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Basalt weathering laws and the impact of basalt weathering on the global carbon cycle

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Abstract

This study attempts to characterise the chemical weathering of basalts and to quantify the flux of carbon transferred from the atmosphere to the ocean during this major process at the surface of the Earth. To this aim, we have compiled different published chemical compositions of small rivers draining basalts. Basaltic river waters are characterised by relatively high Na-normalized molar ratios (Ca/Na: 0.2–3.9; HCO_3/Na: 1–10; Mg/Na: 0.15–6) in comparison with those usually observed for river draining silicates. The data also show the climatic influence on basalt weathering and associated CO_2 consumption. Runoff and temperature are the main parameters controlling the chemical weathering rate and derived CO_2 consumption during basaltic weathering. From these relationships and digital maps, we are able to define the contribution of basalts to the global silicate flux. Taking account of this result, we estimate that the CO_2 flux consumed by chemical weathering of basalts is about 4.08×10^{12} mol/year. The fluxes from the islands of Indonesia and regions of central America represent around 40% of this flux. The global flux of CO_2 consumed by chemical weathering of basalts represents between 30% and 35% of the flux derived from continental silicate determined by Gaillardet et al. [Chem. Geol. 159 (1999) 3]. Finally, it appears that volcanic activity not only acts as a major atmospheric CO_2 source, but also creates strong CO_2 sinks that cannot be neglected to better understand the geochemical and climatic evolution of the Earth.

Keywords: Basalt; Chemical weathering; Atmospheric CO_2 consumption; Rivers; Climate

1. Introduction

Increased interest in pollution and climatic changes has motivated studies of the chemical composition of river waters. As shown in pioneering works (Garrels and Mackenzie, 1971; Berner et al., 1983), it is not the total weathering of continents that affects the carbon cycle, but the weathering of silicates in particular. This process converts atmospheric CO_2 into bicarbonate in streams and, in the long term, leads to carbonate precipitation and sedimentation in the oceans. Since many studies have focused on river geochemistry on a global (Stallard and Edmond, 1983; Négrel et al.,

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1993; Edmond et al., 1995; Dupré et al., 1996; Gaillardet et al., 1997, 1999) and smaller scale (Meybeck, 1986; Bluth and Kump, 1994; White and Blum, 1995; Gislason et al., 1996; Louvat and Allègre, 1997; 1998; Stewart et al., 2001; Millot et al., 2002; Oliva et al., 2003) to quantify the role of silicate weathering in the carbon cycle and to determine the laws governing chemical weathering. The importance of lithology is emphasized by several authors (Meybeck, 1986; Amiotte-Suchet and Probst, 1993) who showed that basalts are among the more easily weathered than other crystalline silicate rocks. According to Louvat in her study on volcanic islands (Louvat, 1997; Louvat and Allègre, 1997, 1998), basalt weathering acts as a major atmospheric CO_2 sink. This idea has been reinforced by the studies of large basaltic provinces, for example that of Taylor and Lasaga (1999) on the rivers flowing through the Columbia River and of Dessert et al. (2001) on the Deccan Traps.

As basalts play a major role in the carbon cycle, it is important to estimate the contribution of basaltic rocks to the continental weathering flux. However, to date, the estimates of these fluxes are uncertain. First, we cannot use the world average river composition because it is difficult to characterize the proportion of elements coming from granitic or basaltic weathering. Further complication arises from the distribution of the different rock types at the Earth's surface. Indeed, many basaltic areas are located near the sea (e.g. volcanic arcs or oceanic islands), so that only small rivers flow through these formations. This explains why the currently available chemical data from rivers do not allow precise estimates of the global weathering flux coming from basalts and why these small "basaltic" rivers are not taken into account in global budgets (Berner et al., 1983; Meybeck, 1987; Gaillardet et al., 1999). However, even if individual river fluxes of dissolved material to the ocean are negligible compared to those of large rivers, these fluxes accumulated over the world-wide surface of volcanic provinces are not negligible. Consequently, to estimate this contribution, it is necessary to determine weathering basalt law and devise the best method of describing how these rocks weather under different climates.

Extensive experimental studies have been dedicated to the determination of the dependence of silicate mineral weathering rates on temperature (Guy and Schott, 1989; Lasaga et al., 1994; Blum and Stillings, 1995). The relationship between climate and chemical weathering has been more difficult to demonstrate in the field. Some authors (Dunne, 1978; Amiotte-Suchet and Probst, 1993; Bluth and Kump, 1994; Louvat, 1997) emphasized the effect of runoff on weathering rates, the effect of temperature (Meybeck, 1986; Velbel, 1993; Louvat, 1997), and the cumulated effect of runoff and temperature (White and Blum, 1995). Moreover, several authors have observed a coupling between chemical and physical denudation rates of silicates for large river systems (Gaillardet et al., 1999) and for small rivers draining granitic or basaltic rocks (Millot et al., 2002). These observations confirm the important role of soils on chemical weathering of silicates. It also appears that parameters such as vegetation, by generating organic acids (Drever, 1994; Oliva et al., 1999; Oelkers and Gislason, 2001), and the age of rock substrates (Gislason et al., 1996; Kennedy et al., 1998) can influence the chemical weathering of silicate.

We are interested at this stage in establishing first-order effects that influence basaltic weathering. Consequently, the aim of this study is to determine the relationship between the chemical weathering of continental basalts and climate, and to provide an initial estimate of the global CO_2 consumption rate during weathering of basalts. For this purpose, we studied selected set river chemistry data of different basaltic provinces where temperature and runoff conditions are contrasted. These relationships are combined with climatic data for each region to calculate the global flux coming from basalt weathering.

2. Origin of data

2.1. Concentrations of major elements

We have selected a set of basaltic regions for which the chemical composition of river water is known: Azores, Java and La Réunion Island (Louvat, 1997; Louvat and Allègre, 1997; 1998), Mount Cameroon (Riotte et al., 2003), Iceland (Gislason et al., 1996; Louvat, 1997), Parana Traps (Benedetti et al., 1994), Deccan Traps (Dessert et al., 2001), and Massif Central, in the South of France (Meybeck, 1986; Négrel and Deschamps, 1996). All these rivers were selectively sampled during the rainy and occasionally dry seasons. The chemical data of the Columbia Plateau (Appendix A) and of Hawaii (Appendix B) are taken from the National Water Data Exchange at the U.S. Geological Survey (monthly or quarterly sampling over a period of at least 3 years). For each region, we have determined discharge-weighted average concentrations (Table 1).

In this study we focus on mean bicarbonate concentration, mean cationic concentration (TDS_{cat}), and mean total dissolved solid (TDS_w) as indicators of chemical weathering of basalts (Table 1). The concentrations are corrected for atmospheric inputs using Cl concentrations and oceanic X/Cl molar ratios (where X = Na, Mg, Ca, K, and SO_4) and assuming that all the Cl content of the dissolved load is of atmospheric origin.

- Riverine bicarbonates reflect atmospheric CO_2 consumption during basalt weathering. Mean concentrations vary between 354 (Columbia Plateau) and 2720 μmol/l (Deccan). The consumption rate of atmospheric CO_2 was calculated using bicarbonate concentrations and runoff values during the sampling period (Table 1). Values of CO_2 consumption rates range from 0.30 and 6.41×10^6 mol/km^2/year and are higher than those for most of the world's largest rivers (Gaillardet et al., 1999).
- TDS_{cat} is the sum of concentrations of major cations (Ca, Mg, Na, and K) corrected for atmospheric inputs. TDS_{cat} range from 7.3 (Columbia Plateau) to 54 mg/l (Deccan) and the cationic fluxes range from 5.6 (M. Central) to 152 t/km^2/year (Java). The specific cationic fluxes for basalts are higher than those for granites (0.8–16.65 mg/l; Millot et al., 2002).
- TDS_w has been calculated from the concentrations of the major dissolved elements (SiO_2, Ca, Mg, Na, K, and SO_4) originating from basalt weathering. TDS_w values range from 20.9 (Iceland) to 84 mg/l (Java). The chemical denudation rate varies between 12.7 and 342 t/km^2/year.

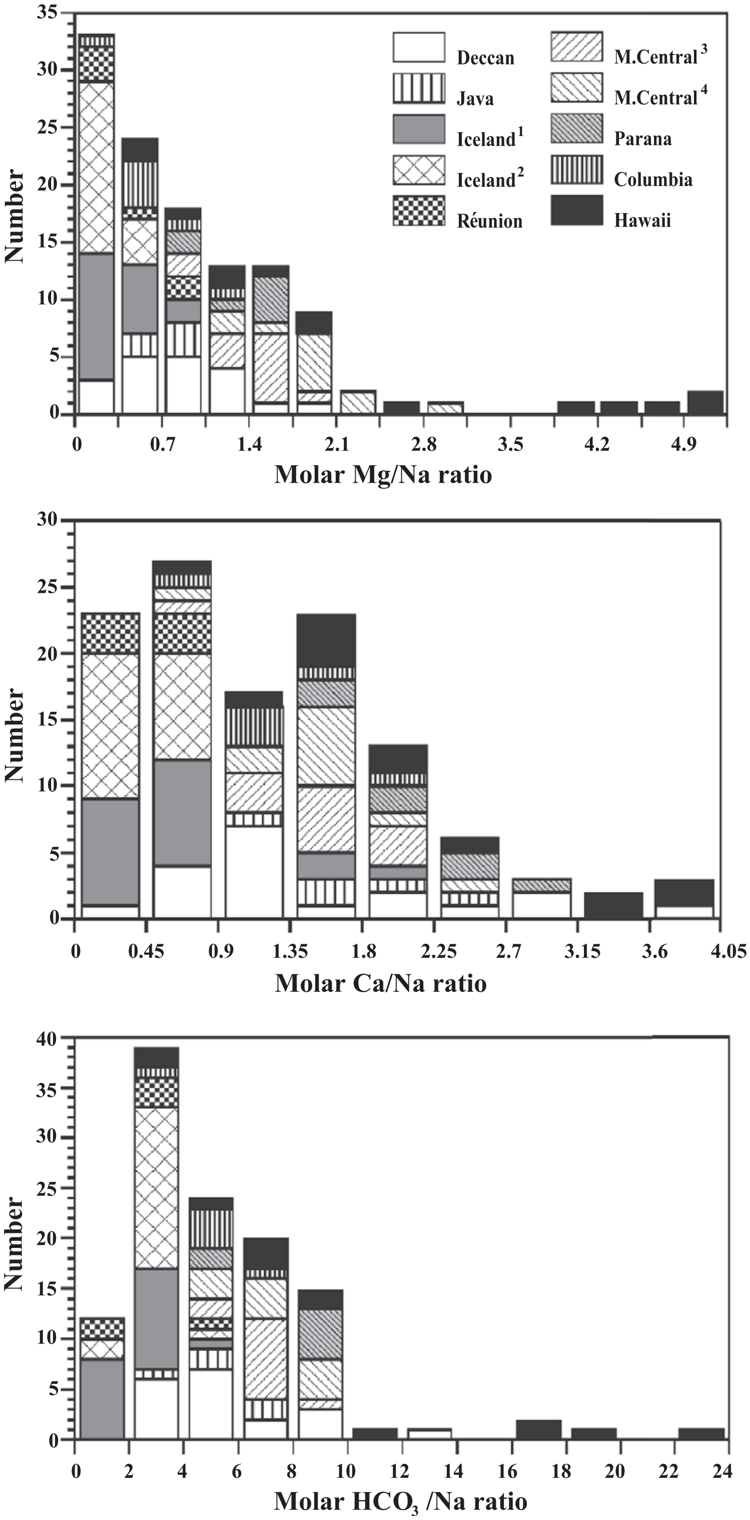
2.2. Elemental ratios of basaltic rivers

The elemental ratios of each river selected in this study are shown in Fig. 1. In contrast to absolute concentrations, elemental ratios avoid dilution and evaporation effects. To quantify basalt weathering, these ratios have been corrected for atmospheric inputs. Most of the HCO_3/Na molar ratios vary between 1 and 10 with a mean value close to 5.3; Ca/Na molar ratios range from 0.2 to 3.15 with a mean value around 1.3; Mg/Na molar ratios vary between 0.15 and 3.15 with a mean value around 1. Some rivers

Table 1
Mean solute concentrations, climatic parameters and rates for basaltic watersheds

Name	HCO_3^- (μmol/l)	TDS_{cat} (mg/l)	TDS_w (mg/l)	Runoff[a] (mm/year)	T[a] (°C)	CO_2 consumption rate (10^6 mol/km^2/year)	Cationic weathering rate (t/km^2/year)	Chemical weathering rate (t/km^2/year)
Columbia Plateau	354	7.3	23.0	1053	7.4[b]	0.37	7.7	24.2
Deccan Traps	2719	54.0	81.0	463	27[b]	1.26	25.1	37.1
Hawaii	407	7.4	21.3	1612	16.0	0.66	11.9	34.4
Iceland[c]	415	10.3	20.9	1883	2.0[b]	0.69	17.7	36.4
Iceland[d]	480	8.8	24.3	2432	2.0[b]	1.11	20.9	57.6
Java	1987	43.0	83.8	4052	24.8[b]	6.41	152.0	342.0
M. Central[e]	686	14.8	32.0	406	8.7[b]	0.30	5.6	12.7
M. Central[f]	698	13.0	32.4	478	8.7[b]	0.35	5.9	13.9
Mt. Cameroon	1623			2120[g]	25.6[b]	3.44		
Parana Traps	817	21.1	63.2	1020	20.2[b]	0.82	20.6	60.2
La Réunion	1311	29.9	57.1	2433	17.0	2.26	48.4	106.0
Sao Miguel	763	19.0	48.3	734	16.0	0.56	13.3	35.3

[a] Mean annual values.
[b] Value determined from the updated version of the IIASA database (Leemans and Cramer, 1991).
[c] Louvat (1997).
[d] Gislason et al. (1996).
[e] Négrel and Deschamps (1996).
[f] Meybeck (1986).
[g] Value determined from Cogley's (1998) database.


Deccan
Java
Iceland^1
Iceland^2
Réunion
M.Central^3
M.Central^4
Parana
Columbia
Hawaii
Number
0
5
10
15
20
25
30
35
0
0.7
1.4
2.1
2.8
3.5
4.2
4.9
Molar Mg/Na ratio
Number
0
5
10
15
20
25
30
0
0.45
0.9
1.35
1.8
2.25
2.7
3.15
3.6
4.05
Molar Ca/Na ratio
Number
0
5
10
15
20
25
30
35
40
0
2
4
6
8
10
12
14
16
18
20
22
24
Molar HCO_3 /Na ratio

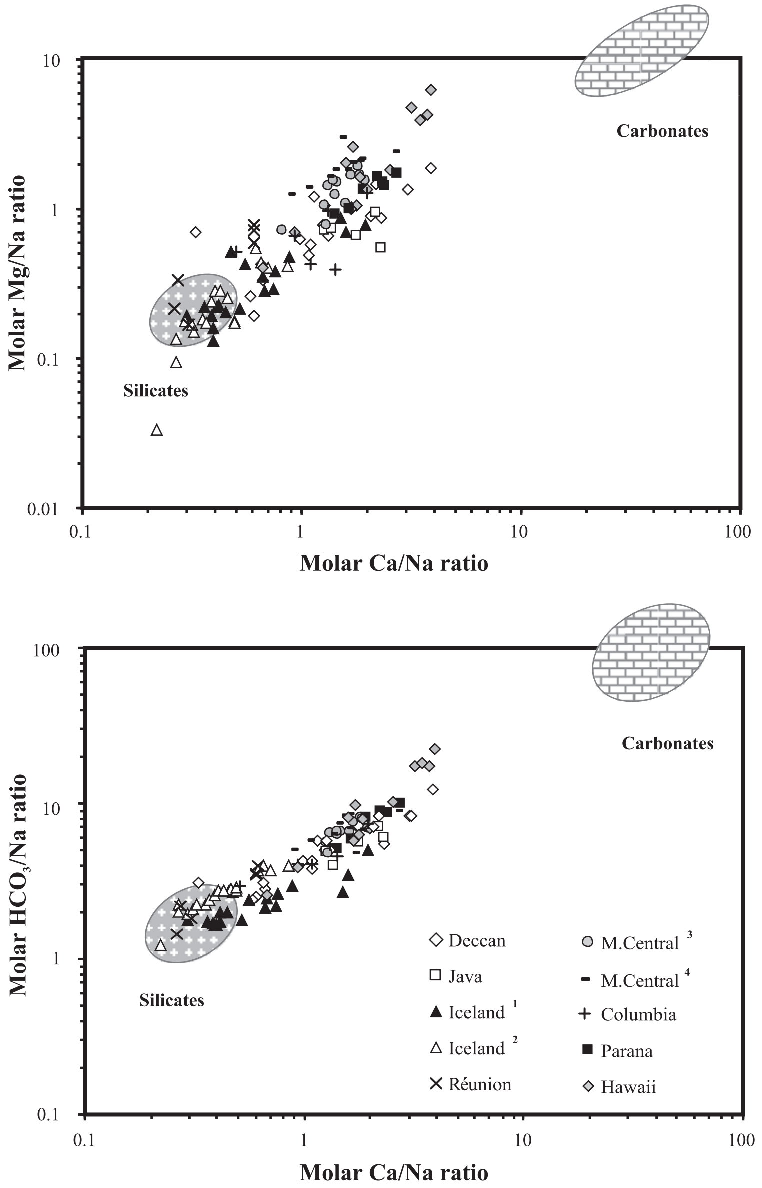


Fig. 2. Mg/Na vs. Ca/Na and HCO_3/Na vs. Ca/Na of basaltic rivers corrected for atmospheric inputs. End member compositions for granitic silicates and carbonates are those determined by Négrel et al. (1993) and by Gaillardet et al. (1999). [1]Louvat (1997); [2]Gislason et al. (1996); [3]Meybeck (1986); [4]Négrel and Deschamps (1996).

exhibit high Na-normalized ratios (HCO_3/Na>10; Ca/Na>3.15; Mg/Na>3.15), in particular rivers from Hawaii. The Na-normalized molar ratios of rivers draining basalts are relatively high in comparison with values usually observed for small streams draining granitoid rocks (Meybeck, 1986; Edmond et al., 1995;

Fig. 1. Distribution of Na-normalised molar ratios determined for each river considered in this study. The molar ratios are corrected for atmospheric inputs by using Cl concentrations and elemental ratios of seawater. [1]Louvat (1997); [2]Gislason et al. (1996); [3]Meybeck (1986); [4]Négrel and Deschamps (1996).

White and Blum, 1995). A good linear relationship is observed between molar ratios in the river waters and we can distinguish two groups of data (Fig. 2). The first, with relatively low molar ratios, is near the granitic silicate end member, while the second group lies between the granitic and carbonate end member. One explanation is that the trend in Fig. 2 is a mixing trend between waters draining granitic silicates and waters interacting with carbonates. As invoked by several authors (Mast and Drever, 1990; White et al., 1999) for granitic watersheds, this trend is consistent with the dissolution of disseminated calcite within basaltic rocks. These variations could also be interpreted as resulting from chemical weathering of more Ca and Mg rich volcanic rocks. The relatively low Ca/Mg ratios of Ca- and Mg-rich rivers (0.7–1.8) are consistent with those of basaltic rocks (0.9–3). While carbonates are characterised by higher Ca/Mg ratios; carbonate minerals are strongly enriched in Ca compared to Mg (Fig. 2). This latter observation suggests that the trend of Fig. 2 reflects principally trends of basalt chemistry. It is noteworthy that the chemical signatures of rivers draining basalts differ from those of rivers draining other silicate rocks (granite, gneiss, shale), with, in particular, higher Na-normalised molar ratios.

3. Effect of climate on chemical weathering

In Figs. 3 and 4 we have plotted various relation characterising the dependence of chemical weathering on runoff and temperature. Note that specific rates and runoff are not independent parameters since the rate is calculated from:

$$f = R_{\mathrm{f}} \times C \qquad (1)$$

where f is the specific weathering flux (10^6 mol/km^2/year or t/km^2/year), R_{f} is runoff and C is bicarbonate concentration (μmol/l), $\mathrm{TDS_{cat}}$ or $\mathrm{TDS_w}$ (mg/l). There is little scatter in the trends in bicarbonate, $\mathrm{TDS_{cat}}$, and $\mathrm{TDS_w}$ concentrations with temperature (Fig. 4) and thus concentrations are relatively independent of runoff as was observed in our previous study (Dessert et al., 2001). In other words, for a constant runoff, an observed increase of CO_2 consumption or weathering rate reflects an increase of temperature (Fig. 3). Nevertheless, this trend is not observed in the case of Hawaii; the temperature of Hawaii is similar to those of Sao Miguel and La Réunion while the concentrations of Hawaiian rivers are relatively low.

The effect of temperature on basalt chemical weathering is shown in Fig. 4 where a good correlation is observed between the different concentrations and surface temperature, except for Hawaii. Although Hawaii does not follow the general trends, it is taken into account in the regression analysis (Fig. 4). Combining these correlation equations with Eq. (1) gives:

$$f_{\mathrm{CO_2}} = R_{\mathrm{f}} \times 323.44\ \exp(0.0642\ T) \qquad (2)$$

where $f_{\mathrm{CO_2}}$ is the specific atmospheric CO_2 consumption rate (mol/km^2/year), R_{f} is in mm/year and T in °C.

$$f_{\mathrm{cat}} = R_{\mathrm{f}} \times 6.69\ \exp(0.0677\ T) \qquad (3)$$

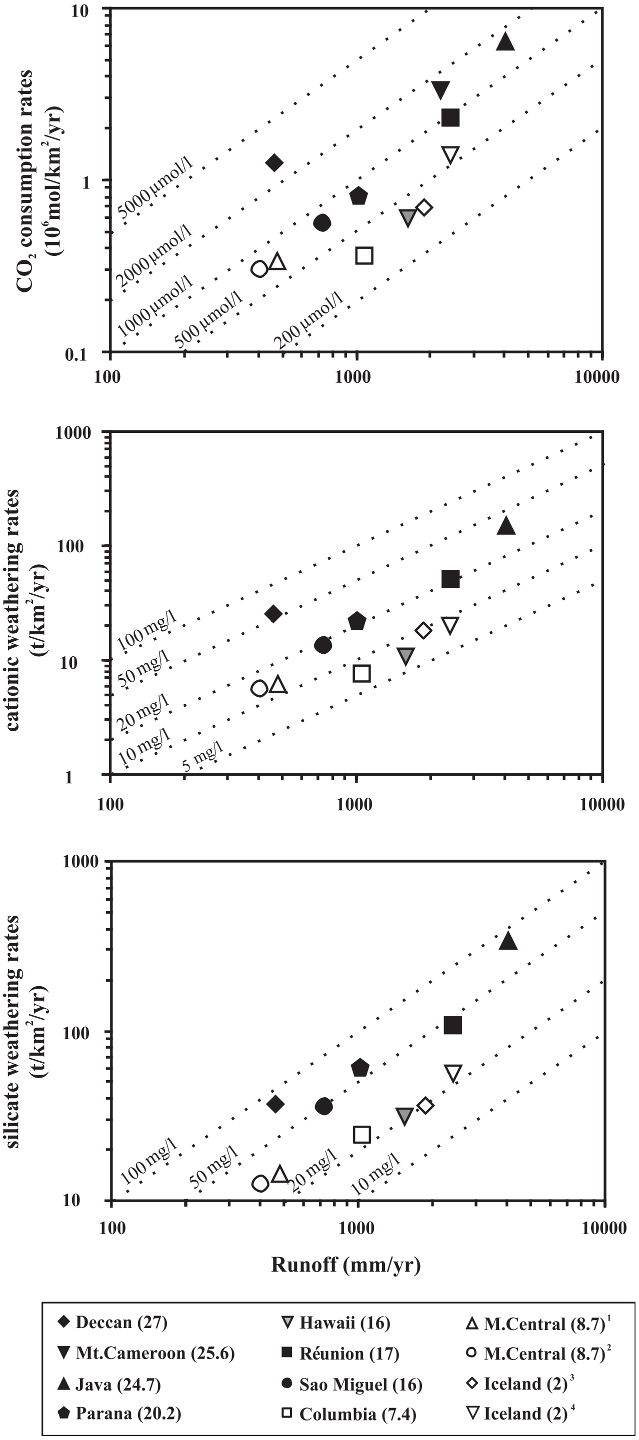
where f_{cat} is the specific cationic weathering rate (t/km^2/year).

$$f_{\mathrm{w}} = R_{\mathrm{f}} \times 18.41\ \exp(0.0553\ T) \qquad (4)$$

where f_{w} is the specific silicate weathering rate (t/km^2/year).

Our relationship differs from those of Amiotte-Suchet and Probst (1995) and Bluth and Kump (1994). These authors observed, from analysis of flux data of watersheds composed of basaltic rocks, a relationship describing weathering fluxes as a function of runoff. Our approach differs from these authors in two respects. We take into account provinces with contrasting temperatures, in contrast to Amiotte-Suchet and Probst who based their study on the data published by Meybeck (1986) on French basaltic basins exhibiting only small temperature variations. For this reason, only the effect of runoff can be

Fig. 3. Plots of mean atmospheric CO_2 consumption rates, cationic weathering rates and silicate weathering rates versus runoff (during sampling period). Dashed straight lines reported in this figure represent constant values of bicarbonate concentrations, $\mathrm{TDS_{cat}}$ and $\mathrm{TDS_w}$. The values in parentheses correspond to the mean surface temperature (during sampling period) determined from an updated version of the IIASA climatological database (Leemans and Cramer, 1991). The black symbols characterise rivers with relatively high surface temperature (>16 °C), the white ones rivers with relatively low temperature (< 10 °C) and the grey one, the particular rivers of Hawaii. [1]Meybeck (1986); [2]Négrel and Deschamps (1996); [3]Louvat (1997); [4]Gislason et al. (1996).


CO_2 consumption rates (10^6mol/km^2/yr)
10
1
0.1
100
1000
10000
5000 µmol/l
2000 µmol/l
1000 µmol/l
500 µmol/l
200 µmol/l
cationic weathering rates (t/km^2/yr)
1000
100
10
1
100
1000
10000
100 mg/l
50 mg/l
20 mg/l
10 mg/l
5 mg/l
silicate weathering rates (t/km^2/yr)
1000
100
10
100
1000
10000
100 mg/l
50 mg/l
20 mg/l
10 mg/l
Runoff (mm/yr)
◆ Deccan (27)
▼ Mt.Cameroon (25.6)
▲ Java (24.7)
⬟ Parana (20.2)
▽ Hawaii (16)
■ Réunion (17)
● Sao Miguel (16)
□ Columbia (7.4)
△ M.Central (8.7)[1]
○ M.Central (8.7)[2]
◇ Iceland (2)[3]
▽ Iceland (2)[4]

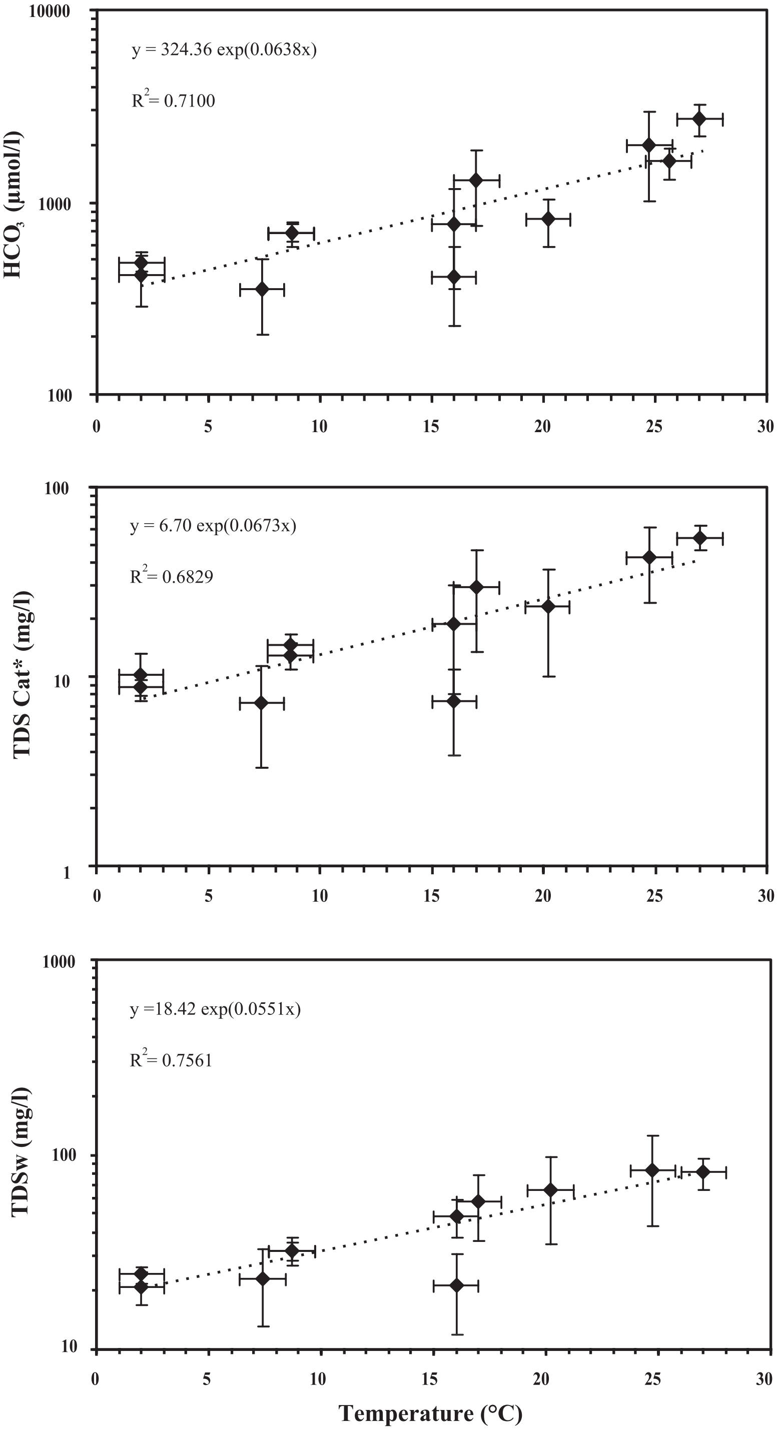


Fig. 4. Plots of mean bicarbonate concentrations, TDS_{cat} and TDS_w versus mean surface temperature. A good relationship is observed between the two parameters (exponential equation).

observed. We use mean data for each basaltic province, while Bluth and Kump (1994) used, for each watershed, river waters sampled once a month over a period of at least 2 years. For these reasons, we attempted to determine from Fig. 3 the relationship between runoff and flux ("runoff law") as it was induced by Amiotte-Suchet and Probst (1995) and Bluth and Kump (1994). The CO_2 consumption rates calculated from this "runoff law" as well as those computed from Eq. (2) are compared with measured values in Fig. 5. These comparisons illustrate that explicit account of temperature improves substantially the description of basalt chemical weathering rates. Note the Hawaii basalt chemical weathering rates are poorly described by both functions. It seems however, according to Chadwick et al. (1999) that Hawaii is not comparable with other basaltic continental settings. Similar trends are also observed for the silicate and cationic weathering rates. This observation confirms the major influence of both runoff and temperature on weathering rates and associated CO_2 consumption as stated previously by several authors (White and Blum, 1995; Louvat, 1997; Dessert et al., 2001).

To conclude, based on the different studies of basaltic rivers, it appears that knowledge of runoff and temperature is sufficient to characterise chemical weathering and the associated atmospheric CO_2 consumption rates. The relationship determined in this study can be used in climate models.

4. Global present-day consumption of atmospheric CO_2 from basalt weathering

Solute fluxes coming from chemical weathering of basalts should be taken into account to estimate global erosion rates. To date, estimates of these fluxes are uncertain. World river average composition data are insufficient because it is difficult to determine the proportion of elements coming from granite or basalt weathering. Moreover the distribution of the different types of rocks at the Earth's surface is difficult to quantify many basaltic terrains are located near the sea (island arc volcanism or oceanic basalt) and are drained by small numerous coastal rivers, from which chemical data are rare. In this paper, we estimate the

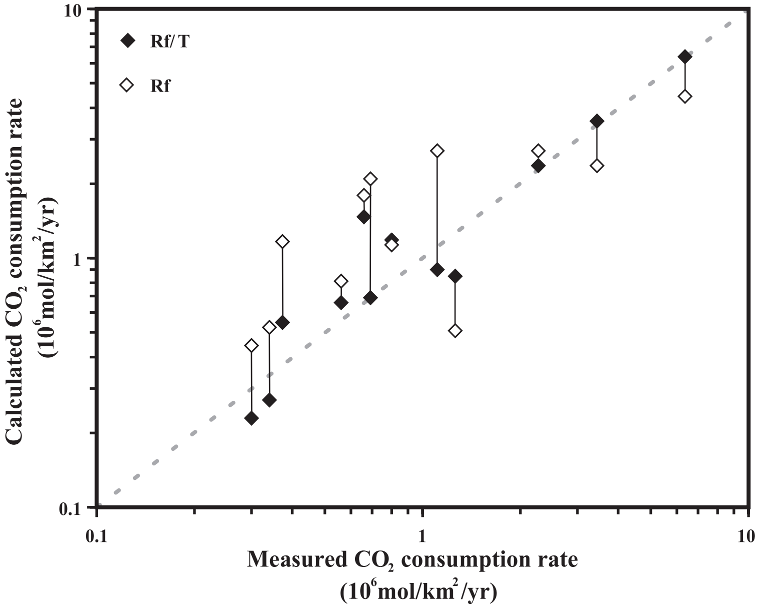


Fig. 5. Calculated CO_2 consumption rates versus measured CO_2 consumption rates. The rates determined from our relationship (as a function of R_f and T) are represented by black symbols and those determined as a function of R_f are represented by white symbols. Globally the rates determined from runoff and temperature are in better agreement with the measured rates than those determined as only a function of runoff.

contribution of basalt weathering to the atmospheric CO_2 consumption flux of each basaltic province using Eq. (2), a global geologic map, and the spatial distribution of runoff and temperature. This approach parallels that of Gibbs, Probst and colleagues (Amiotte-Suchet and Probst, 1993; Amiotte-Suchet and Probst, 1995; Gibbs et al., 1999) who seek to better understand the spatially distributed controls on CO_2 consumption rates. In the case of Amiotte-Suchet and Probst (1995) global CO_2 consumption was estimated using a relationship between runoff and bicarbonate fluxes for different rock types and maps of lithology and drainage intensity with resolution of 1 × 1°. Gibbs et al. (1999) added an Arrhenius-type temperature dependency in their model that modifies the runoff-determined fluxes for each rock type and the CO_2 consumption calculations are performed over a grid of 4.5° latitude by 7.5° longitude.

In our study, we use a digital geologic map with a resolution of 1 × 1° representing the global distribution of exposed basalts. This simplified map has been produced from global geologic maps published by UNESCO (1976), lithologic map of Amiotte-Suchet et al. (2003), and simplified geologic maps (Souther et al., 1984; Mangan et al., 1986; Coffin and Eldholm, 1994; Wignall, 2001). For each grid cell, the mean annual runoff (mm/year) is obtained from a digitised version (Cogley, 1998) of the UNESCO Atlas of world water balance (Korzoun et al., 1977) and mean annual temperature (°C) is taken from an updated (unpublished) version of the IIASA database (Leemans and Cramer, 1991). The details of each major province are listed in Table 2. From the lithological map, we obtain a surface area of 6.8×10^6 km^2 for continental basalts (including continental flood basalt provinces and arc volcanic regions). This surface area represents about 4.6% of the continental surface or about 8.4% of the global silicate area (granite, gneiss, basalt, shales, ...). From the relative surface areas of basalts and of other acid silicates, such as granite and gneiss, an initial estimate of the relative contribution of basalts to the global CO_2 consumption flux can be obtained. The global CO_2 consumption flux from silicate weathering (F_{tot}) can be written as:

$$F_{tot} = 0.916 \times A_{tot} \times f_{sil} + 0.084 \times A_{tot} \times f_{bas} \qquad (5)$$

where f_{bas} and f_{sil} represent CO_2 consumption rates (i.e., fluxes per unit area) coming from the weathering of respectively basalts and other silicates, A_{tot} is the global area of silicates.

Table 2

Characteristic of each basaltic province determined from various digitised maps: combination of lithological maps for the area (see text), Cogley (1998) for runoff and an updated version of the IIASA database (Leemans and Cramer, 1991) for temperature

Name	Area (10^6 km^2)	Runoff (mm/year)	Annual T (°C)	CO_2 consumption flux (10^{12} mol/year)
Ethiopia	0.807	129	21.3	0.121
Siberia	0.796	402	−10	0.053
Parana	0.568	498	19.8	0.318
SE Asia/Indonesia	0.538	1372	25	1.033
Deccan	0.532	424	26.9	0.392
Japan/E Russia	0.373	731	3	0.151
Australia/Tasmania	0.327	176	20.4	0.047
Central America	0.309	763	22.8	0.356
Arabic Peninsula	0.219	4	23.3	0.001
South Africa	0.216	102	19.3	0.024
Patagonia	0.209	151	9.3	0.018
Kamchatka	0.173	750	−3.5	0.031
Columbia River	0.154	371	9.5	0.034
Iceland	0.105	1889	2	0.070
Greenland	0.099	364	−11	0.004
Cascade/Snake River	0.081	404	7.2	0.017
E Canada	0.038	441	1.4	0.006
Other[a]	1.305	420	18.6	0.433
Total	6.849			3.109

[a] Antarctica not included.

From this expression, we can derive the proportion of atmospheric CO_2 consumed by basalt weathering with respect to total CO_2 consumption by silicate weathering:

$$\frac{F_{bas}}{F_{tot}} = \frac{0.084 \times B}{0.916 + 0.084 \times B} \qquad (6)$$

where $B = f_{bas}/f_{sil}$ is the relative weathering rate of basalts compared to other silicates.

According to Dessert et al. (2001) the chemical weathering rate of volcanic rocks is 5–10 times higher than the chemical weathering rate of granite and gneiss. In such a case, the CO_2 consumption flux derived from basalts represents between 31% and 48% of the global flux from all silicates. Although this initial estimate of CO_2 consumption by basalt weathering depends of the choice of the surface area of silicate rocks, it illustrates the non-negligible role played by basalts in the global CO_2 consumption by weathering.

The CO_2 consumption flux can be estimated more precisely from Eq. (2) and the digitised maps of mean annual runoff and temperature to estimate the weathering rate on each $1\times1°$ basaltic pixel of the lithological map. By considering the area of all basaltic provinces, we obtain a total CO_2 consumption flux of 3.11×10^{12} mol/year. The CO_2 consumption fluxes for each basaltic province are listed in Table 2 and represented graphically in Fig. 6. It is interesting to note that the fluxes coming from islands of Indonesia and regions of Central America represent around 40% of the global atmospheric CO_2 flux derived from weathering of basalts. These provinces are not the largest in terms of area, but optimal climatic conditions, including high temperature and runoff, combined to produce high CO_2 consumption rates. Note the rather important contribution of the weathering of Deccan and Parana Traps and the small impact of large basaltic areas as Siberian and Ethiopian Traps. In the case of Siberia, both temperature and runoff are too low to favour the chemical weathering of basalts. In the case of Ethiopia, in spite of high temperature, the runoff is also too low to produce important chemical weathering and associated atmospheric CO_2 consumption rates.

It is important to note that the global estimate is probably too small owing to the fact that, at a resolution of $1\times1°$, we do not take into account small basaltic regions, primarily volcanic islands. However, the studies of Louvat showed that the chemical weathering of volcanic islands is important and not negligible to the carbon cycle. The surface of total volcanic islands was taken from the estimate of Gaillardet et al. (1999) of 1.5×10^6 km^2 which corresponds to the surface of oceanic and arc islands including all small islands as well as larger regions as Kamchatka, Iceland, and Indonesia (total area of 1.1×10^6 km^2). By a simple subtraction we obtain a surface area around 0.4×10^6 km^2 corresponding to small volcanic islands. From the data of Java, Sao Miguel, La Réunion and Hawaii islands (Table 1), we determined a mean specific CO_2 consumption rate of 2.47×10^6 mol/km^2/year. We estimate therefore, that the weathering of these small regions consumes 0.97×10^{12} mol/year. We finally propose that the global present-day CO_2 flux derived from chemical weathering of basalts should approach 4.08×10^{12} mol/year. This number

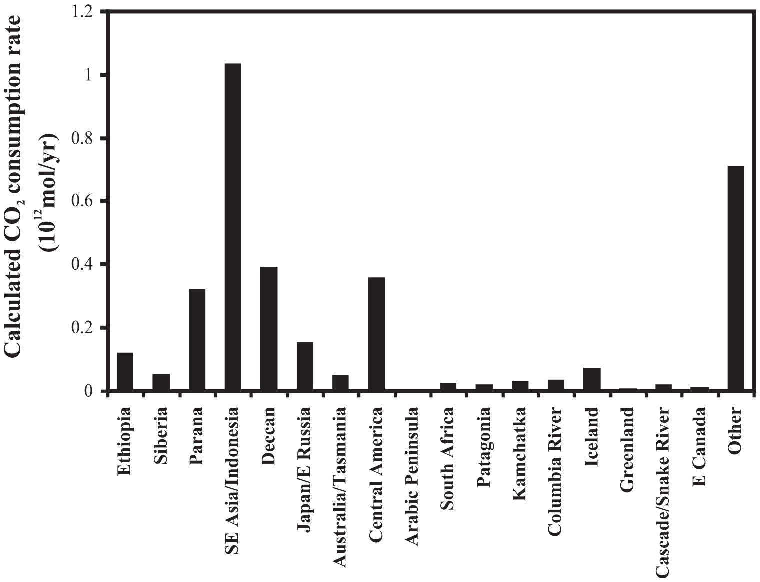


Fig. 6. Diagram showing for each basaltic region, the CO_2 consumption rates (10^{12} mol/year) derived from the weathering of basalts. The regions are presented in decreasing order of surface area.

remains preliminary and more studies (sampling and modelling) are needed to refine this estimate.

5. Implication of basalt weathering in global silicate CO_2 consumption

The global flux of atmospheric CO_2 consumption rates due to silicate weathering has been determined by different methods (Holland, 1978; Berner et al., 1983; Meybeck, 1987; Amiotte-Suchet and Probst, 1995; Gaillardet et al., 1999). In the recent study of Gaillardet et al. (1999), the global CO_2 consumption flux coming from silicate weathering is calculated from the global flux carried by large rivers and from an estimate of CO_2 consumed during weathering of basalts from volcanic arcs and volcanic islands (according the results of Louvat, 1997). In this approach the silicate component is extracted from the total dissolved load by taking into account the major reservoirs that contribute to river chemistry, i.e. rainwater, carbonate and evaporite derived waters. Each end member is characterised by Na-normalized molar ratios. In their first estimate, the silicate end member is characterised by $Ca/Na = 0.35 \pm 0.15$; $HCO_3/Na = 2 \pm 1$; $Mg/Na = 0.24 \pm 0.12$ and gives a CO_2 consumption rate of 11.7×10^{12} mol/year for silicate weathering. In a second estimate, the authors used higher Na-normalized molar ratios ($Ca/Na = 1 \pm 0.4$; $HCO_3/Na = 3 \pm 1$; $Mg/Na = 0.6 \pm 0.2$) and obtained a CO_2 consumption rate of 13.6×10^{12} mol/year.

We observed in this study that Na-normalized molar ratios of rivers draining basalts are relatively high ($Ca/Na>1$; $HCO_3/Na>3$; $Mg/Na>1$). Recent studies (Millot et al., 2002; Oliva et al., 2003) have also observed high Na-normalized molar ratios with $Ca/Na>0.5$, from rivers draining acid silicate rocks (granite, charnockite gneiss, etc.). All these studies show that the silicate end member can also have relatively high Na-normalized molar ratios. These observations show that the global CO_2 consumption rate derived from silicate weathering is probably intermediate between the two estimates of Gaillardet et al. (1999). Our approach provides a CO_2 consumption rate coming from basalt weathering of 4.08×10^{12} mol/year, representing between 30% and 35% of the flux derived from continental silicate as determined by Gaillardet et al. (1999).

Thus, based on studies of basaltic rivers and different digital maps, we conclude that the weathering of continental basalts is of great importance to the global consumption of atmospheric CO_2 by silicate weathering.

6. Discussion: carbon cycle and climate

During the past decades several authors attempted to explain the variations of the Earth's surface temperature over geological times (Walker et al., 1981; Berner et al., 1983). These studies suggested that the Earth's surface temperature is stabilized, on a large scale, by a negative feedback linking atmospheric CO_2, climate, runoff, and silicate weathering. Since then, this mechanism has been used in many models reconstructing the carbon cycle changes over geological time scales (Berner, 1991, 1994; François and Walker, 1992; Goddéris and François, 1995; Wallman, 2001). Other authors (Raymo et al., 1988; Raymo and Ruddiman, 1992) argued that continental orogenesis is the primary factor determining the rate of chemical weathering and thus Earth's climate. This hypothesis is based on the observation that global climate cooled when global cation fluxes were enhanced by weathering associated with the rapid erosion of the Himalaya. Moreover, the organic carbon burial during sediment deposition adds to the increase of chemical weathering and associated CO_2 consumption induced by mountain building (Raymo, 1994; France-Lanord and Derry, 1997; Hayes et al., 1999).

The relationship initially observed between chemical and physical weathering rates of the large rivers (Gaillardet et al., 1999) and on small rivers draining one rock type (Millot et al., 2002) confirms the influence of mountain building on the carbon cycle. On the other hand, the relationships observed on small rivers draining basalts also confirm the dependence of chemical weathering on temperature and thus the existence of a regulation mechanism for atmospheric CO_2. In the case of rivers draining granites, this dependence with temperature is unclear (Millot et al., 2002; Oliva et al., 2003) and it appears that, beside the physical erosion, the dominant factor is runoff. However, even if this parameter is roughly constrained, it was envisaged that global runoff and global surface temperature are linked (Berner et al., 1983).

Our results suggest that the surface area of continental basalts plays a major role in the carbon cycle. It is important to note that the estimate of 4.08×10^{12} mol/year of CO_2 consumed by the weathering of basalts was determined in this study from the present-day basalt surface area. This present-day estimate shows the importance of basalt weathering on the global carbon cycle, an idea previously put forward by several authors (Louvat and Allègre, 1997; Taylor and Lasaga, 1999; Dessert et al., 2001). However, the surface area of basalts, and in particular those of large igneous provinces, has decreased over geologic times. For example, the original area of the Deccan and Siberian Traps were estimated to have been three to five times larger than today. Therefore, we can suppose that the flux coming from basalt weathering has evolved with time. Weathering has also been changed by variations of climate in the past. It is now relatively well established that the emplacement of volcanic rocks induces geochemical and climatic changes over the short-term (Bluth et al., 1992; Genin et al., 1995) and over the long-term (Taylor and Lasaga, 1999; Dessert et al., 2001; Wignall, 2001). On the other hand, the continents have migrated, through plate tectonics, and thus have crossed different climatic zones (e.g. Deccan Traps have been moving from the southern to the northern hemisphere; Courtillot et al., 1999. This set of observations suggests that the different fluxes derived from basalt weathering could have been higher in the past and consequently that the impact of this chemical weathering on geochemical cycles including the carbon cycle could be underestimated.

7. Conclusion

This study examined the chemical composition of river waters in basaltic environments to characterise the chemical weathering of basalts and quantify the atmospheric CO_2 consumption rates associated with basalt weathering.

- The waters from rivers draining basalts have relatively high Na-normalized molar ratios (Ca/Na: 0.2–3.9 ; HCO_3/Na: 1–10 ; Mg/Na: 0.15–6) in comparison with that observed for river waters draining silicates.
- It is suggested that runoff and temperature are the most important parameters controlling chemical weathering rates and the associated atmospheric CO_2 consumption rates. The different fluxes are determined from simple relationships between runoff and surface temperature. Thanks to these relationships, it is now possible to estimate, for any basaltic province, the chemical weathering rates and associated CO_2 consumption rates, provided that the temperature and the runoff are known.
- From these relationships and digital maps, we determined a CO_2 consumption rate of about 4.08×10^{12} mol/year, that represents between 30% and 35% of the flux derived from continental silicate determined by Gaillardet et al. (1999).

This study reinforces the importance of basaltic chemical weathering to the global flux derived from silicate weathering previously emphasized by several authors (Louvat, 1997; Taylor and Lasaga, 1999; Dessert et al., 2001). It appears now that volcanic activity acts not only as a major CO_2 source, but also creates strong CO_2 sinks which cannot be neglected when attempting to improve our understanding the geochemical and climatic evolution of the Earth. In the future, an important effort should be made to obtain more complete data of rivers flowing through basalts to better understand the processes governing chemical weathering of basalts and to emphasize the impact of other parameters including vegetation and topography.

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Appendix A. USGS stream gauging stations of Columbia Plateau

Station number	River name	Location	Period of record	Count	Area km^2	Runoff^{a} (mm/yr)	$HCO_3^{-\,b}$ (μmol/l)	TDS_w^b (mg/l)	TDS_{cat}^b (mg/l)
14048000	John Day	Mcdonalds Ferry, OR	02/90–08/94	18	19,647	72	1413	59.8	28.0
14103000	Deschutes	Biggs, OR	02/85–09/90	33	27,216	180	1198	56.7	24.6
14113000	Klickitat	Pitt, WA	01/80–07/86	51	3362	450	646	43.2	13.2
14123500	White Salmon	Underwood, WA	08/60–09/70	64	1000	1007	511	36.6	10.7
14143500	Washougal	Washougal, WA	10/64–09/70	52	280	2548	174	11.9	3.5
14223500	Kalama	Kalama, WA	10/61–07/70	30	513	2058	325	21.3	6.8

(a) Mean annual runoff determined from daily streamflow (USGS). (b) Mean weighted concentrations.

At the scale of a single watershed and on a long-term seasonal basis, it is observed that concentrations decrease with increasing runoff (except for the Deschutes River where no sampling has been carried out during rising). As it is shown in Fig. 7, this trend is also observed at a broad scale when the six basins are plotted together (this correlation is also observed for TDS_w and TDS_{cat}). No relationship is observed with temperature since the mean annual temperature varies by only a few degrees from one basin to the other. Nevertheless, stream chemistry is not solely controlled by a simple dilution because

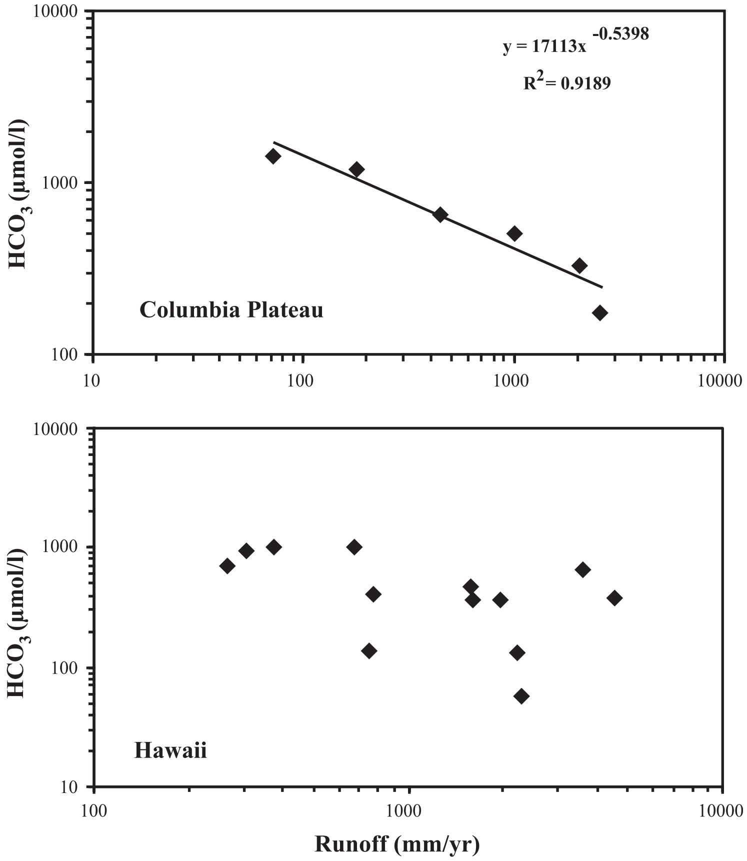

Fig. 7. Bicarbonate concentrations versus runoff for selected rivers of Columbia Plateau and Hawaii.

the bicarbonate flux increases with increasing runoff. These data confirm, on the scale of watershed, the important influence of runoff on river chemistry. Moreover, in this province, the sampling of each basin, in hydrologic terms, seems representative of the annual variations. Since, the mean annual runoff reconstructed from the discharges associated with chemical analyses is similar to those determined by USGS from daily streamflows. Consequently, these data appear to characterise pretty well the global chemical weathering of the basalts of Columbia Plateau.

Appendix B. USGS stream gauging stations of Hawaiian islands

Station number	River name	Location	Period of record	Count	Area (km^2)	Runoff[a] (mm/year)	HCO_3^- [b] (μmol/l)	TDS_w^b (mg/l)	TDS_{cat}^b (mg/l)
16049000	Hanapepe	Kauai	02/71–04/77	14	48	1615	370	16.7	7.0
16060000	Wailua	Kauai	02/71–07/77	13	58	1587	476	20.5	7.7
16103000	Hanalei	Kauai	02/71–11/76	11	49	3600	655	28.3	10.8
16108000	Wainiha	Kauai	08/71–03/77	12	26	4524	384	21.2	6.6
16211600	Makaha	Oahu	04/70–02/77	13	6	265	694	40.9	12.3
16212800	Kipapa	Oahu	03/70–03/77	11	11	755	137	8.5	2.9
16213000	Waikele	Oahu	02/90–09/95	33	118	305	927	66.1	20.1
16229300	Kalihi	Oahu	02/90–08/93	14	13	376	1024	42.5	21.8
16275000	Haiku	Oahu	04/70–02/77	16	2.5	673	1001	43.6	17.8
16400000	Halawa	Molokai	01/75–07/94	66	12	2295	57	7.1	2.0
16405500	Waikolu	Molokai	03/70–06/76	14	5	775	410	29.3	8.5
16618000	Kahakuloa	Maui	01/75–08/95	86	9	1951	364	24.6	7.0
16620000	Honokolau	Maui	01/72–12/76	10	11	2236	130	9.0	2.7

(a) Mean annual runoff determined from daily streamflow (USGS). (b) Mean weighted concentrations.

By considering each individual basin, we observe on a seasonal basis an overall decrease of concentrations with runoff (this decrease is not observed in four basins of Oahu island). However, if we consider all rivers, no relationship appears between concentrations and runoff and the major difficulty is to determine a mean characteristic point of Hawaiian rivers. This random variability is illustrated in Fig. 7 where all Hawaiian rivers are represented (this dispersion is also observed for cationic and silicate weathering rates). Conversely to Columbia province, the mean annual runoff reconstructed from the water discharges associated with chemical analyses is quite different to those determined by USGS from daily streamflow (between 5% and 160% of difference). Moreover, several authors have emphasized the variable factors on Hawaii influencing the chemical weathering as rainfall, soil thickness, rock age, topography, eolian inputs (Kennedy et al., 1998; Chadwick et al., 1999; Stewart et al., 2001). These factors point to the need of more precise data to better understand the chemical weathering of Hawaiian Islands. Nevertheless, the aim of this study is not to characterise the specific chemical weathering of Hawaiian basalts but the chemical weathering of basalts at the global scale. We have thus determined mean values for Hawaii (Table 1) by calculating weighted average of the concentrations for all individual basins. We are aware that it is debatable to use an average river composition to characterise Hawaii. However, it seems also inappropriate to consider separately each Hawaiian watershed in this study where other basaltic provinces (both traps and volcanic islands) are characterized by mean values.

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