minerals contained initially in
- 610 these sediments, the coupled sulphuric acid-carbonate weathering will likely be superseded by slower silicate weathering, reducing any net release of CO₂. When the sediment retention time equals the weathering boost decay time, the effect of landsliding on CO₂ release is maximized. Shorter sediment residence times promote the export of unweathered sediment to the ocean and its burial in marine basins. This can result in a fraction of sulphides contained in the rock mass bypassing the weathering window (Hilton et al. 2014), reducing the potential effect of their exhumation.
- 615 on atmospheric pCO₂ and returning the landscape to a state in which distributed weathering in soils and colluvial fills. dominates. In addition to this inorganic complexity, the export and burial of organic carbon via landsliding also has an impact on the Carbon budget of Taiwan (Hilton et al. 2008, 2011; West et al. 2011). Ultimately, quantification of the processes side by side is important to understanding of the role of mountain erosion in the Earth's Carbon cycle.
- In the Taimali catchment a good analogue for many young mountain belts formed of continental margin sediments widespread, typhoon-triggered landsliding has caused a significant increase in carbonate weathering by Sulphuric acid over timescales shorter than the return time of extreme meteorological drivers of mass wasting. As a result, the area most affected by erosion due typhoon Morakot <u>could beis now</u> a net source of CO₂ to the atmosphere, <u>unless theoffshore burial of organic carbon croded from soils and standing biomass in the landscape offsets this effect. The
 weathering boost is likely to end before the next major meteorological perturbation of southern Taiwan, so that the
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weathering of carbonates and silicates with Carbonic acid must be taken into account when considering the longer-term
effects of erosion in South Taiwan. Future work to quantify the net sequestration or release of CO₂ would require a
closer evaluation of the ratios of acid at work, as well as correction for the extensive secondary precipitation evident in
the catchment.

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6. Conclusions

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Landslides can mobilize and fragment large volumes of rock mass from below the soil and regolith mantle of steep hillslopes. Weathering of this material is promoted by the exposure of fresh rock to <u>infiltrating persistently circulating</u> water and oxygen in landslide debris with very high internal surface area. Under these conditions, weathering is not limited by the <u>kinetics of the reactions</u> of weatherable material. It progresses, instead, at a rate determined primarily by the kinetics of the <u>most labile mineral phases</u>, butdominant chemical reactions, for as long as these phases are abundant in the rockmass the rateall minerals involved in these reactions are abundantly exposed. Thus, the process of weathering is vastly increased in landslides is dominated in the first instance by reaction of the most labile mineral phases in the mobilized rock mass. We have found that these labile phases may be minority constituents, driving the

640 weathering in fresh landslide deposits away from the slower process affecting the surface materials elsewhere in the landscape over longer time scales.

In Southern Taiwan, weathering in landslides triggered during typhoon Morakot in 2009, is governed by the rapid oxidation of pyrite, which is a trace mineral in the pelitic <u>rocksunits</u> of the local substrate. This <u>oxidation generates</u> sulphuric acid, which reacts primarily with ubiquitous carbonates. Five years after landsliding, carbonic acid is a

- 645 weathering agent of lesser importance, <u>while-and</u> the weathering of silicate minerals is <u>not elevated</u>subsumed. In the Taimali catchment in Southeast Taiwan, which sustained landsliding at spatially diverse rates during typhoon Morakot, stream chemistry suggests that landslides have tapped into rock mass with variable amounts of pyrite. Although total dissolved solids concentrations in tributary streams vary with the landslide density in their subcatchments, they are <u>better correlated</u>related first with the Sulphate concentration. In rare subcatchments where older landslides, dating back
- at least 25 years, dominate, TDS and Sulphate concentrations are markedly lower, indicating that the effects of exposure and oxidation of pyrite in landslides may dissipate on decadal time scales in this area.
 Southern Taiwan sits within a rapidly deforming and uplifting mountain belt, formed with calcareous sandstones and pelites from the Asian continental margin. It shares its deformation style and a range of lithologies with many other

young mountain belts at convergent plate boundaries. Bedrock landsliding is the dominant mode of mass wasting and

sediment production in such settings. Therefore, we submit that our observations of landslide weathering in Taiwan may well have wider implications. Moreover, even where pyrite is not significantly present, the importance of labile mineral phases in weathering in recent landslide deposits is evident. In the Western Southern Alps of New Zealand, the rapid reaction of carbonates, which make up only a very small fraction of the rock mass, with carbonic acid dominates the chemistry of landslide seepage and helps set the weathering flux from the mountain belt (e.g. Jacobson et al. 2003, Emberson et al. 2015). The combination of results from these two settings suggests that landslides may affect weathering in other rapidly denuding mountain belts, albeit modulated by lithology.--

Hence, in order to understand chemical weathering rates from mountain belts where erosion is dominated by stochastic mass-wasting it is crucial to consider the way in which this specific erosive process controls weathering. We propose a conceptual model of a rapidly eroding landscape where the background weathering is set by lithology and climate,

overprinted by the impact of landslides. The importance of <u>reactive mineralminor lithological</u> phases, weathering rapidly in small and specific parts of the landscape affected by deep-seated erosion, is a contrast to existing concepts, which do not encompass the stochastic nature of weathering (Dixon and von Blanckenburg 2012; Maher and Chamberlain 2014; Riebe <u>et al. 2003)A full description of the impact of landsliding on weathering should include the effect on the availability of reactive phases in addition to the previously modelled spatial stochasticity (Gabet 2007). , 670
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Ultimately, the importance and impact of landslide weathering will be determined by the rate of the geomorphic process and the mineral composition of the substrate in which it occurs. However, it will also depend on the efficiency with which the products of mass wasting are removed from the landscape and transferred into geological basins, relative to the time needed to leach the most labile mineral phases from the mobile sediment, and to the return time of important

675 mass wasting events in the landscape. In this light, we draw attention to the possibility that disproportionate dissolution of highly labile mineral phases such as pyrite or carbonate as a result of landslide driven weathering may reduce the strength of the <u>linkfeedback</u> between erosion and atmospheric CO₂ drawdown via silicate weathering in fast eroding settings, which in turn would modulate any feedback on erosion through changes in tropical storm intensity (e.g., <u>Knutson et al. 2010</u>). The role of active mountain belts in the global Carbon cycle remains ambiguous.

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 wrote the paper with major input from all authors. Three anonymous reviewers greatly aided the development of the manuscript with constructive commentary. R.E. has benefited from Helmholtz Institutional funding during this project.

690 8. References

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850 Figure Captions

Figure 1

Locations of sampling. Fig.1A: location of Taimali catchment and landslides sampled outside of this river in Taiwan. Fig.1B: Taimali river, with individual streams outlined over a relief map derived from ASTER DEM. Streams are colour coded by the fraction of their area occupied by landslides (scars and deposits); individual landslides are also highlighted as they were mapped from satellite imagery, together with the alluvial fill in the channel. Locations where we sampled landslide seepage are also shown.

Figure 2

Total dissolved solids (TDS) in landslide seepage plotted against the mapped area of the individual landslides.
 Analytical error on TDS measurements are shown.

Cyclic- and hydrothermal-corrected dissolved Sulphate (SO4²⁻) plotted against corrected Total dissolved load (TDS, above) and Calcium (Ca²⁺, below) in seepage from landslides in the Taimali river and surrounds, as well as the Chenyoulan catchment in central Taiwan. Error bars are the total measurement uncertainty.

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Figure 3

 Above: molar ratio of dissolved calcium to sum of dissolved calcium, magnesium, potassium and sodium (Ca²⁺/Na⁺+K⁺ +Ca²⁺+Mg²⁺) plotted against ratio of dissolved sulphate to the same dissolved elements (SO₄²⁻/Na⁺+K⁺+Ca²⁺+Mg²⁺).
 Below: molar ratio of dissolved sodium to sum of dissolved calcium, magnesium, potassium and sodium (Na⁺/Na⁺+K⁺ +Ca²⁺+Mg²⁺) plotted against ratio of dissolved sulphate to the same dissolved elements (SO₄²⁻/Na⁺+K⁺+Ca²⁺+Mg²⁺). Error bars are the propogated total measurement uncertainty.

Normalised volume of landslides (calculated volume divided by catchment area) plotted against total dissolved load (TDS, fig A), dissolved Calcium (Ca²⁺, fig B), and dissolved Sulphate (SO4²⁻, fig C) for streams in the Taimali river (this study) and previous numbers for catchments in the Western Southern Alps (WSA) of New Zealand (Emberson et al. 2016). Vertical error bars are total measurement uncertainty; horizontal error bars are 95% confidence intervals for volumes. Each chemical measurement has been corrected for cyclic and hydrothermal input.

880 Figure 4

<u>Normalised volume of landslides (calculated volume divided by catchment area</u> Dissolved Sulphate (SO₄²⁻) plotted against total dissolved solids (TDS, fig A), dissolved Calcium (Ca²⁺, fig B), and dissolved Sulphate (SO₄²⁻, fig C) for dissolved Silicon (SiO₂, above) and Total dissolved load (TDS, below) in streams in the Taimali river (this study) and previous numbers for catchments in the Western Southern Alps (WSA) of New Zealand (Emberson et al. 2016). Vertical
 error - Error bars are total measurement uncertainty; horizontal error bars are 95% confidence intervals for volumes.
 Each chemical measurement has been corrected for cyclic and hydrothermal input.

Figure 5

(A) Dissolved Sulphate (SO4²⁻) plotted against total dissolved solids (TDS, above) and (B) dissolved sodium (Na⁺,
 below) in streams in the Taimali river. Error bars are total measurement uncertainty.

	<u></u>	0.5	0.7	0.7	0.8	1.1	1.0	1.2	1.1		<u>-0.2</u>		Fractional Channel Fractional Channel
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000	737	<u>154</u>	<u>268</u>	<u>225</u>	<u>161</u>	<u>296</u>	<u>275</u>	217	<u>226</u>		<u>3643</u>		n Elevation
5	<u>747</u>	<u>169</u>	<u>301</u>	<u>305</u>	<u>296</u>	<u>480</u>	<u>348</u>	273	<u>336</u>		<u>46541</u>		e, m Mear
	쾨	<u>106</u>	<u>137</u>	<u>138</u>	<u>1281</u>	346	<u>146</u>	<u>172</u>	<u>134</u>		28		evation range, m Mean Elevation, m
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ç	의	<u>30</u>	뛄	78	<u>61</u>	78	<u>40</u>	<u>129</u>	<u>49</u>		<u>862</u>		Modal Slope
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2.01	<u>C.61</u>	<u>20.0</u>	<u>22.1</u>	<u>27.1</u>	25.5	22.5	<u>22.0</u>	<u>25.1</u>	<u>24.3</u>		<u>24.1</u>	racter	<u>Longitude</u>
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30200 00	<u>C7179.07</u>	120.81979	20.84565	20.85395	20.86786	20.84806	20.84619	20.8435	20.84148		20.93897	stream	0
1 01023 C	CC.0 C7/70.071 047/C.77	22.58458 12	22.56602 120.84565 8.46	22.57761 120.85395	22.57609 120.86786 8.46	22.58842 120.84806	22.59241 120.84619	22.62438 120.8435	22.59696 120.84148 8.41		<u>22.59343</u> <u>120.93897</u> <u>8.43</u> <u>24.1</u>	aimali	Latitude
	7 ++-ciew T	TWS15-48 2	TWS15-50 2	TWS15-52 2	TWS15-53 2	TWS15-88 2	TWS15-89 2	TWS15-93 2	TWS15-94 2	Hot Spring	TWS15-22 2	Table 2: Taimali stream characteristics	Sample Name

Ta

Sample Name Latitude	. 1	<u>Longitude</u>	<u>Modal Slope</u>	Elevation 18	<u>nge, m</u> abc	Elevation range, m Mean Elevation, m above s.l.	<u>Fractional</u> <u>Catchment</u> landslide area	<u>Fractional</u> <u>Catchment</u> landslide volume	<u>Catchment area</u> , <u>m²</u>	Fractional Channel Fractional Channel Fill pre Morakot Fill Post Morakot	Fractional Channel Fill Post Morakot
22.59175	3175	120.93216	× 91	26	1030	734	0.0002	0.0001	<u>3129659</u>		
22.59484	<u> 1484</u>	120.94132	ST H	80	<u>661</u>	508			<u>496256</u>		
22.58802	3802	120.92323	хти -	32	<u>285</u>	<u>364</u>			8467		
22.58496	<u>3496</u>	120.92101	x M	22	732	547	0.0089	0.0203	1023265		
22.58763	<u> 3763</u>	120.91415	्या	<u>24</u>	1196	790	0.0010	0.0014	<u>9861469</u>		
22.58	22.58109	120.91301	x 91	28	<u> 799</u>	<u>643</u>	0.0030	0.0077	3359874		
22.57508	7508	120.88979	× 91	23	872	723	0.0350	0.0414	3652240		
22.57	7427	120.88347	x 90	28	<u>1126</u>	740	0.1665	0.7246	<u>5461061</u>	0.0325	0.0328
22.56	1265	120.87585	×*#	33	781	711	0.0180	0.0857	1211594		
22.57	7248	120.82725	41	<u>40</u>	745	<u>868</u>	0.0740	0.1187	357320		
22.58458	<u> 3458</u>	120.81979	x - H	33	<u>2653</u>	1473	0.1170	0.2774	<u>62281990</u>	0.0218	0.032
22.56	<u>5602</u>	120.84565	x 91	23	1007	898	0.0690	0.1005	4423928		
22.57761	19/1	120.85395	894 1	31	1308	795	0.0880	0.2111	25323473	0.0466	0.1268
22.57609	609	120.86786	x 91	<u>26</u>	<u>959</u>	<u>805</u>	0.1480	1.2017	3012380	0.0214	0.0332
22.58842	<u>8842</u>	120.84806	x - 11	31	225	<u>631</u>	0.1293	<u>0.1646</u>	145453		
22.59241	<u> 1241</u>	120.84619	x 91	<u>29</u>	<u>484</u>	<u>666</u>	0.0170	0.0637	596003		
22.62	22.62438	120.8435	×-18	33	<u>777</u>	1115	0.1660	0.3246	<u>5670550</u>	0.0113	0.0348
22.59696	<u> 9696</u>	120.84148	र वा	<u>26</u>	1032	<u>806</u>	0.1150	0.1833	4462662	0	0.0512

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Fractional Channel Fill Post Morakot	
Fractional Channel Fill pre Morakot	
Calcite saturation index	
Catchment area, m ^ª	
Fractional Catchment landslide volume	
Fractional Catchment landslide area	
<u>Landslide</u> volume, m ³	
<u>Landslide</u> area, m² (±10%) →	
TDS, https://	
DIC, µmol/f (±10%)	
SO4^{2-,} µmol/1 (±10%)	
СІ-, µшо / (±10%)	
Sr ²⁺ - µmol/ 1 (±2%)	
Si, µmol/ł (±3%)	
<mark>Nta⁺;</mark> µtmol/ 1 (±6%)	
Mg ²⁺ - pumol/1 (±4%)	
Ca ²⁺ , µmol/l K ⁻ , µmol/l Li ⁺ , µmol/l (±2%) (±2%)	
K ⁺ , µmol/1 (±5%)	
i a ²¹, µmol/l (±3%)	
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365	201	535	585	195	367	226	282	582	527	567	162	173	199	15 4	13	4 4	202	530	375		587	274	552	572	569	587	190	201	206	186	219	530	1 5	368
571	200	469	253	699	334	317	365	115	571	381	36/	452	406	807	963	541	312	302	609		983	638	381	1 5	513	377	326	381	318	322	330	247	169	301
1016	1117	85 7	925	6/9E	185	373	179	240	4	545	1267	3476	1643	1081	1412	1187	677	153	486		1189	<u>1</u> 448	478	629	876	537	6//	562	614	974	350	2	106	137
1.92	lbdl	Hed	lbdl	5.82	IP4	194	lbdl	lbdl	Hbd	lbdl	3.42	1.52	4.38	4.80	61:9	3.45	lbdl	lbdl	3.14		9.94	5:71	194	194	1941	lbdl	lbdl	lbdl	lbdl	HP4	1941	lbdl	hd	hdl
13	4	52	4	81	64	5	168	4	8	09	25	<u>tt</u>	56	57	8	26	30	112	120		58	4	5	윎	θ	61	57	50	17	3 6	30	18	30	25
																							4		++									
2716) 2862	5 <u>15</u> 34	5 <u>1324</u>	+ 1019	4 3374	5 2123	4462	5 <u>3946</u>) 2588	5 2975	t 1475	3 <u>1666</u>	4337	3 <u>1680</u>	5 1926	5 1373	t 3308	3 4658	¢ 6954		t 8344	4 3813	4 1164		+ 1801	7 892	6 1535	s 1357	5 <u>1309</u>) 2627	1326	5 1732) <u>2703</u>	+ 2069
7.63 22.2	8:23 21.0	8.06 21.6	8.45 21.3	8.68 21.1	7.67 21.7	8.36 19.5	7.30 21.4	8.26 21.5	8.29 21.0	<u>8.26 24.5</u>	7.49 22.4	39 27.8	7.99 23.7	33 20.8	54 <u>19.5</u>	8.46 20.5	8.43 22.4	7.76 29.8	8.19 30.5		32:4	46 <u>23.4</u>	8.42 20.5		8.34 20.1	8.42 19.7	8 .51 20.6	36 19.8	8.47 20.5	<u>8.40 19.9</u>	47 18.3	5 3 18.5	7.77 20.0	8-46 22-1
												309 8.39		692 7.83	718 7.64						86.1 - 1.98	918 7.46						301 8.56			585 8.47	725 8.53		
7 120.90475	8 120.94849	5 120.92747	3 120.92238	2 120.89299	4 120.87698	8 <u>120.87584</u>	3 120.82355	+ 120.8213	6 120.8312	8 120.85339	7 120.68069	2 120.69309	4 120.66478	9 120.71692	7 120.71718	5 120.88542	9 <u>120.848</u> 4	3 120.84421	5 120.846		23.61785 120.89787	2 120.89918	5 120.93216		2 120.92323	6 120.92101	3 120.91415	9 120.91301	8 120.88979	7 120.88347	+ 120.87585	8 120.82725	8 120.81979	2 120 84565
22.52297	<u>22.59528</u>	22.58965	22.58543	22.57942	22.57541	<u>22.56998</u>	<u>22.57503</u>	22.58521	22.57556	<u>22.57318</u>	<u>22.51877</u>	22.53412	22.76694	22.73699	22.73627	22.44895	22.58739	22.60323	22.61525		23.6178:	23.62402	22.59175	22.5 9484	<u>22.58802</u>	<u>22.58496</u>	22.58763	22.58109	<u>22.57508</u>	22.57427	<u>22.56971</u>	<u>22.57248</u>	22.58458	00995 CC
TWS15-17	TWS15-26	TWS15-31	TWS15-33	TWS15-38	TWS15-41	TWS15-42	TWS15-46	TWS15-47	TWS15-49	TWS15-51	TWS15-63	TWS15-65	TWS15-68	TWS15-72	TWS15-73	TWS15-86	TWS15-87	06-51SWT	16-51SWT	Chenyoulan seepage	CH1307-101	CH1310-40 Taimali Streams	TWS15-23	TWS15-25	TWS15-32	TWS15-34	TWS15-35	TWS15-36	TWS15-39	TWS15-40	TWS15-43	TWS15-44	TWS15-48	1WS15-50

0.1268	0.0332			0.0348	0.0512		
0.0466	0.0214			0.0113	θ		
0.7	0.8	‡	0:1	1 ;	‡		6.0
25323473	3012380	145453	596003	5670550	4462662		
0.2111	1.2017	0.1646	0.0637	0.3246	0.1833		
0.0880	0.1480	0.1293	0.0170	0.1660	0.1150		
7374	7657	12709	6275	<u>12287</u>	8388		100150
1574	0661	3068	5571	2880	2711		110£
2280	2138	3734	948	3761	1933		235
56	37	4	35	50	5 6		40124
1 1	1 4	17	6	55	12		øþ
225	161	296	275	217	226		3643
305	296	480	348	273	336		46541
138	1281	346	146	172	134		38
IP4	hdł	lbd	lbd	lbd	lbd		1191
78	61	78	40	129	61		862
2736	1678	4642	1902	4815	5965		#
27.1	25.5	22.5	<u>22.0</u>	25.1	24.3		24.1
8.19	8.46	8.25	8.46	8.27	8.41		8.43
120.85395	120.86786	120.84806	120.84619	120.8435	120.84148		120.93897
22.57761	22.57609 4	22.58842 H	22.59241 -	<u>22.62438</u>	22.59696		22.59343 120.93897
TWS15-52	TWS15-53	TWS15-88	TWS15-89	TWS15-93	TWS15-94	Hot Spring	TWS15-22

All samples are shown uncorrected for either cyclic or hydrothermal input. ** Charge balance calculations suggest no DIC, meaning a calcite saturation index is not possible to calculate.











