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Notes

^{231}Pa excesses in arc volcanic rocks: Constraint on melting rates at convergent margins

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ABSTRACT

The ^{231}Pa - ^{235}U disequilibria provide greater ability to constrain the rate of mantle melting in convergent margin settings than the more often analyzed ^{238}U - ^{230}Th - ^{226}Ra systems, which are strongly affected by fluid-addition processes from the subducting slab. Here we present new ^{231}Pa - ^{235}U data for 12 samples from the Kick'em Jenny (KEJ) submarine volcano in the Southern Lesser Antilles to define the melting rate at a subduction zone with one of the lowest convergence rates. The KEJ samples have the highest average $(^{231}\text{Pa})/(^{235}\text{U})$ yet measured in global arcs, consistent with other studies of the Southern Lesser Antilles lavas. These results reinforce the previously noted negative correlation between average $(^{231}\text{Pa})/(^{235}\text{U})$ and convergence rate in all arc settings globally.

We develop a model to explain this negative correlation and to better constrain melting rates at convergent margins. Assuming that the corner flow velocity is coupled to and equal to the subducting slab velocity, the melting rate, directly reflecting the flux of water added, becomes a linear function of subduction rate. This physical model is then coupled to three different melting models previously developed for calculating U-series disequilibria (reactive porous flow, dynamic, and flux melting). All three models reproduce the globally observed negative correlation between subduction rate and ^{231}Pa excess. Although the style of melting cannot be easily discriminated, the good correspondence between models and observation provides an example of how geochemical and geophysical models can be linked to provide a self-consistent model of melt generation in convergent margin settings.

Keywords: ^{231}Pa - ^{235}U , melting process, subduction rate, convergent margin.

INTRODUCTION

Determining the origin of convergent margin magmas is essential to understanding the formation of continental crust and recycling of subducted materials back into the convecting mantle. Because hydrous fluids released from subducted slabs by dehydration reactions depress the solidus of mantle peridotite, it is generally thought that arc magmatism dominantly reflects partial melting of hydrated mantle peridotite in the overriding plate (e.g., Gill, 1981). However, there remains uncertainty about the relationship between the rate of plate convergence and the rate of magma production and output.

U-series disequilibria data provide important temporal constraints on the production and ascent of magmas in arc settings. Numerous studies have shown that many young arc rock suites show a negative correlation between $(^{238}\text{U})/(^{232}\text{Th})$ and $(^{230}\text{Th})/(^{238}\text{U})$ (Fig. 1) as well as correlation between $(^{226}\text{Ra})/(^{230}\text{Th})$ and indicators of fluid addition such as Ba/Th (e.g., Turner et al., 2003). Both $(^{230}\text{Th})/(^{238}\text{U})$ and $(^{226}\text{Ra})/(^{230}\text{Th})$ are observed to vary widely in a given arc, and this is usually interpreted to reflect addition of U- and Ra-rich fluids to the mantle source followed by ultrafast migration of magma to the surface (e.g., Sigmarsson et al., 2002). Because of the wide range in

these disequilibria and their dependence on the amount of fluid component, one cannot easily use ^{238}U - ^{230}Th - ^{226}Ra data to assess the rate of peridotite melting in the mantle wedge beneath arc volcanoes.

Although fewer analyses of $(^{231}\text{Pa})/(^{235}\text{U})$ in young arc lavas exist at present, the available data definitively show that ^{231}Pa excess is relatively large and ubiquitous in arc settings (Pickett and Murrell, 1997; Turner et al., 2006). The origin of large $(^{231}\text{Pa})/(^{235}\text{U})$ disequilibria is generally attributed to melting processes (Turner et al., 2003). The ^{231}Pa excesses are unlikely to reflect fluid addition because of the low solubility of ^{231}Pa (Brenan et al., 1994; Keppler, 1996). In contrast, the ratio of U and Pa partition coefficients between mantle solid phases and melt, $D_{\text{U}}^{\text{solid/melt}}/D_{\text{Pa}}^{\text{solid/melt}}$, likely ranges from 100 to 1000 (Lundstrom et al., 1994; Sims et al., 1999), meaning that large ^{231}Pa excess could be generated by melting processes such as in mid-ocean ridge and intraplate settings simulated in in-growth melting models (e.g., McKenzie, 1985). In-growth melting models have more recently been applied to explain U-series data from arc magmas (Bourdon et al., 2003; Dosseto et al., 2003; Turner et al., 2006).

Here we examine the relationship between ^{231}Pa - ^{235}U disequilibria and subduction rate for the global data set consisting of previously published convergent margin data plus new data for

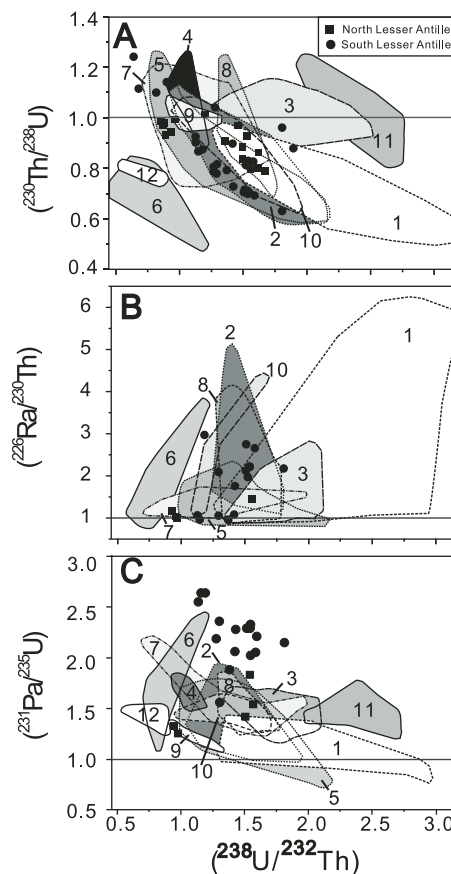


Figure 1. U/Th vs. U-series data for arc lavas. Data for global arcs are from Turner et al. (1996, 1997, 1998, 1999, 2000, 2001, 2006), Elliott et al. (1997), Turner and Hawkesworth (1997, 1998), Pickett and Murrell (1997), Bourdon et al. (1999), Turner and Foden (2001), Thomas et al. (2002), Dosseto et al. (2003), George et al. (2003), Zellmer et al. (2003), and Asmerom et al. (2005). Arcs: 1—Tonga; 2—Vanuatu; 3—Kamchatka; 4—Costa Rica; 5—Kermadec; 6—Sunda; 7—Alaska; 8—Aleutians; 9—Mexico; 10—Mariana; 11—Nicaragua; 12—Philippine.

samples from the slowly converging Southern Lesser Antilles (SLA) arc. These latter samples provide an important end member of high ^{231}Pa excess supporting the inverse relationship between convergence rate and ^{231}Pa excess. We couple a variety of in-growth-based melting models to a physical model linking convergence rate with melting rate to show that the global ^{231}Pa data set is consistent with a simple model whereby the rate of melting of the mantle wedge is linearly proportional to the convergence rate.

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TECTONIC SETTING, SAMPLING, AND METHODS

The Lesser Antilles (LA) arc provides an excellent case study for using ^{231}Pa excesses to understand the melting process in a slow subduction zone. The Atlantic oceanic lithosphere subducts beneath the Caribbean plate with a convergence rate of 2–4 cm/yr (Macdonald et al., 2000). The arc can be divided into two segments, a northern zone defined by portions north of Martinique, and a southern zone to the south. The northern zone has a slab dip of 50° – 60° and the southern zone ranges from a slab dip of 45° – 50° in the north to almost vertical in the south (Wadge and Shepherd, 1984). The change in obliquity of subduction along the arc results in a significantly lower convergence rate in the south (~ 1.3 cm/yr) than the north (3.7 cm/yr) (Jarrard, 1986; Speed et al., 1993; Macdonald et al., 2000). Magma production is greatest in the central arc, possibly reflecting the variation in the obliquity of subduction (Macdonald et al., 2000). The LA arc has the thickest crust (~ 35 km) of all oceanic subduction zones (Gill, 1981; Plank and Langmuir, 1998). Geochemical studies show southward increases in sediment component accompanied by decreasing fluid component (Turner et al., 1996).

We analyzed 12 samples from Kick'em Jenny (KEJ), a submarine volcano ($12^\circ 30' \text{N}$, $61^\circ 38' \text{W}$) ~ 8 km north of Grenada in the SLA arc (Devine and Sigurdsson, 1995; collected by H. Sigurdsson). All samples are fresh and porphyritic (plagioclase and amphibole phenocrysts) and span a compositional range from basalts to basaltic andesites with SiO_2 from 47.2 to 55.5 wt%. The U–Pa data of KEJ samples were analyzed by isotope dilution mass spectrometry using the multicollector–inductively coupled plasma–mass spectrometer in the Department of Geology at the University of Illinois at Urbana-Champaign, following established methods in Regelous et al. (2004).

RESULTS

KEJ samples have $(^{231}\text{Pa})/(^{235}\text{U})$ values from 1.56 to 2.64. Because these are submarine samples, eruption ages are uncertain. However, of the 12 KEJ lavas 8 have significant ^{226}Ra excess [$(^{226}\text{Ra})/(^{230}\text{Th})$ of 1.76–2.97], meaning that radioactive decay of $(^{231}\text{Pa})/(^{235}\text{U})$ since eruption is minimal because the half-life of ^{231}Pa (32,760 yr) is much longer than that of ^{226}Ra (1599 yr). For the 4 samples with ^{226}Ra – ^{230}Th disequilibrium $< 5\%$, $(^{231}\text{Pa})/(^{235}\text{U})$ ranges from 1.88 to 2.64, suggesting youthfulness of these samples. Therefore, none of the samples has been age corrected. No obvious correlation between ^{231}Pa excess and SiO_2 or MgO is observed, suggesting that assimilation and fractional crystallization have not strongly affected the $(^{231}\text{Pa})/(^{235}\text{U})$ of most samples.

Overall, these new data confirm the high ^{231}Pa excess (2.15) originally observed at KEJ (Pickett and Murrell, 1997) and agree well with the results of other SLA samples from nearby St. Vincent and Ile de Caille Islands, results that range in $(^{231}\text{Pa})/(^{235}\text{U})$ from 2.0 to 2.6 (Turner et al., 2006). The one exception to the generally high ^{231}Pa excess in the SLA arc is a sample from Grenada with a relatively uncertain age [$(^{231}\text{Pa})/(^{235}\text{U}) = 1.14$; Turner et al., 2006]. These new ^{231}Pa data reinforce the negative correlation between average $(^{231}\text{Pa})/(^{235}\text{U})$ and subduction rate observed for arcs globally (Fig. 2A) (Turner et al., 2006).

DISCUSSION

It is generally agreed that partial melting of the mantle wedge strongly depends on the amount of water added from the subducting slab (Davies and Bickle, 1991). Experimental studies of peridotite melting indicate that degree of melting is linearly related to the H_2O content of the melt (Hirose and Kawamoto, 1995) and

peridotite (Gaetani and Grove, 1998). A direct link between water flux and magma output has been inferred for the Mariana backarc (Stolper and Newman, 1994), the Aleutian arc (George et al., 2003), and South America, where the Chile ridge subducts (Turner et al., 2003). A recent parameterization of experiments also indicates a nearly one-to-one relationship between water added and degree of melting (Katz et al., 2003). Assuming that the subducted oceanic crust has a constant water content, the flux of water added to the mantle wedge would be linearly related to subduction rate (Davies and Bickle, 1991; George et al., 2003). Therefore, as long as the corner flow velocity of the mantle wedge matches that of the subducting slab, the water concentration in the overlying mantle source remains independent of the subduction rate; that is, new mantle material and water flow into the melting region at the same rate, resulting in a constant water content in the peridotite that is melting. Accordingly, the melting rate, the mass of melt produced per unit time, is directly pro-

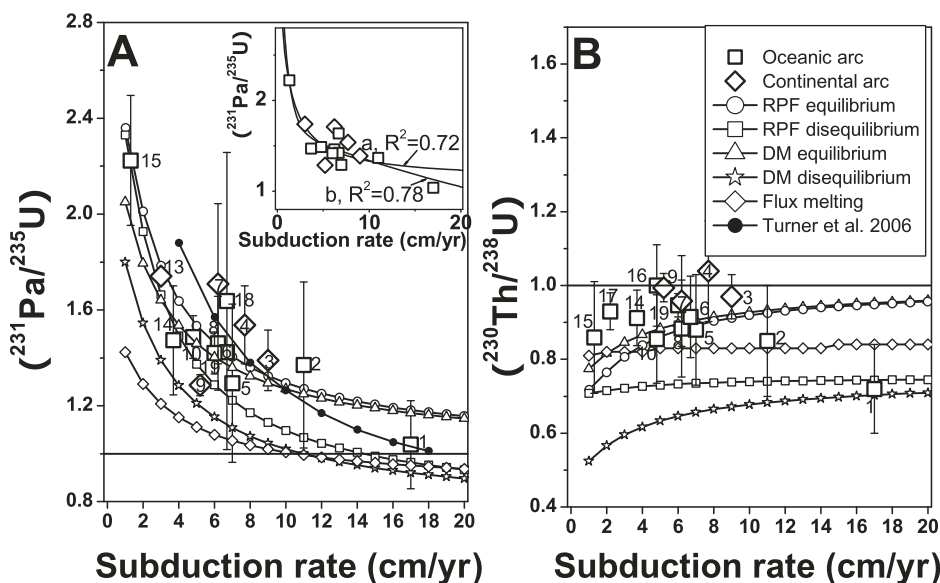


Figure 2. Average $(^{231}\text{Pa})/(^{235}\text{U})$ and $(^{230}\text{Th})/(^{238}\text{U})$ of global arcs vs. subduction rate. Melting rate (Γ) is defined as mass of melt produced (M) per unit time (t) per volume of peridotite (V) entering melting zone. Thus, $\Gamma = M/(Vt)$. Assuming similar convergence amount (d) and melting degree (F) in global arcs, $t = d/v_s$ and $M = F\rho_s V$ (v_s , subduction rate; ρ_s , mantle density). Therefore, melting rate should be linearly related to subduction rate with equation $\Gamma = F\rho_s v_s/d$. Such correlation can be combined with fluxing melting, dynamic melting (DM), and reactive porous flow (RPF) models, all producing similar negative correlation between subduction rate and $(^{231}\text{Pa})/(^{235}\text{U})$. In the RPF and DM model, the effect of recent fluid addition on U-series disequilibrium is also tested by varying initial status of mantle wedge. For the mantle source in disequilibrium, e , $(^{231}\text{Pa})/(^{235}\text{U})$ and $(^{230}\text{Th})/(^{238}\text{U})$ are 0.75. Global $(^{231}\text{Pa})/(^{235}\text{U})$ and subduction rates were nonlinearly fitted using KaleidaGraph software. Two curves in inset of A show that R^2 is 0.72 (a) and 0.78 (b) for the fittings, according to format of equations of DM and RPF models, respectively. The $(^{231}\text{Pa})/(^{235}\text{U})$ data are from Pickett and Murrell (1997), Bourdon et al. (1999), Thomas et al. (2002), Dosseto et al. (2003), Asmerom et al. (2005), Turner et al. (2006), and this study, with error bars of 1 standard deviation of each arc. Average $(^{230}\text{Th})/(^{238}\text{U})$ is from DuFrane et al. (2006) and Figure 1. Subduction rates are from compilation in Turner et al. (2006) based on Jarrard (1986) and Plank and Langmuir (1988), except northern Lesser Antilles (LA) (Jarrard, 1986) and Southern Lesser Antilles (SLA) (Speed et al. 1993). Arcs: 1–12 as in Figure 1; 13—Eolian-Stromboli; 14—Northern LA; 15—SLA; 16—Bicol; 17—Bataan; 18—Java-Merapi; 19—Luzon.

portional to the subduction rate. The melting rate variations produced by this model as a function of subduction rate are then incorporated into different models of two-phase flow to assess the generation of U-series disequilibria as a function of convergence rate (see the Appendix in the GSA Data Repository¹ for details).

This melting rate model whereby water flux and melting rate are directly tied to convergence rate generally agrees with previous models of arc magmatism (Bourdon et al., 2003; DuFrane et al., 2006; Turner et al., 2003, 2006) and estimates from numerical models (Davies and Bickle, 1991). We emphasize that the correlation between melting rate and subduction rate in the physical model does not consider the effects of the subducted slab dip angle or thermal structure, peridotite enrichment and/or depletion in the mantle wedge or thickness of the overlying crust, and decoupling between subducted slab and mantle corner flow (Turner and Hawkesworth, 1998), all of which could also affect melting processes in an arc (Katz et al., 2003).

Because $D_{\text{mineral/melt}}$, D_{Th} , D_{Pa} , D_{Ra} are very small (<0.01), the generation of disequilibria during melting is often attributed to residence time differences reflecting differential partitioning of elements between moving melt phase and slower moving or stationary solid phase. This means that the generation of ^{230}Th - ^{238}U and ^{231}Pa - ^{235}U disequilibria is sensitive to melting rate, not degree of melting. In our model, both the mantle wedge flow velocity and water flux from the slab are directly tied to the slab subduction rate, resulting in a steady-state melting condition.

Recent studies have tried to relate the average $(^{230}\text{Th})/(^{238}\text{U})$ to the crustal thickness or subduction rate of a particular arc (e.g., DuFrane et al., 2006; Garrison et al., 2006). Although the majority of arc samples have ^{238}U excess, both ^{230}Th excesses and ^{238}U excesses can and often do exist in the same arc (e.g., Condomines and Sigmarsson, 1993), reflecting both the variations in fluid component added and possibly changes in the sense of $D_{\text{clinopyroxene/melt}} D_{\text{U-Th}}$ (Wood et al., 1999). Accordingly, the standard deviation of average $(^{230}\text{Th})/(^{238}\text{U})$ of a particular arc is usually quite large (e.g., Fig. 12 in DuFrane et al., 2006) making it difficult to discriminate any trend in the data. Thus, using the average $(^{230}\text{Th})/(^{238}\text{U})$ of an arc to constrain melting rates in arc settings might be questionable.

For comparing melting rate variations, ^{231}Pa excesses provide a distinct advantage over $(^{230}\text{Th})/(^{238}\text{U})$. Unlike the source of mid-ocean ridge basalts and ocean island basalts, which can be assumed in secular equilibrium at the ini-

tiation of melting, the mantle wedge is likely to have an initial ^{238}U (and ^{235}U) excess due to recent addition of U-rich fluids. Aging or contribution of sediment melt can dilute or eliminate the fluid effect (Turner et al., 2003), making the initial condition even more uncertain. There are three major advantages to using $(^{231}\text{Pa})/(^{235}\text{U})$ to examine melting rates relative to $(^{230}\text{Th})/(^{238}\text{U})$ disequilibria. First, melting-generated $(^{230}\text{Th})/(^{238}\text{U})$ variations are small relative to the $(^{230}\text{Th})/(^{238}\text{U})$ generated by fluids or sediment melts. In contrast, melting-generated $(^{231}\text{Pa})/(^{235}\text{U})$ variations are very large relative to those of fluid and sediment melt-induced starting compositions. Second, no shift in sense of relative U-Pa partitioning occurs within the melt column, unlike the case for U-Th, where $D_{\text{Th/U}}$ can vary from <1 to >1 (Wood et al., 1999). Third, the shorter half-life of ^{231}Pa results in this system having less memory of fractionations generated by fluids or sediment melts. This last effect can be seen in our models, which vary the initial state of the mantle source to assess the effect of recent contributions of materials from the subducted slab (Fig. 2).

Oxygen fugacity (fO_2) of the mantle wedge may also be an important variable affecting U-series disequilibria in arcs. The mantle wedge is more oxidizing than either oceanic or cratonic mantle (Parkinson and Arculus, 1999), and the partitioning of U between mantle peridotite and silicate melt strongly depends on fO_2 (Beattie, 1993; Lundstrom et al., 1994). Given the high but possibly variable fO_2 in the mantle wedge of typical arc settings, we decrease D_U but keep D_{Th} , D_{Pa} , and D_{Ra} constant in the in-growth melting models to assess this variable. Results show that decreasing D_U results in lower ^{231}Pa excess but greater ^{238}U excess. Therefore, reconciling the high ^{231}Pa and ^{238}U excesses in the SLA lavas may provide constraints on the range of D_U at the higher fO_2 of arc settings. For example, to produce simultaneous high ^{231}Pa and ^{238}U excesses using a reactive porous flow model, we decrease D_U by a factor of two to 0.0026. However, the use of a smaller D_U (1.56×10^{-3}) results in lower ^{231}Pa excess than that observed at KEJ, while a larger D_U (5.2×10^{-3}) produces slight ^{230}Th excess, also inconsistent with the observations.

The advantage of using $(^{231}\text{Pa})/(^{235}\text{U})$ compared to $(^{230}\text{Th})/(^{238}\text{U})$ for melting rate determination is apparent (Fig. 2). The $(^{230}\text{Th})/(^{238}\text{U})$ varies widely in a given arc (e.g., Turner et al., 1996; Elliott et al., 1997; George et al., 2003), making it difficult to definitively observe a trend in average $(^{230}\text{Th})/(^{238}\text{U})$ with subduction rate (Fig. 2B). This variation is consistent with our models, showing that this probably reflects variations in initial conditions. In contrast, the trends in $(^{231}\text{Pa})/(^{235}\text{U})$ are defined by convergence rate end members, Tonga and SLA. Tonga has the highest convergence rate and lowest average ^{231}Pa excess, while the SLA has the opposite fea-

tures. Data for intermediate convergence rates are more ambiguous. This wide range in ^{231}Pa excess for a given arc could reflect variations in initial $(^{231}\text{Pa})/(^{235}\text{U})$ due to varying slab input. However, all three models consistently show that at low convergence rate, this initial condition becomes insignificant such that all three produce high ^{231}Pa excess and negatively curved trends between subduction rate and ^{231}Pa excess. With a slow subduction rate, the three models can also produce ^{226}Ra excess (not shown). For example, $(^{226}\text{Ra})/(^{230}\text{Th})$ generated by the reactive porous flow (RPF) model is as high as 3.6, consistent with data of KEJ samples. Thus, the best test of our model will be analyses of other arc setting volcanoes where convergence rate is low. Notably, the average $(^{231}\text{Pa})/(^{235}\text{U})$ values of young hotspot lavas also show negative correlation with buoyancy flux of upwelling regions (Bourdon et al., 2006). In this case, average $(^{231}\text{Pa})/(^{235}\text{U})$ as a function of melting rate can be associated with other geophysical parameters such as buoyancy flux of upwelling buoyant hotspots (Bourdon et al., 2006).

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¹GSA Data Repository item 2007248, Appendix, $(^{231}\text{Pa})/(^{235}\text{U})$ data, and models, is available online at www.geosociety.org/pubs/ft2007.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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