



Estimating silicate weathering timescales from geochemical modeling and spring water residence time in the Kirishima volcanic area, southern Japan

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ABSTRACT

Kyushu Island in southern Japan is located within an area subjected to one of the highest rates of chemical weathering in the world. This area is purported to have the highest discharge rate of dissolved silica and other cations within the Japanese Island arc. To understand the timescale of chemical weathering in such a weathering ‘hot spot’, details of water–rock interaction are characterized using a conventional geochemical model for the Kirishima volcanic area. The model is focused on source water (spring water) with different residence times previously estimated by chlorofluorocarbons (CFCs) concentration analysis. The geochemical and geological features of the spring water samples suggest that the primary reactant minerals are pyroxenes and plagioclase. It was also found from activity–diagram analysis that kaolinite and Ca-smectite are the major precipitable secondary minerals from the system. The occurrence of these reactions was quantitatively confirmed by stoichiometric calculations of measured dissolved ions. In combination with previous estimates of spring water residence ages, it is concluded that saturation of dissolved silica and simultaneous transition of phase precipitation from kaolinite to Ca-smectite starts in ca. 20 years after aqueous recharge. Our findings imply the usefulness of geochemical data as a function of water–rock interaction timescale in certain field. In future work, these findings can be compared to data from other areas to improve understanding of global heterogeneity in chemical weathering rates.

1. Introduction

Chemical weathering is one of the important processes controlling biogeochemical cycles in Earth surface environments (Alexandre et al., 1997; Aufdenkampe et al., 2011; Elderfield, 2010; Filippelli and Souch, 1999; Lerman, 1988). Silica, the second most abundant element in the Earth’s crust, is one of these important cycled materials. Dissolved silica (DSi) is generally produced through chemical weathering at the Earth’s surface, and is a valuable component of marine ecosystems (Bernard et al., 2011; Kristiansen and Hoell, 2002; Ragueneau et al., 2000). Moreover, chemical weathering reactions in terrestrial environments are known to consume atmospheric CO₂, the effect of which, together with intensity of chemical weathering, has been debated in reconstructions of Earth’s paleoclimate over the past several million years (Berner et al., 1983; Brady, 1991; Maher and Chamberlain, 2014; Pearson and Palmer, 2000; Raymo and Ruddiman, 1992; Willenbring and Von Blanckenburg, 2010).

Globally, the largest fluxes of DSi from land to ocean are from volcanic arcs and flood basalt areas subjected to relatively high

temperature and high humidity conditions, such as in tropical and subtropical regions of the Pacific and other continental margins (Dessert et al., 2003, 2001; Dürr et al., 2011; Gaillardet et al., 1999, 2011; Ibarra et al., 2016; Schopka et al., 2011; Schopka and Derry, 2012; von Blanckenburg, 2004). Japan is situated in such a high DSi flux zone. However, its rate varies spatially according to different lithology and runoff rate (Hartmann et al., 2014; Hartmann and Moosdorf, 2011). Among these, Kyushu Islands (Fig. 1) are considered to be the most significant ‘hot spot’ areas in terms of chemical weathering activity (Hartmann et al., 2010). A river in this region has been investigated, and found to have evolved its chemical composition of increasing DSi saturation downstream, owing to active silicate weathering and secondary mineral precipitation (Anazawa et al., 2006).

In chemical weathering, its timescale is considered to be the main factor controlling material cycling. To best understand such a rate, estimation of precipitation reaction rates of secondary minerals is also important (Bouchez and Gaillardet, 2014; Maher, 2010, 2011; Nugent et al., 1998). However, the weathering rates when evaluated in the field are significantly different from that found in laboratory experiments

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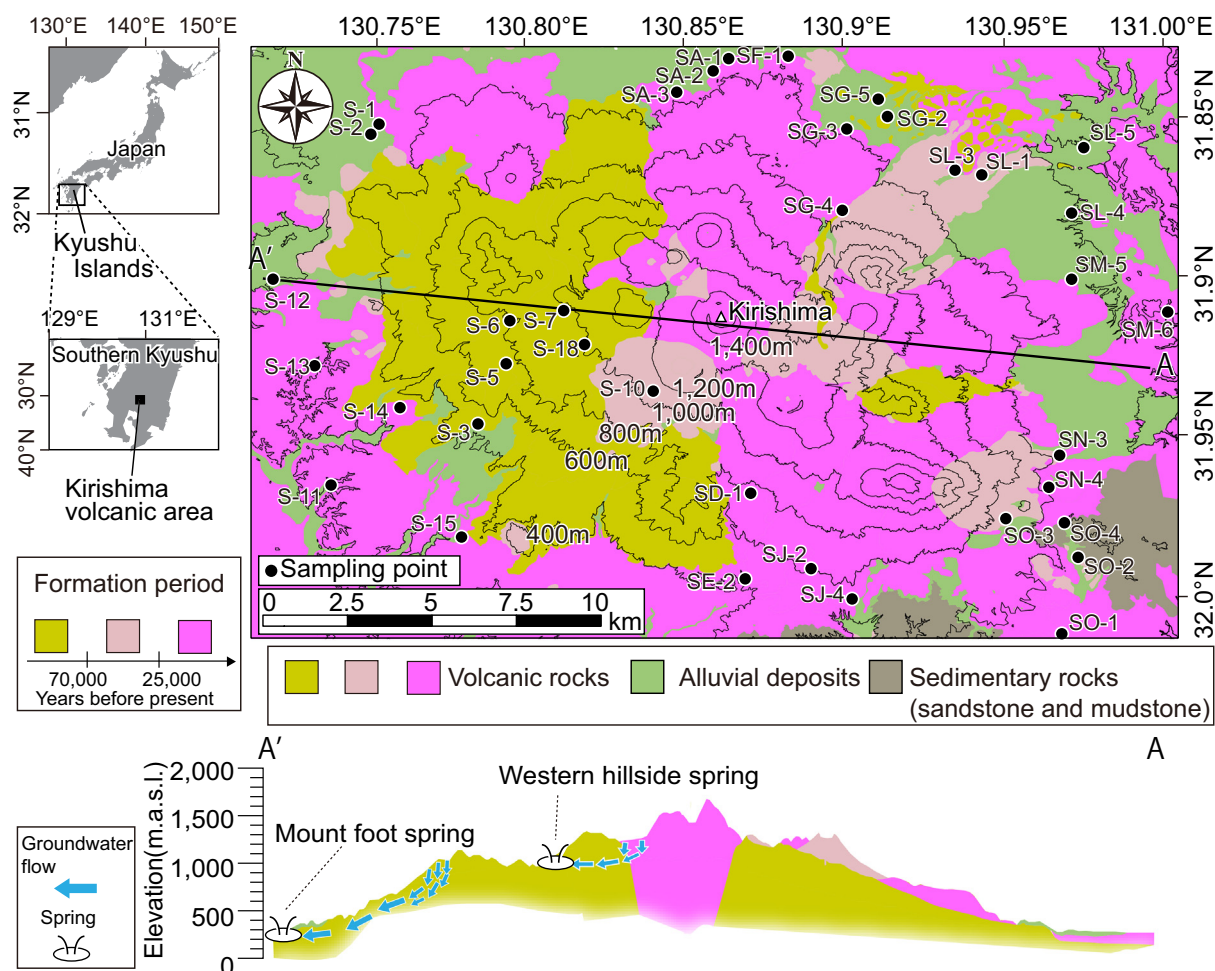


Fig. 1. Simplified geological map and schematic hydrogeological cross section for the study area. The geological classification is after Imura and Kobayashi (2001) and Sawamura and Matsui (1957). Spring water sampling points for 2013 survey are shown in the map. Spring water discharge systems are also shown in the cross section (see text for details).

(White and Brantley, 2003; Yokoyama and Matsukura, 2006); improved estimation of precipitation reaction rates in the field is thus needed (Alekseyev et al., 1997; Maher et al., 2009).

In situ hydrogeological conditions affect the chemical weathering rate, inclusive of precipitation reactions (Ludwig et al., 2012; Zhu, 2005) and it can be proposed that groundwater residence time is a useful parameter for determining water–rock interaction time since it contains factors such as topography, geological properties and amount of precipitation in the target area. Therefore, to monitor timescales of geochemical evolution, groundwater (spring water) can be used, as it has been established that it typically evolves geochemically along the flow path (Chapelle and McMahon, 1991; Garrels and Mackenzie, 1967; Gastmans et al., 2010; Ikeda, 1989; Pawar et al., 2008). For example, Kenoyer and Bowser (1992a, 1992b) investigated secondary mineral precipitation using groundwater residence time deduced by hydrogeological analyses in a sandstone aquifer in Wisconsin, USA, and suggested that precipitable secondary minerals altered from a gibbsite phase to a kaolinite phase between a 40-year period (accuracy within 40%). This result implies a presence of time-dependent weathering phase change placed on certain hydrogeological system. However, there is very limited knowledge about the timescale of chemical weathering in the hot spot area of Kyushu.

In the past two decades, there has been significant development in the methodology for evaluating relatively young groundwater residence times. The method uses chemical tracers such as chlorofluorocarbons (CFCs) and SF_6 (Alley, 2002; Busenberg and Plummer, 1992, 2000;

Cook and Solomon, 1997), although direct correlations between chemical weathering rates and measurable groundwater age parameters have been rarely discussed. A total of 25 spring water samples have been analyzed for CFC concentrations in the years 2007–2013 in the Kirishima volcanic area in southern Kyushu (Fig. 1) to obtain absolute average ages for each spring (Ide et al., 2016; see also Appendix A). Therefore, this study aims to clarify the time-dependent evolution of chemical weathering and the saturation point of DSI by using hydrochemistry and residence time in spring water collected from the Kirishima volcanic area.

2. Outline of the study area

The Kirishima volcanic area (Fig. 1) has a surface area of approximately 471 km² with a maximum elevation of 1700 m above sea level (m.a.s.l.). It is composed of multiple volcanoes that have variously erupted over the past 0.3 Ma (Imura, 1992; Sawamura and Matsui, 1957). Average annual precipitation of the study area for the year 2007 to 2013 is approximately 2530 mm y⁻¹ (Japan Meteorological Agency, 2002), with June to August being the predominantly rainy season in which more than half the annual precipitation occurs. According to Tsukada (1987, 1988) over 200 natural springs discharge at the base of surrounding mountains are mostly within the elevation range of 200 to 1200 m.a.s.l. (Fig. 1).

Basement rocks of this area consist of pelagic sedimentary rocks (sandstone and mudstone) of accretionary prism of the Paleogene

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