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Marc F. Benedetti[1*], Aline Dia[2*], Martine Gérard[3], Jacques Boulègue[1], François Chabaux[4], Bertrand Fritz[4], Jean Riotte[4], Catherine Chauvel[5], Marc Bulourde[2], Bernard Déruelle[6] and Philippe Ildefonse[7+],

1 Laboratoire de Géochimie et Métallogénie, CNRS-UMR 7047, Université Pierre & Marie Curie, Case 124, 4 place Jussieu, 75252 Paris Cedex 05, France
2 Géosciences Rennes, CNRS-UMR 6118, Campus Beaulieu, Av. du Général Leclerc, 35042 Rennes Cedex, France
3 Institut de Recherche pour le Développement (IRD), 32 avenue Henry Varagnat, 93143 Bondy Cedex, France
4 Centre de Géochimie de la Surface, Ecole et Observatoire des Sciences de la Terre, 1 Rue Blessig, 67084 Strasbourg Cedex, France
5 Laboratoire de Géodynamique des Chaînes Alpine, Observatoire de Grenoble, 1381 rue de la Piscine, 38041 Grenoble Cedex 09, France
6 Magmatologie et Géochimie CNRS-UMR 7047, Université Pierre & Marie Curie, Case 124, 4 place Jussieu 75252 Paris Cedex 05, France
7 Laboratoire de Minéralogie-Cristallographie, Université Pierre & Marie Curie, UMR CNRS 7590, 4 Place Jussieu, 75252 Paris Cedex 05, France

+ Deceased

corresponding authors emails: benedett@cicrp.jussieu.fr, dia@univ-rennes1.fr

Abstract

This study was dedicated to the early stage of the weathering of historic basaltic flows located in Mount Cameroon. Spring and rivers were sampled all around the volcano. We report here the basic chemistry of the waters as well as strontium and uranium isotopic ratios.

The combination of the molar proportions of solute obtained with the modal amounts of the minerals in the basalts gives a prediction of what should be the relative molar concentrations of major compounds in the weathering waters issuing from Mount Cameroon. The measured Alkalinity/Si and Mg/Si ratios are higher than the calculated ones while the measured Ca/Si ratio is equal to the calculated value. We suggest that the Si depletion of Mount Cameroon waters is caused by biological pumping, trapping of Si in Fe-silicate minerals such as Si containing ferrihydrite and Si interaction with bacterial cell wall leading to the formation of allophane type minerals which were observed in Mount Cameroon soil profiles. Calcium uptake by plants explains the lower Ca/alkalinity ratios measured in the water samples.

The water-rock ratio (R) calculated from the strontium isotopic compositions of the water samples, ranges from 29,452 to 367,450. The calculated weathering rates (W_R) range from 1 to 20 mm/kyr and from 1 to 103 mm/kyr for high and low altitudes, respectively and agree with both the thickness and the age of paleosoils found in the same area and with previously published estimates from coupled reaction-transport models. This difference emphasizes the role of vegetation and rainfall at lower altitudes as compared to what happens at high altitudes.

1. Introduction

Rock weathering depends on many factors such as climate, relief, lithology, tectonics, and vegetative cover and controls the morphology at the Earth's surface and the solute acquisition of waters flowing towards the ocean. As silicate weathering influences the CO_2 mass balance in the atmosphere (Berner, 1992 ; 1999), riverine erosion is likely to be involved in the long term climate evolution. Among rocks of volcanic origin, basalts are particularly sensitive to chemical erosion (Berner and Berner, 1996). While numerous mineralogical and geochemical studies have been carried out on basalt weathering (Crovisier et al., 1992; White and Hochella, 1992; Daux et al., 1994, Gislason et al., 1996; Dahlgren et al., 1999) the chemistry of the waters involved in the weathering processes (chemical and mechanical) has received far less attention (Gislason and Eugster, 1987a; Négrel et al., 1993; Benedetti et al., 1994; Dupré et al., 1996; Gaillardet et al., 1997; Louvat and Allègre, 1998). Laboratory-derived weathering rates for basaltic rocks are available (Crovisier et al., 1987; Gislason and Eugster, 1987b). Non-equilibrium weathering model based on weathering mineral reactions adjusted taking into account pseudomorphic replacement and their kinetics was proposed by Wang et al. (1995).

The water held within the internal surface areas of rocks or soils is subject to nanodomain influences which are different than bulk water and that may be the reason why it is difficult to compare laboratory and field-based studies (Hochella, 2002). The role of plants in controlling rates and products of weathering may also account for differences between laboratory and field-based studies (Lucas, 2001; Hinsinger et al., 2001). Then, the need to document weathering rates of geologic materials under field conditions still persists because any attempt to extrapolate these experimental weathering rates to the natural environment remains hazardous. Field weathering rate data have been estimated from both elemental budgets (Velbel, 1986; Benedetti et al., 1994; Edmond et al., 1995; Gaillardet et al., 1995; 1997; White and Brantley, 1995; Louvat and Allègre, 1997; 1998; Aiuppa et al., 2000; Picouet et al., 2002) or isotope composition (Innocent et al., 1997; Vigier et al., 2001) of watershed surface waters.

Mount Cameroon is an ideal target to study such processes because its basaltic composition helps to constrain the influence of other factors such as climate and/or vegetative cover. Also, its

location in a humid tropical area implies (i) that an abundant vegetation grows on it depending, however, on the age of the volcanic substrate and on the altitude and (ii) that weathering alteration should be fast. Water samples were collected from springs, streams and rivers around Mount Cameroon, during the dry season (December 1996) and at the beginning of the small rainy season (March 1997) in order to assess the influence of climate on surface water chemistry. The meteoric waters are extremely dilute and therefore should respond readily to the weathering of the terranes. Thus, the study of surface water samples in such a context of high rainfalls should allow us to define solute acquisition processes, saturation states of waters with respect to primary and secondary minerals, and estimates of mass transfer through water/rock ratio and weathering rate.

The aims of this paper are, through major-, trace-element concentrations as well as Sr and U isotope composition measurements of the dissolved load, (i) to define and to interpret the chemistry of waters recovered from Mount Cameroon, (ii) to establish the source of dissolved constituents and (iii) to estimate water/rock ratios and the chemical denudation rates as well as the duration of the weathering and the possible emphasizing effects of the vegetation and differential climatic conditions (rainfall, elevation…) on the chemical weathering of basalt and transfer to solution.

2. Geologic and Climatic Settings

Mount Cameroon belongs to the Cameroon Line, an intraplate volcanic alignment extending from the Gulf of Guinea into the African continent. This chain has been active for at least 65 Myr (Lee et al., 1994) (Fig.1a). Mount Cameroon is the only active volcano of the Cameroon Line. Its volcanic activity started 11 Ma ago (Upper Miocene) and persists leading to the deposition of tephra and massive basaltic lava flows on the flanks of the volcano. Historical volcanic lava flows range from 1815 to 1999 AD (Déruelle et al., 1987; Bulourde et al., 2002).

Since Mount Cameroon is located in a humid tropical area characterized by extreme rainfalls and elevated temperatures all year long, rocks undergo intense weathering conditions favouring their rapid alteration. However, climatic conditions are not uniform on the volcano.

The combination of high relief (4071m) and proximity of the sea leads to strong local climatic contrasts (Fig.1b). Mean annual temperatures decrease from 26-29°C at sea level to 0°C at the top of the edifice and this large temperature drop is associated to a decrease in rainfalls. The highest rainfall is recorded on the southwest flank of Mount Cameroon where rainfall can reach 12 meters per year. Lower rainfalls occur on the opposite flank because it is partially sheltered from the oceanic influence (for example : 1800 mm/year at Ekona). Due to the altitudinal gradient, the mountains moderate the diurnal range of air temperatures and maintain atmospheric humidity levels. At high elevation, a high diffusion coefficient for water vapor under low air pressure and a high radiation load result in evapotranspiration rates ranging from 50 to 90% higher in tropical mountains than in middle-latitude ones (Leuschner, 2000). At lower altitudes, evapotranspiration can represents 60 to 80 % of the annual rainfall (Njitchoua et al., 1999).

The climatic conditions promote the growth of abundant vegetation on Mount Cameroon. The vegetation cover depends both on the age of the volcanic substrate and the altitude. The most recent lava flows are colonized by pioneer species such as mosses and lichens. Jackson and Keller (1970) investigated the role of lichens in the chemical weathering of basaltic lava flows. These authors suggest that chemical weathering is greatly accelerated for lichen-covered rocks comparatively to bare rocks. This finding is questioned by Cochran and Berner (1996) for whom lichens only play a minor role. Depending on the altitude and rainfall, the Mount Cameroon basaltic terranes are colonized by grasses, shrubs or trees (the forest being present up to altitudes of about 2000 m). This type of vegetation cover has been shown to have drastic effects on the weathering rate of basalts. The effect of plants on weathering was both estimated by Benedetti et al. (1994), Moulton and Berner (1998) and Hinsinger et al. (2001) who showed that weathering rates are two to five times higher in vegetated areas than in bare areas for basaltic rocks and recently reviewed by Lucas (2001). Considering these environmental characteristics, Mount Cameroon water samples may be ideal targets to record and quantify chemical mobility during the very first stage of the chemical weathering of basalts.

3. Sample collection

Springs, streams and rivers from Cameroon were sampled at different times and elevations. Some of them were sampled twice, in the dry season (December 1996) and at the beginning of the rainy season (May 1997). Twelve water samples were collected during the first field trip; twenty four samples were recovered during the May 1997 rainy season as well, as four rain-water samples at Buea. The water sample locations are reported in Fig. 1b.

Samples used for determination of dissolved major-, trace-element, and Sr and U isotope concentrations were collected in pre-cleaned, acid-washed polyethylene bottles and stored after filtration (with 0.1 μm cellulose acetate filter). Unacidified samples were stored for analysis of anions (Cl^-, SO_4^{2-}, NO_3^-). Aliquots for trace-element and Sr isotope analyses were acidified with 15N distilled nitric acid.

4. Analytical Methods

Several parameters were measured in the field at the time of the sampling or within 2 hours of sampling (pH and alkalinity) and are reported in Table 1. The pH was measured with a combination electrode after a calibration with standard solutions, the precision of pH measurements was 0.05 pH units.

4.1. Major and Trace elements

Alkalinity was measured in the field within two hours after sampling (Gran titration) with a precision of 1%. Laboratory measurements of major cations were done by flame atomic absorption spectrometry (precision ± 1%). The detection limits for Ca and Mg are 7.5μM and 8.5μM, respectively. Chloride, nitrate and sulfate concentrations were measured by Dionex chromatography (accuracy ± 1% and detection limit = 1μM). Silica was determined by inductively coupled plasma emission spectroscopy (detection limit 18μM and precision ±2%). Total aluminium and iron, were determined by Zeeman corrected electrothermal atomic

absorption spectrometry (precision ±5-10%) and the detection limits were 18nM and 18nM, respectively.

4.2. Sr isotopes

Conventional chromatographic method was used to separate Sr using Sr-Spec Eichrom (Pin et al., 1994) and a combination of HNO_3 and H_2O. Total blanks for Sr were less than 60 pg and were negligible as compared to the Sr amounts of the samples. Sr concentrations were determined by ICP-MS (HP® 4500) while isotopic analyses were performed at Géosciences Rennes by thermal ionisation mass spectrometry using a Finnigan® MAT 262 multicollector mass spectrometer. During the course of this study the NBS 987 Sr standard yielded a mean $^{87}Sr/^{86}Sr$ ratio of 0.710270 ±11.

4.3. Uranium isotopes (Riotte et al. 2002 data)

The U isotopic compositions were determined by T.I.M.S. with 5 to 20 ng of U. After preconcentration by coprecipitation with Fe hydroxide, U was separated and purified by ion exchange chromatography, as described in Chabaux et al. (1995). The U isotopic compositions were determined on a VG Sector mass spectrometer equipped with a Daly detector operating in analogical mode. The analytical uncertainties of the $^{234}U/^{235}U$ ratios measured were about 5‰, except for a few low U content samples, whose uncertainties can reach 20‰ (see Table 2). The external reproducibility of the U isotopic analyses was tested with regular measurements of the HU1 U Standard supposed to be at secular equilibrium. The regular measurements of this standard over 3 years (about 80 determinations), gave a mean ($^{234}U/^{238}U$) activity ratio of 0.9995±0.002 (2σ error) by using the most recent value given for the half life of ^{234}U, that is T=(2,455±0.006).10^5 yr (Akovali Y.A., 1994; Audi et al., 1997; Cheng et al., 2000). The new U half-life value will be used here to convert $^{234}U/^{238}U$ atomic ratios into ($^{234}U/^{238}U$) activity ratios (Table 2). The blanks of the whole procedure are less than 40 pg; they always remain negligible as compared to the U amounts analyzed in the samples.

5. Results

5.1. Major elements and trace elements

Concentrations of major elements and trace elements are reported in Tables 1 and 2. The sample waters are mostly near neutrality with pH ranging from 6.39 to 8.05. Waters sampled in the low altitude region have the highest ionic charge (2000 to 3000 μM) and those draining the higher altitude have the lowest ionic charge (<1300 μM). Waters are characterised by a good charge balance, as indicated by the normalised inorganic charge balance ($\square^{+}$-$\square^{-}$/ $\square^{+}$). Values range from 1 to 7%.

The dissolved silica concentration varies between 276 and 1034μM. The dissolved silica concentration is systematically the lowest for the high altitude samples (< 568μM). Concentrations in the water samples are similar to those from rivers flowing through the Deccan Traps and the Réunion Island rivers (200 to 800 μM - Louvat et al., 1997, Dessert et al., 2001). Bicarbonate ion (251 to 3677μM) is always the dominant anion species, followed by Cl^{-}, SO_4^{2-} and F^{-} (see Table 1 for values) with concentrations systematically smaller than those found in the Réunion island (Louvat et al., 1997). The lower anionic concentration characterizes samples originated from high altitude. This observation is also true for cationic content. Among major cations, Na^{+} is dominant (50 to 1324μM). Ca^{2+} and Mg^{2+} (40 to 775μM) have the same range of concentrations whereas concentrations of K^{+} are smaller (11 to 238μM). The cations concentrations are systematically smaller than the one reported for rivers flowing through the Deccan Traps or the Réunion island (Louvat and Allègre, 1998, Dessert et al., 2001)

The concentrations of Sr vary between 0.548 and 1.768μM. Fe and Al concentrations range from 20 to 1134 nM. Al and Fe concentrations are smaller than that of waters flowing on other basaltic rocks (Benedetti et al., 1994, Louvat and Allègre, 1998, Aiuppa et al., 2000). U concentrations were analyzed by Riotte et al. (2002). Most U concentrations are low and range from 0.03 to 1.34nM. They are lower than the U concentrations of Etnean springs (Aiuppa et al., 2000) ranging from 0.4 to 33nM.

5.2. Sr isotopes

The Sr isotope compositions are reported in Table 2. The water samples display $^{87}Sr/^{86}Sr$ ratios varying between 0.703397 (CM 19) to 0.704868 (CM 49). Most of the water samples exhibit Sr isotope compositions extremely close to the fresh lava flow $^{87}Sr/^{86}Sr$ ratios (Bulourde et al., 2002) suggesting that at this time scale the transfer of elements from basalt and associated soils through solute acquisition remains limited. The $^{87}Sr/^{86}Sr$ ratios measured in the rainwater samples (ranging in between 0.70778 and 0.706219) are more radiogenic than the mean basalt value composition, which is significant for some of the water samples whose $^{87}Sr/^{86}Sr$ ratios are shifted towards more radiogenic values than that of the basaltic substrate. But, these water samples (as for example: CM 48 or CM 50) remain always below the rainwater mean value (0.706762), which suggests that these shifts towards more radiogenic values are more likely to be due to basalt/meteoric water interaction and subsequent solute acquisition by meteoric waters rather than to contamination by seawater.

The first set of samples collected during the dry season have similar $^{87}Sr/^{86}Sr$ values, while the $^{87}Sr/^{86}Sr$ values for samples recovered during the rainy season show a larger spread. This is particularly true at low altitude and especially for the samples collected close to sea level. This result will be further discussed and compared to analogous observations made with uranium isotope data. No systematic trends are observed between Sr isotope data and pHs, alkalinities or altitudes (see Tables 1 and 2).

5.3. Uranium isotope data from Riotte et al. (2002)

The U isotope data and U contents that are reported in Table 2 are taken from Riotte et al. 2002. These uranium analyses are part of a paper dedicated to the U colloidal transport and the origin of the ^{234}U-^{238}U fractionation in Mount Cameroon surface waters (Riotte et al., 2002). The $(^{234}U/^{238}U)$ ratio range widely from 1.038 to 1.397. The narrow range of variation of the samples recovered at high altitude was interpreted as recording meteoric weathering of the bedrock while larger ^{234}U-^{238}U fractionations displayed by the samples from medium and low altitudes may be

linked to either the nature and the intensity of the weathering processes, or deep water supplies characterized by different uranium signature (Riotte et al., 2002).

6. Discussion

6.1 Major ion geochemistry

Major ion concentrations show a strong correlation with the altitude of the sample. As evidenced in Fig. 2 where chloride concentrations are reported versus the sample altitude, the higher the altitude is, the lower is the Cl^- concentration while there is no simple relationship between the Cl^- concentration and the distance from the coast. This does reflect the input of the oceanic monsoon chloride rich rainwater for low altitude samples. For higher altitudes, the decrease of Cl^- is due to smaller rainfall inputs caused by the rapid decrease of the absolute humidity and temperature with altitude, resulting in a smaller condensation.

Correlation between elements shown in Figs. 3 and 4 can be interpreted within the mass balance approach for small watershed initially developed by Garrels and MacKenzie (1967). The major mineral phases identified by Déruelle et al. (1987) in the basalts are : olivine (17%), pyroxene minerals (20%), plagioclase (50%) and oxides (13%). The chemical formula of the reacting minerals (i.e. all minerals except oxides) and the precipitating minerals used in the mass balance calculations are given in Table 3. The two end products of the weathering are amorphous iron oxides and allophane type minerals both evidenced in the soil profiles by Gérard et al. (1999). The combination of the molar proportions of solute obtained with the modal amounts of the minerals in the basalts give a prediction of what should be the relative molar concentrations of major compounds in the weathering waters issuing from Mount Cameroon. The calculated ratios were compared to the measured ratios of the different samples in Table 4. Measured Alkalinity/Si and Mg/Si ratios were higher than the calculated ones while the measured Ca/Si ratio was equal to the calculated value. The measured and calculated Mg/Alkalinity ratios were equal while the measured Ca/Alkalinity value is much smaller than the calculated value.

Two hypotheses can explain the higher measured Alkalinity/Si, Mg/Si and the smaller Ca/Alkalinity ratios. The first one is the existence of an external source of Mg and CO_2 in the

system and the second one is an extra sink for Si and Ca that was not taken into account in the mass balance calculations. The extra source could be hydrothermal Mg, CO_2-rich fluids resulting from volcanic activity. However, the low temperature of the water samples and the lack of ^{18}O shift with respect to the meteoric line (not shown here) exclude the influence of hydrothermal water-rock interactions (Aiuppa et al., 2000). Several mechanisms can explain the Si depletion of Mount Cameroon waters. The first one is the uptake of Si by plants (Hinsinger et al., 2001 and reference therein). This process is supported by the occurrence of phytolites in some of the A horizons of the soil profiles developed onto Mount Cameroun basalts. The second mechanism is the sequestration of Si ions. Their strong affinity for hydroxy-Fe ions leads to the formation of Fe-silicate minerals such as Si containing ferrihydrite (Vempati and Loeppert, 1989) detected in the soil profiles (Gérard et al., 1999). The third process is preferential interaction of Si ions with bacteria involved in the weathering process (Thorseth et al., 1992; Kawano and Tomita, 2001; Welch and Banfield, 2002). Interaction of Si ions with bacterial cell wall takes place either by cation bridging (Urrutia and Beveridge, 1994; Fein et al., 2002), or direct interaction with reactive groups that leads to the preferential formation of allophane type minerals that were observed during this work (Gérard et al.,1999). Calcium uptake by plants can also explain the lower Ca/Alkalinity ratios measured in the water samples (Hinsinger et al., 2001). The lack of agreement between the Ca/Mg ratios can result from either preferential uptake of Ca by plants (Hinsinger et al., 2001), or differential kinetics of dissolution of olivine (i.e. a Mg source) and plagioclase (i.e. a Ca). The latter has a slower dissolution rate than the former (Schnoor, 1990).

In Fig. 3 the regression line indicates that when Alkalinity tends to zero, 228 μMol of silica should be detected in solution. Thus, a source of Si is undergoing weathering without any impact on the alkalinity mass balance value. The weathering of quartz can account for such a trend. However, minerals of the quartz group are not found in such basaltic lavas. A source of SiO_2 are the easily dissolving phytolites evidenced in some soil profiles of Mount Cameroon. Atmospheric inputs of quartz particles are another potential source of SiO_2. Chauvel et al. (2002) evidenced inputs from Sahara dust based on Nd and Sr isotopic data in soils of Mount Cameroon. Saharan dust are well known in Cameroon especially in winter when the Harmattan

wind carry airborne particles from the mechanical erosion from Tibesti and Hoggar massifs (Caquineau et al., 2002). These particles are mechanically disaggregated and there is no subsequent chemical fractionation occurring during their transport as compared to the chemical composition of the parent-rock. This result in a Si-rich material input to the top levels of soils as compared to what is expected in soils developed onto basalts.

The proportion particles needed to account for the 228 μMol of silica shift in Fig. 3 can be estimated with the amount of water actually flowing through the weathering profiles and the rate of formation of those profiles. The amount of recharge available to enter the aquifers on an annual basis is about equal to average annual precipitation minus water losses (average annual runoff and evapotranspiration). Runoff is directly related to rainfall, topography, soil type, and land use, and can range from 10 to 40 percent of the average annual precipitation. At the top of Mount Cameroun, the annual pan-evaporation rates could range from 80 to 90% such as what happens at lower altitude, because of clear sky, strong winds and dry air as well as a high diffusion coefficient for water vapor under low air pressure and a high radiation load (Leuschner, 2000). This could mean that only 10 to 20% of the average annual precipitation would be available. If we take a average precipitation of 3000 mm for the high altitude area (Sieffermann, 1973, Njitchoua et al., 1999); a water flow ranging from 300 to 600 mm/year is supposed to enter the aquifers. These water flows correspond to a fluxes ranging from 0.068 to 0.136 moles of Si per year and m^2 or 4.09 to 8.19 g of SiO_2 per year and m^2. Based on Sr isotopic data, an average weathering rate of 5 10^{-5} m/yr is calculated for Mount Cameroon area (see discussion section 6.3 on water rock ratio and weathering rates). For one square meter and a mean density of 2, this rate corresponds to 100g of rock undergoing weathering. The amount of SiO_2 calculated above to account for the 228μMol of silica shift represents 4.09 to 8.19% of the material undergoing alteration. This extra amount of SiO_2 is in the range of the amount of atmospheric particle inputs calculated by Chauvel et al. (2002).

6.2 The Sr isotope record

These surface water data show that the Sr isotopic compositions reflect mostly the nature of the drained bedrocks, i.e. basalt and associated soils developed on it and therefore result from basalt/meteoric water interaction and subsequent solute acquisition by meteoric waters. The variations of Sr concentrations and isotopic compositions are illustrated in Fig. 5 as a conventional $^{87}Sr/^{86}Sr$ ratio versus 1/Sr diagram. Spring waters (recovered at high altitude) collected during the dry season are characterized by almost uniform Sr isotope compositions slightly higher than those of the basaltic lavas (Bulourde et al., 2002). The $^{87}Sr/^{86}Sr$ ratios of the samples collected during the rainy period reveal larger shifts particularly at low altitudes. This might result from mixing between different water sources and/or contributions from soil horizons and weathered profiles as suggested by the comparison with the uranium isotope data (Fig. 6).

While the major ion chemistry does correlates with the altitude of the sample (Fig.2) both during dry and rainy seasons, the Sr isotope data do not whichever the sampling period was (Table 2) (Fig. 7). It appears therefore difficult to explain the Sr isotope signature as reflecting both the seawater contamination in the rainfall at low altitude and smaller rainfall inputs at higher altitudes.

Accounting for the measured chemical compositions of the surface waters, a conventional Water/Rock ratio (R) was estimated from the $^{87}Sr/^{86}Sr$ ratios as previously done elsewhere (Innocent et al. 1997, and references therein). The isotopic composition $(^{87}Sr/^{86}Sr)_{rain}$ and the Sr concentration of the local rain $c_{rain,}$ was estimated by the mean of the average of the data obtained on the four rain samples collected at Buea (i.e., 0.706762 and 2.125ppb), respectively. $(^{87}Sr/^{86}Sr)_{basalt}$ and c_{basalt} are the Sr isotope composition and concentration of the fresh basaltic lava, respectively. They were estimated by averaging the analyses (Sr content and isotope composition) of four fresh massive basalt from 1909, 1922, 1959 and 1982 (Bulourde et al., 2002). The freshness of this predicted basaltic material was checked through both thin section observation and a major-, trace-element concentration, Sr, Nd and Pb isotope investigation to be sure that these samples could be considered as a good reference material (Bulourde et al., 2002).

$(^{87}Sr/^{86}Sr)_{basalt} - (^{87}Sr/^{86}Sr)_{water}$

$$R = \frac{}{(^{87}Sr/^{86}Sr)_{water} - (^{87}Sr/^{86}Sr)_{rain}} (c_{basalt}\, d_{basalt} / c_{rain,})$$

where $(^{87}Sr/^{86}Sr)_{basalt} = 0.703325$ $c_{basalt} = 1045$ ppm $d_{basalt} = 2.8$ (basalt density)

$(^{87}Sr/^{86}Sr)_{rain} = 0.706762$ $c_{rain,} = 2.125$ ppb

The Water/Rock ratios (R) range between 29,452 and 96,446 for the samples recovered during the dry period and between 34,929 and 1 121, 800 for the rainy season samples, respectively (Table 2). These ratios are much higher than values obtained for high-temperature water/rock interactions (Albarède et al., 1981; Michard et al., 1984; Piepgras and Wasserburg, 1985). These data from Mount Cameroon are quite comparable to previously reported ratios deduced from meteoric weathering of basalt from Iceland (Daux, 1992).

6.3 Rate and duration of the weathering

The weathering rate, W_R, is related to the water rock ratio (R) according to the following equation (Innocent et al., 1997):

W_R = WI/R, where WI is the water input in mm per year (i.e. rainfall – evapotranspiration).

Assuming that the water recharge area of the high altitude rivers and springs is mainly located at the top of the volcano at altitudes above 2000m and that only 10 to 20% of the average annual precipitation would be available (Leuschner, 2000). If we take an average precipitation of 3000 mm for the high altitude area (Sieffermann, 1973, Njitchoua et al., 1999); a water flow (WI) ranging from 300 to 600 mm/year is supposed to enter the aquifers. The water rock ratio for high altitude springs ranges from 29,452 to 235,520 and the W_R is ranging from 1.3 to 20.4 mm/kyr. At lower altitudes evapotranspiration can represents 60 to 80 % of the annual rainfall that ranges from 3,000 to 9,000 mm/yr (Njitchoua et al., 1999), which let between 600 mm/yr and 3,600 mm/yr to enter the system. The water rock ratios for the mid to low altitude samples

range from 34,929 to 367,450 and the W_R is ranging from 1.6 to 103 mm/kyr. There is a significant variation between the maximum estimated weathering rates at low and high altitudes emphasizing role of the vegetation, temperature and rainfall at lower altitudes. The 5 fold increase in the maximum weathering rate at low altitudes is in agreement with previously reported vegetation impact on weathering rates (Benedetti et al., 1994; Moulton and Berner, 1998; Hinsinger et al., 2001).

The range value is in good agreement with the weathering rates reported for basaltic rocks corresponding to similar climatic conditions by Moreira Nordemann (1980) and Benedetti et al. (1994) for the Parana basalts in Brazil, as well as weathering rates based on the water/rock ratios estimated by Innocent et al. (1997) for the same area. It seems that a narrower range is reported in field-derived W_R studies for ultrabasic rocks in New Caledonia (i.e. 29-47 mm/kyr) (Trescases, 1975). Surprisingly and recognising that the equation used to calculate the weathering rate is oversimplified, the rates are in very good agreement with the rate calculated by Wang et al (1995) (i.e. 50 mm/kyr) using a model that takes into account rate laws, effective surface areas, reaction transport feedbacks.

These weathering rates could be underestimated since they do not take into account the suspended material. However in this study, the suspended part is negligible since very small amounts of suspended material were recovered on the filters during the water filtration process.

This means that in about 1500 years a weathered horizon with a thickness ranging from 0.2 to 20 cm can be created which corresponds to the thickness of the paleosoil profiles observed in the lower part of the volcano corresponding to older lavas. A paleosoil with a coal level located at 35 cm depth and dated at 1395±100 years (^{14}C dating) was studied in the area (Gérard et al., 1999). In other words, it took 1400 years for the soil to develop after the lava eruption over the previous soil profile. The coal level corresponds to the burned remains of the vegetation, growing on the previous soil profile that was covered by the lava flow 1400 years ago. However, as shown by Wirrmann et al., (2001) since 5400 yr B.P. a general trend towards drier climatic conditions is recorded suggesting that the mean annual rainfall value was higher than the present day one. This suggests that a coal level at about 35 cm depth corresponds to an average annual rainfall ranging from 7,363 to 58,880 mm/yr with water rock ratios ranging from 29,452 to

235,520, respectively. The lower range of the calculated rainfall values agrees with the presumed increase of the mean annual rainfall over this time range (Nguetsop et al., 1998; Wirrmann et al., 2001; Wirrmann personal communication).

Conclusions

The geochemistry of springs and river water samples recovered all around Mount Cameroon allowed us to identify some of the processes controlling the major and trace element behaviour in such water samples.

The mass balance calculations alone cannot account for the observed element ratios in the water of Mount Cameroon. The measurements and calculations indicate that Si and Ca geochemistry is partly controlled by plants via biological pumping as previously shown by other studies for different weathering systems. Atmospheric inputs of minerals from the quartz group or phytolites issued from plants have to be included in the mass balance calculations to account for the concentrations of Si and alkalinity values in the waters all around Mount Cameroon.
Water/rock ratios were estimated by means of both Sr isotope compositions and Sr contents. The values are in good agreement with previously published datasets. Knowledge of both these water/rock ratios and the water inputs calculated weathering rates of the basaltic lavas that range from 1 to 103 mm/kyr. These values suggest that in the Mount Cameroon area chemical weathering is the major driving force and that physical and biological disaggregation are apparently second order rate processes in the weathering of fresh basalt. The 5 fold increase between the maximum estimated weathering rates at high and low altitudes emphasizes the role of the vegetation, the temperature and the rainfall at lower altitudes.

This finding is supported by the age of Mount Cameroon paleosoils that assesses the elapsed time of the weathering based on the rates obtained from the water-rock ratio. However, future studies dedicated to the weathering at a microscopic scale should be undergone in order to, either rule out, or evidence the potential role of micro-organisms during the weathering of the lavas.

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Figure captions

Figure 1a: General location map showing the location of the Cameroon Line in West Africa (modified from Lee et al., 1994).

Figure 1b: Mount Cameroon sampling location map showing altitudes (m) and rainfalls expressed in mm per year (Sieffermann, 1973). The sample locations are reported as full circles for samples recovered during the wet season (march 1997), open circles dry season (December 1996) while samples recovered at both seasons are represented by patterned circles.

Figure 2: Chloride concentrations expressed in □M as function of the altitude (m). LA corresponds to samples recovered at low altitude (below 400 m), while MA corresponds to samples whose altitude range in between 400 and 1850m and HA characterizes samples higher than 1850 m. The open symbols correspond to dry season collected samples (December 1996) while the full symbols correspond to samples recovered during the short rainy season (March 1997). Circles are reported for low altitude samples. Medium altitude samples are plotted with diamonds while high altitude samples are reported with triangles.

Figure 3: (a) Alkalinity (□M) is reported versus dissolved silica (□M). **(b)** Mg (□M) versus dissolved silica (□M) and **(c)** Ca (□M) versus dissolved silica (□M). LA corresponds to samples recovered at low altitude (below 400 m), while MA corresponds to samples whose altitude range in between 400 and 1850m and HA characterizes samples higher than 1850 m. The open symbols correspond to dry season collected samples (December 1996) while the full symbols correspond to samples recovered during the short rainy season (March 1997). Circles are reported for low altitude samples. Medium altitude samples are plotted with diamonds while high altitude samples are reported with triangles.

Figure 4: (a) Mg (□M) is reported versus Alkalinity (□M). **(b)** Ca (□M) versus Alkalinity (□M) and **(c)** Ca (□M) versus Mg (□M). LA corresponds to samples recovered at low altitude

(below 400 m), while MA corresponds to samples whose altitude range in between 400 and 1850m and HA characterizes samples higher than 1850 m. The open symbols correspond to dry season collected samples (December 1996) while the full symbols correspond to samples recovered during the short rainy season (March 1997). Circles are reported for low altitude samples. Medium altitude samples are plotted with diamonds while high altitude samples are reported with triangles.

Figure 5: $^{87}Sr/^{86}Sr$ isotopic ratios are reported versus (1/Sr) expressed in $(\mu mol/L)^{-1}$. LA corresponds to samples recovered at low altitude (below 400 m), while MA corresponds to samples whose altitude range in between 400 and 1850m and HA characterizes samples higher than 1850 m. The open symbols correspond to dry season collected samples (December 1996) while the full symbols correspond to samples recovered during the short rainy season (March 1997). Circles are reported for low altitude samples. Medium altitude samples are plotted with diamonds while high altitude samples are reported with triangles. Basalt and associated soil profile developed onto sample $^{87}Sr/^{86}Sr$ isotopic ratios are also reported (Bulourde, 2001; Bulourde et al., 2002). The arrow figures the rain data out of scale.

Fig. 6: $^{234}U/^{234}U$ isotopic ratios are reported versus U/Sr ratios. LA corresponds to samples recovered at low altitude (below 400 m), while MA corresponds to samples whose altitude range in between 400 and 1850m and HA characterizes samples higher than 1850 m. The open symbols correspond to dry season collected samples (December 1996) while the full symbols correspond to samples recovered during the short rainy season (March 1997). Circles are reported for low altitude samples. Medium altitude samples are plotted with diamonds while high altitude samples are reported with triangles. The patterned domain corresponds to low altitude samples located in the northeast part while the low and medium altitude samples from the east and west flanks of the volcano correspond to the light grey domain. Samples recovered at high altitudes exhibit both low $^{234}U/^{234}U$ and U/Sr ratios.

Figs. 7: $^{87}Sr/^{86}Sr$ isotopic ratios are reported versus altitudes (m). LA corresponds to samples recovered at low altitude (below 400 m), while MA corresponds to samples whose altitude range

in between 400 and 1850m and HA characterizes samples higher than 1850 m. The open symbols correspond to dry season collected samples (December 1996) while the full symbols correspond to samples recovered during the short rainy season (March 1997). Circles are reported for low altitude samples. Medium altitude samples are plotted with diamonds while high altitude samples are reported with triangles.

Table captions :

Table 1 : Physico-chemical parameters of the water samples recovered all around the Mount Cameroon. n.m. refers to not measured, < q.l. refers to below the quantification limit and ** refers to a spring catchment for water supply. All samples were filtered at 0.1µm. Samples for cation content analysis were acidified.

Table 2: Sr and U concentrations expressed in µM and nM, respectively, as well as $^{87}Sr/^{86}Sr$ and $^{234}U/^{238}$ U isotopic ratios are reported. Error bars at the 95% confidence limit (2σ) are also reported as well as R corresponding to water/rock ratios (see text for calculation). Uranium data are taken from Riotte et al. 2002.

Table 3: Reactions involving the major minerals used in the mass-balance calculations. $Al(OH)_3°$ and $Fe(OH)_3°$ stand for all dissolved species of Fe and Al.

Table 4: Comparison between calculated and measured element ratios. r^2 is the correlation coefficient corresponding to the regression lines in Figs. 3 and 4.