



## Temperature dependence of basalt weathering



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### ABSTRACT

The homeostatic balance of Earth's long-term carbon cycle and the equable state of Earth's climate are maintained by negative feedbacks between the levels of atmospheric CO<sub>2</sub> and the chemical weathering rate of silicate rocks. Though clearly demonstrated by well-controlled laboratory dissolution experiments, the temperature dependence of silicate weathering rates, hypothesized to play a central role in these weathering feedbacks, has been difficult to quantify clearly in natural settings at landscape scale. By compiling data from basaltic catchments worldwide and considering only inactive volcanic fields (IVFs), here we show that the rate of CO<sub>2</sub> consumption associated with the weathering of basaltic rocks is strongly correlated with mean annual temperature (MAT) as predicted by chemical kinetics. Relations between temperature and CO<sub>2</sub> consumption rate for active volcanic fields (AVFs) are complicated by other factors such as eruption age, hydrothermal activity, and hydrological complexities. On the basis of this updated data compilation we are not able to distinguish whether or not there is a significant runoff control on basalt weathering rates. Nonetheless, the simple temperature control as observed in this global dataset implies that basalt weathering could be an effective mechanism for Earth to modulate long-term carbon cycle perturbations.

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### 1. Introduction

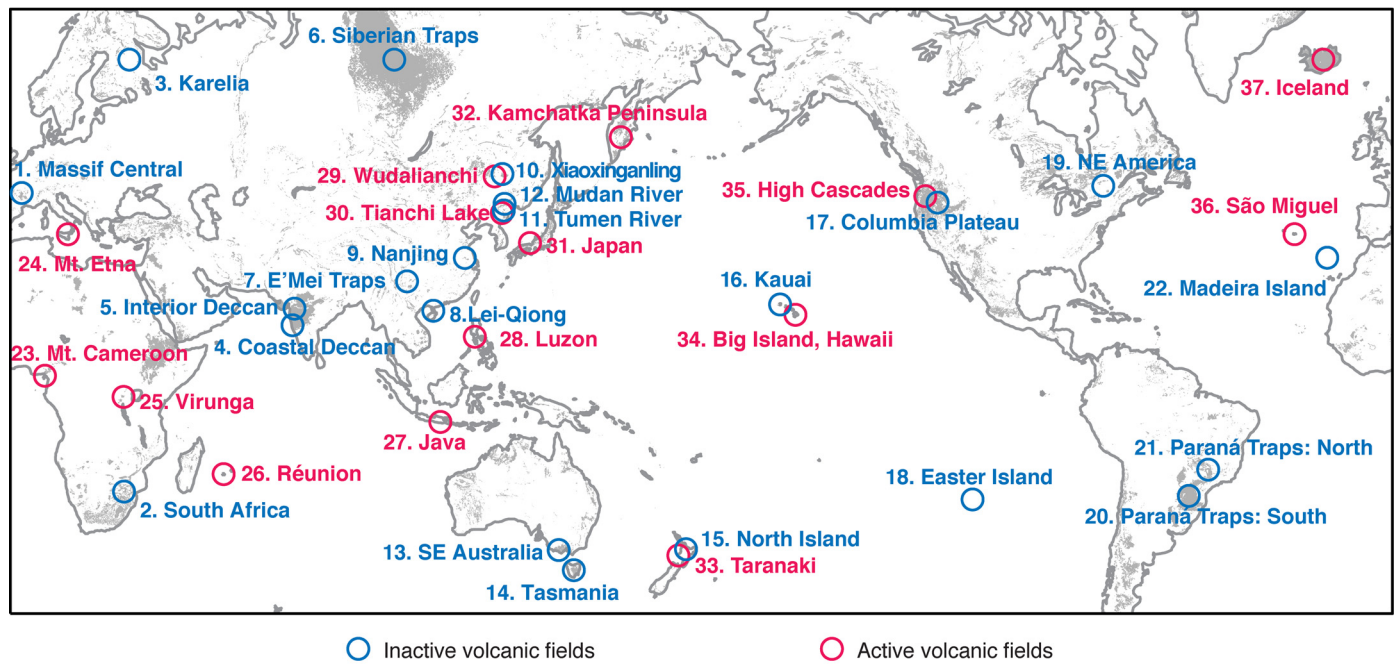
The control of climate on the rate of silicate mineral weathering and thus the burial of atmospheric CO<sub>2</sub> in carbonate minerals over geologic time provides a negative feedback stabilizing changes in Earth's atmospheric CO<sub>2</sub> concentration (*p*CO<sub>2</sub>). Such a *p*CO<sub>2</sub>-weathering feedback is believed to have maintained the homeostatic balance of the long-term carbon cycle and the habitability of Earth's surface over timescales  $> \sim 10^5$  yrs (Walker et al., 1981; Berner et al., 1983; MacKenzie and Andersson, 2013). The limited reservoir of carbon in the atmosphere and oceans implies that disturbances in carbon cycle fluxes will result in changes in *p*CO<sub>2</sub>, and thus climate through the greenhouse effect of CO<sub>2</sub>. Changing climate will drive changes in the consumption of CO<sub>2</sub> by weathering, opposite to the direction of initial change, until carbon

fluxes into and out of the atmosphere–ocean system are balanced and a new equilibrium is established (Berner and Caldeira, 1997).

The temperature dependence of silicate mineral weathering rates is believed to have played an important role in the *p*CO<sub>2</sub>-weathering feedbacks together with the effects of runoff and biotic interactions (e.g., Berner and Kothavala, 2001; Arvidson et al., 2006). As predicted by chemical kinetics, rates of mineral dissolution show distinct temperature dependence in well-controlled lab experiments (Kump et al., 2000). Although there is also evidence for the temperature dependence of silicate weathering fluxes in field settings (White and Blum, 1995; Oliva et al., 2003), the inferred relationships are often complicated by multiple effects on weathering. Some of the other factors that might influence field-scale weathering fluxes, such as precipitation, runoff, and vegetation, co-vary with temperature, and the importance of variations in erosion rate further confounds simple interpretation (Riebe et al., 2004; West et al., 2005; Ferrier and Kirchner, 2008; Dixon et al., 2012; West, 2012). Moreover, natural weathering sys-

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**Fig. 1.** Distribution of the basaltic fields in our compilation. Shaded area shows the distribution of basaltic rocks based on global lithological map (Hartmann and Moosdorf, 2012). Red and blue circles are for active and inactive basaltic fields, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

tems that approach equilibrium will be controlled by thermodynamic rather than kinetic limits, with greater importance of water flow than temperature in controlling total alkalinity fluxes that consume  $\text{CO}_2$  (Maher, 2011).

Studies on single lithologies may reduce the complexities associated with rock types (White and Blum, 1995; Oliva et al., 2003; Hartmann, 2009; Ferrier et al., 2012). The relatively homogeneous composition among geographic regions makes basaltic rocks an ideal target for such investigation (Bluth and Kump, 1994). Previous studies have demonstrated that basalt weathering is globally significant due to rapid weathering rates and the resulting large global weathering fluxes, despite limited areal coverage of basalts (Gislason et al., 1996; Louvat and Allègre, 1997, 1998; Gaillardet et al., 1999; Dessert et al., 2003; Schopka et al., 2011). An empirically observed relationship between mean annual temperature (MAT, in  $^\circ\text{C}$ ) and the concentration of dissolved bicarbonate (resembling alkalinity) in stream water draining basaltic rocks has been proposed (Dessert et al., 2003):

$$K_{\text{CO}_2} = \text{RUN} \times C = \text{RUN} \times c_2 e^{c_1 \text{MAT}} \quad (1)$$

where  $K_{\text{CO}_2}$  ( $\text{mol}/\text{km}^2/\text{yr}$ ) is the consumption rate of atmospheric  $\text{CO}_2$  associated with basalt weathering,  $\text{RUN}$  ( $\text{mm}/\text{yr}$ ) the annual runoff,  $C$  ( $\mu\text{mol}/\text{L}$ ) the concentration of bicarbonate in stream water, and  $c_1$  ( $1/^\circ\text{C}$ ) and  $c_2$  ( $\mu\text{mol}/\text{L}$ ) the constants that define the empirical relationship between temperature and concentration of bicarbonate. Such mathematical laws are the basic building blocks of numerical models calculating the evolution of the partial pressure of atmospheric  $\text{CO}_2$  and Earth's climate over geological timescales (e.g., Berner et al., 1983; Donnadiu et al., 2006; Godd eris et al., 2014; Mills et al., 2014).

The model of Dessert et al. (2003) provides a first-order description of basalt weathering but does not account for some important observations. One such observation is that dilution with increasing runoff has been widely recognized for basaltic rivers (Bluth and Kump, 1994). In contrast, the model of Dessert et al. (2003) requires that the concentrations of bicarbonate are independent of runoff (Eq. (1)).

Another observation not explicitly considered in the model of Dessert et al. (2003) is that emplacement age of basaltic rocks influences observed weathering fluxes, with younger volcanic fields showing higher weathering reactivity (Bluth and Kump, 1994; Amiottte-Suchet and Probst, 1995; Louvat et al., 2008; Hartmann, 2009; Rad et al., 2013; Freire et al., 2014). Several potential mechanisms, operating at different scales, could explain why weathering rates are influenced by emplacement age. At the scale of mineral surfaces, changes in surface area, etch pit size and density, and coatings of secondary phases can impact mineral dissolution rates (White and Brantley, 2003). At the catchment scale, regolith development can change hydraulic conductivity with consequent changes in infiltration rates and surface water-ground water partitioning (Lohse and Dietrich, 2005; Schopka and Derry, 2012). At the scale of the volcanic edifice, active volcanic fields commonly support hydrothermal circulation as well as degassing of  $\text{CO}_2$ ,  $\text{SO}_2$ , and other reactive gases. Volcanic  $\text{CO}_2$  degassing can contribute significantly to the alkalinity fluxes from active volcanic centers (Aiuppa et al., 2000; Riv e et al., 2013).

New observations on the chemistry of rivers draining basaltic fields provide an opportunity to update prior compilations (Dessert et al., 2003) and re-examine the relationships between basalt weathering and climatic factors. In this study, we compile data from 37 basaltic regions based on published datasets, together with new data from several Chinese basaltic fields (Fig. 1). We separate the data compilation into 22 inactive volcanic fields (IVFs) and 15 active volcanic fields (AVFs) and find that the role of temperature is clarified if only IVFs are considered (Fig. 2(a)).

## 2. Methods

### 2.1. Proxy for weathering rate

Estimation of weathering rates relies on using the mineral-water interfacial area as a scaling factor (Navarre-Sitchler and Brantley, 2007). Rates inferred from the flux of weathering-derived dissolved ions in a river are typically normalized to the geographic surface area of the river catchment. Following Dessert et al. (2001),

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