Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of Volcanology and Geothermal Research 140 (2005) 171–191

Journal of volcanology and geothermal research

www.elsevier.com/locate/jvolgeores

Magma evolution and ascent at volcanic arcs: constraining petrogenetic processes through rates and chronologies

G.F. Zellmer[a,*], C. Annen[b], B.L.A. Charlier[c,1], R.M.M. George[d], S.P. Turner[d], C.J. Hawkesworth[e]

[a]*Lamont-Doherty Earth Observatory, 61 Route 9W, Palisades, New York 10964, USA*
[b]*Département de Minéralogie, Université de Genève, 1205 Genève, Switzerland*
[c]*Department of Geological Sciences, University of Durham, South Road, Durham DH1 3LE, UK*
[d]*GEMOC, Department of Earth and Planetary Sciences, Macquarie University, Sydney, NSW 2109, Australia*
[e]*Department of Earth Sciences, University of Bristol, Wills Memorial Building, Queens Road, Bristol BS8 1RJ, UK*

Received 4 October 2003; received in revised form 12 March 2004; accepted 15 July 2004

Abstract

Large ^{226}Ra excesses in a number of mafic arc magmas, and geophysical observations of earthquake hypocenters locations, indicate that mafic melts can get transferred from source to surface within days to ~1 kyr. Decreasing ^{226}Ra–^{230}Th disequilibria with increasing SiO_2 in co-magmatic suites of individual arc volcanoes suggest that magma differentiation often occurs on timescales of a few thousand years in closed systems, or less in open systems. However, the rapid decrease of global ^{238}U–^{230}Th disequilibria with increasing SiO_2 within basaltic andesite compositions suggests that rapid closed system differentiation does not typically produce more evolved compositions from mafic parental magmas with large ^{238}U excesses. In the case of rapid magmatic evolution, open system processes are frequently involved in the production of andesites and dacites, and the data imply that the open system component is close to ^{238}U–^{230}Th secular equilibrium. MELTS modeling of magma evolution at Santorini in the Aegean arc provides corroborating evidence from an individual well-studied arc volcano, that closed system fractional crystallization fails to produce the more evolved compositions, and increasing ^{87}Sr/^{86}Sr ratios with increasing SiO_2 from andesites to rhyolites suggests assimilation of old crust is involved. These observations can be conceptualized through thermal modeling of basaltic sill injections into the lower and upper crust. Melt production in deep crustal hot zones provides an explanation for low ^{238}U–^{230}Th disequilibria of the evolved magmatic component, as significant accumulation of evolved melts through partial re-melting of previously intruded basalts requires >100 kyr incubation time at typical magmatic flux rates. Thermal modeling also provides a framework for understanding assimilation of old upper crustal rocks in more mature magmatic systems. Further insights into magmatic evolution can be gained from dating of minerals from mafic, intermediate and felsic arc magmas. (a) Many U–Th mineral isochrons in *mafic* arc magmas yield ages significantly older than those obtained from Ra–Th mineral isochrons from the same samples. A crystal size distribution case study from Soufrière, St.

* Corresponding author. Now at: Department of Geology and Geophysics, SOEST, University of Hawaii at Manoa, 1680 East West Road, Honolulu, Hawaii, HI 96822, USA. Tel.: +1 808 956 5033; fax: +1 808 956 5512.
E-mail address: gzellmer@hawaii.edu (G.F. Zellmer).
[1] Present address: Department of Earth Sciences, The Open University, Walton Hall, Milton Keynes MK7 6AA, UK.

0377-0273/$ - see front matter
doi:10.1016/j.jvolgeores.2004.07.020

Vincent, indicates that this may result from the abundance of cumulate xenocrysts within more mafic arc magmas. (b) The majority of U–Th mineral isochrons in *intermediate* arc magmas yield ages within error of eruption age, indicating that other dating techniques that may be more suitable for deciphering the shorter timescales yielded by these compositions. In a case study of crystals of remobilized andesites from the Soufrière Hills volcano, Montserrat, intra-crystalline trace element diffusion modeling yields crystallization timescales of the order of 10–1000 years. Varying temperature-time histories of individual crystals can be interpreted to reflect multi-stage magma ascent and the development of ephemeral subvolcanic magma reservoirs. (c) U–Th mineral isochrons from *felsic* arc magmas yield very variable ages from >200 ka to shortly prior to eruption. A case study from the Taupo Volcanic Zone has shown that both remobilization of previous intrusives and melting of old country rock are occurring, giving rise to a this range in ages. Both intermediate and felsic magmas therefore can be shown to have multi-stage ascent histories. Future work will have to address the reasons for the apparent inter-volcano variability in time-averaged subvolcanic magma volumes within the arc crust. These include differences in magma supply rate; thickness, composition and thermal structure of the crust; and magma composition and volatile content, which may vary between different tectonic settings.

Keywords: magma evolution; ascent; volcanic arc; petrogenetic process

1. Introduction

Recently, a number of different techniques have been employed to study the rates and chronologies at arcs (for a summary, see Hawkesworth et al., 2004). Whole-rock data, mineral isochrons (for recent reviews, see e.g., Condomines et al., 2003; Turner et al., 2003a), in-situ analysis of major mineral phases (e.g., Gerlach and Grove, 1982; Nakamura, 1995; Zellmer et al., 1999; Costa et al., 2003; Zellmer et al., 2003b) and accessory mineral phases (e.g., Reid et al., 1997; Charlier and Zellmer, 2000; Reid and Coath, 2000; Charlier et al., 2003), and crystal size distribution (CSD) studies (e.g., Marsh, 1988; Higgins, 1996a, 1996b, 2000; Turner et al., 2003c) are all holding clues to the processes responsible for magmatic differentiation in arc crust. The timing of magmatic differentiation and of episodes of crystallization provides constraints on the dynamics of the magmatic plumbing system beneath arcs. Magmatic differentiation by crystallization has long been inferred from the abundance of crystals within erupted magmas (Bowen, 1928), yet in recent years it has become evident that many of these crystals are xenocrysts or ‘antecrysts’ (term coined by W. Hildreth, and implying that they are derived from young plutons or crystal mushes) and are not representative of fractionating assemblages that could be associated with the magmatic differentiation of their host (e.g., Hawkesworth et al., 2000). Differentiation studies therefore need to include the temporal compositional evolution of liquids (or whole-rocks for elements that preferentially partition into the liquid).

Using a variety of available data, his contribution aims to answer some of the remaining issues regarding the rates and processes of magma evolution and ascent at volcanic arcs:

- At what rate do arc magmas ascent from source to surface, and what is their ascent history?
- Does magmatic evolution occur dominantly by closed system crystal fractionation, or does it involve open system processes such as magma mixing, crustal assimilation, and advection of cumulate crystals?
- How do the rates of such closed and open system processes differ?
- What is the age and history of the crystals carried in arc magmas? Where and how do they form, and what insights do they provide about the magmatic processes at volcanic arcs?
- How do these rates and processes of arc magma evolution and ascent change from mafic through intermediate to felsic magma compositions?

In order to address these questions, this study initially considers whole rock U–Th–Ra data of individual arc volcanoes, which can be used in conjunction with the global arc dataset to infer timescales of magma differentiation in both closed and open systems. MELTS modeling from Santorini corroborates the conclusion that rapid differentiation to andesites and dacites often requires open system processes such as assimilation of more evolved compositions. It will then be shown that observations from whole-rock studies can be understood in the context of thermal modeling of repeated basaltic intrusions into the arc crust. The second part of this paper considers the insights that can be gained from mineral chronology. After a general overview of available U–Th–Ra mineral ages from arc volcanoes, three case studies from Soufrière (St. Vincent), Montserrat, and Taupo, are used to show how a variety of techniques can be employed to provide further insights into the crystallization histories of mafic, intermediate and felsic magmas, respectively. The paper concludes with a brief outlook on future work necessary to gain more detailed insights into the fundamental parameters that govern rates and chronologies of magma ascent, storage and evolution in arcs.

2. Insights from global trends in U-series whole-rock data

One of the most straightforward ways to assess the time scales of differentiation sensu lato (i.e. irrespective of whether this is caused by crystal fractionation, magma mixing, assimilation or some combination thereof) is to investigate how whole rock radioactive disequilibria vary with indices of differentiation. For example, if differentiation takes of the order of the half-life of the isotope being plotted, then disequilibria will decrease with decreasing MgO and increasing SiO_2. Several groups have demonstrated systematic decreases in U-series disequilibria with increasing differentiation using global datasets (Reagan et al., 2003; Turner et al., 2003a). It is apparent from Fig. 1a that the largest excesses of ^{226}Ra over ^{230}Th at the time of eruption are found in basalts and basaltic andesites, with (^{226}Ra/^{230}Th) activity ratios of up to ~6. ^{226}Ra has a half-life of 1.6 kyr, and any ^{226}Ra excesses over ^{230}Th therefore decay to secular equilibrium within ~8 kyr. The observation that the most mafic samples show the highest (^{226}Ra/^{230}Th) ratios suggests that ^{226}Ra excesses are a primary feature of mantle derived magmas, and that transfer of some mafic magmas from source to surface may only take of the order of 1 kyr or less. This is consistent with geophysical evidence that magma ascent through the crust may be extremely fast and occur on timescales of days. For example, preeruptive earthquakes prior to the 1975 Great Tolbachik fissure eruption in Kamchatka were recorded to initiate at the base of the crust less than 10 days prior to eruption, becoming shallower with time. Thus, magma ascent through the crust demonstrably can be as fast as 1–3.5 km per day (Fedorov and Markhinin, 1983). Even faster magma ascent rates of $\geq$26 km per day have been estimated from the preservation of ultramafic hornblende-peridotite xenoliths from central Mexico, which reached the surface so rapidly that they were not affected by dissolution in their host magma during ascent from the mantle (Blatter and Carmichael, 1998).

A decrease in ^{226}Ra–^{230}Th disequilibria with increasing silica within intra-oceanic arc suites is evident in Fig. 1a. Since the early 1990's, diminishing ^{226}Ra excesses towards more evolved magmas have been interpreted to reflect differentiation from basalts to andesites and dacites within millennia (e.g., Gill and Williams, 1990; Turner et al., 2001). This assumes that: (a) the magmatic system studied consists of a number of magma pulses, which all originate from the same parental melt, or from melts with similar primary disequilibria; (b) some of these magmas erupt after small amounts of differentiation, while others differentiate further prior to eruption; and (c) different pulses of magma coexisted as separate batches at any one time. The above conditions will most plausibly be met in lava suites from individual arc volcanoes (e.g. Condomines et al., 1995). Very detailed geochemical studies of such suites can elucidate if different magma compositions are comagmatic, and can also give insights into whether rapid magmatic evolution occurs through closed system or open system differentiation.

For example, Sangeang Api volcano erupts a comagmatic suite of potassic, cumulate-xenolith bearing lavas that span 47–54% SiO_2 in the Sunda reararc (Turner et al., 2003b). Geochemical data suggest that the compositional variation in these products reflects polybaric, fractional crystallization as the magmas passed across the Moho. U-series data show near constant (^{230}Th/^{232}Th) in lavas, bulk cumulates and separated minerals. Thus, it would appear that differentiation occurred faster than could be recorded

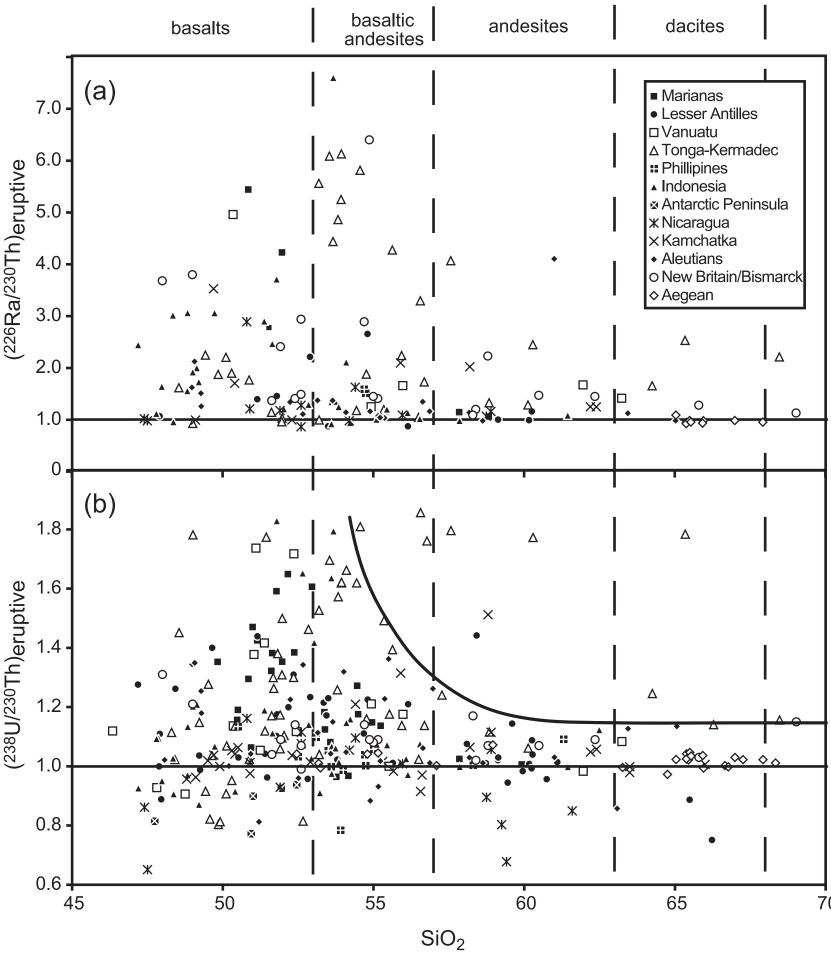

Fig. 1. Initial U-series activity ratios as a function of silica content for whole-rock samples from a selection of volcanic arcs, analyzed by TIMS (Woodhead, 1989; McDermott and Hawkesworth, 1991; Gill et al., 1993; Reagan et al., 1994; Turner et al., 1996, 1997, 1998, 1999, 2001; Elliott et al., 1997; Hawkesworth et al., 1997; Heath et al., 1998a, 1998b; Zellmer, 1998; Williams, 2001; Turner and Foden, 2001; George et al., 2003a; Zellmer et al., 2003a) Activity ratios of 1.0 indicate secular equilibrium. (a) ($^{226}Ra/^{230}Th$) activity ratios at the time of eruption. (b) ($^{238}U/^{230}Th$) activity ratios at the time of eruption. Symbols as in (a). The thick solid curve indicates the general decrease of ^{238}U excesses over ^{230}Th towards more evolved compositions. See text for discussion.

by the half-life of ^{230}Th. However, ($^{226}Ra/^{230}Th$) systematically decreases with increasing SiO_2 and Th, suggesting that the time scale for differentiation was of the order of a few 1000 years. In this instance, this time scale is corroborated by Ra–Th ages of minerals from cumulates. In a similar study, George et al. (2003b) have studied the tholeiitic Akutan volcano and the calc-alkaline Aniakchak volcano in the Aleutian arc. Historic tholeiitic magmas at Akutan have ($^{226}Ra/^{230}Th$) activity ratios that decrease from 1.18 to almost equilibrium values as SiO_2 increases from 54.3–57.3 wt.%, whereas ($^{238}U/^{230}Th$) ratios do not vary significantly (1.16–1.23). At Aniakchak, the magnitude of the ($^{226}Ra/^{230}Th$) activity ratios in calc-alkaline lavas is the same, but decreases over a considerably larger silica range (57–68 wt.%). ($^{238}U/^{230}Th$) ratios straddle the equiline between 0.85 and 1.13. The Akutan lavas can be explained by closed system magmatic evolution whereas curvilinear trace element trends and a significantly larger range in $^{87}Sr/^{86}Sr$ isotope ratios in the Aniakchak data compared to Akutan appear to require the combined effects of fractional crystallization, assimilation and magma mixing. The comparable range in ^{226}Ra–^{230}Th disequilibria of the two volcanoes suggests that the time scale of crustal residence of magmas was similar and on the order of several thousand years, but implies that the open-system processes at Aniakchak permitted more time-efficient differentiation.

The persistence of ^{226}Ra excesses in some siliceous island arc lavas (Fig. 1a) is in contrast to the rapid decrease of ^{238}U excesses with increasing differentiation (Fig. 1b). As ^{230}Th has a half-life of ~76 kyr (Cheng et al., 2000), (^{238}U/^{230}Th) activity ratios should not be affected significantly on timescales of <10 kyr. However, with the exception of some Tongan samples, large ^{238}U-excesses of up to 80% are restricted to mafic magmas with <55 wt.% SiO_2, and initial ^{238}U excesses do not exceed 20% in the majority of the more evolved samples. A similar decrease of U excesses with increasing SiO_2 is displayed in a number of continental arcs (Reagan et al., 2003). While low (^{238}U/^{230}Th) dacites may have evolved from mafic parents with limited initial ^{238}U excesses, the lack of concordance between ^{226}Ra–^{230}Th and ^{238}U–^{230}Th disequilibria implies that those more siliceous compositions which show marked ^{226}Ra excesses cannot be generated by closed system magmatic evolution from parental magmas with large ^{238}U excesses. The large ^{226}Ra excesses in some dacites may therefore be created during differentiation, e.g. by mixing with less evolved compositions that are in greater ^{226}Ra–^{230}Th disequilibrium.

From the above case studies and the global U-series systematics it can be concluded that rapid closed system differentiation can occur within the more mafic samples, and that rapid differentiation to more evolved compositions may often require open system processes such as assimilation or magma mixing, implying that the assimilated component is close to secular equilibrium and thus old. It should however be noted that rapid magmatic evolution is not always the norm at arcs, and that there is, for example, some evidence for closed system evolution from basalts to andesites and even dacites, e.g. in Japan (Kuno, 1950; Fujinawa, 1988, 1990; Tamura et al., 2000; Kimura et al., 2001). Unfortunately, detailed U-series studies have not yet been conducted in these settings. To demonstrate that there may often be a profound change in the differentiation mechanism from initially closed system to subsequently open system evolution, a case study of Santorini in the Aegean Volcanic arc is presented below.

3. Evolution to more evolved compositions: an example from Santorini, Aegean volcanic arc

3.1. Regional tectonics and seismic observations

The Aegean volcanic arc is formed by northeastward subduction of the African plate beneath the Aegean microplate at the Hellenic trench system, which is composed of a sequence of northwest–southeast trending thrust faults connected through transform faults (Fig. 2). Clusters of shallow earthquakes (<65 km) occur near the transform faults, suggesting that the subducting African plate fractures to accommodate subduction to the north and transform motion to the south of each transform fault. Intermediate depth earthquakes (65–120 km) occur within the subducting slab because crustal thickness does not exceed 55 km anywhere in the Aegean (Makris, 1978). A zone of intermediate depth earthquakes runs northeastwards from the most prominent cluster of shallow earthquakes at 35 °N 23 °E, in the direction of subduction and towards Santorini. Immediately adjacent to this zone of earthquakes, the subducting slab is largely aseismic, suggesting that most of the deformation within the slab takes place where previous structural weakening occurred due to trench segmentation (cf. Fig. 2). The deepest earthquakes (>120 km) are found furthest away from the trench, beneath the active volcanic arc. The volcanic centers of Milos, Santorini, and most notably Nisyros are situated above clusters of deep earthquakes. As fractures in the subducting slab are likely to facilitate slab fluid release into the overlying mantle wedge, thereby increasing the amount of mantle melting, it is argued here that slab fracturing and mantle melt production are intimately associated. Santorini is situated above a linear zone of intermediate depth earthquakes that are interpreted to represent a very prominent and localized zone of slab deformation, resulting in greater activity at this volcanic site compared to the other active volcanic centers along the arc.

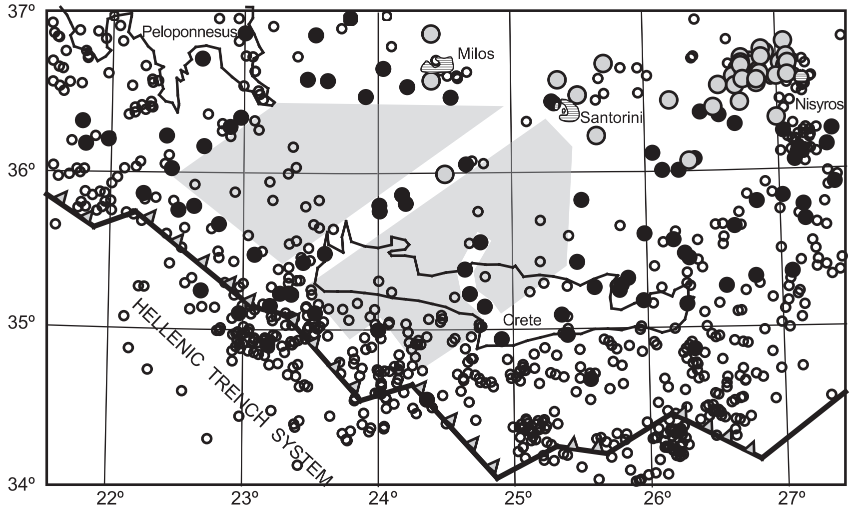

Fig. 2. Earthquake epicenter map of the South Aegean, UTM projection. The position of the South Aegean volcanic centers of Milos, Santorini and Nisyros are indicated (hatched), as are the coastlines of the Peloponnesus peninsula and Crete. The thick dark line shows the Ionian Trench system in the southwest of the Hellenic Trench, where the African Plate subducts northeastwards beneath the Aegean microplate. Earthquake epicenters are plotted for shallow (<65 km, small open circles), intermediate (65–120 km, black circles), and deep (>120 km, light grey circles) teleseismic and microseismic events (Papazachos et al., 1991; Hatzfeld and Martin, 1992; Hatzfeld et al., 1993; Papazachos and Panagiotopoulos, 1993; Engdahl et al., 1998). Standard errors on epicenter locations and focal depths are generally better than ±10 km (Engdahl et al., 1998). Grey shading indicates areas where intermediate and deep earthquakes are absent. See text for discussion.

3.2. MELTS fractional crystallization modeling at Santorini

MELTS modelling (Ghiorso and Sack, 1995) has been conducted to see if magmatic evolution at Santorini can be explained by closed system fractional crystallization. Although the MELTS calibration database is dominated by basaltic compositions, a considerable number of more evolved compositions have also been taken into account, notably at the relatively high total alkali contents also displayed by samples from Santorini (Zellmer et al., 2000). This suggests that the liquid–solid equilibria predicted by MELTS may be applicable to the entire compositional range of volcanic products erupted at Santorini during the last 200 kyr (Fig. 3). One of the less evolved samples is used as starting composition, and initial pressure, temperature and oxygen fugacity constraints are taken from the literature (see Table 1 and caption to Fig. 3 for details). As the H_2O content of a cooling magma strongly influences the onset and extent of plagioclase crystallization, and as plagioclase fractional crystallization affects the Al_2O_3 content of the melt, Fig. 3a serves to constrain the H_2O content of the starting composition. High H_2O contents inhibit early plagioclase fractionation. To model the relatively rapid decrease in Al_2O_3 with increasing SiO_2 within the basaltic andesites (53–57 wt.% SiO_2), early plagioclase removal is required, thus constraining the H_2O content of the starting composition to ~0.7 wt.%, consistent with previous estimates (Huijsmans, 1985) for the more mafic compositions. Closed system fractional crystallisation can explain differentiation to 58–60 wt.% SiO_2, i.e. andesitic compositions. At more evolved compositions and temperatures less than 1050 °C, however, MELTS predicts extensive plagioclase fractionation, which is not observed in the data. Increasing the modelling pressure to 3 kbar to suppress plagioclase fractionation does not have a significant effect on the modelled fractionation trends (Fig. 3b). These observations indicate that at more evolved compositions, closed system fractional crystallisation cannot be invoked to explain the observed differentiation trends at Santorini.

3.3. Alternative differentiation processes

Difficulties in modeling whole-rock differentiation trends purely by closed system fractional crystallization indicate that other, open system processes may be

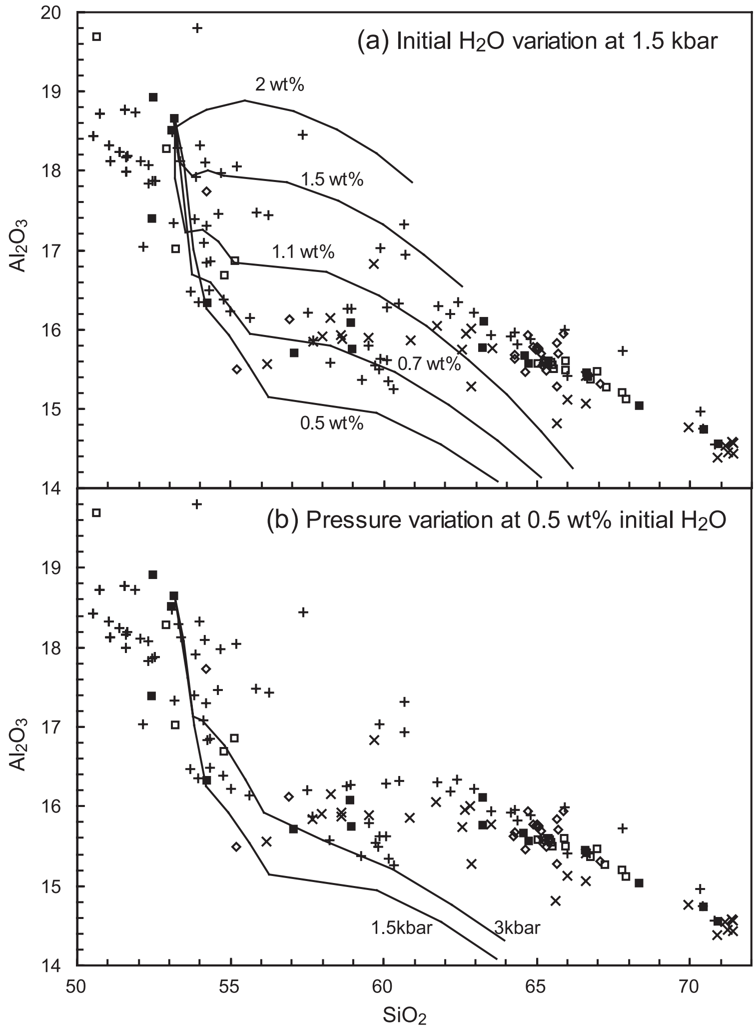

Fig. 3. Magmatic evolution of Santorini WR samples erupted during the last 200 kyr. These include the second eruptive cycle (+,■) and the Kameni lavas (◇, □). Data sources: + and ◇ are from Huijsmans (1985), × from Edwards (1994), and ■ and □ from Zellmer et al. (2000). Although a range of at least 5 wt.% in the SiO_2 content of the parental magmas is required to explain the observed scatter, the mafic sample "as2-Oia-1" from Zellmer et al. (2000) has been chosen here as starting composition for MELTS modeling. A starting temperature of 1160 °C was assumed, based on olivine-liquid geothermometry estimates on samples of similar SiO_2 content (cf. Table 1). As pre-eruptive pressures have a limited range from 1–3 to 1–2 kbar for mafic and felsic samples, respectively, isobaric crystallization was assumed. Oxygen fugacities are buffered slightly above the Ni–NiO buffer (Huijsmans, 1985), and so MELTS modeling was conducted one log-unit above the quartz-fayalite-magnetite (QFM) buffer. (a) Effect of variable initial water content at 1.5 kbar. Major inflections in the modeled magmatic evolution are the result of changes in the fractionating phase assemblage, from plag only in all but the most hydrous model through plag+px to plag+px+mt. (b) Effect of variable pressure of crystallization at and initial H_2O content of 0.5 wt.%.

important. For example, mixing of mafic and felsic melts could potentially produce intermediate magma compositions. At Santorini, there is some evidence for magma mixing, based on the occurrence of compositionally banded pumices and hybrid andesites (Druitt et al., 1996), resorption textures within phenocrysts (Stamatelopoulou-Seymour et al., 1990) and crystal size distribution studies on dacitic lavas (Higgins, 1996a). However, inflections of the major element differentiation trends at variable SiO_2 abundances, namely at 56 wt.% SiO_2 for Fe_2O_3, MgO and Al_2O_3 (Fig. 3), at 59 wt.% SiO_2 for TiO_2, at 62 wt.% SiO_2

Table 1
Estimates of pre-eruptive temperatures, pressures, oxygen fugacities and water contents of Santorini magmas, after (Huijsmans, 1985) and (Barton and Huijsmans, 1986)

	Temperature (°C)	Pressure (kbar)	Oxygen fugacity	Water content (wt.%)
Basalts	1150–11200	1–3	~10^{-7}	<0.5
Basaltic andesites	1050–1150	n.d.	n.d.	n.d.
Andesites	960–1050	1–3	~10^{-10}	<1.5
Rhyolites	800–980	1–2	10^{-10}–10^{-12}	3–4

for P_2O_5 (Fig. 4a), and at 68 wt.% SiO_2 for Na_2O, limit the SiO_2 range for any mixing. If mixing across a large compositional range was the dominant differentiation mechanism, distinct inflections in major element trends would not be observed. The samples with relatively low P_2O_5 abundances at intermediate compositions may be an exception (Fig. 4a), and their petrogenesis may be interpreted to reflect relatively uncommon mixing processes of less evolved (<~58 wt.% SiO_2) with much more felsic (>~65 wt.% SiO_2) material.

Assimilation of upper crustal host rocks is another complicating factor in the geochemical evolution of magma compositions at arcs. In continental arcs, crustal assimilation will result in increasing crustal isotopic signatures with increasing degrees of differentiation. There is, for example, evidence for crustal assimilation in the more evolved samples from Santorini, where $^{87}Sr/^{86}Sr$ ratios increase with increasing SiO_2 due to assimilation of old crust that has a radiogenic Sr signature (Fig. 4b). However, in oceanic

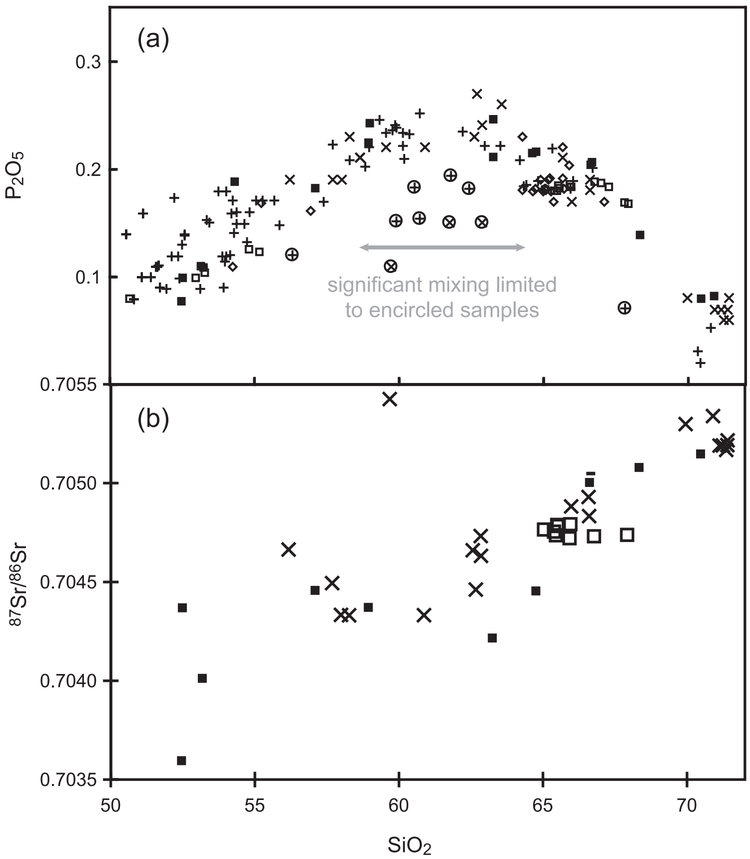

Fig. 4. (a) Symbols as in Fig. 3. In Santorini, P_2O_5 versus SiO_2 shows a prominent inflection between the andesites and dacites. This suggests that while magma mixing may occur over short SiO_2 ranges, mixing between mafic and felsic magmas is limited. The encircled samples may be an exception. (b) $^{87}Sr/^{86}Sr$ isotopes increase steadily towards higher SiO_2 content in the more evolved samples. This suggests that the petrogenesis of these samples involves assimilation of radiogenic crust.

arcs and in magmatic systems where the crustal host rock is dominated by previous magmatic intrusions with an isotopic signature similar to the evolving magma, assimilation may be cryptic to long-lived isotopes and may not be evident in whole-rock geochemical trends (Reagan et al., 2003; Zellmer et al., 2003a).

In summary, the geochemical evolution from basalts to basaltic andesites may be dominated by closed system fractional crystallization. Although closed-system differentiation to intermediate and felsic compositions has also been reported in some areas, petrogenesis of the more evolved compositions often seems to involve open system processes. These include magma mixing and assimilation of crustal rocks, either as bulk rocks or as partial melts. At Santorini, magma mixing is generally restricted to limited SiO_2 ranges, as evidenced by distinct inflections in the differentiation trends. However, $^{87}Sr/^{86}Sr$ systematics indicate crustal assimilation, particularly at >60 wt.% SiO_2.

4. Insights to differentiation and assimilation from thermal modeling

Using U-series isotope data, it was argued above that magmatic evolution to andesites and more evolved compositions often requires open system processes, with the assimilated component close to ^{238}U–^{230}Th secular equilibrium. However, MELTS modeling successfully reproduced evolutionary trends of the mafic samples at Santorini. Using thermal modeling, we explore in this section how deep crustal hot zones near the base of the lower arc crust can explain the evolution of mafic magmas and the origin of secular equilibrium in more differentiated compositions. In addition, the potential for assimilation of radiogenic old crust will be assessed.

The successive injection of basalt sills at the base of the lower crust can be simulated numerically (Annen and Sparks, 2002). Heat transfer from basalt into the surrounding rocks is computed using finite differences. A thermal anomaly in the intrusion area builds up with time. Eventually melts start to accumulate between injection events by incomplete crystallization of the injected basalt and possibly by partial melting of the crust. The simulation was run with basalts containing 0.3% H_2O and injected at 1300 °C, and basalts containing 2% H_2O and injected at 1100 °C in order to bracket the range of common basalt water content and corresponding liquidus temperature from "hot spot-type" to "arc-type" basalts. Melt fraction curves for basalt and crustal rocks were calibrated using published data (Mysen, 1981; Green, 1982; Wyllie, 1983; Clemens and Vielzeuf, 1987; Foden and Green, 1992).

The model was also tested with a fertile (amphibolite) and a refractory (mafic granulite) lower crust. In the case of an amphibolitic lower crust, the generation of melt from the crust involves dehydration melting. Flux melting was not considered. The solidus temper-

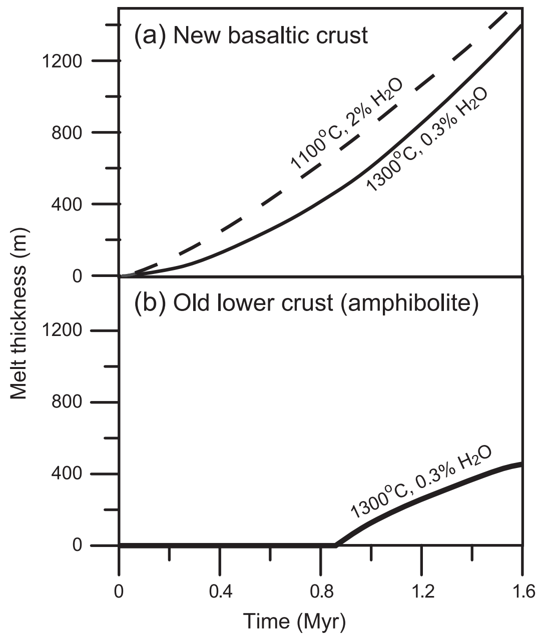

Fig. 5. Thermal model of periodic dike injection of basaltic magma at the Moho, adapted from Annen and Sparks (2002). Melt thickness within (a) the new basaltic crust composed of freshly intruded basalt and (b) the old amphibolitic lower crust, are plotted as a function of time. More melt is generated in the new basaltic crust because its solidus (wet granite solidus) is lower than the amphibolite solidus (amphibole breakdown temperature). If the old crust is granulitic instead of amphibolitic it does not partially melt because of its high solidus temperature. Note that if the basalt temperature is 1100 °C, the amphibolitic crust does not melt either. The rate of melt production is larger within the 1100 °C/2% H_2O basalt than within the 1300 °C/0.3% H_2O basalt because of the steeper slope of the melt fraction curve. For more details on modeled solidus temperatures and melt fraction curves, see Annen and Sparks (2002).

ature of fertile crust corresponds to the breakdown temperature of amphibole. The solidus temperature of the quenched melts corresponds to the wet granite solidus, because water concentrates in the evolved melts that are the residual from differentiation of initially basaltic intrusions. The rates and degrees of melting and crystallization strongly depend on the basalt intrusion rate (Annen and Sparks, 2002). Here, an intrusion rate of 2×10^{-3} km^3 $year^{-1}$ is chosen, representative of magmatic arcs based on volcanic output rates and assumptions on intrusive to extrusive ratio (Crisp, 1984; Shaw, 1985).

The main results of this model are presented in Fig. 5. Because the solidus temperature of the quenched melts (about 650 °C at 30 km depth) is much lower than the lower crust solidus temperature (about 1020 °C at 30 km for an amphibolite), melt starts to accumulate in the freshly intruded basalt layer. The magma volume in the deep crustal hot zone of newly intruded basaltic crust increases steadily, as shown for two magma temperatures and melt H_2O contents in Fig. 5a. Thus, the first melts that form are small degree residual liquids of the intruded basalts, and these residual liquids represent the most evolved compositions in this system. However, significant volumes of evolved melts only accumulate after an intrusive period of the order of >100 ka, and any initial U-excesses of the early intruded basalts, from which these more evolved compositions are produced, have therefore been reduced significantly by decay.

It is apparent from Fig. 5b that melting of old lower crust only occurs after a long period of time (~1 Myr), and only if it is hydrous (amphibolite) and the intruded basalts are hot (1300 °C). Thus, assimilation of significant amounts of old crust during differentiation from basalts to andesites may be rare.

However, assimilation of old crust is evident in more evolved compositions from isotopic constraints, e.g. from increasing $^{87}Sr/^{86}Sr$ ratios with increasing zSiO_2 at Santorini as shown in Fig. 4b. Geobarometric constraints suggest that magmatic differentiation to

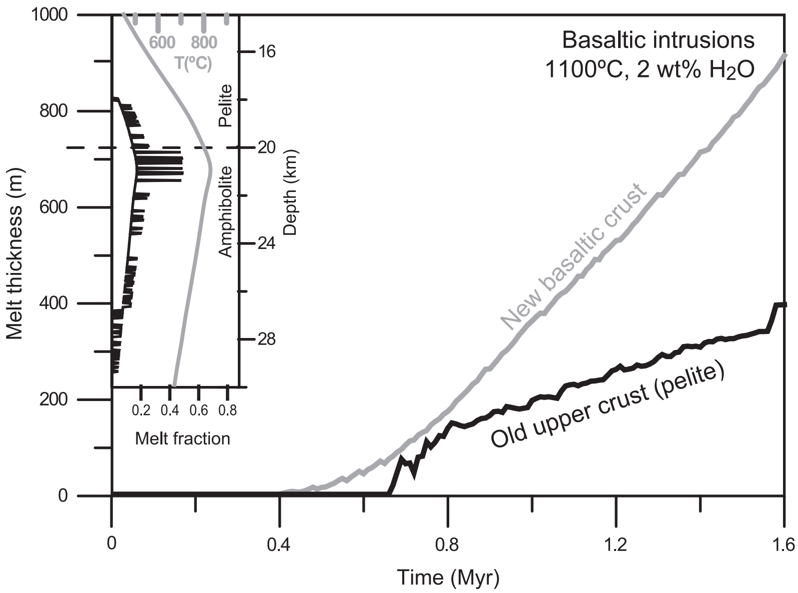

Fig. 6. Thermal model of periodic dike injection of basaltic magma at random depths close to the lower crust–upper crust boundary, adapted from Annen and Sparks (2002). Melt thickness within the new basaltic crust and the old pelitic upper crust are plotted as a function of time. The rate of melt production within the new basaltic crust is lower than in Fig. 5a because of the colder initial temperature of the surrounding crust at mid-crustal depth. The inset shows melt fraction and temperature distribution as a function of depth after the injection of 160 sills, 50 m thick, over 1.6 Myr for this particular model run. High melting degree peaks in the melt fraction curves correspond to partially melted screens of pelitic crust sandwiched by basaltic sills, which themselves contain a low melt fraction of residual melts. The lower crust (Amphibolite)–upper crust (Pelite) boundary is initially set at 20 km depth. Basaltic sills were randomly injected between 18 and 22 km. Portions of both basaltic sills and pelitic screens are displaced downward with time as a result of new sill intrusions.

more evolved compositions occurs at intermediate or upper crustal levels. The process of magmatic intrusion of low temperature, hydrous mafic magmas into intermediate crustal levels can also be modeled thermally. The upper crust was modeled either as a pelite (fertile case) or as a granodiorite (unfertile case). In the case of a pelitic crust the solidus corresponds to the breakdown temperature of muscovite, i.e. about 700 °C at 20 km depth. After an incubation period, melt starts to accumulate by incomplete crystallization of newly injected basalt and eventually by partial melting of the upper crust. It is found that upper crustal lithologies are likely to be assimilated after <1 Myr incubation time (Fig. 6). Previous intrusions will be remelted due to the heat supplied by new intrusions, which cease to freeze after about 500 kyr of sustained activity. Finally, it is also apparent from Fig. 6 that old upper crustal melts become increasingly voluminous as the magmatic system is maturing. This result will be discussed in a later section.

5. Constraints on magmatic processes from dating of minerals

To gain further insights into the rates of differentiation from andesites to more evolved compositions at subduction zones, crystals as a record of magmatic evolution provide another route to investigate the dynamics of magmatic systems. Many attempts have recently been made to constrain crystallization ages and crystal residence times using U-series mineral isochron studies, CSD studies, trace element diffusion within and between crystals, and in situ U–Th and U–Pb dating of accessory phases. In the following, an overview of U-series mineral isochron ages from arc magmas across the entire compositional range will be given. Then, some examples will be presented to demonstrate how a variety of geochronological tools can be used to gain insights into the petrogenetic processes in the mafic, intermediate and felsic magmatic regimes.

5.1. Insights from U-series mineral isochron dating

U–Th dating of mineral separates has been used for decades to constrain the timing of crystallization in volcanic rocks (for a review, see Condomines et al., 2003). This method relies on the fact that different minerals display different relative mineral-melt partition coefficients of U and Th. Thus closed system crystallization starts with a range of U/Th but constant (^{230}Th/^{232}Th) ratios in the mineral phases, and therefore variable amounts of ^{238}U–^{230}Th disequilibrium at the time of crystallization. Subsequent radioactive decay towards secular equilibrium modifies the (^{230}Th/^{232}Th) ratio by ingrowth/decay of ^{230}Th, which has a half-life of ~76 kyr. Similarly, crystallization also leads to fractionation of Ra from Th, followed by subsequent ingrowth/decay of ^{226}Ra, which has a half-life of ~1.6 kyr. For a detailed account of the U-series dating methodology, the reader is referred to a recent review by Condomines et al. (2003). Here it will be sufficient to state that crystallization ages of mineral assemblages can be calculated given the assumptions that (a) crystallization occurs in a closed system, (b) all phases crystallized at the same time, and (c) crystallization was rapid compared to the half-life of the system.

Fig. 7 summarizes the available initial U–Th–Ra crystallization ages in arcs, i.e. corrected for post-eruptive radioactive decay. Early data from α-counting (Hemond and Condomines, 1985; Gill and Williams, 1990) was excluded from this compilation, because the uncertainties in (^{230}Th/^{232}Th) ratios were too large to obtain well-constrained ages. In order to ensure consistency and facilitate comparison, the ages and associated 2σ errors have been recalculated using Ludwig's (2001) Isoplot Model 1 fits through the published mass-spectrometric U-series mineral data, rather than just compiling the calculated ages from the literature. U–Th mineral ages are rarely constrained to better than $\pm$15 ka at the 95% confidence limit, because U–Th fractionation between major mineral phases is generally small and there is often considerable scatter in the mineral data, indicating that one or more of the above assumptions may not always be appropriate. Nevertheless, the following points are apparent:

(1) Samples with ^{238}U–^{230}Th mineral ages significantly greater than 10 ka often preserve ^{226}Ra–^{230}Th disequilibria even though the latter system should return to secular equilibrium within 8 ka (Fig. 7b). This is evident in all analyzed mafic samples that typically yield U–Th mineral ages between 10 and 75

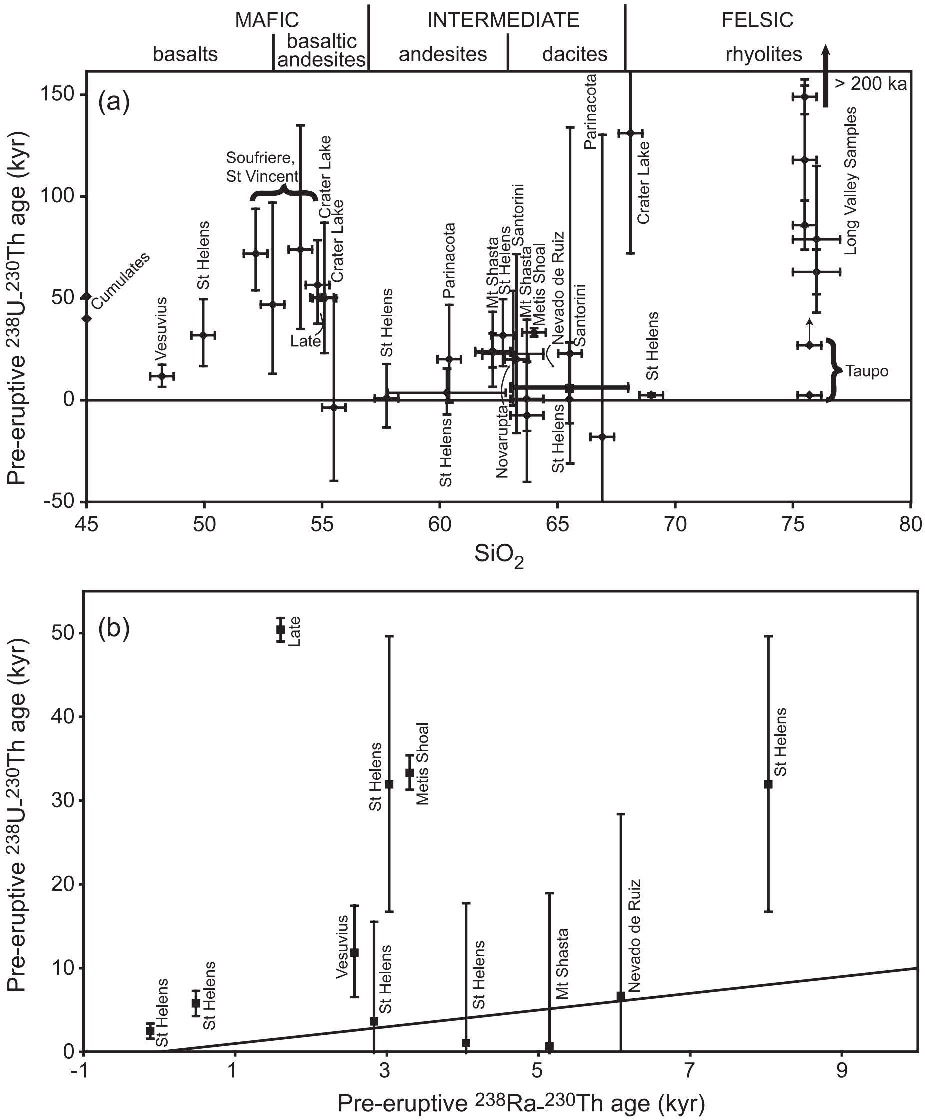

Fig. 7. Initial U-series mineral ages are recalculated from the published data: Mount St. Helens (Volpe and Hammond, 1991), Mt. Shasta (Volpe, 1992), Nevado de Ruiz (Schaefer et al., 1993), Long Valley (Reid et al., 1997; Reid and Coath, 2000; Heumann et al., 2002), Vesuvius and the Vesuvius cumulate (Black et al., 1998), Soufrière on St. Vincent (Heath et al., 1998b), Parinacota (Bourdon et al., 2000), Taupo (Charlier and Zellmer, 2000), Crater Lake and Novarupta (Reagan et al., 2003), Late, Metis Shoal, and the Soufrière cumulate (Turner et al., 2003c), and Santorini (Zellmer et al., 2000). (a) Ages are plotted versus SiO_2 of the whole rock to compare pre-eruptive mineral ages in mafic, intermediate and felsic compositions. Silica content of the cumulates is not known, but have been set to 45 wt.% here to allow plotting. (b) ^{238}U–^{230}Th ages plotted against available ^{226}Ra–^{230}Th ages. Uncertainties of Ra–Th ages have not been plotted. The equal age line is given. See text for discussion.

ka, but also true for some of the more evolved compositions, e.g. some Mount St. Helens samples and Metis Shoal.

(2) In contrast to the relatively old initial U–Th mineral ages obtained from mafic samples, the majority of those obtained from intermediate magma compositions are within error of eruption age (Fig. 7a). This seems to indicate that the proportion of old crystals often decreases towards the more porphyritic evolved compositions, despite the generally more

complicated morphologies observed in the crystals of such samples.

(3) Felsic magmas have crystallization ages ranging from >200 ka to a few ka prior to eruption (Fig. 7a). It is important to note that some of these ages reflect crystallization of accessory phases (mainly zircon), which in contrast to more common minerals is strongly dependent on the degree of trace element saturation. Hence, different crystallization conditions are dated by analyzing accessory phases.

Using examples from selected volcanic centers in the mafic, intermediate and felsic regimes, these observations will be discussed in terms of petrogenetic processes, with supporting evidence from a number of additional geochronological tools.

5.2. Mafic compositions—an example from Soufrière, St. Vincent

Apparently disparate U–Th and Ra–Th ages are frequently observed, most notably in mafic arc lavas that have old pre-eruptive U–Th mineral ages (cf. Fig. 7a). This indicates that the assumption of rapid growth of the entire crystal population is violated. Crystal size distribution studies can be used to identify and quantify processes such as crystal nucleation, growth and dissolution, and mixing of crystal populations. The crystal size distribution of the 1979 Upper Rabacca basalt from Soufrière, St. Vincent, is given as an example in Fig. 8. It has a markedly concave-up shape, and crystals >2 mm in size, which make up 23% of the population, are larger than expected from the size distribution of the smaller crystals. Such data can be interpreted by overgrowth of young rims onto older crystals prior to eruption (Turner et al., 2003c), or by a mixture of young and old crystals in the analyzed separates (Cooper and Reid, 2003). In the absence of microanalytical data on individual crystals it is not possible to decide between these scenarios, but they need not be mutually exclusive.

The abundance of old crystal cores in mafic magmas can be understood in the framework of deep crustal hot zones, where older, partially crystallized melts coexist with young melt of the last basaltic injection that is currently crystallizing. Crystals formed from mixtures of these magmas will display a large range in ages, and the largest crystals

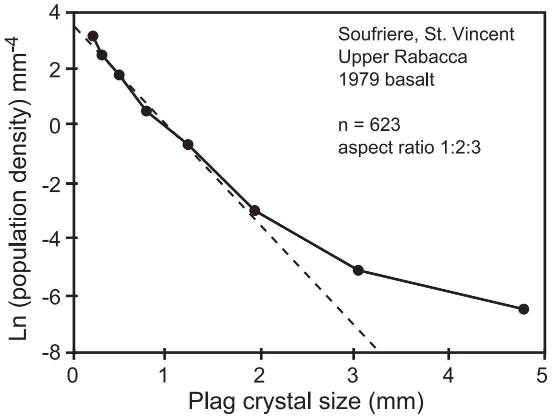

Fig. 8. Crystal size distribution plot of the 1979 basalt lava erupted at Upper Rabacca, Soufrière, St. Vincent, adapted from Turner et al. (2003c), with *n* as the number of crystals measured. Stereological conversions from 2D to 3D measurements were determined following the methods of Higgins (2000).

will have the greatest proportion of old, earlier crystallized cores. Alternatively, mixtures of young and old crystals may be generated if crystals formed earlier are removed from low viscosity melts and stored in cumulates, to be entrained into younger magmas at a later stage. Rapid crystal settling has previously been suggested by Hawkesworth et al. (2000) to explain the absence of old crystal ages in very low viscosity mafic magmas from different tectonic settings. During differentiation the viscosity of magmas increases, allowing re-entrainment of old cumulate crystals prior to eruption. Two other lines of evidence point to entrainment of cumulate crystals. Firstly, although the uncertainties are significant, two analyzed pre-eruptive U–Th cumulate ages of 40 (+91/−49) ka and 51 (+14/−13) ka, 2σ (Black et al., 1998; Turner et al., 2003c) are comparable to the pre-eruptive U–Th mineral ages obtained from mafic samples. Secondly, intra-crystalline trace element disequilibria are preserved in some plagioclase phenocrysts from the Upper Rabacca eruption (Zellmer et al., 1999), indicating that crystals were stored at comparatively low temperatures, pertaining to cumulate mush zones.

5.3. Intermediate compositions—an example from the Soufrière Hills volcano, Montserrat

Despite the great diversity of crystals in silicic andesites and dacites, most of the available initial U–

Th mineral ages obtained from intermediate magma compositions are within error of eruption age (Fig. 7a). This may indicate that the proportion of entrained old crystals is lower in these more evolved magmas that often have a greater overall crystallinity, and that the complicated crystal morphologies observed in these rocks are produced over a comparatively short timescale. A different dating method, capable of resolving shorter timescales, is required to obtain more detailed information about the pre-eruptive crystal growth processes of such samples.

Intra-crystalline trace element diffusion is a powerful technique to elucidate crystal residence time, if temperature can be constrained independently. Here, we use the currently active Soufrière Hills volcano on Montserrat in the Lesser Antilles as an example for a typical intermediate arc volcano. Its eruption products are dominantly andesites, that carry an abundance of mafic inclusions, interpreted by Murphy et al. (1998; 2000) and others to represent less evolved magma that remobilized previously intruded andesitic material. The dominant crystal phase within the andesites is plagioclase feldspar, which contains multiple resorption textures, indicating complex growth histories. Zellmer et al. (2003b) have used Sr diffusion systematics within plagioclase phenocrysts to elucidate the timescales of crystal growth in this system. The basis of this dating method is that the initial distribution of Sr between different growth zones within an individual crystal is different from its equilibrium distribution (Fig. 9). As a batch of H_2O saturated magma is brought up to higher levels within the crust, decompression will lead to degassing induced crystallization of more and more sodic plagioclase, which can be modeled using the calibrations of Yoder et al. (1957) and Housh and Luhr (1991). Sr partitions preferentially into the growing crystals (D_{Sr}>1), leading to decrease of Sr concentration within the liquid. However, as D_{Sr} steadily increases with decreasing X_{An}, the decrease in the liquid is compensated by an increase in the proportion of Sr taken up into the growing crystal, such that the Sr concentration within the crystal remains approximately constant. Thus, in contrast to the equilibrium distribution of Sr, which favors more sodic plagioclase, the initial distribution of Sr across the crystal is one of relatively constant concentration, independent of the anorthite content of the crystal (Fig. 9). Zellmer et al. (2003b) modeled the

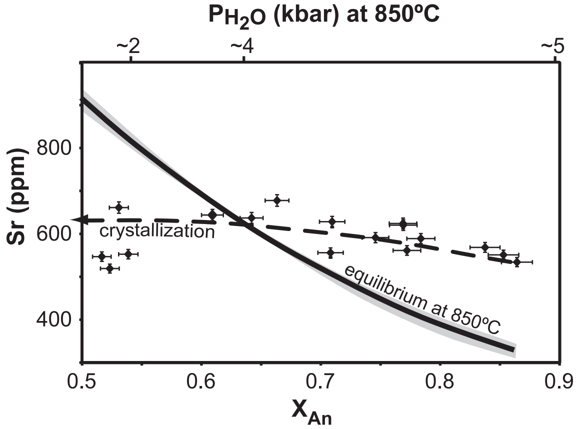

Fig. 9. Sr concentration versus anorthite content in a plagioclase crystal from the Gages andesitic lava dome, Soufrière Hills volcano, Montserrat. Compositions crystallised under H_2O-saturated conditions during isothermal degassing in response to decompression at 850 °C (dashed line) are compared with compositions predicted by bulk traverse equilibrium partitioning (solid line).The shaded area is the $\pm 2\sigma$ uncertainty range in the bulk traverse equilibrium partitioning. Initially flat Sr profiles are consistent with closed system crystallization of plagioclase. Variations in Sr concentrations are likely to be due to subsequent diffusion, initially towards local equilibrium partitioning, and ultimately towards bulk traverse equilibrium partitioning (solid line). The diffusive evolution of the Sr concentrations is complex because of the interplay between diffusion and partitioning as a function of local anorthite content. Adapted from Zellmer et al. (2003b).

subsequent diffusion of Sr towards equilibrium for a number of plagioclase crystals and obtained variable temperature-time histories. Within individual rock samples crystal ages varied by more than an order of magnitude, with an overall variation between ~10 and ~1000 years at a magmatic temperature of 850 °C, typical for Montserrat.

These data were interpreted to indicate repeated crystallization of individual andesitic magma batches in the upper crust, each producing its own crystals with individual temperature-time histories. Remobilization of these individual magma batches by hotter, less evolved magma, which is now represented by the mafic inclusions, resulted in amalgamation of these individual andesite bodies and juxtaposition of crystals with different growth histories immediately prior to eruption. In conclusion, petrogenesis at Montserrat involves multi-stage crystallization and remobilization processes. Young crystal ages argue for rapid crystallization and—in the case of diffusion dating—rapid cooling in ephemeral magma chambers. Such conditions are met in systems where crystallization is due to decompression induced degassing, and where individual magma batches are small. In addition, it should be noted that the minerals formed during rapid crystallization at shallow depth post-date the deeper differentiation processes that produced the andesitic melt compositions at Montserrat.

5.4. Felsic compositions—an example from the Taupo Volcanic Zone, New Zealand

In order to understand the large range in pre-eruptive crystallization ages in felsic compositions, we again focus on an individual volcanic centre. Taupo, in the Taupo volcanic zone (TVZ), New Zealand, has erupted rhyolitic compositions for many tens of kyrs (Fig. 10a). There is evidence for multi-stage zircon crystal growth of the 26.5 ka Oruanui eruption (Charlier and Zellmer, 2000), and Charlier et al. (in review) have recently studied in more detail the distribution of $^{238}U/^{230}Th$ zircon ages from the Oruanui rhyolite and Oruanui-type magmas from the earlier Tihoi and Okaia eruptions. Zircon age distributions were obtained from these rhyolites using an ion microprobe, and are given in Fig. 10b. It is immediately apparent that each of these eruptions has two peaks in zircon crystallization ages, a common older peak at ~100 ka before present and a younger peak representing crystallization immediately prior to eruption. Charlier et al. (in review) conclude that these rhyolites were generated from the same source region which represents a previous magmatic intrusion that crystallized zircon ~100 kyr ago. The absence of the Tihoi pre-eruptive zircon ages in the Okaia and Oruanui rhyolites (Fig. 10b) indicate that these eruptive products were not erupted from the same magma chamber as the Tihoi rhyolites. Instead, each magma batch was remobilized from the same igneous source just prior to eruption. Thus, magma remobilization and multi-stage ascent histories (cf. Cottrell et al., 1999) may be very common for both intermediate and felsic magmas.

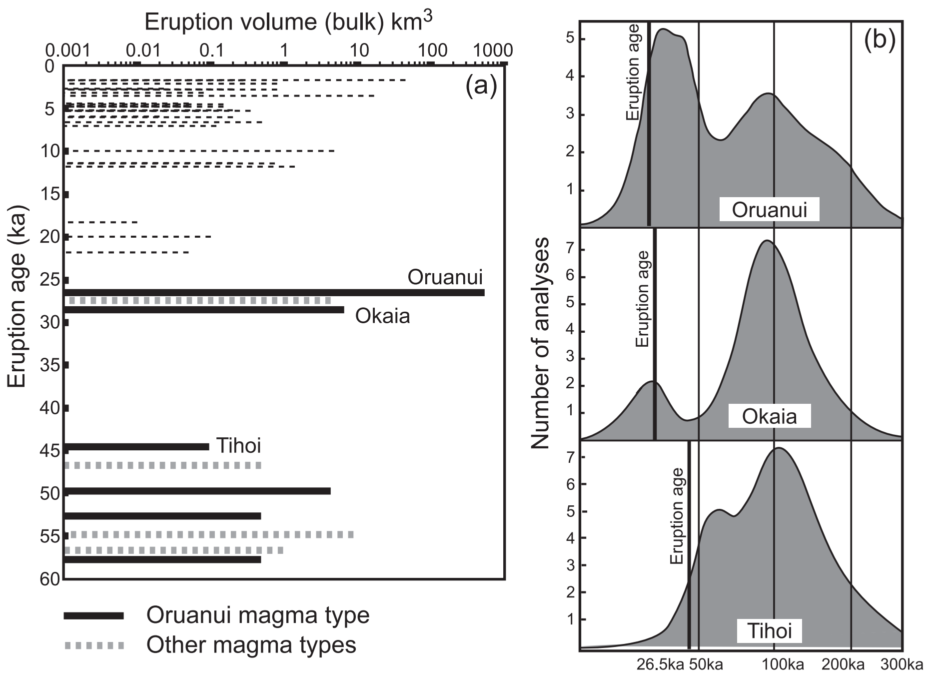

Fig. 10. (a) Plot of stratigraphy versus log scale eruption volume for all post 60 ka eruptives from Taupo volcano. (b) Comparison of probability density functions from Tihoi, Okaia and Oruanui SIMS U–Th zircon model age histograms. Note the close coincidence of the ~100 ka peak in all of the analyzed samples, and the lack of young Tihoi-aged crystals in the Okaia sample. A log-scale was used to facilitate the calculation of cumulative probability curves from data with asymmetrical age uncertainties.

Finally, it is apparent from thermal modeling (Fig. 6) that old upper crustal melts become increasingly voluminous as the magmatic system is maturing, and that high silica rhyolite petrogenesis therefore probably involves a significant proportion of crustal melting. Charlier et al. (in review) found a number of Proterozoic zircons within some of the eruptive products of the TVZ, suggesting that not only assimilation of previously intruded material, but also of old country rock is occurring in this system.

6. Future work

This review of geochronological data has revealed a wide range of magma residence times in arc crust (days, e.g. Tolbachik, to ~100 kyr, e.g. for generation of evolved melts in lower crustal hot zones), and an similarly wide range of pre-eruptive crystal residence times (years to ≥100 kyr). In Fig. 11, magma eruption rates are plotted against pre-eruption ages for a number of volcanoes from a variety of tectonic settings. It is evident that young pre-eruption ages are dominantly found in mafic magmas, whereas large volume rhyolites often yield older ages. The diagram is contoured for magma volumes, representing that part of the subvolcanic magma reservoir that eventually becomes erupted. Strikingly, magma volumes vary by more than three orders of magnitude between different volcanic centers. Future work should aim to elucidate the factors influencing the volume of magma stored in subvolcanic crustal reservoirs, as a better

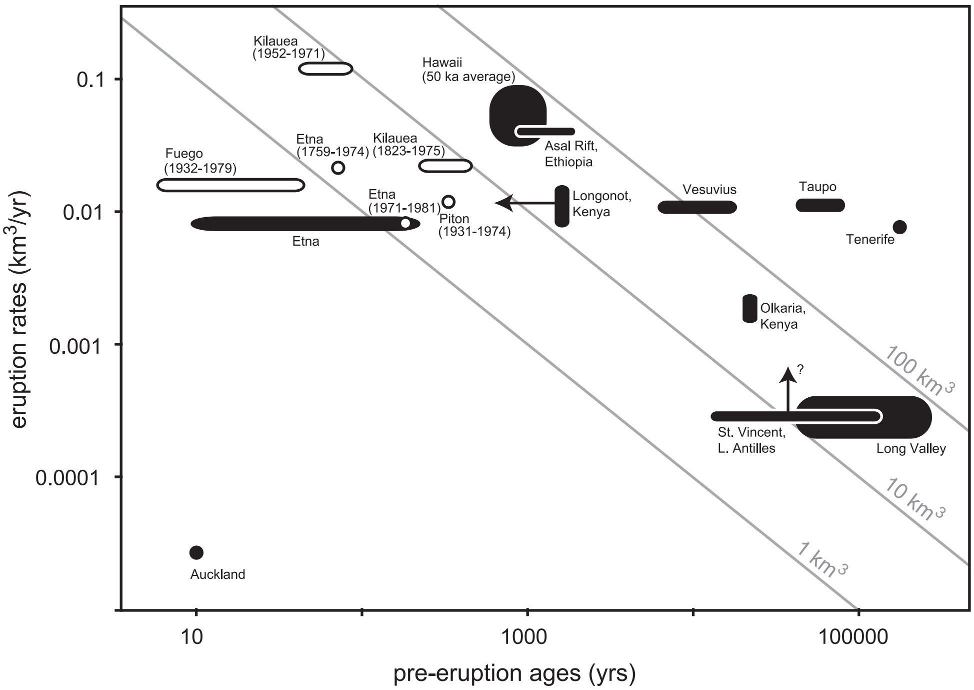

Fig. 11. A plot of pre-eruption ages against eruption rates of volcanoes from a variety of tectonic settings, adapted from Hawkesworth et al. (2004), with uncertainties where data was available. Eruption rates are integrated over the time periods given by Crisp (1984), unless indicated otherwise. Pre-eruption ages are constrained from estimates of magma output rate and magma chamber volume (open symbols, Pyle, 1992), or derived from geochronological studies of minerals and whole-rocks (black symbols, Condomines et al., 1995; Reid et al., 1997; Black et al., 1998; Heath et al., 1998b; Hawkesworth et al., 2000; Reid and Coath, 2000; Cooper et al., 2001; Heumann and Davies, 2002; Heumann et al., 2002, and this study). Ages for Longonot, Etna, and all open symbols apart from Fuego are maximum values. Eruption rates are from Crisp (1984) and Pyle (1992). Longonot and Olkaria eruption rates are those given as "Kenya (phonolites only)" and "Kenya (silicic only)", respectively, and thus are maximum values. Eruption rates of St. Vincent assumed to be one order of magnitude lower than those of entire Lesser Antilles arc. Note that the contoured crustal volume of magma that eventually becomes erupted varies by more than three orders of magnitude between individual volcanic centres.

understanding of these will be a key to improved long-term hazard mitigation strategies at sites of active and dormant volcanism. Parameters to be considered include magma supply rate; thickness, composition and thermal structure of the crust; and magma composition and volatile content, which vary between different tectonic settings. Integration of these variables into petrogenetic models will allow prediction of the long-term evolution of areas of active arc magmatism. Unfortunately, there are currently too few reliable data to assess the relative importance of these variables systematically. In future, a combination of many of the above dating techniques, with an even greater focus on crystal morphology and in-situ approaches, will therefore be necessary to obtain more accurate geochonological information.

7. Conclusions

(1) Large ^{226}Ra excesses in a number of mafic arc magmas indicate that mafic melts can get transferred from source to surface rapidly, within less than ~1 kyr. In some cases, geophysical evidence suggests that magma transfer is even faster, occurring within days.

(2) The evolution of ^{226}Ra–^{230}Th disequilibria in co-magmatic suites of individual arc volcanoes suggest that magma differentiation can occur on timescales of a few thousand years only. There is evidence for both closed system and open system differentiation.

(3) The generally lower maximum (^{238}U/^{230}Th) ratios in intermediate rocks compared with the large ratios that are observed in some basalts suggests that rapid closed system differentiation is not a generic mode for the production of more evolved compositions. Instead, many case studies have shown a role for open system processes which, whilst not ubiquitous, certainly can assist in the evolution from basalt to more silicic compositions. In some instances, decreases in (^{238}U/^{230}Th) ratios may reflect the time taken for magmatic evolution. However, where the preservation of ^{226}Ra excesses appear to rule this out, the most likely explanation may be assimilation of material close to U–Th equilibrium. In such cases, the decrease in (^{238}U/^{230}Th) results from mixing and has no simple time significance other than to indicate that evolution took less than 350 kyr.

(4) MELTS modeling of magma evolution at Santorini in the Aegean arc provides further evidence that closed system fractional crystallization cannot produce the more evolved compositions at this volcano, and there is isotopic evidence for assimilation of old crust.

(5) Thermal modeling of melt production in deep crustal hot zones provides an explanation for insignificant ^{238}U–^{230}Th disequilibria of evolved magmatic assimilants: significant volumes of evolved melts accumulate only after >100 kyr of intrusive history, increasing the melt fraction of the basaltic magma body with every new injection. This leaves significant time for radioactive decay of potential initial disequilibria. Thermal modeling also provides a framework for understanding assimilation of old upper crustal rocks in more mature magmatic systems.

(6) U–Th mineral isochrons in analyzed mafic arc magmas yield pre-eruptive ages of 43 ± 26 ka (1 std, n=9), significantly older than ages obtained from Ra–Th mineral isochrons. A CSD case study from Soufrière, St. Vincent, in combination with U-series data from cumulates, indicates that this may result from the abundance of cumulate xenocrysts within more mafic arc magmas.

(7) Most analyzed U–Th mineral isochrons from intermediate arc magmas yield ages within error of eruption age, indicating that other dating techniques are more suitable for these compositions. A case study of remobilized andesites from the Soufrière Hills volcano, Montserrat, has employed intra-crystalline trace element diffusion to show that crystallization timescales are rapid (of the order of 10–1000 years), and that the temperature-time history of individual crystals is extremely complex.

(8) U–Th mineral isochrons in felsic arc magmas yield very variable ages of >200 ka to shortly prior to eruption. A case study from the Taupo Volcanic Zone has shown that both remobilization of previous intrusives and melting of old country rock are occurring, giving rise to a this range in ages. Both intermediate and felsic magmas therefore can be shown to frequently have multi-stage ascent histories.

Acknowledgements

We thank Steve Sparks, Louise Thomas, and Steve Blake for useful discussions. The comments of James Jackson, Iris Zöllner and Hans-Joachim Mönicke are gratefully acknowledged. GFZ was funded by a Lamont-Doherty Research Fellowship and acknowledges additional financial support by Dave Walker and Steve Goldstein. CA was funded by a Swiss National Science Foundation Fellowship. This contribution benefitted greatly from the constructive reviews of Jim Gill, Mark Reagan and an anonymous reviewer.

References

Annen, C., Sparks, R.S.J., 2002. Effects of repetitive emplacement of basaltic intrusions on thermal evolution and melt generation in the crust. Earth and Planetary Science Letters 203, 937–955.

Barton, M., Huijsmans, J.P.P., 1986. Post-caldera dacites from the Santorini volcanic complex, Aegean Sea, Greece: an example of the eruption of lavas of near-constant composition over a 2200 year period. Contributions to Mineralogy and Petrology 94, 472–495.

Black, S., Macdonald, R., De Vivo, B., Kilburn, C.R.J., Rolandi, F., 1998. U-series disequilibria in young (A.D. 1944) Vesuvius rocks: preliminary implications for magma residence times and volatile addition. Journal of Volcanology and Geothermal Research 82, 97–111.

Blatter, D.L., Carmichael, I.S.E., 1998. Hornblende peridotite xenoliths from central Mexico reveal the highly oxidized nature of subarc upper mantle. Geology 26, 1035–1038.

Bourdon, B., Wörner, G., Zindler, A., 2000. U-series evidence for crustal involvement and magma residence times in the petrogenesis of Parinacota volcano, Chile. Contributions to Mineralogy and Petrology 139, 458–469.

Bowen, N.L., 1928. The Evolution of the Igneous Rocks. Princeton University Press, Princeton.

Charlier, B.L.A., Zellmer, G.F., 2000. Some remarks on U–Th mineral ages from igneous rocks with prolonged crystallisation histories. Earth and Planetary Science Letters 183, 457–469.

Charlier, B.L.A., Peate, D.W., Wilson, C.J.N., Lowenstern, J.B., Storey, M., Brown, J.A., 2003. Crystallisation ages in coeval silicic magma bodies: ^{238}U–^{230}Th disequilibrium evidence from the Rotoiti and Earthquake Flat eruption deposits, Taupo Volcanic Zone, New Zealand. Earth and Planetary Science Letters 206, 441–457.

Charlier, B.L.A., Wilson, C.J.N., Lowenstern, J.B., Blake, S., van Calsteren, P.W., Davidson, J.P. Magma generation, mobilisation and cannibalism at a large, hyperactive silicic volcano (Taupo, New Zealand) revealed by U/Th and U/Pb systematics in zircon. Journal of Petrology, in review.

Cheng, H., Edwards, R.L., Hoff, J., Gallup, C.D., Richards, D.A., Asmerom, Y., 2000. The half-lives of uranium-234 and thorium-230. Chemical Geology 169, 17–33.

Clemens, J.D., Vielzeuf, D., 1987. Constraints on melting and magma production in the crust. Earth and Planetary Science Letters 86, 287–306.

Condomines, M., Tanguy, J.C., Michaud, V., 1995. Magma dynamics at Mt Etna: constraints from U–Th–Ra–Pb radioactive disequilibria and Sr isotopes in historical lavas. Earth and Planetary Science Letters 132, 25–41.

Condomines, M., Gauthier, P.-J., Sigmarsson, O., 2003. Timescales of magma chamber processes and dating of young volcanic rocks. In: Bourdon, B., Henderson, G.M., Lundstrom, C.C., Turner, S.P. (Eds.), Uranium-Series Geochemistry, Reviews in Mineralogy and Geochemistry. The Mineralogical Society of America, Washington, pp. 125–174.

Cooper, K.M., Reid, M.R., 2003. Re-examination of crystal ages in recent Mount St. Helens lavas: implications for magma reservoir processes. Earth and Planetary Science Letters 213, 149–167.

Cooper, K.M., Reid, M.R., Murrell, M.T., Clague, D.A., 2001. Crystal and magma residence at Kilauea volcano, Hawaii: ^{230}Th–^{226}Ra dating of the 1955 East Rift Eruption. Earth and Planetary Science Letters 184, 703–718.

Costa, F., Chakraborty, S., Dohmen, R., 2003. Diffusion coupling between trace and major elements and a model for calculation of magma residence times using plagioclase. Geochimica et Cosmochimica Acta 67, 2189–2200.

Cottrell, E., Gardner, J.E., Rutherford, M.J., 1999. Petrologic and experimental evidence for the movement and heating of the pre-eruptive Minoan rhyodacite (Santorini, Greece). Contributions to Mineralogy and Petrology 135, 315–331.

Crisp, J.A., 1984. Rates of magma emplacement and volcanic output. Journal of Volcanology and Geothermal Research 20, 177–211.

Druitt, T., Lanphere, M., Vougioukalakis, G., 1996. Field workshop Santorini, 8.-15.9.1996, Guide and Excursions. IAVCEI, Commission on Explosive Volcanismt.

Edwards, L., 1994. Magma cyclicity and isotopic variation on Santorini volcano, Aegean Sea, Greece. PhD thesis, University of Bristol, Bristol, 372 pp.

Elliott, T., Plank, T., Zindler, A., White, W., Bourdon, B., 1997. Element transport from slab to volcanic front at the Mariana arc. Journal of Geophysical Research 102 (B7), 14991–15019.

Engdahl, E.R., van der Hilst, R., Buland, R., 1998. Global teleseismic earthquake relocation with improved travel times and procedures for depth determination. Bulletin of the Seismological Society of America 88, 722–743.

Fedorov, S.A., Markhinin, Y.K. (Eds.), The Great Tolbachik Fissure Eruption. Cambridge University Press, Cambridge, England, 341 pp.

Foden, J.D., Green, D.H., 1992. Possible role of amphibole in the origin of andesite: some experimental and natural evidence. Contributions to Mineralogy and Petrology 109, 479–493.

Fujinawa, A., 1988. Tholeiitic and calc-alkaline magma series at Adatara volcano, northeast Japan: 1. Geochemical constraints on their origin. Lithos 22, 135–158.

Fujinawa, A., 1990. Tholeiitic and calc-alkaline magma series at Adatara volcano, northeast Japan: 2. Mineralogy and phase relations. Lithos 24, 217–236.

George, R., Turner, S., Hawkesworth, C., Morris, J., Nye, C., Ryan, J., Zeng, S.-H., 2003a. Melting processes and fluid and sediment transport rates along the Alaska-Aleutian arc from an integrated U–Th–Ra–Be isotope study. Journal of Geophysical Research 100 (B5), 2252 (doi:10.1029/2002JB001916).

George, R.M.M., Turner, S.P., Hawkesworth, C.J., Bacon, C.R., Nye, C., Stelling, P., Dreher, S., 2003b. Chemical versus temporal controls on the evolution of tholeiitic and calc-alkaline magmas at two volcanoes in the Alaska-Aleutian arc. Journal of Petrology 45, 203–219.

Gerlach, D.C., Grove, T.L., 1982. Petrology of Medicine Lake Highland volcanics: characterisation of endmembers of magma mixing. Contributions to Mineralogy and Petrology 80, 147–159.

Ghiorso, M.S., Sack, R.O., 1995. Chemical mass transfer in magmatic processes IV. A revised and internally consistent thermodynamic model for the interpolation and extrapolation of liquid–solid equilibria in magmatic systems at elevated temperatures and pressures. Contributions to Mineralogy and Petrology 119, 197–212.

Gill, J.B., Williams, R.W., 1990. Th isotope and U-series studies of subduction-related volcanic rocks. Geochimica et Cosmochimica Acta 54, 1427–1442.

Gill, J.B., Morris, J.D., Johnson, R.W., 1993. Timescale for producing the geochemical signature of island arc magmas: U–Th–Po and Be–B systematics in recent Papua New Guinea lavas. Geochimica et Cosmochimica Acta 57, 4269–4283.

Green, T.H., 1982. Anatexis of mafic crust and high pressure crystallization of andesite. In: Thorpe, R.S. (Ed.), Andesites: Orogenic Andesites and Related Rocks. Wiley, New York.

Hatzfeld, D., Martin, C., 1992. Intermediate depth seismicity in the Aegean defined by teleseismic data. Earth and Planetary Science Letters 113, 267–275.

Hatzfeld, D., Besnard, M., Makropoulos, K., Voulgaris, N., Kouskouna, V., Hatzidimitriou, P., Panagiotopoulos, D., Karakaisis, G., Deschamps, A., Lyon-Caen, H., 1993. Subcrustal microearthquake seismicity and fault plane solutions beneath the Hellenic Arc. Journal of Geophysical Research 98 (B6), 9861–9870.

Hawkesworth, C.J., Turner, S.P., McDermott, F., Peate, D.W., van Calsteren, P., 1997. U–Th isotopes in arc magmas: implications for element transfer from the subducted crust. Science 276, 551–555.

Hawkesworth, C.J., Blake, S., Evans, P., Hughes, R., Macdonald, R., Thomas, L.E., Turner, S.P., Zellmer, G.F., 2000. Time scales of crystal fractionation in magma chambers—integrating physical, isotopic and geochemical perspectives. Journal of Petrology 41 (7), 991–1006.

Hawkesworth, C., George, R., Turner, S., Zellmer, G., 2004. Time scales of magmatic processes. Earth and Planetary Science Letters 218, 1–16.

Heath, E., Macdonald, R., Belkin, H., Hawkesworth, C., Sigurdsson, H., 1998a. Magmagenesis at Soufriere Volcano, St. Vincent, Lesser Antilles Arc. Journal of Petrology 39 (10), 1721–1764.

Heath, E., Turner, S., Macdonald, R., Hawkesworth, C.J., van Calsteren, P., 1998b. Long magma residence times at an island arc volcano (Soufriere, St. Vincent) in the Lesser Antilles: evidence from ^{238}U–^{230}Th isochron dating. Earth and Planetary Science Letters 160, 49–63.

Hemond, C., Condomines, M., 1985. On the reliability of the ^{230}Th–^{238}U dating method applied to young volcanic rocks—discussion. Journal of Volcanology and Geothermal Research 26, 365–369.

Heumann, A., Davies, G.R., 2002. U–Th disequilibrium and Rb–Sr age constraints on the magmatic evolution of peralkaline rhyolites from Kenya. Journal of Petrology 43, 557–577.

Heumann, A., Davies, G.R., Elliott, T., 2002. Crystallization history of rhyolites at Long Valley, California, inferred from combined U-series and Rb-Sr isotope systematics. Geochimica et Cosmochimica Acta 66, 1821–1837.

Higgins, M.D., 1996. Magma dynamics beneath Kameni volcano, Thera, Greece, as revealed by crystal size and shape measurements. Journal of Volcanology and Geothermal Research 70, 37–48.

Higgins, M.D., 1996. Crystal size distributions and other quantitative textural measurements in lavas and tuff from Egmont volcano (Mt. Taranaki), New Zealand. Bulletin of Volcanology 58, 194–204.

Higgins, M.D., 2000. Measurement of crystal size distributions. American Mineralogist 85, 1105–1116.

Housh, T.B., Luhr, J.F., 1991. Plagioclase-melt equilibria in hydrous systems. American Mineralogist 76, 477–492.

Huijsmans, J.P.P., 1985. Calc-alkaline lavas from the volcanic complex of Santorini, Aegean Sea, Greece. A petrological, geochemical and stratigraphic study. PhD, Rijksuniversiteit te Utrecht, Utrecht, 316 pp.

Kimura, J.-I., Tanji, T., Yoshida, T., Iizumi, S., 2001. Geology and geochemistry of lavas at Nekoma volcano: implications for origin of Quaternary low-K andesite in the north-eastern Honshu arc, Japan. The Island Arc 10, 116–134.

Kuno, H., 1950. Petrology of Hakone Volcano. Geological Society of America Bulletin 61, 957–1014.

Ludwig, K.R., 2001. Isoplot/Ex ver. 2.49, A Geochronological Toolkit for Microsoft Excel. Berkeley Geochronology Centre, Berkeley, California.

Makris, J., 1978. The crust and upper mantle of the Aegean region from deep seismic soundings. Tectonophysics 46, 269–284.

Marsh, B.D., 1988. Crystal size distribution (CSD) in rocks and the kinetics and dynamics of crystallisation: I. Theory. Contributions to Mineralogy and Petrology 99, 277–291.

McDermott, F., Hawkesworth, C.J., 1991. Th, Pb, and Sr isotope variations in young island arc volcanics and oceanic sediments. Earth and Planetary Science Letters 104 (1), 1–15.

Murphy, M.D., Sparks, R.S.J., Barclay, J., Carroll, M.R., Lejeune, A.-M., Brewer, T.S., Macdonald, R., Black, S., Young, S., 1998. The role of magma mixing in triggering the current eruption at the Soufriere Hills volcano, Montserrat, West Indies. Geophysical Research Letters 25 (18), 3433–3436.

Murphy, M.D., Sparks, R.S.J., Barclay, J., Carroll, M.R., Brewer, T.S., 2000. Remobilization of andesite magma by intrusion of

mafic magma at the Soufriere Hills volcano, Montserrat, West Indies. Journal of Petrology 41, 21–42.
Mysen, B.O., 1981. Melting curves of rock and viscosity of rock forming melts. In: Touloukian, Y.S., Judd, W.R., Roy, R.F. (Eds.), Physical Properties of Rocks and Minerals. Data Series on Material Properties. McGraw-Hill/CINDAS, pp. 361–407.
Nakamura, M., 1995. Continuous mixing of crystal mush and replenished magma in the ongoing Unzen eruption. Geology 23 (9), 807–810.
Papazachos, B., Kiratzi, A., Papadimitriou, E., 1991. Regional focal mechanisms for earthquakes in the Aegean area. Pure and Applied Geophysics 136 (4), 405–420.
Papazachos, B.C., Panagiotopoulos, D.G., 1993. Normal faults associated with volcanic activity and deep rupture zones in the southern Aegean volcanic arc. Tectonophysics 220, 301–308.
Pyle, D.M., 1992. The volume and residence time of magma beneath active volcanoes determined by decay-series disequilibria methods. Earth and Planetary Science Letters 112, 61–73.
Reagan, M.K., Morris, J.D., Herrstrom, E.A., Murrell, M.T., 1994. Uranium series and beryllium isotope evidence for an extended history of subduction modification of the mantle below Nicaragua. Geochimica et Cosmochimica Acta 58 (19), 4199–4212.
Reagan, M.K., Sims, K.W.W., Erich, J., Thomas, R.B., Cheng, H., Edwards, R.L., Layne, G., Ball, L., 2003. Timescales of differentiation from mafic parents to rhyolite in North American continental arcs. Journal of Petrology 44, 1703–1726.
Reid, M.R., Coath, C.D., 2000. In situ U–Pb ages of zircons from the Bishop Tuff: no evidence for long crystal residence times. Geology 28 (5), 443–446.
Reid, M.R., Coath, C.D., Harrison, T.M., McKeegan, K.D., 1997. Prolonged residence times for the youngest rhyolites associated with Long Valley Caldera: ^{230}Th–^{238}U ion microprobe dating of young zircons. Earth and Planetary Science Letters 150, 27–39.
Schaefer, S.J., Sturchio, N.C., Murrell, M.T., Williams, S.N., 1993. Internal ^{238}U-series systematics of pumice from the November 13, 1985, eruption of Nevado del Ruiz, Colombia. Geochimica et Cosmochimica Acta 57, 1215–1219.
Shaw, H.R., 1985. Links between magma-tectonic rate balances, plutonism, and volcanism. Journal of Geophysical Research 90 (B13), 11275–11288.
Stamatelopoulou-Seymour, K., Vlassopoulos, D., Pearce, T.H., Rice, C., 1990. The record of magma chamber processes in plagioclase phenocrysts at Thera Volcano, Aegean Volcanic Arc, Greece. Contributions to Mineralogy and Petrology 104, 73–84.
Tamura, Y., Yuhara, M., Ishii, T., 2000. Primary arc basalts from Daisen Volcano, Japan: equilibrium crystal fractionation versus disequilibrium fractionation during supercooling. Journal of Petrology 41, 431–448.
Turner, S.P., Foden, J., 2001. U, Th and Ra disequilibria, Sr, Nd and Pb isotope and trace element variations in Sunda Arc lavas; predominance of a subducted sediment component. Contributions to Mineralogy and Petrology 142, 43–57.
Turner, S., Hawkesworth, C., van Calsteren, P., Heath, E., Macdonald, R., Black, S., 1996. U-series isotopes and destructive plate margin magma genesis in the Lesser Antilles. Earth and Planetary Science Letters 142 (1–2), 191–207.
Turner, S., Hawkesworth, C., Rogers, N., Bartlett, J., Worthington, T., Hergt, J., Pearce, J., Smith, I., 1997. ^{238}U–^{230}Th disequilibria, magma petrogenesis, and flux rates beneath the depleted Tonga-Kermadec island arc. Geochimica et Cosmochimica Acta 61 (22), 4855–4884.
Turner, S.P., McDermott, F., Hawkesworth, C.J., Kepezhinskas, P., 1998. A U-series study of lavas from Kamchatka and the Aleutians: constraints on source composition and melting processes. Contributions to Mineralogy and Petrology 133, 217–234.
Turner, S.P., Peate, D.W., Hawkesworth, C.J., Eggins, S.M., Crawford, A.J., 1999. Two mantle domains and the time scales of fluid transfer beneath the Vanuatu Arc. Geology 27, 963–966.
Turner, S., Evans, P., Hawkesworth, C., 2001. Ultrafast source-to-surface movement of melt at island arcs from ^{226}Ra–^{230}Th systematics. Science 292, 1363–1366.
Turner, S.P., Bourdon, B., Gill, J., 2003a. Insights into magma genesis at convergent margins from U-series isotopes. In: Bourdon, B., Henderson, G.M., Lundstrom, C.C., Turner, S.P. (Eds.), Uranium Series Geochemistry, Reviews in Mineralogy and Geochemistry. The Mineralogical Society of America, Washington, pp. 255–315.
Turner, S.P., Foden, J., George, R.M.M., Evans, P., Varne, R., Elburg, M., Jenner, G., 2003b. Rates and processes of potassic magma generation at Sangeang Api volcano, east Sunda arc, Indonesia. Journal of Petrology 44, 491–515.
Turner, S.P., George, R.M.M., Jerram, D.A., Carpenter, N., Hawkesworth, C.J., 2003c. Some case studies of plagioclase growth and residence times in island arc lavas from the Lesser Antilles and Tonga, and a model to reconcile apparently disparate age information. Earth and Planetary Science Letters 214, 279–294.
Volpe, A.M., 1992. ^{238}U–^{230}Th–^{226}Ra disequilibrium in young Mt. Shasta andesites and dacites. Journal of Volcanology and Geothermal Research 53, 227–238.
Volpe, A.M., Hammond, P.E., 1991. ^{238}U–^{230}Th–^{226}Ra disequilibria in young Mount St. Helens rocks: time constraint for magma formation and crystallisation. Earth and Planetary Science Letters 107, 475–486.
Williams, C.A., 2001. The Geochemistry of Mt. Misery Volcano, St. Kitts, Lesser Antilles: A Combined U-Series Disequilibria and Crystal Size Distribution Study. PhD thesis, The Open University, Milton Keynes.
Woodhead, J.D., 1989. Geochemistry of the Mariana arc (western Pacific): source composition and processes. Chemical Geology 76, 1–24.
Wyllie, P.J., 1983. Experimental studies on biotite- and muscovite-granites and some crustal magmatic sources. In: Atherton, M.P., Gribble, C.D. (Eds.), Migmatites, Melting and Metamorphism. Shiva Publishing, Cambridge, MA, pp. 12–26.
Yoder, H.S., Stewart, D.B., Smith, J.R., 1957. Ternary feldspars. Carnegie Institution of Washington Year Book 56, 206–214.
Zellmer, G.F., 1998. Petrogenetic processes and their timescales beneath Santorini, Aegean Volcanic Arc, Greece. PhD thesis. The Open University, Milton Keynes, 264 pp.

Zellmer, G.F., Blake, S., Vance, D., Hawkesworth, C., Turner, S., 1999. Plagioclase residence times at two island arc volcanoes (Kameni islands, Santorini, and Soufriere, St. Vincent) determined by Sr diffusion systematics. Contributions to Mineralogy and Petrology 136 (4), 345–357.

Zellmer, G.F., Turner, S.P., Hawkesworth, C.J., 2000. Timescales of destructive plate margin magmatism: new insights from Santorini, Aegean volcanic arc. Earth and Planetary Science Letters 174 (3–4), 265–282.

Zellmer, G.F., Hawkesworth, C.J., Sparks, R.S.J., Thomas, L.E., Harford, C., Brewer, T.S., Loughlin, S., 2003a. Geochemical evolution of the Soufrière Hills volcano, Montserrat, Lesser Antilles Volcanic Arc. Journal of Petrology 44, 1349–1374.

Zellmer, G.F., Sparks, R.S.J., Hawkesworth, C.J., Wiedenbeck, M., 2003b. Magma emplacement and remobilisation timescales beneath Montserrat: insights from Sr and Ba zonation in plagioclase phenocrysts. Journal of Petrology 44, 1413–1431.