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► To cite this version:

Xiaobai Ruan, Albert Galy. On the significance of periglacial conditions in active mountain belts for chemical weathering processes: Insights from the Chayu area, SE Tibet. Chemical Geology, 2021, 585, pp.120581. 10.1016/j.chemgeo.2021.120581. hal-03400683

HAL Id: hal-03400683 https://hal.science/hal-03400683

Submitted on 5 Jan 2024

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Version of Record: https://www.sciencedirect.com/science/article/pii/S0009254121005246 Manuscript_c58e69740d89bf504a0a493c435bbcdc

Ruan & Galy

On the significance of periglacial conditions in active mountain belts for
chemical weathering processes: insights from the Chayu area, SE Tibet
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Highlight
• Periglacial conditions in active mountain belts enhance chemical weathering.
• The weathering processes in the Chayu catchment are dominated by the preferential dissolution of reactive phases such as trace carbonate and sulphide and the preferential release of K from sheet silicate (biotite, chlorite).
• In the south, under monsoonal climate, such preferential weathering can be seen in bedrock landslide scars on granitoid lithology.
• In the north, the periglacial conditions coupled with physical erosion dominated by landsliding provide a strong rock exposure mechanism and the preferential weathering is widespread.
Abstract
Chemical weathering is a key player of the long-term (Ma) timescale carbon
cycle. Weathering processes under alpine periglacial environment could be important
during the Quaternary glacial-interglacial cycles, but such timescale is difficult to
fully apprehend for the weathering impact on C cycles. The Chayu river catchment in
SE Tibet spans a geographical gradient from a periglacial environment in the north to
a monsoonal environment in the south and we have investigated the various chemical
weathering along this climatological gradient. The lithology of the catchment is
dominated by granifold rocks. Due to the active tectonic activity and the periglacial environment, mass wasting deposits including landslide deposits, talus and rock

33 glaciers are found in the catchment. In order to figure out the influence of mass 34 wasting process on water chemistry, we sampled seepage water from the mass 35 wasting deposits and nearby stream water for comparison. Cyclic input contributes 36 5.7 to 8.2 % to the dissolved load of the water samples, suggesting the dominance of 37 weathering contribution within this granitoid catchment. The contributions from 38 sulfuric acid and carbonate to chemical weathering are estimated based on the 39 elemental data and the oxidation of sulfide as the source of dissolved sulfate. In the 40 monsoonal South Chayu, the K*/Si, Ca*/Si ratios of the bedrock landslide seepages 41 are higher than the streams by 265% and 117%, respectively. Together with the high 42 contribution of carbonate (~80%) and sulfuric acid (20 to 40%), the results indicates 43 that landslide deposits have strong influences on local water chemistry by processes 44 identical to bedrock landslides of greywacke and carbonate lithology in active 45 mountain belts, suggesting that the enhanced weathering by landslide process can 46 apply to granitoid regions as well. In the North Chayu with periglacial environment, 47 the chemistry of the seepages and streams are nearly identical, and are similar to the 48 landslide seepage water in the south. Such result highlights the role of the periglacial 49 critical zone without the influence of mass wasting deposits that possibly enhances 50 chemical weathering. The high porosity created by repeat freeze-thaw cycles provides 51 more fresh mineral surfaces for water-rock interaction. But, the low temperature and 52 the partial availability of water, restricted to thaw periods, provide strong kinetic 53 limitations that inhibit the silicate weathering. Such features of the alpine periglacial 54 critical zone led to the preferential weathering of the most reactive mineral phases including calcite, sulfide, and the interlayer K⁺ of sheet silicates (biotite, chlorite), 55 56 resulting in the characteristic periglacial water chemistry. The influence of periglacial 57 environment on local water chemistry may apply beyond the North Chayu and may 58 play an important role in modulating the chemical weathering within the transition 59 between Quaternary glacial and interglacial period.

60

61 **1. Introduction**

The Quaternary climatic cycles may bring fluctuations of chemical weathering rate and flux from regional to global scale (e.g. Colin et al., 1999; Foster and Vance, 2006; Hein et al., 2017; Vance et al., 2009; Wan et al., 2017; Wilson et al., 2015). As a key feature of the climatic cycles, the expansion and shrinkage of ice sheet induce downstream climatic impact to the broad high and middle latitudes regions (e.g. Ruddiman, 2014), including the occurrence of periglacial environment over large area of land surface during glacial periods (e.g. Hubberten et al., 2004; Marshall et al., 69 2021). Periglacial environment is defined as "the climatic conditions, processes, 70 landforms, landscapes, sediments and soil structures associated with cold, nonglacial 71 environments" (Murton, 2021). The conditions of chemical weathering under 72 periglacial environment can be peculiar since repeated freeze-thaw cycles can 73 increase the porosity in bedrocks (e.g. Hallet et al., 1991; Marshall et al., 2021, 2015; 74 Sun et al., 2020) that might have played a role in the response of weathering to 75 Quaternary climate change (Foster and Vance, 2006; Vance et al., 2009).

76 Periglacial conditions are also prevailing in the cold alpine part of mountain belts, 77 and the periglacial frost cracking process is not just caused by low temperatures but 78 also by the large amplitude of diurnal or seasonal temperature variations in such 79 setting (e.g. Draebing and Krautblatter, 2019; Mair et al., 2020). This process is also 80 responsible for the ubiquitous talus deposits beneath bedrock cliffs covering valley 81 slopes (Hales and Roering, 2007), and cause high rate of physical erosion in alpine 82 mountain areas (e.g. Egholm et al., 2015; Hales and Roering, 2007). However, 83 bedrock landsliding is the dominant mass wasting process of active mountain belts 84 (Hovius et al., 2000, 1997) and the chemical weathering flux can be significantly 85 impacted by typical reactions occurring within the landslide deposits (Emberson et al., 86 2016a). The characteristics of such chemical reactions (a strong kinetic limitation and 87 the preferential weathering of the most reactive phases) are promoted by a high 88 surface area of fresh rocks and high porosity rock mass (Emberson et al., 2018, 2017, 89 2016b). The processes of frost cracking and landsliding share the commonality of 90 breaking bedrocks and creating fresh mineral surfaces for chemical weathering. 91 Therefore, it is plausible to argue that the frost cracking under periglacial environment 92 may have strong impact on the composition of local water as well.

93 In this study, we investigated the water chemistry in the Chayu catchment in 94 Southeast Tibet, a place that spans large geographical gradient from periglacial to 95 monsoonal environment, dominated by granitoid lithology, and experiencing active 96 tectonic activity. The aim of this study is first to test the hypothesis that the mass 97 wasting deposits in periglacial environment has the effect of enhancing chemical 98 weathering and preferential weathering of labile phases, identical to the weathering of 99 bedrock landslide deposits (Emberson et al., 2018, 2017, 2016b); and second to test if 100 elevated chemical weathering in landslide deposits extends to granitoid lithologies as 101 well. By comparing the seepage waters from mass wasting deposits and the nearby 102 stream waters in two different environments in the Chayu catchment, we found (1) the 103 chemistry of both seepage and stream waters are similar and concentrated under the 104 periglacial environment, and suggestive of the preferential dissolution of calcite and

105 leaching of K from biotite coupled with sulfuric acid under periglacial environment, 106 and (2) the bedrock landslide seepages are more concentrated than streams under 107 monsoonal environment on granitoid lithology extending the characteristics of 108 bedrock landslides chemical reactions to this lithology.

109

110 **2. Study area and sampling protocol**

111 The Chayu river is the upper part of the Lohit river, an eastern tributary of the Brahmaputra. It drains the eastern Himalaya for an area of ~18000 km². The 112 113 geography of the catchment is diverse and related to the variation in altitude. The 114 north of the catchment (North Chayu) is in a semi-arid periglacial alpine environment. 115 The surrounding mountains peak above 6000 m and the riverbed is above 3500 m. As 116 altitude decrease, the south of the catchment (South Chayu) reaches a subtropical 117 monsoonal forest environment. The lowest point of the riverbed in this area is around 118 1000 m. Near Chayu city, where the riverbed altitude is ~2000m, the mean annual 119 temperature (MAT) is 11.9° and the mean annual precipitation (MAP) is about 800 120 mm (http://www.weather.com.cn/cityintro/101140404.shtml). 30 km NW of North Chayu, outside the catchment at 3850 m, the observed MAT is of 2.4° at the Ranwu 121 122 lake (Ju et al., 2017). With a mean altitude around 4600m of the North Chayu (Table 123 4), and a local thermal gradient of -6.1° /km, the MAT for North Chayu is estimated to 124 be -2.2°C. A rough estimation for MAP in the North Chayu from a satellite 125 observation-based model is less than 200 mm/yr (Anders et al., 2006).

Mass wasting deposits are well developed in the valley, especially in the north of 126 127 the catchment. Several types of mass wasting deposits can be identified based on our field survey combined with satellite images (e.g. Google Earth or ArcGIS World 128 129 Imagery). The most prevailing type is talus (also referred to as "scree" in some 130 literature), which is the accumulation of loose rock debris, usually presented as cones, 131 at the foot of steep slopes (Ballantyne, 2002). Such landscape is frequently found in 132 alpine periglacial environment over the world (e.g. Hales and Roering, 2007 and the 133 references therein). Materials of the talus are mostly supplied by rockfall of the upper 134 rock cliffs (Ballantyne, 2002), triggered by frost cracking processes caused by the 135 stress during pore water freezing and ice segregation (e.g. Murton et al., 2006). The 136 talus deposit can experience continuous accumulation or reworking for century to millennium long (e.g. Curry and Morris, 2004). Despite the temperature-related 137 138 processes, rock fall triggering mechanisms also include earthquakes (e.g. Matsuoka 139 and Sakai, 1999) and unloading during deglaciation (e.g. Hinchliffe and Ballantyne, 140 1999). During the Last Glacial Maximum, the high-altitude part of the Chayu valley

141 could have been covered by glacier, because the maximum extension of LGM end 142 moraines in the surrounding eastern Himalaya areas (Namcha Barwa and Parlung 143 Tsangpo Valley) are between 2800 and 3000m (Hu et al., 2020; Zhou et al., 2010). 144 However, considering the much lower MAP of the North Chayu, which means the LGM glacier in North Chayu should not reach the elevation as low as the two places, 145 146 the newly supplied fresh rock debris on the North Chayu talus, and the intense 147 tectonic activity in the area (e.g. Mukhopadhyaya and Dasgupta, 2015), the talus 148 development in North Chayu cannot be fully ascribed to the Holocene deglaciation. 149 The second type of deposit is rock glacier, which corresponds to thick talus deposits 150 that contain permafrost and formed by the subsequent creep and deformation of such 151 rock/ice mixture (Barsch, 1992; Haeberli et al., 2006). Rock glaciers are often seen in 152 continental climate (Barsch, 1992), and favour places with high talus production rates 153 (Janke, 2007). The north part of the Chayu catchment being on the leeward slope of 154 the Mt. Kangri Garpo, it provides suitable climatic conditions for the rock glaciers. In 155 the Chayu catchment, some "talus" fans are indeed fed by the frontal collapse of rock glaciers (e.g. the talus at 29°19'54.3"N, 97°5'51.0"E). The third type of deposit is 156 157 associated to bedrock landslide, which refers to the catastrophic movement of mass 158 bedrock along failures by gravity (Korup et al., 2007).

159 Geologically, the Chayu river catchment belongs to the eastern part of the Lhasa 160 terrane, east of the Namche Barwa Himalayan Syntaxis. The Lhasa terrane was the 161 latest geological block rifted from Gandwana, drifted northward and accreted to 162 Eurasia before its collision with the Indian continent (Zhu et al., 2011). The collision 163 with the Indian plate has been accommodated by the Tibet and Lhasa terrane through 164 strong deformation along strike-slip faults responsible for the eastern extrusion and 165 EW extension (e.g. Tapponnier, 2001). In the studied area, a dextral strike-slip fault, 166 the Jiali Fault, goes across the eastern Lhasa terrane, and the fault splits into two 167 branches in the Chayu river catchment (Fig. 1), the Parlung fault in the north and the 168 Puqu fault in the south (Lee et al., 2003). The consequences of the dextral strike-slip 169 are a N-S zonation of the lithology distribution, and similarities with the regions to the 170 west. In the Chayu river catchment, there are three basic lithological units: (1) the 171 metamorphic Precambrian basement; (2) the Mesozoic cover, and (3) the Eastern 172 Himalayan Batholith (EHB). The Precambrian basement contains an older part 173 bearing schist, leptynite, gneiss, and minor amphibolite and marble (Xu et al., 2013), 174 and a younger Neoproterozoic part made of greenschist-facies suite of clastic rocks 175 (Xie et al., 2007). The Mesozoic cover is composed of Ordovician-Devonian 176 carbonate rocks and Carboniferous-Permian slate (Dong et al., 2019). The EHB is the 177 dominant unit in the study area (Chiu et al., 2009; Dong et al., 2019; Lin et al., 2012;

178 Pan et al., 2012; Zhu et al., 2011). In the Chayu river catchment, intrusions of the 179 EHB mainly occurred in the Early Cretaceous but also in the Paleocene. The limited 180 mineralogy investigation for the EHB in the studied area show that it is constituted by 181 K-feldspar ($\sim 40\%$), plagioclase ($\sim 30\%$), quartz ($\sim 20\%$), biotite (~ 5 to 10%) and 182 accessary minerals such as hornblende, muscovite, apatite, and opaque phases (Chiu 183 et al., 2009; Lin et al., 2012). Based on the age, the location relative to the Parlung 184 and Puqu faults and the petrology, the EHB can be divided in three units, which are 185 the Demulha Batholith, the Azhagong Batholith and the Chayu Batholith along the 186 river path from north to south (Fig. 1, Lin et al., 2012). The elemental compositions of 187 these three batholiths are distinctive (Lin et al., 2012). Abundances of CaO (wt%) is 188 the highest in the Azhagong Batholith and the lowest in the Demulha Batholith and 189 anti-correlated with abundances of K₂O%. According to AFT thermochronology, this area experienced strong exhumation (2.3-10 mm a⁻¹) in the Quaternary (Sun et al., 190 191 2015) associated to a significant earthquake activity (e.g. Mukhopadhyaya and 192 Dasgupta, 2015). This is corroborated by the basin hypsometry that indicates intense 193 crustal shortening and uplift for the Chayu catchment (Zhang et al., 2020).

194 Field survey and sampling for the Chayu river catchment was conducted in 195 September 2019. A series of samples are collected corresponding to seepages from the 196 mass wasting deposit, bedrocks, and water and solid samples for streams nearby the 197 sampled seepages. Seepages are usually found at points where water exfiltrate from 198 the bottom of the mass wasting deposits (Table 1). For the landslide sampled in the 199 south catchment, seepages water is sourced from the scar of the landslide and flowing 200 on the surface of deposit. Some large deposits had several seepages, and we only 201 collected samples for those with high fluxes but also representative of the range of pH 202 and conductivity. Streams close the seepage sampling sites were also sampled. In the 203 south catchment, three streams near the landslide deposit (Fig. 1) correspond to small 204 $(3.6 \text{ to } 24 \text{ km}^2)$ catchment draining the same lithology under similar climatic 205 condition and with no evident mass wasting deposit in their upstream catchment. In 206 the north, the greater occurrence of mass wasting deposits, and the poor accessibility 207 implied that only one sampled stream (RYG 153) is without evident deposits. Streams 208 having larger upstream areas (> 100 km²) but with some deposits (RYG 114, RYG 209 126 and RYG 161) were also sampled. The locations of the seepages and stream 210 samples are shown in Fig. 1. The seepages sampled in the north catchment are mostly 211 from the Demulha Batholith, while the seepages in the south are from Chayu 212 Batholith. This study is far from being extensive since some mass wasting deposits 213 were unreachable (e.g. the bedrock landslide deposit at 28°57'45.9"N, 97°27'46.6"E, 214 on the riverside of a tributary too remote to access).

215

216 **3. Methods**

217 **3.1 Sampling and analytical methods**

The water samples are collected using HDPE syringe, filtered in situ through 0.2 µm nylon Millipore filter and stored in two HDPE bottles and one tube free of air. The bottles and tube are fully rinsed with filtered sample water before using. A bottle, for cations element analysis, was acidified with distilled nitric acid immediately after sampling. The second bottle is for anion analysis. The conductivity, temperature, and pH for the water samples are measured in situ with the thermo-hygrometer HANNA 9565.

225 The major elements were measured on a Thermo Scientific X-series 2 ICP-MS in 226 CRPG, using USGS M214, P62 and T223 as external standards and quality control 227 (QC). There was no systematic drift during the measurement, and the relative standard 228 deviation for all the elements are always below 10%. The Si concentration in water is 229 determined by the colorimetric method of the Mo blue complex (Mullin and Riley, 230 1955) in CRPG, with an uncertainty below 10%. The F⁻, Cl⁻, SO₄²⁻ and NO₃⁻ are 231 measured on Dionex ICS-1000 Ion Chromatograph at GFZ with USGS M214, P62, 232 M212 and M222 as QC. The uncertainty of anions is less than 10% (Emberson et al., 233 2016b). Bicarbonate concentration is estimated by charge balance. Previous studies of 234 the dissolved chemistry of Himalayan and Southern Tibetan rivers that have measured 235 the bicarbonate by titration in the field show no systematic difference with the values 236 calculated by charge balance (e.g. Galy and France-Lanord, 1999; Hren et al., 2007). 237 The individual differences are actually within the 10% uncertainty of the 238 quantification of the others anions and cations. Therefore, the entire dataset of anions 239 and cations (Table 3) should be considered with uncertainties of 10%. The TDS is 240 calculated by summing the weight of elements of Si and all the cations, anions in per 241 unit volume.

242

243 **3.2 Mapping the deposits and the stream catchments**

The areas of the sampled mass wasting deposits are mapped through ArcGIS using the detailed GPS location recorded during the field survey, and the satellite image of ArcGIS World Imagery. The surface covered by fresh rocks on the deposits are usually much brighter than the surface colonized by vegetation including lichen. We mapped the area colonized by vegetation on each deposit and calculated the vegetation coverage (Table 1). For the sampled streams, we extract their catchments with the Hydrology tools in ArcGIS based on the 30-m resolution ASTER GDEM (Advanced Spaceborne Thermal Emission and Reflection Radiometer Global Digital Elevation Model) data (Geospatial Data Cloud, http://www.gscloud.cn/), and for streams that is too small to be extracted, the catchments were mapped on ArcGIS by hands. Then, the average altitude of the area or catchment of each deposit and stream is calculated (Table 1 and 2).

256

257 **4. Results**

258 The cation, anions, silicon concentrations and TDS of the seepages and streams 259 are listed in Table 3. The pH value and water temperature are also listed, as well as the 260 altitude of the sampling sites. The data are grouped by the North Chayu and the South 261 Chayu. In the North Chayu, the seepages have a wide range of TDS, varying from 55 262 to 225 mg/L but describing a negative trend with the sampling altitude. The streams 263 have variable TDS as well but slightly lower than the seepages (49 to 173 mg/L). In 264 the South Chayu, the TDS values are generally lower than in the North Chayu, and 265 the seepages have higher TDS values (102 to 139 mg/L) than the streams (29 to 77 266 mg/L).

267 In terms of anions, the HCO_3^- is the dominant load. It makes up 71 to 95 Eq% (%) 268 in equivalent units) of total anion in the north seepages, 59 to 90 Eq% in the north 269 streams, 54 to 72 Eq% in the south seepages and 70 to 76 Eq% in the south streams 270 (Fig. 2). Cl⁻ has concentrations between 4.4 and 14.6 µmol/L and corresponds to a 271 minor component (<2 Eq% of total anion), suggesting that 1) no minor evaporitic 272 lithologies are present, and 2) cyclic input is very limited in the Chayu catchment. SO_4^{2-} is the second most important anion, which makes up 4 to 28 Eq% of total anion 273 274 in the north seepages, 9 to 41 Eq% to the north streams, 19 to 36 Eq% in south seepages and 11 to 12 Eq% in the south streams (Fig. 2). In terms of cations, Ca^{2+} is 275 276 dominant in the north streams and all the seepages (57 to 87 Eq% of total cation) but 277 appears to be lower in the south streams (52 to 69 Eq%). Na⁺ shows the opposite 278 systematics, making up 13 to 23 Eq% in the south streams, which is higher than all 279 the other water samples (2 to 11 Eq%). K⁺ is systematically higher in the South Chayu 280 and the highest K⁺ concentration is observed in the seepages. The proportion of 281 dissolved Si in TDS span a similar range in the seepages and the streams in the north, 282 varying from 2.8 to 12.4% of TDS. The Si load in TDS is higher in the South Chayu. For seepages it makes up 9.9 to 14.4% and for streams it makes up 19.6 to 37.1%. The 283 284 comparison between the seepages and the streams shows a greater difference in the South Chayu, where the concentrations of SO_4^{2-} , Ca^{2+} and K^+ in the seepages are 285

clearly more elevated than in the streams. In the North Chayu, seepages and streamsare similar in composition.

288 In comparison with the rivers draining silicate lithology in (1) the Lhasa terrane 289 (e.g. Hren et al., 2007; Yu et al., 2021, Fig. 2) and (2) the southern flank of the Himalaya (e.g. Galy and France-Lanord, 1999), the north Chayu seepages and streams 290 291 are similar with the Nyang River, in the Eastern part of the Lhasa terrane while the 292 south Chayu water samples with greater contribution of the alkalis is closer to the 293 rivers draining silicate lithologies (e.g. Lhasa River and southern Himalayan flank 294 rivers draining gneiss). However, the surface water samples from the Chavu 295 catchment have a lower Cl⁻ concentration and Cl⁻/anions ratio (Fig. 2), which is 296 different from many of the Lhasa terrane rivers impacted by hot-springs and/or 297 evaporite formations (Hren et al., 2007; Yu et al., 2021).

298

299 **5. Discussion**

300 5.1 Cyclic input correction

The rain composition (Table 4) from the network of Southeastern Tibet Stations (SET, Liu et al., 2013) is used in our study to correct for the cyclic input. The closest station to the Chayu valley is near Nyinchi city, about 200 km to the west to our study area. Based on the low Cl⁻ concentrations in our samples, and the lack of correlation between Cl⁻ and SO_4^{2-} , the weathering of evaporite lithology is unlikely. Therefore, we adopt the assumption that all the dissolved chloride in surface water is from cyclic input, and the correction is based on the following standard equation:

308
$$[X]^{*} = [X]_{sample} - [X]_{rain} \times ([Cl^{-}]_{sample} / [Cl^{-}]_{rain})....(1)$$

309 Where [X] is the measured concentration of any species, and [X]* denotes the 310 concentration corrected for the cyclic input. The atmospheric contribution to our 311 samples represents 5.6 to 20.0 Eq% for Na and 0.8 to 21.5 Eq% for K, whereas Mg 312 and Ca are less impacted (0.1 to 5.2 Eq% and 1.5 to 14.5 Eq%, respectively). The 313 highest cyclic contributions are found in some seepages from the North Chayu, 314 associated with a small discharge of these seepages. More generally, the cyclic 315 contribution in the seepages is lower in the South Chayu than in the North Chayu. The 316 Chayu catchment is also poorly impacted by human activities, and this is specially the 317 case for the sampled seepages and streams. So [X]* is likely to be representative of 318 natural mineral weathering.

320 **5.2 Source of sulphate**

321 We use X_{SO_4} value to represent the proportion of sulphate in anions corrected 322 for the cyclic input, defined by the following equation:

323
$$X_{SO_4} = 2 \times \left[SO_4^{2^-} \right]^* / \left(2 \times \left[SO_4^{2^-} \right]^* + \left[HCO_3^{-^-} \right]^* \right) \dots (2)$$

324 Since there is no evident evaporite unit in the lithology of the Chayu catchment (Fig. 1) and given the lack of correlation between Cl^{-} and SO_4^{2-} , the dissolved 325 sulphate can be considered as the result of sulphide oxidation. All the X_{SO_4} values 326 327 are higher than 0.04 and 70% are above 0.1, suggesting the prevalence of sulphide 328 weathering in the Chayu catchment. In the Lhasa terrane, sulphide minerals are found 329 associated with Cu-bearing deposits hosted in granitoid rocks (e.g. Fu et al., 2017; 330 Hou et al., 2015), with S-bearing assemblages such as sphalerite + galena + pyrite + 331 chalcopyrite (Hou et al., 2015). However, the highest sulphate concentrations are 332 found in the streams RYG 125, RYG 126 and RYG 161. They are within a small area 333 of 300 km², with RYG 125 and 126 draining mostly pre-Cambrian metamorphic sedimentary rocks, and RYG 161 draining Mesozoic sedimentary formations (Fig. 1). 334 335 The prevalence of sulphate in the Chayu water samples could indicate the contribution of the oxidation of disseminated sulphide in the granitoids while high $[SO_4^{2-}]$ in some 336 337 streams are related to sedimentary lithology locally enriched in sulphide, although 338 little documented in that part of the Himalayas and Tibet but recognised ~800km to 339 the West in the Tethys Himalaya (e.g. Feng et al., 2017).

340

341 **5.3 Carbonate weathering**

342 The contribution of carbonate weathering (X_{carb}) can be estimated using the 343 dissolved load corrected for cyclic input (e.g. Galy and France-Lanord, 1999):

344
$$X_{carb} = (2 \times Ca^*_{carb} + 2 \times Mg^*_{carb})/(Na^*_{riv} + K^*_{riv} + 2 \times Ca^*_{riv} + 2 \times Mg^*_{riv}).....(3)$$

Where Na_{riv}^* , K_{riv}^* , Ca_{riv}^* and Mg_{riv}^* are measured concentrations corrected for cyclic input. The Ca_{carb}^* and Mg_{carb}^* are calculated by subtracting silicate-derived Ca and Mg from the Ca_{riv}^* and Mg_{riv}^* (Eq. (4) and Eq. (5)). This assumes that there are no evaporite-derived cations in the water, likely to be the case given the very low dissolved chlorine concentration (Table 3).

350
$$Ca_{carb}^* = Ca_{riv}^* - Ca_{sil}^* = Ca_{riv}^* - Na^* \times (Ca/Na)_{sil}$$
.....(4)

351
$$Mg_{carb}^* = Mg_{riv}^* - Mg_{sil}^* = Mg_{riv}^* - K^* \times (Mg/K)_{sil}$$
.....(5)

Of course, the (Ca/Na)sil and (Mg/K)sil (expressed in molar ratio) have to 352 353 correspond to the local composition of the silicate endmember (e.g. Galy and 354 France-Lanord, 1999). According to the mean rock composition in each unit of EHB 355 (Lin et al., 2012), the (Ca/Na)_{sil} for the Chayu Batholith in South Chayu and the 356 Demulha Batholith in the North Chayu are 0.277 and 0.117, respectively, and the 357 mean (Mg/K)_{sil} for the Chayu Batholith is 0.049. For the Demulha Batholith, the MgO 358 content is not formally listed in Lin et al. (2012), however, their limit of quantification 359 and the overall trend between MgO and SiO₂ suggest that an average MgO content of 360 0.10±0.02 (weight%). Therefore, we use a (Mg/K)_{sil}=0.023±0.007 for the Demulha Batholith. The X_{carb} is based on the bulk chemistry of the silicate endmember, and is 361 362 likely to only be an inaccurate estimate for the relative contribution of carbonate and 363 silicate as soon as incongruent weathering becomes important. For instance, a 364 possible incongruent release of K could be the result of the faster release of K during 365 the weathering of biotite (e.g. Chae et al., 2006; Malmström and Banwart, 1997) and 366 may significantly change the Mg/K ratio of the weathered silicate endmember. The 367 bulk (Mg/K)_{sil} has to be regarded as an upper limit of the (Mg/K)_{sil} ratios, and we also 368 consider the ratios to be 80% less according to the biotite dissolution experiments 369 (Malmström and Banwart, 1997). In this case, the (Mg/K)_{sil} for the Chayu Batholith 370 and Demulha Batholith used in our estimation for X_{carb} values are 0.008-0.049 and 0.005-0.023, respectively. Results shows that the variation of (Mg/K)_{sil} caused by the 371 372 potential incongruent release of K has little impact on the estimation of X_{carb} (Table 3). 373 A faster dissolution of Ca-bearing silicates (Ca-plagioclase or epidote, e.g. Blum and 374 Stillings 1995; Oliva et al. 2003) would correspond to an incongruent dissolution of 375 silicates and would raise the inaccuracy of the relative contribution of carbonate and 376 silicate, so only large variations in the X_{carb} can be discussed. Based on the source rock (Mg/K)_{sil} ratios, the X_{carb} is 0.85 to 0.97 and 0.91 to 0.97 for the seepages and 377 378 streams in the North Chayu, respectively, and the values are 0.78 to 0.80 and 0.51 to 379 0.73 in the South Chayu, respectively. So, it is clear that (1) carbonate still contributes 380 the majority solute load (>50%) despite lithologies dominated by granitoids and (2) 381 the North Chayu water samples have higher X_{carb} values than the South Chayu.

All the mass wasting deposits in this study have X_{carb} values above 0.8. In pure granitoid deposits and watersheds (RYG 153, 129, 114, 96, 97 and 99), disseminated calcite is likely the most important carbonate source (e.g. White et al., 1999). For streams partly draining sedimentary lithologies (RYG 125, 126 and 161), sedimentary carbonate formations could be the dominant source.

388 **5.4 Comparison between the seepages and streams**

389 The influence of mass wasting deposits on chemical weathering has already been 390 recognised in the Western Southern Alps (WSA) and in Taiwan by a direct 391 comparison of the seepages from the mass wasting deposits with the nearby streams 392 (Emberson et al., 2018, 2017, 2016a). This approach allows to reduce the effects of 393 different climate and lithology, so it will be applied to the periglacial North Chayu and 394 the monsoonal South Chayu separately. In WSA and Taiwan, the differences between 395 seepages and streams are emphasised when the K*/Si and Ca*/Si ratios and the X_{carb} 396 and X_{SO_A} values are considered. Here, the differences of those indicators are always 397 larger in the South Chayu than in the North Chayu (Fig. 3).

398 5.4.1 The South Chayu

The X_{carb} and X_{SO_4} values and the K*/Si and Ca*/Si ratios are elevated in the seepages. By comparison to the streams, those values are 29.5%, 350.0%, 264.6%, 116.7% higher in seepages, respectively.

402 In the South Chayu, the seepages are from a bedrock landslide and such contrast 403 with the nearby streams is in agreement with the studies of bedrock landslide 404 weathering from the WSA and Taiwan (Emberson et al., 2018, 2017, 2016a, 2016b) 405 although their lithologies are different from the Chayu catchment. Bedrock landslides 406 expose fresh mineral surfaces by generating landslide scar and grind the mobile rock 407 mass together with conducting water into debris and fractures (Emberson et al., 408 2016a). Higher TDS, K*/Si and Ca*/Si ratios, and X_{carb} and X_{SO₄} values in seepages 409 have been associated to processes characteristic of the initiation of chemical 410 weathering with abundant fresh mineral surfaces and fluid supply (Emberson et al., 411 2017, 2016b, 2016a): the fast dissolution of disseminated calcite (White et al., 1999) 412 and sulphide (e.g. Nicholson et al., 1988; Wildaman et al., 2004) and the fast release 413 of interlayer K from biotite (Malmström and Banwart, 1997). Results in the South 414 Chayu, fitting well with previous results for bedrock landslide weathering in 415 sedimentary or metamorphic lithologies indicate that such processes are also 416 pronounced in granitoid bedrock landslides.

417 **5.4.2 The North Chayu**

The results from the North Chayu are in sharp contrast with the results from the South Chayu. For the north seepages and streams, the K*/Si ratios are 0.08 to 0.46 and 0.2 to 0.75, respectively; the Ca*/Si ratios are 3.61 to 11.19 and 3.18 to 13.28, respectively; the X_{SO_4} are 0.04 to 0.28 and 0.09 to 0.41, respectively; and the X_{carb} is 0.85 to 0.97 and 0.91 to 0.97, respectively (Table 3). Regarding these parameters, there are two major differences between the North and South Chayu (Fig. 3): (1) the
differences between the seepages and streams are negligible in the North not in the
South, and (2) the values are high in both the North Chayu stream and seepage waters.

426 The lack of sizable difference between the seepages and the streams may indicate 427 variable weathering conditions in the mass wasting deposits in North Chayu. Given 428 the variable nature of the studied mass wasting deposits (Table 1), weathering 429 conditions might be more diverse than those of fresh bedrock landslides. Indeed, the 430 freshness of the material in talus deposit varies greatly since they can be continuously 431 supplied by new rock mass and that they could be of different age leading to variable 432 weathering rates and yields (Emberson et al., 2016a; Taylor and Blum, 1995). One 433 rough way to estimate the recent activity of a landslide and the relative age of the 434 deposits is by vegetation coverage, because the colonization of vegetation on the 435 deposit needs a stable surface (Innes, 1985). However, in North Chayu, the vegetation 436 coverage of the mass wasting deposits (Table 1) correlate neither with the K*/Si ratios $(R^2 = 0.03, p = 0.04)$ nor the Ca*/Si ratios ($R^2 = 0.11, p = 0.06$) of the corresponding 437 438 seepages, which suggest that the age is not responsible for the large variations in the K*/Si and Ca*/Si ratios of the seepages in the North Chayu. 439

440 The negligible difference between the seepages and streams can be a statistical 441 artefact of the large range in the K*/Si and Ca*/Si ratios and X_{SO_4} (Fig. 3), but two 442 other possibilities should be considered. First, the streams sampled in the North 443 Chayu are significantly influenced by mass wasting deposits. The satellite image 444 shows that talus deposits are ubiquitous on both sides of the valleys for those streams, 445 and there is just one small stream (RYG 153) among those we have sampled, which 446 has no deposits. It is worth noticing that the influence of deposits is widespread for 447 the streams in North Chayu. However, the dissolved load in the streams corresponds 448 to the mixing between a pure mass wasting deposits end-member and the weathering 449 of areas without any deposits. And some streams (RYG 129, 125, 126 and 161) have 450 K*/Si, Ca*/Si, X_{carb} and X_{SO₄} even higher than some seepages (RYG 117, 131, 139-143 and 155, Tables 3). Assuming adding water coming from a pure mass 451 452 wasting deposit end-member in variable proportion seems to have little impact on the 453 dissolved load of the streams. Therefore, the chemical composition of the water from 454 mass wasting deposits and the water characteristic of chemical weathering in North 455 Chayu outside the landslides (vegetated slopes or meadows at the bottom of the 456 valleys) are likely to be similar.

457 Three factors have been indicated as the key factors enhancing weathering in 458 bedrock landslides deposits (Emberson et al., 2018, 2016a): the larger mineral surface 459 for water-rocks interaction, the more favourable hydrological conditions, and the 460 occurrence of fresh rocks. Our results suggest that similar conditions should be more 461 widespread in North Chayu than just restricted to landslides and mass wasting 462 deposits in order to explain the chemistry of the streams.

463 Regarding the geomorphic processes and the climate in this region, the key 464 factor to such conditions is likely to be the periglacial environment. Under periglacial 465 environment, the repeat freeze-thaw process caused by the low temperature and large 466 amplitude of diurnal and seasonal temperature change (Marshall et al., 2021) 467 increases the density of pores and fractures in bedrock (Sun et al., 2020), as well as 468 the permeability (Takarli et al., 2008), thus increasing the surface area for water-rock interaction. The estimated MAT in the North Chayu is within the range (-4 to -15°C) 469 470 suggested to be most effective in making fractures by ice segregation growth (Walder 471 and Hallet, 1985). However, the low temperature of the periglacial environment can in 472 turn exert kinetic limitations on chemical weathering. In addition, in the North Chayu 473 the water supply is limited. Water availability is an important factor limiting the 474 efficacy of chemical weathering (Maher and Chamberlain, 2014). The fresh surfaces 475 area created by freeze-thaw cycles are not readily exposed to chemical weathering since they are ice-filled for a period of time, limiting the availability of fluids (Hall et 476 477 al., 2012). Only during the thaw, the rock fragments are available for weathering. 478 Moreover, low temperatures reduce the silicate weathering rate but also increase 479 carbonate solubility. A change in MAT from 11.9°C to -2.2°C, for the comparison of 480 the South Chayu with the North Chayu, induces a 15% increase in the dissolved 481 carbonate, while the silicate weathering rate decreases by 63% to 90% (using the apparent activation energy of 74 \pm 29 kJ mol⁻¹ of West et al., 2005 and a change in K_{sp} 482 483 for CaCO₃ by 133%, e.g. Bufe et al., 2021). Therefore, silicate weathering in the 484 periglacial environment is likely to be limited by the low temperature while carbonate 485 weathering is slightly increased. In addition water availability is restricted to thaw 486 periods, so periglacial environment could greatly inhibit silicate weathering. But, the 487 freeze-thaw cycles are exposing fresh surfaces, enhancing the preferential weathering 488 of the most reactive phases, e.g. the disseminated calcite and pyrite, and the release of 489 K from the biotite. The importance of such weathering of reactive mineral phases is, 490 however, only relative because silicate weathering is strongly inhibited. The positive 491 correlation between the Ca*/Si ratios and the X_{SQ_4} values for the North Chayu 492 streams (Fig. 4) corroborates the carbonate weathering enhanced by sulphide 493 oxidation. And the positive correlations of K*/Si and Ca*/Si ratios and X_{carb} and X_{SO}, 494 values with TDS* in both the seepage and stream water samples from North Chayu

495 (Fig. 5) suggest the weathering of the labile phases dominate the dissolved load in the496 North Chayu, irrespective to the relative occurrence of mass wasting deposits.

497 To sum up, we attribute the dissolved characteristics in the North Chayu to the 498 periglacial environment. The frost process creates large surfaces areas for water-rock 499 interaction, while the hydrological conditions and low temperature just favour the 500 preferential weathering of labile phases and reduce the silicate weathering rate. As a 501 result, the chemical weathering has specific characteristics: high TDS*, K*/Si and 502 Ca*/Si ratios and X_{carb} and X_{SO4} values.

503

504 **5.5 Implication on periglacial weathering**

505 **5.5.1** Comparison with alpine weathering of granitoids

506 The particularity of the periglacial weathering in the North Chayu can be further 507 supported by the comparison with the large dataset of granitoid watersheds (White 508 and Blum, 1995; Oliva et al., 2003; Gurumurthy et al., 2012; Wu et al., 2013; Zhang 509 et al., 2016; Sun et al., 2019). For a direct comparison, that dataset has been corrected 510 for cyclic input following the same method used for the Chayu catchment. The K*/Si 511 and Ca*/Si ratios do not correlate with MAT, where MAT has been estimated using a 512 local thermal gradient of -6.1°/km and the mean altitude of the corresponding 513 catchment (Fig. 6). The data span a MAT range of 0 to 30°C, and their K*/Si ratios 514 are <0.37 and Ca*/Si ratios are <2.33. A watershed with a K*/Si of 0.66 corresponds 515 to very high [K⁺] of 212 µmol/L in river water, and such values are likely to witness a 516 significant contribution from fertilizer (Sun et al., 2019) and are not considered in the 517 dataset. So with these, a rough range of water compositions draining granitoid 518 lithology under most of the environmental conditions on the earth surface is defined. 519 The data from the South Chayu streams are compatible to this range (Fig. 6a, b). In 520 contrast, the North Chayu data, draining mostly granitoid lithology as well, are much 521 higher, highlighting the effect of its periglacial environment. Although the North 522 Chayu is distinctive to all the other watersheds in the world granitoid dataset, we 523 regard that the alpine periglacial environment is not rare on the Earth surface, but are 524 only rarely studied. The alpine periglacial environment should draw attention since it 525 nurtures the headwaters of most mountain rivers.

526

527 5.5.2 Implications on continental weathering during glacial-interglacial cycles

528 Alpine periglacial weathering may play an important role in modulating the 529 chemical weathering during the Quaternary glacial-interglacial cycles. Several 530 hypotheses have been proposed to explain the records for past continental weathering. 531 The cyclic expansion and retreat of continental ice sheet could rejuvenise the surfaces 532 affected by chemical weathering and so boost the weathering rate (Crocket et al., 2012; 533 Foster and Vance, 2006; Vance et al., 2009). The cyclic latitudinal migration of the inter-tropical convergence zone and associated variation in monsoon strength could 534 535 drive higher interglacial chemical flux (e.g. Hein et al., 2017; Wilson et al., 2015). 536 The exposure of continental shelf could introduce additional carbonate weathering 537 flux into the ocean during glacial periods (e.g. Stoll and Schrag, 1998), and probably 538 silicate weathering flux in tropical areas (e.g. Wan et al., 2017), although there are 539 also studies suggesting stable weathering flux during the Quaternary climate cycles 540 (e.g. von Blanckenburg et al., 2015).

541 Enhanced chemical weathering under periglacial environment is likely to require 542 significant fractured bedrock allowing deep-water penetration before the freeze-thaw 543 processes take place. Indeed, the Chayu valley is within a seismic belt (e.g. 544 Mukhopadhyaya and Dasgupta, 2015) and affected by high magnitude earthquake 545 capable of triggering landslides (e.g. Coudurier-Curveuer et al., 2020; Zhao et al. 546 2021). On the other hand, in areas sculptured by ice sheet or glacial (proglacial 547 settings) fluctuations and exposed to weathering (Foster and Vance, 2006) the 548 chemical weathering rate could be limited since the bedrock surface smoothed by ice 549 motion could lack fractures. Therefore, the most remarkable increases in chemical 550 weathering rate after ice sheet rejuvenation (Foster and Vance, 2006) will take place 551 in active mountain belts where new fractures are constantly generated due to tectonic 552 activity. Temperate areas can also become a periglacial environment during the glacial 553 expansion, e.g. central Eurasia (Hubberten et al., 2004) and mid-latitude North 554 American (Marshall et al., 2021). Among those areas, the periglacial weathering 555 mechanisms should be more significant in the active mountain belts where fractures 556 exist, and less on the stable terrains. Therefore, the alpine regions in active mountain 557 belts are likely to be much more sensitive to the Quaternary climatic change through 558 periglacial weathering.

559

560 **6.** Conclusion

In the Chayu catchment in the eastern Himalaya draining granitoid lithology, we studied the dissolved load of the seepages water from mass wasting deposits and the streams water in two regions with different climatic conditions. In the water samples studied, the cations are dominated by Ca^{2+} except for the South Chayu streams in which Na⁺ and K⁺ become important and the anions are dominated by HCO_3^- . The 566 cyclic input represents 5.7 to 8.2 % of the dissolved load. The SO4²⁻ of the water 567 samples are mainly from the oxidation of sulphide mineral in granitoid lithology, with 568 the minor contribution from sedimentary sulphide for some North Chayu streams. 569 Carbonate weathering contribute to a large part for the dissolved load despite the 570 granitoid lithology, and the carbonate contribution is more significant in the North 571 Chayu.

572 In the South Chayu with sub-tropical monsoonal environment, the seepages are 573 found to be systematically higher in TDS*, K*/Si and Ca*/Si ratios, and X_{carb} and 574 X_{SO_4} values than the nearby streams. The differences between landslide seepage and 575 stream are identical to the previous studies about landslide weathering with other 576 lithologies (e.g. greywacke or sedimentary rock enriched with carbonate), suggesting 577 the enhanced weathering by landslide process still holds in granitoid regions.

578 In the North Chayu with alpine periglacial environment, the differences between 579 the seepages and streams are negligible and the values are high. This highlights the impact of the periglacial environment. The high K*/Si, Ca*/Si and X_{SO4} values in all 580 the periglacial water samples suggest the preferential weathering of reactive mineral 581 582 phases, likely related to their fresh exposure by freeze-thaw cycles. Besides, 583 earthquake is likely an important parameter, promoting landslides and exposing 584 bedrocks in the area, since seismic activity is both frequent and strong around the 585 Eastern Himalayan Syntaxis (e.g. Mukhopadhyaya and Dasgupta, 2015) and the 586 Chayu catchment is in a landslide-prone area (e.g. Coudurier-Curveuer et al., 2020; 587 Zhao et al. 2021). Therefore, the freeze-thaw process coupled with a physical erosion 588 dominated by landsliding provide a strong rock exposure mechanism in alpine 589 periglacial setting of the North Chayu. The preferential weathering of the reactive 590 mineral phases is also more dominant since the partial availability of water, restricted 591 to thaw periods and low temperature favour the preferential weathering of labile 592 mineralogical phases and mineralogical sites and reduces the weathering rates of 593 silicate. Alpine periglacial weathering is important since the headwaters of most 594 mountain rivers originates from periglacial environment and granitoids are prevalent 595 among the active mountain belts. In addition, the alpine periglacial weathering in 596 active mountain belt could play an important role in modulating the chemical 597 weathering during the Quaternary glacial-interglacial cycles.

599 Acknowledgement

This work was supported by the Second Tibetan Plateau Scientific Expedition and Research (STEP) program (Grant No. 2019QZKK0707). We thank Yibo Yang and Chengcheng Ye for their assistance in the field, Bernhard Zimmermann and Aimeryc Schumacher for their assistance in elemental analysis and Jérôme Lavé for discussions on the morphology of landslides. Thanks to Niels Hovius and Christoff Andermann for the access to their analytical facilities.

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902 **1. Caption**

903

Fig. 1. Simplified geological map indicating the sampling locations. a) The entire
Chayu catchment. b) zoomed-in map of the North Chayu. c) zoomed-in map of the
South Chayu. Geology and river system are from Tibetan Institute of Geological
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Fig. 2. Ternary plots representing the relative abundance, expressed in Eq/L, of the major cation and anion of the studied Chayu water samples with the rivers draining the Lhasa terrane (Hren et al., 2007; Yu et al., 2021) as a comparison. Ca^{2+} is dominant and Cl^{-} is negligible in the samples of this study. The K⁺ and Na⁺ are more significant in the South Chayu than in the North Chayu water samples.

915

Fig. 3. The K*/Si, Ca*/Si ratios (corrected for cyclic input), X_{SO_4} and X_{carb} values of the water samples, grouped by location and nature. The vertical bars in the panel of K*/Si and Ca*/Si indicate the corresponding composition of the batholiths, with the red bar for the Demulha Batholith in the North Chayu and the green bar for the Chayu Batholith in the South Chayu.

921

922 Fig. 4. Ca*/Si ratios *versus* X_{SO_4} values for the North Chayu streams and seepages. 923 The black line represents the linear fit for the North Chayu streams with 95% 924 confidence interval presented by the grey shade. For the linear fit, the correlation 925 coefficient (R) is 0.931, and the P-value is 0.007.

926

927 Fig. 5. TDS* versus a) K*/Si ratios, b) Ca*/Si ratios and c) X_{SO4} values of the water samples. The red lines and the pink shades represent the linear fits and the 95% 928 929 confidence interval, respectively, for the North Chayu streams except the sample RYG 930 114 (this stream shows a significantly lower TDS* compared with other streams with similar K*/Si and Ca*/Si ratios, thus is considered to be diluted by the melt water 931 932 from an upstream glacier). The linear fit is characterised by a correlation coefficient 933 (R) and a P-value of 0.986 and 0.002 in panel b, while those values are 0.926, and 934 0.024 in panel c.

Fig. 6. Mean annual Temperature (MAT) *versus* a) K*/Si ratio and b) Ca*/Si ratio for
the studied Chayu water samples and granitoid watersheds elsewhere (White and
Blum, 1995; Oliva et al., 2003; Gurumurthy et al., 2012; Wu et al., 2013; Zhang et al.,
2016; Sun et al., 2019). The colder North Chayu has higher K*/Si and Ca*/Si ratios
than other granitoid watersheds from literature, highlighting factors other than
temperature that enhance chemical weathering in this periglacial environment.

Table	1	Inform	ation	about	tl	he	sampled r	mass	wasting	deposits.
Sample ID		Deposit No.	Long.	Lat.	Altitude ^[1]	Relief	Type of mass wasting deposit	Su	rface Area	Vegetation Cover
					m	m			km ²	
RYG 109, 110		ZL-1	97.1699	29.2948	4133	1312	Talus fan		0.779	39%
RYG 119, 120		ZL-2	97.0376	29.3370	4944	673	Rock glacier		0.232	13%
RYG 131		ZL-3	97.0947	29.3341	4306	247	Talus fan / rock glacier		0.199	62%
RYG 139-142, 117		ZL-4	97.1095	29.3296	4263	310	Talus fan		0.231	29%
RYG 143		ZL-5	97.1500	29.3273	3973	322	Talus fan		0.213	97%
RYG 155		ZL-6	97.1104	29.3227	4284	385	Rock glacier		0.091	54%
RYG 158		ZL-7	97.0571	29.3382	4390	225	Talus fan		0.113	85%
RYG 147-149		ZL-8	97.1617	28.5993	2010	1220	Bedrock landslide		0.921	51%

[1] The mean altitude of the deposit area.

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Table 2 Information about the sampled streams

Sample ID	Region	Long.	Lat.	Catchment Area	Mean Catchment Altitude	Relief	Estimated MAT
				km ²	m	m	°C
RYG 129	North Chayu	97.1653	29.3228	4.76	4836	1778	-3.6
RYG 153	North Chayu	97.1147	29.3219	0.122	4405	492	-1.0
RYG 114	North Chayu	97.1347	29.3318	189.1	5168	1992	-5.6
RYG 125	North Chayu	97.1892	29.3054	53.93	4927	2146	-4.2
RYG 126	North Chayu	97.1875	29.3142	141.5	4886	2159	-3.9
RYG 161	North Chayu	97.1819	29.1712	354.6	4655	2791	-2.5
RYG 96	South Chayu	97.2369	28.5960	8.35	2961	2261	7.8
RYG 97	South Chayu	97.1149	28.5807	24.0	3592	2947	4.0
RYG 99	South Chayu	97.1738	28.6018	3.59	2999	2183	7.6

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Table	3		Cher	mical			data		of			the		W	ater		samples
Sample ID	Туре	Altitude	pН	Temp.	Na ⁺	Mg^{2+}	K+	Ca ²⁺	H4SiO4	F -	Cl-	SO4 ²⁻	NO ₃ -	HCO3 ⁻	TDS	Xcarb ^[1]	Xcarb ^[2]
		m		°C	µmol/l	µmol/l	µmol/l	µmol/l	µmol/l	µmol/l	µmol/l	µmol/l	µmol/l	µmol/l	mg/L		
North Catchment																	
RYG 109	Seepage	3710	7.49	7.5	59.8	396.6	33.6	834.7	95.8	11.3	5.0	359.0	BQL	1823	201	0.96	0.96
RYG 110	Seepage	3856	8.15	10.7	44.3	332.6	42.9	1024.1	90.1	7.3	6.4	239.0	BQL	2309	225	0.97	0.97
RYG 119	Seepage	4661	7.84	2.9	31.3	22.3	16.7	297.7	78.0	1.5	6.3	48.0	BQL	584	62	0.93	0.93
RYG 120	Seepage	4662	7.5	1.6	38.7	25.8	14.9	310.1	74.1	1.9	6.5	47.0	2.5	620	64	0.92	0.92
RYG 131	Seepage	4216	7.3	8.4	99.1	74.7	29.4	510.4	108.2	5.0	8.7	42.0	10.0	1190	114	0.89	0.89
RYG 139	Seepage	4149	8.05	5.8	103.2	79.9	24.7	495.9	82.3	3.3	12.1	37.0	3.4	1187	110	0.89	0.89
RYG 140	Seepage	4149	7.07	6.7	90.2	80.9	26.4	461.7	84.7	1.8	12.3	30.0	11.9	1115	104	0.89	0.89
RYG 141	Seepage	4149	8.26	3.8	95.6	97.9	19.8	462.9	80.5	2.0	8.5	31.0	BQL	1164	106	0.89	0.89
RYG 117	Seepage	4143	8.24	3.8	98.7	166.2	14.1	423.0	68.4	1.9	8.1	43.0	9.8	1186	108	0.90	0.90
RYG 142	Seepage	4147	7.85	7.1	90.7	205.3	13.4	408.3	68.0	2.0	7.8	51.0	5.3	1214	110	0.91	0.91
RYG 143	Seepage	3857	8.19	5.5	208.4	198.1	27.8	627.8	90.4	35.0	14.6	129.0	10.0	1571	155	0.85	0.85
RYG 155	Seepage	4137	7.55	4.2	67.0	87.8	17.7	320.7	64.1	2.0	9.7	88.0	5.9	707	76	0.90	0.90
RYG 158	Seepage	4397	7.67	4.0	38.4	28.0	6.3	254.2	61.6	BQL	4.9	13.0	1.2	577	55	0.92	0.92
RYG 129	Stream	3785	8.22	4.8	98.7	278.1	20.5	452.5	52.7	2.1	5.5	191.0	9.1	1182	124	0.91	0.91
RYG 153	Stream	4127	7.8	5.4	35.0	47.2	16.1	246.5	74.1	0.9	4.4	30.0	BQL	573	58	0.91	0.91
RYG 114	Stream	3949	7.94	10.3	32.4	60.0	19.8	217.7	24.2	5.2	5.7	52.0	BQL	493	49	0.91	0.91
RYG 125	Stream	3581	7.38	6.5	47.7	291.8	25.4	563.9	50.6	3.8	5.3	359.0	3.4	1054	136	0.96	0.96
RYG 126	Stream	3572	7.78	5.6	42.2	443.5	17.8	683.9	50.6	3.9	4.9	472.0	3.5	1360	173	0.97	0.97
RYG 161	Stream	3311	8.19	8.9	73.4	400.0	23.9	584.3	50.9	3.7	12.1	352.0	5.6	1341	157	0.95	0.95
South Catchment																	
RYG 147	Seepage	1957	8.3	20.2	73.0	53.8	115.1	443.0	117.1	93.1	6.2	114.0	BQL	854	102	0.80	0.81
RYG 148	Seepage	2016	8.12	20.3	86.2	61.8	127.9	501.6	117.5	100.2	5.2	147.0	BQL	942	114	0.80	0.81
RYG 149	Seepage	2125	9.12	24.6	88.2	78.8	193.3	576.1	208.7	119.9	5.7	283.0	43.6	856	139	0.78	0.79
RYG 96	Stream	1886	7.86	15.2	104.6	48.5	55.7	288.4	158.1	107.6	7.0	47.0	BQL	626	77	0.73	0.74
RYG 97	Stream	1650	7.5	15.8	52.5	11.1	13.5	82.0	104.7	26.8	4.7	14.0	BQL	192	29	0.60	0.61
RYG 99	Stream	1787	7.62	19.2	114.0	35.0	49.2	126.7	220.0	80.1	6.4	30.0	BQL	340	57	0.51	0.52

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BQL: below quantification limit

[1] Calculated with the $(Mg/K)_{sil}$ equals to the Mg/K ratio of the corresponding batholith.

[2] Calculated with the (Mg/K)_{sil} equals to 20% of the Mg/K ratio of the corresponding batholith.



Fig. 2







Fig. 4



▲ S Stream● S Seepage a 1.2 N Stream 1 N Seepage RYG 114 0.4 0.2 0 b 16 12 Ca*/Si RYG 114 8 4 Δ 0 С 0.5 0.4 **°S** 0.3 **×** 0.2 RYG 114 0.2 0.1 0 120 160 200 240 **TDS*** 0 40 80

Fig. 5

