

Chemical erosion rates in the upper Blue Nile Basin and related atmospheric CO2 consumption

Luc Bastian, Nathalie Vigier, Marie Revel, Gezahegn Yirgu, Dereje Ayalew, Raphaël Pik

▶ To cite this version:

Luc Bastian, Nathalie Vigier, Marie Revel, Gezahegn Yirgu, Dereje Ayalew, et al.. Chemical erosion rates in the upper Blue Nile Basin and related atmospheric CO2 consumption. Chemical Geology, 2019, 518, pp.19-31. 10.1016/j.chemgeo.2019.03.033 . hal-02134565

HAL Id: hal-02134565 https://hal.science/hal-02134565v1

Submitted on 22 Oct 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



2

Chemical erosion rates in the upper Blue Nile Basin and related atmospheric CO₂ consumption

3	
4	Luc Bastian ^{1,2} , Nathalie Vigier ² , Marie Revel ¹ , Gezahegn Yirgu ³ , Dereje Ayalew ³ , Raphaël
5	Pik^4
6	
7	¹ Geoazur, Université de la Cote d'Azur, CNRS, OCA, IRD, Geoazur, 250 rue Albert
8	Einstein, 06500 Valbonne, France
9	² Laboratoire d'Océanographique de Villefranche-sur-Mer (LOV), CNRS, Sorbonne
10	Université, 181 chemin du Lazaret, 06230 Villefranche-sur-Mer, France
11	³ School of Earth Sciences, Addis Ababa University, Addis Ababa, Ethiopia
12	⁴ Centre de Recherche Pétrographique et Géochimiques (CRPG), UMR 7358 CNRS-
13	Université de Lorraine, 15 rue Notre Dame des Pauvres, BP20, Vandoeuvre-les-Nancy,
14	Cedex, France
15	
16	
17	*corresponding author: bastian@geoazur.unice.fr
18	
19	

Abstract

2021

22

23

24

25

26

27

28

29

30

31

32

33

34

35

36

37

38

39

40

41

42

43

44

45

46

Silicate weathering of basaltic rocks constitutes a non-negligible sink of atmospheric CO₂ but the role it plays in the regulation of past and future global climate is still matter of debate. In this study, silicate weathering rates for various sub-basins of the Ethiopian Traps, emplaced 30 million years ago, and the corresponding atmospheric CO₂ consumption rates are evaluated. For this, major and trace elements were measured in the dissolved phases and in the sedimentary particles carried and deposited by the main rivers flowing through this steep region. Lithium isotopes and major elements were also measured in the extracted clay fractions in order to infer complementary information on weathering processes in this region. Clay δ^7 Li values correlate positively with Mg/Ti ratios, and are best explained by varying ratios of leaching versus clay formation rate. Although located in a region annually submitted to monsoon, average silicate weathering rate (16.1 tons/km²/year) and CO₂ consumption rate (0.65x10¹² mol/year) are estimated to be low 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 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 monitoring station data, we show that runoff intensity is a key parameter that explains this difference. We determine that, at present, the weathering of the Ethiopian Traps plays a 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 African Humid Period (14-8 kyr), a significant increase of Monsoon precipitation may have resulted in much higher weathering rates and related CO₂ consumption (0.91-1.5x10¹² mol/year). This study therefore evidences the potential importance of this region in the past, and the need to quantify more precisely the variations of the monsoon intensity and its impact on tropical watersheds for reconstructing past CO₂ levels.

47 48

Key words

50 51

49

Ethiopian Traps, silicate weathering rate, lithium isotopes, CO₂ consumption, basalt, African monsoon

53

1. Introduction

54 55

56

57

58

59

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85

86

87

Chemical and physical erosion, which describe respectively mineral dissolution and soil formation, rock cracking and particle transport are key processes in landscape evolution. Various parameters can control weathering (chemical erosion) at a watershed scale, among 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 al., 2001; Gaillardet et al., 1999; Li et al., 2016; P Louvat and Allègre, 1998; West et al., 2005). Over geological timescale (>10⁵ years), silicate weathering, with organic matter burial, are major sinks of atmospheric carbon. Weathering of silicate rocks consumes atmospheric CO₂ and transforms it to dissolved HCO₃⁻ (Gaillardet et al., 1999), which further precipitates with calcium carbonate in the ocean. As CO₂ is a greenhouse gas, silicate rock weathering can 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 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 this is not consensual thus far. Other mountainous regions may have been of importance, in particular those rich in basaltic lithologies, and are little considered yet (Li and Elderfield, 2013). Indeed, all silicate rocks do not present the same sensitivity to weathering. Among 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, 1997; P Louvat and Allègre, 1998) indicate that basaltic rocks are weathered 8 time faster 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 estimated as an important sink of CO₂ at the global scale (Dessert et al., 2003) and it is calculated to be responsible of 30% of the present-day CO₂ consumption (Gaillardet et al., 1999). The Deccan Traps were investigated in great details (e.g. Das et al., 2005; Dessert et 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 not been studied yet. For example, weathering of the Ethiopian Traps (upper Blue Nile Basin) is not well constrained, despite the importance of this region in terms of climate, size, tectonics and age (30 Ma). A recent carbon-climate modeling study highlights that the Ethiopian Traps were the world-wide most important hotspot of silicate weathering during the 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 ⁶Li (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 δ^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; 120

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 sediments deposited in terraces, lake or delta (Bastian et al., 2017; Dosseto et al., 2015) to trace past variations of silicate weathering has also been demonstrated (Bastian et al., 2017; Dosseto et al., 2015; Lechler et al., 2015). By studying the Nile Delta during the last 32kyr, Bastian et al (2017) show that clays deposited by the Nile River were mostly smectite formed in the Ethiopian Traps. Their δ^7 Li values co-vary with climate proxies, and in particular with precipitation variations. The controls of lithium isotope composition for clays formed in the Ethiopian Traps are therefore important to investigate further, and should help to refine our understanding of the relationships between climate and basalt weathering. In this study, we target to provide chemical erosion rates and related CO₂ 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 CO₂ consumption rate during an intensive runoff period, well-known and well evidenced in Nile delta sedimentary cores, the African Humid Period (14.5-8kyr).

2. Study area

The Ethiopian Continental Flood Basalt (CFB) province is located in the North and West Ethiopian highlands (Fig 1 & Fig S1). It covers a surface of about 800,000 km² and has an estimated volume ranging between 1 and 1.5 million km³ (Rochette et al., 1998). They erupted 30 Ma ago over a short time period, 1-2 Ma (Hofmann et al., 1998). The lave pile thickness varies from 500 to 2000 m with maximum thickness of 3000 m in the northern part of the plateau (Mohr. P, 1988). This CFB province is mainly composed by transitional basalts 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). 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 elevation up to 4600 meters. The Ethiopian Highlands have been uplifted since at least 25 Ma (Pik et al., 2003) with concomitant development of the two main basins of the Nile and Tekeze rivers. The plateau presents important border escarpments with steep slopes, especially along the roughly N-S marginal graben on its Eastern side (Fig. 1). These highlands have been dissected by rapid development and propagation of the Nile and Tekeze basins

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 around 21°C. Thus, 90% of the annual precipitations occur during the summer month (Korecha and Barnston, 2007) (Fig 1). Precipitations display an East-West gradient in Ethiopia: rainfall increase from 200 mm/year on the Indian Ocean to 1800 mm/year in the Highlands. At present, the Blue Nile River (major affluent of the Nile River) and its tributaries drain the Ethiopian Traps. Thus, this high topography region is estimated to contribute to 60 % of the total water discharge of the main Nile and to 60 % of its terrigenous 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

lead to stronger precipitation every 21 kyrs. These strong runoff variations are recorded by

Ethiopian lake sediments (Amaral et al., 2013) and by the sediments transported by the Nile

(Shanahan et al., 2015), in relation with the precession parameter (Tuenter et al., 2003) which

River and deposited in its delta (Blanchet et al., 2014; Castañeda et al., 2016; Revel et al.,

174 2015, 2010).

3. Sampling and methods

3.1. Sampling

All samples come from the northern Ethiopian plateau (Fig 1), which encompasses the large catchment of the Blue Nile (64 000 km²), several smaller sub-basins (1000 to 6000 km²), as well as small catchments emplaced directly on the top of the plateau (5 to 100 km²). Two 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 filtered on the field with 0.2µm filters and immediately acidified with purified nitric acid. Sands and muds were sampled on the river banks after mixing 5 different aliquots of about 100g each. The difference between mud and sand is the visible granulometry: sand samples correspond to coarse sized mineral (> 2mm grain) rich-particles and the mud samples to the

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

191 192

3.2. Runoff Modeling

193 194

195

196

197

198

199

200

201

202

203

The hydrology of the Blue Nile upper basins is strongly controlled by seasonal precipitations, with strong rains and flooding observed during the summer monsoon, from June to September (e.g. Conway, 2009). These conditions are not appropriate for providing a reliable estimate of 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 calculations, we used a numerical model based on the IDRISI © GIS software. This model was run using the plateau topography and available public rainfall data. In order to avoid 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 calibrated discharge and runoff data for each sub-basin are presented in Table 1.

204

205 3.3. Analytical protocol

206

207 3.3.1. Chemical analyses

208

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

215

3.3.2. Lithium isotope analyses of the clay fractions

217

216

Clays were extracted from the mud samples, by decantation of the sediments in 50ml of ultrapure water mixed with 20µl 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/HNO₃/HCl solution in a clean laboratory at LOV, using 223 ultrapure acids. The solution was evaporated and the residue dissolved in 1.00N HCl. The 224 total dissolution of samples was systematically verified by the absence of residue after 20 min 225 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 ≈60ng 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-collector inductively coupled plasma spectrometer (MS-ICP-MS) at the Ecole Nationale Supérieur de Lyon (CNRS National Facilities). Sample was introduced through an Aridus II desolvating system before ionization to increase sensitivity (1V 7 Li / ppb Li). We used the jet and X cones, as described in (Balter and Vigier, 2014). To correct for instrumental mass bias, we used the standard bracketing technique, using the LSVEC standard. The total blank procedure is systematically lower than 10pg Li. The accuracy of isotopic analysis was verified 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 δ^7 Li value 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 δ^7 Li values are 30.3±0.4% (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 compare well with published values (Carignan et al., 2007; Millot et al., 2004).

4. Results

4.1. Runoff and discharge

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 m³/yr (for the Ulet Wenz River) to 1.8_x10^{10} m³/yr (for the Blue Nile river), with a mean value of 19_x10^8 m³/yr (Table 1).

- For the Blue Nile river, the calculated discharge is close to the annual discharge measured at
- 258 the Roseires Dam Station (http://www.grdc.sr.unh.edu/html/Polygons/P1663800.html) :
- $4.7_x 10^{10}$ m³/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
- 263 Traps (463 mm/year). It is also lower than the one determined for the Siberian Traps (324
- 264 mm/yr). Compared to small tropical basaltic areas such as Hawaii (1612 mm/yr) and the
- Reunion Island (2433 mm/yr), the Ethiopian runoff is also low. This is partly due to their
- topography, their small surface area compared to the Ethiopian Traps (800,000 km² vs 16760
- 267 km² in Hawaii and 2512 km² in the Reunion Island), and the occurrence of frequent
- 268 hurricanes and tropical storms. In Iceland, high runoff (1883-2432 mm/yr) values are mostly
- due to ice melting, which introduces water in the riverine system during the summer season
- 270 (Gislason et al., 2009).
- 271
- 4.2. Major and trace elements
- 273
- 274 4.2.1 River waters
- 275
- In waters, the dominant cations are, in increasing order of concentrations: Si, Mg, Na, and Ca,
- 277 with mean values of 447 (\pm 219), 1768 (\pm 502), 712 (\pm 332) and 883 (\pm 417) μ mol/I
- 278 respectively. These concentrations are corrected from the atmospheric input using Cl
- 279 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 (± 415), 68
- 281 (± 26) and 574 (± 285) μ mol/l respectively.
- On average, molar Ca*/Na* and Mg*/Na* ratio are 2.5 (± 0.9) and 0.4 (± 0.6), respectively
- 283 (Table 2). For comparison, average Ca/Na and Mg/Nain Ethiopian basalts are 3.9 (± 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).
- 285 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%)
- 288 compared to basaltic rocks.
- The dominant anions are, in increasing order of concentrations: HCO₃⁻ SO₄²-, Cl⁻, NO₃⁻, and
- 290 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).

4.2.2 River bed particles

Sands are enriched in SiO₂ compared to mud samples (Table 3), $(51.9 \pm 10.6 \%)$ on average vs $43.2 \pm 2.6 \%$ for muds) and Na₂O (2.2 ± 0.8 % vs 1.1 ± 0.5 %), while depleted in Al₂O₃ (11.7 $\pm 3.4 \%$ vs 13.0 $\pm 3.5 \%$ in mud), CaO (7.0 $\pm 3.4 \%$ vs 4.7 $\pm 2.1 \%$ in mud) and Fe₂O₃ (11.4 \pm $3.4 \% \text{ vs } 13.5 \pm 1.5\%$). TiO₂ and MgO (5.7 ± 4.0 % vs 5.8 ± 3.1%) concentrations are similar between sands and muds: ($\approx 3.1 \pm 1.4$ wt % and $\approx 5.8 \pm 3.1$ wt % respectively). Si/Al ratio is lower by ≈30% in muds compared to sands, which is consistent with occurrence of aluminosilicate minerals (clays/smectite) in the collected muds. The chemical composition of <2µm clay fractions extracted from mud samples, contains, on average, 24.2% (± 4.7 %)</p> Al_2O_3 , 22.2% (± 1.8 %) Fe_2O_3 , and 15.1% (± 6.9 %) MgO. We note that the mud samples 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)

were preferentially used for assessing weathering processes in this region.
Some trace elements measured in the solid phases can be used to study chemical erosion

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 Sr/Th versus U/Th (Fig 3, Table 2 and 3), when compared to bedrocks, Ethiopian river waters display elevated U/Th and Sr/Th ratios (respectively >2 and > 8405) while the solid phases display low U/Th and Sr/Th values (respectively <0.3 and <87.8). Sr and U are two mobile elements during water-rock interactions and are usually depleted in the solid phase, in contrast with Th, which is an immobile element that can easily adsorb, except in organic-rich environments (Dupré et al., 1999). Figure 3 also indicates that the river particles are mostly derived from basaltic rocks, as previously described, and that the influence from rhyolite weathering is minimal, except for one sample (Nazero Wenz), which is located in a rhyolite rich area (Figure S1).

4.3 Lithium isotope compositions of clay size (<2µm) fractions

Ethiopian bedrock samples exhibit a mean δ^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 %o, (Genske et al., 2014)) clays deposited by Ethiopian rivers are significantly enriched in ⁶Li: clay δ^7 Li values range from 0.3% (± 0.5%) to 4% (± 0.5%), with a mean value of 2.0% ($\pm 1.2\%$). 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 δ^7 Li values is observed in suspended particles (SPM) of rivers draining basaltic regions e.g. in Iceland (with values from -1.3 to 7.5%, (von Strandmann et al., 2006)) and in the Azores archipelago (3.5 to 9.5%, (von Strandmann et al., 2010)). Also in soils, the range of published δ^7 Li for bulk soils samples is large (-15 to 10%, (Clergue et al., 2015; Pistiner and Henderson, 2003; Ryu et al., 2014; Schmitt et al., 2012; von 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 endmember (Dellinger et al., 2014). Here, clays were extracted from river sediments in order to minimize the influence of primary minerals.

5. Discussion

5.1. Assessing weathering processes at the basin scale

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

secondary phases (F_{lea}^{Li}/F_{sp}^{Li}). In rivers currently draining the Ethiopian Traps, clay fractions display variable δ^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 δ^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⁺ essentially substitutes to Mg²⁺ 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 δ^7 Li values have been shown to be assertially explained by the ratio of leaching varsus aloy formation rates, and thus reflect

Using a simple modeling, based on mass balance, clay δ' Li values have been shown to be essentially explained by the ratio of leaching versus clay formation rates, and thus reflect different weathering intensities (Bastian et al., 2017; Bouchez et al., 2013; Vigier et al., 2009; 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, considering a simple mass balance between bedrock, rivers and soils at the watershed scale. It is defined such that all the lithium released by rock dissolution is either incorporated into secondary phases formed in weathering profiles or released into superficial waters (and *in fine* in rivers).

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

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. δ_{lea} , δ_{sp} , δ_{riv} are the Li isotopic signature (δ^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 Δ_{land} . After simplification, the isotope composition of the secondary phases is given by:

386
$$\delta_{\rm sp} = \frac{\Delta_{\rm land}}{F_{\it lea}^{\it Li}/F_{\it sp}^{\it Li}} + \delta_{\rm BedRock} - \Delta_{\rm land} \qquad \text{Equation (2)}$$

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 to F_{lea}^{Li} , then $\delta_{sp} = \Delta_{land}$.

391

392

393

394

395

396

The positive correlation between clay $\delta^7 \text{Li}$ 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 $\delta^7 \text{Li}$, 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_{rlea} = 0). Thus Mg/Ti ratio of secondary phases can be described as:

397

398
$$({^{Mg}/_{Ti}})_{sp} = \frac{1}{F_{lea}^{Mg}/_{F_{sp}^{Mg}} \cdot T_{leav}^{mean}/_{Mg_{BedRock}}}$$
 Equation (3)

399

400

The relation between the Li and the Mg flux becomes:

401

412

413

414

415

417

418

419

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

with β 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 δ^7 Li and low Mg/Ti values,

405 (ie Jara Wenz (ETH05-11), Borkena Wenz (ETH05-13), Robi Wenz (ETH05-4), Meri Wenz

406 (ETH05-28), Tikur Wenz (ETH05-19)). For these small basins, the weathering model

407 indicates that soil formation rate (F_{sp}) is low compared to the leaching rate (F_{lea}) . Other areas

display high clay δ^7 Li values associated to high Mg/Ti ratios (Ketching Abeba Wenz

409 (ETH05-25), Gereb Ara Wenz (ETH05-40), Tekeze (ETH0535), Hormat Wenz (ETH05-47),

410 Ala-Wiha (ETH05-20), Nazero Wenz (ETH05-6)).

Following our modeling, the various δ^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 (β parameter). For a

large part, the β 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 δ^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 $\delta^7 \text{Li}$ 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.

425

426

420

421

422

423

424

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

427428

429

430

431

432

433

434

435

436

437

438

439

440

441

442

443

444

445

446

447

448

449

450

451

452

453

As discussed above, the relatively large range of clay Mg/Ti and δ^7 Li reflects a significant variation of weathering conditions in the Ethiopian Traps region while the lithology is dominantly basaltic. It is possible to use the chemical composition of river waters in order to quantify present-day weathering rates (Table 2) at the scale of each corresponding watershed (e.g. Gaillardet et al., 1999; Stallard and Edmond, 1983). The Total Dissolved Solid (TDS) is defined by the sum of major mobile cations (Ca, Mg, K and Na) concentrations in the dissolved phases of rivers. These concentrations are corrected from the atmospheric input using Cl concentration and oceanic X/Cl molar ratios, following the method detailed in (Das et al., 2005). TDS* values (* indicate corrected from atmospheric inputs) estimated from 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 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 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), which are also characterized by elevated annual temperature (27°C and 24.8 °C respectively). These three regions display river TDS* values significantly greater than in Hawaii (7.4 mg/l) 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 parameter which controls river TDS* (Dessert et al., 2003; Li et al., 2016). 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/km²/year, with a mean value of 16.1 tons/km²/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/km²/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/km²/year, 48.4 tons/km²/year 456 for La Reunion, and 152 tons/km²/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, 458 459 other parameters may also play a role, in particular temperatures, the age of the basaltic rocks (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). 463 Overall, both river particles and water composition can provide valuable information on 464 chemical erosion rate in the various basins. However here there is no correlation between 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

475

476

5.3. CO₂ consumption and SO₄²⁻ concentration due to silicate weathering

477478

479

480

481

482

483

484

485

From river water chemical compositions, it is possible to estimate the CO₂ consumption rates related to silicate weathering. In the literature, several methods have been used, using corrections from source rocks in mixed lithology basins, calcium concentration in silicate rivers (Goddéris et al., 2013), or HCO₃⁻ concentrations in carbonate-free watersheds (e.g. Das et al., 2005). Since the Ethiopian Traps are essentially constituted of silicate rocks, with little vegetated areas, major cations concentrations in rivers have only been corrected for atmospheric inputs, as described in section 5.1. HCO₃⁻ concentrations are determined using the following electronic mass balance:

486

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

489

490

491

492

493

494

495

496

497

498

499

500

501

502

503

504

505

506

507

508

509

510

511

512

513

514

515

516

517

518

519

520

where all concentrations are corrected for atmospheric inputs (see section 4.2.1). The average pH of the Ethiopian rivers (7.5-8.2, (MERID, 2005)) makes it to possible to assume that HCO₃⁻ is the dominant anion compared to CO₃². Thus, in order to determine the CO₂ consumption related to silicate weathering, we consider that 1 mole of HCO₃- exported by rivers corresponds to 1 mole of consumed atmospheric CO₂. CO₂ consumption rates are then calculated using the river discharge and the basin surface area (for specific rates). Some recent studies (Calmels et al., 2007; Li et al., 2008; Torres et al., 2016) show that sulfate mineral dissolution produces sulfuric acid that can enhance bedrock and mineral dissolution, in particular in sedimentary or carbonate rich context. Any silicate weathering due to sulfuric acid may lead to an overestimation of the CO₂ consumption rate if this effect is not considered into the calculations. In the rivers studied here, dissolved sulfate concentrations do not 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 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 amounts of nitrate in waters (see section 4.2.1). The use of Equation (5) to calculate HCO₃⁻ allows us to avoid a potential bias due to sulfuric acid in the determination of CO₂ consumption rate. Table 2 show that specific CO₂ consumption rate due to basalt dissolution in Ethiopian Traps vary from $0.11x10^6$ mol/km²/yr (for the Ulet-Wenz basin) to 1.33×10^6 mol/km²/yr (for the Ketchiin Abeba Wenz), and is $0.76 \times 10^6 \frac{\text{mol/km}^2}{\text{yr}}$ on average. The CO₂ consumption rate cover a large range, possible due to local – small scale - effects. Thus, for the comparison with literature data, we use the two values estimated for the Blue Nile River, which integrates most waters draining the Ethiopian Traps. The Blue Nile River was sampled at two different seasons (ETH04-2; Juin-04 and ETH05-52; Mars -05). The two samples lead to similar CO₂ consumption rate, of 0.49 and 0.52 mol/km²/yr respectively, but are low compared to other basaltic regions. For comparison, this flux is estimated to be 1.26 10⁶ mol/km²/yr for the Deccan Traps and 6.41 10⁶ mol/km²/yr for Java Island (Dessert et al., 2003). The CO₂ consumption rate due to silicate weathering in the Blue Nile basin is slightly higher, than the value published by (Dessert et al., 2003) for the Ethiopian Traps, i.e. $0.15x10^6$ mol/km²/yr. When using the Dessert et al., (2003) theoretical equation $fCO_2 = R_{f x}323.44 \exp(0.0642 \text{ T})$ where fCO₂, R_f and T are respectively the specific atmospheric CO₂ consumption

consumption rate is also slightly lower (0.16x10⁶ mol/km²/year) that our calculated value. 522 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 CO₂ consumption flux (in mol/year). The average CO₂ 524 consumption rate is calculated to be 0.65×10^{12} mol/year. This is significantly higher than in 525 Iceland (0.070 x10¹² mol/year), the Reunion Island (0.006 x10¹² mol/year) or the Azores 526 (0.0004 x10¹² mol/year) (see Fig. 6). However, this value remains lower than for the Deccan 527 (0.392 x10¹² mol/year) or than SE Asia (1.033 x10¹² mol/year). The upper Blue Nile represent 528 a CO₂ consumption close to the global average ((Gaillardet et al., 1999), see Fig. 6), and only 529 7.5 % of the total CO₂ consumed by silicate weathering on Earth. It corresponds to 7.8.10³Gt 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 CO₂ 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

(mol/km²/year), the runoff (mm/year) and the temperature (°C), the corresponding

LMDZ and ORCHIDEE models, determined an increase of precipitation in the Ethiopian Traps of 1.4 to 2.5 times larger during the AHP ((Marzin and Braconnot, 2009); Vadsaria and Ramstein, Pers. Comm.). This corresponds to a runoff ranging between 340 and 600 mm/years. By applying this factor, CO₂ consumption rate during African Humid Period may thus have increased up to values of 0.91-1.5x10¹² mol/yr (brown bar in Fig. 6), which corresponds to the most elevated current rates. These estimations are in line with the study of (Lefebvre 2013), which showed that the importance of this region in the carbon cycle during the Oligocene period. However, it is important to note that during the Oligocene, the global temperature was also higher, while for the AHP, the runoff increase in North Africa is likely not related to a significant increase of temperature (De Boer et al., 2013).

These results also suggest a potential increase of global CO₂ 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.

6. Conclusion

This study examines the chemical composition of river water and sediments in the Ethiopian Traps in order to characterize silicate weathering and corresponding CO₂ consumption in this region. Lithium isotopic composition of the clay fractions was also examining and present a positive correlation with clay Mg/Ti ratios, in relation with variable weathering conditions in this region. The average weathering rate (16.1 tons/km²/yr) and CO₂ consumption rate (0.65.10¹² 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 CO₂ consumption rate for both periods. The determined CO₂ consumption rate for the African Humid Period range

between 0.91 and 1.5.10¹² mol/yr. This indicates that during humid periods the Ethiopian 588 traps could have played a significate role on carbon cycle and on climate. 589 Overall, this study brings out the key role of a region that is understudied thus far in terms of 590 weathering and climate paleo-variations. More generally, the influence on global climate of 591 tropical basins influenced by monsoon would deserve to be assessed more precisely, in 592 particular those of basaltic lithology. 593 594 595 596 Acknowledgments 597 598 This work was supported by BQR Geoazur, CNRS INSU-SYSTER grants and ANR 599 INTOCC. We thank P. Telouk (LGLTPE) for his assistance with the ENS-Lyon Neptune 600 plus. We thank also P.H Blard and B. Marty (CRPG) for their help on the field and fruitful 601 discussions. 602

- Amaral, P.G.C., Vincens, A., Guiot, J., Buchet, G., Deschamps, P., Doumnang, J.C.,
- Sylvestre, F., 2013. Palynological evidence for gradual vegetation and climate changes
- during the African Humid Period termination at 13°N from a Mega-Lake Chad
- sedimentary sequence. Clim. Past 9, 223–241. https://doi.org/10.5194/cp-9-223-2013
- Ayalew, D., Barbey, P., Marty, B., Reisberg, L., Yirgu, G., Pik, R., 2002. Source, genesis, and
- timing of giant ignimbrite deposits associated with Ethiopian continental flood basalts.
- Geochim. Cosmochim. Acta 66, 1429–1448. https://doi.org/10.1016/S0016-
- 613 7037(01)00834-1
- Balter, V., Vigier, N., 2014. Natural variations of lithium isotopes in a mammalian model.
- 615 Metallomics 6, 582. https://doi.org/10.1039/c3mt00295k
- Bastian, L., 2017. Thèse de doctorat Bastian Luc Impact des variations de la mousson
- Africaine sur l'érosion chimique des silicates dans le bassin versant du Nil depuis.
- Bastian, L., Revel, M., Bayon, G., Dufour, A., Vigier, N., 2017. Abrupt response of chemical
- weathering to Late Quaternary hydroclimate changes in northeast Africa. Sci. Rep. 7,
- 620 44231. https://doi.org/10.1038/srep44231
- Bayon, G., Dennielou, B., Etoubleau, J., Ponzevera, E., Toucanne, S., Bermell, S., 2012. Use
- in Iron Age Central Africa. Science (80-.). 335, 1219–1222.
- 623 https://doi.org/10.1126/science.1215400
- Bayon, G., Skonieczny, C., Delvigne, C., Toucanne, S., Bermell, S., Ponzevera, E., André, L.,
- 625 2016. Environmental Hf-Nd isotopic decoupling in World river clays. Earth Planet. Sci.
- 626 Lett. 438, 25–36. https://doi.org/10.1016/j.epsl.2016.01.010
- Benedetti, M.F., Menard, O., Noack, Y., Carvalho, A., Nahon, D., 1994. Water-rock
- interactions in tropical catchments: field rates of weathering and biomass impact. Chem.
- Geol. 118, 203–220. https://doi.org/10.1016/0009-2541(94)90177-5
- Berner, E., Berner, R.A., 1987. The Global Water Cycle. Geochemistry Environ. Prentice
- 631 Hall.
- Blanchet, C.L., Frank, M., Schouten, S., 2014. Asynchronous changes in vegetation, runoff
- and Erosion in the Nile River watershed during the Holocene. PLoS One 9, 1–18.
- https://doi.org/10.1371/journal.pone.0115958
- Blanchet, C.L., Tjallingii, R., Frank, M., Lorenzen, J., Reitz, A., Brown, K., Feseker, T.,
- Brückmann, W., 2013. High- and low-latitude forcing of the Nile River regime during
- the Holocene inferred from laminated sediments of the Nile deep-sea fan. Earth Planet.
- 638 Sci. Lett. 364, 98–110. https://doi.org/10.1016/j.epsl.2013.01.009
- Bouchez, J., Gaillardet, J., von Blanckenburg, F., 2014. Weathering Intensity in Lowland
- River Basins: From the Andes to the Amazon Mouth. Procedia Earth Planet. Sci. 10,
- 280–286. https://doi.org/10.1016/j.proeps.2014.08.063
- Bouchez, J., Von Blanckenburg, F., Schuessler, J.A., 2013. Modeling novel stable isotope
- ratios in the weathering zone. Am. J. Sci. 313, 267–308.
- https://doi.org/10.2475/04.2013.01
- Brady, P. V, Carroll, S.A., 1994. Direct effects of CO2 and temperature on silicate

- weathering: Possible implications for climate control. Pergamon Gecchimica
 Cosmochim. Acta 58, 1853–1856. https://doi.org/10.1016/0016-7037(94)90543-6
- Brady, P. V, Dorn, R.I., Brazel, A.J., Clark, J., Moore, R.B., Glidewell, T., 1999. Direct measurement of the combined effects of lichen, rainfall, and temperature on silicate
- weathering. Geochim. Cosmochim. Acta 63, 3293–3300. https://doi.org/10.1016/S0016-
- 651 7037(99)00251-3
- 652 Calmels, D., Gaillardet, J., Brenot, A., France-Lanord, C., 2007. Sustained sulfide oxidation 653 by physical erosion processes in the Mackenzie River basin: Climatic perspectives.
- Geology 35, 1003–1006. https://doi.org/10.1130/G24132A.1
- 655 Carignan, J., Vigier, N., Millot, R., 2007. Three secondary reference materials for lithium
- isotope measurements: Li7-N, Li6-N and LiCl-N solutions. Geostand. Geoanalytical Res.
- 657 31, 7–12. https://doi.org/10.1111/j.1751-908X.2007.00833.x
- 658 Castañeda, I.S., Schouten, S., Pätzold, J., Lucassen, F., Kasemann, S., Kuhlmann, H.,
- Schefuß, E., 2016. Hydroclimate variability in the Nile River Basin during the past
- 28,000 years. Earth Planet. Sci. Lett. 438, 47–56.
- https://doi.org/10.1016/j.epsl.2015.12.014
- Chadwick, O.A., Derry, L.A., Vitousek, P.M., Huebert, B.J., Hedin, L.O., 1999. Changing
- sources of nutrients during four million years of ecosystem development Terrestrial
- biogeochemists traditionally distinguish atmospherically derived from rock-derived
- elements. Nat. www.nature.com 397.
- 666 Clergue, C., Dellinger, M., Buss, H.L., Gaillardet, J., Benedetti, M.F., Dessert, C., 2015.
- Influence of atmospheric deposits and secondary minerals on Li isotopes budget in a
- highly weathered catchment, Guadeloupe (Lesser Antilles). Chem. Geol. 414, 28–41.
- https://doi.org/10.1016/j.chemgeo.2015.08.015
- Conway, D., 1997. A water balance model of the Upper Blue Nile in Ethiopia. Hydrol. Sci. J.
 42, 265–286. https://doi.org/10.1080/02626669709492024
- Das, A., Krishnaswami, S., Sarin, M.M., Pande, K., 2005. Chemical weathering in the
- Krishna Basin and Western Ghats of the Deccan Traps, India: Rates of basalt weathering
- and their controls. Geochim. Cosmochim. Acta 69, 2067–2084.
- 675 https://doi.org/10.1016/j.gca.2004.10.014
- De Boer, E.J., Hooghiemstra, H., Vincent Florens, F.B., Baider, C., Engels, S., Dakos, V.,
- Blaauw, M., Bennett, K.D., 2013. Rapid succession of plant associations on the small
- ocean island of Mauritius at the onset of the Holocene. Quat. Sci. Rev. 68, 114–125.
- https://doi.org/10.1016/j.quascirev.2013.02.005
- Decarreau, A., Vigier, N., Pálková, H., Petit, S., Vieillard, P., Fontaine, C., 2012. Partitioning
- of lithium between smectite and solution: An experimental approach. Geochim.
- 682 Cosmochim. Acta 85, 314–325. https://doi.org/10.1016/j.gca.2012.02.018
- Dellinger, M., Bouchez, J., Gaillardet, J., Faure, L., Moureau, J., 2017. Tracing weathering
- regimes using the lithium isotope composition of detrital sediments. Geology 45, 411–
- 685 414. https://doi.org/10.1130/G38671.1
- Dellinger, M., Gaillardet, J., Bouchez, J., Calmels, D., Louvat, P., Dosseto, A., Gorge, C.,
- Alanoca, L., Maurice, L., 2015. Riverine Li isotope fractionation in the Amazon River
- basin controlled by the weathering regimes. Geochim. Cosmochim. Acta 164, 71–93.

- 689 https://doi.org/10.1016/j.gca.2015.04.042
- 690 Dellinger, M., Gaillardet, J.Ô., Bouchez, J., Calmels, D., Galy, V., Hilton, R.G., Louvat, P.,
- France-Lanord, C., 2014. Lithium isotopes in large rivers reveal the cannibalistic nature
- of modern continental weathering and erosion. Earth Planet. Sci. Lett. 401, 359–372.
- 693 https://doi.org/10.1016/j.epsl.2014.05.061
- 694 Dessert, C., Dupre, B., Francois, L.M., Schott, J., Gaillardet, J., Chakrapani, G., Bajpai, S.,
- 695 2001. Erosion of Deccan Traps determined by river geochemistry; impact on the global
- climate and the (super 87) Sr/ (super 86) Sr ratio of seawater. Earth Planet. Sci. Lett. 188,
- 697 459–474.
- Dessert, C., Dupré, B., Gaillardet, J., François, L.M., Allègre, C.J., 2003. Basalt weathering
- laws and the impact of basalt weathering on the global carbon cycle. Chem. Geol. 202,
- 700 257–273. https://doi.org/10.1016/j.chemgeo.2002.10.001
- 701 Dosseto, A., Vigier, N., Joannes-Boyau, R., Moffat, I., Singh, T., Srivastava, P., 2015. Rapid
- response of silicate weathering rates to climate change in the Himalaya. Geochemical
- 703 Perspect. Lett. 10–19. https://doi.org/10.7185/geochemlet.1502
- Dupré, B., Viers, J., Dandurand, J.L., Polve, M., Bénézeth, P., Vervier, P., Braun, J.J., 1999.
- Major and trace elements associated with colloids in organic-rich river waters:
- 706 Ultrafiltration of natural and spiked solutions. Chem. Geol. 160, 63–80.
- 707 https://doi.org/10.1016/S0009-2541(99)00060-1
- 708 Elliott, T., Thomas, A., Jeffcoate, A., Niu, Y., 2006. Lithium isotope evidence for subduction-
- 709 enriched mantle in the source of mid-ocean-ridge basalts. Nature 443, 565–568.
- 710 https://doi.org/10.1038/nature05144
- Fielding, L., Najman, Y., Millar, I., Butterworth, P., Ando, S., Padoan, M., Barfod, D.,
- Kneller, B., 2017. A detrital record of the Nile River and its catchment. J. Geol. Soc.
- 713 London. 174, 301–317. https://doi.org/10.1144/jgs2016-075
- Forman, S.L., Wright, D.K., Bloszies, C., 2014. Variations in water level for Lake Turkana in
- the past 8500 years near Mt. Porr, Kenya and the transition from the African Humid
- Period to Holocene aridity. Quat. Sci. Rev. 97, 84–101.
- 717 https://doi.org/10.1016/j.quascirev.2014.05.005
- 718 Gaillardet, J., Dupré, B., Louvat, P., Allègre, C.J., 1999. Global silicate weathering and CO
- consumption rates deduced 2 from the chemistry of large rivers. Chem. Geol. 159, 3–30.
- 720 https://doi.org/http://dx.doi.org/10.1016/S0009-2541(99)00031-5
- Garcin, Y., Junginger, A., Melnick, D., Olago, D.O., Strecker, M.R., Trauth, M.H., 2009. Late
- 722 Pleistocene-Holocene rise and collapse of Lake Suguta, northern Kenya Rift. Quat. Sci.
- 723 Rev. 28, 911–925. https://doi.org/10.1016/j.quascirev.2008.12.006
- Garzanti, E., Andò, S., Padoan, M., Vezzoli, G., El Kammar, A., 2015. The modern Nile
- sediment system: Processes and products. Quat. Sci. Rev. 130, 9–56.
- 726 https://doi.org/10.1016/j.quascirev.2015.07.011
- Genske, F.S., Turner, S.P., Beier, C., Chu, M.F., Tonarini, S., Pearson, N.J., Haase, K.M.,
- 728 2014. Lithium and boron isotope systematics in lavas from the Azores islands reveal
- 729 crustal assimilation. Chem. Geol. 373, 27–36.
- 730 https://doi.org/10.1016/j.chemgeo.2014.02.024
- 731 Gislason, S.R., Arnorsson, S., Armannsson, H., 1996. Chemical weathering of basalt as

- deduced from the composition of precipitation, rivers and rocks in SW Iceland. Am. J.
- 733 Sci. 296, 837–907.
- Gislason, S.R., Oelkers, E.H., 2003. Mechanism, rates, and consequences of basaltic glass
- dissolution: II. An experimental study of the dissolution rates of basaltic glass as a
- function of pH and temperature. Geochim. Cosmochim. Acta 67, 3817–3832.
- 737 https://doi.org/10.1016/S0016-7037(00)00176-5
- Gislason, S.R., Oelkers, E.H., Eiriksdottir, E.S., Kardjilov, M.I., Gisladottir, G., Sigfusson,
- B., Snorrason, A., Elefsen, S., Hardardottir, J., Torssander, P., Oskarsson, N., 2009.
- Direct evidence of the feedback between climate and weathering. Earth Planet. Sci. Lett.
- 741 277, 213–222. https://doi.org/10.1016/j.epsl.2008.10.018
- Goddéris, Y., Brantley, S.L., François, L.M., Schott, J., Pollard, D., Déqué, M., Dury, M.,
- 743 2013. Rates of consumption of atmospheric CO2 through the weathering of loess during
- the next 100 yr of climate change. Biogeosciences 10, 135–148.
- 745 https://doi.org/10.5194/bg-10-135-2013
- Hoelzmann, P., Schwalb, A., Roberts, N., Cooper, P., Burgess, A., 2010. Hydrological
- response of an east-Saharan palaeolake (NW Sudan) to early-Holocene climate.
- 748 Holocene 20, 537–549. https://doi.org/10.1177/0959683609356584
- Hofmann, C., Courtillot, V., Féraud, G., Rochette, P., Yirgus, G., Ketefo, E., Pik, R., 1998.
- 750 Timing of the Ethiopian flood basalt event and implications for plume birth and global
- change. Nature 394, 192–195. https://doi.org/10.1038/nature02336.1.
- Huh, Y., Chan, L.H., Chadwick, O.A., 2004. Behavior of lithium and its isotopes during
- weathering of Hawaiian basalt. Geochemistry, Geophys. Geosystems 5.
- 754 https://doi.org/10.1029/2004GC000729
- 755 Kisakurek, B., James, R.H., Harris, N.B.W., 2005. Li and δ7Li in Himalayan rivers: Proxies
- for silicate weathering? Earth Planet. Sci. Lett. 237, 387–401.
- 757 https://doi.org/10.1016/j.epsl.2005.07.019
- Kisakürek, B., Widdowson, M., James, R.H., 2004. Behaviour of Li isotopes during
- continental weathering: The Bidar laterite profile, India. Chem. Geol. 212, 27–44.
- 760 https://doi.org/10.1016/j.chemgeo.2004.08.027
- Korecha, D., Barnston, A.G., 2007. Predictability of June–September Rainfall in Ethiopia.
- 762 Mon. Weather Rev. 135, 628–650. https://doi.org/10.1175/MWR3304.1
- Lamb, H.F., Bates, C.R., Bryant, C.L., Davies, S.J., Huws, D.G., Marshall, M.H., Roberts,
- H.M., 2018. 150,000-year palaeoclimate record from northern Ethiopia supports early,
- multiple dispersals of modern humans from Africa. Sci. Rep. 8, 1–7.
- 766 https://doi.org/10.1038/s41598-018-19601-w
- Lanckriet, S., Schwenninger, J.L., Frankl, A., Nyssen, J., 2015. The Late-Holocene
- geomorphic history of the Ethiopian Highlands: Supportive evidence from May Tsimble.
- 769 Catena 135, 290–303. https://doi.org/10.1016/j.catena.2015.08.011
- Lechler, M., Pogge von Strandmann, P.A.E., Jenkyns, H.C., Prosser, G., Parente, M., 2015.
- Lithium-isotope evidence for enhanced silicate weathering during OAE 1a (Early Aptian
- 772 Selli event). Earth Planet. Sci. Lett. 432, 210–222.
- 773 https://doi.org/10.1016/j.epsl.2015.09.052
- Leemans, Cramer, 1991. The IIASA Database for Mean Monthly Values of Temperature,

- Precipitation, and Cloudiness on the Global Terrestrial Grid. International Institute for Applied Systems Analysis, Laxenburg, Austria. 1991.
- Lefebvre, V., Donnadieu, Y., Goddéris, Y., Fluteau, F., Hubert-Théou, L., 2013. Was the
 Antarctic glaciation delayed by a high degassing rate during the early Cenozoic? Earth
 Planet. Sci. Lett. 371–372, 203–211. https://doi.org/10.1016/j.epsl.2013.03.049
- Li, G., Elderfield, H., 2013. Evolution of carbon cycle over the past 100 million years.
 Geochim. Cosmochim. Acta 103, 11–25. https://doi.org/10.1016/j.gca.2012.10.014
- Li, G., Hartmann, J., Derry, L.A., West, A.J., You, C.F., Long, X., Zhan, T., Li, L., Li, G.,
 Qiu, W., Li, T., Liu, L., Chen, Y., Ji, J., Zhao, L., Chen, J., 2016. Temperature
- dependence of basalt weathering. Earth Planet. Sci. Lett. 443, 59–69.
- 785 https://doi.org/10.1016/j.epsl.2016.03.015
- Li, S.L., Calmels, D., Han, G., Gaillardet, J., Liu, C.Q., 2008. Sulfuric acid as an agent of
 carbonate weathering constrained by δ13CDIC: Examples from Southwest China. Earth
 Planet. Sci. Lett. 270, 189–199. https://doi.org/10.1016/j.epsl.2008.02.039
- Louvat, P., 1997. Etude géochimique de l'érosion fluviale d'iles volcaniques à l'aide des bilans d'éléments majeurs et traces. PhD Thesis Université, 322pp.
- Louvat, P., Allègre, C.J., 1998. Riverrine erosion rates on Sao Miguek volcanic island, Azores
 archipelago. Chem. Geol. 148, 177–200.
- Louvat, P., Allègre, C.J., 1998. Riverine erosion rates on Sao Miguel volcanic island, Azores
 archipelago. USDA For. Serv. Gen. Tech. Rep. RMRS-GTR 148, 177–200.
 https://doi.org/10.1016/S0009-2541(98)00028-X
- Louvat, P., Allègre, C.J., 1997. Present denudation rates on the island of réunion determined by river geochemistry: Basalt weathering and mass budget between chemical and mechanical erosions. Geochim. Cosmochim. Acta 61, 3645–3669. https://doi.org/10.1016/S0016-7037(97)00180-4
- Marzin, C., Braconnot, P., 2009. The role of the ocean feedback on Asian and African monsoon variations at 6 kyr and 9.5 kyr BP. Comptes Rendus Geosci. 341, 643–655. https://doi.org/10.1016/j.crte.2009.09.001
- MERID, F., 2005. National Nile Basin Water Quality Monitoring Baseline Report for Ethiopia.
- 805 Meybeck, M., 1986. Composition chimique des ruisseaux non pollués de France. Sci. Geol. 806 3–77.
- Millot, R., Gaillardet, J., Dupré, B., Allègre, C.J., 2002. The global control of silicate weathering rates and the coupling with physical erosion. new insights from rivers Can. Shield 196, 83–98.
- Millot, R., Guerrot, C., Vigier, N., 2004. Accurate and high-precision measurement of lithium isotopes in two reference materials by MC-ICP-MS. Geostand. Geoanalytical Res. 28, 153–159. https://doi.org/10.1111/j.1751-908X.2004.tb01052.x
- Millot, R., Vigier, N., Gaillardet, J., 2010. Behaviour of lithium and its isotopes during weathering in the Mackenzie Basin, Canada. Geochim. Cosmochim. Acta 74, 3897–3912. https://doi.org/10.1016/j.gca.2010.04.025
- Mohr. P, Z.B., 1988. The Ethiopian flood basalt province. McDougall, J.D. _Ed., Cont. flood

- basalts. Kluwer Acad. Publ., Dordr. 63–110. https://doi.org/10.1007/978-94-015-7805-
- 818 9_3
- Négrel, P., Allègre, C.J., Dupré, B., Lewin, E., 1993. Erosion sources determined by inversion
- of major and trace element ratios and strontium isotopic ratios in river water: the Congo
- Basin case. Earth Planet. Sci. Lett. 120, 59–76.
- Négrel, P., Deschamps, P., 1996. Natural and anthropogenic budgets of a small watershed in
- the Massif Central (France): chemical and strontium isotopic characterization of water
- and sediments. Aquat. Geochem 2, 1–27.
- Nyssen, J., Vandenreyken, H., Poesen, J., Moeyersons, J., Deckers, J., Haile, M., Salles, C.,
- Govers, G., 2005. Rainfall erosivity and variability in the Northern Ethiopian Highlands.
- J. Hydrol. 311, 172–187. https://doi.org/10.1016/j.jhydrol.2004.12.016
- Pik, R., Deniel, C., Coulon, C., Yirgu, G., Hofmann, C., Ayalew, D., 1998. The northwestern
- 829 Ethiopian Plateau flood basalts: classification and spatial distribution of magma types. J.
- Volcanol. Geotherm. Res. 81, 91–111. https://doi.org/10.1016/S0377-0273(97)00073-5
- Pik, R., Deniel, C., Coulon, C., Yirgu, G., Marty, B., 1999. Isotopic and trace element
- signatures of Ethiopian flood basalts: Evidence for plume-lithosphere interactions.
- 833 Geochim. Cosmochim. Acta 63, 2263–2279. https://doi.org/10.1016/S0016-
- 834 7037(99)00141-6
- Pik, R., Marty, B., Carignan, J., Lavé, J., 2003. Stability of the Upper Nile drainage network
- 836 (Ethiopia) deduced from (U-Th)/He thermochronometry: Implications for uplift and
- erosion of the Afar plume dome. Earth Planet. Sci. Lett. 215, 73–88.
- https://doi.org/10.1016/S0012-821X(03)00457-6
- Pistiner, J.S., Henderson, G.M., 2003. Lithium-isotope fractionation during continental
- weathering processes. Earth Planet. Sci. Lett. 214, 327–339.
- https://doi.org/10.1016/S0012-821X(03)00348-0
- Pogge von Strandmann, P.A.E., Frings, P.J., Murphy, M.J., 2017. Lithium isotope behaviour
- during weathering in the Ganges Alluvial Plain. Geochim. Cosmochim. Acta 198, 17–31.
- https://doi.org/10.1016/j.gca.2016.11.017
- Pokrovsky, O.S., Schott, J., Kudryavtzev, D.I., Dupré, B., 2005. Basalt weathering in Central
- Siberia under permafrost conditions. Geochim. Cosmochim. Acta 69, 5659–5680.
- 847 https://doi.org/10.1016/j.gca.2005.07.018
- Revel, M., Colin, C., Bernasconi, S., Combourieu-Nebout, N., Ducassou, E., Grousset, F.E.,
- 849 Rolland, Y., Migeon, S., Bosch, D., Brunet, P., Zhao, Y., Mascle, J., 2014. 21,000 Years
- of Ethiopian African monsoon variability recorded in sediments of the western Nile
- deep-sea fan. Reg. Environ. Chang. 14, 1685–1696. https://doi.org/10.1007/s10113-014-
- 852 0588-x
- Revel, M., Ducassou, E., Grousset, F.E., Bernasconi, S.M., Migeon, S., Revillon, S., Mascle,
- J., Murat, A., Zaragosi, S., Bosch, D., 2010. 100,000 Years of African monsoon
- variability recorded in sediments of the Nile margin. Quat. Sci. Rev. 29, 1342–1362.
- 856 https://doi.org/10.1016/j.quascirev.2010.02.006
- Revel, M., Ducassou, E., Skonieczny, C., Colin, C., Bastian, L., Bosch, D., Migeon, S.,
- Mascle, J., 2015. 20,000 years of Nile River dynamics and environmental changes in the
- Nile catchment area as inferred from Nile upper continental slope sediments. Quat. Sci.

- Rev. 130. https://doi.org/10.1016/j.quascirev.2015.10.030
- Riotte, J., Chabaux, F., Benedetti, M.F., Dia, A., Gérard, M., Boulégue, J., Etamé, J., 2003. U
- colloidal transport and origin of the 234U/238U fractionation in surface waters: new
- insights from Mount Cameroon. Chem. Geol. 202, 363–379.
- Rochette, P., Tamrat, E., Féraud, G., Pik, R., Courtillot, V., Ketefo, E., Coulon, C., Hoffmann,
- 865 C., Vandamme, D., Yirgu, G., 1998. Magnetostratigraphy and timing of the Oligocene
- 866 Ethiopian traps. Earth Planet. Sci. Lett. 164, 497–510. https://doi.org/10.1016/S0012-
- 867 821X(98)00241-6
- Rothacker, L., Dosseto, A., Francke, A., Chivas, A.R., Vigier, N., Kotarba-Morley, A.M.,
- Menozzi, D., 2018. Impact of climate change and human activity on soil landscapes over
- the past 12,300 years. Sci. Rep. 8, 1–7. https://doi.org/10.1038/s41598-017-18603-4
- 871 Ryu, J.S., Vigier, N., Lee, S.W., Lee, K.S., Chadwick, O.A., 2014. Variation of lithium
- isotope geochemistry during basalt weathering and secondary mineral transformations in
- Hawaii. Geochim. Cosmochim. Acta 145, 103–115.
- https://doi.org/10.1016/j.gca.2014.08.030
- 875 Schmitt, A.D., Vigier, N., Lemarchand, D., Millot, R., Stille, P., Chabaux, F., 2012. Processes
- controlling the stable isotope compositions of Li, B, Mg and Ca in plants, soils and
- waters: A review. Comptes Rendus Geosci. 344, 704–722.
- 878 https://doi.org/10.1016/j.crte.2012.10.002
- Shanahan, T.M., Mckay, N.P., Hughen, K.A., Overpeck, J.T., Otto-Bliesner, B., Heil, C.W.,
- King, J., Scholz, C.A., Peck, J., 2015. The time-transgressive termination of the African
- humid period. Nat. Geosci. 8, 140–144. https://doi.org/10.1038/ngeo2329
- Stallard, R.F., Edmond, J.M., 1983. Geochemistry of the Amazone. J. Geophys. Res. 88,
- 883 9671–9688.
- Torres, M.A., West, A.J., Clark, K.E., Paris, G., Bouchez, J., Ponton, C., Feakins, S.J., Galy,
- V., Adkins, J.F., 2016. The acid and alkalinity budgets of weathering in the Andes–
- Amazon system: Insights into the erosional control of global biogeochemical cycles.
- Earth Planet. Sci. Lett. 450, 381–391. https://doi.org/10.1016/j.epsl.2016.06.012
- Tuenter, E., Weber, S.L., Hilgen, F.J., Lourens, L.J., 2003. The response of the African
- summer monsoon to remote and local forcing due to precession and obliquity. Glob.
- Planet. Change 36, 219–235. https://doi.org/10.1016/S0921-8181(02)00196-0
- 891 Verney-Carron, A., Vigier, N., Millot, R., 2011. Experimental determination of the role of
- diffusion on Li isotope fractionation during basaltic glass weathering. Geochim.
- 893 Cosmochim. Acta 75, 3452–3468. https://doi.org/10.1016/j.gca.2011.03.019
- Vigier, N., Bourdon, B., Lewin, E., Dupré, B., Turner, S., Chakrapani, G.J., van Calsteren, P.,
- Allègre, C.J., 2005. Mobility of U-series nuclides during basalt weathering: An example
- from the Deccan Traps (India). Chem. Geol. 219, 69–91.
- 897 https://doi.org/10.1016/j.chemgeo.2005.02.010
- 898 Vigier, N., Decarreau, A., Millot, R., Carignan, J., Petit, S., France-Lanord, C., 2008.
- Quantifying Li isotope fractionation during smectite formation and implications for the
- 900 Li cycle. Geochim. Cosmochim. Acta 72, 780–792.
- 901 https://doi.org/10.1016/j.gca.2007.11.011
- Vigier, N., Gislason, S.R., Burton, K.W., Millot, R., Mokadem, F., 2009. The relationship

- between riverine lithium isotope composition and silicate weathering rates in Iceland.
- 904 Earth Planet. Sci. Lett. 287, 434–441. https://doi.org/10.1016/j.epsl.2009.08.026
- Vigier, N., Goddéris, Y., 2015. A new approach for modeling Cenozoic oceanic lithium
- isotope paleo-variations: The key role of climate. Clim. Past 11, 635–645.
- 907 https://doi.org/10.5194/cp-11-635-2015
- 908 von Strandmann, P.A.E.P., Burton, K.W., James, R.H., van Calsteren, P., Gislason, S.R.,
- 909 2010. Assessing the role of climate on uranium and lithium isotope behaviour in rivers
- 910 draining a basaltic terrain. Chem. Geol. 270, 227–239.
- 911 https://doi.org/10.1016/j.chemgeo.2009.12.002
- von Strandmann, P.A.E.P., Burton, K.W., James, R.H., van Calsteren, P., Gíslason, S.R.,
- Mokadem, F., 2006. Riverine behaviour of uranium and lithium isotopes in an actively
- glaciated basaltic terrain. Earth Planet. Sci. Lett. 251, 134–147.
- 915 https://doi.org/10.1016/j.epsl.2006.09.001
- von Strandmann, P.A.E.P., Opfergelt, S., Lai, Y.J., Sigfússon, B., Gislason, S.R., Burton,
- 917 K.W., 2012. Lithium, magnesium and silicon isotope behaviour accompanying
- weathering in a basaltic soil and pore water profile in Iceland. Earth Planet. Sci. Lett.
- 919 339–340, 11–23. https://doi.org/10.1016/j.epsl.2012.05.035
- Walker, J.C.G., Hays, P.B., Kasting, J.F., 1981. Weathering rates in 106 gm km -2 yr -1 of
 dissolved Si02. Parentheses signify. J. Geophys. Res. 86, 9776–9782.
- West, A.J., Galy, A., Bickle, M., 2005. Tectonic and climatic controls on silicate weathering.
- 923 Earth Planet. Sci. Lett. 235, 211–228. https://doi.org/10.1016/j.epsl.2005.03.020
- Wimpenny, J., Gíslason, S.R., James, R.H., Gannoun, A., Pogge Von Strandmann, P.A.E.,
- Burton, K.W., 2010. The behaviour of Li and Mg isotopes during primary phase
- dissolution and secondary mineral formation in basalt. Geochim. Cosmochim. Acta 74,
- 927 5259–5279. https://doi.org/10.1016/j.gca.2010.06.028
- Wolfenden, E., Ebinger, C., Yirgu, G., Deino, A., Ayalew, D., 2004. Evolution of the
- 929 northern Main Ethiopian rift: Birth of a triple junction. Earth Planet. Sci. Lett. 224, 213–
- 930 228. https://doi.org/10.1016/j.epsl.2004.04.022
- 231 Zakharova, E.A., Pokrovsky, O.S., Dupré, B., Zaslavskaya, M.B., 2005. Chemical weathering
- of silicate rocks in Aldan Shield and Baikal Uplift: Insights from long-term seasonal
- measurements of solute fluxes in rivers. Chem. Geol. 214, 223–248.
- 934 https://doi.org/10.1016/j.chemgeo.2004.10.003

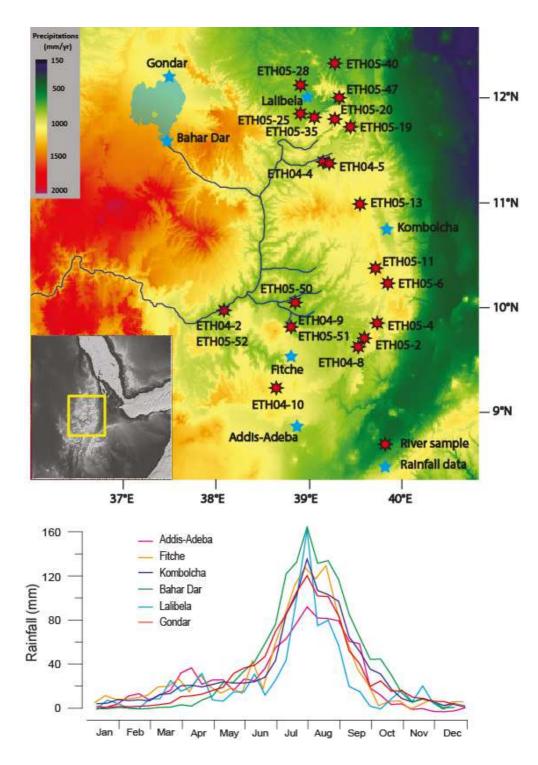


Fig 1: A. Map showing samples localization and the studied area. Sample name on the map are: 1: Jara Wenz (ETH05-11); 2:Borkena Wenz (ETH05-13); 3: Robi Wenz (ETH05-4); 4: Meri Wenz (ETH05-28); 5: Tikur Wenz (ETH05-19); 6: Ketchin Abeba Wenz (ETH05-25); 7: Gereb Ara Wenz (ETH05-40); 8: Tekeze (ETH05-35); 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 the Ethiopian Traps. Reported values corresponds to the average of rainfall between 1961 and 1997 for Gondar, Bahar Dar, Kombolcha and Addis Ababa, between 1991 and 1997 for Fiche and between 1994 and 1997 for Lalibellathe.

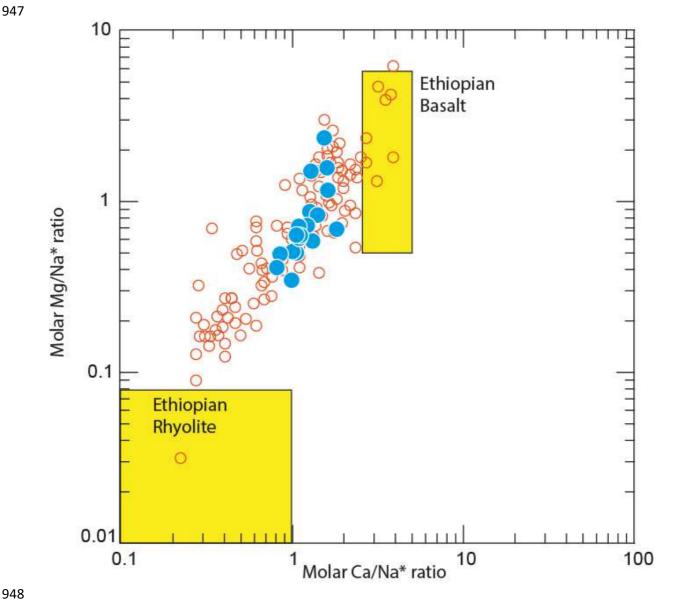


Fig 2: Mg/Na molar ratio as a function of Ca/Na molar ratio, for river dissolved phases. Both ratios are corrected from atmospheric inputs as described in section 4.2.1. The blue circles are for samples analyzed in this study. Orange circles are for other basaltic regions reported in Dessert et al., (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)

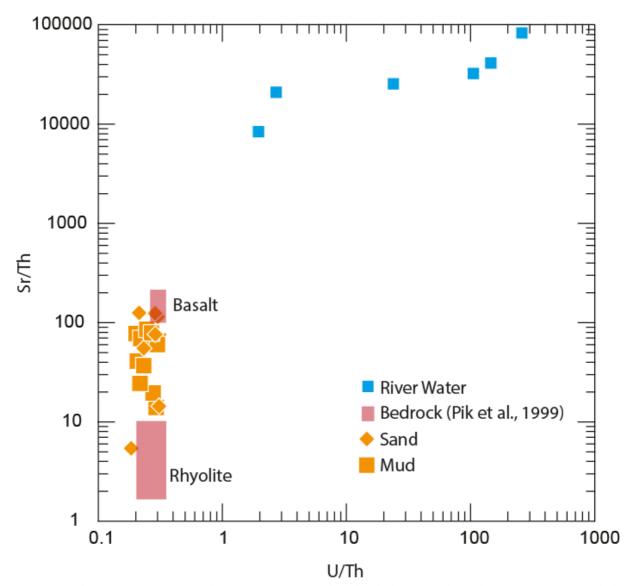


Fig 3: Sr/Th as a function of U/Th (wt ratios) in the dissolved phases and solid phases of the sampled rivers. Bedrock values come from Pik et al., 1999.

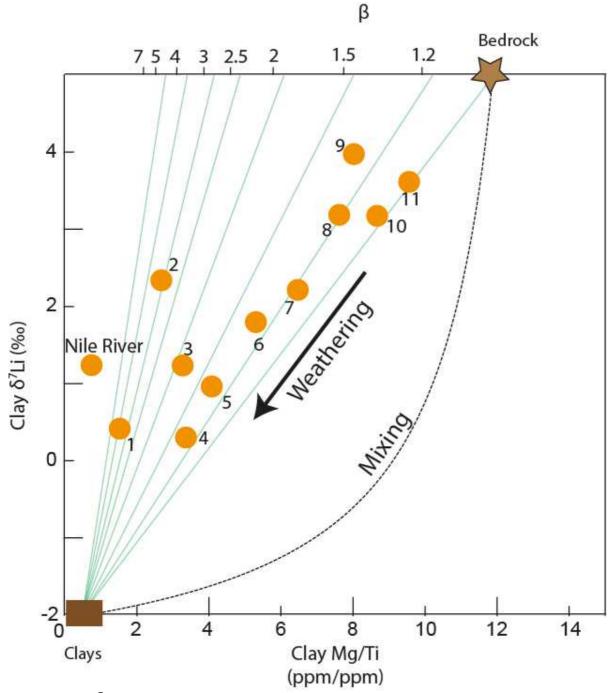


Fig 4: Clay $\delta^7 \text{Li}$ as a function of clay Mg/Ti for the main Blue Nile Basin rivers (blue diamonds). Green lines correspond to the result of the model described in section 5.1. for different β values. The dotted line corresponds to the theoretical mixing curve between bedrock and isotopically fractionated clay endmembers. Mixing model conditions: Bedrock $\delta 7 \text{Li} = 5\%$, [Li] = 10 ppm (this study); clays $\delta^7 \text{Li} = -2.3\%$ (clay $\delta^7 \text{Li}$ measured in the most recent Nile delta fan sediments, [Li] = 100 ppm (Bastian, 2017; Bastian et al., 2017). River numbers are the same as in figure 1.

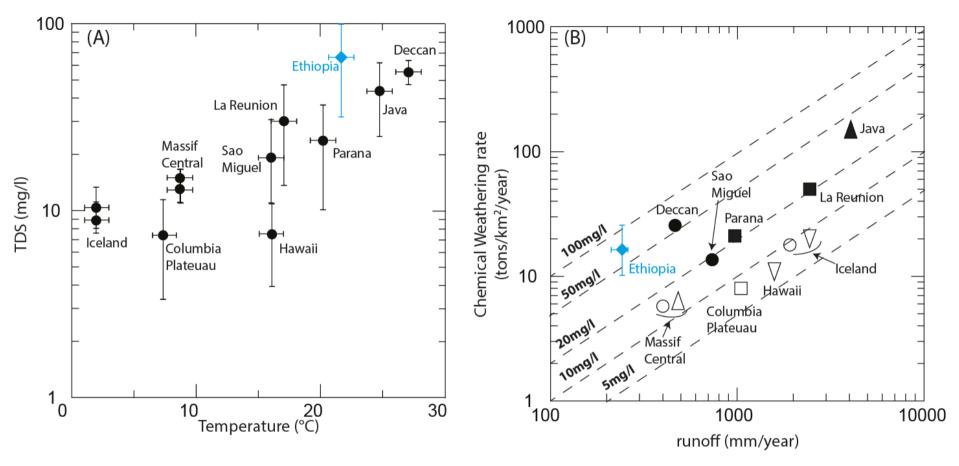


Fig 5: (A) TDS* (Total Dissolved Solids corrected for atmospheric inputs) as a function of temperature. The blue diamond corresponds to the Ethiopian Traps (average based on 17 data, see Table 3), and the black circles come from the compilation of (Dessert et al., 2003) (Benedetti et al., 1994; Dessert et al., 2001; Gaillardet et al., 1999; Gislason et al., 1996; Leemans and Cramer, 1991; Louvat, 1997; Pascale Louvat and Allègre, 1998; 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.

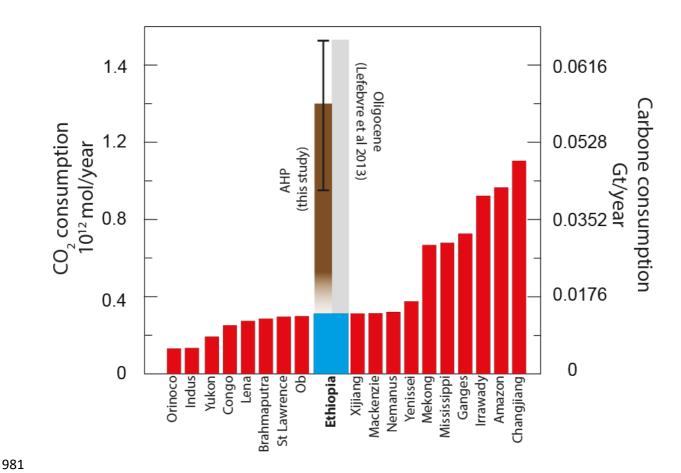


Fig 6: Total CO₂ consumption in mol/year (Gaillardet et al., 1999) and annual CO₂ consumption in GtC/yr due to silicate weathering for the largest rivers system. The blue bar corresponds to the current CO₂ consumption rate estimated in Ethiopia based on our measurements in river waters. The brown bar indicate the CO₂ 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. Table 2: Chemical composition of river waters, Total Dissolved Solid (TDS), silicate weathering rate and related CO₂ consumption rate for the major basins of the Ethiopian 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)

1003 Supplementary informations

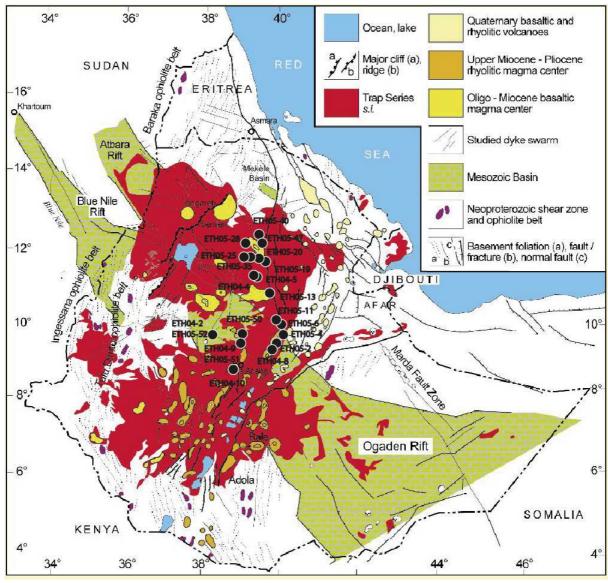


Fig S1: Ethiopian geological map. The black circles indicate our sampling sites.

(https://esdac.jrc.ec.europa.eu/images/Eudasm/Africa/images/maps/download/afr_etgm.jpg)

Table 1: Characteristics of the studied watersheds.

Sample number	Sample name	Elevation (m)	Mean elevation (m)	Area (km²)	Annual discharge (m³/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 2: Chemical composition of river waters, Total Dissolved Solid (TDS), silicate weathering rate and CO₂ consumption rate for the major basins of the Ethiopian Traps

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 ⁻ µmol/l	Cl ⁻ µmol/l	NO ₃ - µmol/l	SO ₄ ²⁻ µmol/l	Mg* (µmol/l)	Ca* (µmol/l)	Na* (µmol/l)	K* (µmol/l)	F ⁻ * µmol/l	NO ₃ -* µmol/l	SO ₄ ²⁻ * μmol/l	HCO ₃ -* µmol/l	TDS* (µmol/l)	TDS* (mg/l)	Total chemical weathering tons/km²/year	CO2 10 ⁶ 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	8.0	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

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)

14010 0. 011011	mear composition or carra,	That and day samples (day fractions have been separated using physical decantation to																							Clay										
Sample	Sample	Sand SiO ₂ Al ₂ O ₃ Fe ₂ O ₃ MnO MgO CaO Na ₂ O K2O TiO ₂ Sr Th U									$egin{array}{c ccccccccccccccccccccccccccccccccccc$												Clay												
	•	SiO ₂	Al_2O_3	Fe ₂ O ₃	₃ MnO	MgO	CaO	Na ₂ O	K20	TiO_2	Sr	Th	U	SiO ₂	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na ₂ O	K20	TiO_2	Sr	Th	U	δ7Li	K20	CaO	MgO	Fe2O3	Al2O3	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	8.0	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	8.0	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	8.0	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	8.0	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