



Evolution of carbon cycle over the past 100 million years

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Abstract

It is generally accepted that progressive cooling of global climate since the Late Cretaceous results from decreasing partial pressure of atmospheric CO₂ (*p*CO₂). However, details on how and why the carbon cycle evolved and how it would affect *p*CO₂ have not been fully resolved. While the long-term decline of *p*CO₂ might be caused by the decrease of volcanic degassing through the negative feedback between *p*CO₂ and silicate weathering, seafloor spreading, the major control of CO₂ degassing, seems to have remained relatively constant. Alternative explanation, known as ‘uplift driven climate change’ hypothesis, proposes that tectonic uplift may have enhanced the sink of atmospheric CO₂ by silicate weathering, and thus produced the decline of *p*CO₂. However, increasing weathering sink of CO₂ could deplete atmosphere all of its CO₂ within several million years while holding volcanic outgassing constant. In this work, major fluxes of long-term carbon cycle are calculated based on a reverse model constrained by marine C, Sr and Os isotopic records and the spreading rate of sea floor. Weathering of island basalt and continental silicate rocks are separated in the new model. The results indicate a long-term decline of island basalt weathering in consistent with the global cooling trend over the past 100 million years. Dramatic changes of the CO₂ fluxes associated continental silicate weathering, reverse weathering, volcanic degassing and the growth of organic carbon reservoir have been observed. Disturbance of atmospheric CO₂ cycle by these fluxes seems to be maintained by the concomitant adjustments of island basalt weathering that were sensitive to the *p*CO₂ controlled environment factors such as temperature and runoff. The negative feedbacks between *p*CO₂ and weathering of island basalt might have played a significant role in stabilizing the long-term carbon cycle.

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1. INTRODUCTION

Global climate has experienced dramatic change since the Late Cretaceous. During the latest portion of Cretaceous approximately 100–65 million years ago, global temperature reached the highest level of the last ~200 million years with no glaciation in both poles (Bice et al., 2006). Progressive cooling afterward transited the earth from a greenhouse state into an icehouse world with ice caps at high latitudes, first in Antarctic and later in the North Hemisphere (Zachos et al., 2001). It is believed that decreasing partial pressure of CO₂ in atmosphere (*p*CO₂)

is the primary determinant for the long-term cooling trend and for the growth of continental-scale ice sheet in polar regions (Deconto and Pollard, 2003). Although growing evidences confirmed a long-term decrease of *p*CO₂ (Pearson and Palmer, 2000; Pagani et al., 2005; Tripati et al., 2009), the reason behind the decline of *p*CO₂ is still under debates.

Reduction of CO₂ degassing has been proposed by BLAG carbon cycle model as the primary driver for the decline of *p*CO₂ through the negative feedbacks between *p*CO₂ and silicate weathering (Berner et al., 1983). In this model, weathering reaction is largely controlled by the climatic factors that are sensitive to the changes of *p*CO₂, such as temperature and runoff. Thus, balance of carbon cycle could be maintained by the negative feedbacks between

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$p\text{CO}_2$ and silicate weathering (Walker et al., 1981; Berner et al., 1983). Due to the limited capacity of atmosphere, a decrease of CO_2 outgassing would result in rapid drop of $p\text{CO}_2$ until new balance is reached so that the decreasing CO_2 consumption by silicate weathering in response to the decline of $p\text{CO}_2$ could compensate the reduction of CO_2 degassing. However, the rate of sea floor spreading, which is believed as the major control of CO_2 degassing (Berner et al., 1983), have remained relatively constant especially over the past 65 million years (Rowley, 2002; Muller et al., 2008).

Alternative explanation, known as ‘uplift driven climate change’ hypothesis (Raymo et al., 1988; Raymo and Ruddiman, 1992), suggested that the late Cenozoic tectonic uplift, mainly Tibetan Plateau, may have enhanced physical erosion and monsoonal rainfall, which finally helped draw-down of atmospheric CO_2 by silicate weathering. The post-Eocene decline of $p\text{CO}_2$ and global cooling is coincident with the time of major Cenozoic orogenesis (Raymo and Ruddiman, 1992). Increasing production of terrigenous sediment and raising $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{187}\text{Os}/^{188}\text{Os}$ ratios of seawater also support a late Cenozoic increase in both physical and chemical denudation (Hay et al., 1988; Edmond, 1992; Peucker-Ehrenbrink et al., 1995). However, increasing weathering sink of CO_2 would deplete atmosphere all of its CO_2 within several million years while holding volcanic outgassing constant (Berner and Caldeira, 1997).

The ‘uplift driven climate change’ hypothesis argues that the sequestration of atmosphere CO_2 by silicate weathering is a strong function of continental relief, and the importance of $p\text{CO}_2$ -weathering feedback which prevents runaway icehouse or greenhouse in BLAG carbon cycle model is implicitly negated (Raymo and Ruddiman, 1992). As no evidence pointing an increase of volcanic degassing, two potential negative feedbacks were proposed to compensate the increasing erosional drawdown of CO_2 (Raymo and Ruddiman, 1992): (1) decreasing accumulation of sedimentary organic carbon due to increasing oxygen concentration of seawater in response to climate cooling (Raymo, 1994); (2) increasing precipitation of silicate minerals in the deep sea or decreasing weathering of seafloor basalt in response to the elevated $p\text{H}$ of seawater under lower $p\text{CO}_2$ (Francois and Walker, 1992). However, growth of organic carbon reservoir seems to have increased as a result of increasing preservation efficiency in regions with high sedimentation rate of terrigenous materials (France-Lanord and Derry, 1997). Precipitation of silicate minerals in the deep sea, known as reverse weathering, may be largely controlled by the supply of degraded weathering product (Michalopoulos and Aller, 1995). Although reverse weathering may increase in response to increasing weathering product, it cannot compensate all of the increasing consumption of CO_2 by continental silicate weathering since the net effect of weathering and reverse weathering should remove CO_2 from atmosphere. The dynamics of seafloor basalt weathering is poorly understood and is supposed to be largely controlled by the spreading rate of seafloor rather than $p\text{H}$ (Wallmann, 2001).

The fundamental difference between BLAG carbon model (Berner et al., 1983) and the ‘uplift driven climate change’

hypothesis (Raymo and Ruddiman, 1992) is related to the two extreme mechanisms that control the rate of silicate weathering, i.e., the weathering limited regime and the supply limited regime respectively. Weathering regime largely depends on the extent of physical erosion (Kump et al., 2000; West et al., 2005). Under the weathering limited regime, fresh rocks are sufficiently exposed and weathering rate is controlled by kinetic parameters of weathering reaction that are closely linked to $p\text{CO}_2$, e.g., temperature. While under the supply limited weathering regime, physical erosion is weak, weathering of fresh rock is inhibited by the protection of soil cover and weathering rate largely depends on the rate of physical erosion (Kump et al., 2000; West et al., 2005).

Generally, the large continents are not effectively eroded, weathering of continental silicate seems to be largely supply limited (Gaillardet et al., 1999). Kump and Arthur (1997) argued that the increasing sequestration of atmospheric CO_2 in active orogens such as Himalayas may have been offset by reduction of weathering elsewhere under a kinetically controlled weathering regime due to the decline of temperature and runoff in response to the drop of $p\text{CO}_2$. Weathering of island basalt may play a significant role in this mechanism. Most of the oceanic islands locate at tectonically active regions, and are generally characterized by mountainous relief with particularly high rate of physical erosion (Milliman and Meade, 1993). Thus, weathering of island basalt might be largely subjected to the weathering limited regime, which shows high weathering flux and strong environmental influence such as temperature, runoff and plant cover (Moulton and Berner, 1998; Dessert et al., 2003; Louvat et al., 2008; Gaillardet et al., 2011; Schopka et al., 2011). Updated estimates on the cumulative surface area and weathering rate of island basalt (Dessert et al., 2003; Allègre et al., 2010) indicate that weathering of island basalt consumes up to 5.0×10^{12} mol atmospheric CO_2 per year at the present, which is more than half of that consumed by continental silicate weathering (Gaillardet et al., 1999). This value might be even higher when the underground weathering flux is considered (Schopka and Derry, 2012). Thus, weathering of island basalt may act an important role in regulating the long-term balance of carbon cycle through the $p\text{CO}_2$ -weathering feedbacks.

Marine isotopic records of carbon, strontium and osmium could help resolve the ancient carbon cycle processes because carbon fluxes derived from different reservoirs are characterized by distinct $\delta^{13}\text{C}$ values and the processes that control the long-term carbon cycle also control the strontium and osmium isotopic compositions of seawater. A set of early works has established the framework to calculate ancient carbon cycle fluxes mainly based on marine C and Sr isotopic records (Kump, 1989; Derry and France-Lanord, 1996; Godd ris and Fran ois, 1996; Kump and Arthur, 1997; Francois and Godd ris, 1998; Wallmann, 2001; Berner, 2006; Kashiwagi et al., 2008). In these works, normally four equations were involved regarding the mass balances of carbon and alkalinity, and the isotopic balances of carbon and strontium in ocean–atmosphere system. Some works may favor the mass balance equation of atmospheric CO_2 instead of oceanic alkalinity. As a matter of fact, mass balance equation of atmospheric CO_2 could be

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