Chemical Geology 365 (2014) 30–42

Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

A reactive-transport model for examining tectonic and climatic controls on chemical weathering and atmospheric CO_2 consumption in granitic regolith

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ARTICLE INFO

Article history:
Received 15 August 2013
Received in revised form 26 November 2013
Accepted 27 November 2013
Available online 12 December 2013

Editor: Dr. J. Fein

Keywords:
Reactive-transport model
Chemical weathering
Regolith development
Carbon cycle

ABSTRACT

We developed a 1D reactive-transport model for examining how tectonic and climatic parameters, namely uplift rate, water seepage velocity, and temperature, control chemical weathering and atmospheric CO_2 consumption rates during regolith development. The model consists of mass-conservation equations describing how mineral and solute concentrations change temporally and spatially (vertically) during weathering of granite containing 30% plagioclase (An_{20}) and up to 3% accessory calcite. The equations are coupled by volumetric weathering rates, which depend on mineral abundances, intrinsic dissolution rate constants, and surface areas as well as the departure of pore water from thermodynamic equilibrium. We numerically solve a non-steady-state, non-dimensional version of the model. By eliminating the need to prescribe regolith thickness, non-dimensionalization introduces two key scaling parameters that drive variations in the model results: the rock residence time (τ_r, the time for rock to travel upward through the model domain) and the water residence time (τ_w, the time for water to travel downward through the model domain). We use the model to examine fundamental properties of chemical weathering, especially reaction front propagation, with the primary aim of characterizing weathering regimes and identifying factors that maximize the dissolution of plagioclase because only silicate weathering regulates atmospheric CO_2 levels over geological timescales.

Weathering regimes are commonly classified as transport (or supply)-limited versus weathering (or kinetically)-limited, but as outgrowths of geomorphology, these terms primarily refer to the role of τ_r in regulating regolith thickness. Our findings suggest that the paradigm should be expanded to include kinetic controls determined by τ_w. Both regimes can experience far-from- and near-equilibrium mineral dissolution. However, transport-limited regimes generally have lower τ_w/τ_r ratios compared to kinetically-limited regimes. Under transport limitation, thick to thin reaction fronts propagate downward indefinitely yielding extensive mineral depletion, deeply weathered regolith, and high ratios of silicate-to-carbonate weathering. Under kinetic limitation, thin to non-existent reaction fronts evolve to steady-state yielding minimal mineral depletion, no regolith development, and low ratios of silicate-to-carbonate weathering. Kinetically-limited regimes typifying tectonically active mountain ranges have low long-term atmospheric CO_2 consumption rates. Transport-limited regimes typifying tectonically stable cratons also have low long-term atmospheric CO_2 consumption rates, but because much of the Earth's surface is transport-limited, we infer that such environments have the greatest impact on the global atmospheric CO_2 consumption flux. The model output leads us to hypothesize that the maximum contribution should occur for extensive regions of silicate bedrock consolidated in warm, wet climates because the combination of tectonic stability, high temperatures, and rapid seepage velocities accelerates reaction front propagation, facilitates calcite depletion, and sustains deep regolith development over long timescales. Our findings point to the importance of internal climate feedbacks for stabilizing long-term atmospheric CO_2 levels.

1. Introduction

Silicate and carbonate weathering both consume atmospheric CO_2, but when balanced by carbonate precipitation in the oceans, only silicate weathering affects atmospheric CO_2 concentrations and hence, the degree of greenhouse warming (Urey, 1952; Walker et al., 1981; Berner et al., 1983). Understanding controls on the absolute magnitude of silicate weathering rates as well as the relative ratio of silicate-to-carbonate

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0009-2541/$ – see front matter
http://dx.doi.org/10.1016/j.chemgeo.2013.11.028

weathering therefore has important implications for modeling the evolution of Earth's climate over geological timescales. Several studies have attempted to elucidate tectonic and climatic controls on the link between weathering and climate (Raymo and Ruddiman, 1992; Riebe et al., 2001; Jacobson et al., 2003; Chamberlain et al., 2005; West et al., 2005; Moore et al., 2013), yet defining exact relationships has proved enigmatic. For example, various lines of evidence suggest that mountain uplift should accelerate the drawdown of atmospheric CO_2 by silicate weathering (Raymo et al., 1988; Raymo and Ruddiman, 1992), but efforts to trace carbon flow by apportioning dissolved Ca among lithological sources have revealed that carbonate weathering dominates the geochemistry of rivers draining active orogens (Blum et al., 1998; Jacobson and Blum, 2003; Quade et al., 2003; Hren et al., 2007; Wolff-Boenisch et al., 2009; Moore et al., 2013). This occurs even for rivers draining silicate bedrock because granite, schist, and other silicate rocks that commonly compose mountains contain trace amounts of calcite, which easily dissolves during uplift and erosion (Mast et al., 1990; Blum et al., 1998; Jacobson and Blum, 2000, 2003; White et al., 2005). In the Southern Alps of New Zealand, for instance, uplift and erosion rates span three orders of magnitude across a region of silicate bedrock hosting 3% accessory calcite (Templeton et al., 1998; Jacobson et al., 2003). Total silicate plus carbonate weathering rates for the most rapidly uplifting region of the mountain range are about one order of magnitude higher than the global mean and rank among the highest chemical weathering rates in the world (Jacobson and Blum, 2003; Moore et al., 2013). However, major ion and Ca isotope mixing models suggest that the fraction of Ca from carbonate weathering increases with increasing tectonic activity, from ~50 to 60% in regions experiencing the lowest uplift rates to >90% in regions experiencing the highest uplift rates (Jacobson and Blum, 2003; Jacobson et al., 2003; Moore et al., 2013). The net implication is that long-term atmospheric CO_2 consumption rates in the most rapidly uplifting region are no higher than the global mean (Jacobson and Blum, 2003; Jacobson et al., 2003; Moore et al., 2013). Other conundrums surround the effects of climate. For example, silicate weathering rates are expected to increase with temperature, yet watershed elemental fluxes do not readily conform with Arrhenius-style relationships (White and Blum, 1995; Huh and Edmond, 1999; Kump et al., 2000; Riebe et al., 2004; von Blanckenburg, 2005; Maher, 2010; Turner et al., 2010). This may reflect a combination of previously underappreciated factors, including water residence times in soils and the more significant effect of temperature on thermodynamic solubility instead of kinetic rate constants (Maher, 2010). These and other similar observations have raised important questions about the type of tectonic and climatic conditions that maximize the effect of silicate weathering on long-term climate change.

In recent years, studies have employed a variety of analytical and numerical models to quantify chemical weathering during soil and regolith development (White et al., 1999, 2001, 2008, 2009; Chamberlain et al., 2005; Waldbauer and Chamberlain, 2005; Hren et al., 2007; Brantley et al., 2008; Ferrier and Kirchner, 2008; Yoo and Mudd, 2008; Brantley and White, 2009; Gabet and Mudd, 2009; Hilley et al., 2010; Lebedeva et al., 2010; Maher, 2010; Moore et al., 2012; West, 2012). Reactive-transport modeling in particular offers an exceptionally powerful technique for describing and predicting the spatial and temporal evolution of complex Earth surface systems, as the interplay between fundamental phenomena, such as kinetics, thermodynamics, and material transport, can be simultaneously considered in the context of coupled mass-conservation equations (Steefel et al., 2005; Steefel, 2008; Maher, 2010). In this study, we present a simple 1D reactive-transport model for examining tectonic and climatic controls on chemical weathering and atmospheric CO_2 consumption during regolith production. We apply the model to granite containing plagioclase and variable amounts of accessory calcite, and we study effects related to rock uplift rates, water seepage velocities, and temperature. Our overall approach, which centers around reaction front propagation (e.g., White, 1995; Brantley et al., 2008; Lebedeva et al., 2010; Moore et al., 2012), resembles the numerical treatment previously established for sandstone core flood experiments (Fogler and McCune, 1976; Lund and Fogler, 1976; Hekim and Fogler, 1980; Hinch and Bhatt, 1990) but includes several notable modifications, including consideration of mineral-specific reaction rates as opposed to lumped, bulk-rock parameters, a focus on weathering products instead of reactants, and allowance for solid phase advection. We derive analytical solutions for non-steady-state dimensional and dimensionless equations, but in this contribution, we primarily focus on steady-state (or quasi-steady-state) output obtained by numerically solving the dimensionless equations. We use the model to investigate fundamental controls on regolith development, with particular emphasis placed on classifying weathering regimes and constraining the combination of tectonic and climatic parameters that optimize the long-term drawdown of atmospheric CO_2.

2. Methods: 1D reactive-transport model

Here, we formulate a 1D reactive-transport model for regolith development, which mainly applies to ridge top systems. The model does not consider lateral transport, only vertical regolith development. In Section 2.1, we present weathering reactions as well as mass-conversation equations for mineral and solute concentrations and their associated boundary and initial conditions. In Section 2.2, we derive an expression for the reaction front velocity, which allows us to transform the model from a system of partial differential equations to a system of ordinary differential equations that can be analytically solved for infinitely thick model domains. In Section 2.3, we introduce a non-dimensional version of the model, which allows us to examine the characteristic behavior of the equations without having precise knowledge of the weathering zone depth. Section 2.4 presents a numerical scheme for solving the non-dimensional model, and finally, in Section 2.5, we tabulate and describe input parameters.

2.1. Theoretical overview and general formulation

Fig. 1 provides a conceptual diagram for the model. Table 1 defines symbols and includes references to relevant equations, mainly the first occurrence of the symbol but also expressions where values for the

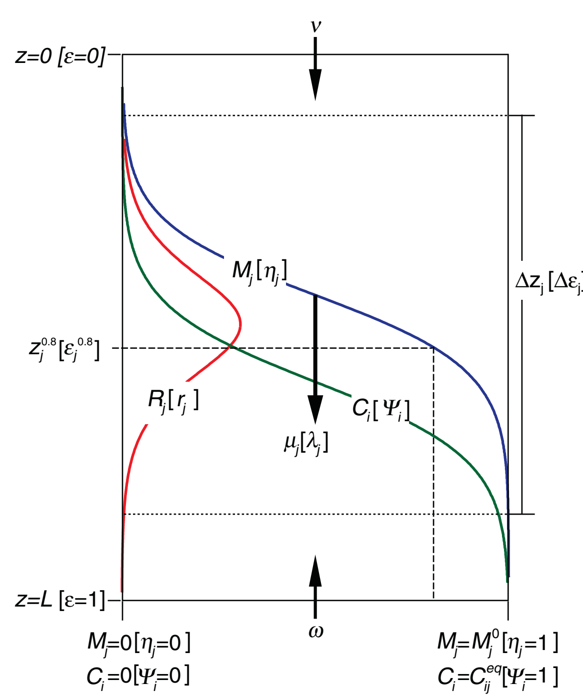

Fig. 1. Schematic depth profile illustrating key model variables for dimensional and non-dimensional versions of the model. Non-dimensional variables are provided in square brackets.