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Mobile evaporite enhances the cycle of physical-chemical erosion in badlands

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9 Abstract. Chemical weathering driven by physical erosion is one a of the manifestations of natural 10 processes that strongly affects chemical and solid matter budgets at the Earth's surface. However, the influence of extreme climatic erosion on chemical weathering dynamics is poorly understood. Badland 11 landscapes formed in highly erodible, homogeneous substrates have the potential to respond to 12 individual events on scales that are rapid enough for direct observation. Here, we assess the 13 14 geochemical and grain-size composition of suspended sediment and riverine chemistry measurements collected from two catchments during the 2017 Nesat and Haitang typhoons in southwestern Taiwan. 15 16 During the typhoons, the sodium adsorption ratio Na⁺ concentration covaried with suspended sediment concentration, which we attributed to sodium-induced deflocculation. Evaporite weathering at peak 17 18 rainfall is succeeded by peak silicate weathering at maximum discharge, which dominates the 19 weathering signal of the event. Overall, our observations suggest that initial weathering of near-surface 20 evaporite enhances the physical erosion of silicate rock during extreme rainfall events.

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22 **1. Introduction**

23 Chemical weathering induced by physical erosion controls nutrient supply to ecosystems (Milligan and Morel, 2002), reflects dynamic surface processes (e.g., Calmels et. al., 2011; Clift et. al., 2014; 24 25 Emberson et. al., 2016; Meyer et. al., 2017), and regulates the global carbon cycle and the evolution 26 of Earth's long-term climate (Berner et al., 1983; Ram et al., 1992; Gaillardet et al., 1999). In most 27 landscapes, physical erosion and chemical weathering operate on geological timescales that may be 28 difficult to observe on human timescales (e.g., Maher et al., 2014). However, in many landscapes 29 studies show that most erosion dominantly occurs during stochastic events, such as storms (e.g., Hartshorn et al., 2002; Lee et al., 2020; Wang et al., 2021). In particular, typhoons are able to transport 30 31 large volumes of water and dissolved solids within hours to days, allowing us to observe the 32 interactions between physical erosion and chemical weathering in landscapes impacted by them. 33 Nevertheless, observations of the interaction between extreme physical erosion and chemical 34 weathering dynamics are limited (Meyer et. al., 2017). Furthermore, the Lack of high-frequency 35 stream water sampling leads to a fundamental difficulty in constraining the dynamic behavior between 36 physical erosion and chemical weathering during a high discharge period (e.g., a typhoon), which could 37 have key implications for our ability to quantify topographic responses to these events.the 38 quantification of topographic responses.

39

40 Badlands are landscapes characterized by highly erodible and weathered substrates, which that are 41 largely devoid of vegetation. The high erodibility of these landscapes provides a unique opportunity 42 to investigate and quantify denudation processes that operate at short timescales (Cheng et al., 2019; 43 Yang et al, 2019, 2021a; 2021b). Badlands is typically dominated by mudstones and clays, and Soils 44 soils that contain clays saturated in sodium ions are particularly vulnerable to erosion by water. Sodium 45 ions alter the layer charge of double-layered clay minerals (i.e. smectite) and cause the clays to 46 deflocculate, which refers to the process of breaking up the clay (and ultimately the soil) into finer particles that are more easily washed away by water (e.g., Faulkner et al., 2004; Mitchell et al., 1993; 47

Rengasamy and Olsson, 1991; Rengasamy et al., 1984; Sherard et al., 1976; Kašanin-Grubin et. al.,
2018). Additionally, mineral assemblage affects the stability of soil aggregates; for example, small
amounts of smectite in kaolinitic materials cause it to be more dispersive and unstable (Levy et al.,
1993).

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53 Previous studies in the badlands of southwestern (SW)SW Taiwan have revealed that dissolving halite 54 and gypsum dissolve at depth and migrate to the hillslope surface and deposit in desiccation cracks 55 during the dry season (Higuchi et al., 2013, 2015; Nakata and Chigira, 2009). Others have observed 56 that This produces pore waters found in the near-surface mudstone have with a concentration of Na⁺ 57 concentrations of 1–3 million µmol/L at 1–2 cm depth (Nakata and Chigira, 2009). Mud cracks lead 58 to the properties of the mudstone, e.g., rock density, water permeability, and ion concentration between 59 the surface (a few centimeters to 10 cm depth) and bedrock are different (Fig. S1). For example, the 60 bedrock hardly participates in physical erosion during a rainfall event due to low permeability. We hypothesize that the dissolved dissolving halite and gypsum re-crystallizes in the near-surface and is 61 62 are deposited in the mudstone cracks through capillary action during the dry season. Subsequent 63 extreme precipitation dissolves the evaporite, and the dissolved Na⁺which enhances erosion by clay dispersity and further exposes more weatherable materials, forming a positive feedback cycle. 64 65 Assuming a mudstone substrate that is primarily comprised of silicate minerals, we expect that the 66 concentration of the evaporite ions should be consistent with the changes in the sediment concentration 67 and the concentration of silicate ions.

68

To investigate <u>this potential feedbackthe relationship</u> between evaporite dissolution and erosion, we use suspended sediment concentrations (SSC) and stream chemistry data from two catchments in the badlands of SW Taiwan (Fig. 1), <u>collected at collected with a temporal resolution of 3 hours over 3</u> <u>days</u>. We interpret our observations in <u>the badlands to reflect how the excess sodium that re-precipitates</u> <u>at surface in dry season enhances physical erosion and chemical weathering in the following-during a</u> typhoon event., and the importance of this process for exposing fresh bedrock available for weathering
 in the following dry season.

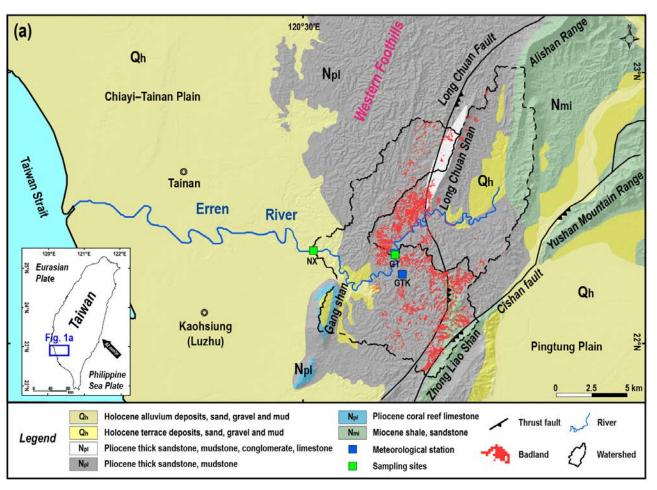
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77 2. Geological and Meteorological Setting

78 In Taiwan's badlands, the annual precipitation exceeds is about 2 m, and 90% of the rainfall is 79 concentrated in the rainy season. The rainy season lasts from May to October and reaches its peak in 80 August, with over 400 mm of precipitation within a single month. In contrast, less than 40 mm of 81 average monthly rainfall is measured from November to April. We collected river water samples from 82 two sites downstream of the studied badland areas. The first site, Nanxiong Bridge (NX), is located at the midstream of the Erren River and has a drainage area of 175 km². This area includes badlands 83 covering an area of 4.37 km², which accounts for 2.49% of the total catchment area (Fig.1). The Erren 84 85 River catchment is predominantly underlain by Plio-Pleistocene mudstones, which are several kilometers thick, and mainly feature illite (30.54%) and chlorite (28.70%) minerals (Tsai, 1984a). 86 During the dry seasons, the pore water chemistry in the near-surface mudstones is mainly composed 87 of Na⁺, Cl⁻, Ca²⁺and SO₄²⁻ (Nakata and Chigira, 2009). 88

89

90 The gauging station at Nanxiong Bridge (NX) provides hourly discharge data for calculating sediment and solute fluxes. The annual average discharge of Nanxiong Bridge station is 10.2 m³/s, and the 91 typhoon season accounts for 84% of the total discharge. The meteorological station at Gutingkeng 92 (GTK) is located 5.5 km from Nanxiong Bridge and provides hourly precipitation data. Our second 93 sampling site is Guting (GT) Bridge, with an upstream drainage area of 79 km² and a badlands area of 94 95 1.87 km², corresponding to 2.37% of the total area. Guting Bridge is located adjacent to a badlands 96 conservation area, so the riverine water chemistry reflects the weathering products derived from the 97 adjacent hillslopes. Due to a lack of stream discharge observations at Guting Bridge, we use hourly 98 precipitation data at GTK, which is less than 1 km from the sampling site, to quantify the impact of 99 the typhoon events.



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Figure 1. Location of sampling sites and geology of the study area. (a) The geological map of the study area (Source: Central Geological Survey, 2013). The green squares are sampling sites; hourly stream discharge data were obtained from the Nanxiong Bridge (NX) hydrometric station (Water Resources Agency). The blue square is the meteorological station, which provides hourly precipitation data (Central Weather Bureau, <u>https://dbar.pccu.edu.tw/</u>).

107

3. Methods and Materials

109 3.1 Water Sampling

110 We collected 42 stream samples from the two sampling sites for the typhoon period of July 2017.

111 During sample collection, two 1000 ml PE bottles were dropped 1 to 2 meters below the water surface

- 112 of the river simultaneously. Suspended sediment concentration (SSC) was subsequently calculated
- 113 from the water collected in one of the PE bottles, and riverine chemistry was determined from water
- 114 collected in the other bottle. Samples were filtered *in situ*, and the filtrate was preserved in the
- refrigerator for laboratory analysis. Additionally, 31 samples were collected from September 2014 to

December 2016 in the second half of every month at Nanxiong Bridge for non-typhoon periods, usingthe same sampling procedure.

- 118
- 119 3.2 Dissolved load and sediment chemistry analysis

For the riverine dissolved load, we measured major dissolved anions (Cl^{-} , SO_4^{2-} , NO^{2-} , NO^{3-} , F^{-}) on an 120 Ion chromatography (IC, Metrohm Basic-883 plus), and we measured major dissolved cations (Na⁺, 121 K^+ , Mg^{2+} , Sr^{2+} , Ba^{2+} , Si^{4+}) on an ICP-OES (PerkinElmer, Optima 2100DV). We measured bulk 122 sediment chemistry from two samples of suspended sediment collected from Guting Bridge at low 123 flow before the typhoon event (2.26 m^3/s) and at the peak of runoff (724.32 m^3/s). About 0.7 g of dried 124 125 sediment sample was combusted in the muffle furnace at 650°C for 2 hours and then weighed to obtain 126 the loss on ignition (LOI). Afterwards, an aliquot of ~100 mg from the residue was digested with a 127 mixture of concentrated HF and aqua regia. After digestion and drying, the sample was dissolved in 0.3 N HNO₃ for elemental determination. Major elemental concentrations of sediment samples were 128 obtained by ICP-OES (Varian 720-ES) at the GFZ German Research Centre for Geosciences. 129

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131 **3.3 Grain size of suspended load**

132 Before measuring grain size, we removed the non-clastic deposition, i.e., sea salt, organic matter, and 133 carbonate. To remove sea salt, ~1 g of dried sediment sample was added to 15 ml of distilled water, 134 placed in a shaker, and shaken at a speed of 4000 rpm for 5 minutes. The centrifuged supernatant was 135 then poured out and these steps were repeated 3 times. To remove organic matter, 10 ml of a 15% H₂O₂ solution was added to the sediment and placed in an ultrasonic oscillator for 24 hours. After 136 137 adding a second 10 ml of H₂O₂ (15%) to confirm the completion of the reaction, the mixture was 138 centrifuged and the supernatant containing the organic matter was removed. The sediment was then 139 washed by adding 30 ml of distilled water, and the supernatant was again removed after centrifugation. This washing step was repeated 3 times to remove residual H_2O_2 in the centrifuge tube. To remove the 140

141 carbonates, we added 10 ml of 10% HCl solution to the centrifuge tube and allowed the acid to react 142 with the sediments for 24 hours. An additional 10 ml of HCl was then added to confirm the 143 completeness of the reaction. The sample was then centrifuged, and the supernatant was decanted to 144 remove the carbonates. The sample was then rinsed with 30 ml of distilled water, centrifuged, and 145 decanted. This step was performed 3 times to remove any residual HCl.

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To disperse sediment agglomeration, we added 10 ml of 1% Na(PO₃)₆ solution to the sediment and let
the sample react for more than half a day. The grain size of the sediment samples was obtained by
Laser Diffraction Particle Size Analyzer LA950 at the GFZ German Research Centre for Geosciences.
By using LA950, we measured grains in the size range of between 100 nm to about 3 cm.

151

152 3.4 Calculation of the enriched ratio and sodium adsorption ratio (SAR)

In order to classify the supply of different ion sources during the typhoon event, we used the enriched ratio of concentration as a reference. The enriched ratio is the ion concentration at a certain time divided by the ion concentration at the first observation. A value greater than 1 represents a point in time when the sample is more concentrated relative to the first observation, whereas a value smaller than 1 represents a point in time when the sample is more diluted relative to the first observation. <u>T-he</u> first observation was sampled 6 hours before the typhoon which represents the background value of river water chemistry in this study.

160

Dissolved calcium and magnesium can stabilize soil aggregates and therefore <u>facilitate enhance</u> water permeability (Nadler et al., 1996). By contrast, excess sodium can disperse soil particles through deflocculation, thereby reducing water permeability (Hanson et al., 1999). The potential for material dispersion in badlands is generally determined by measuring the presence and behavior of sodium and is quantified by the sodium absorption ratio (SAR), (1):

$$166 \quad SAR = \frac{Na^+}{\sqrt{\left(\frac{Ca^{2+}+Mg^{2+}}{2}\right)}}$$

Here, the cation measurements are expressed in milliequivalents per liter (meq/L). For pore water,
When when SAR is greater than 13, the excess sodium causes soil particles to repel each other,
preventing the formation of soil aggregates (Seelig, 2000; Horneck et al., 2007). Given the influence
of soil structure, SAR value for irrigation water smaller than 3 is low, from 3 to 9 is medium and above
9 is high (Ayers and Westcot, 1985).

172

173 3.5 Calculation of TDS and chemical weathering rate

174 Riverine TDS is widely used to estimate chemical weathering rates of river catchments (e.g. Gaillardet

et al. 1999). In this study, riverine TDS (in units of μ mol/L) is expressed as:

$$176 \quad TDS = TDS_{rain} + TDS_{evaporite} + TDS_{sil} + TDS_{carb} \tag{2}$$

where the contributions from precipitation (TDS_{rain}), evaporite (TDS_{evaporite}), silicate weathering 177 178 (TDS_{sil}) and carbonate weathering (TDS_{carb}) are considered. We calculated the proportions of ion 179 contributions from rainwater, evaporite, silicate and carbonate for Ca, Mg, Na, Cl, and SO₄ with the MEANDIR inversion model (Kemeny and Torres, 2021), a MATLAB script for inverting fractional 180 181 contributions of end-members, and for constraining the chemical compositions of those end-members 182 with Monte Carlo propagation of uncertainty. To exclude the input of precipitation (TDS_{rain}) from 183 riverine TDS, we used local rainwater Cl⁻ concentrations with an average value of 68 µmol/L (Lu, 2014), and also the ratios of Cl⁻and SO₄/Cl²⁻, Na/Cl⁺, K/Cl⁺, Mg/Cl²⁺, Ca/Cl²⁺ in rainfall based on the 184 rainfall chemistry from 2007 to 2013 reported by Lu (2014) (Table $1\frac{SO_4^2}{Cl^2} = 0.35$, Na⁺/Cl² = 0.90, 185 $K^+/Cl^-=0.09$, $Mg^{2+}/Cl^-=0.18$, $Ca^{2+}/Cl^-=0.35$) (Lu, 2014). Thus, we estimated the annual deposition 186 187 of those cations using equation (3):-

$$[X]_{norain} = [X]_{river} - [X]_{rain}$$
(3)

$$TDS_{rain} = \sum [X]_{rain}$$
(4)

Here [X]_{norain} reflects the remaining concentration of ion X after the removal of atmospheric inputs;
[X]_{river} is the concentration of ion X in river water, and [X]_{rain} is the concentration of ion X from atmospheric deposition. In the second step, we corrected for evaporite inputs (TDS_{evaporite}) using the following equation:

194
$$[X]_{NSS} = [X]_{norain} - [X]_{evap} = [X]_{norain} - \left([Cl]_{norain} \times \left(\frac{X}{Cl}\right)_{evap}\right)$$
(5)

L

195
$$TDS_{evaporite} = \sum [X]_{evap}$$
 (6)

where $[X]_{NSS}$ is the concentration of ion X after the removal of ions attributed to evaporites, $[X]_{evap}$. $[X/CI]_{evap}$ is the ratio of ion X and Cl by using the end-member molar ratios of evaporite reported by Burke et al. (2018), of which K/Cl is referred to Chao et al., (2011) (Table 1)SO4²⁻/Cl⁻=0.4, Na⁺/Cl⁻ = 1, Mg²⁺/Cl⁻=0.10, Ca²⁺/Cl⁻=0.5, Burke et al., 2018; K⁺/Cl⁻=0.026, Chao et al., 2013). Then, after the correction for evaporite, the chemical weathering budget can be divided into contributions by silicate (TDS_{sil}) and carbonate weathering (TDS_{carb}), expressed as:

202
$$TDS_{sil} = [Na]_{sil} + [K]_{sil} + [Mg]_{sil} + [Ca]_{sil} + [SiO_2]_{sil}$$
 (7)

$$203 \quad TDS_{carb} = [Mg]_{carb} + [Ca]_{carb} + [HCO_3]_{carb}$$
(8)

204
$$[HCO_3]_{carb} = \frac{1}{2}([Mg]_{crab} + [Ca]_{crab})$$
 (9)

where [Na]_{sil} and [K]_{sil} are riverine [Na]_{NSS} and [K]_{NSS} concentration<u>s</u>, respectively. We used endmember values for silicate- and carbonate-dominated rocks reported by Gaillardet et al. (1999), (Table 1). We agree that the use of global endmembers leads to a larger range of estimations, but is still appropriate in discussing trends in weathering rates, which gave ratios of Ca/Na =0.35 and Mg/Na =0.24 for silicates, and Ca/Na = 50 and Mg/Na =10 for carbonates.

210 <u>Considering the hydrological response, we use flow weighted method to calculate the flux of solute</u>

		<u>.s:</u>			
Flux _{(rain, evap, s}	$_{\rm sil, carb)} = (m \times$	$\frac{\sum_{i=1}^{n} \text{TDS}_{(rain, ev}}{\sum_{i=1}^{n} c}$	ap, sil, carb)i Qi	$\langle Q_t \rangle / catchme$	ent area
where m is the co	onversion factor	for a specific u	nit (ton/km ² /	yr). Q _i is the h	ourly discharge
corresponding to	sampling time.	Qt is total disch	arge during	the year or dur	ing the typhoon
	1 1 0 2				
Table 1 Input end					
	u-members for t	he mixing mode	<u>el.</u>		
End-member	<u>SO₄/Cl</u>	<u>he mixing mode</u> <u>Na/ Cl</u>	<u>el.</u> <u>K/ Cl</u>	<u>Mg/Cl</u>	<u>Ca/Cl</u>
*				<u>Mg/Cl</u> 0.18	<u>Ca/Cl</u> <u>0.35</u>
End-member	<u>SO4/C1</u>	<u>Na/ Cl</u>	<u>K/ Cl</u>		
End-member Precipitation	<u>SO₄/Cl</u> <u>0.35</u>	<u>Na/ Cl</u> <u>0.90</u>	<u>K/Cl</u> <u>0.09</u>	0.18	0.35
End-member Precipitation	<u>SO4/Cl</u> <u>0.35</u> <u>0.6±0.6</u>	<u>Na/ Cl</u> <u>0.90</u> <u>1.0±0</u>	<u>K/Cl</u> <u>0.09</u>	0.18	0.35

219 **4. Results**

220 4.1 Geochemistry of river water and suspended sediment

(Ilyong at al. 2012) arenaged as

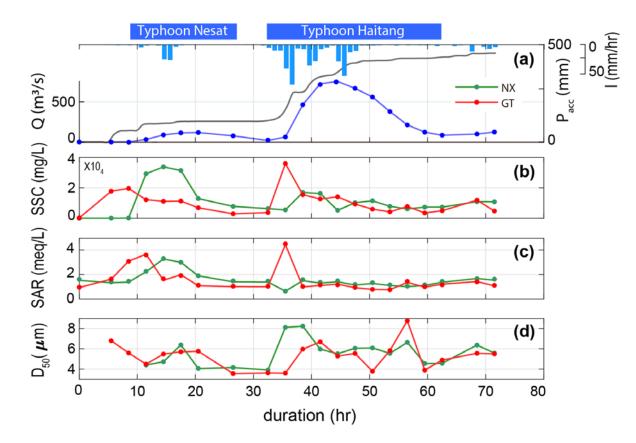
221 In 2017, the Nesat and Haitang typhoons brought 579 mm of rainfall over three days, with a maximum 222 intensity of 74 mm/hr. The discharge at Nanxiong Bridge demonstrated that the climatic co-response 223 has two pulses (Fig. 2). Since the time interval between the two typhoons was less than 6 hours, we 224 define the two typhoons as one typhoon event and distinguish between a first and second discharge 225 pulse. We quantify time relative to the onset of the typhoon (0 hr). The first pulse occurred from 8.5 to 32.5 hr, with a mean water discharge of $66.2 \text{ m}^3/\text{s}$. The second pulse that occurred from 32.5 to 62.5226 227 hr had a 5.5 times higher mean discharge of 369.2 m³/s. The maximum discharge (753.2 m³/s) was 228 observed during the second pulse at 44.5 hr (July 31th 31st, 2017, at 6:00 a.m.) (Fig. 2).

229

At Nanxiong Bridge, SSC has a statistically significant positive correlation with SAR ($\rho = 0.51$, p < 0.05). SSC has two peaks, one during each during the both pulses, but SAR only shows a peak during the first pulse. During the first pulse, SSC ranged from 10 to 33757 mg/L and SAR increased from 8.21.44 and to 17.73.14. During the second pulse, SSC increased from 5445 to 16900 mg/L and SAR

234 is steadilyremained about 7.31.44.

235 The median grain size (D_{50}) ranged from 3.9 to 8.2 µm, with an average value of 5.6 µm during the second pulse, and exhibited a positive correlation with discharge ($\rho = 0.40$). At Guting Bridge, SSC 236 237 has a statistically significant positive correlation with SAR ($\rho = 0.69$, p < 0.05) during the survey. SSC 238 ranged from 164 to 19538 mg/L before the first pulse and ranged from 2857 to 35920 mg/L during the 239 second pulse, while SAR showed a mean of $\frac{8.21.46}{20.4}$ and two peaks with a value over $\frac{20.4}{20.4}$ during both 240 pulses. D₅₀ ranged from 3.6 to 8.8 µm, with an average value of 5.3 µm during the second pulse, (Fig. 241 2). In terms of sediment chemistry at Guting Bridge, major elements of the two selected sediment 242 samples show that calcium and sodium accounted for about 10% of the mass loss between the typhoon 243 event (5.5 hr of duration) and the peak of discharge (41.5 hr of duration) (Table. S4).



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Figure 2. Timeseries SSC, SAR and median grain size of suspended sediment (D₅₀) at two sampling
sites. The blue line denotes hourly discharge (Q) at Nanxiong Bridge, and the blue bar denotes hourly
precipitation (I) at Gutingkeng station. The gray line denotes precipitation accumulation (P_{acc}), the
green line denotes the Nanxiong Bridge (NX) dataset, and the red line denotes the Guting Bridge (GT)
dataset.

251 The fractional proportions of TDS at Nanxiong Bridge during baseflow show that precipitation, 252 evaporites, silicates, and carbonates contribute 3.0±1.1% (uncertainty gives the standard error of the 253 mean), 28.7±14.6%, 26.9±6.5%, and 41.4±13.2%, respectively (Fig. 3a). During the typhoon event, 254 the proportion of TDS at Nanxiong Bridge attributed to TDS_{rain} is 6.3±2.4%. TDS_{evaporite} contributes 255 $32.4\pm13.6\%$ and increases from 27.4% to 61.1% at the incipient first pulse. TDS_{sil} contributes 256 $39.5\pm15.2\%$, which is 12.6% higher than the non-typhoon period. TDS_{carb} contributes 21.8±11.5 % 257 (Fig. 3b), which is 19.6% lower than the non-typhoon period. The fractional proportions of TDS at the 258 Guting Bridge show that 6.5±2.1% of TDS is contributed by TDS_{rain}. TDS_{evaporite} contributes 24.8±16.2% and increases from 13.6% to 61.6% at the incipient second pulse, when the SSC and SAR 259 peak simultaneously. TDS_{sil} and TDS_{carb} contribute 39.5±15.2% and 27.5±16.7%, respectively (Fig. 260 261 3c).

262

263 Enriched ratios of less than <u>1-one</u> indicate dilution, and values greater than <u>1-one</u> indicate concentration. 264 Since we set the ion concentration of rainfall to be constant during the typhoon event, the enriched 265 ratio of precipitation is constant throughout the observation period. At Nanxiong Bridge, the evaporites enriched ratio increases from 0.4 to 1.7 between the two pulses and decreases to 0.1 at the discharge 266 267 peak. The silicates enriched ratio increases from 1 to 1.5 before the first pulse and decreases to 0.1 at 268 the peak of discharge, then returns to 1 before the observation ends. The concentration attributed to 269 carbonates is always diluted. The evaporites and carbonates enriched ratio have has a statistically 270 significant negative correlation with discharge (evaporites: $\rho = -0.67$, carbonates: -0.60, p<0.05) and the silicate enriched ratio has a negative correlation with discharge ($\rho = -0.32$), indicating dilution by 271 272 typhoon rainfall (Fig. 3d). At Guting Bridge, the evaporites enriched ratio has two peaks during the two pulses with a value of 5.2 at the first peak, a value of 4.7 at the second peak. After the event, the 273 274 value returns to about 1.2. Notably, the evaporites enriched ratios during the both pulses are similar, 275 but the peak discharge of the second pulse is 5.5 times higher than that of the first pulse. The silicate

276 enriched ratio has an analogous pattern with the evaporites enriched ratio, but the enriched ratio is 277 smaller. Similar to Nanxiong Bridge, the carbonates enriched ratio is always diluted at Guting Bridge 278 (Fig. 3e). The evaporite and silicate enriched ratio shows a statistically significant positive correlation 279 $(\rho = 0.96, p < 0.05)$, and the evaporite and silicate enriched ratios have a statistically significant positive 280 correlation with SAR ($\rho = 0.86$, $\rho = 0.84$, p<0.05). We also use the concentration-discharge (cQ) 281 relationship of each ion at rising and recession limb, as well as baseflow at Nanxiong Bridge to assess 282 the state of dilution behavior (Fig. S2). Overall, our results show that all ions are in a dilution, and the 283 dilution in recession limb is stronger than that in rising limb, except for SO₄ during baseflow (θ =0.07). The concentration of Na, Cl and K during baseflow have a higher variability than the values during 284 285 the event. Additionally, Na, Cl, and SO₄ increase the concentration with increasing flow at the certain 286 period of rising limb.

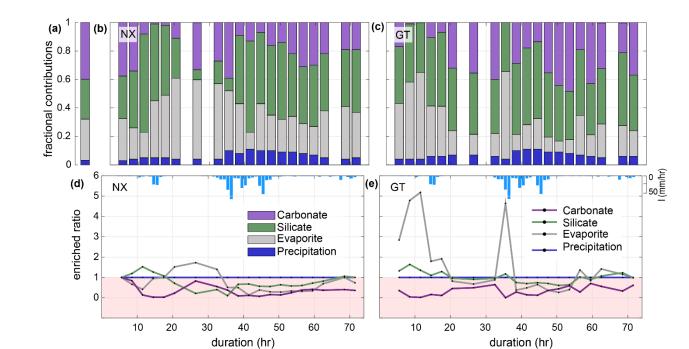




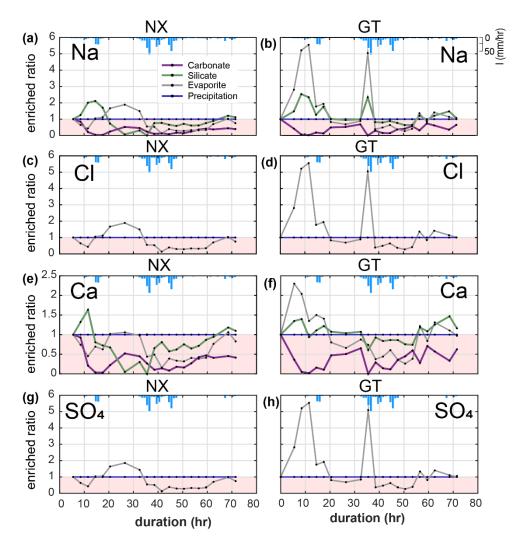
Figure 3. Timeseries illustrating TDS sources during the typhoon event at the two sampling sites. 289 Fig.3a shows the average proportion of TDS for the non-typhoon period from September 2014 to 290 291 December 2016 at Nanxiong Bridge; Fig.3b-c denotes the endmember contributions to TDS at 292 Nanxiong Bridge dataset and Guting Bridge dataset from the typhoon period; the purple bar denotes TDS_{carb} (Eq. 8); green denotes TDS_{sil} (Eq. 7); the gray bar denotes TDS_{evaporite} (Eq. 6); the blue bar 293 denotes TDS_{rain} (Eq. 4). Fig.3d-e denotes the enriched ratio of ion concentrations by TDS sources from 294 295 the Nanxiong Bridge dataset and Guting Bridge dataset during the typhoon period. The purple line 296 denotes TDS_{carb}, the green line denotes TDS_{sil}, the gray line denotes TDS_{evaporite}, the blue line denotes the TDS_{rain}, and blue bar denotes hourly precipitation (I) at GTK station. 297

4.2 Evaporite, <u>silicate and carbonate</u> dissolution over time

We calculated the enriched ratios of ions (i.e., Na^+ , Cl^- , Ca^{2+} and SO_4^{2-}) that are sourced from evaporites (i.e., halite (NaCl) and gypsum (CaSO₄)). The variability in the concentrations of each of these ions reflects the overall trends in TDS (Fig. 3d-e & Fig. 4).

302

At Nanxiong Bridge, all evaporite and carbonate ions have a statistically significant negative 303 correlation with discharge. The enriched ratios in evaporite Na^+ , Cl^- and SO_4^{2-} have the same trend (Fig. 304 4), which show an initial decrease during the first pulse, followed by an increase to 2 between the two 305 pulses, and a final decrease during the second pulse. Evaporite Ca²⁺ shows a similar trend with 306 evaporite Na⁺, Cl⁻ and SO₄²⁻, but the values are below 1. The enriched ratios of silicate Na⁺, Ca²⁺, and 307 SO_4^2 show an increase during the first pulse and a decrease to less than 1 before the rainfall peak, 308 309 followed by an increase from about 0.06 to 1.11 at the end of observation. At Guting Bridge, all evaporite ions have a statistically significant positive correlation with the corresponding silicate ions 310 \$11 $(Na^+, \rho = 0.98; Ca^+, \rho = 0.81; SO_4^{2-} = 0.98, p < 0.05)$. Evaporite Na⁺, Cl⁻, and SO₄²⁻ each have two peaks 312 that occur prior to the maximum rainfall and reflect a factor of 5 increase in the enriched ratio. Compared with Nanxiong Bridge (downstream), the enriched ratio in evaporite Ca²⁺ at Guting Bridge 313 concentrates at the onset of the first pulse and after peak discharge. Additionally, the enriched ratios of 314 carbonate at Guting Bridge are similar to Nanxiong Bridge, and are always below 1. 315

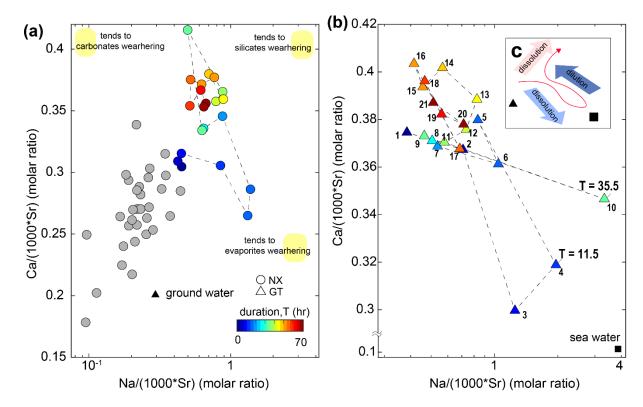


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Figure 4. Time-series patterns in enriched ratio at two sampling sites. NX denotes the Nanxiong Bridge
dataset and GT denotes Guting Bridge dataset. The pink area indicates enriched values below 1. Blue
bar denotes hourly precipitation (I) at Gutingkeng station.

321 Gaillardet et al. (1999) documented that dissolved ions ratios of Ca/Sr and Na/Sr are distinct for carbonates (low Na/Sr, high Ca/Na) versus silicates or evaporites (high Na/Sr, low Ca/Na). We use 322 323 these ratios to elucidate potential mixing between carbonates, and silicates, and evaporite 324 endmenberss (Fig. 5). At Nanxiong Bridge, non-typhoon ratios of Na/(1000*Sr) and Ca/(1000*Sr) are 325 0.23–0.68 and 0.19–0.35, respectively (Table S4). These values increase markedly during the typhoon 326 events, with enriched_ ratios of Na⁺ exceeding 5 at T = 11.5 and 35.5 hr. The high concentration of Na⁺, Cl⁻ and SO₄²⁻ (as illustrated in the enriched ratio) indicate that there is enhanced dissolution of 327 328 evaporites at the onset of the typhoon event, especially at Guting Bridge. Subsequently, the 329 concentration of Na⁺ decreased with sustained rainfall. Then, the ratios approach the
330 silicates/carbonates weathering (high Na/Sr, high Ca/Sr ratios) after the peak discharge.

331



332

Figure 5. Molar ratio mixing diagrams of Erren River waters for (a-b) Na/(1000*Sr) versus 333 334 Ca/(1000*Sr), circles denote dataset at Nanxiong Bridge, and triangles denote dataset at Guting Bridge. 335 Colorbar denotes survey duration. Gray circles denote the dataset at Nanxiong Bridge during baseflow 336 conditions from 2014 to 2016. The black triangle illustrates the groundwater endmember (Chao et al., 337 2011); the black square illustrates the seawater endmember. Numbers in the triangle represent the time 338 sequence, 1 represents the start point, and 21 represents the end point. Yellow areas indicate trends in 339 weathering types rather than the locations of endmember. (c) Illustration of dynamic weathering. The 340 red line indicates the direction of change with time. The light blue arrow denotes dissolution of 341 evaporite, the dark blue arrow denotes dilution from rainfall, and the red arrow denotes dissolution of 342 suspended sediment.

343

344 5. Discussion

5.1 Relationships between dissolved evaporite and river water chemistry

Before the survey, the monthly rainfall of the study area was 72.5 mm, which is only 18% of the

average monthly rainfall, implying that it provides a relatively dry environment for accumulating 347 evaporites on the slope surface. Under maximum rainfall intensity, Na⁺, Cl⁻ and SO₄²⁺ at Guting Bridge 348 349 show markedly increased concentrations at the onset of the typhoon, peaks in enriched ratios that \$50 exceed 5 (Fig. 4), and the greatest contribution of dissolved ions from evaporites (Fig. 3). In addition, \$51 Calculated with pore water chemistry during the dry season from the same study site, the sodium absorptions ratio (SAR) is 240.8 and exceeds the threshold value of 13. During the typhoon event, the \$52 \$53 river water SAR has a maximum value of 4.41 at Guting Bridge (3.14 at Nanxiong Bridge), suggesting 354 soil deflocculation within river is weaker than on the hillslopes. However, the sodium absorptions ratio (SAR) has a statistically significant positive correlation with TDS_{evaporite} ($\rho = 0.86$, p<0.05) at Guteng \$55 Bridge (upstream). During the typhoon event, the SAR increases from 4.8 to 24.9 and exceeds the 356 \$57 threshold value of 13 at the incipient first pulse and at the incipient second pulse. This pattern indicates 358 that excess sodium is effective at inducing material dispersion at hillslopes and thus, contributing to a \$59 higher suspended sediment load. The trend of river water SAR is able to reflect the extent of dissolved 360 Na⁺ from hillslope. (Fig. 2).

361

These observations and results suggest that rainwater in the typhoon event rapidly dissolves the evaporites on the slope surface, which produces high measured concentrations of Na⁺, Cl⁻, and SO4²⁺ during the time of peak precipitation (30-40 hr of duration). Furthermore, the dissolution of the nearsurface evaporite deposits should be most heavily influenced by runoff from the hillslopes, so we expect that excess sodium and enhanced erosion will be most significant on the hillslopes.

367

At Nanxiong Bridge, we observe a 10-hour delay in the peak enriched ratio relative to the SAR (Fig. 3d) and overall lower enriched ratios relative to Guteng Bridge (Fig. 3d-e). We suggest that dilution and the transport distance from the badlands is are responsible for this. The two catchments have a similar areal extent of badlands within the total catchment area, which is about 2.49% at Nanxiong Bridge catchment and 2.37% in Guting Bridge catchment. Badlands contribute considerable evaporite solutes (Chou, 2008), but the higher downstream drainage area will result in dilution of the solutes
without additional inputs. Additionally, Nakata and Chigira (2009) have observed that salt dissolution
induces an increase in electrical conductivity during intermittent rainfall events and decreases
gradually after rainfall events when evaporation and salt precipitate. Therefore, re-crystalization during
the transportation is to be expected.

378

379 **5.2** From evaporite dissolution to silicate weathering

380 Our observations show that silicate weathering during the typhoon event contributes 16.8 381 ton/km²/yrthe water chemistry of the typhoon event is dominated by silicate weathering at 16.8 382 ton/km²/day, corresponding to contributing 16.6% to of the annual silicate weathering flux (Table S3). 383 Additionally, we observed a change in dominant chemical weathering mechanism during the typhoon event. We rule out significant contributions from groundwater and deep seawater after peak discharge, 384 385 since ratios shift to higher Na/Sr, and Ca/Sr ratios relative to the non-typhoon ratio (Fig. 5a-b), and the Ca/Sr ratio of mud volcanoes in the study site is one order of magnitude less than river water (Chao 386 et al., 2011). Carbonate weathering is the primary contributor of Ca^{2+} for most of the world's large 387 rivers (Gaillardet et al. 1999), but the increased Na⁺ and consistently enriched ratio of carbonate Ca²⁺ 388 389 does not make this a likely main contributor to the Erren River during the typhoon. We thus suggest 390 that the primary contributor to weathering is from enhanced silicate dissolution. This interpretation is supported by the temporal evolution of the enriched ratio of silicate Ca^{2+} , which gradually increases 391 392 after the discharge peak, to approach a value of about 1 at the end of survey (Fig. 4e&f). As such, in the waning of the event, excess Ca^{2+} originates from a silicate source. Therefore, we suggest that the 393 394 ratios shift to higher Na/Sr, Ca/Sr ratios is due to enhanced silicate weathering during the typhoon. We 395 also observe that the masses of Na and Ca are reduced by 10.6% and 9.9%, respectively, in the 396 suspended sediment during the course of the typhoon event (Table S6).—

397

398 Given that the sediment transported in the channel is supplied by physical erosion, we suggest that 399 physical erosion in our study site enhances silicate chemical weathering, which is consistent with 400 previous studies (Chung, 2002; Chou, 2008). Thus Moreover, we associate the change in weathering 401 regime during the course of the typhoon with abrasive erosion of silicate sediments in the channel. 402 Mechano-chemical dissolution of weakly bound ions, e.g., F⁻ from the fresh muscovite surfaces is driven by abrasion under high energy sediment transport with reorganization of the river bed 403 404 (Andermann et al., 2022). Mudstones is are mainly composed of silicate minerals (e.g., illite and 405 chlorite minerals) (Tsai, 1984a), and <u>a few swelling clay minerals</u> (e.g., montmorillonite), which 406 provide an abundant silicate pool. We suggest that high suspended sediment concentrations, combined 407 with high energy flow during the typhoon, caused increased silicate input from the weathered silicates 408 in the suspended sediment. This trend can explain about 10% of the reduced mass and it has also been 409 observed on in typhoon-driven silicate chemical weathering from silicate minerals at surface (Meyer 410 et. al., 2017). Importantly, the global annual silicate weathering flux of rivers is 15.7 ton/km²/yr 411 (Gaillardet et al. 1999), relative to our value of 16.8 ton/km²/yr.the silicate weathering flux that we 412 calculate in this study is comparable to the global annual flux of rivers (Gaillardet et al. 1999), 413 suggesting that individual stochastic events may have global relevance.

414

415 **5.3 Typhoon-controlled cycles of physical and chemical erosion**

416 Evaporites, including halite (NaCl) and gypsum (CaSO₄), are found in few sedimentary environments, 417 and they are often excluded from the estimation of CO₂ consumption global chemical weathering 418 eycles (Gaillardet et al., 1999). Compared to silicate rocks, the relation between evaporites weathering 419 and physical erosion has rarely been discussed. Through the interactions among riverine chemistry, 420 suspended sediment properties, and previous soil water chemistry studies, we suggest a positive feedback cycle of physical-chemical erosion driven by mobile dissolved evaporite (Fig. 5). The 421 feedback cycle includes three steps. (1) precipitation and deposition of evaporite during the dry season 422 423 in near-surface mudstone desiccation cracks through capillary transport (Higuchi et al., 2013, 2015; Nakata and Chigira, 2009). In the dry season, exposed bedrock with low water content develops desiccation cracks (Allen, 1982; Goehring et al., 2010; Kindle, 1917; Seghir and Arscott, 2015; Xiaa and Hutchinson, 2000), providing space for the re-precipitation of evaporite minerals. Using evidence from core samples in mudstone bedrock at the study site, the depth of the crack of about 20 cm can be regarded as the thickness of the weathering layer. Higuchi et al. (2013) suggested that the weathering layer in the top 10 cm of mudstone can easily be eroded by intense rainfall. Erosion exposes fresh bedrock, which would dry in the following dry season and further produce weatherable material.

431

432 (2) Rainfall dissolves the evaporites, producing sodic water that increases physical erosion during 433 typhoon events. The resulting dissolved sodium causes higher hillslope erosion by deflocculation, leading to increased suspended sediment in the channels. In the study site, hillslope erosion rate is 434 435 about 9-30 cm/year (Higuchi et al., 2013; Yang et al., 2021a). At Nanxiong Bridge, the denudation rate 436 approaches about 142,857 ton/km²/yr, measured from river suspended load (Dadson et. al., 2003), and 437 the chemical weathering flux is 124-2357 ton/km²/yr (Chou, 2008; this study). The high hillslope 438 erosion rate ensures a steady supply of freshly exposed bedrock, allowing for high chemical weathering 439 rates.

440

(3) Physical erosion enhances silicate weathering and bedrock exposure on hillslopes. Clay minerals
in mudstone deposits are abraded from the abundantly available sediment and provide material for
silicate weathering in streams. Ultimately, with frequent typhoon events and high temperatures in the
study area, this dynamic cycle could repeat several times a year.

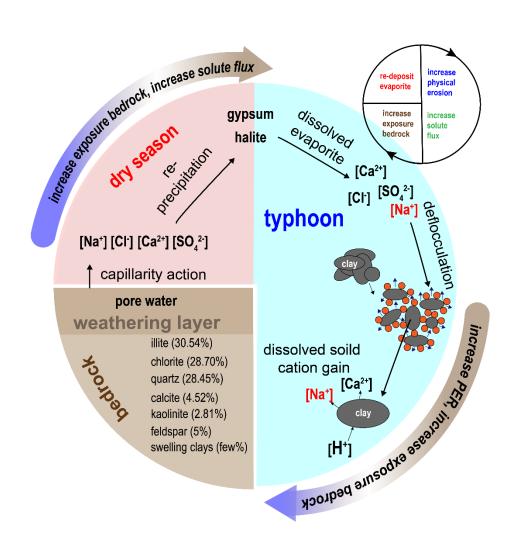


Figure. 6. Cycle of feedback between physical erosion rate (PER) and solute flux chemical weathering
rate (CWR) in badlands catchment. Red blocks represent dry season conditions. Blue region represents
typhoon conditions. Brown region represents the bedrock and indicates the type and proportion of
minerals of mudstone (Tsai, 1984b).

450

451 6. Conclusion

We presented major element compositions of stream water from two sites in the Erren River catchment at three-hour intervals during a three-day typhoon event in 2017. At the-Guteng Station (upstream), TDS_{evaporite} is covariant with TDS_{sil}, the sodium adsorption ratio, and the suspended sediment concentration, which can be assigned to dissolved evaporite (e.g., halite and gypsum). The excess sodium in the evaporite deposits causes material dispersion through deflocculation, which enhances the suspended sediment flux. Our observations show that the water chemistry of the typhoon event is **dominated**-mainly contributed by silicate weathering at 16.8 ton/km²/yrday and evaporite weathering at 10.9 ton/km²/yr, in contrast with baseflow (non-typhoon) conditions that are mainly
 contributeddominated by carbonate weathering. Moreover, during the course of the typhoon, we
 observed a shift from predominantly evaporite weathering during peak precipitation to silicate
 weathering at peak discharge.

463

464 Combining the observation of riverine chemistry, suspended sediment properties, and previous soil 465 water chemistry studies, we propose a feedback cycle between physical erosion and chemical 466 weathering in badlands topography, illustrating that precipitation of evaporites during the dry season 467 produces sodic water during typhoon events and preferentially triggers higher local erosion. The 468 enhanced hillslope erosion and abrasive effects of clay in a high discharge stream enhance bedrock exposure on hillslopes and silicate weathering, respectively. Newly exposed bedrock then produces 469 470 more weathered material. Although measurements of bedrock mineral chemistry and Sr isotope are 471 still needed for confirming sources of excess sodium and calcium (Fig. 5), we suggest that the 472 conceptual model could provide an insight into landscape change of badlands. The results from our 473 study suggest that high erosion rates in mudstone badlands of the Erren River catchment is are due to 474 both weakened lithology and to the interaction between evaporites and hillslope erosion.

475

Data availability. Relevant data supporting the findings of the study are available in the Supplementary
Information, or from the corresponding author upon request. Source data are provided with this paper.

- *Author contributions.* C.-J.Y. designed the study and conducted field surveys, data analysis, and
 modelling. P.-H. C. conducted data analysis. <u>S. X. conducted modelling. T. Y. T. provided the verified</u>
 <u>data. J.-C.L. and J.-C. Huang contributed to the scientific discussion, interpretation, and paper</u>
 preparation.C.-J.Y., E. D. E. and J.M.T. wrote the paper with input
- 483 of all authors. S. X. conducted modelling. T. Y. T. provided the verified data. J.-C.L. and J.-C. Huang
- 484 contributed to the scientific discussion, interpretation, and paper preparation.

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