



# Mobile evaporite enhances the cycle of physical–chemical erosion in badlands

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





## 22 1. Introduction

23 Chemical weathering induced by physical erosion controls nutrient supply to ecosystems (Milligan 24 and Morel, 2002), reflects dynamic surface processes (e.g., Calmels et. al., 2011; Clift et. al., 2014; Emberson et. al., 2016; Meyer et. al., 2017), and regulates the global carbon cycle and the evolution 25 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 (e.g., Maher 28 et al., 2014). However, studies show that most erosion occurs during stochastic events, such as storms 29 (e.g., Lee et al., 2020; Wang et al., 2021). In particular, typhoons are able to transport large volumes 30 of water and dissolved solids within hours to days, allowing us to observe the interactions between 31 physical erosion and chemical weathering. Nevertheless, observations of the interaction between 32 extreme physical erosion and chemical weathering dynamics are limited (Meyer et. al., 2017). Lack of high-frequency stream water sampling leads to a fundamental difficulty in constraining the dynamic 33 34 behavior between physical erosion and chemical weathering during a high discharge period (e.g., a 35 typhoon), which could have key implications for the quantification of topographic responses.

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37 Badlands are landscapes characterized by highly erodible and weathered substrates, which are largely 38 devoid of vegetation. The high erodibility of these landscapes provides a unique opportunity to investigate and quantify denudation processes that operate at short timescales (Cheng et al., 2019; 39 40 Yang et al, 2019, 2021a; 2021b). Soils that contain clays saturated in sodium ions are particularly 41 vulnerable to erosion by water. Sodium ions alter the layer charge of double-layered clay minerals (i.e. 42 smectite) and cause the clays to deflocculate, which refers to the process of breaking up the clay (and 43 ultimately the soil) into finer particles that are more easily washed away by water (e.g., Faulkner et al., 44 2004; Mitchell et al., 1993; Rengasamy and Olsson, 1991; Rengasamy et al., 1984; Sherard et al., 1976; 45 Kašanin-Grubin et. al., 2018). Additionally, mineral assemblage affects the stability of soil aggregates; 46 for example, small amounts of smectite in kaolinitic materials cause it to be more dispersive and 47 unstable (Levy et al., 1993).





#### 48

49 Previous studies in the badlands of SW Taiwan have revealed that dissolving halite and gypsum at 50 depth migrate to the slope surface and deposit in desiccation cracks during the dry season (Higuchi et 51 al., 2013, 2015; Nakata and Chigira, 2009). This produces pore water in the near-surface mudstone 52 with a concentration of Na<sup>+</sup> of 1-3 million  $\mu$ mol/L at 1-2 cm depth (Nakata and Chigira, 2009). We 53 hypothesize that the dissolving halite and gypsum re-crystallizes near-surface and is deposited in the 54 mudstone cracks through capillary action during the dry season. Subsequent extreme precipitation 55 dissolves the evaporite, which enhances erosion by clay dispersity and further exposes more 56 weatherable materials, forming a positive feedback cycle. Assuming a mudstone substrate that is 57 primarily comprised of silicate minerals, we expect that the concentration of the evaporite ions should 58 be consistent with the changes in the sediment concentration and the concentration of silicate ions.

59

To investigate the relationship 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), collected with a temporal resolution of 3 hours. We interpret our observations in badlands to reflect how the excess sodium enhances physical erosion and chemical weathering during a typhoon event, and the importance of this process for exposing fresh bedrock available for weathering in the following dry season.

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# 67 2. Geological and Meteorological Setting

In Taiwan's badlands, the annual precipitation exceeds 2 m, and 90% of the rainfall is concentrated in the rainy season. The rainy season lasts from May to October and reaches its peak in August, with over 400 mm of precipitation within a single month. In contrast, less than 40 mm of average monthly rainfall is measured from November to April. We collected river water samples from 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 km<sup>2</sup>. This area includes badlands covering an area of 4.37

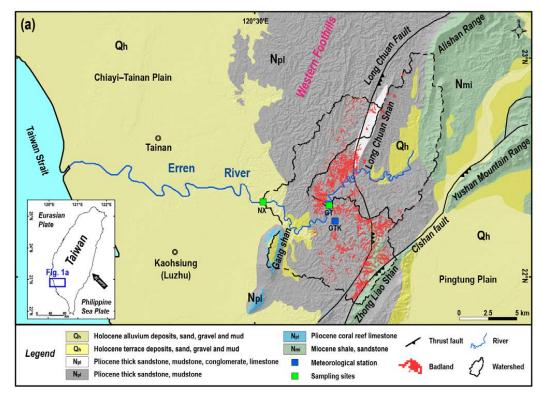




- 74 km<sup>2</sup>, which accounts for 2.49% of the total catchment area (Fig.1). The Erren River catchment is 75 predominantly underlain by Plio-Pleistocene mudstones, which are several kilometers thick, and mainly feature illite (30.54%) and chlorite (28.70%) minerals (Tsai, 1984a). During the dry seasons, 76 the pore water chemistry in the near-surface mudstones is mainly composed of Na<sup>+</sup>, Cl<sup>-</sup>, Ca<sup>2+</sup> and SO4<sup>2-</sup> 77 78 (Nakata and Chigira, 2009). 79 80 The gauging station at Nanxiong Bridge (NX) provides hourly discharge data for calculating sediment 81 and solute fluxes. The annual average discharge of Nanxiong Bridge station is 10.2 m<sup>3</sup>/s, and the 82 typhoon season accounts for 84% of the total discharge. The meteorological station at Gutingkeng 83 (GTK) is located 5.5 km from Nanxiong Bridge and provides hourly precipitation data. Our second sampling site is Guting (GT) Bridge, with an upstream drainage area of 79 km<sup>2</sup> and a badlands area of 84
- sampling site is Guting (G1) Bridge, with an upstream drainage area of 79 km<sup>2</sup> and a badlands area of 1.87 km<sup>2</sup>, corresponding to 2.37% of the total area. Guting Bridge is located adjacent to a badlands conservation area, so the riverine water chemistry reflects the weathering products derived from the adjacent hillslopes. Due to a lack of stream discharge observations at Guting Bridge, we use hourly precipitation data at GTK, which is less than 1 km from the sampling site, to quantify the impact of 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>).

97

## 98 3. Methods and Materials

99 3.1 Water Sampling

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

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

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





- 106 December 2016 in the second half of every month at Nanxiong Bridge for non-typhoon periods, using
- 107 the same sampling procedure.
- 108

## 109 3.2 Dissolved load and sediment chemistry analysis

For the riverine dissolved load, we measured major dissolved anions (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sup>2-</sup>, NO<sup>3-</sup>, F<sup>-</sup>) on an 110 111 Ion chromatography (IC, Metrohm Basic-883 plus), and we measured major dissolved cations (Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Sr<sup>2+</sup>, Ba<sup>2+</sup>, Si<sup>4+</sup>) on an ICP-OES (PerkinElmer, Optima 2100DV). We measured bulk 112 113 sediment chemistry from two samples of suspended sediment collected from Guting Bridge at low 114 flow before the typhoon event  $(2.26 \text{ m}^3/\text{s})$  and at the peak of runoff  $(724.32 \text{ m}^3/\text{s})$ . About 0.7 g of dried 115 sediment sample was combusted in the muffle furnace at 650°C for 2 hours and then weighed to obtain 116 the loss on ignition (LOI). Afterwards, an aliquot of ~100 mg from the residue was digested with a 117 mixture of concentrated HF and aqua regia. After digestion and drying, the sample was dissolved in 118 0.3 N HNO<sub>3</sub> for elemental determination. Major elemental concentrations of sediment samples were 119 obtained by ICP-OES (Varian 720-ES) at the GFZ German Research Centre for Geosciences.

120

#### 121 3.3 Grain size of suspended load

122 Before measuring grain size, we removed the non-clastic deposition, i.e., sea salt, organic matter, and 123 carbonate. To remove sea salt, ~1 g of dried sediment sample was added to 15 ml of distilled water, placed in a shaker, and shaken at a speed of 4000 rpm for 5 minutes. The centrifuged supernatant was 124 125 then poured out and these steps were repeated 3 times. To remove organic matter, 10 ml of a 15% 126 H<sub>2</sub>O<sub>2</sub> solution was added to the sediment and placed in an ultrasonic oscillator for 24 hours. After 127 adding a second 10 ml of  $H_2O_2$  (15%) to confirm the completion of the reaction, the mixture was 128 centrifuged and the supernatant containing the organic matter was removed. The sediment was then 129 washed by adding 30 ml of distilled water, and the supernatant was again removed after centrifugation. 130 This washing step was repeated 3 times to remove residual  $H_2O_2$  in the centrifuge tube. To remove the





131	carbonates, we added 10 ml of 10% HCl solution to the centrifuge tube and allowed the acid to react
132	with the sediments for 24 hours. An additional 10 ml of HCl was then added to confirm the
133	completeness of the reaction. The sample was then centrifuged, and the supernatant was decanted to
134	remove the carbonates. The sample was then rinsed with 30 ml of distilled water, centrifuged, and
135	decanted. This step was performed 3 times to remove any residual HCl.
136	
137	To disperse sediment agglomeration, we added 10 ml of 1% Na(PO <sub>3</sub> ) <sub>6</sub> solution to the sediment and let
138	the sample react for more than half a day. The grain size of the sediment samples was obtained by
139	Laser Diffraction Particle Size Analyzer LA950 at the GFZ German Research Centre for Geosciences.
140	By using LA950, we measured grains in the size range of between 100 nm to about 3 cm.
141	
142	3.4 Calculation of the enriched ratio and sodium adsorption ratio (SAR)
143	In order to classify the supply of different ion sources during the typhoon event, we used the enriched
144	ratio of concentration as a reference. The enriched ratio is the ion concentration at a certain time
145	divided by the ion concentration at the first observation. A value greater than 1 represents a point in
146	time when the sample is more concentrated relative to the first observation, whereas a value smaller
147	than 1 represents a point in time when the sample is more diluted relative to the first observation.
148	
149	Dissolved calcium and magnesium can stabilize soil aggregates and therefore facilitate water
150	permeability (Nadler et al., 1996). By contrast, excess sodium can disperse soil particles through
151	deflocculation, thereby reducing water permeability (Hanson et al., 1999). The potential for material
152	dispersion in badlands is generally determined by measuring the presence and behavior of sodium and
153	is quantified by the sodium absorption ratio (SAR), (1):

154 
$$SAR = \frac{Na^+}{\sqrt{\left(\frac{Ca^{2+}+Mg^{2+}}{2}\right)}}$$
 (1)





(3)

- 155 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).

157

# 158 **3.5** Calculation of TDS and chemical weathering rate

- 159 Riverine TDS is widely used to estimate chemical weathering rates of river catchments (e.g. Gaillardet
- 160 et al. 1999). In this study, riverine TDS (in units of  $\mu$ mol/L) is expressed as:

$$161 \quad TDS = TDS_{rain} + TDS_{evaporite} + TDS_{sil} + TDS_{carb} \tag{2}$$

where the contributions from precipitation (TDS<sub>rain</sub>), evaporite (TDS<sub>evaporite</sub>), silicate weathering 162 163  $(TDS_{sil})$  and carbonate weathering  $(TDS_{carb})$  are considered. We calculated the proportions of ion 164 contributions with the MEANDIR inversion model (Kemeny and Torres, 2021), a MATLAB script for inverting fractional contributions of end-members, and for constraining the chemical compositions of 165 those end-members. To exclude the input of precipitation (TDS<sub>rain</sub>) from riverine TDS, we used local 166 rainwater Cl<sup>-</sup> concentrations with an average value of 68  $\mu$ mol/L (Lu, 2014), and also the ratios of Cl<sup>-</sup> 167 and SO42-, Na+, K+, Mg2+, Ca2+ in rainfall based on the rainfall chemistry from 2007 to 2013 (SO42-/ 168 169  $Cl^{-} = 0.35$ ,  $Na^{+}/Cl^{-} = 0.90$ ,  $K^{+}/Cl^{-} = 0.09$ ,  $Mg^{2+}/Cl^{-} = 0.18$ ,  $Ca^{2+}/Cl^{-} = 0.35$ ) (Lu, 2014). Thus, we 170 estimated the annual deposition of those cations using equation (3):

171 
$$[X]_{norain} = [X]_{river} - [X]_{rain}$$

172 
$$TDS_{rain} = \sum [X]_{rain}$$
 (4)

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

177 
$$[X]_{NSS} = [X]_{norain} - [X]_{evap} = [X]_{norain} - \left( [Cl]_{norain} \times (\frac{X}{Cl})_{evap} \right)$$
(5)

178 
$$TDS_{evaporite} = \sum [X]_{evap}$$
 (6)





- 179 where  $[X]_{NSS}$  is the concentration of ion X after the removal of ions attributed to evaporites,  $[X]_{evap}$ .
- 180  $[X/Cl]_{evap}$  is the ratio of ion X and Cl by using the end-member molar ratios of evaporite (SO<sub>4</sub><sup>2-</sup>/Cl<sup>-</sup> =
- 181  $0.4, \text{Na}^+/\text{Cl}^- = 1, \text{Mg}^{2+}/\text{Cl}^- = 0.10, \text{Ca}^{2+}/\text{Cl}^- = 0.5, \text{Burke et al., 2018; K}^+/\text{Cl}^- = 0.026, \text{Chao et al., 2013}.$
- 182 Then, after the correction for evaporite, the chemical weathering budget can be divided into
- 183 contributions by silicate (TDS<sub>sil</sub>) and carbonate weathering (TDS<sub>carb</sub>), expressed as:

184 
$$TDS_{sil} = [Na]_{sil} + [K]_{sil} + [Mg]_{sil} + [Ca]_{sil} + [SiO_2]_{sil}$$
 (7)

$$185 \quad TDS_{carb} = [Mg]_{carb} + [Ca]_{carb} + [HCO_3]_{carb}$$
(8)

186 
$$[HCO_3]_{carb} = \frac{1}{2}([Mg]_{crab} + [Ca]_{crab})$$
 (9)

where [Na]<sub>sil</sub> and [K]<sub>sil</sub> are riverine [Na]<sub>NSS</sub> and [K]<sub>NSS</sub> concentration, respectively. We used
endmember values for silicate- and carbonate-dominated rocks reported by Gaillardet et al. (1999),
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.

191

192 **4. Results** 

### 193 4.1 Geochemistry of river water and suspended sediment

194 In 2017, the Nesat and Haitang typhoons brought 579 mm of rainfall over three days, with a maximum 195 intensity of 74 mm/hr. The discharge at Nanxiong Bridge demonstrated that the climatic co-response 196 has two pulses (Fig. 2). Since the time interval between the two typhoons was less than 6 hours, we 197 define the two typhoons as one typhoon event and distinguish between a first and second discharge 198 pulse. We quantify time relative to the onset of the typhoon (0 hr). The first pulse occurred from 8.5 to 199 32.5 hr, with a mean water discharge of  $66.2 \text{ m}^3$ /s. The second pulse that occurred from 32.5 to 62.5200 hr had a 5.5 times higher mean discharge of  $369.2 \text{ m}^3/\text{s}$ . The maximum discharge ( $753.2 \text{ m}^3/\text{s}$ ) was 201 observed during the second pulse at 44.5 hr (July 31th, 2017, at 6:00 a.m.) (Fig. 2).



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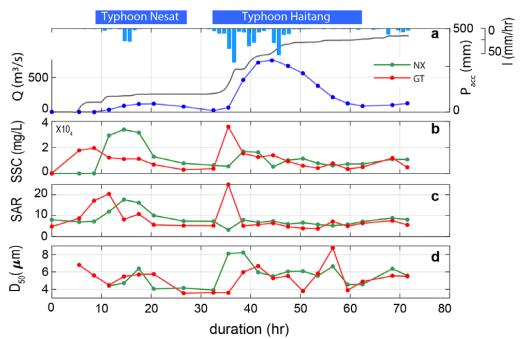
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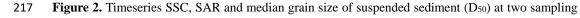


204 0.05). SSC has two peaks during the both pulses, but SAR only shows a peak during the first pulse. During the first pulse, SSC ranged from 10 to 33757 mg/L and SAR increased from 8.2 and to 17.7. 205 206 During the second pulse, SSC increased from 5445 to 16900 mg/L and SAR is steadily about 7.3. 207  $D_{50}$  ranged from 3.9 to 8.2 µm, with an average value of 5.6 µm during the second pulse, and exhibited 208 a positive correlation with discharge ( $\rho = 0.40$ ). At Guting Bridge, SSC has a statistically significant 209 positive correlation with SAR ( $\rho = 0.69$ , p < 0.05) during the survey. SSC ranged from 164 to 19538 210 mg/L before the first pulse and ranged from 2857 to 35920 mg/L during the second pulse, while SAR 211 showed a mean of 8.2 and two peaks with a value over 20 during both pulses. D<sub>50</sub> ranged from 3.6 to

At Nanxiong Bridge, SSC has a statistically significant positive correlation with SAR ( $\rho = 0.51$ , p <

8.8 µm, with an average value of 5.3 µm during the second pulse, (Fig. 2). In terms of sediment
chemistry at Guting Bridge, major elements of the two selected sediment samples show that calcium
and sodium accounted for about 10% of the mass loss between the typhoon event (5.5 hr of duration)
and peak of discharge (41.5 hr of duration) (Table. S4).





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<sub>acc</sub>), the
green line denotes the Nanxiong Bridge (NX) dataset, and the red line denotes the Guting Bridge (GT)
dataset.

222

The fractional proportions of TDS at Nanxiong Bridge during baseflow show that precipitation, 223 224 evaporites, silicates, and carbonates contribute  $3.0\pm1.1\%$ ,  $28.7\pm14.6\%$ ,  $26.9\pm6.5\%$ , and  $41.4\pm13.2\%$ , 225 respectively (Fig. 3a). During the typhoon event, the proportion of TDS at Nanxiong Bridge attributed to TDS<sub>rain</sub> is  $6.3\pm2.4\%$ . TDS<sub>evaporite</sub> contributes  $32.4\pm13.6\%$  and increases from 27.4% to 61.1% at the 226 227 incipient first pulse. TDS<sub>sil</sub> contributes  $39.5\pm15.2\%$ , which is 12.6% higher than the non-typhoon 228 period. TDS<sub>carb</sub> contributes 21.8±11.5 % (Fig. 3b), which is 19.6% lower than the non-typhoon period. 229 The fractional proportions of TDS at the Guting Bridge show that  $6.5\pm2.1\%$  of TDS is contributed by 230 TDS<sub>rain</sub>. TDS<sub>evaporite</sub> contributes 24.8±16.2% and increases from 13.6% to 61.6% at the incipient second 231 pulse, when the SSC and SAR peak simultaneously. TDSsil and TDScarb contribute 39.5±15.2% and 232 27.5±16.7%, respectively (Fig. 3c).

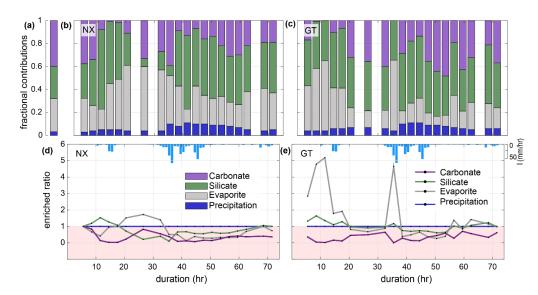
233

234 Enriched ratios less than 1 indicate dilution, and values greater than 1 indicate concentration. Since we 235 set the ion concentration of rainfall to be constant during the typhoon event, the enriched ratio of 236 precipitation is constant throughout the observation period. At Nanxiong Bridge, the evaporites 237 enriched ratio increases from 0.4 to 1.7 between the two pulses and decreases to 0.1 at the discharge 238 peak. The silicates enriched ratio increases from 1 to 1.5 before the first pulse and decreases to 0.1 at the peak of discharge, then returns to 1 before the observation ends. The concentration attributed to 239 240 carbonates is always diluted. The evaporites and carbonates enriched ratio have a statistically 241 significant negative correlation with discharge (evaporites:  $\rho = -0.67$ , carbonates: -0.60, p<0.05) and 242 the silicate enriched ratio has a negative correlation with discharge ( $\rho = -0.32$ ), indicating dilution by 243 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 244 245 value returns to about 1.2. Notably, the evaporites enriched ratios during the both pulses are similar,





but the peak discharge of the second pulse is 5.5 times higher than that of the first pulse. The silicate enriched ratio has an analogous pattern with the evaporites enriched ratio, but the enriched ratio is smaller. Similar to Nanxiong Bridge, the carbonates enriched ratio is always diluted at Guting Bridge (Fig. 3e). The evaporite and silicate enriched ratio shows a statistically significant positive correlation  $(\rho = 0.96, p<0.05)$ , and the evaporite and silicate enriched ratios have a statistically significant positive correlation with SAR ( $\rho = 0.86, \rho = 0.84, p<0.05$ ).



253 Figure 3. Timeseries illustrating TDS sources during the typhoon event at the two sampling sites. 254 Fig.3a shows the average proportion of TDS for the non-typhoon period from September 2014 to December 2016 at Nanxiong Bridge; Fig.3b-c denotes the endmember contributions to TDS at 255 256 Nanxiong Bridge dataset and Guting Bridge dataset from the typhoon period; the purple bar denotes TDS<sub>carb</sub> (Eq. 8); green denotes TDS<sub>sil</sub> (Eq. 7); the gray bar denotes TDS<sub>evaporite</sub> (Eq. 6); the blue bar 257 258 denotes TDSrain (Eq. 4). Fig.3d-e denotes the enriched ratio of ion concentrations by TDS sources from 259 the Nanxiong Bridge dataset and Guting Bridge dataset during the typhoon period. The purple line denotes TDS<sub>carb</sub>, the green line denotes TDS<sub>sil</sub>, the gray line denotes TDS<sub>evaporite</sub>, the blue line denotes 260 261 the TDS<sub>rain</sub>, and blue bar denotes hourly precipitation (I) at GTK station.



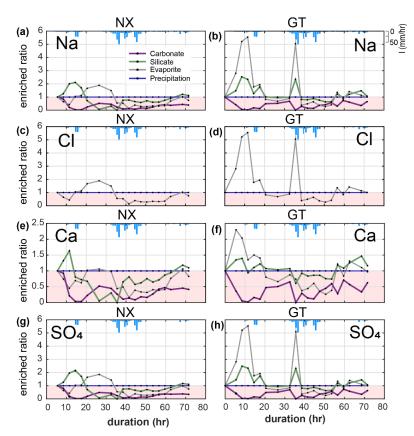


#### 262 **4.2 Evaporite dissolution over time**

We calculated the enriched ratios of ions (i.e., Na<sup>+</sup>, Cl<sup>-</sup>, Ca<sup>2+</sup> and SO4<sup>2-</sup>) that are sourced from evaporites 263 264 (i.e., halite (NaCl) and gypsum (CaSO<sub>4</sub>)). The variability in the concentrations of each of these ions 265 reflects the overall trends in TDS (Fig. 3d-e & Fig. 4). 266 At Nanxiong Bridge, all evaporite and carbonate ions have a statistically significant negative 267 correlation with discharge. The enriched ratios in evaporite  $Na^+$ ,  $Cl^-$  and  $SO4^{2-}$  have the same trend (Fig. 268 4), which show an initial decrease during the first pulse, followed by an increase to 2 between the two 269 pulses, and a final decrease during the second pulse. Evaporite  $Ca^{2+}$  shows a similar trend with 270 evaporite Na<sup>+</sup>, Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, but the values are below 1. The enriched ratios of silicate Na<sup>+</sup>, Ca<sup>2+</sup>, and 271  $SO_4^{2-}$  show an increase during the first pulse and a decrease to less than 1 before the rainfall peak, 272 273 followed by an increase from about 0.06 to 1.11 at the end of observation. At Guting Bridge, all 274 evaporite ions have a statistically significant positive correlation with the corresponding silicate ions (Na<sup>+</sup>=0.98; Ca<sup>+</sup>=0.81; SO4<sup>2-</sup>=0.98, p<0.05). Evaporite Na<sup>+</sup>, Cl<sup>-</sup>, and SO4<sup>2-</sup> each have two peaks that 275 276 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 277 concentrates at the onset of the first pulse and after peak discharge. Additionally, the enriched ratios of 278 279 carbonate at Guting Bridge are similar to Nanxiong Bridge, and are always below 1.







280

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.

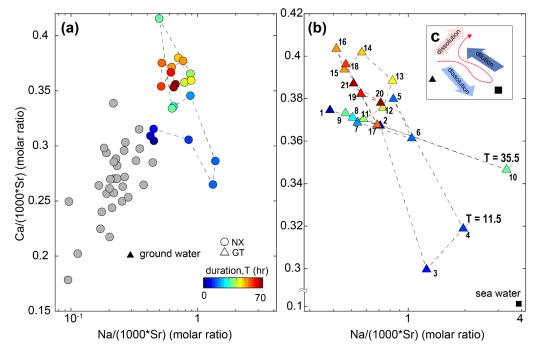
285 Gaillardet et al. (1999) documented that dissolved ions ratios of Ca/Sr and Na/Sr are distinct for 286 carbonates (low Na/Sr, high Ca/Na) versus silicates or evaporites (high Na/Sr, low Ca/Na). We use 287 these ratios to elucidate potential mixing between carbonates and silicates/evaporites (Fig. 5). At Nanxiong Bridge, non-typhoon ratios of Na/(1000\*Sr) and Ca/(1000\*Sr) are 0.23-0.68 and 0.19-0.35, 288 respectively (Table S4). These values increase markedly during the typhoon events, with enriched 289 ratios of Na+ exceeding 5 at T = 11.5 and 35.5 hr. The high concentration of Na<sup>+</sup>, Cl<sup>-</sup> and SO4<sup>2-</sup> (as 290 291 illustrated in the enriched ratio) indicate that there is enhanced dissolution of evaporites at the onset of 292 the typhoon event, especially at Guting Bridge. Subsequently, the concentration of Na<sup>+</sup> decreased with





- 293 sustained rainfall. Then, the ratios approach the silicates/carbonates weathering (high Na/Sr, high
- 294 Ca/Sr ratios ) after the peak discharge.

295



296

297 Figure 5. Molar ratio mixing diagrams of Erren River waters for (a-b) Na/(1000\*Sr) versus 298 Ca/(1000\*Sr), circles denote dataset at Nanxiong Bridge, and triangles denote dataset at Guting Bridge. 299 Colorbar denotes survey duration. Gray circles denote the dataset at Nanxiong Bridge during baseflow 300 conditions from 2014 to 2016. The black triangle illustrates the groundwater endmember (Chao et al., 301 2011); the black square illustrates the seawater endmember. Numbers in the triangle represent the time 302 sequence, 1 represents the start point, and 21 represents the end point. (c) Illustration of dynamic weathering. The red line indicates the direction of change with time. The light blue arrow denotes 303 304 dissolution of evaporite, the dark blue arrow denotes dilution from rainfall, and the red arrow denotes 305 dissolution of suspended sediment.

306

## 307 5. Discussion

#### 308 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





evaporites on the slope surface. Under maximum rainfall intensity, Na<sup>+</sup>, Cl<sup>-</sup> and SO<sub>4</sub><sup>2+</sup> at Guting Bridge 311 312 show markedly increased concentrations at the onset of the typhoon, peaks in enriched ratios that 313 exceed 5 (Fig. 4), and the greatest contribution of dissolved ions from evaporites (Fig. 3). In addition, 314 the sodium absorptions ratio (SAR) has a statistically significant positive correlation with TDS<sub>evaporite</sub> 315  $(\rho = 0.86, p < 0.05)$  at Guteng Bridge (upstream). During the typhoon event, the SAR increases from 316 4.8 to 24.9 and exceeds the threshold value of 13 at the incipient first pulse and at the incipient second 317 pulse. This pattern indicates that excess sodium is effective at inducing material dispersion and thus, 318 contributing to a higher suspended sediment load (Fig. 2).

319

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  $SO4^{2+}$ 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.

325

326 At Nanxiong Bridge, we observe a 10-hour delay in the peak enriched ratio relative to the SAR (Fig. 327 3d) and overall lower enriched ratios relative to Guteng Bridge (Fig. 3d-e). We suggest that dilution 328 and the transport distance from the badlands is responsible for this. The two catchments have a similar 329 areal extent of badlands within the total catchment area, which is about 2.49% at Nanxiong Bridge 330 catchment and 2.37 % in Guting Bridge catchment. Badlands contribute considerable evaporite solutes 331 (Chou, 2008), but the higher downstream drainage area will result in dilution of the solutes without 332 additional inputs. Additionally, Nakata and Chigira (2009) have observed that salt dissolution induces 333 an increase in electrical conductivity during intermittent rainfall events and decreases gradually after 334 rainfall events when evaporation and salt precipitate. Therefore, re-crystalization during transportation is to be expected. 335





#### 336

# 337 5.2 From evaporite dissolution to silicate weathering

Our observations show that the water chemistry of the typhoon event is dominated by silicate 338 339 weathering at 16.8 ton/km<sup>2</sup>/day, contributing 16.6% to the annual silicate weathering flux (Table S3). 340 Additionally, we observed a change in dominant chemical weathering mechanism during the typhoon 341 event. We rule out significant contributions from groundwater and deep seawater after peak discharge, 342 since ratios shift to higher Na/Sr, and Ca/Sr ratios relative to the non-typhoon ratio (Fig. 5a-b), and 343 the Ca/Sr ratio of mud volcanoes in the study site is one order of magnitude less than river water (Chao et al., 2011). Carbonate weathering is the primary contributor of  $Ca^{2+}$  for most of the world's large 344 rivers (Gaillardet et al. 1999), but the increased Na<sup>+</sup> and consistently enriched ratio of carbonate Ca<sup>2+</sup> 345 346 does not make this a likely contributor to the Erren River. We thus suggest that the primary contributor to weathering is from enhanced silicate dissolution. This interpretation is supported by the temporal 347 348 evolution of the enriched ratio of silicate  $Ca^{2+}$ , which gradually increases after the discharge peak, to 349 approach a value of about 1 at the end of survey (Fig. 4e&f). As such, in the waning of the event, 350 excess  $Ca^{2+}$  originates from a silicate source. Therefore, we suggest that the ratios shift to higher Na/Sr, 351 Ca/Sr ratios is due to silicate weathering. We also observe that the masses of Na and Ca are reduced by 10.6% and 9.9%, respectively, in the suspended sediment during the course of the typhoon event 352 353 (Table S6).

354

Given that the sediment transported in the channel is supplied by physical erosion, we suggest that physical erosion in our study site enhances silicate chemical weathering, which is consistent with previous studies (Chung, 2002; Chou, 2008). Thus, we associate the change in weathering regime during the course of the typhoon with abrasive erosion of silicate sediments in the channel. Mudstone is mainly composed of silicate minerals (e.g., illite and chlorite minerals) (Tsai, 1984a), and few swelling clay minerals (e.g., montmorillonite), which provide an abundant silicate pool. We suggest





- that high suspended sediment concentrations combined with high energy flow during the typhoon, caused increased silicate input from the weathered silicates in the suspended sediment. This trend can explain about 10% of the reduced mass and it has also been observed on typhoon-driven silicate chemical weathering from silicate minerals at surface (Meyer et. al., 2017). Importantly, the silicate weathering flux that we calculate in this study is comparable to the global annual flux of rivers (Gaillardet et al. 1999), suggesting that individual stochastic events may have global relevance.
- 367

### 368 5.3 Typhoon-controlled cycles of physical and chemical erosion

369 Evaporites, including halite (NaCl) and gypsum (CaSO<sub>4</sub>), are found in few sedimentary environments, 370 and they are often excluded from global chemical weathering cycles (Gaillardet et al., 1999). 371 Compared to silicate rocks, the relation between evaporites weathering and physical erosion has rarely 372 been discussed. Through the interactions among riverine chemistry, suspended sediment properties, 373 and previous soil water chemistry studies, we suggest a positive feedback cycle of physical-chemical 374 erosion driven by mobile dissolved evaporite (Fig. 5). The feedback cycle includes three steps. (1) 375 precipitation and deposition of evaporite during the dry season in near-surface mudstone desiccation 376 cracks through capillary transport (Higuchi et al., 2013, 2015; Nakata and Chigira, 2009). In the dry 377 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 378 379 the re-precipitation of evaporite minerals. Using evidence from core samples in mudstone bedrock at 380 the study site, the depth of the crack of about 20 cm can be regarded as the thickness of the weathering 381 layer. Higuchi et al. (2013) suggested that the weathering layer in the top 10 cm of mudstone can easily 382 be eroded by intense rainfall. Erosion exposes fresh bedrock, which would dry in the following dry 383 season and further produce weatherable material.

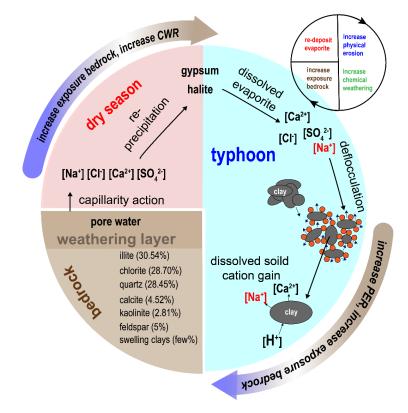
384

(2) Rainfall dissolves the evaporites, producing sodic water that increases physical erosion duringtyphoon 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
  about 9-30 cm/year (Higuchi et al., 2013; Yang et al., 2021a). At Nanxiong Bridge, the denudation rate
  approaches about 142,857 ton/km<sup>2</sup>/yr, measured from river suspended load (Dadson et. al., 2003), and
  the chemical weathering flux is 124-235 ton/km<sup>2</sup>/yr (Chou, 2008; this study). The high hillslope
  erosion rate ensures a steady supply of freshly exposed bedrock, allowing for high chemical weathering
  rates.
- 393
- (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
- 397 study area, this dynamic cycle could repeat several times a year.



**Figure. 6.** Cycle of feedback between physical erosion rate (PER) and 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).

403

# 404 6. Conclusion

405 We presented major element compositions of stream water from two sites in the Erren River catchment 406 at three-hour intervals during a three-day typhoon event in 2017. At the Guteng Station (upstream), 407 TDS<sub>evaporite</sub> is covariant with TDS<sub>sil</sub>, the sodium adsorption ratio, and the suspended sediment 408 concentration, which can be assigned to dissolved evaporite (e.g., halite and gypsum). The excess 409 sodium in the evaporite deposits causes material dispersion through deflocculation, which enhances 410 the suspended sediment flux. Our observations show that the water chemistry of the typhoon event is 411 dominated by silicate weathering at 16.8 ton/km<sup>2</sup>/day, in contrast with baseflow (non-typhoon) 412 conditions that are dominated by carbonate weathering. Moreover, during the course of the typhoon, 413 we observed a shift from predominantly evaporite weathering during peak precipitation to silicate 414 weathering at peak discharge.

415

416 Combining the observation of riverine chemistry, suspended sediment properties, and previous soil 417 water chemistry studies, we propose a feedback cycle between physical erosion and chemical 418 weathering in badlands topography, illustrating that precipitation of evaporites during the dry season produces sodic water during typhoon events and preferentially triggers higher local erosion. The 419 420 enhanced hillslope erosion and abrasive effects of clay in a high discharge stream enhance bedrock 421 exposure on hillslopes and silicate weathering, respectively. Newly exposed bedrock then produces 422 more weathered material. Although measurements of bedrock mineral chemistry and Sr isotope are 423 still needed for confirming sources of excess sodium and calcium (Fig. 5), we suggest that the 424 conceptual model could provide an insight into landscape change of badlands. The results from our 425 study suggest that high erosion rates in mudstone badlands of the Erren River catchment is due to both

426 weakened lithology and to the interaction between evaporites and hillslope erosion.





- 427 Data availability. Relevant data supporting the findings of the study are available in the Supplementary
- 428 Information, or from the corresponding author upon request. Source data are provided with this paper.
- 429
- 430 Author contributions. C.-J.Y. designed the study and conducted field surveys, data analysis, and
- 431 modelling. P.-H. C. conducted data analysis. C.-J.Y., E. D. E. and J.M.T. wrote the paper with input
- 432 of all authors. S. X. conducted modelling. T. Y. T. provided the verified data. J.-C.L. and J.-C. Huang
- 433 contributed to the scientific discussion, interpretation, and paper preparation.
- 434
- 435 *Competing interests.* The authors declare that they have no competing interests.
- 436
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