



**HAL**  
open science

## Atmospheric contribution to cations cycling in highly weathered catchment, Guadeloupe (Lesser Antilles)

Celine Dessert, Clémentine Clergue, Alain Maurice Adolphe Antoine Rousteau, Olivier Crispi, Marc F Benedetti

### ► To cite this version:

Celine Dessert, Clémentine Clergue, Alain Maurice Adolphe Antoine Rousteau, Olivier Crispi, Marc F Benedetti. Atmospheric contribution to cations cycling in highly weathered catchment, Guadeloupe (Lesser Antilles). *Chemical Geology*, 2020, 531, pp.119354. 10.1016/j.chemgeo.2019.119354 . hal-02403791

**HAL Id: hal-02403791**

**<https://hal.science/hal-02403791>**

Submitted on 21 Dec 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.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

1 **Atmospheric contribution to cations cycling in highly weathered catchment, Guadeloupe**  
2 **(Lesser Antilles)**

3

4 Céline Dessert<sup>1,a</sup>, Clémentine Clergue<sup>1</sup>, Alain Rousteau<sup>2</sup>, Olivier Crispì<sup>1,b</sup>, Marc F. Benedetti<sup>1</sup>

5

6 <sup>1</sup>Université de Paris, Institut de physique du globe de Paris, CNRS, F-75005 Paris, France

7 <sup>2</sup>Laboratoire de biologie et de physiologie végétales, UMR EcoFoG, CNRS, Cirad, INRA, Université des  
8 Antilles, Université de Guyane, 97159 Pointe-à-Pitre, France

9

10 <sup>a</sup> corresponding author: E-mail address: [dessert@ipgp.fr](mailto:dessert@ipgp.fr) (Céline Dessert)

11 <sup>b</sup> present address: Laboratoire d'océanographie biologique, UMR 7621, Laboratoire Arago, BP 44,  
12 66651 Banyuls-sur-Mer cedex, France

13

14 **Keywords:** Atmospheric deposit, Saharan dust, cation-nutrient recycling, Sr and Nd isotopes, Critical  
15 Zone.

16

17 **Abstract:**

18 The important fertilizing role of atmospheric dust, and particularly African dust, in tropical rainforests  
19 is increasingly recognized but still poorly quantified. To better evaluate dust input into the Caribbean  
20 basin, we sampled critical zone compartments of a small forested volcanic catchment in Guadeloupe  
21 (soils, parent rock, atmospheric dust, plants, soil solutions, stream and rain waters). The aims of this

22 study are to track sources of cation nutrients (Ca, Mg, K, Sr) developed on highly weathered soil in the  
23 rainforest of Guadeloupe, to quantify plant recycling of these nutrients, and to identify constraints on  
24 regolith development and its associated nutrient pool.

25 In the Quiock Creek catchment, a large isotopic range of  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\epsilon\text{Nd}$  values was observed despite  
26 the small scale of observation. Sr isotopic composition of the dissolved load varied from 0.7084 in  
27 rainfall to 0.7110 in soil solution, whereas it ranges between 0.7068 and 0.7153 for soil samples and  
28 between 0.7096 and 0.7102 for plants. The Nd isotopic composition varied between -8.39 in near-  
29 surface soil samples to 2.71 in deeper soil. All samples had an intermediate signature between that of  
30 the bedrock endmember ( $^{87}\text{Sr}/^{86}\text{Sr}=0.7038$ ;  $\epsilon\text{Nd}=4.8$ ) and the atmospheric endmember (sea salt:  
31  $^{87}\text{Sr}/^{86}\text{Sr}=0.7092$  and Saharan dust:  $^{87}\text{Sr}/^{86}\text{Sr}=0.7187$ ,  $\epsilon\text{Nd}=-11.5$ ).

32 The regolith was built on pyroclastic deposits, but, because of extreme leaching, the regolith has lost  
33 its original bedrock signature and inherited an exogenous atmospheric signature. Our results show that  
34 only the chemical weathering of the fresh near-surface minerals can provide nutrients to the ecosystem  
35 (first 30 cm). However, this dust weathering is too low to sustain the tropical forest ecosystem on its  
36 own. The cationic mass balance at the catchment scale, as well as the Sr isotopic signature, show that  
37 cation and Sr fluxes are of atmospheric origin only and that original bedrock no longer participates in  
38 nutrient cycles. The vegetation reflects the  $^{87}\text{Sr}/^{86}\text{Sr}$  of the dissolved pool of atmospheric Sr.

39 At the soil-plant scale, the cation-nutrient fluxes provided by vegetation (litter fall + leaf excretion) are  
40 major compared to input and output fluxes. The annual Ca, K, Sr and Mg fluxes within the vegetation  
41 are, respectively, 31, 28, 20 and 3 times greater than the exported fluxes at the outlet of the basin. The  
42 residence time of nutrients in the vegetation is 16 years for K and close to 45 years for Sr, Ca and Mg.  
43 These results emphasize the highly efficient vegetative turnover that dominates the nutrient cycle in  
44 the Quiock Creek catchment.

45 This first characterization of biogeochemical cycles in the Guadeloupean rainforest suggests that the  
46 forest community of Quiock Creek is sustained by a small near-surface nutrient pool disconnected from

47 the deep volcanic bedrock. We also demonstrated that, even with efficient nutrient recycling, Saharan  
48 dust plays a significant role in maintaining ecosystem productivity in Guadeloupe over long-time scales.

## 49 1. Introduction

50 The base cations calcium, potassium and magnesium, are nutrient elements essential for plant  
51 development as they are building blocks of tissues and chlorophyll and have a number of metabolic  
52 functions. They also play a major role in neutralizing soil acidity and maintaining biological activity. In  
53 ecosystems, base cations in soils and plants originate from in situ mineral weathering (*e.g.* Schlesinger,  
54 1997). However, numerous studies have shown that, under specific circumstances, these elements can  
55 be provided by the atmosphere through precipitation or dust (*e.g.* Dahms, 1993; Hedin et al., 1994;  
56 Kennedy et al., 1998). Forest ecosystems are generally poor in nutrients and many studies have  
57 suggested that atmospheric deposits are significant sources of nutrients (*e.g.* Hedin and Likens, 1996;  
58 Okin et al., 2004; Soderberg and Compton, 2007; Pett-Ridge, 2009; Yu et al., 2015; Schuessler et al.,  
59 2018). Nutrient depletion in soils is particularly high in tropical areas, where temperature and  
60 precipitation combine to intensively leach nutrients from rocks and soil. In such contexts, the soil is  
61 usually strongly impacted by atmospheric inputs (*e.g.* Whipkey et al., 2000; Kurtz et al., 2001; Poszna  
62 et al., 2002; Pett-Ridge et al., 2009b). Cation nutrients, essential for plants, are thus expected to be  
63 strongly influenced by atmospheric inputs in highly weathered soils. Several studies conducted in  
64 Hawaii (Kennedy et al., 1998; Chadwick et al., 1999; Stewart et al., 2001), along chrono- and climato-  
65 sequence gradients, have shown that the more soils are leached, the more associated plants are  
66 influenced by atmospheric inputs. Other studies conducted in tropical areas of Costa-Rica (Bern et al.,  
67 2005; Porder et al., 2006) have shown that the soil-nutrient pool could still be influenced by bedrock  
68 weathering. The authors have suggested that rock-derived input was maintained by high erosion rates  
69 or deep plant roots.

70 The transport of African dust into the Caribbean basin is relatively well documented, either by  
71 measuring contemporary fluxes (*e.g.* Prospero et al., 1981; Prospero 1999) or by studying Quaternary  
72 age geological limestone records found on the western Atlantic islands of Barbados, the Florida Keys,  
73 and the Bahamas (*e.g.* Muhs et al., 1990; 2007). However, the number of soil record studies examining  
74 the impact of Saharan dust inputs on nutrient cycling over pedogenic timescales of hundreds to

75 thousands of years is limited. Studies conducted in the Luquillo Mountains in Puerto Rico (Pett-Ridge  
76 et al., 2009a; McClintock et al., 2015) have revealed that Saharan dust contributes significantly to  
77 nutrient inputs into weathered soils. Recent studies (Dessert et al., 2015; Clergue et al., 2015; Fries et  
78 al., 2019) have also tended to show that the soils of Guadeloupe can be strongly impacted by  
79 atmospheric deposits. These few studies did not take into account internal nutrient cycles in the  
80 watershed (e.g. litterfall, leaf excretion) and did not focus on the influence of dust on plants. However,  
81 the characterization of these cycles is important from the perspective of ecological theory and the  
82 understanding of Earth's Critical Zone.

83 As it is impossible to directly measure the cation-nutrient flux between rock, soil and vegetation, base  
84 cation cycles in forested ecosystems are not straightforward to characterize. Sr and its radiogenic  
85 isotopes have been shown to be proxies for cation-nutrient cycling in the atmosphere-soil-plant system  
86 (e.g. Capo et al., 1998; Bern et al., 2005; Belanger et al., 2012; Stille et al., 2012). As calcium has a  
87 hydrate ionic radius and an ionic charge similar to those of Sr, its behaviour can be deduced from the  
88  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio. K and Mg chemistry differs from that of Ca and Sr, but these lithophile elements  
89 ultimately originate from the same sources. Their behaviour can be inferred from Sr isotope data  
90 combined with on site element flux calculations. The efficient use of Sr isotopes as a cation-nutrient  
91 cycle proxy necessitates that different Sr sources exhibit different  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios.

92 In Guadeloupe, possible common sources of Sr and base cation nutrients (volcanic rock, sea salt and  
93 Saharan dust) differ in Sr isotopic signature. The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of volcanic bedrock is between 0.70341  
94 and 0.70382 (White and Patchett, 1984; White and Dupré, 1986). The signature of sea salt is 0.70917  
95 (e.g. Dia et al., 1992) and the  $^{87}\text{Sr}/^{86}\text{Sr}$  mean ratio of Saharan dust is 0.71788 (Biscaye et al., 1974;  
96 Grousset et al., 1988; Grousset et al., 1992; Rognon et al., 1996; Grousset et al., 1998; Pett-Ridge et al.,  
97 2009). We looked at Sr isotopes, Nd isotopes, and base cation nutrients to identify their sources. Like  
98 Sr, Nd isotope ratios are used to trace Nd sources, as they are not affected by biological and weathering  
99 processes (e.g. Liu et al., 2013). The Nd signature in volcanic rock from Guadeloupe ranges from 3.02  
100 to 5.05 (White and Patchett, 1984; White and Dupré, 1986) and the signature of Saharan dust varies

101 between -10.5 and -14.5 (Grousset et al., 1992; Dia et al., 2006). Nd and Sr differ in their mobility (Nd  
102 is less mobile than Sr) and are distributed differently in surface compartments, thus the comparison  
103 between these two isotopic systems provides complementary information on constraints in the study  
104 area.

105 In this study we report Sr isotope ratios in all compartments of a small catchment (rainfall, throughfall,  
106 soil solution, bulk soil, unweathered rock, river water, litter and atmospheric dusts) and Nd isotope  
107 ratios of soils and unweathered rock. The catchment is located in the tropical rainforest of Guadeloupe,  
108 in the Caribbean (FWI). This area is covered by highly weathered soils built on mono-lithological  
109 volcanoclastic deposits. Despite the poverty of the soils (Buss et al., 2010; Clergue et al., 2015), a dense  
110 rainforest is developed on it (Rousteau et al., 1994, 1996). The aims of this study are to: 1) Track sources  
111 of cation nutrients (Ca, Mg, K) of the rainforest of Guadeloupe developed on a highly weathered soil;  
112 2) Quantify the vegetation recycling of cation nutrients; 3) Identify the origin of “rock-derived” cation-  
113 nutrient pool in the soil and give constraints on regolith development.

114

## 115 **2. Site description**

116 The studied catchment (16°17'N, 61°70'W) is located on Basse-Terre Island, the volcanic part of the  
117 Guadeloupe archipelago in the French West Indies (Fig. 1). The Quiock Creek drains a watershed of 8  
118 ha, ranging from 200 m to 350 m in altitude, located in the primary tropical rainforest of the  
119 Guadeloupe National Park. The Quiock Creek catchment is monitored by the ObsErA observatory  
120 (OZCAR research infrastructure; Gaillardet et al., 2018) dedicated to the study of weathering and  
121 erosion processes under tropical climatic conditions (Clergue et al., 2015; Lloret et al., 2016; Fries et  
122 al., 2019; Guérin et al., 2019).

123 The climate on site is characterized by high mean annual temperature (25°C) and precipitation (3500  
124 mm $\text{yr}^{-1}$ ). The mean annual runoff is 1,130 mm. Evapotranspiration is high, between 60 and 70%  
125 (Clergue et al., 2015; Guérin et al., 2019) and tropical storms and hurricanes contribute significantly to  
126 the hydrology of the site (Zahibo et al., 2007).

127 Quiock Creek is covered by a very thick ferralitic soil (>15 m thick; Colmet-Daage and Bernard, 1979,  
128 IRD soil map) built on Pleistocene andesitic pyroclastic deposits (Boudon et al., 1988). The regolith  
129 profile of Quiock Creek mainly consists of secondary minerals, with kaolinite/halloysite as the main  
130 mineral (95% by weight of the bulk soil; Buss et al., 2010) and is highly depleted in mobile elements  
131 (Buss et al., 2010; Clergue et al., 2015). All these features are characteristic of a highly weathered soil.  
132 Despite the high nutrient depletion of these soils, they support a tropical rainforest (Rousteau et al.,  
133 1996; Van Laere et al., 2016). This rainforest is characterized by three dominant species: *Amanoa*  
134 *caribaea* Kr. and Urb. (Phyllanthaceae), *Tapura latifolia* Benth (Dichapetalaceae) and *Dracryodes excelsa*  
135 *Vahl* (Burseraceae). The litter flux produced by this oldgrowth rainforest was determined each two  
136 weeks using 10 litter traps of one square meter size each monitored for two years (2011–2013). Annual  
137 litter flux reached 7.8 t.ha<sup>-1</sup>.yr<sup>-1</sup> of dry matter (Rousteau, 2014), comparable to the rainforest flux in  
138 northeastern Puerto Rico (Weaver and Murphy, 1990). The litter production is seasonal, with the  
139 highest litter fall occurring in June-August (40% of the annual litter fall). Finally, the biomass measured  
140 on site is equal to 376.8 t.ha<sup>-1</sup> (Rousteau, 2014).

141 Atmospheric inputs to the watershed come in two forms: wet deposits and dry deposits. Wet deposits  
142 consist of rainfall, while dry deposits include sea salt aerosols, Saharan dust and volcanic ash. Because  
143 of the location of the study site in the heart of the National Park, anthropic contributions are negligible.

144 One of the peculiarities of the Caribbean is the major input of Saharan dust (SD) up to 100 kg.ha<sup>-1</sup>.yr<sup>-1</sup>  
145 measured in Guadeloupe (J. Molinié, personal communication) which is consistent with amounts  
146 reported in the literature for the Caribbean basin (Glaccum and Prospero, 1980; Mahowald et al., 2006;  
147 Pett-Ridge et al., 2009; Yu et al., 2015). Throughout the Caribbean region, Saharan dust is deposited on  
148 the ground or on the canopy by rain. These dust deposits are mainly composed of mica/illite and quartz  
149 (Prospero et al., 1970; Glaccum and Prospero, 1980). For instance, in the Quiock Creek soil profile, the  
150 dull aspect of quartz, characteristic of aerial transport, suggests that Saharan dust is likely the source  
151 of quartz.



152 Another exogenous input to the top soil comes from the neighbouring volcano Soufriere Hills on  
153 Montserrat, located 86 km NW of Guadeloupe. It erupted on July 18, 1995, after a century of  
154 quiescence. Since 1995, six recognized ash fall episodes have impacted Guadeloupe ([www.mvo.ms](http://www.mvo.ms);  
155 activity report of OVSG 2000). The two most important events took place in September 1996 and  
156 February 2010 and produced ash-fall deposits on Guadeloupe that were thicker than 1 mm,  
157 corresponding to fluxes of about 50 to 100 g m<sup>-2</sup> of ash each (Cadelis et al., 2013; OVSG reports). The  
158 volcanic ashes were mainly composed of crystalline silica, mostly cristobalite (Baxter et al., 1999).

159 All these atmospheric sources (sea salt and dust) have been shown to highly impact soil in the  
160 Caribbean Basin (e.g. Borg and Banner, 1996; Herwitz et al., 1996; Muhs et al., 2009; Pett-Ridge et al.,  
161 2009; Opfergelt et al., 2012; Clergue et al., 2015; Dessert et al., 2015; Fries et al., 2019).

162

### 163 **3. Sample collection**

164 All compartments of the watershed were sampled between 2011 and 2013. Water samples consisted  
165 of rainfall, throughfall, soil solution and river water, and solid samples consisted of soil, rock, litter and  
166 dust.

#### 167 **3.1. Water samples**

168 We collected water samples during contrasting hydrological periods. River samples (QC, n=39) were  
169 collected manually once a month and more frequently during six field trips. Rainfall (RF, n=9), and  
170 throughfall (TF, n=48) were sampled during six field trips between 2011 and 2013. They both were  
171 collected in polypropylene bottles with a funnel located one meter above the ground in order to avoid  
172 contamination from soil particles. Rainfall were collected on site on the roof of the “Maison de la Forêt”  
173 (RF-MF) or at the “Observatoire Volcanologique et Sismologique de Guadeloupe” (RF-OVSG). These  
174 samples correspond to several rainfall events cumulated over 1 to 5 days, depending on the sample.  
175 Throughfall waters were collected by three collectors randomly positioned in the Quiock Creek  
176 catchment (collector n°1 to n°3) and were sampled every 1 to 5 days, depending on the sample. Soil

177 solutions (SS) were sampled using nested tension lysimeters equipped with porous ceramic cups  
178 located between 15 and 823 cm in depth (Buss et al., 2010). These soil solutions were sampled each  
179 month and the sampling dates selected for isotopic measurement were 10-26-2012 and 06-28-2013.  
180 Conductivity and pH were measured on site for the Quiock Creek samples and after collection at the  
181 OVSG for the other samples. For alkalinity measurements, non-filtered 20 mL aliquots were used. The  
182 remaining water was filtered through 0.2  $\mu\text{m}$  cellulose filters and divided into two subsamples. One  
183 non-acidified aliquot was stored in polypropylene bottles and used for anion and silica analyses. The  
184 second aliquot was acidified (2%  $\text{HNO}_3$ ), stored in polypropylene bottles and used for cation, trace  
185 element and Sr isotope measurements. Strontium isotopic compositions were measured on 26  
186 selected samples corresponding to 3 samples of rainfall, 6 samples of throughfall, 4 samples of stream  
187 water and 13 samples of soil solution.

### 188 3.2. Solid samples

189 We analyzed andesite (04-GW-12) and basalt (00-GU-44 and 03-GU-75) samples from Samper et al.  
190 (2007) to characterize the unweathered parent rock (Fig. 1). A soil profile (n=27) was collected with a  
191 hand-auger down to 12.5 m depth during lysimeter installation in 2007 (Buss et al., 2010). We collected  
192 Saharan dust during a dust event deposit on July 1 and 5, 2013. It corresponds to rainfall particulate  
193 matter obtained by filtration on a 0.2  $\mu\text{m}$  cellulose filter. The filter was then dried and stored in a Petri  
194 dish. We also analysed 5 samples of litter, collected during one of the most productive periods  
195 (between June 10 and 24, 2011). "Leaves 1" corresponds to a mixture of litter from different species  
196 (collected in the same litter collector) whereas "Leaves 2" corresponds to *Amanoa caribaea* leaves only.  
197 "Branches 1" and "Branches 2" correspond to a mixture of branches collected in two different litter  
198 collectors. Finally, "Roots" (April 2012) are from *Amanoa caribaea*.

199

## 200 4. Analytical methods and flux calculation

### 201 4.1. Solid sample digestion

202 Soil and litter samples were first dried in an oven (60°C) for 3 days and crushed in an agate mortar (soil  
203 samples) or in a ball mill (litter samples). Soil and rock samples (100 mg) were dissolved in a mixture of  
204 HNO<sub>3</sub>/HF for 2 days at 120 °C. This mixture was then evaporated and dissolved twice in 2mL 6N HCl  
205 and finally stored in 1N HCl. Saharan dust samples collected on cellulose filters underwent an initial  
206 step of filter digestion in diluted 5N HNO<sub>3</sub> at 100°C for 1 day and were treated with the same protocol  
207 as for rock and soil samples. Litter samples (500 mg) were first dissolved in 10 mL of 16N HNO<sub>3</sub> in 50  
208 mL Teflon beakers for 24h at room temperature and then digested in a DigiPREP block digestion system  
209 for another 24h at 90°C. After an evaporation step at 60°C, the samples were dissolved once more in  
210 16N HNO<sub>3</sub> for 24h at 90°C. Finally, after evaporation, 0.5 mL HF/ H<sub>3</sub>BO<sub>3</sub> were added to fully dissolve the  
211 phytoliths. These samples were then stored in 50 mL of 2N HNO<sub>3</sub>.

#### 212 4.2. Major and trace element analyses

213 We measured base cations by ICP AES (ThermoFisher iCap 6200 series) at the Institut de Physique du  
214 Globe de Paris (IPGP). Anions in soil solutions were measured with a Dionex DX 120 chromatograph at  
215 the IPGP and the OVSG/IPGP. We quantified Sr concentrations using HR-ICP-MS (Element II  
216 ThermoScientific) at IPGP. The limit of quantification is 20 ppt and repeated measurement of the SLRS-  
217 5 standard lead to an external reproducibility of 3%.

218 Major elements in solid samples were measured by ICP AES (ThermoFisher iCap 6200 series) at the  
219 IPGP. Trace elements in solid samples were measured with an ICP-MS X series mass spectrometer  
220 (Thermo Electron) at the Institut des Sciences de la Terre de Paris (ISTeP) of the Université Pierre et  
221 Marie Curie. We followed the bracketing method using BEN rock standards for bracketing and BEN,  
222 GSN and LKSD2 to validate the measurements. The standard values measured in this study are  
223 compared to the certified values to validate the data (Table S1). We measured major element and Sr  
224 concentrations in litter samples using a calibration curve, such as for water samples. The accuracy of  
225 this measurement was checked by repeated analyses of a vegetation standard NIST SRM 1515.

#### 226 4.3. Sr and Nd purification

227 Sr and Nd isotope analyses were performed on the Neptune multicollector ICP-MS (Thermo Scientific)  
228 at IPGP using the Apex sample inlet system. A prior step of purification is needed in order to avoid a  
229 matrix effect during measurement.

230 The separation of Sr from the rest of the matrix was automated for water samples with HPIC (High  
231 Performance Ion Chromatography) as fully described in Meynadier et al. (2006). Sr contained in  
232 solubilized solid samples was separated from the rest of the matrix on Sr-Spec resin in 3N HNO<sub>3</sub> media.  
233 The elution step was performed with milliQ H<sub>2</sub>O. The resin was replaced after each separation to avoid  
234 memory effects. The purified solution of Sr was evaporated and dissolved in 0.05N HNO<sub>3</sub> for isotopic  
235 measurement. The minimum amount required for Sr isotopic measurement is 50-100 ng Sr in 1 mL.  
236 For Nd separation, a two-step procedure was applied (Richard et al., 1976; Caro et al., 2006). First, rare  
237 earth elements were separated from the rest of the matrix on TRU-Spec resin in 2N HNO<sub>3</sub> media. Then,  
238 the separation of the different rare earth elements was performed on Ln-spec resin in 0.25N HCl media.  
239 The purified Nd solution was evaporated and dissolved in 3N HNO<sub>3</sub> for isotopic measurement. The  
240 minimum amount required for Nd isotopic measurement is 50 ng Nd in 1 mL.

#### 241 4.4. Sr and Nd isotope analyses

242 For Sr, the method used corresponds to the measurement of 100 ratios of 4 second iteration time per  
243 sample or standard. Mass interferences with Kr (84 and 86) were corrected using <sup>83</sup>Kr/<sup>84</sup>Kr and <sup>83</sup>Kr/<sup>86</sup>Kr,  
244 measured in the blank at the beginning of each session. Mass interference with Rb (87) was corrected  
245 using the <sup>85</sup>Rb/<sup>87</sup>Rb ratio measured in a low concentrated Rb solution (2.5 ppb) after Kr measurement.  
246 The mass bias was corrected using the <sup>86</sup>Sr/<sup>88</sup>Sr ratio of 0.1194. Each measurement was normalized to  
247 the NBS value certified as <sup>87</sup>Sr/<sup>86</sup>Sr = 0.71025, which was measured every 3 samples. The standard error  
248 based on repeated measurement of the NBS standard was 2% = 0.00001.

249 The εNd was calculated relative to the CHUR (Chondritic Uniform Reservoir) as follows:

$$250 \quad \epsilon Nd = \left( \frac{{}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{sample}}}{{}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{CHUR}}} - 1 \right) \times 10000 \quad (1)$$

251 The CHUR value is 0.512638 (Jacobsen and Wasserburg, 1980). The method used corresponds to the  
252 measurement of 100 ratios of 4 second iteration time per sample or standard. Mass interferences with  
253 Sm (144, 148 and 150) were corrected using  $^{147}\text{Sm}/^{144}\text{Sm} = 4.83870$ . The mass bias was corrected using  
254 the  $^{146}\text{Nd}/^{144}\text{Nd}$  ratio of 0.72190. Each measurement was normalized to the NIST value certified as  
255  $^{143}\text{Nd}/^{144}\text{Nd} = 0.511418 \pm 6 \cdot 10^{-6}$  (Caro et al., 2006), which was measured every 3 samples. The external  
256 error was  $0.18 \epsilon$  ( $2\sigma$ ).

#### 257 4.5. Cation flux and residence time calculation

258 Elemental fluxes were obtained by multiplying the water/matter flux by the mean concentrations or  
259 determined from the following equations. Water fluxes were determined in Clergue et al. (2015).  
260 Throughfall water flux is  $3079 \text{ mm yr}^{-1}$ . The Quiock Creek discharge ranges between 150 and  $400 \text{ L min}^{-1}$   
261 with a mean value of  $172 \text{ L min}^{-1}$ , corresponding to a runoff of  $1130 \text{ mm yr}^{-1}$ . Dry matter litter flux  
262 reaches  $7.8 \text{ t ha}^{-1} \text{ yr}^{-1}$  (Rousteau 2014).

263 Throughfall elementary fluxes ( $F_{\text{TF}}$ ) are described by equation 2, where  $F_{\text{EX}}$  is the flux of cations exuded  
264 by plants, and  $F_{\text{RF}}$  the rainfall flux.

$$265 \quad F_{\text{TF}} = F_{\text{EX}} + F_{\text{RF}} \quad (2)$$

266 The rainfall compartment is influenced by both sea salt aerosols ( $F_{\text{SW}}$ ) and Saharan dust leaching ( $F_{\text{SD}}$ )  
267 and  $F_{\text{RF}}$  is equal to:

$$268 \quad F_{\text{RF}} = F_{\text{SW}} - F_{\text{SD}} \quad (3)$$

269 We consider that the flux of Cl in throughfall ( $F\text{-Cl}_{\text{TF}}$ ) is exclusively from rainfall.  $F_{\text{RF}}$  is determined from  
270 the  $F\text{-Cl}_{\text{TF}}$  and  $(X/\text{Cl})$  rainfall ratios:

$$271 \quad F_{\text{RF}} = F\text{-Cl}_{\text{TF}} \times (X/\text{Cl})_{\text{RF}} \quad (4)$$

272 with  $\text{Na}/\text{Cl}_{\text{RF}} = 0.63$ ,  $\text{Ca}/\text{Cl}_{\text{RF}} = 0.09$ ,  $\text{Mg}/\text{Cl}_{\text{RF}} = 0.09$ ,  $\text{K}/\text{Cl}_{\text{RF}} = 0.05$ ;  $1000\text{Sr}/\text{Cl}_{\text{RF}} = 0.56$  (mean mass ratio  
273 determined from rainfall data; see following section)

274 and  $F_{EX}$  is equal to:

$$275 \quad F_{EX} = F_{TF} - F_{RF} \quad (5)$$

276 Assuming that all the Cl flux in throughfall has a sea salt origin, we used  $F\text{-Cl}_{TF}$  and  $X/Cl$  sea salt ratios  
277 ( $Na/Cl_{SW} = 0.86$ ,  $Ca/Cl_{SW} = 0.02$ ,  $Mg/Cl_{SW} = 0.10$ ,  $K/Cl_{SW} = 0.02$ ,  $1000Sr/Cl_{SW} = 0.16$  and mass ratio from  
278 Brewer et al., 1975) to determine  $F_{SW}$  for each cation:

$$279 \quad F_{SW} = F\text{-Cl}_{TF} \times (X/Cl)_{SW} \quad (6)$$

280 and  $F_{SD}$  is equal to:

$$281 \quad F_{SD} = F_{RF} - F_{SW} \quad (7)$$

282 The vegetation flux ( $F_{VEG}$ ) is described by equation 8, where  $F_{EX}$  is the flux of cations exuded by plants,  
283 and  $F_{LF}$  the litter fall flux.

$$284 \quad F_{VEG} = F_{EX} + F_{LF} \quad (8)$$

285 The standing stock of cations in the forest (Stock) is determined from biomass ( $376.8 \text{ t}\cdot\text{ha}^{-1}$ ; Rousteau  
286 et al., 2014) and mean cationic concentrations into plants. The residence time ( $T_{res}$ ) of each element in  
287 the vegetation is determined by the following equation:

$$288 \quad T_{res} = \text{Stock} / F_{VEG} \quad (9)$$

289

## 290 **5. Results**

291 All the compartments of the catchment were characterized for their element concentrations and  
292 isotopic compositions. Table 1 presents results of major elements and Sr concentrations only in water  
293 samples for which Sr isotopic composition was measured. A full list of the concentrations of major  
294 cations we have used in this paper are taken from Clergue et al. (2015) of which the range in

295 concentrations, means and median values are summarized in Table 2. New trace element and isotopic  
296 compositions for solid samples are presented in Table 3.

### 297 5.1. Atmospheric samples

298 The pH of rainfall ranged from 4.1 to 5.7 (Clergue et al., 2015). The major cation in rainfall was Na,  
299 whose concentration varied between 41 and 165  $\mu\text{mol/L}$ . The mean concentration of K, Ca and Mg  
300 were 4, 7 and 12  $\mu\text{mol/L}$ , respectively. Sr concentration ranged from 9 and 39 nmol/L (Table 2S) and Sr  
301 isotopic composition varied between 0.70841 and 0.71029 (Fig. 2). These data were in agreement with  
302 previously published data in the Puerto Rican and Guadeloupean rainforests (Gioda et al., 2013;  
303 Dessert et al., 2015).

304 The concentrations of major elements in Saharan dust collected in Guadeloupe, were higher than those  
305 in the soil samples (except for Al and Fe; see Clergue et al., 2015) and lower than in the rock samples  
306 (Table 3). The average values for Ca (1830 ppm), Na (940 ppm) and K (5300 ppm) were lower than those  
307 reported in the literature for dust samples collected in West Africa (> 2600 ppm for Ca and Na, > 9800  
308 ppm for K; Moreno et al., 2006). Mg concentrations around 5200 ppm were in the range of those  
309 reported in the literature (> 4600 ppm; Moreno et al., 2006). The concentration of Sr concentration  
310 was around 50 ppm, 1.5 to 5 times lower than the values reported in the literature for dust samples  
311 (Moreno et al., 2006) and 10 times higher than soil Sr concentrations. Concerning Sr isotopic data, the  
312 dust samples were the most radiogenic of all samples collected on the site. The isotopic signature,  
313 around 0.7187, is consistent with previous estimations and measurements (Biscaye et al., 1974;  
314 Grousset et al., 1988; Grousset et al., 1992; Rognon et al., 1996) which gave a value of 0.7179 for  
315 Saharan dust in the Caribbean (Pett-Ridge et al., 2009b).

### 316 5.2. Throughfall samples

317 The pH of the throughfall was slightly higher than that of the rainfall, ranging from 5.1 to 6.3 (Clergue  
318 et al., 2015). The major cation in the throughfall was Na, whose concentration varied between 17 and  
319 264  $\mu\text{mol/L}$  (Table 2). Mean concentration of K, Ca and Mg were 39, 17 and 17  $\mu\text{mol/L}$  respectively. Sr

320 concentration ranged from 8 and 136 nmol/L (Table S2) and Sr isotopic composition varied between  
321 0.70953 and 0.70978 (Table 1, Fig. 2). These data were in agreement with the ranges published in the  
322 literature for tropical rainforests (e.g. Forti and Neal, 1992; McDowell, 1998).

323 The K/Cl ratio in throughfall was ten times higher than the K/Cl ratio in rainfall, attesting to a strong  
324 enrichment of K in throughfall (Fig. 3). To a lesser extent, Sr, Ca and Mg were also enriched in  
325 throughfall. The enrichment of throughfall compared to rainfall indicates that these elements are  
326 partially released by plants. This exudation process has been largely described in the literature  
327 (Vitousek and Sanford, 1986; Dezzio and Chacon, 2006).

328 Throughfall is slightly more radiogenic than sea salt (Fig. 2). This observation can be explained by a  
329 mixture in throughfall of sea salt Sr and Sr from Saharan dust leachates or plant exudates. It is  
330 impossible to distinguish between the two latter sources only on the basis of Sr isotopes.

### 331 5.3. Litter samples

332 Major element concentrations in litter samples were very different from concentrations in the other  
333 solid samples (Table 3). The most abundant element in the litter was Ca, with concentrations ranging  
334 between 1 and 3%. This is more than 100 times the Ca concentration in the soil and is similar to the  
335 parent rock Ca concentration. The K concentration, intermediate between 0.1 and 1.1%, and the Mg  
336 concentration, ranging between 0.2 and 0.6%, were also higher than the K and Mg concentrations in  
337 the soil. These concentrations are consistent with values in the literature for litter samples (e.g. Liao et  
338 al., 2006; Riotte et al., 2014). Similarly, the Sr concentration in the litter varied from 80 to 200 ppm,  
339 which is one or two orders of magnitude higher than the Sr concentration in the soil (up to 13 ppm).  
340 The isotopic signature of the litter varied between 0.70966 and 0.71019 (Fig. 2). This signature differed  
341 from the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of local bedrock, but was similar to that of samples of vegetation growing on  
342 highly depleted volcanic soil as reported in the literature (Whipkey et al., 2000; Stewart et al., 2001;  
343 Chadwick et al., 2009).

### 344 5.4. Soil solution samples



345 The soil solutions were acidic, with pH values ranging from 4.2 to 5.4. This can be explained by the low  
346 cation content, which plays a key role in neutralizing the acidity of soil solutions. Na and Cl were the  
347 most abundant elements, with mean concentrations of 179  $\mu\text{mol/L}$  and 199  $\mu\text{mol/L}$ , respectively (Table  
348 2). Average Mg, K and Ca concentrations were 24, 5 and 3 respectively. Sr concentrations in soil  
349 solutions ranged between 7 and 101 nmol/L (Table S2), and isotopic composition varied between  
350 0.70996 and 0.71097 (Table 1). Soil solutions were more radiogenic than local rocks and sea salts,  
351 attesting to an exogenous source of Sr in the soil solution (Fig. 2). In superficial soil horizons (up to 90  
352 cm), Sr concentrations were systematically higher than in the deeper horizons (mean value of 53 and  
353 16 nmol/L respectively). To a lesser extent, Ca and Mg were also slightly higher in superficial horizons  
354 than in the deepest. On the contrary, K concentrations were constant over the entire soil profile.

355 Fig. 3 shows the cation concentrations normalized to Cl for all dissolved samples. Na/Cl ratios were  
356 relatively homogeneous in all water samples and close to the sea salt ratio. In general, soil solutions  
357 were depleted in nutrient cations compared to throughfall. This was particularly true for K, which was  
358 probably due to efficient K uptake by plant root systems and microorganisms in the first few  
359 centimeters of soil. As previously noted, soil solutions of superficial horizons were slightly enriched in  
360 Sr, Ca and Mg compared to deeper soil solutions, which indicates a potential atmospheric source of  
361 cations. It was even observed that the surface solutions were sometimes enriched in Mg with respect  
362 to throughfall.

### 363 5.5. Soil and rock samples

364 The soil profile is characterized by a very high proportion of secondary minerals (95% wt. of secondary  
365 minerals and 70 %wt. of kaolinite and halloysite; Buss et al., 2010). The chemical composition of the  
366 soil indicated that it was highly depleted in mobile cations compared to unweathered rock in  
367 Guadeloupe (Table 3). For example, the Ca concentration varied from 23 to 306 ppm, which  
368 corresponds to about 100% depletion compared to volcanic rock, which contains 4.85% to 7.45% of Ca.  
369 Sr concentration in soil varied between 2 and 13 ppm and was very low compared to unweathered rock

370 (Table 3). To correct for dilution effects and volume change, Sr concentration in the soil samples was  
371 normalized to an immobile element (Ti). Fig. 4 presents mass transfer coefficient ( $\tau$ ) values for Sr:

$$372 \tau_{\text{Sr}} = \left[ \frac{(\text{Sr}/\text{Ti})_{\text{soil}}}{(\text{Sr}/\text{Ti})_{\text{rock}}} \right] - 1 \quad (10)$$

373  $\tau_{\text{Sr}}$  was negative, representing the depletion of Sr in soil compared to unweathered rock. Sr had a  
374 depletion rate of around  $98 \pm 2\%$ . The soil profile was systematically less depleted in its first 15 cm and  
375 at depths of 305 cm, 427 cm, 1219 cm and 1250 cm. These depths corresponded to horizons enriched  
376 in quartz and K-feldspar.

377 The entire profile was more radiogenic (0.70677 – 0.71527) than the unweathered volcanic rock  
378 sampled in Guadeloupe (Fig. 2 and 4). Quartz horizons exhibited the highest Sr isotopic ratio. This  
379 observation attests of exogenic source(s) of Sr to the soil. The  $\epsilon_{\text{Nd}}$  of the profile varied between -8.39  
380 and 2.44 and was different from unweathered rock ranging from 4.34 to 5.45 (Table 3). The Nd  
381 signature of the quartz horizons was very negative and attested again of an exogenic contribution of  
382 Nd to the profile.

### 383 5.6. Quiock Creek water samples

384 Conductivity in Quiock Creek ranged from 22.5 to 68.2  $\mu\text{S}$  and pH was acidic, ranging from 4.3 to 5.9  
385 (see Clergue et al., 2015). The major cation was Na, with a mean concentration of 234  $\mu\text{mol/L}$ . Mean  
386 concentrations of K, Ca and Mg were 5, 10 and 29  $\mu\text{mol/L}$ , respectively (Table 2). Sr concentration  
387 ranged from 35 and 70  $\text{nmol/L}$  (Table S2) and Sr isotopic composition varied between 0.70922 and  
388 0.70939.

389

## 390 6. Discussion

### 391 6.1. Vegetation cycling

#### 392 6.1.1. Impact of atmospheric deposits on the cation-nutrient budget

393 A key question is to what extent atmospheric deposition (wet and dry) impacts the forest's nutrient  
394 source. Many biogeochemical studies have shown that Sr, and particularly the  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic ratio,  
395 is a good tracer of ecosystem Ca sources (e.g. Capo et al., 1998; Bern et al., 2005; Belanger et al., 2012;  
396 Stille et al., 2012; Pett-Ridge et al., 2009a). K and Mg chemistry differs from Ca and Sr, but these  
397 lithophilic elements ultimately originate from the same sources. It is therefore possible to acquire some  
398 qualitative information on the cycles of these elements using Sr as a proxy.

399 The internal water cycle of our basin is relatively homogeneous from a Sr isotopic point of view, with a  
400 mean  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of 0.7097 for throughfall and 0.7104 for soil solutions (up to 12 meters deep; Fig.  
401 2). This shows that the dissolved pool of Sr is dominated by sea salts ( $^{87}\text{Sr}/^{86}\text{Sr}$  value equal to 0.7092),  
402 with a small influence of more radiogenic sources coming from Saharan dust leaching (Fig. 2). Note also  
403 that the Sr isotopic composition of litter of 0.7099 is in the range of that of the dissolved pool of Sr.  
404 This has already been well described in several studies, which have shown that vegetation exhibits the  
405 signature of the exchangeable Sr pool (e.g. Kennedy et al., 1998; Poszwa et al., 2002). Our results  
406 emphasize that atmospheric inputs contribute significantly to actively cycled Sr in the Quiock Creek  
407 basin. This hypothesis is consistent with the literature for similar contexts with highly depleted volcanic  
408 soils and strong atmospheric input (e.g. Whiptkey et al., 2000; Stewart et al., 2001; Chadwick et al.,  
409 2009; Schuessler et al., 2018). As an example, Pett-Ridge et al. (2009a) reported that the proportions  
410 of foliar Sr derived from sea salt and dust vary between 33% and 59% in the Puerto Rican rainforest.  
411 Similarly, in the Quiock Creek catchment, the  $^{87}\text{Sr}/^{86}\text{Sr}$  of litter is slightly more radiogenic than sea salt  
412 aerosols, emphasizing the influence of Saharan Sr on the ecosystem.

413 Based on the Sr isotopic composition, the weathering of local volcanic rock seems not to contribute in  
414 the plant Sr pool. It can be deduced that this is also true for the Ca, K and Mg pools. This observation  
415 is consistent with the idea that tropical forests developed on highly nutrient-depleted soils derive their  
416 nutrients from superficial atmospheric sources and from litter recycling (Jordan, 1982). It may be for  
417 this reason that surface soil solutions are slightly less depleted in cations than deeper solutions (>91  
418 cm; Fig. 3), reflecting the cationic inputs of throughfall, dust leaching/weathering and litter

419 remineralization. We also suggest that a little near-surface nutrient pool sustains the forest community  
420 of Quiock Creek.

421 Tropical forests are largely described as dependent on atmospheric sources, as observed in this study,  
422 but they are also known to recycle nutrients very efficiently. In the following section, we calculate the  
423 vegetation fluxes in the catchment and estimate the cation-nutrient recycling fluxes.

#### 424 6.1.2. Vegetation cycling at the soil-plant profile scale

425 Nutrients contained in plants are released during leaf excretion, litter leaching and litter  
426 decomposition. These bio-available elements are partly taken up by the vegetation from the soil  
427 solution and, therefore, are recycled. The proportion of nutrients recycled by plants depends on the  
428 amounts of available nutrients in the soil. As an example, the study by Holmden and Belanger (2010)  
429 shows that in a Boreal forest ecosystem, the proportion of recycled Ca from litter fall can represent  
430 from 80% and 90% of the annualized Ca inputs to the forest Ca cycle. The authors also note that the  
431 proportion of recycled Ca increases with decreasing weathering flux.

432 The internal-cationic fluxes calculated in the Quiock Creek catchment are summarized in Table 4 and  
433 Fig. 5. The vegetation flux (FVEG) varies between  $8.4 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  for Na and  $141.5 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  for Ca and  
434 represents the biggest elemental flux, except for Na. These results are in agreement with those of the  
435 review by Proctor et al. (1987) on primary rainforests and the study by McDowell (1998) in a Puerto  
436 Rican rainforest. For Ca, Mg and Sr, most of the vegetation flux is provided by litter fall. Concerning K,  
437 its flux provided by leaf excretion ( $41.2 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ) is twice as abundant as the litter fall flux ( $20.9 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ). This confirms that canopy leaching is the main source of K for the top soil (e.g. McDowell 1998;  
438 Riotte et al., 2014). Finally, the Na flux released by vegetation is relatively weak compared to rainfall  
439 flux. Na is not a nutrient for plants and it exhibits conservative behaviour when rainwater passes  
440 through the canopy. We note that for Sr, K, Ca, and, to a lesser extent Mg, plant recycling is highly  
441 efficient. The vegetation flux (FVEG) of Sr, K, and Ca are respectively, 20, 28 and 31 times greater than  
442 flux exported at the outlet of the basin (FQC). The vegetation flux of Mg is 3 times greater than the flux

444 exported by the Quiock Creek. This vegetation turnover efficiency is related to the degree of soil  
445 leaching and thus the nutrient pool derived from weathering minerals. In the tropical context of our  
446 study, it is not surprising to observe such a high turnover, because of the nutrient paucity in soils. The  
447 same range of solute turnover has been measured in the tropical watershed of Mule Hole in India  
448 (Riotte et al., 2014), where vegetation recycling fluxes represented 10-15 times the exported fluxes.  
449 More recently, in a tropical watershed in Sri Lanka, Schuessler et al. (2018) estimated this recycling  
450 factor between 2 and 7 for Mg, 3 and 10 for Ca and 9 and 29 for K.

451 Rousteau et al. (2014) have shown that vegetation in the Quiock Creek catchment is at equilibrium with  
452 respect to carbon. If the ecosystem is close to equilibrium, the nutrient fluxes taken up would be equal  
453 to FVEG (litter fall + leaf excretion). Numerous studies (e.g. Proctor, 1987) have shown that in tropical  
454 rainforests, decomposition is invariably rapid and that litter turnover takes less than a year. Even though  
455 we did not measure the decomposition rate of litter, our field observations confirm the speed of this  
456 process. From the standing stocks of cations in the forest and the annual-cationic FVEG fluxes (Table  
457 4), we determined the residence time ( $T_{res}$ ) of each element in the vegetation. The element presenting  
458 the shortest residence time in the vegetation is K, namely 16 years. This residence time is consistent  
459 with the 10 year period determined for K in Puerto Rican forest (McDowell 1998). This short residence  
460 time is justified by the fact that K is one of the most important elements for plant metabolism (as a  
461 catalytic agent for carbohydrate synthesis and the transfer of other nutrients) and is abundantly  
462 excreted by the leaves. For other elements, the residence time is slightly longer than those determined  
463 for K: 44.6 years for Sr, 44.9 for Ca, 45.7 for Mg and 48.2 for Na. These results highlight that vegetation  
464 recycling dominates the nutrient cycle, and, in light of the Sr isotopic signature, we can deduce that  
465 these nutrients have the particularity of being initially of atmospheric origin. Therefore, the Quiock  
466 Creek ecosystem is sustained by a little near-surface nutrient pool, which seems disconnected from the  
467 deep bedrock.

468 Our study is a first step in the characterization of biogeochemical cycles at the plant-soil scale in the  
469 Guadeloupean rainforest. To better characterize and quantify the nutrient recycling, it would be useful

470 for example, to study the seasonality of vegetation fluxes, investigate the N and P cycles, or use the  
471 isotopic fractionations of the elements involved in these biogeochemical processes (Ca, Mg, Si, B,...).

## 472 6.2. Soil cation-nutrient pool and weathering

### 473 6.2.1. Origin of cation-nutrient pool in the soil

474 After studying geochemical cycles at the plant-soil scale, it is necessary to study the soil profile to  
475 determine whether soil is still able to provide nutrients to the forest.

476 In the entire soil profile (Fig. 2), the Sr isotopic signature of the solid phase is high relative to rock,  
477 which is consistent with exogenous contributions of Sr. Fig. 6 presents the Sr isotopic signature of soil  
478 as a function of the Sr depletion rate ( $\tau_{Sr}$ ). The more the sample is rich in Sr, the more the soil Sr  
479 signature is radiogenic. This implies that the main source enriching the soil in Sr is more radiogenic than  
480 bedrock. The two potential sources of Sr that could enrich the soil in radiogenic Sr are: 1) Sea salts with  
481 a signature of 0.70917 (Dia et al., 1992) or 2) Saharan dust with a signature of 0.7187 (this study). This  
482 indicates that the soils are so leached that they have almost lost their original rock signature and have  
483 inherited an exogenous atmospheric signature. As previously discussed (Section 5.5) at depths of 15,  
484 30, 305, 427, 1219 and 1250 cm, the high  $^{87}Sr/^{86}Sr$  ratios ( $> 0.712$ ) are correlated with the highest  
485 primary mineral content (Quartz + Feldspar), which could correspond to different dust deposit  
486 horizons. All the other soil samples are not enriched in primary minerals but, nevertheless, are  
487 characterized by Sr isotopic ratios that indicate a mixture of bedrock and exogenous atmospheric  
488 sources. This indicates that Sr derived from atmospheric deposits is transferred to the soil exchangeable  
489 pool and may be incorporated into secondary minerals. This recycled Sr in the soil exchangeable pool  
490 could be taken up by plants and reinforces the hypothesis that the nutrients involved in the Quiock  
491 Creek ecosystem are of atmospheric origin. This strong atmospheric influence on the soil profile has  
492 been described in the study by Clergue et al. (2015), devoted to Li dynamic in the same catchment. The  
493  $\delta^7Li$  measured in the soil profile also indicated that atmospheric deposition (wet and dry) was a main  
494 source of Li in the bulk soil.

495 Because both wet and dry atmospheric deposits are sources of radiogenic Sr, it is not possible to  
496 discriminate between sea salt and Saharan sources in soil only on the basis of the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio.  
497 Coupled with Sr isotopes, Nd isotopes are useful for identifying the different contributions to the soil  
498 profile. For example,  $\epsilon\text{Nd}$  of rock ranges from 4.34 to 5.45 (Table 3) and  $\epsilon\text{Nd}$  of Saharan dust ranges  
499 from -10.5 to -14.5 (Grousset et al., 1992; Dia et al., 2006; Scheuven et al., 2013; McClintok et al.,  
500 2015). Sea salt exhibits an extremely low Nd concentration and Saharan dust input seems to be the  
501 only atmospheric contribution able to shift the isotopic composition of the soil. The Nd isotopic  
502 composition of soil varies between -8.39 and 2.71 and could be explained by a mixture of rock and dust  
503 endmembers. The quartz-feldspar horizons are characterized by very negative  $\epsilon\text{Nd}$ , indicating a strong  
504 Saharan Nd contribution. The amount of quartz and Nd isotopes provide two independent proxies of  
505 long-term dust additions to the soil profile. The superficial horizon (the first 30 centimetres) is a  
506 contemporary depositional surface while the other horizons attest to past deposition. The deepest  
507 quartz horizons (305, 427, 792 and 1219-1250 cm) may represent paleo-surfaces of dust deposits,  
508 interspersed between multiple events of volcanic deposition. The variable thickness of these horizons  
509 may be partly explained by a physical soil mixing process resulting from bioturbation.

510 In Fig. 7, the soil samples are plotted in a two-component mixing diagram for  $^{87}\text{Sr}/^{86}\text{Sr}$  versus  $\epsilon\text{Nd}$ . The  
511 curvature of the different mixing lines depends on the Sr/Nd ratio of each endmember. The dotted  
512 curve represents the mixing line between the unweathered rock endmember ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.7038$ ,  $\epsilon\text{Nd}$   
513 = 4.8, Sr/Nd = 17.9; from this study and the andesite Nd content of 16 ppm from Taylor and McLennan  
514 1985) and the Saharan endmember ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.7187$ ,  $\epsilon\text{Nd} = -11.5$ , Sr/Nd = 3.4; from this study and  
515 Grousset et al., 1998). This mixing line cannot explain the isotopic composition of the soil samples,  
516 indicating that dust is not mixed with fresh rock but with a Sr-depleted weathered rock. Four mixing  
517 lines with depleted rock endmembers are shown on Fig. 7. Their Sr/Nd ratios decreased from 10 to 0.1  
518 in the most depleted case.

519 If we first consider soil horizons enriched in quartz-feldspar, this mixing model confirms that these  
520 particular layers are strongly influenced by the dust endmember. It also emphasized that the

521 proportion of Saharan Sr in the soil decreases with depth, from 65% in the first 15 cm, to 20% in the  
522 deepest quartz-feldspar horizons. Finally, our results show that the degree of rock depletion also  
523 increases with depth. Indeed, surface horizons (15 and 30 cm) can be explained by mixing of dust with  
524 moderately depleted rock ( $10 < \text{Sr}/\text{Nd} < 5$ ) whereas the deeper horizons involve a mixture with a very  
525 depleted rock ( $\text{Sr}/\text{Nd} < 1$ ). The superficial volcanic rock signature is inherited from the deposit of volcanic  
526 ashes in Guadeloupe linked to very recent activity of neighbour volcano Soufriere Hills on Montserrat  
527 (Baxter et al., 1999; Cadelis et al., 2013). The accumulation of Saharan dust in the upper soil is also  
528 observed in quartz-diorite and volcanoclastic soils in Puerto Rico (McClintock et al., 2015), which is too  
529 far from the volcanic arc of the Lesser Antilles to be impacted by recent ash deposits. In Puerto Rico,  
530 the calculated soil dust content ranges between 0 and 8%, and Nd from dust in the upper 20 cm of soil  
531 ranges between 0 and 92% (McClintock et al., 2015), which is consistent with what we have observed  
532 in Guadeloupe.

533 If we now consider soil horizons non-enriched in quartz-feldspar, we note that their isotopic  
534 composition still reflects a slight influence of the dust endmember and that they have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios  
535 close to those of rainfall or that are slightly more radiogenic. The rock Sr isotope signature in these  
536 horizons has been completely overprinted by the atmospheric signature (rainfall and dust-derived Sr).  
537 The slight influence of dust on the Nd signature in the entire soil profile is clear evidence for the  
538 mobilization of dust-derived Nd and its redistribution in the soil. This is especially the case for the  
539 horizons at depths of 61 and 91 cm, which do not contain quartz and feldspar, but whose Sr and Nd  
540 isotope signatures are strongly influenced by the dust endmember (25 to 35%). It is possible that the  
541 Sr and Nd come from the leaching of Saharan minerals contained in the upper soil and that are  
542 reincorporated into the deeper soil levels. Even if Nd mobility is much lower than Sr mobility, it seems  
543 to increase under extreme leaching conditions. This mobilization of dust-derived Nd into highly  
544 weathered soil profiles has been observed in Puerto Rico, where the Nd isotope signature of Saharan  
545 dust has been observed noted at depths of 2 and 3 m (Pett-Ridge et al., 2009b). Our mixing model  
546 cannot explain all the data because of the influence of a third end-member, namely sea salt Sr derived



547 from rainwater. Because sea salts are not a significant source of Nd, the addition of sea salts does not  
548 influence the Nd isotopic composition of soil. Except for the dust horizons, andesitic and dust-derived  
549 Sr have been leached extensively and the sea salt Sr signature dominates in the entire profile.

550 In the Quiock Creek catchment, we observe a strong impact of atmospheric sources on the evolution  
551 of the chemical composition of the soil with depth and with time. Elements are continuously provided  
552 at the surface by fresh dust minerals on a highly depleted soil. Then, we observe a “reversed”  
553 weathering profile, with the horizon the most enriched in primary minerals (Saharan and volcanic) and  
554 having the strongest andesitic isotopic signature at the top of the soil profile. The impact of  
555 atmospheric sources on volcanic soil has been demonstrated in different tropical contexts. In a soil  
556 chronosequence in Hawaii, it was shown that the older the soils, the bigger the impact of atmospheric  
557 inputs (Kennedy et al., 1998; Whiptkey et al., 2000; Kurtz et al., 2001; Chadwick et al., 2009). Saharan  
558 dust was shown to affect soil composition in Puerto Rico (Pett-Ridge et al., 2009b; McClintock et al.,  
559 2015) and in Mount Cameroon (Dia et al., 2006). In Guadeloupe, Mg and Li isotopes reflect the  
560 influence of sea salt and dust on soil (Opfergelt et al., 2012; Clergue et al., 2015; Fries et al., 2019). Our  
561 study supports these prior observations and the need to fully account for these inputs when studying  
562 weathering and cation-nutrient cycling under tropical climatic conditions.

#### 563 6.2.2. Does chemical weathering provide elements to the catchment flux?

564 The comparison between rainfall fluxes and output flux from Quiock Creek provides information on  
565 processes taking place in the soil. Elementary budgets (output flux – input flux) are negative (Fig. 5).  
566 This may mean that the catchment stores cation nutrients in the soil or in the vegetation. However, it  
567 can also mean that for these elements, inputs from rainfall are underestimated. In fact, their mean  
568 concentrations in rain are highly variable and hard to estimate. In any case, it means that these  
569 elements are not enriched as they move through the soil and thus there is no weathering flux for these  
570 elements in the catchment. The Sr isotopic composition of Quiock Creek, which is close to the sea salt  
571 aerosol signature, supports the idea that the Sr comes from atmospheric input only (Fig. 5). The fact

572 that the Quiock Creek  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio is very different from the volcanic rock signature, indicates that  
573 the local rock no longer contributes to the weathering flux. The absence of weathering flux in the  
574 Quiock Creek catchment and its peculiar Sr isotopic composition are linked with the intense weathering  
575 of the soil, which is highly depleted in major elements (Clergue et al., 2015). This situation is peculiar  
576 in Guadeloupe, as in all other catchments studied, the weathering flux is equivalent, or even twice as  
577 high, as the rainfall flux (Dessert et al., 2015). This situation is also peculiar in the Caribbean, for  
578 example in Puerto-Rico, where numerous catchments are surveyed by the USGS. In the Puerto-Rican  
579 catchments, which are subject to the same climate and are covered by rainforest, soils are not as  
580 depleted as in Guadeloupe (e.g. Ferrier et al., 2010). The Rio Icacos basin in Puerto Rico is the site of  
581 the highest measured chemical solute fluxes for a catchment on granodiorite (Turner et al., 2003).

582 Clergue et al. (2015) showed that in this catchment, part of the weathering flux of Li originates from  
583 the main secondary mineral dissolution (i.e. kaolinite). Using the kaolinite dissolution rate (Carroll et  
584 al., 1988), the authors showed that kaolinite dissolution can explain the weathering flux of Li measured  
585 on site. The same calculations were made for Mg and Sr, using the average concentration of those  
586 elements in kaolinite. They show that kaolinite can provide Mg and Sr by dissolution, with a respective  
587 flux of  $0.012 \text{ kg}_{\text{Mg}} \text{ ha}^{-1} \text{ yr}^{-1}$  and  $0.002 \text{ kg}_{\text{Sr}} \text{ ha}^{-1} \text{ yr}^{-1}$ . However, these fluxes are much lower than the  
588 rainfall fluxes and consistent with the absence of measurable weathering flux for some major elements  
589 and Sr.

## 590 7. Conclusion

591 In the Quiock Creek catchment, a large range of  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\epsilon\text{Nd}$  values is observed despite the small  
592 scale of observation. Sr isotopic composition varies from 0.70677 in deep bulk soil samples to 0.71882  
593 in dust samples. The Nd isotopic composition varies just as much, between -8.39 in near-surface soil  
594 samples to 2.71 in deeper soil (732 cm depth). For the Saharan dust endmember, we used a mean  $\epsilon\text{Nd}$   
595 value of -11.5, determined from the literature. These large isotopic ranges, coupled with major and  
596 trace elements, have helped to better understand nutrient dynamic in tropical ecosystems in  
597 Guadeloupe.

598 The regolith profile was built on pyroclastic deposits, but, because of extreme leaching, it has almost  
599 lost its original andesitic signature and inherited an exogenous atmospheric signature. Only Saharan  
600 dust is likely to have influenced the Nd balance throughout the entire 12m-thick profile, while the Sr  
601 balance is influenced by both Saharan dust and sea salt aerosols. Sr and Nd isotopes, combined with  
602 mineralogy and mobile element concentrations in the regolith, argue for a multi-layered soil profile  
603 which has been built by successive volcanoclastic episodes, separated by periods of dust deposition.  
604 Deep horizons enriched in quartz-felspar are then interpreted as paleo-soils impacted by Quaternary  
605 Saharan dust deposits, whereas the near-surface soil horizon is currently impacted by fresh dust  
606 deposits. Our results show that only the chemical weathering of the near-surface fresh minerals can  
607 provide nutrients to the ecosystem, the deeper regolith (> 30 cm) being nutrient depleted. However,  
608 this dust weathering flux is too low to sustain the tropical forest ecosystem.

609 The cationic mass balance at the catchment scale and the Sr isotopic signature of Quiock Creek indicate  
610 that cation and Sr fluxes are of atmospheric origin. Thus, in this highly weathered catchment, the  
611 original andesitic bedrock no longer participates in the weathering fluxes of Quiock Creek. This  
612 conclusion leads us to question the notion of "bedrock" in catchment involving on a deep weathered  
613 formation. Is it still an andesitic bedrock or is it now a lateritic bedrock?

614 The dissolved Sr cycle at the catchment scale is mainly dominated by sea salts, with a small contribution  
615 of Sr coming from the Saharan dust leaching. The vegetation exhibits the signature of the dissolved  
616 pool of Sr, indicating that the nutrient pool derives from atmospheric inputs.

617 At the soil-plant scale, the cation-nutrient fluxes provided by vegetation (litter fall + leaf excretion) are  
618 major compared to input and output fluxes. The Ca, K, Sr and Mg fluxes each year in the vegetation  
619 are, respectively, 31, 28, 20 and 3 times greater than the exported fluxes at the outlet of the basin. The  
620 residence time of nutrients in vegetation is 16 years for K and close to 45 years for Sr, Ca and Mg. These  
621 results emphasize the highly efficient plant turnover that dominates the nutrient cycle in the Quiocq  
622 Creek catchment.

623 This first characterization of biogeochemical cycles in a Guadeloupean forest suggests that the forest  
624 community of Quiocq Creek is sustained by a small near-surface nutrient pool, disconnected from the  
625 deep andesitic bedrock.

626

## 627 **Acknowledgements**

628 This work has been financially supported by the INSU-CNRS and the Région Guadeloupe. Parts of this  
629 work were supported by the IPGP multidisciplinary program PARI and by the Paris–IdF region SESAME  
630 Grant no. 12015908. This work could not have been done without logistical support from 1) both INSU-  
631 CNRS observatories in Guadeloupe run by the IPGP: the Observatoire de l'Eau et de l'Erosion aux  
632 Antilles (OBSERA) and the Observatoire Volcanologique et Sismologique de Guadeloupe (OVSG) and,  
633 2) the Parc National de Guadeloupe (PNG). We thank in particular N. Liagre, M. Valy and F. Bastaraud  
634 from PNG for their assistance with the field work. We also thank X. Quidelleur and P. Lahitte (Paris Sud  
635 11 University) for providing us with rock samples from Guadeloupe. Major element concentrations  
636 from water and solid samples were measured by L. Cordier and C. Gorge. Thanks are extended to B.  
637 Caron from IsTeP-UPMC and J.-L. Birck, A. Cogez, L. Faure, A. Michel, M. Tharaud from IPGP for their  
638 analytical assistance. The equipment and services of the Laboratoire de biologie et de physiologie

639 végétale de l'Université des Antilles was used for the biological and ecological aspects of the study. We  
640 thank M. Daullet for his assistance in the laboratory. Special thanks to V. Virapin, who processed and  
641 weighed most of the collected litter samples and E. Mira who made and placed the litter traps. This  
642 study greatly benefited from comments from A. Cogez, M. Dulormne, F. Bussière, J. Molinié and R.  
643 Losno. We gratefully acknowledge Susan Johnson for her helpful comments on the original version of  
644 this paper. We have a special thought for our colleague and friend Agnès Michel, who passed away in  
645 2018. She was involved in the solid sample analyses and was always keen to develop new analytical  
646 experiments. We gratefully acknowledge two anonymous reviewers and the editor for their helpful and  
647 critical comments on the original manuscript. IPGP contribution # 4079.

648 **References**

- 649 Baxter, P.J., Bonadonna, C., Dupree, R., Hards, V.L., Kohn, S.C., Murphy, M.D., Nichols, A., Nicholson, R.  
650 A., Norton, G., Searl, A., Sparks, R.S.J., Vickers, B.P., 1999. Cristobalite in volcanic ash of Soufriere Hills  
651 Volcano, Monserrat. British West Indies. Science 283, 1142-5.
- 652 Bélanger, N., Holmden, C., Courchesne, F., Côté, B., Hendershot, W.H., 2012. Constraining soil mineral  
653 weathering  $87\text{Sr}/86\text{Sr}$  for calcium apportionment studies of deciduous forest growing on soils  
654 developed from granitoid igneous rocks. Geoderma 185-186, 84-96.
- 655 Bern, C.R., Townsend, A.R., Lang Farmer, G., 2005. Unexpected dominance of Parent-material strontium  
656 in a tropical forest on highly weathered soils. Ecology 86, 626-632.
- 657 Biscaye, P. E., Chesselet, R., Prospero, J. M., 1974. Rb-Sr,  $87\text{Sr}/86\text{Sr}$  isotope system as an index of  
658 provenance of continental dusts in the open Atlantic Ocean. Journal de Recherches Atmospheriques 8,  
659 819-829.
- 660 Borg, L.E., Banner, J.L., 1996. Neodymium and strontium isotopic constraints on soil sources in  
661 Barbados, West Indies. Geochim. Cosmochim. Acta 60 (21), 4193–4206.
- 662 Boudon, G., Dagain, J., Semet, M. P., Westercamp, D., 1988. Le massif volcanique de la Soufrière  
663 (département de la Guadeloupe, Petites Antilles). In: BRGM (Ed.), Carte géologique au 1/20.000ème.
- 664 Brewer, P. G., Riley, J. P., Skirrow, C. (1975) Chemical oceanography. Academic Press, New York, 1, 417.
- 665 Buss H.L., White A.F., Dessert C., Gaillardet J., Blum A.E., Sak P.B., 2010. Depth profiles in a tropical  
666 volcanic critical zone observatory: Basse-Terre, Guadeloupe. In: Torres-Alvarado, I.S., Birkklee, P. (Eds.),  
667 Proceedings of the 13th International conference on water-Rock Interaction. Taylor & FrancisGroup,  
668 London, UK.

669 Cadelis, G., Tourres, R., Molinie, J., Petit, R.H., 2013. Exacerbations of asthma in Guadeloupe (French  
670 West Indies) and volcanic eruption in Montserrat (70 km from Guadeloupe). *Revue des Maladies*  
671 *Respiratoires* 30, 203-214.

672 Capo, R.C., Stewart, B.W., Chadwick, O.A., 1998. Strontium isotopes as tracers of ecosystem processes:  
673 theory and methods. *Geoderma* 82, 197-225.

674 Caro, G., Bourdon, B., Birck, J.L., Moorbath, S., 2006. High-precision  $^{142}\text{Nd}/^{144}\text{Nd}$  measurements in  
675 terrestrial rocks: constraints on the early differentiation of the Earth's mantle. *Geochim. Cosmochim.*  
676 *Acta* 70 (1), 164–191.

677 Carroll-Webb, S. A., & Walther, J. V. (1988) A surface complex reaction model for the pH-dependence  
678 of corundum and kaolinite dissolution rates. *Geochim. Cosmochim. Acta* 52 (11), 2609-2623.

679 Chadwick, O. A., Derry, L. A., Vitousek, P. M., Huebert, B. J., Hedin, L. O., 1999. Changing sources of  
680 nutrients during four million years of ecosystem development. *Nature* 397(6719), 491-497.

681 Chadwick, O. A., Derry, L. A., Bern, C. R., Vitousek, P. M., 2009. Changing sources of strontium to soils  
682 and ecosystems across the Hawaiian Islands. *Chem. Geol.* 267(1), 64-76.

683 Clergue, C., Dellinger, M., Buss, H. L., Gaillardet, J., Benedetti, M.F., Dessert, C., 2015. Influence of  
684 atmospheric deposits and secondary minerals on Li isotopes budget in a highly weathered catchment,  
685 Guadeloupe (Lesser Antilles). *Chem. Geol.* 414, 28-41.

686 Colmet-Daage, F., Bernard, Z., 1979. Contribution à l'Atlas des départements d'Outre-mer: Guadeloupe.  
687 Carte des sols de la Guadeloupe, Grande-Terre, Marie-Galante. Carte des pentes et du modelé de la  
688 Guadeloupe, Grande-Terre, Marie-Galante. ORSTOM, Antilles.

689 Cuevas, E., Lugo, A.E., 1998. Dynamics of organic matter and nutrient return from litterfall in stands of  
690 ten tropical tree plantation species. *Forest Ecology and Management* 112, 263-279.

691 Dahms, D.E., 1993. Mineralogical evidence for eolian contribution to soils of late quaternary moraines,  
692 Wind River Mountains, Wyoming, USA. *Geoderma*, 59, 175-196.

693 Dessert, C., Lajeunesse, E., Lloret, E., Clergue, C., Crispi, O., Gorge, C., Quidelleur, X., 2015. Controls on  
694 chemical weathering on a mountainous volcanic tropical island: Guadeloupe (French West Indies).  
695 *Geochim. Cosmochim. Acta* 171, 216-237.

696 Dezzeo, N., Chacon, N., 2006. Nutrient fluxes in incident rainfall, throughfall, and stemflow in adjacent  
697 primary and secondary forests of the Gran Sabana, southern Venezuela. *Forest ecology and*  
698 *management* 234, 218-226.

699 Dia, A., Chauvel, C., Bulourde, M., Gérard, M., 2006. Eolian contribution to soils on Mount Cameroon:  
700 Isotopic and trace element records. *Chem. Geol.* 226, 232-252.

701 Dia, A. N., Cohen, A. S., O'Nions, R. K., Shackleton, N. J., 1992. Seawater Sr isotope variation over the  
702 past 300 kyr and influence of global climate cycles. *Nature* 356, 786 – 788.

703 Ferrier, K.L., Kirchner, J.W. Riebe, C.S., Finkel, R.C., 2010. Mineral-specific chemical weathering rates  
704 over millennial timescales: measurements at Rio Icacos, Puerto Rico. *Chem. Geol.* 277 (1-2), 101-114.

705 Forti, M. C., Neal, C., 1992. Hydrochemical cycles in tropical rainforests: an overview with emphasis on  
706 central Amazonia. *Journal of Hydrology* 134(1), 103-115.

707 Fries D.M., James, R.H., Dessert, C., Bouchez, J., 2019. The response of Li and Mg isotopes to rain events  
708 in a highly-weathered catchment. *Chem. Geol.* 519, 68-82.

709 Gaillardet J., Braud I., Hankard F., et al., 2018. OZCAR: The French Network of Critical Zone  
710 Observatories. *Vadose Zone Journal*, 17, doi:10.2136/vzj2018.04.0067.

711 Gioda, A., Mayol-Bracero, O.L., Scatena, F.N., Weathers, K.C., Mateus, V.L., McDowell, W.H., 2013.  
712 Chemical constituents in clouds and rainwater in the Puerto Rican rainforest: Potential sources and  
713 seasonal drivers. *Atmospheric Environment* 68, 208-220.



714 Glaccum, R.A., Prospero, J.M., 1980. Saharan aerosols over the tropical north Atlantic mineralogy. *Mar.*  
715 *Geol.* 37, 295–321.

716 Grousset, F. E., Biscaye, P. E., Zindler, A., Prospero, J., Chester, R., 1988. Neodymium isotopes as tracers  
717 in marine sediments and aerosols: North Atlantic. *Earth Planet. Sci. Lett.* 87, 367-378.

718 Grousset, F. E., Rognon, P., Coudé-Gaussen, G., Pédemay, P., 1992. Origins of peri-Saharan dust deposits  
719 traced by their Nd and Sr isotopic composition. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* 93, 203-212.

720 Grousset, F. E., Parra, M., Bory, A., Martinez, P., Bertrand, P., Shimmield, G., Ellam, R. M., 1998. Saharan  
721 wind regimes traced by the Sr-Nd isotopic composition of subtropical Atlantic sediments: last glacial  
722 maximum vs today. *Quaternary Science Reviews* 17, 395-409.

723 Guérin, A., Devauchelle, O., Robert, V., Kitou, T., Dessert, C., Quiquerez, A., Allemand, P., Lajeunesse,  
724 E., 2019. Stream-discharge surges generated by groundwater flow. *Geophysical Research Letters*, doi:  
725 10.1029/2019GL082291.

726 Hedin, L.O., Granat, L., Likens, G.E., Adri Buishand, T., Galloway, J.N., Butler, T.J., Rodhe, H., 1994. Steep  
727 declines in atmospheric base cations in regions of Europe and North America. *Nature* 367, 351-354.

728 Heartsill-Scalley, T., Scatena, F.N., Estrada, C., W.H. McDowell, W.H., Lugo, A.E., 2007. Disturbance and  
729 long-term patterns of rainfall and throughfall nutrient fluxes in a subtropical wetforest in Puerto Rico.  
730 *Journal of Hydrology* 333, 472-485.

731 Herwitz, S.R., Muhs, D.R., Prospero, J.M., Mahan, S., Vaughn, B., 1996. Origin of Bermuda's clay-rich  
732 Quaternary paleosols and their paleoclimatic significance. *J. Geophys. Res.-Atmos.* 101, 23389–23400.

733 Holmden, C., Bélanger, N., 2010. Ca isotope cycling in a forested ecosystem. *Geochim. Cosmochim. Acta*  
734 74, 995-1015.

735 Jacobsen, S.B., Wasserburg, G.J., 1980. Sm-Nd isotopic evolution of chondrites. *Earth Planet. Sci. Lett.*  
736 50 (1), 139-155.

737 Jordan, C.F, 1982. The nutrient balance of an Amazonian rain forest. *Ecological Society of America* 63  
738 (3), 647-654.

739 Kennedy, M. J., Chadwick, O. A., Vitousek, P. M., Derry, L. A., Hendricks, D. M., 1998. Changing sources  
740 of base cations during ecosystem development, Hawaiian Islands. *Geology* 26(11), 1015-1018.

741 Kurtz, A. C., Derry, L. A., Chadwick, O. A., 2001. Accretion of Asian dust to Hawaiian soils: isotopic,  
742 elemental, and mineral mass balances. *Geochim. Cosmochim. Acta* 65(12), 1971-1983.

743 Liao, J. H., Wang, H. H., Tsai, C. C., & Hseu, Z. Y., 2006. Litter production, decomposition and nutrient  
744 return of uplifted coral reef tropical forest. *Forest ecology and management* 235(1), 174-185.

745 Liu X-M., Rudnick R.L., McDonough W.F., Cummings M.L., 2013. Influence of chemical weathering on  
746 the composition of the continental crust: Insights from Li and Nb isotopes in bauxite profiles developed  
747 on Columbia River Basalts. *Geochim. Cosmochim. Acta* 115, 73-91.

748 Lloret E., C. Dessert, H. L. Buss, C. Chaduteau, S. Huon, P. Alberic, Benedetti M.F., 2016. Sources of  
749 dissolved organic carbon in small volcanic mountainous tropical rivers, examples from Guadeloupe  
750 (French West Indies) *Geoderma* 282, 129–138.

751 Mahowald, N.M., Muhs, D.R., Levis, S., Rasch, P.J., Yoshioka, M., Zender, C.S., Luo, C., 2006. Change in  
752 atmospheric mineral aerosols in response to climate: last glacial period, pre-industrial, modern, and  
753 doubled carbon dioxide climates. *J. Geophys. Res.-Atmos.* 111 (D10).

754 McClintock, M.A., Brocard, G., Willenbring, J., Tamaya, C., Porder, S., Pett-Ridge, J.C., 2015. Spatial  
755 variability of African dust in soils in a montane tropical landscape in Puerto Rico. *Chem. Geol.* 412, 69-  
756 81.

757 McDowell, W.H., 1998. Internal nutrient fluxes in a Puerto Rican rain forest. *Journal of Tropical Ecology*  
758 14, 521-536.

759 Meynadier, L., Gorge, C., Birck, J.-L., Allègre, C.J., 2006. Automated separation of Sr from natural water  
760 samples or carbonate rocks by high performance ion chromatography. *Chem. Geol.* 227, 26-36.

761 Moreno, T., Querol, X., Castillo, S., Alastuey, A., Cuevas, E., Herrmann, L., Mounkaila, M., Elvira, J.,  
762 Gibbons, W., 2006. Geochemical variations in aeolian mineral particles from the Sahara–Sahel Dust  
763 Corridor. *Chemosphere* 65, 261–270.

764 Muhs, D.R., Bush, C.A., Stewart, K.C., Rowland, T.R., Crittenden, R.C., 1990. Geochemical evidence of  
765 Saharan dust parent material for soils developed on Quaternary limestones of Caribbean and Western  
766 Atlantic islands. *Quaternary Research* 33, 157-177.

767 Muhs, D.R., Budahn, J., Prospero, J.M., Carey, S.N., 2007. Geochemical evidence for African dust inputs  
768 to soils of western Atlantic islands: Barbados, the Bahamas and Florida. *J. Geophys. Res.* 112, F02009.

769 Okin, G.S., Mahowald, N., Chadwick, O.A., Artaxo, P., 2004. Impact of desert dust on the  
770 biogeochemistry of phosphorus in terrestrial ecosystems. *Glob. Biogeochem. Cycles* 18, GB2005.

771 Opfergelt, S., Georg, R.B., Delvaux, B., Cabidoche, Y.M., Burton, K.M., Halliday, A.N., 2012. Mechanisms  
772 of magnesium isotope fractionation in volcanic soil weathering sequences, Guadeloupe. *Earth Planet.*  
773 *Sci. Lett.* 341–344, 176–185.

774 OVSG-IPGP Website. Volcano and seismic activity bulletin of La Soufrière of Guadeloupe.  
775 <http://www.ipgp.fr/fr/ovsg/observatoire-volcanologique-sismologique-de-guadeloupe>.

776 Pett-Ridge, J.C., 2009. Contributions of dust phosphorus cycling in tropical forests of the Luquillo  
777 Mountains, Puerto Rico. *Biogeochemistry* 94, 63-80.

778 Pett-Ridge, J.C., Derry, L.A., Barrows, J.K., 2009a. Ca/Sr and  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios as tracers of Ca and Sr cycling  
779 in the Rio Icacos watershed, Luquillo Mountains, Puerto Rico. *Chem. Geol.* 267(1), 32-45.

780 Pett-Ridge, J.C., Derry, L.A., Kurtz A.C., 2009b. Sr isotopes as a tracer of weathering processes and dust  
781 inputs in a tropical granitoid watershed, Luquillo Mountains, Puerto Rico. *Geochim. Cosmochim. Acta*  
782 73, 25-43.

783 Porder, S., Clark, D.A., Vitouseck, P.M., 2006. Persistence of rock-derived nutrients in the wet tropical  
784 forests of La Selva, Costa Rica. *Ecology* 87, 594-602.

785 Poszwa, A., Dambrine, E., Ferry, B., Pollier B., Loubet, M., 2002. Do deep tree roots provide nutrients  
786 to the tropical rainforest? *Biogeochemistry* 60, 97-118.

787 Proctor, J., 1987. Nutrient cycling in primary and old secondary rainforests. *Applied Geography* 7, 135-  
788 152.

789 Prospero, J.M., 1999. Long-term measurements of the transport of African mineral dust to the  
790 southeastern United States: implications for regional air quality. *Journal of Geophysical Research* 104,  
791 15917–15927.

792 Prospero, J.M., Bonatti, E., Schubert, C., Carlson, T.N., 1970. Dust in Caribbean atmosphere traced to  
793 an African dust storm. *Earth Planet. Sci. Lett.* 9, 287–290.

794 Prospero, J.M., Glaccum, R.A., Nees, R.T., 1981. Atmospheric transport of soil dust from Africa to South  
795 America. *Nature* 289, 570–572.

796 Schlesinger, W.H., 1997. *Biogeochemistry: an analysis of global change*. Academic Press, San Diego,  
797 California, USA.

798 Richard, P., Shimizu, N., Allègre, C.J., 1976.  $^{143}\text{Nd}/^{146}\text{Nd}$ , a natural tracer: an application tooceanic  
799 basalts. *Earth Planet. Sci. Lett.* 31 (2), 269–278.

800 Riotte, J., Marechal, J. C., Audry, S., Kumar, C., Bedimo, J. B., Ruiz, L., ... & Braun, J. J., 2014. Vegetation  
801 impact on stream chemical fluxes: Mule Hole watershed (South India). *Geochim. Cosmochim. Acta* 145,  
802 116-138.

803 Rognon, P., Coudé-Gaussen, G., Revel, M., Grousset, F. E., Pédemay, P., 1996. Holocene Saharan dust  
804 deposition on the Cape Verde Islands: sedimentological and Nd-Sr isotopic evidence. *Sedimentology*  
805 43, 359-366.

806 Rousteau, A., 1996. Structures, flores, dynamiques: réponses des forêts pluviales des Petites Antilles  
807 aux milieux montagnards. In: Guillaumet, J.L., Belin, M., Puig, H. (Eds.), *Phytogéographie Tropicale:*  
808 *Réalités et Perspectives*. ORSTOM, Paris, pp. 307–321.

809 Rousteau, A., Dessert, C., Dulormne, M., 2014. Bilan des travaux réalisés dans le cadre du projet  
810 “Vulnérabilité des écosystèmes insulaires guadeloupéens aux changements climatiques”. Région  
811 Guadeloupe funding.

812 Rousteau, A., Portecop, J., Rollet, B., 1994. Carte écologique de la Guadeloupe. Université Antilles-  
813 Guyane, ONF, Conseil Général, Parc National de la Guadeloupe, Guadeloupe.

814 Samper, A., Quidelleur, X., Lahitte, P., Mollex, D., 2007. Timing of effusive volcanism and collapse events  
815 within an oceanic arc island: Basse-Terre, Guadeloupe archipelago (Lesser Antilles Arc). *Earth Planet.*  
816 *Sci. Lett.* 258, 175–191.

817 Schuessler, J.A., von Blanckenburg, F., Bouchez, J., Uhlig, D., Hewawasam, T., 2018. Nutrient cycling in  
818 a tropical montane rainforest under a supply-limited weathering regime traced by elemental mass  
819 balances and Mg stable isotopes. *Chem. Geol.* 497, 74-87.

820 Soderberg, K., Compton, J.S., 2007. Dust as a Nutrient Source for Fynbos Ecosystems, South Africa.  
821 *Ecosystems* 10, 550-561.

822 Stewart, B. W., Capo, R. C., Chadwick, O. A., 2001. Effects of rainfall on weathering rate, base cation  
823 provenance, and Sr isotope composition of Hawaiian soils. *Geochim. Cosmochim. Acta* 65(7), 1087-  
824 1099.

825 Stille, P., Schmitt, A.-D., Labolle, F., Pierret, M.-C., Gangloff, S., Cobert, F., Lucot, E., Guéguen, F., Brioschi,  
826 L., Steinmann, M., Chabaux, F., 2012. The suitability of annual tree growth rings as environmental

827 archives: Evidence from Sr, Nd, Pb and Ca isotopes in spruce growth rings from the Strengbach  
828 watershed. *C. R. Geoscience* 344, 297-311.

829 Taylor, S.R. and McLennan, S.M., 1985. *The Continental Crust: Its Composition and Evolution*. Blackwell,  
830 Oxford, 1-312.

831 Turner, B. F., Stallard, R. F., & Brantley, S. L., 2003. Investigation of in situ weathering of quartz diorite  
832 bedrock in the Rio Icacos basin, Luquillo Experimental Forest, Puerto Rico. *Chem. Geol.* 202, 313-341.

833 Van Laere, G., Gall, Y. and Rousteau, A., 2016. The forest Ecosystems Observatory in Guadeloupe (FWI).  
834 *Caribbean Naturalist*, Special Issue n°1: 108-115.

835 Vitousek, P. M., Sanford, R. L., 1986. Nutrient cycling in moist tropical forest. *Annu. Rev. Ecol. Syst.* 17,  
836 137-167.

837 Weaver, P.L., Murphy, P.G., 1990. Forest structure and productivity in Puerto Rico's Luquillo Mountains.  
838 *Biotropica* 22 (1), 69–82.

839 Whipkey, C. E., Capo, R. C., Chadwick, O. A., Stewart, B. W., 2000. The importance of sea spray to the  
840 cation budget of a coastal Hawaiian soil: a strontium isotope approach. *Chem. Geol.* 168(1), 37-48.

841 White, W.M., Dupré, B., 1986. Sediment subduction and magma genesis in the Lesser Antilles: isotopic  
842 and trace element constraints. *J. Geophys. Res.* 91, 5927–5941.

843 White, W.M., Patchett, J., 1984. Hf-Nd-Sr isotopes and incompatible element abundances in island arcs;  
844 implications for magma origins and crust-mantle evolution. *Earth and Planetary Science Letters* 67,  
845 167-185.

846 Wilcke, W., Velescu, A., Leimer, S., Bigalke, M., Boy, J., Valarezo, C., 2017. Biological versus geochemical  
847 control and environmental change drivers of the base metal budgets of a tropical montane forest in  
848 Ecuador during 15 years. *Biogeochemistry* 136, 167-189.

849 Xu, Y., Losno, R., Dessert, C., Monna, F., Robert, V., Chane-Teng, J., 2017. Compositional analysis of  
850 Saharan dust input to Guadeloupe (FWI). Goldschmidt Conference, Paris, 2017 August 13-18,  
851 16d/3045.

852 Yu, H., Chin, M., Yuan, T., Bian, H., Remer, L.A., Prospero, J.M., Omar, A., Winker, D., Yang, Y., Zhang, Y.,  
853 Zhang, Z., Zhao, C., 2015. The fertilizing role of African dust in the Amazon rainforest: A first multiyear  
854 assessment based on data from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations.  
855 Geophys. Res. Lett. 42, 1984-1991.

856 Zahibo, N., Pelinovsky, E., Talipova, T., Rabinovich, A., Kurkin, A., Nikolkina, I., 2007. Statistical analysis  
857 of cyclone hazard for Guadeloupe, Lesser Antilles. Atmos. Res. 84, 13-29.

858 Figure caption

859 Fig. 1. The Quiock Creek catchment (16°17'N, 61°70'W) in the Guadeloupe National Park. The MF rain  
860 gauge is located in a clearing, on the roof of the “Maison de la Forêt”, and the OVSG rain gauge is  
861 located on the roof of the observatory. The soil profile was sampled during lysimeter installation (see  
862 Buss et al., 2010).

863 Fig. 2. Sr isotopic composition of dissolved and solid samples collected in the Quiock Creek catchment.  
864 Sea salt Sr isotopic ratio = 0.70917 (Dia et al., 1992); volcanic rock = 0.7038 (this study); Saharan dust  
865 = 0.7187 (this study).

866 Fig. 3. Molar ratios ( $\mu\text{mol}/\mu\text{mol}$  ratios, except for the Sr/Cl ratio expressed in  $\text{nmol}/\mu\text{mol}$ ) of dissolved  
867 load in the different compartments of the Quiock Creek catchment. RF: rainfall, TF: throughfall, SS: soil  
868 solution (above and below 91 cm depth), QC: Quiock Creek, SW: seawater/sea salt.

869 Fig. 4. Soil profiles. Quartz + feldspar content (from Buss et al., 2010), mass transfer coefficient ( $\tau$ ) for  
870 Sr in soil samples relative to bedrock, and isotopic signature of Sr and Nd.  $\tau$  is calculated considering Ti  
871 to be immobile and illustrates the extreme depletion of Sr in the soil compared to unweathered rock  
872 (values closed to -1).

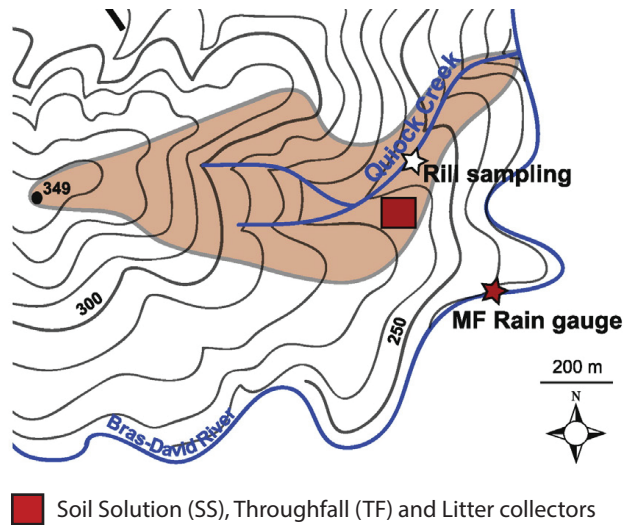
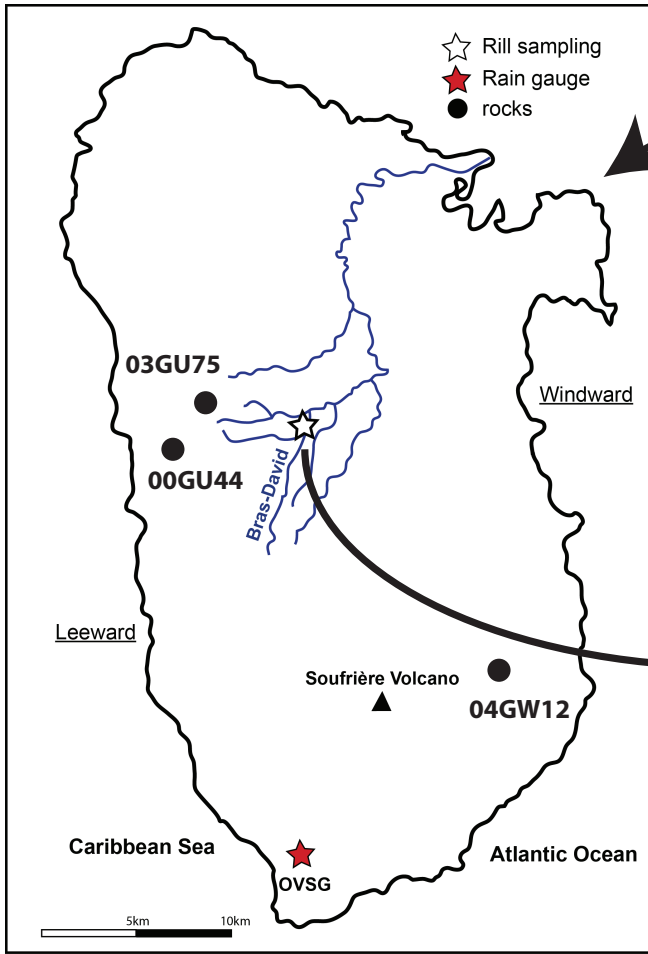
873 Fig. 5. Cation fluxes in the Quiock Creek catchment expressed in  $\text{kg}/\text{ha}/\text{yr}$ . FSW is the sea salt flux, FSD  
874 the Saharan dust leaching flux, FRF the rainfall flux, FTF the throughfall flux, FLF the litter fall flux, FEX  
875 the leaf excretion flux and FQC the Quiock Creek flux.

876 Fig. 6. Sr mass transfer coefficient as a function of Sr isotopic ratio in soil samples. Sea salt Sr isotopic  
877 ratio = 0.70917 (Dia et al., 1992); volcanic rock = 0.7038 (this study); Saharan dust = 0.7187 (this study).  
878 Samples enriched in quartz-feldspar are symbolized by white square.

879 Fig. 7. Two-component mixing diagram of  $\epsilon\text{Nd}$  vs. Sr isotopic ratio in soil samples. Horizon depths (cm)  
880 are added and samples enriched in quartz-feldspar are symbolized by white squares. The solid lines



881 represent different mixing curves between a bedrock (fresh and depleted) end-member and a “Saharan  
882 dust” end-member. The mixing proportions are expressed in % of dust Sr. The black dotted curve  
883 represents a mixture of fresh rock ( $^{87}\text{Sr}/^{86}\text{Sr} = 0.7038$ ,  $\epsilon\text{Nd} = 4.8$ ,  $\text{Sr}/\text{Nd} = 17.9$ ) and dust ( $^{87}\text{Sr}/^{86}\text{Sr} =$   
884  $0.7187$ ,  $\epsilon\text{Nd} = -11.5$ ,  $\text{Sr}/\text{Nd} = 3.4$ ). The other curves show a mixture between of depleted rock ( $^{87}\text{Sr}/^{86}\text{Sr}$   
885  $= 0.7038$ ,  $\epsilon\text{Nd} = 4.8$ ,  $\text{Sr}/\text{Nd}$  varying from 10 to 0.1 for the most depleted rock) and dust. The samples  
886 are explained by a mixture of Saharan dust and depleted rock. The soil horizons enriched in quartz-  
887 feldspar are strongly influenced by dust (20 to 65% of dust Sr). The near-surface horizon (first 30 cm) is  
888 the most impacted by dust and can be explained by the mixing of dust with moderately depleted rock.  
889 This emphasizes the contemporary, or very recent, deposition of both Saharan dust and volcanic ash in  
890 Guadeloupe (recent volcanic activity of nearby Montserrat). The deeper quartz-feldspar horizons are  
891 paleo-surfaces of dust deposits interspersed with volcanic deposits. With time, andesitic and dust-  
892 derived Sr has been leached extensively and the sea salt Sr signature dominates today in the entire  
893 profile.



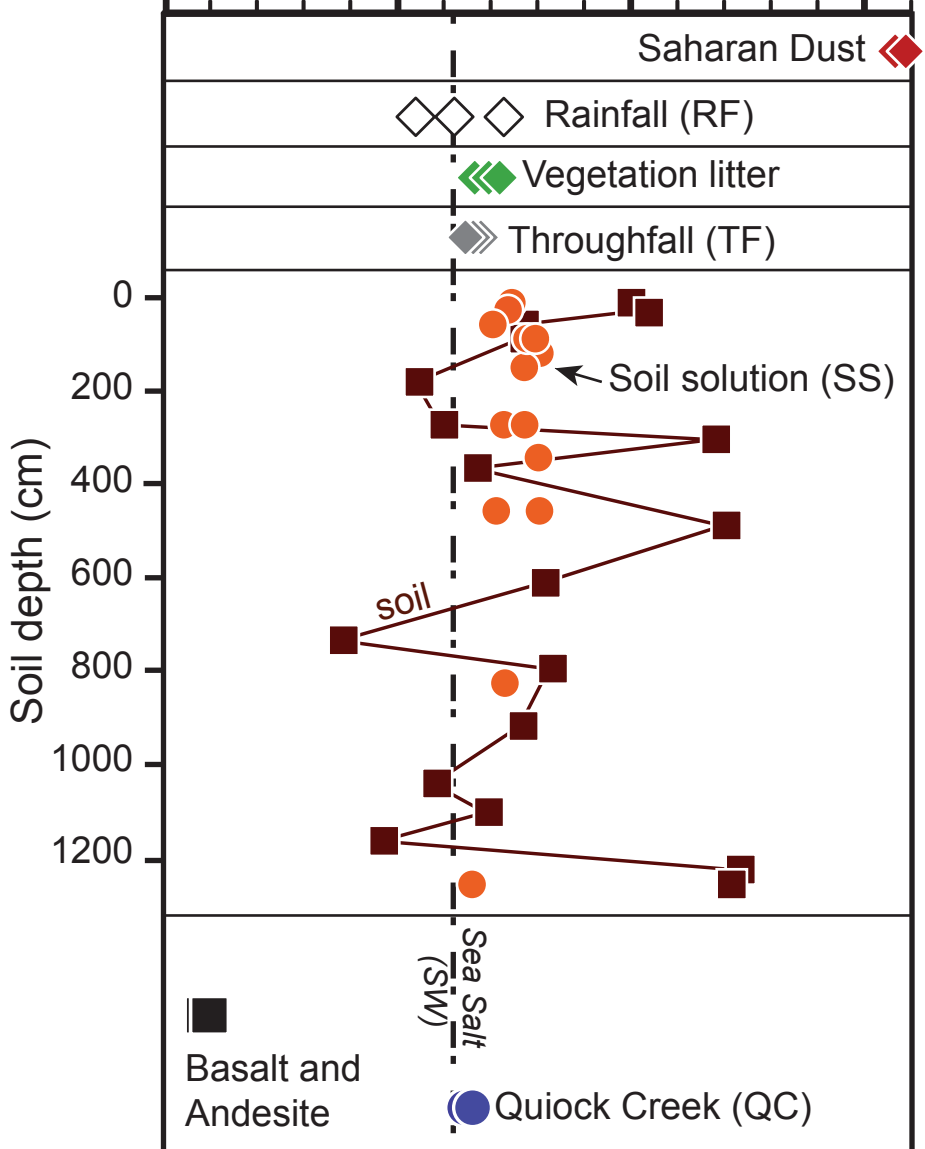
# $^{87}\text{Sr}/^{86}\text{Sr}$

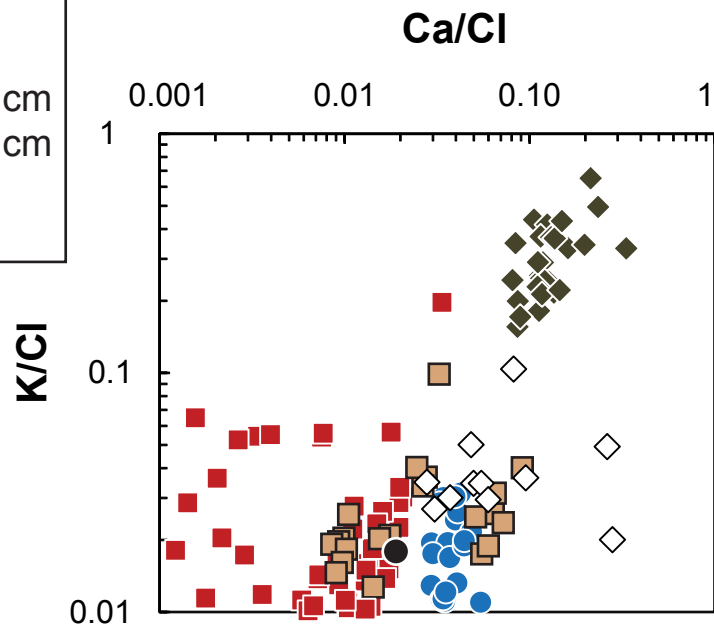
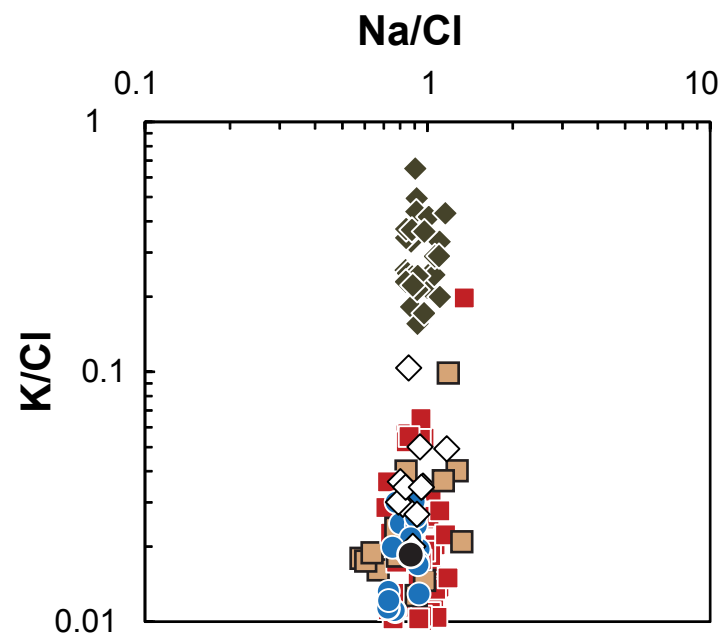
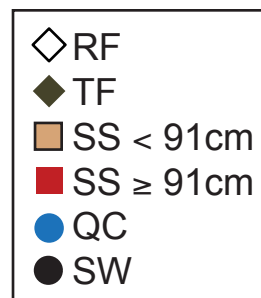
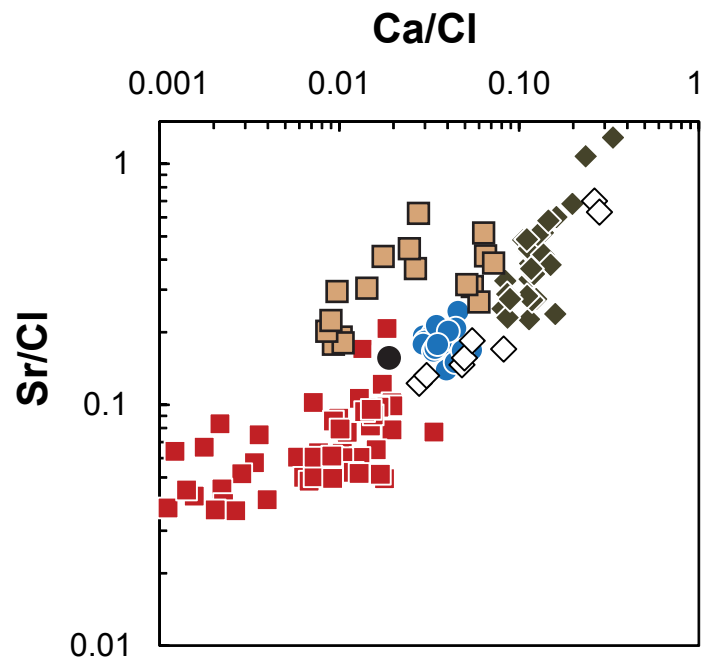
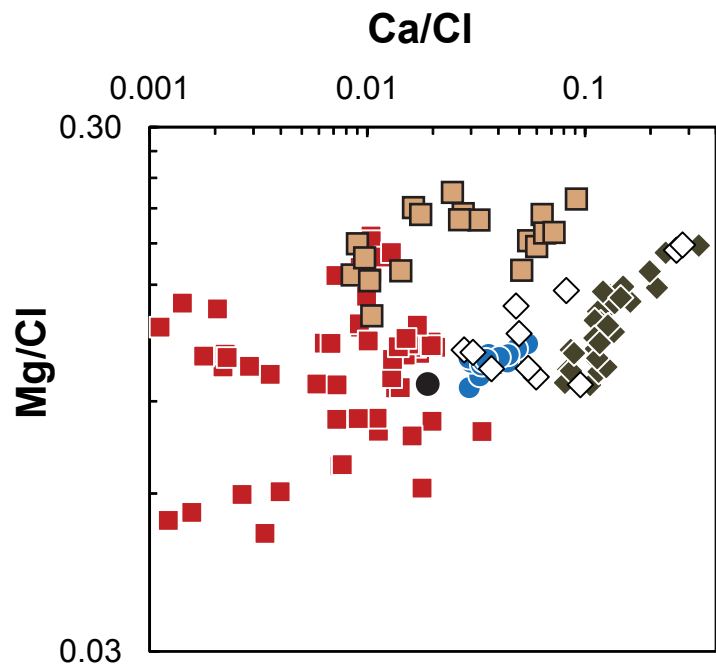
0.703

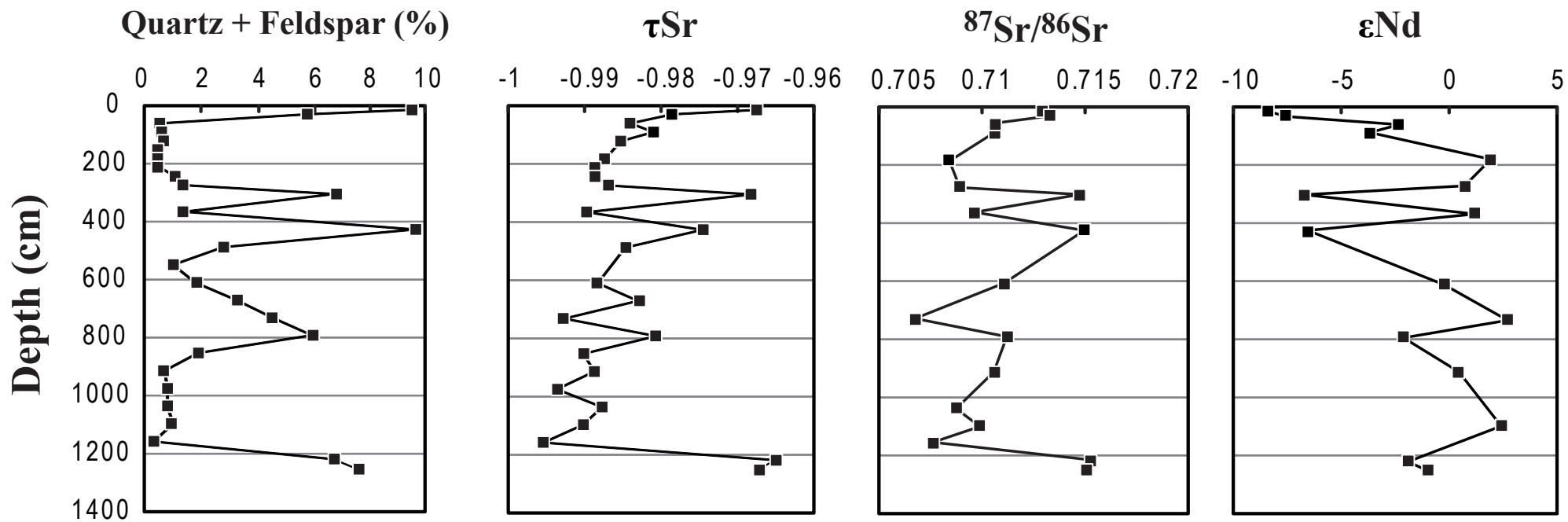
0.708

0.713

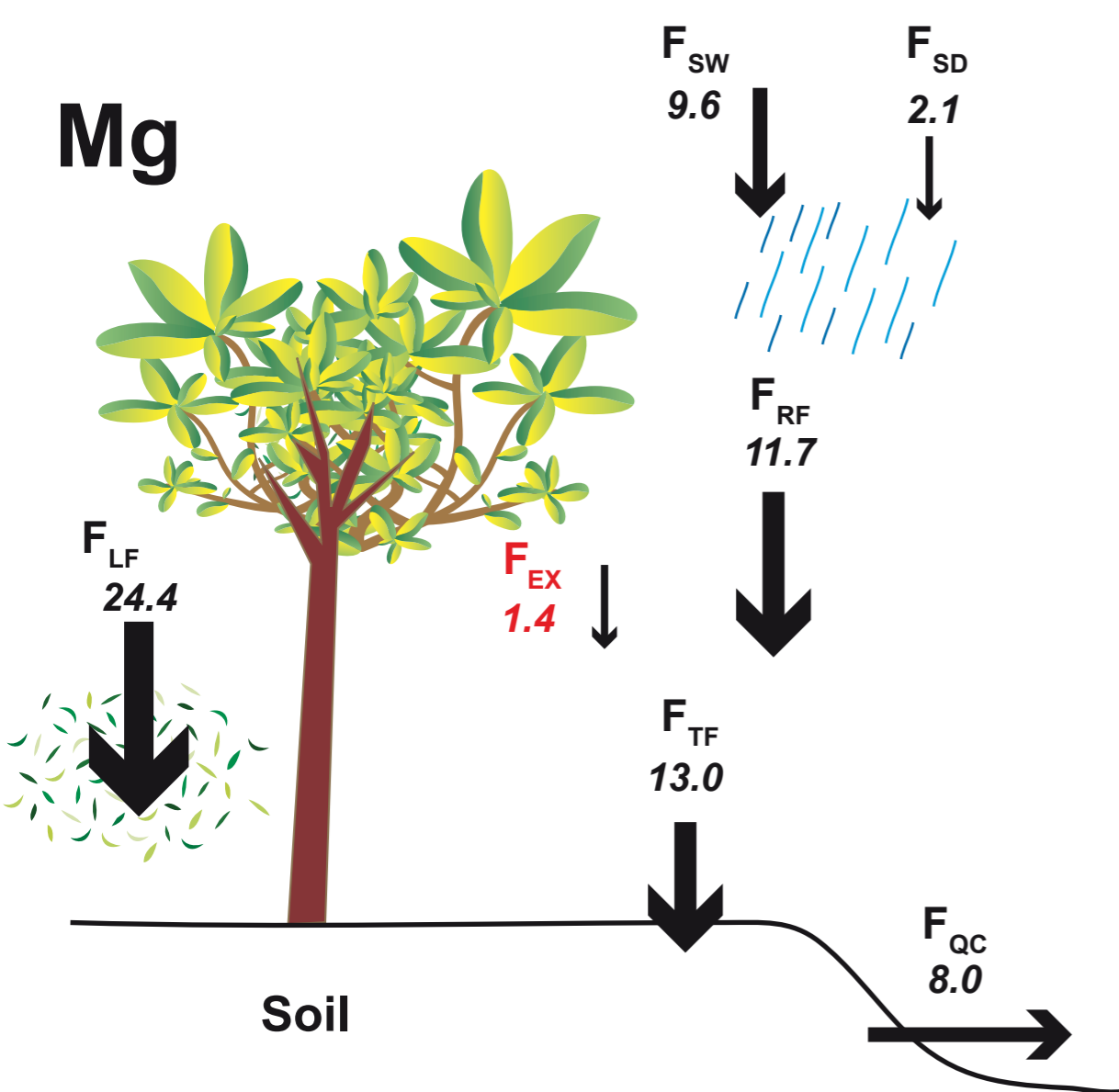
0.718



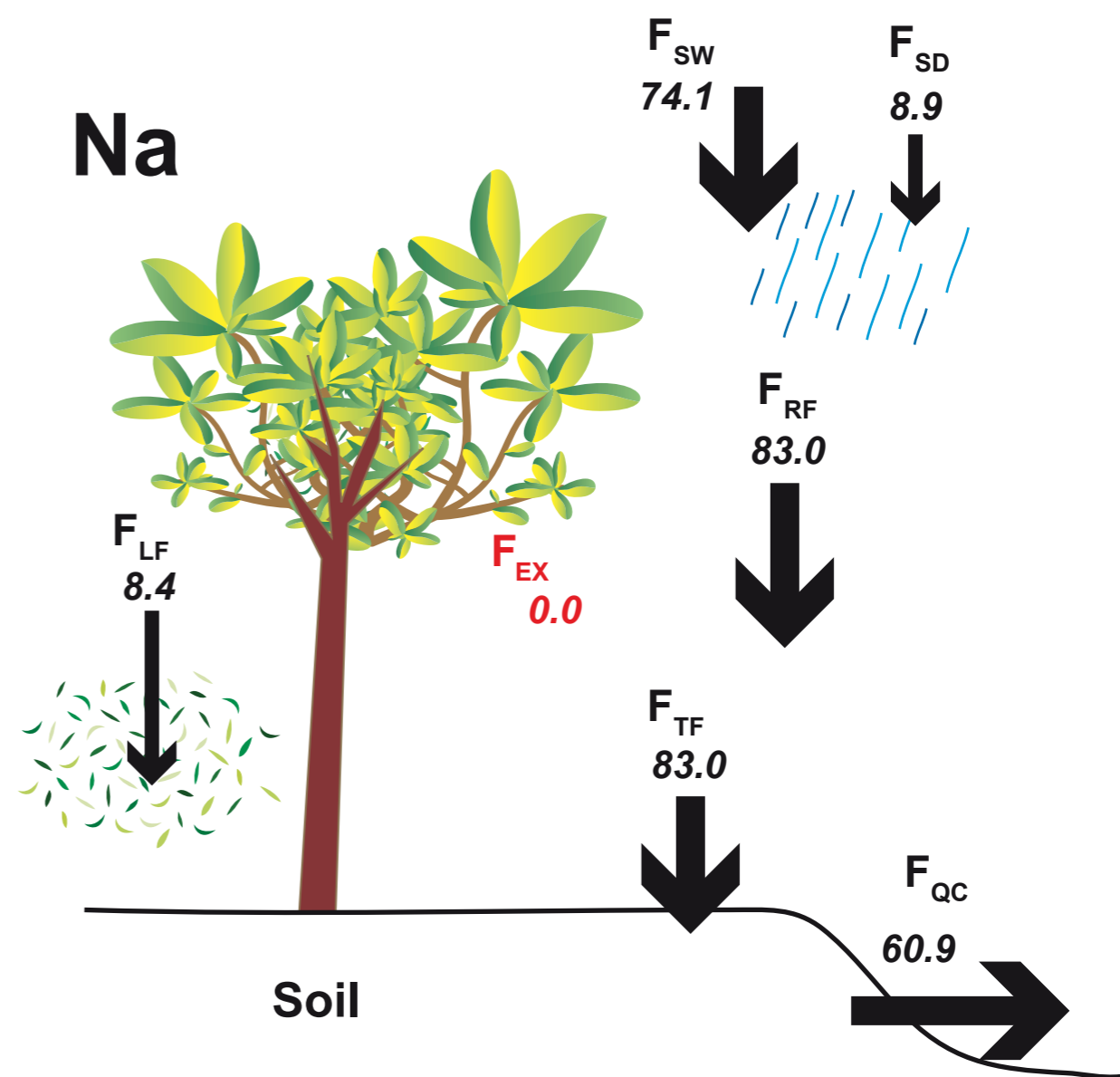




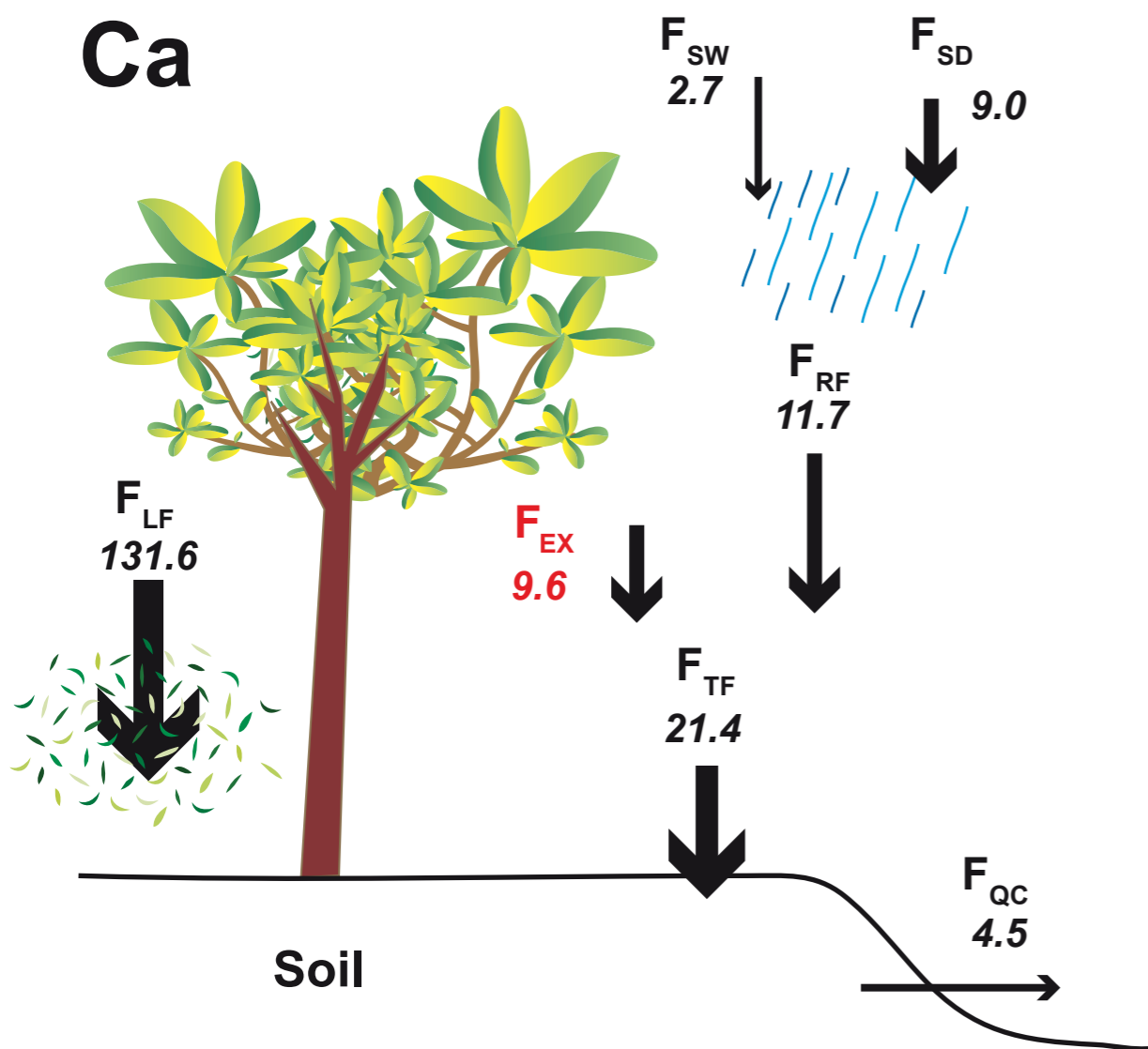
**Mg**



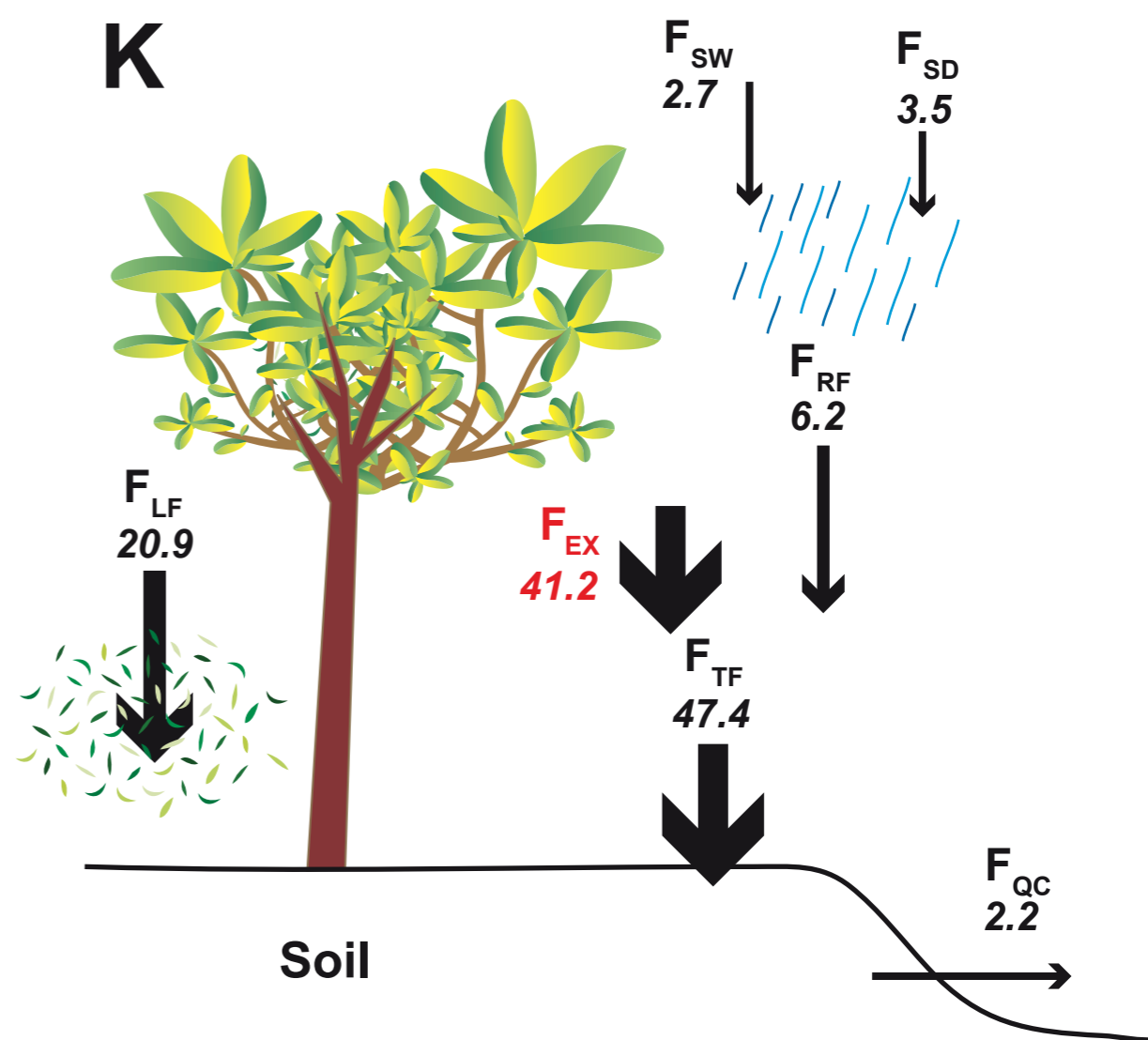
**Na**



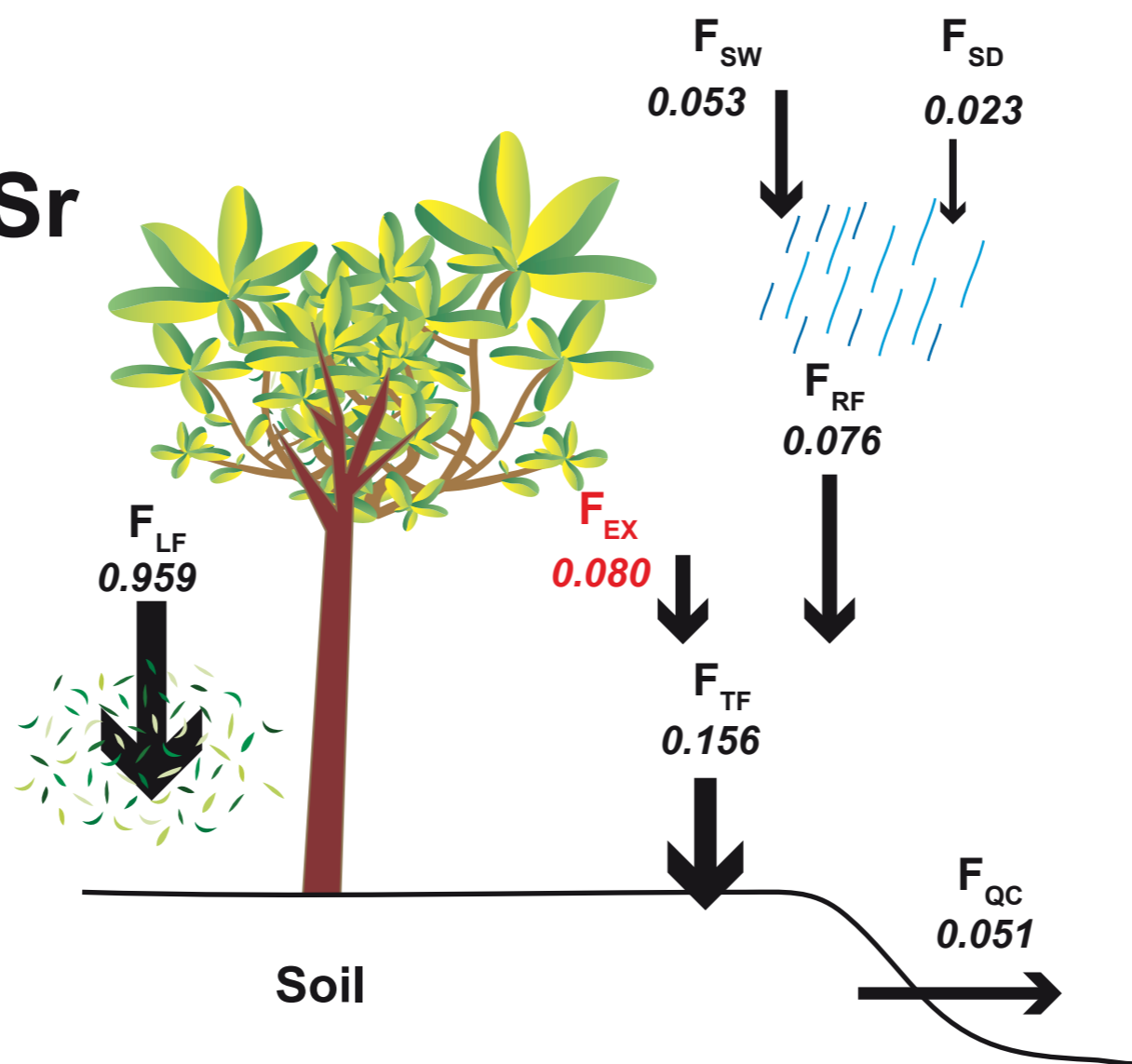
**Ca**

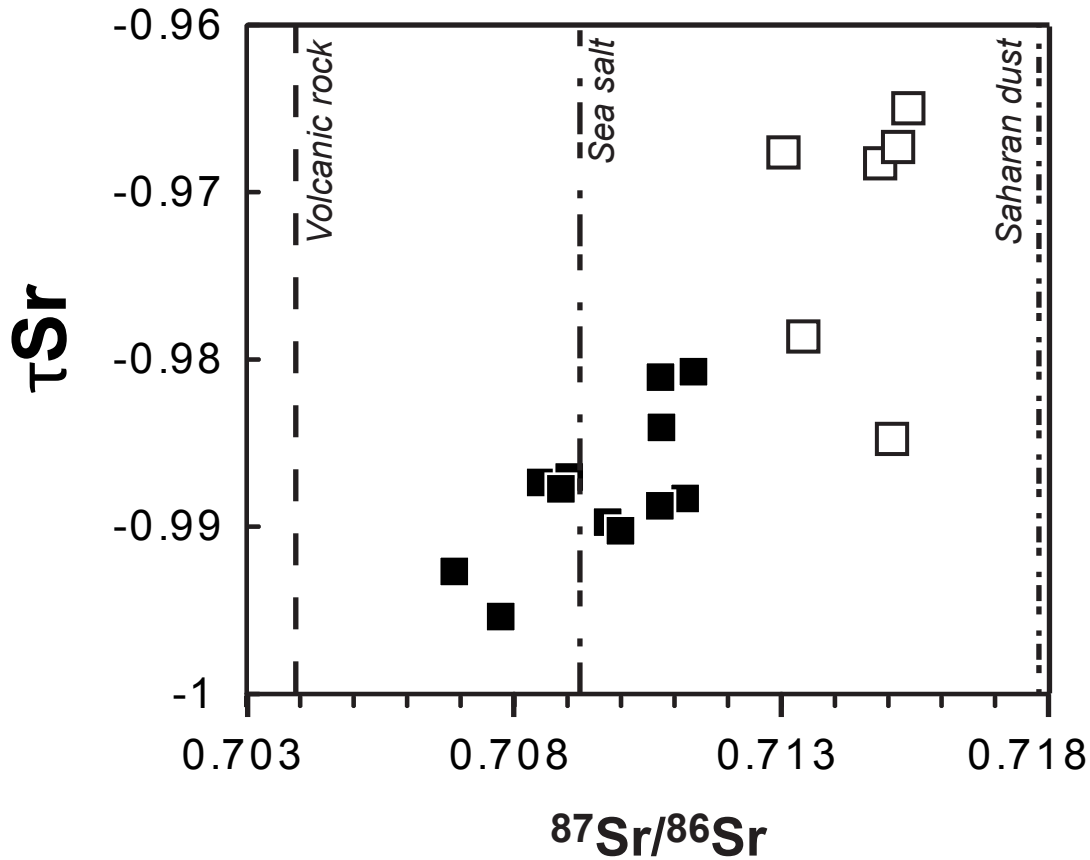


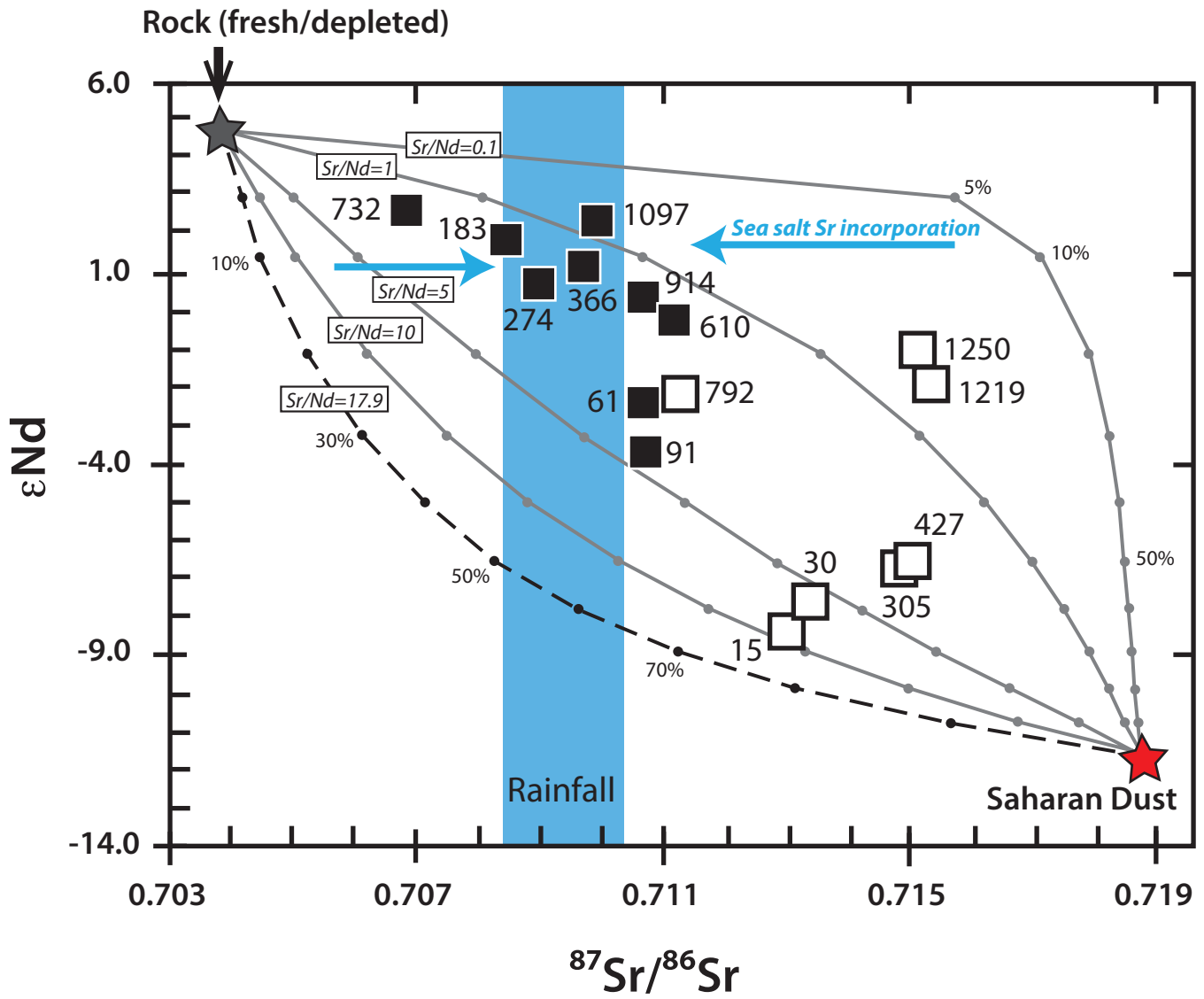
**K**



**Sr**









## Tables\_Results

Table 1: Major element concentrations are from Clergue et al. (2015). Sr concentrations and Sr isotopic signature in water samples are new data.

Sample name	Date dd/mm/yy	pH	Na μmol/L	Cl μmol/L	Ca μmol/L	Mg μmol/L	K μmol/L	Sr μmol/L	<sup>87</sup> Sr/ <sup>86</sup> Sr
<i>Rainfall (RF)</i>									
RF-OVSG	20/03/13	4.8	82	85	2	10	3	0.010	0.70918
RF-OVSG	04/04/13	5.1	76	88	7	13	9	0.015	0.70841
RF-OVSG	15/07/13	nd <sup>a</sup>	96	nd	7	14	5	0.034	0.71029
<i>Throughfall (TF)</i>									
TF collector n°2	21/11/11	nd	25	27	6	4	18	0.121	0.70965
TF collector n°1	10/08/12	6.1	81	92	15	13	31	0.056	0.70978
TF collector n°3	21/03/13	5.6	71	64	6	7	13	0.015	0.70953
TF collector n°1	03/04/13	nd	155	168	14	19	26	0.048	0.70969
TF collector n°3	03/04/13	nd	154	158	14	18	27	0.043	0.70972
TF collector n°1	25/06/13	5.6	117	132	17	17	32	0.067	0.70968
<i>Soil solution (SS)<sup>b</sup></i>									
GBD2_91	26/10/12	5.1	226	211	3	21	3	0.022	0.71070
GBD2_122	26/10/12	4.9	171	145	2	16	2	0.009	0.71097
GBD2_152	26/10/12	4.6	153	141	1	22	1	0.008	0.71063
GBD2_274	26/10/12	4.6	209	241	4	28	5	0.022	0.71020
GBD2_344	26/10/12	4.5	207	244	0	30	2	0.009	0.71094
GBD2_457	26/10/12	4.5	194	168	2	14	4	0.013	0.71004
GBD2_15	28/06/13	nd	118	130	2	21	2	0.039	0.71037
GBD2_30	28/06/13	5.4	147	150	1	27	2	0.034	0.71029
GBD2_61	28/06/13	4.8	126	163	12	31	4	0.063	0.70996
GBD2_91	28/06/13	5.3	194	207	3	25	2	0.020	0.71087
GBD2_274	28/06/13	4.7	161	221	2	26	2	0.017	0.71064
GBD2_457	28/06/13	5.4	159	167	2	14	3	0.010	0.71097
GBD2_823	28/06/13	nd	210	243	1	15	13	0.010	0.71022
<i>Quiock Creek</i>									
QC	23/04/12	nd	184	229	11	25	6	0.056	0.70939
QC	03/08/12	nd	248	271	9	29	5	0.052	0.70922
QC	26/10/12	4.7	254	271	8	30	4	0.048	0.70933
QC	21/03/13	4.8	211	274	15	32	3	0.046	0.70928

<sup>a</sup> nd=not determined.

<sup>b</sup> For soil solution sample names GBD2\_X, X=depth in cm.

Table 2: Range of concentration in the different compartments (mean value/median value) from Clergue et al. (2015).

	K ( $\mu\text{mol/L}$ )	Na ( $\mu\text{mol/L}$ )	Mg ( $\mu\text{mol/L}$ )	Ca ( $\mu\text{mol/L}$ )	Cl ( $\mu\text{mol/L}$ )
RF* (n=13)	1 – 9 (4/3)	41 – 165 (89/79)	1 – 22 (12/11)	1 – 15 (7/7)	49 – 167 (92/85)
TF (n=48)	10 – 108 (39/32)	17 – 264 (117/102)	3 – 40 (17/17)	3 – 36 (17/16)	23 – 264 (125/99)
SS (n=75)	1 – 39 (5/3)	112 – 266 (179/182)	11 – 58 (24/25)	0.3 – 14 (3/2)	85 – 345 (199/203)
QC (n=57)	3 – 12 (5/5)	184 – 282 (234/239)	24 – 35 (29/29)	7 – 15 (10/10)	229 – 315 (272/274)

\*: MF + OVSG rainfall.

Table 3: Chemical and isotopic (Sr and Nd) composition of soil, rock, Saharan dust and vegetation samples. Some Nd isotopic composition of soils (highlighted) and Ca, K, Mg, Na, Ti concentrations are from Clergue et al. (2015).

Sample name	Ca ppm	K ppm	Mg ppm	Na ppm	Ti ppm	Sr ppm	$^{87}\text{Sr}/^{86}\text{Sr}$	$\epsilon\text{Nd}$
<i>Saharan Dust</i>								
Saharan 1	1747	5474	5355	951	nd	52	0.71865	nd
Saharan 2	1918	5134	5059	930	nd	49	0.71882	nd
<i>Vegetation Litter</i>								
Leaves 1	30533	5987	5441	1356	nd	203	0.71019	nd
Leaves 2	10237	2302	1960	1857	nd	80	0.71010	nd
Branches 1	14234	1315	2730	581	nd	112	0.70966	nd
Branches 2	12468	1122	2397	504	nd	98	0.70975	nd
Roots	16410	11390	5676	2459	nd	94	0.70976	nd
<i>Soil<sup>a</sup></i>								
GBD2_15	306	1087	1977	681	8573	13	0.71293	<u>-8.39</u>
GBD2_30	147	643	1339	404	8873	9	0.71331	-7.57
GBD2_61	45	356	1189	356	7913	6	0.71065	<u>-2.35</u>
GBD2_91	43	455	1207	295	6894	6	0.71064	-3.66
GBD2_183	41	174	1287	444	7674	4	0.70840	<u>1.92</u>
GBD2_274	28	249	1043	248	7374	4	0.70893	0.74
GBD2_305	47	1026	1499	353	8033	12	0.71474	<u>-6.70</u>
GBD2_366	35	194	1068	344	8153	4	0.70964	1.19
GBD2_427	28	615	1262	158	10132	7	0.71496	-6.54
GBD2_610	34	326	1419	264	7314	4	0.71109	-0.21
GBD2_732	36	317	825	160	6834	2	0.70677	<u>2.71</u>
GBD2_792	43	621	880	278	5096	4	0.71125	-2.12
GBD2_914	29	321	1196	339	7074	4	0.71062	0.42
GBD2_1036	48	258	1234	347	6714	4	0.70877	nd
GBD2_1097	23	242	946	291	5995	3	0.70988	<u>2.44</u>
GBD2_1158	41	215	1341	324	9052	2	0.70764	nd
GBD2_1219	78	1276	1612	277	7674	12	0.71527	-1.89
GBD2_1250	60	971	1312	215	8273	12	0.71508	-0.97
<i>Volcanic Rock<sup>b</sup></i>								
04GW12	48462	7308	9060	27328	5373	244	0.70388	5.45
00GU44	74521	5175	29483	20282	5466	331	0.70380	4.34
03GU75	68417	4325	26106	21669	5796	283	0.70385	4.67

<sup>a</sup> for soil sample names GBD2\_X, X=depth in cm

<sup>b</sup> rock samples from Samper et al. (2007).

Table 4: Elemental annual flux ( $\text{kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ), stock ( $\text{kg}\cdot\text{ha}^{-1}$ ) and residence time (yr) in the Quiock Creek catchment.

	<b>Na</b>	<b>Ca</b>	<b>Mg</b>	<b>K</b>	<b>Sr</b>
<b>Rainfall flux (FRF)</b>	83.0	11.7	11.7	6.2	0.076
<i>Sea Salt flux (FSW)</i>	74.1	2.7	9.6	2.7	0.053
<i>Saharan dust leaching flux (FSD)</i>	8.9	9.0	2.1	3.5	0.023
<b>Throughfall flux (FTF)</b>	83.0	21.4	13.0	47.4	0.156
<i>Sea Salt flux (FSW)</i>	74.1	2.7	9.6	2.7	0.053
<i>Saharan dust leaching flux (FSD)</i>	8.9	9.0	2.1	3.5	0.023
<i>Leaf excretion flux (FEX)</i>	0.0	9.6	1.4	41.2	0.080
<b>Vegetation flux (FVEG)</b>	8.4	141.5	25.8	62.1	1.039
<i>Litter fall flux (FLF)</i>	8.4	131.9	24.4	20.9	0.959
<i>Leaf excretion flux (FEX)</i>	0.0	9.6	1.4	41.2	0.080
<b>Vegetation Stock</b>	405.1	6355.9	1180.1	1010.6	46.3
<b>Residence time (Tres)</b>	48.2	44.9	45.7	16.0	44.6
<b>Quiock Creek flux (FQC)</b>	60.9	4.5	8.0	2.2	0.051

Calculations are described by equations 2 to 9.

Table 1S: ICP AES (for major elements) and ICP MS (for Sr) analyses of rock and vegetation standard reference materials.

	Al	Ca	Fe	K	Mg	Na	Sr
	ppm	ppm	ppm	ppm	ppm	ppm	ppm
<i>Rock standards</i>							
BEN certif <sup>1</sup>	53312	99071	89880	11534	79291	23594	1370
BEN (this study)	54289	99897	90935	11739	79269	23846	1655
	52140	95138	89888	11397	76588	23325	
GSN certif <sup>1</sup>	77665	17857	26250	38419	13868	27971	570
GSN (this study)	77987	17562	25798	35813	13629	26838	531
	77673	17467	25594	35454	13600	26723	554
	80519	17834	26123	37214	13770	27587	
<i>Vegetation standard</i>							
NIST_SRM 1515 (certif)	286	15260	83	16000	2700	24.4	25
NIST_SRM 1515 (this study)	283	14441	69	15216	2421	55	
	290	14349	73	15544	2478	57	
	283	14872	75	16102	2662	42	
	286	15014	76	16307	2682	42	

<sup>1</sup> Recommended values of GSN, BEN are taken from CRPG certificate of analyze (<http://helium.crpq.cnrs-nancy.fr/SARM/pages/geostandards.html#>).

Table 2S : Sr concentration in throughfall, rainfall and Quiock Creek samples

Sample name	Date	Sr $\mu\text{mol/l}$	Sample name	Date	Sr $\mu\text{mol/l}$	Sample name	Date	Sr $\mu\text{mol/l}$
<b>Throughfall</b>								
TF col n°1	14/11/2011	0.136	TF col n°3	25/03/2013	0.041	QC	16/03/2012	0.064
TF col n°2	14/11/2011	0.129	TF col n°1	27/03/2013	0.070	QC	04/04/2012	0.057
TF col n°1	21/11/2011	0.025	TF col n°3	27/03/2013	0.049	QC	09/04/2012	0.058
TF col n°2	21/11/2011	0.121	TF col n°1	29/03/2013	0.070	QC	13/04/2012	0.058
TF col n°3	21/11/2011	0.038	TF col n°3	29/03/2013	0.050	QC	19/04/2012	0.070
TF col n°1	25/11/2011	0.035	TF col n°1	01/04/2013	0.018	QC	23/04/2012	0.056
TF col n°2	25/11/2011	0.026	TF col n°3	01/04/2013	0.016	QC	26/04/2012	0.056
TF col n°3	25/11/2011	0.035	TF col n°1	03/04/2013	0.048	QC	08/06/2012	0.054
TF col n°1	09/04/2012	0.012	TF col n°3	03/04/2013	0.043	QC	03/08/2012	0.052
TF col n°2	09/04/2012	0.067	TF col n°1	25/06/2013	0.067	QC	05/08/2012	0.053
TF col n°3	09/04/2012	0.037	TF col n°3	25/06/2013	0.062	QC	10/08/2012	0.053
TF col n°1	13/04/2012	0.126	TF col n°1	26/06/2013	0.048	QC	16/08/2012	0.050
TF col n°2	13/04/2012	0.076	TF col n°3	26/06/2013	0.036	QC	07/09/2012	0.050
TF col n°3	13/04/2012	0.049	TF col n°1	28/06/2013	0.043	QC	05/10/2012	0.050
TF col n°2	16/04/2012	0.027				QC	23/10/2012	0.049
TF col n°3	16/04/2012	0.023	<b>Rainfall</b>			QC	26/10/2012	0.048
TF col n°1	19/04/2012	0.119	RF-MF	03/08/2012	0.039	QC	30/11/2012	0.037
TF col n°2	19/04/2012	0.113	RF-MF	10/08/2012	0.034	QC	14/12/2012	0.035
TF col n°3	19/04/2012	0.109	RF-OVSG	20/03/2013	0.010	QC	18/01/2013	0.037
TF col n°1	23/04/2012	0.011	RF-OVSG	21/03/2013	0.011	QC	01/02/2013	0.037
TF col n°2	23/04/2012	0.010	RF-OVSG	27/03/2013	0.021	QC	01/03/2013	0.046
TF col n°3	23/04/2012	0.008	RF-OVSG	31/03/2013	0.009	QC	20/03/2013	0.045
TF col n°1	05/08/2012	0.031	RF-OVSG	01/04/2013	0.017	QC	21/03/2013	0.046
TF col n°2	05/08/2012	0.032	RF-OVSG	04/04/2013	0.015	QC	25/03/2013	0.042
TF col n°3	05/08/2012	0.038	RF-OVSG	15/07/2013	0.034	QC	29/03/2013	0.039
TF col n°1	10/08/2012	0.056				QC	01/04/2013	0.039
TF col n°2	10/08/2012	0.060	<b>Quiock Creek</b>			QC	03/04/2013	0.042
TF col n°3	10/08/2012	0.121	QC	14/10/2011	0.059	QC	24/05/2013	0.056
TF col n°1	16/08/2012	0.098	QC	10/11/2011	0.069	QC	21/06/2013	0.048
TF col n°2	16/08/2012	0.115	QC	23/11/2011	0.068	QC	25/06/2013	0.051
TF col n°3	16/08/2012	0.136	QC	09/12/2011	0.060	QC	26/06/2013	0.052
TF col n°1	21/03/2013	0.017	QC	06/01/2012	0.064	QC	28/06/2013	0.047
TF col n°3	21/03/2013	0.015	QC	03/02/2012	0.061	QC	02/07/2013	0.049
TF col n°1	25/03/2013	0.084						