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Assimilation of the plutonic roots of the Andean arc controls variations in U-series disequilibria at Volcan Llaima, Chile

O. Reubi [a,b,*], B. Bourdon [a,1], M.A. Dungan [b], J.M. Koornneef [a], D. Sellés [c,d], C.H. Langmuir [c], S. Aciego [a]

[a] ETH Zurich, Institute of Geochemistry and Petrology, Switzerland
[b] University of Geneva, Earth Sciences, Switzerland
[c] Harvard University, Earth and Planetary Sciences, USA
[d] Sernageomin, Chile

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ABSTRACT

U-series disequilibria provide important constraints on the processes and time scales of melt production, differentiation, and transport in subduction settings. Such constraints, which are essential for understanding the chemical evolution of the continental crust, are conventionally based on the assumption that the U-series disequilibria measured in mafic lavas are produced during mantle metasomatism and melting, and that intracrustal differentiation and assimilation have limited impacts. Here we show that mantle-derived U-series disequilibria in mafic lavas erupted at Volcán Llaima, Chile are significantly diminished by assimilation of plutonic rocks forming Llaima's subvolcanic basement. This contamination process is extremely subtle in terms of "classical" indicators of crustal assimilation like Sr, Nd or Pb isotopes because it is a manifestation of assimilative recycling of the plutonic roots of the arc. This process results in variations in U-series disequilibria and incompatible trace element ratios that are significant compared to regional and global variability in arc magmas. Furthermore, it yields linear correlations between U-series excesses and incompatible trace element ratios that are generally interpreted as slab-fluid indicators and chronometers, or tracers of sediment recycling in subduction zone. Cannibalization of ancestral magmas by ascending melts warrants careful evaluation when considering the components and chemical fluxes in subduction zones. Linear arrays defined by activity ratios of U-series nuclides with different half-lives may be the most reliable indicators of assimilative recycling of ancestral intrusive magmas.

1. Introduction

Disequilibria between short-lived nuclides produced in radioactive decay chains of U and Th are excellent time-dependent tracers of magmatic processes. U-series disequilibria measured in arc magmas can provide constraints on the processes and time scales of mantle metasomatism, melt production, magma differentiation, and mass transport above subduction zones (see, Turner et al., 2003, and references herein). These constraints are essential for quantifying chemical fluxes through subduction zones, with important implications for the chemical evolution of the continental crust, the dynamics volcanic eruptions, and the formation of ore deposits.

Arc lavas generally have excesses of ^{238}U over ^{230}Th [i.e. (^{230}Th/^{238}U)<1; where parentheses denote activity ratios] and large excesses of ^{226}Ra relative to ^{230}Th. As both U and Ra are fluid mobile elements, and these excesses correlate with trace element ratios indicative of metasomatism by fluids (e.g. Ba/Th), they are generally attributed to recent addition of slab-fluid to the mantle wedge (Allegre and Condomines, 1982; Elliott et al., 1997; Gill and Williams, 1990; McDermott and Hawkesworth, 1991; Turner and Hawkesworth, 1997; Turner et al., 2000). The broad linear correlations between (^{230}Th/^{232}Th) and (^{238}U/^{232}Th) observed for suites of lavas from single arc have been interpreted as an indication that the time span between fluid addition to the mantle wedge and eruption is typically between 20 and 150 kyr (Sigmarsson et al., 1990; Turner and Hawkesworth, 1997; Turner et al., 2001). Preservation of ^{226}Ra excesses in arc magmas indicate much shorter time scales, possibly as short as few hundred years (Turner et al., 2001). These apparent discrepancies in the time scales of slab fluid addition have been interpreted to result from multistage dehydration events in which the first fluid fluxes contain ^{238}U and ^{226}Ra, when the last fluids expelled contain only ^{226}Ra produce by in-growth from residual ^{230}Th in the slab (Turner et al., 2000). The excesses of ^{231}Pa over ^{235}U observed in the majority of arc lavas are taken as an indication that the effects of partial melting processes overprint those of slab-fluid (Bourdon et al., 1999; Huang and Lundstrom, 2007; Pickett and Murrell, 1997; Thomas et al., 2002; Turner et al., 2006). The broad negative correlation between average (^{231}Pa/^{235}U) and convergence rate in all arc settings has been

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* Corresponding author.
E-mail address: olivier.reubi@erdw.ethz.ch (O. Reubi).

[1] Now at: Ecole Normale Supérieure de Lyon and CNRS, France.

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interpreted as an indication that mantle melting rates strongly control ^{231}Pa excesses and are coupled to the subduction rate (Huang and Lundstrom, 2007; Turner et al., 2006).

These constraints on the time scales and processes of mantle metasomatism and partial melting are obtained from regional to global data sets. However, detailed studies on single volcanoes often reveal significant ranges in U-series disequilibria with respect to regional and global variations (e.g. Turner et al., 2007; Yokoyama et al., 2006), demonstrating that second-order local processes operating over relatively short time scales may modify mantle-derived U-series disequilibria. Several recent studies have shown that processes affecting the magmas during ascent through the crust (e.g. crustal assimilation, crystallization) can have significant leverage on U-series disequilibria (e.g. Bourdon et al., 2000; Garrison et al., 2006; Handley et al., 2008; Huang et al., 2008; Price et al., 2007; Yokoyama et al., 2006). To which extent crustal processes may have modified the signature of primary genetic processes (e.g. fluid addition to the mantle wedge, partial melting regime) needs to be considered to ascertain the reliability of time scales inferred from regional to global U-series data sets.

Here, we investigate the extent and cause of local and short-term temporal variability in U-series disequilibria in lavas from Volcán Llaima, Chile, one of the most historically active volcanoes in South America. To this end, we measured isotope concentrations of U, Pa, Th and Ra by mass spectrometry on multiple samples from each historic eruptive event for which vents and timing are reasonably well documented (Naranjo and Moreno, 2005).

2. Geological setting

Volcán Llaima, Chile is one of the most historically active volcanoes in South America. It is located at 38.7 °S in the Andean Southern Volcanic Zone (SVZ; 33–46 °S) (Fig. 1). The SVZ overlay the section of the subducting slab bounded to the north and south respectively by the subducted Juan Fernandez Ridge and the Chile Rise. The Nazca oceanic plate being subducted beneath the central SVZ is ~30 Ma old and converges on the Andean margin at a rate of ~10 cm/yr (Tassara et al., 2007). The SVZ segment in which Llaima is located is underlain by 30–40 km thick continental crust and is characterized by the predominance of basaltic andesite stratovolcanoes whose magmas show relatively minor elemental or isotopic (Nd–Sr–Pb–O) indications of crustal contamination on their magma chemistry (Hickey et al., 1986; Hickey-Vargas et al., 2002; Tormey et al., 1991). The Andean crust thickens to 55 km from 36 to 33 °S and andesite to dacite magmas with geochemical features indicative of significant crustal contamination predominate (Davidson et al., 1988; Hickey et al., 1986; Hildreth and Moorbath, 1988). The subvolcanic basement beneath Llaima comprises Tertiary diorite to granodiorite plutons related to earlier magmatism of the arc (Lucassen et al., 2004) (Fig. 1).

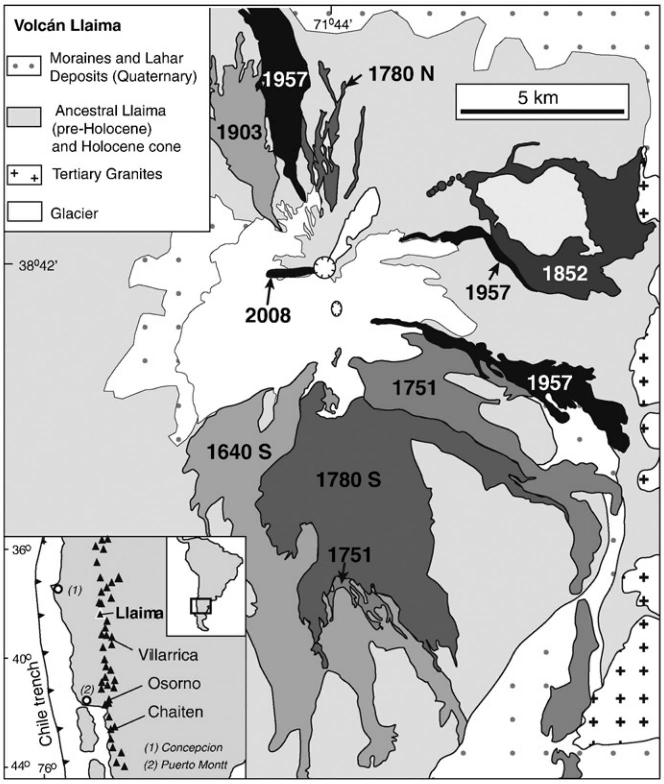

Fig. 1. Geological map of Volcán Llaima modified from Naranjo and Moreno (2005). The map focuses on the Holocene cone. Historic lava flows are represented by grey shaded areas labeled with the eruption year. Note the Tertiary granites that outcrop on the eastern side of Volcán Llaima. Inset map shows that location of Volcán Llaima in the Andean Southern Volcanic Zone.

Holocene volcanic activity at Llaima began with caldera collapse and eruption of the basaltic to andesitic (~52–58% SiO_2, our unpublished data) Curacautin Ignimbrite (~13.5 ka). Eruptions from a N–S central-vent system and oblique-trending flank fissures buried this caldera during the late Holocene. Historical volcanic activity of Llaima consists predominantly of Strombolian events. Six major effusive eruptions occurred between 1640 and 1957 (Fig. 1). The most recent eruptive activity started in May 2007 and it has been marked by short but vigorous Strombolian phases in January 2008 and April 2009, both of which subsequently produced small lava flows. Samples representative of the historical activity are presented here and were selected from a sample collection that includes more than 450 samples analyzed for major elements by XRF (Dungan et al., unpublished results). All historical samples are evolved basalts (>51% SiO_2, <6.5% MgO) to basaltic andesites (<55% SiO_2, >3.9% MgO) and contain mineral assemblages with widely variable proportions of plagioclase, olivine, one or two pyroxenes, and spinel.

3. Analytical techniques

Major elements were determined by X-ray fluorescence (XRF) on fused glass discs on a Phillips PW 2400 spectrometer equipped with a rhodium tube at the University of Lausanne (Switzerland), following the methods described by Pfeifer et al. (1991). Trace element concentrations were determined using a Thermo X-series ICP-MS quadrupole at Harvard University. The samples (50 mg) were processed using the method described in Bezos et al. (2009). ^{72}Ge, ^{103}Rh, ^{115}In, ^{169}Tm, and ^{209}Bi were used for internal standard normalization and the following rocks were used for calibration curves: BHVO-2, BCR-2, AGV-1, DNC-1, and JB-2, as well as the in-house standard Mid-Atlantic Ridge basalt (MAR). The in-house standard K1919 was also analyzed and used for drift correction. Trace element data obtained by ICP-MS are accurate to within ±5% on the basis of duplicate analyses.

Neodymium isotopic ratios were measured on a Nu Instrument® MC-ICP-MS at ETH Zurich. For each sample, about 100 mg of rock powder was digested in a mixture of HF–HNO_3, followed by neodymium separation by extraction chromatography using Eichrom TRU and LN spec resins columns following the procedure of Pin and Zalduegui (1997). The sample solutions were introduced using an Apex desolvation system. $^{143}Nd/^{144}Nd$ isotope ratios were corrected for mass fractionation using a $^{146}Nd/^{144}Nd$ ratio of 0.7219 and normalized to the JNdi-1 standard value of 0.512115 (Tanaka et al., 2000). Repeated measurements of JNdi-1 during the analysis period yielded a mean $^{143}Nd/^{144}Nd = 0.512111 \pm 29$ (2 S.D. $n = 14$).

Full details of the chemical separation and analytical techniques used for U-series analyses are given in Koornneef et al. (2010), and an outline is given here. U, Pa, Th, and Ra concentrations and isotope ratios were determined on single aliquots (about 800 mg) spiked with ^{236}U, ^{229}Th, ^{233} Pa, and ^{228}Ra tracers before dissolution in HF–HNO_3