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# Chemical erosion rates in the upper Blue Nile Basin and related atmospheric CO<sub>2</sub> consumption

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- 20 Abstract
- 21

Silicate weathering of basaltic rocks constitutes a non-negligible sink of atmospheric CO<sub>2</sub> but 22 the role it plays in the regulation of past and future global climate is still matter of debate. In 23 this study, silicate weathering rates for various sub-basins of the Ethiopian Traps, emplaced 24 30 million years ago, and the corresponding atmospheric  $CO_2$  consumption rates are 25 evaluated. For this, major and trace elements were measured in the dissolved phases and in 26 the sedimentary particles carried and deposited by the main rivers flowing through this steep 27 region. Lithium isotopes and major elements were also measured in the extracted clay 28 fractions in order to infer complementary information on weathering processes in this region. 29 Clay  $\delta^7$ Li values correlate positively with Mg/Ti ratios, and are best explained by varying 30 ratios of leaching versus clay formation rate. 31

32 Although located in a region annually submitted to monsoon, average silicate weathering rate (16.1 tons/km<sup>2</sup>/year) and CO<sub>2</sub> consumption rate ( $0.65 \times 10^{12}$  mol/year) are estimated to be low 33 34 when compared to other basaltic regions such as the Deccan Traps, and volcanically active islands of the tropical zone. This is surprising since the concentrations of Total Dissolved 35 36 Solids of the Ethiopian rivers are among the highest ones. With a 2D rainfall model that takes into account the detailed topography of the region, annual occurrence of the Monsoon, and 37 monitoring station data, we show that runoff intensity is a key parameter that explains this 38 difference. We determine that, at present, the weathering of the Ethiopian Traps plays a 39 40 negligible role in the carbon cycle. However, simple calculations, which integrate recent knowledge on African climate variations and on weathering controls, illustrate that during the 41 African Humid Period (14-8 kyr), a significant increase of Monsoon precipitation may have 42 resulted in much higher weathering rates and related  $CO_2$  consumption (0.91-1.5x10<sup>12</sup>) 43 mol/year). This study therefore evidences the potential importance of this region in the past, 44 45 and the need to quantify more precisely the variations of the monsoon intensity and its impact on tropical watersheds for reconstructing past CO<sub>2</sub> levels. 46

47 48

#### 49 Key words

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<sup>Ethiopian Traps, silicate weathering rate, lithium isotopes, CO<sub>2</sub> consumption, basalt, African
monsoon</sup> 

#### 54 **1. Introduction**

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Chemical and physical erosion, which describe respectively mineral dissolution and soil 56 formation, rock cracking and particle transport are key processes in landscape evolution. 57 Various parameters can control weathering (chemical erosion) at a watershed scale, among 58 59 them lithology, tectonics, relief, soil thickness, age, vegetation cover and climate parameters (Bayon et al., 2016; Bouchez et al., 2013; Brady et al., 1999; Clergue et al., 2015; Dessert et 60 al., 2001; Gaillardet et al., 1999; Li et al., 2016; P Louvat and Allègre, 1998; West et al., 61 2005). Over geological timescale (> $10^5$  years), silicate weathering, with organic matter burial, 62 are major sinks of atmospheric carbon. Weathering of silicate rocks consumes atmospheric 63 64  $CO_2$  and transforms it to dissolved  $HCO_3^-$  (Gaillardet et al., 1999), which further precipitates with calcium carbonate in the ocean. As  $CO_2$  is a greenhouse gas, silicate rock weathering can 65 66 be a critical feedback control on the climate (Brady and Carroll, 1994; Walker et al., 1981). However, at present, the relative importance of the various continental areas and various types 67 68 of lithologies having played a key role in the carbon cycle is still debated, in particular over the Cenozoic. The emplacement of the Himalayas (and its uplift) may have been critical but 69 70 this is not consensual thus far. Other mountainous regions may have been of importance, in 71 particular those rich in basaltic lithologies, and are little considered yet (Li and Elderfield, 72 2013). Indeed, all silicate rocks do not present the same sensitivity to weathering. Among 73 them, basaltic rocks can be weathered more easily than any other igneous ones (e.g. Dessert et al., 2003). Some studies (Dessert et al., 2003; Gaillardet et al., 1999; Louvat and Allègre, 74 1997; P Louvat and Allègre, 1998) indicate that basaltic rocks are weathered 8 time faster 75 76 than granitic rocks. Millot et al., (2002) show that the silicate weathering rate is 10 to 100 times larger for basaltic regions than for granitic areas. Basalt weathering has thus been 77 estimated as an important sink of CO<sub>2</sub> at the global scale (Dessert et al., 2003) and it is 78 79 calculated to be responsible of 30% of the present-day CO<sub>2</sub> consumption (Gaillardet et al., 1999). The Deccan Traps were investigated in great details (e.g. Das et al., 2005; Dessert et 80 81 al., 2001; Kisakürek et al., 2004; Vigier et al., 2005), as well as the Siberian Traps (e.g. Pokrovsky et al., 2005; Zakharova et al., 2005). However, some large basaltic provinces have 82 not been studied yet. For example, weathering of the Ethiopian Traps (upper Blue Nile Basin) 83 is not well constrained, despite the importance of this region in terms of climate, size, 84 tectonics and age (30 Ma). A recent carbon-climate modeling study highlights that the 85 Ethiopian Traps were the world-wide most important hotspot of silicate weathering during the 86 87 Oligocene (Lefebvre et al., 2013). The invoked reason is that, during this period, the

Ethiopian Traps were located within the equatorial latitudes, affected by high precipitation 88 rates and high temperatures, two parameters known to strongly increase basaltic weathering 89 rates (Dessert et al., 2003; Goddéris et al., 2013). At present the Ethiopian Traps are 90 influenced by monsoonal climate, with most precipitation occurring during the summer 91 period. The monsoon intensity in North-East Africa fluctuated significantly in the past, with, 92 for instance, particularly high peaks during high insolation periods such as the African Humid 93 period (14.5 to 8 ka, (Lamb et al., 2018; Shanahan et al., 2015)). Numerous studies show that 94 monsoon variations controlled physical erosion and the Blue Nile sediment flux during the 95 96 Quaternary period (Revel et al., 2015, 2014, 2010). As shown by lithium isotope data for the 97 Nile Basin (Bastian et al., 2017) and for the Himalayas (Dosseto et al., 2015), or hafnium 98 isotopes for the Congo Basin (Bayon et al., 2012), continental silicate weathering can respond rapidly to monsoon fluctuations. Thus, the impact of the Ethiopian Traps on the atmospheric-99 100 ocean carbon system deserves to be assessed both at present and in the past.

In fact, from sedimentary records located in the Nile deep sea fan, the importance of physical 101 102 erosion in this region, and the reactivity of sediment transport to hydro-climate change have extensively been highlighted. Neodymium isotopes measured in the silt fractions (Blanchet et 103 104 al., 2014, 2013, Revel et al., 2015, 2010) of Quaternary sediments have evidenced co-105 variations of terrigenous flux from the Blue Nile River with monsoon intensity. More recently, (Bastian et al., 2017) have explored the use of Li isotope composition of the clay 106 107 fractions to determine paleo-variations of silicate weathering in this region. Indeed, lithium isotope composition of river waters and sediments can be used to characterize the weathering 108 intensity (Bouchez et al., 2014, 2013, Dellinger et al., 2017, 2015, 2014; Pogge von 109 Strandmann et al., 2017). Lithium is powerful to trace silicate weathering because it is 110 principally derived from the chemical erosion of silicate minerals (Kisakurek et al., 2005; 111 Millot et al., 2010). During chemical erosion in soils or in weathering profiles, no significant 112 fractionation during rock dissolution can be highlighted (Pistiner and Henderson, 2003; Ryu 113 et al., 2014; Verney-Carron et al., 2011; Wimpenny et al., 2010). In contrast, large isotope 114 fractionations occur during clay neoformation, in favor of the light <sup>6</sup>Li (into octahedral sites). 115 At a watershed scale, simple modeling taking into account most recent knowledge on the 116 magnitude of Li isotope fractionation indicate that high clay  $\delta^7$ Li may correspond to low ratio 117 of dissolution/clay neoformation rate (Bastian et al., 2017; Bouchez et al., 2013), or low 118 119 weathering intensity (Dellinger et al., 2015).

A large amount of Li studies is focused on the river dissolved phases (Millot et al., 2010;
Pogge von Strandmann et al., 2017; Vigier et al., 2009). However, recently, the potential of

river suspended particles (Dellinger et al., 2017, 2015, 2014; Millot et al., 2002), and of 122 sediments deposited in terraces, lake or delta (Bastian et al., 2017; Dosseto et al., 2015) to 123 trace past variations of silicate weathering has also been demonstrated (Bastian et al., 2017; 124 Dosseto et al., 2015; Lechler et al., 2015). By studying the Nile Delta during the last 32kyr, 125 Bastian et al (2017) show that clays deposited by the Nile River were mostly smectite formed 126 in the Ethiopian Traps. Their  $\delta^7$ Li values co-vary with climate proxies, and in particular with 127 precipitation variations. The controls of lithium isotope composition for clays formed in the 128 Ethiopian Traps are therefore important to investigate further, and should help to refine our 129 130 understanding of the relationships between climate and basalt weathering.

In this study, we target to provide chemical erosion rates and related  $CO_2$  consumption rate in the region of the Blue Nile River, the major river draining the Ethiopian Traps basins. For this, we use the first large dataset of chemical compositions of water and particles from most rivers draining the Ethiopian Traps. The objectives are (1) to study weathering rates, their controls, and their relationships with clay Li isotope composition in this region, and (2) to give an estimation of  $CO_2$  consumption rate during an intensive runoff period, well-known and well evidenced in Nile delta sedimentary cores, the African Humid Period (14.5-8kyr).

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#### 139 **2. Study area**

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The Ethiopian Continental Flood Basalt (CFB) province is located in the North and West 141 142 Ethiopian highlands (Fig 1 & Fig S1). It covers a surface of about 800,000 km<sup>2</sup> and has an estimated volume ranging between 1 and 1.5 million km<sup>3</sup> (Rochette et al., 1998). They 143 erupted 30 Ma ago over a short time period, 1-2 Ma (Hofmann et al., 1998). The lave pile 144 thickness varies from 500 to 2000 m with maximum thickness of 3000 m in the northern part 145 of the plateau (Mohr. P, 1988). This CFB province is mainly composed by transitional basalts 146 147 of low-Ti and high-Ti affinities (Pik et al., 1999, 1998), and is covered by a rhyolite terminal formation on the western part of the plateau, along the main escarpment (Ayalew et al., 2002). 148 149 The Ethiopian Traps exhibits a mean altitude of ~ 3000 m with flat under lying remnants of the original Oligo-Miocene surface. The volcanoes emplaced on the plateau present a higher 150 elevation up to 4600 meters. The Ethiopian Highlands have been uplifted since at least 25 Ma 151 (Pik et al., 2003) with concomitant development of the two main basins of the Nile and 152 Tekeze rivers. The plateau presents important border escarpments with steep slopes, 153 especially along the roughly N-S marginal graben on its Eastern side (Fig. 1). These highlands 154 have been dissected by rapid development and propagation of the Nile and Tekeze basins 155

towards its most and early uplifted Eastern edge (Pik et al., 2003), which acted as the main tectonic drainage divide since Oligo-Miocene times. This tectonic divides is separating the two large westward basins (Nile and Tekeze) from the short and parallel eastward basins developed along the marginal graben after initiation of extension 25 Ma ago (Wolfenden et al., 2004).

This tropical region is submitted to the African monsoon, with a mean annual temperature 161 around 21°C. Thus, 90% of the annual precipitations occur during the summer month 162 (Korecha and Barnston, 2007) (Fig 1). Precipitations display an East-West gradient in 163 Ethiopia: rainfall increase from 200 mm/year on the Indian Ocean to 1800 mm/year in the 164 Highlands. At present, the Blue Nile River (major affluent of the Nile River) and its 165 tributaries drain the Ethiopian Traps. Thus, this high topography region is estimated to 166 contribute to 60 % of the total water discharge of the main Nile and to 60 % of its terrigenous 167 168 sediments (Fielding et al., 2017; Garzanti et al., 2015; Nyssen et al., 2005). In the past, during the last 30 kyr, the positioning and the intensity of the monsoon rain belt has oscillated 169 170 (Shanahan et al., 2015), in relation with the precession parameter (Tuenter et al., 2003) which lead to stronger precipitation every 21 kyrs. These strong runoff variations are recorded by 171 172 Ethiopian lake sediments (Amaral et al., 2013) and by the sediments transported by the Nile River and deposited in its delta (Blanchet et al., 2014; Castañeda et al., 2016; Revel et al., 173 2015, 2010). 174

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#### 176 **3. Sampling and methods**

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178 3.1. Sampling

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All samples come from the northern Ethiopian plateau (Fig 1), which encompasses the large 180 catchment of the Blue Nile (64 000 km<sup>2</sup>), several smaller sub-basins (1000 to 6000 km<sup>2</sup>), as 181 well as small catchments emplaced directly on the top of the plateau (5 to 100 km<sup>2</sup>). Two 182 183 sampling campaigns have been carried out in June 2004 (ETH04) and in March 2005 (ETH05). At each sample site, sand, mud and river waters were collected. Waters were 184 filtered on the field with 0.2µm filters and immediately acidified with purified nitric acid. 185 Sands and muds were sampled on the river banks after mixing 5 different aliquots of about 186 100g each. The difference between mud and sand is the visible granulometry: sand samples 187 correspond to coarse sized mineral (> 2mm grain) rich-particles and the mud samples to the 188

finest grain deposits. At the laboratory, clay-size fractions were extracted from the mudsamples before chemical and isotope analyses (see section 3.3.2).

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192 3.2. Runoff Modeling

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The hydrology of the Blue Nile upper basins is strongly controlled by seasonal precipitations, 194 with strong rains and flooding observed during the summer monsoon, from June to September 195 (e.g. Conway, 2009). These conditions are not appropriate for providing a reliable estimate of 196 197 annual discharge and runoff for each studied zone, using classical methods. In order to provide these estimations for each sampled sub-basin, necessary for weathering rate 198 199 calculations, we used a numerical model based on the IDRISI © GIS software. This model 200 was run using the plateau topography and available public rainfall data. In order to avoid 201 unconstrained biases linked to evapotranspiration, the output has been convincingly calibrated with current discharge data available for the upper Blue Nile basin (Conway, 1997). These 202 203 calibrated discharge and runoff data for each sub-basin are presented in Table 1.

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205 3.3. Analytical protocol

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207 3.3.1. Chemical analyses

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209 Major and trace element concentrations were analyzed in waters and sediments using ICP-**ICP-MS** SARM 210 AES and at the National Facilities in Nancy (France) (http://helium.crpg.cnrs-nancy.fr/SARM/index.html). Anions have been measured using ionic 211 chromatography Dionex ICS3000-S with AG11-HC/AS11-HC column. Corresponding 212 uncertainties and detection limits are described (http://helium.crpg.cnrs-213 in nancy.fr/SARM/pages/roches.html). 214

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216 3.3.2. Lithium isotope analyses of the clay fractions

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Clays were extracted from the mud samples, by decantation of the sediments in 50ml of ultrapure water mixed with  $20\mu$ l of sodium hexametaphosphate (100mg/l). This separation allows to extract particles with a diameter and a density lower than 2 µm and 1.8 respectively. Clay fractions were finely crushed into an agate mortar and 10 mg of the fine powder was mineralized using a concentrated HF/HNO<sub>3</sub>/HCl solution in a clean laboratory at LOV, using ultrapure acids. The solution was evaporated and the residue dissolved in 1.00N HCl. The
total dissolution of samples was systematically verified by the absence of residue after 20 min
of centrifuging at 4000rpm.

For major and trace element analyses, an aliquot was diluted 300 times and analyzed by ICP-OES. For lithium isotope analyses, an aliquot containing  $\approx 60$ ng Li was introduced on a cationic resin column and eluted with 1.00N HCl, following the method published in (Bastian et al., 2017; Vigier et al., 2008). This separation was performed twice and allowed Li recovery and purification. For all samples, the total cationic concentration of the sample was constrained to be less than 10% of the total cationic capacity of the column resin. All seawater and rock reference materials were submitted to the exact same protocol.

Lithium isotope measurements were performed using a Neptune Plus (Thermofisher) multi-233 collector inductively coupled plasma spectrometer (MS-ICP-MS) at the Ecole Nationale 234 Supérieur de Lyon (CNRS National Facilities). Sample was introduced through an Aridus II 235 desolvating system before ionization to increase sensitivity (1V<sup>7</sup>Li / ppb Li). We used the jet 236 and X cones, as described in (Balter and Vigier, 2014). To correct for instrumental mass bias, 237 we used the standard bracketing technique, using the LSVEC standard. The total blank 238 procedure is systematically lower than 10pg Li. The accuracy of isotopic analysis was verified 239 240 by measuring Li7-N reference solution (Carignan et al., 2007) and various reference materials (BE-N basalt, GH granite and seawater). Without chemical purification, an average  $\delta^7$ Li value 241 242 of 30.2±0.4% (2SD, n=32) is obtained for the pure Li7-N standard, in agreement with published and theoretical value. After chemical purification, mean  $\delta^7$ Li values are 30.3±0.4‰ 243 244 (2SD, n=22), 5.12±0.9% (2SD, n=5), 1.64 ±0.6% (2SD, n=3) and 31.1±0.6% (2SD, n=6), for Li7-N, BE-N basalt, GH granite and seawater respectively. All values for reference materials 245 246 compare well with published values (Carignan et al., 2007; Millot et al., 2004).

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### 248 **4. Results**

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- 250 4.1. Runoff and discharge
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As described in section 3.2, we have estimated runoff values using a specific numerical model including published data for rainfall and discharge, and regional topography. The runoff values determined for 19 sub-basins range from 190 to 273 mm/yr with a mean value of 242 mm/yr. The corresponding calculated discharges range from  $11_x10^5$  m<sup>3</sup>/yr (for the Ulet Wenz River) to  $1.8_x10^{10}$  m<sup>3</sup>/yr (for the Blue Nile river), with a mean value of  $19_x10^8$  m<sup>3</sup>/yr (Table 1). For the Blue Nile river, the calculated discharge is close to the annual discharge measured at the Roseires Dam Station (http://www.grdc.sr.unh.edu/html/Polygons/P1663800.html) :  $4.7_x 10^{10} \text{ m}^3/\text{yr}$ . The estimated average runoff value of 242 mm/yr for the Ethiopian Traps is in line with the value published in ((Dessert et al., 2003), 129 mm/year). These two comparisons indicate that our model estimates representative values for the runoff and river discharge.

The average runoff for this region is two times lower than the estimated value for the Deccan 262 Traps (463 mm/year). It is also lower than the one determined for the Siberian Traps (324 263 mm/yr). Compared to small tropical basaltic areas such as Hawaii (1612 mm/yr) and the 264 265 Reunion Island (2433 mm/yr), the Ethiopian runoff is also low. This is partly due to their 266 topography, their small surface area compared to the Ethiopian Traps (800,000 km<sup>2</sup> vs 16760 km<sup>2</sup> in Hawaii and 2512 km<sup>2</sup> in the Reunion Island), and the occurrence of frequent 267 hurricanes and tropical storms. In Iceland, high runoff (1883-2432 mm/yr) values are mostly 268 269 due to ice melting, which introduces water in the riverine system during the summer season 270 (Gislason et al., 2009).

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272 4.2. Major and trace elements

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274 4.2.1 River waters

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In waters, the dominant cations are, in increasing order of concentrations: Si, Mg, Na, and Ca, with mean values of 447 ( $\pm$  219), 1768 ( $\pm$  502), 712 ( $\pm$  332) and 883 ( $\pm$  417) µmol/l respectively. These concentrations are corrected from the atmospheric input using Cl concentration and oceanic X/Cl molar ratios, following the method detailed in (Berner and Berner, 1987). After corrections the concentration of Mg\*, Na\*, and Ca\* are 880 ( $\pm$  415), 68 ( $\pm$  26) and 574 ( $\pm$  285) µmol/l respectively.

282 On average, molar Ca\*/Na\* and Mg\*/Na\* ratio are 2.5 ( $\pm$  0.9) and 0.4 ( $\pm$  0.6), respectively

283 (Table 2). For comparison, average Ca/Na and Mg/Nain Ethiopian basalts are  $3.9 (\pm 1.3)$  and

 $3.2 (\pm 2.7)$  respectively, and in the Ethiopian rhyolite are 0.07 ( $\pm 0.06$ ) and 0.03 ( $\pm 0.04$ ).

When compared with other regions of similar lithologies, river water chemical composition displays roughly a single trend between basalts and rhyolites (Fig. 2). From this trend, it can be estimated that the dissolved contribution coming from rhyolite weathering is low (<15%) compared to basaltic rocks.

The dominant anions are, in increasing order of concentrations:  $HCO_3^- SO_4^{2-}$ ,  $Cl^-$ ,  $NO_3^-$ , and F, with mean values of 3170 (406 to 6039) 202 (15 to 880), 156 (15 to 348), 71 (4 to 292) and

- 10 (5 to 21) µmol/l respectively. The anion concentrations are typical of those measured in
  other rivers draining basaltic regions (e.g. Dessert et al., 2001).
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- 4.2.2 River bed particles
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Sands are enriched in SiO<sub>2</sub> compared to mud samples (Table 3),  $(51.9 \pm 10.6 \%$  on average vs 296  $43.2 \pm 2.6$  % for muds) and Na<sub>2</sub>O (2.2 ± 0.8 % vs 1.1 ± 0.5 %), while depleted in Al<sub>2</sub>O<sub>3</sub> (11.7 297  $\pm$  3.4 % vs 13.0  $\pm$  3.5 % in mud), CaO (7.0  $\pm$  3.4 % vs 4.7  $\pm$  2.1 % in mud) and Fe<sub>2</sub>O<sub>3</sub> (11.4  $\pm$ 298 299 3.4 % vs 13.5  $\pm$  1.5%). TiO<sub>2</sub> and MgO (5.7  $\pm$  4.0 % vs 5.8  $\pm$  3.1%) concentrations are similar between sands and muds: ( $\approx$ 3.1 ± 1.4 wt % and  $\approx$ 5.8 ± 3.1 wt % respectively). Si/Al ratio is 300 lower by  $\approx 30\%$  in muds compared to sands, which is consistent with occurrence of 301 aluminosilicate minerals (clays/smectite) in the collected muds. The chemical composition of 302 303  $<2\mu$ m clay fractions extracted from mud samples, contains, on average, 24.2% (± 4.7 %) Al<sub>2</sub>O<sub>3</sub>, 22.2% ( $\pm$  1.8 %) Fe<sub>2</sub>O<sub>3</sub>, and 15.1% ( $\pm$  6.9 %) MgO. We note that the mud samples 304 305 collected in river banks are not representative of the clay fraction separated at the laboratory and rather reflects a mixing between clays and primary phases. Thus, clay fractions (<2µm) 306 307 were preferentially used for assessing weathering processes in this region.

308 Some trace elements measured in the solid phases can be used to study chemical erosion degree or intensity (Louvat and Allègre, 1997; P Louvat and Allègre, 1998). In the diagram 309 Sr/Th versus U/Th (Fig 3, Table 2 and 3), when compared to bedrocks, Ethiopian river waters 310 display elevated U/Th and Sr/Th ratios (respectively >2 and > 8405) while the solid phases 311 display low U/Th and Sr/Th values (respectively <0.3 and <87.8). Sr and U are two mobile 312 elements during water-rock interactions and are usually depleted in the solid phase, in contrast 313 with Th, which is an immobile element that can easily adsorb, except in organic-rich 314 environments (Dupré et al., 1999). Figure 3 also indicates that the river particles are mostly 315 derived from basaltic rocks, as previously described, and that the influence from rhyolite 316 weathering is minimal, except for one sample (Nazero Wenz), which is located in a rhyolite 317 318 rich area (Figure S1).

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320 4.3 Lithium isotope compositions of clay size ( $<2\mu m$ ) fractions

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Ethiopian bedrock samples exhibit a mean  $\delta^7$ Li value of 5±0.5‰ (n=3) (Bastian et al., 2017) (Fig. 4), which compares well with the range published for intraplate basalts. Compared to these rocks, or to tholeiitic basalts (3.1 to 5.2 ‰, (Elliott et al., 2006)), and to OIB basalts (3.1  $5.2 \%_0$ , (Genske et al., 2014)) clays deposited by Ethiopian rivers are significantly enriched in <sup>6</sup>Li: clay  $\delta^7$ Li values range from  $0.3\%_0 (\pm 0.5\%_0)$  to  $4\%_0 (\pm 0.5\%_0)$ , with a mean value of  $2.0\%_0 (\pm 1.2\%_0)$ . This is consistent with preferential enrichment in light lithium isotope during smectite neoformation, as seen experimentally (Vigier et al., 2008) and in basaltic soils (Huh et al., 2004; Kisakürek et al., 2004; Pistiner and Henderson, 2003; Ryu et al., 2014).

In the literature, a large range of  $\delta^7 Li$  values is observed in suspended particles (SPM) of 330 rivers draining basaltic regions e.g. in Iceland (with values from -1.3 to 7.5%, (von 331 Strandmann et al., 2006)) and in the Azores archipelago (3.5 to 9.5%, (von Strandmann et al., 332 2010)). Also in soils, the range of published  $\delta^7$ Li for bulk soils samples is large (-15 to 10%), 333 (Clergue et al., 2015; Pistiner and Henderson, 2003; Ryu et al., 2014; Schmitt et al., 2012; von 334 335 Strandmann et al., 2012)). These observed variations are possibly due to mixing between clay and primary minerals, because these samples are not necessarily representative of clay pure 336 337 endmember (Dellinger et al., 2014). Here, clays were extracted from river sediments in order to minimize the influence of primary minerals. 338

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#### 340 **5. Discussion**

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# 5.1. Assessing weathering processes at the basin scale

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As described in section 4.2, chemical and isotopic compositions of river waters and particles 344 can give indications on weathering rates and/or intensity at the watershed scale. Some 345 elements are particularly mobile during basalt alteration (e.g. Sr, K, Na), and preferentially 346 lost from weathering profiles after a short period of time (Chadwick et al., 1999; Ryu et al., 347 2014). Thus, as described in section 4.2, Sr and U are depleted in sands and mud samples, 348 while enriched into the dissolved phases (Fig. 2) of the corresponding rivers. Also, Na, Ca and 349 Mg concentrations are typical of waters draining basaltic rocks (Fig. 3). However, using 350 elementary ratios, or elementary concentrations, it is often difficult to distinguish the role of 351 352 lithology from the one of dissolution or leaching process, because of the chemical diversity of the drained bedrocks. As described in previous works (Pistiner and Henderson, 2003; Vigier 353 et al., 2008; Wimpenny et al., 2010), lithium isotopes provide a complementary mean to 354 examine chemical erosion processes (Bastian et al., 2017; Dellinger et al., 2015). Bastian et 355 al., (2017), have analyzed 55 clay fractions extracted from Nile deep sea fan core sediments. 356 Their  $\delta^7$ Li values vary between 4 and -1.2 % during the last 32,000 years. In their study, clay 357  $\delta^7$ Li were related to the ratio between dissolved Li flux and the flux of Li incorporated in the 358

secondary phases ( $F_{lea}^{Li}/F_{sp}^{Li}$ ). In rivers currently draining the Ethiopian Traps, clay fractions display variable  $\delta^7$ Li values, from 0.3% to 4.0%, a range that is similar to the 0-32 kyr clays deposited in the Nile delta. Clay  $\delta^7$ Li and Mg/Ti ratios display a positive correlation (shown in fig. 4), which suggests that both proxies are impacted by similar processes during basalt weathering. This is not surprising since Li<sup>+</sup> essentially substitutes to Mg<sup>2+</sup> during smectite formation (Decarreau et al., 2012; Vigier et al., 2008), and smectite is the most abundant phase carried by rivers draining the Ethiopian Traps (Garzanti et al., 2015).

Using a simple modeling, based on mass balance, clay  $\delta^7$ Li values have been shown to be 366 essentially explained by the ratio of leaching versus clay formation rates, and thus reflect 367 different weathering intensities (Bastian et al., 2017; Bouchez et al., 2013; Vigier et al., 2009; 368 369 Vigier and Goddéris, 2015; von Strandmann et al., 2012). The behavior of Li isotopes at the scale of a watershed or of a continent has been described with the following equations, 370 considering a simple mass balance between bedrock, rivers and soils at the watershed scale. It 371 is defined such that all the lithium released by rock dissolution is either incorporated into 372 secondary phases formed in weathering profiles or released into superficial waters (and in fine 373 in rivers). 374

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$$F_{lea}^{Li}\delta_{lea} = F_{sp}^{Li}\delta_{sp} + F_{riv}^{Li}\delta_{riv} \qquad \text{equation (1)}$$

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where  $F_{lea}^{Li}$ ,  $F_{sp}^{Li}$ , and  $F_{riv}^{Li}$  are the Li flux related to dissolution, secondary phase precipitation and soil water outflow respectively.  $\delta_{lea}$ ,  $\delta_{sp}$ ,  $\delta_{riv}$  are the Li isotopic signature ( $\delta^7$ Li value) of the dissolved phase, the secondary phase, the river water and the bedrock, respectively. Since basalt dissolution is isotopically congruent,  $\delta_{lea} = \delta_{Bedrock}$  (Verney-Carron et al., 2011; Wimpenny et al., 2010). The isotope fractionation between dissolved phase and the secondary phases is noted  $\Delta_{land}$ . After simplification, the isotope composition of the secondary phases is given by:

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$$\delta_{\rm sp} = \frac{\Delta_{\rm land}}{F_{lea}^{Li}/F_{\rm sp}^{Li}} + \delta_{\rm BedRock} - \Delta_{\rm land} \qquad \text{Equation (2)}$$

387

388 This equation states that if  $F_{sp}^{Li}$  is close to  $F_{lea}^{Li}$ , then  $\delta_{sp} = \delta_{BedRock}$ . If  $F_{sp}^{Li}$  is low compared 389 to  $F_{lea}^{Li}$ , then  $\delta_{sp} = \Delta_{land}$ .

The positive correlation between clay  $\delta^7$ Li and Mg/Ti cannot be explained by a binary mixing between bedrocks and Li-rich clays, as illustrated in Figure 4. Thus, a similar mass balance between rocks and weathering products, as for  $\delta^7$ Li, was applied to clay Mg/Ti. For solving it, we assumed that no Ti is released into water during dissolution (due to the high immobility of this element during water/rock interactions) (Ti<sub>*rlea*</sub> = 0). Thus Mg/Ti ratio of secondary phases can be described as:

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 $({}^{Mg}/_{Ti})_{sp} = \frac{1}{\frac{F_{lea}^{Mg}}{F_{lea}^{Mg}} + \frac{Ti_{clay}^{mean}}{F_{sp}^{Mg}}}$ Equation (3)

399

400 The relation between the Li and the Mg flux becomes:

401

402 
$$\frac{F_{lea}^{Li}}{F_{sp}^{Li}} \cdot \beta = \frac{F_{lea}^{Mg}}{F_{sp}^{Mg}} \qquad \text{Equation (4)}$$

403 with  $\beta$  reflecting the Li/Mg ratio of the clay fraction (normalized to the source rock one).

As shown in Figure 4, some basins are characterized by low clay  $\delta^7$ Li and low Mg/Ti values, (ie Jara Wenz (ETH05-11), Borkena Wenz (ETH05-13), Robi Wenz (ETH05-4), Meri Wenz (ETH05-28), Tikur Wenz (ETH05-19)). For these small basins, the weathering model indicates that soil formation rate (F<sub>sp</sub>) is low compared to the leaching rate (F<sub>lea</sub>). Other areas display high clay  $\delta^7$ Li values associated to high Mg/Ti ratios (Ketching Abeba Wenz (ETH05-25), Gereb Ara Wenz (ETH05-40), Tekeze (ETH0535), Hormat Wenz (ETH05-47), Ala-Wiha (ETH05-20), Nazero Wenz (ETH05-6)).

Following our modeling, the various  $\delta^7$ Li and Mg/Ti values are best explained by variations of the weathering intensity, leading also to a large range of clay Li/Mg ( $\beta$  parameter). For a large part, the  $\beta$  values are greater than 1 (Fig. 4). This indicates that Li is more retained in the weathering profiles than Mg (compared to source rocks). This is consistent with observations made in various Hawaiian weathering profiles where kaolinite prevail (Ryu et al., 2014).

Thus, the trend between clay  $\delta^7$ Li and clay Mg/Ti is likely due to variable weathering conditions. However, no clear relationship with the runoff, the topography or vegetation can be highlighted for this group of rivers. Since Mg is involved in the vegetation cycle, and Li is not, perhaps a loss of Mg in relation with recent plant and root activity can also explain part of this variability. More work would certainly need to be undertaken in the most vegetated areas of this region since no convincing correlation between  $\delta^7$ Li or Mg/Ti and physical parameters such as the slope, runoff, vegetation and land use can be highlighted here. This suggests that, at present, basalt weathering and soil production rate in the Blue Nile Basin are variable in space, and that this variation is likely to be a complex function of several parameters.

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#### 5.2. Weathering rates and controls in the Upper Blue Nile Basin

- As discussed above, the relatively large range of clay Mg/Ti and  $\delta^7$ Li reflects a significant 428 variation of weathering conditions in the Ethiopian Traps region while the lithology is 429 dominantly basaltic. It is possible to use the chemical composition of river waters in order to 430 quantify present-day weathering rates (Table 2) at the scale of each corresponding watershed 431 (e.g. Gaillardet et al., 1999; Stallard and Edmond, 1983). The Total Dissolved Solid (TDS) is 432 defined by the sum of major mobile cations (Ca, Mg, K and Na) concentrations in the 433 dissolved phases of rivers. These concentrations are corrected from the atmospheric input 434 using Cl concentration and oceanic X/Cl molar ratios, following the method detailed in (Das 435 et al., 2005). TDS\* values (\* indicate corrected from atmospheric inputs) estimated from 436 437 concentrations measurement in the dissolved phase of the Blue Nile Basin rivers range from 9.5 to 111.0 mg/l, with a mean value of 67 mg/l. In the diagram TDS\* versus temperature, the 438 439 Blue Nile tributaries plot along the same trend defined by all other basaltic regions studied thus far (Fig. 5a). This first indicates the good representative nature of our sampling and 440 441 methods. The average TDS\* for the Ethiopian Traps is as high as the one determined for the Deccan Traps (54 mg/l) and for Java Island (43 mg/l) (Dessert et al., 2003; Li et al., 2016), 442 443 which are also characterized by elevated annual temperature (27°C and 24.8 °C respectively). 444 These three regions display river TDS\* values significantly greater than in Hawaii (7.4 mg/l) 445 and in Iceland (8.8-10.3 mg/l), displaying both lower surface temperatures on average (16 and 2 °C respectively). This trend confirms that, for basaltic basins, temperature is a key 446 parameter which controls river TDS\* (Dessert et al., 2003; Li et al., 2016). 447
- Using the TDS\* values, the estimated annual discharge, and the watershed area, it is possible to determine silicate weathering rates. These rates in the Ethiopian Traps basins range from 2.6 to 24.4 tons/km<sup>2</sup>/year, with a mean value of 16.1 tons/km<sup>2</sup>/year (Table 2). For the Blue Nile River, sampled at the output of the Ethiopian Traps (see map Fig. 1), the silicate weathering rate is 11.3 tons/km<sup>2</sup>/year. The mean value estimated for chemical erosion rate in the Ethiopian Traps, based on 17 data (Table 2), is consistent with the positive correlation

previously observed with the runoff (see Fig. 5b, (Dessert et al., 2003)). In contrast to TDS\*, 454 weathering rates in Ethiopia are low compared to other regions of similar lithology. For 455 instance, the weathering rate in the Deccan Traps is 25.1 tons/km<sup>2</sup>/year, 48.4 tons/km<sup>2</sup>/year 456 for La Reunion, and 152 tons/km<sup>2</sup>/year for Java (Dessert et al., 2003). Lower runoff in the 457 Ethiopian Traps (242 mm/years) compared to these regions is certainly a key factor. However, 458 other parameters may also play a role, in particular temperatures, the age of the basaltic rocks 459 (Li et al., 2016), the soil age (Ryu et al., 2014; Vigier et al., 2005), and formation of glassy 460 material in volcanically active area (e.g. Java Island and Iceland) (Gislason et al., 2009; 461 462 Gislason and Oelkers, 2003; Vigier et al., 2009).

Overall, both river particles and water composition can provide valuable information on 463 chemical erosion rate in the various basins. However here there is no correlation between 464 river particles and water chemical compositions. This is likely to be due to a recent 465 466 decoupling between soil (or weathering profile) formation and river aqueous phase. Indeed, large soil reservoir where clays have formed can encompass different weathering conditions, 467 468 issued from past climate variations, and may therefore not be representative of the current state. Recent observations in the Nile deltaic sediments show that both chemical and physical 469 470 erosion responded rapidly to hydro-climate change in the last 30kyr (Bastian et al., 2017). 471 This argues for a residence time of clay-rich sediments that is not above a few thousand years at most, in coherence with elevated topography of the Traps where clays mainly come from. 472 The observed decoupling between clays and waters must then have occurred just recently, 473 perhaps due to intensification of anthropic activities in this region (Lanckriet et al., 2015) 474

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# 476 **5.3.** CO<sub>2</sub> consumption and SO<sub>4</sub><sup>2-</sup> concentration due to silicate weathering

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From river water chemical compositions, it is possible to estimate the CO<sub>2</sub> consumption rates 478 related to silicate weathering. In the literature, several methods have been used, using 479 corrections from source rocks in mixed lithology basins, calcium concentration in silicate 480 481 rivers (Goddéris et al., 2013), or HCO<sub>3</sub><sup>-</sup> concentrations in carbonate-free watersheds (e.g. Das et al., 2005). Since the Ethiopian Traps are essentially constituted of silicate rocks, with little 482 vegetated areas, major cations concentrations in rivers have only been corrected for 483 atmospheric inputs, as described in section 5.1. HCO<sub>3</sub><sup>-</sup> concentrations are determined using 484 the following electronic mass balance: 485

487 
$$[HCO_3^-] = 2[Ca^{2+}] + [K^+] + 2[Mg^{2+}] - 2[SO_4^{-2}] - [F^-] - [NO_3^-] Equation (5)$$

where all concentrations are corrected for atmospheric inputs (see section 4.2.1). The average 489 pH of the Ethiopian rivers (7.5-8.2, (MERID, 2005)) makes it to possible to assume that 490  $HCO_3^{-1}$  is the dominant anion compared to  $CO_3^{2^{-1}}$ . Thus, in order to determine the  $CO_2$ 491 consumption related to silicate weathering, we consider that 1 mole of HCO3<sup>-</sup> exported by 492 rivers corresponds to 1 mole of consumed atmospheric CO<sub>2</sub>. CO<sub>2</sub> consumption rates are then 493 calculated using the river discharge and the basin surface area (for specific rates). Some recent 494 studies (Calmels et al., 2007; Li et al., 2008; Torres et al., 2016) show that sulfate mineral 495 dissolution produces sulfuric acid that can enhance bedrock and mineral dissolution, in 496 particular in sedimentary or carbonate rich context. Any silicate weathering due to sulfuric 497 acid may lead to an overestimation of the CO<sub>2</sub> consumption rate if this effect is not considered 498 into the calculations. In the rivers studied here, dissolved sulfate concentrations do not 499 500 correlate to dissolved nitrate but rather with elements that are mobile during silicate dissolution (for example Ca and Mg). This suggests that sulfates are not significantly related 501 502 to fertilizer addition but may mostly derive from weathering and oxidizing processes. This is not surprising since this region is not intensively cultivated, and is also supported by small 503 504 amounts of nitrate in waters (see section 4.2.1). The use of Equation (5) to calculate  $HCO_3^-$ 505 allows us to avoid a potential bias due to sulfuric acid in the determination of CO<sub>2</sub> 506 consumption rate.

Table 2 show that specific CO<sub>2</sub> consumption rate due to basalt dissolution in Ethiopian Traps 507 508 vary from 0.11x10<sup>6</sup> mol/km<sup>2</sup>/yr (for the Ulet-Wenz basin) to 1.33 x10<sup>6</sup> mol/km<sup>2</sup>/yr (for the Ketchiin Abeba Wenz), and is 0.76x10<sup>6</sup> mol/km<sup>2</sup>/yr on average. The CO<sub>2</sub> consumption rate 509 cover a large range, possible due to local - small scale - effects. Thus, for the comparison 510 with literature data, we use the two values estimated for the Blue Nile River, which integrates 511 most waters draining the Ethiopian Traps. The Blue Nile River was sampled at two different 512 seasons (ETH04-2; Juin-04 and ETH05-52; Mars -05). The two samples lead to similar CO<sub>2</sub> 513 consumption rate, of 0.49 and 0.52 mol/km<sup>2</sup>/yr respectively, but are low compared to other 514 basaltic regions. For comparison, this flux is estimated to be 1.26 10<sup>6</sup> mol/km<sup>2</sup>/yr for the 515 Deccan Traps and 6.41 10<sup>6</sup> mol/km<sup>2</sup>/yr for Java Island (Dessert et al., 2003). The CO<sub>2</sub> 516 consumption rate due to silicate weathering in the Blue Nile basin is slightly higher, than the 517 value published by (Dessert et al., 2003) for the Ethiopian Traps, i.e. 0.15x10<sup>6</sup> mol/km<sup>2</sup>/yr. 518 When using the Dessert et al., (2003) theoretical equation  $fCO_2 = R_{f x} 323.44 \exp(0.0642 \text{ T})$ 519 where fCO<sub>2</sub>, R<sub>f</sub> and T are respectively the specific atmospheric CO<sub>2</sub> consumption 520

521 (mol/km<sup>2</sup>/year), the runoff (mm/year) and the temperature (°C), the corresponding 522 consumption rate is also slightly lower ( $0.16 \times 10^6$  mol/km<sup>2</sup>/year) that our calculated value.

In order to determine the relative role of the Blue Nile Basin in the present-day carbon cycle, 523 one needs to estimate its total  $CO_2$  consumption flux (in mol/year). The average  $CO_2$ 524 consumption rate is calculated to be  $0.65 \times 10^{12}$  mol/year. This is significantly higher than in 525 Iceland (0.070  $\times 10^{12}$  mol/year), the Reunion Island (0.006  $\times 10^{12}$  mol/year) or the Azores 526  $(0.0004 \text{ x}10^{12} \text{ mol/year})$  (see Fig. 6). However, this value remains lower than for the Deccan 527  $(0.392 \times 10^{12} \text{ mol/year})$  or than SE Asia  $(1.033 \times 10^{12} \text{ mol/year})$ . The upper Blue Nile represent 528 a CO<sub>2</sub> consumption close to the global average ((Gaillardet et al., 1999), see Fig. 6), and only 529 7.5 % of the total CO<sub>2</sub> consumed by silicate weathering on Earth. It corresponds to  $7.8.10^3$ Gt 530 of carbon consumed by year, which is the equivalent of 0.4% of the total human carbon 531 emission per year. Our results demonstrate that, at present, the Ethiopian Traps weathering 532 533 has a negligible impact on the carbon cycle, and thus on global climate.

In the past, this could however be different. First, as described in the Introduction, the 534 535 modeling study from Lefebvre et al., (2013), show that, during the Oligocene, the Ethiopian Traps was likely the most important region on Earth in terms of silicate weathering. More 536 537 recently, since 100kyr, the North East African region is known to have been submitted to 538 extreme variations of precipitation and runoff. The monsoon rain belt oscillated together with the precession cycle of 21,000 years. This induced succession of "humid" and "arid" periods 539 that affected sediment flux and physical erosion in this region. This is evidenced in lakes 540 where arid periods are characterized by hiatus or paleo-soils in the sedimentary records and 541 water level variations (for lake Turkana, lake Abhé (Forman et al., 2014; Garcin et al., 2009; 542 Hoelzmann et al., 2010)). This is also observed in the marine cores of the Nile deep-sea-fan, 543 with much higher sedimentation rates during humid periods (Revel et al., 2015, 2014, 2010). 544

Based on both observations and modeling, it is possible to provide first extrapolation of our 545 results to some critical period of the Quaternary period. Indeed, recent studies illustrate that 546 during this time, silicate weathering rates were able to respond rapidly to climate change 547 548 (Bastian et al., 2017; Dosseto et al., 2015; Rothacker et al., 2018). This region was submitted to large changes of the African Monsoon intensity (Shanahan et al., 2015) while runoff likely 549 plays a key role on basalt weathering and related CO<sub>2</sub> consumption ((Goddéris et al., 2013), 550 see also Fig. 5b and 8). Since 6 kyr, the climate in the Ethiopian Traps is characterized by 551 relatively low monsoonal precipitation and low runoff. However, between 15 and 8 kyr, 552 during the African Humid Period (AHP), this region was submitted to much higher 553 precipitation rates (Blanchet et al., 2013; Revel et al., 2014). Climate modeling studies, using 554

LMDZ and ORCHIDEE models, determined an increase of precipitation in the Ethiopian 555 Traps of 1.4 to 2.5 times larger during the AHP ((Marzin and Braconnot, 2009); Vadsaria and 556 Ramstein, Pers. Comm.). This corresponds to a runoff ranging between 340 and 600 557 mm/years. By applying this factor, CO<sub>2</sub> consumption rate during African Humid Period may 558 thus have increased up to values of  $0.91-1.5 \times 10^{12}$  mol/yr (brown bar in Fig. 6), which 559 corresponds to the most elevated current rates. These estimations are in line with the study of 560 (Lefebvre 2013), which showed that the importance of this region in the carbon cycle during 561 the Oligocene period. However, it is important to note that during the Oligocene, the global 562 temperature was also higher, while for the AHP, the runoff increase in North Africa is likely 563 not related to a significant increase of temperature (De Boer et al., 2013). 564

These results also suggest a potential increase of global CO<sub>2</sub> consumption during more humid periods due to increase of silicate weathering. Indeed, other large basaltic regions (e.g. Deccan, Parana) are also located in the tropical zone influenced by monsoon. This is likely a key for understanding past rapid climate change and the influence of tropical basins on the carbon cycle.

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#### 571 **6.** Conclusion

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573 This study examines the chemical composition of river water and sediments in the Ethiopian 574 Traps in order to characterize silicate weathering and corresponding CO<sub>2</sub> consumption in this 575 region. Lithium isotopic composition of the clay fractions was also examining and present a 576 positive correlation with clay Mg/Ti ratios, in relation with variable weathering conditions in 577 this region.

The average weathering rate (16.1 tons/km<sup>2</sup>/yr) and CO<sub>2</sub> consumption rate (0.65.10<sup>12</sup> mol/yr) determined for the Ethiopian Traps are lower than for other basaltic regions (e.g. Deccan or SE Asia). This is bet explained by the low runoff values observed in Ethiopia at present (242 mm/yr), despite regular occurrence of the monsoon. This demonstrates that, at present, the Ethiopian Traps weathering has a negligible impact on the carbon cycle, and thus on climate.

In the recent or the remote past, such as during the African Humid Period (from 14.500 to 8.000 years BP) or during the Oligocene (30 Ma) when this region was under equatorial latitudes, the runoff was increased to much higher values that modelers have estimated. Based on these results and weathering models, we find comparable results for CO<sub>2</sub> consumption rate for both periods. The determined CO<sub>2</sub> consumption rate for the African Humid Period range between 0.91 and  $1.5.10^{12}$  mol/yr. This indicates that during humid periods the Ethiopian traps could have played a significate role on carbon cycle and on climate.

590 Overall, this study brings out the key role of a region that is understudied thus far in terms of 591 weathering and climate paleo-variations. More generally, the influence on global climate of 592 tropical basins influenced by monsoon would deserve to be assessed more precisely, in 593 particular those of basaltic lithology.

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Fig 1: A. Map showing samples localization and the studied area. Sample name on the map are: 1:
Jara Wenz (ETH05-11); 2:Borkena Wenz (ETH05-13); 3: Robi Wenz (ETH05-4); 4: Meri Wenz
(ETH05-28); 5: Tikur Wenz (ETH05-19); 6: Ketchin Abeba Wenz (ETH05-25); 7: Gereb Ara Wenz
(ETH05-40); 8: Tekeze (ETH05-35); 9: Hormat Wenz (ETH05-47); 10: Ala-Wiha (ETH05-20); 11:
Nazero Wenz (ETH05-6) .B. Measured rainfall (mm/week) temporal variations in various places of

943 the Ethiopian Traps. Reported values corresponds to the average of rainfall between 1961 and 1997

944 for Gondar, Bahar Dar, Kombolcha and Addis Ababa, between 1991 and 1997 for Fiche and between

945 1994 and 1997 for Lalibellathe.





Fig 2: Mg/Na molar ratio as a function of Ca/Na molar ratio, for river dissolved phases. Both ratios are corrected from atmospheric inputs as described in section 4.2.1. The blue circles are for samples analyzed in this study. Orange circles are for other basaltic regions reported in Dessert et al., (2003).
Bedrock data come from (Pik et al., 1999). (Benedetti et al., 1994; Dessert et al., 2001; Gaillardet et al., 1999; Gislason et al., 1996; Leemans and Cramer, 1991; Louvat, 1997; Pascale Louvat and Allègre, 1998; Louvat and Allègre, 1997; Meybeck, 1986; Négrel et al., 1993; Négrel and Deschamps, 1996; Riotte et al., 2003)



Fig 3: Sr/Th as a function of U/Th (wt ratios) in the dissolved phases and solid phases of the sampled
rivers. Bedrock values come from Pik et al., 1999.



966 Fig 4: Clay  $\delta^7 Li$  as a function of clay Mg/Ti for the main Blue Nile Basin rivers (blue 967 diamonds). Green lines correspond to the result of the model described in section 5.1. for 968 different  $\beta$  values. The dotted line corresponds to the theoretical mixing curve between 969 bedrock and isotopically fractionated clay endmembers. Mixing model conditions: Bedrock 970  $\delta7Li = 5\%$ , [Li] = 10 ppm (this study); clays  $\delta^7Li = -2.3\%$  (clay  $\delta^7Li$  measured in the most 971 recent Nile delta fan sediments, [Li] = 100 ppm (Bastian, 2017; Bastian et al., 2017).

972 *River numbers are the same as in figure 1.* 



973

Fig 5: (A) TDS\* (Total Dissolved Solids corrected for atmospheric inputs) as a function of temperature. The blue diamond corresponds to the Ethiopian Traps (average based on 17 data, see Table 3), and the black circles come from the compilation of (Dessert et al., 2003) (Benedetti et al., 1994; Dessert et al., 2001; Gaillardet et al., 1999; Gislason et al., 1996; Leemans and Cramer, 1991; Louvat, 1997; Pascale Louvat and Allègre, 1997; Meybeck, 1986; Négrel et al., 1993; Négrel and Deschamps, 1996; Riotte et al., 2003) and corresponds to most other basaltic areas studied thus far. (B) Plot of chemical erosion rate as a function of runoff (average based on 17 data, see Table 3). The blue diamond reflects the Ethiopian Traps (this study) and the other symbols come from (Dessert et al., 2003) and display other basaltic basins with active zones (in black) and inactive zone (in white). The dashed lines correspond to the TDS\* concentrations.



Fig 6: Total CO<sub>2</sub> consumption in mol/year (Gaillardet et al., 1999) and annual CO<sub>2</sub>
consumption in GtC/yr due to silicate weathering for the largest rivers system. The blue bar
corresponds to the current CO<sub>2</sub> consumption rate estimated in Ethiopia based on our
measurements in river waters. The brown bar indicate the CO<sub>2</sub> consumption rate estimated
for the African Humid Period (AHP, see text for more details, and the grey one is for the
Oligocene and was published by (Lefebvre et al., 2013).

*Table 1: Characteristics of the studied watersheds.* 

991 Table 2: Chemical composition of river waters, Total Dissolved Solid (TDS), silicate
992 weathering rate and related CO<sub>2</sub> consumption rate for the major basins of the Ethiopian
993 Traps.

Table 3: Chemical composition of sand, mud and clay samples (clay fractions have been
physically separated following the procedure described in part 3.3.2)





*Fig S1: Ethiopian geological map. The black circles indicate our sampling sites.* 

<sup>1006 (</sup>https://esdac.jrc.ec.europa.eu/images/Eudasm/Africa/images/maps/download/afr\_etgm.jpg)
1007

Sample number	Sample name	Elevation (m)	Mean elevation (m)	Area (km²)	Annual discharge (m <sup>3</sup> /year)	calculated runoff (mm/an)
ETH04-2	Nil (Juin-04)	1158	2277	64214	1.8E+10	273
ETH 05 -52	Nil (Mars-05)	1158	2277	64214	1.8E+10	273
ETH04-9	Jemma Juin-04)	1290	2516	5878	1.4E+09	232
ETH05-51	Jemma (Mars-05)	1290	2516	5878	1.4E+09	232
ETH 05 -50	Wench't Wenz	1384	2520	2853	7.2E+08	252
ETH 05 -13	Borkena Wenz	1948	2658	24	6.5E+06	268
ETH 05 -19	Tikur Wiha (Woldia)	1729	2500	171	3.9E+07	227
ETH04-4	Wegel Tena	1651	2625	945	2.2E+08	234
ETH04-5	Bashillo	1649	2719	2668	7.1E+08	266
ETH05-40	Gereb Ara Wenz	1496	2243	58	1.1E+07	190
ETH05-20	Ala Wiha (Godg Wabit)	2028	2874	75	1.6E+07	218
ETH05-47	Hormat Wenz	1523	2346	134	2.7E+07	200
ETH 05 -25	Ketchin Abeba Wenz	1998	2848	220	4.8E+07	220
ETH 05 -28	Meri Wenz	2005	2591	131	2.6E+07	198
ETH 05 -35	Tekeze amont	2133	3020	493	1.1E+08	219
ETH04-10	plateau-N-Addis	2486	2697	56	1.5E+07	265
ETH04-8	Plateau E-Addis	2880	3267	32	8.5E+06	267
ETH 05 -2	Ulet Wenz	3034	3250	4	1.1E+06	270
ETH 05 -4	Robi Wenz	1389	2071	219	5.5E+07	252
ETH 05 -11	Jara Wenz	1656	2575	77	2.0E+07	265
ETH 05 -6	Nazero Wenz	1422	1879	171	4.4E+07	259

Table 1: Characteristics of the studied watersheds.

I able 2: Chemical composition of river waters, I otal Dissolved Solid (IDS), silicate weathering rate and	omposition of river waters. Total Dissolved Solid (TDS), silicate weathering rate and	1 C
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Sample number	Sample name	Si (µmol/l)	Mg (µmol/l)	Ca (µmol/l)	Na (µmol/l)	K (µmol/l)	Al (µmol/l)	Sr (µg/l)	Th (µg/l)	U (µg/l)	F <sup>-</sup> µmol/l	Cl <sup>-</sup> µmol/l	NO <sub>3</sub> <sup>-</sup> µmol/l	SO4 <sup>2-</sup> µmol/l	Mg* (µmol/l)	Ca* (µmol/l)	Na* (µmol/l)	K* (µmol/l)	F⁻* µmol/l	NO <sub>3</sub> <sup>-</sup> * µmol/l	SO4 <sup>2-</sup> * µmol/l	HCO <sub>3</sub> <sup>-</sup> * µmol/l	TDS* (µmol/l)	TDS* (mg/l)	Total chemical weathering tons/km <sup>2</sup> /year	CO2 10 <sup>6</sup> mol/km2/year
ETH04-2	Nil (Juin-04)	142	304	530	494	70	3405	135	0.005	0.1	8.5	76	43	146	296	529	427	68	8.5	43	146	1808	1320	40.9	11.2	0.49
ETH 05 -52	Nil (Mars-05)	154	341	584	414	61	398	174	< L.D.	0.2	8.9	70	4	171	334	582	352	60	8.9	4	171	1897	1328	41.9	11.4	0.52
ETH04-9	Jemma Juin-04)	332	323	688	650	141	3946	162	0.011	0.6	12.2	116	100	252	311	686	548	139	12.2	100	252	2075	1683	53.1	12.4	0.48
ETH05-51	Jemma (Mars-05)	660	684	1173	959	98	93	371	< L.D.	0.5	17.7	127	14	269	671	1171	847	96	17.7	14	269	4070	2785	86.5	20.1	0.95
ETH 05 -50	Wench't Wenz	631	585	1035	1219	92	264	257	< L.D.	0.6	14.2	165	8	402	568	1032	1073	89	14.2	8	402	3551	2762	83.3	21.0	0.90
ETH 05 -13	Borkena Wenz	465	499	1334	734	59	26	265	< L.D.	0.3	8.5	345	292	125	464	1327	429	52	8.5	292	125	3546	2273	76.4	20.5	0.95
ETH 05 -19	Tikur Wiha (Woldia)	667	789	1110	688	70	198	237	< L.D.	0.9	9.7	138	115	98	775	1107	566	68	9.7	115	98	4089	2516	78.9	17.9	0.93
ETH04-4	Wegel Tena	639	775	1141	901	65	3625	236	0.006	0.8	9.1	273	77	272	748	1135	659	60	9.1	77	272	3880	2602	81.2	19.0	0.91
ETH04-5	Bashillo	498	663	1022	940	60	4823	227	0.007	0.7	10.1	164	16	182	647	1019	795	57	10.1	16	182	3809	2518	77.1	20.5	1.02
ETH 05 -25	Ketchin Abeba Wenz	526	1565	1357	1055	71	62	488	< L.D.	0.9	8.8	348	132	212	1530	1350	748	65	8.8	132	212	6039	3692	111.0	24.4	1.33
ETH 05 -28	Meri Wenz	473	1365	1386	869	69	458	672	< L.D.	0.5	9.2	184	11	880	1346	1382	706	65	9.2	11	880	4466	3500	106.9	21.2	0.89
ETH 05 -35	Tekeze amont	453	1768	1170	761	48	101	360	< L.D.	1.2	7.3	298	129	205	1739	1164	497	43	7.3	129	205	5826	3442	102.0	22.4	1.28
ETH04-10	plateau-N-Addis	197	169	304	273	56	443	115	< L.D.	0.0	7.7	52	154	22	164	303	228	55	7.7	154	22	1014	750	23.5	6.2	0.27
ETH04-8	Plateau E-Addis	153	88	160	147	51	3359	53	0.006	0.0	6.4	54	21	15	83	159	99	50	6.4	21	15	582	391	12.6	3.4	0.16
ETH 05 -2	Ulet Wenz	199	59	133	102	20	2225	44	0.002	0.0	5.1	15	26	27	57	133	89	20	5.1	26	27	406	299	9.5	2.6	0.11
ETH 05 -4	Robi Wenz	539	250	721	734	73	307	114	< L.D.	0.4	12.2	56	59	70	244	720	684	72	12.2	59	70	2478	1720	53.3	13.4	0.62
ETH 05 -6	Nazero Wenz	877	581	1161	1155	100	68	174	0.002	0.5	21.3	168	8	90	565	1158	1007	97	21.3	8	90	4354	2826	87.1	22.5	1.13

 $CO_2$  consumption rate for the major basins of the Ethiopian Traps

Sampla	Samala							Sand												Mud											Clay				
Sample	Sample	SiO <sub>2</sub>	$AI_2O_3$	Fe <sub>2</sub> O <sub>3</sub>	MnO	MgO	CaO	Na <sub>2</sub> O	K2O	TiO <sub>2</sub>	Sr	Th	U	SiO <sub>2</sub>	$AI_2O_3$	$Fe_2O_3$	MnO	MgO	CaO	Na <sub>2</sub> O	K2O	TiO <sub>2</sub>	Sr	Th	U	δ7Li	K2O	CaO	MgO	Fe2O3	AI2O3	TiO2	Sr	Ba	Mn
number	name	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(ppm)	(ppm)	(ppm)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(ppm)	(ppm)	(ppm)	(‰)	(%)	(%)	(%)	(%)	(%)	(%)	(ppm)	(ppm)	(ppm)
ETH04-2	Nil (June-04)	50.3	12.6	12.7	0.2	4.4	8.5	1.9	1.0	2.7	361.9	3.2	1.0	41.1	21.1	14.4	0.2	1.5	1.4	0.3	0.8	2.2	48.7	3.4	1.0	1.2	0.7	1.3	1.4	12.8	19.5	2.0			1448.0
ETH 05 -52	Nil (March-05)																																		
ETH04-9	Jemma June-04)	78.2	6.8	5.3	0.1	1.4	3.1	1.5	1.1	1.3	156.7	4.0	1.0																						
ETH05-51	Jemma (March-05)																																		
ETH 05 -50	Wench't Wenz	56.6	13.6	9.2	0.2	3.3	5.0	2.5	2.0	1.8	362.9	6.7	1.5																						
ETH 05 -13	Borkena Wenz	45.3	16.3	12.4	0.2	2.7	5.7	2.3	0.8	2.5				45.4	16.2	10.9	0.2	2.4	4.2	1.4	0.9	2.1	279.0	3.9	0.9	2.3	0.4	3.1	1.9	6.2	11.7	0.7	156.3	195.7	1100.2
ETH 05 -19	Tikur Wiha (Woldia)	52.0	11.5	10.8	0.2	5.8	7.8	2.5	2.0	3.2				44.7	13.2	11.8	0.1	4.8	3.0	0.7	1.3	2.6	256.8	2.9	0.7	1.0	0.6	2.9	3.7	6.9	9.7	0.9	148.0	182.1	669.1
ETH04-4	Wegel Tena	52.3	11.3	11.1	0.2	6.3	8.6	2.6	1.8	3.2	436.8	5.8	1.7																						
ETH04-5	Bashillo	47.6	11.2	13.6	0.2	6.6	10.0	2.1	1.2	3.9	429.9	3.5	1.0	43.0	12.8	13.1	0.2	5.5	5.2	1.2	1.2	3.5													
ETH05-40	Gereb Ara Wenz	46.9	10.5	12.6	0.2	7.1	9.3	2.4	1.7	4.7				42.8	10.8	12.6	0.2	7.2	5.2	1.2	1.3	4.0	314.6	5.1	1.5	2.2	0.6	3.8	6.0	7.0	7.3	0.9	225.8	174.3	778.3
ETH05-20	Ala Wiha (Godg Wabit)	43.8	7.7	13.6	0.2	12.8	10.7	1.1	0.8	3.9				41.4	7.2	15.1	0.2	12.2	10.1	0.7	0.7	5.1	175.3	7.0	1.5	3.2	0.4	3.7	7.9	6.7	6.5	0.9	281.9	128.8	821.4
ETH05-47	Hormat Wenz	44.4	7.1	13.6	0.2	13.0	11.6	1.3	0.9	4.2				40.7	9.3	14.7	0.2	9.0	6.3	1.0	1.0	5.4	331.5	4.1	1.1	4.0	0.4	3.7	7.3	7.0	7.2	0.9	283.5	150.0	821.2
ETH 05 -25	Ketchin Abeba Wenz	45.1	9.9	13.8	0.2	8.9	8.9	2.0	1.4	4.8				41.1	11.9	14.4	0.2	6.6	4.5	0.9	1.1	4.4	361.0	5.7	1.6	1.8	0.5	2.8	4.9	7.2	9.8	0.9	237.2	182.3	1020.9
ETH 05 -28	Meri Wenz	45.5	9.9	14.9	0.2	8.1	8.7	2.1	1.5	5.7				42.3	13.2	13.7	0.2	5.3	4.4	1.6	1.5	4.1	404.7	6.1	1.9	0.3	0.7	3.0	4.3	7.7	10.3	1.3	259.9	284.1	1411.5
ETH 05 -35	Tekeze amont	44.9	8.8	13.5	0.2	12.1	10.9	1.7	1.0	4.5				41.0	11.4	13.3	0.2	7.8	2.9	0.6	1.0	3.0	197.1	5.2	1.2	3.2	0.4	2.2	6.2	7.4	8.3	0.8	158.2	152.3	1050.4
ETH04-10	plateau-N-Addis	46.9	15.5	13.9	0.2	4.3	7.9	2.6	1.4	2.8	571.3	4.6	1.0																						
ETH04-8	Plateau E-Addis																						157.0	7.8	2.2										
ETH 05 -2	Ulet Wenz	38.7	20.2	14.8	0.4	1.5	0.9	0.4	0.9	3.5	112.5	7.9	2.4																						
ETH 05 -4	Robi Wenz	56.9	13.7	9.0	0.2	2.1	4.7	3.0	2.4	2.1				49.3	14.7	11.2	0.2	2.2	4.2	1.9	1.5	2.6	337.0	8.1	1.7	1.2	0.6	3.2	3.5	7.3	10.0	1.1	230.2	211.0	1013.9
ETH 05 -11	Jara Wenz	66.0	13.0	5.7	0.1	1.6	2.7	3.6	3.5	1.0				46.0	14.4	11.3	0.2	4.6	5.0	1.7	1.2	2.0	367.1	4.6	0.9	0.4	0.6	3.0	1.4	5.8	7.4	0.9	187.1	193.9	1019.6
ETH 05 -6	Nazero Wenz	72.4	11.7	4.0	0.1	0.5	1.0	3.6	4.2	0.5	79.4	14.9	2.7													3.6	0.3	3.0	1.9	2.6	3.7	0.2	232.0	141.0	482.0

Table 3: Chemical composition of sand, mud and clay samples (clay fractions have been separated using physical decantation following the predure described in part 3.3.2)