Magma Mixing, Recharge and Eruption Histories Recorded in Plagioclase Phenocrysts from El Chichón Volcano, Mexico

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Consistent core-to-rim decreases of $^{87}Sr/^{86}Sr$ ratios and coincident increases in Sr concentrations in plagioclase phenocrysts of varying size (~ 1 cm to 2 mm) are reported from samples of the 1982 and pre-1982 (~ 200 ka) eruptions of El Chichón Volcano. Maximum $^{87}Sr/^{86}Sr$ ratios of ~ 0.7054, significantly higher than the whole-rock isotopic ratios ($\sim 0.7040–0.7045$), are found in the cores of plagioclase phenocrysts, and minimum $^{87}Sr/^{86}Sr$ ratios of ~ 0.7039 are found near some of the rims. Plagioclase phenocrysts commonly display abrupt fluctuations in An content (up to 25 mol %) that correspond to well-developed dissolution surfaces. The isotopic, textural and compositional characteristics suggest that these plagioclase phenocrysts grew in a system that was periodically recharged by higher-temperature magma with a lower $^{87}Sr/^{86}Sr$ ratio and a higher Sr concentration. Rim $^{87}Sr/^{86}Sr$ ratios in plagioclase phenocrysts of rocks from the 200 ka eruption indicate that, at that time, the magma had already attained the lowest recorded $^{87}Sr/^{86}Sr$ value of the system (~ 0.7039). In contrast, cores from plagioclase phenocrysts of the 1982 eruption, inferred to have grown in the past few thousand years, have the highest recorded $^{87}Sr/^{86}Sr$ ratios of the system. Collectively, the Sr isotopic data (for plagioclase and whole rock), disequilibrium textural features of the phenocrysts, known eruption frequencies, and inferred crystal-residence times of the plagioclases are best interpreted in terms of an intermittent magma chamber model. Similar processes, including crustal contamination, magma mixing, periodic recharge by addition of more mafic magma to induce plagioclase disequilibrium (possibly triggering eruption) and subsequent re-equilibration, apparently were operative throughout the 200 ky history of the El Chichón magma system.

INTRODUCTION

The bulk compositions of volcanic rocks typically represent the integrated effects of fractional crystallization, magma mixing and contamination acting on magmas since they originally separated from their source. However, to characterize magma sources and magma-generation mechanisms, a more complete understanding and quantification of these processes and the timescales over which they operate is required. Bulk isotopic analyses of magmatic differentiates provide only an 'averaged' result that is unlikely to identify end-member compositions, and are difficult to interpret. Isotopic signatures of end-members may be preserved in early-formed crystals from magmas that have subsequently hybridized, melt inclusions from precursor, less-differentiated magmas, or xenocrysts from wallrock assimilants (Davidson et al.,

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1998; Knesel *et al.*, 1999; Tepley *et al.*, 1999). Crystal-isotope stratigraphy (textural, chemical and isotopic analysis of single crystals and growth zones within crystals) is especially successful in identifying the endmembers involved and recording the pathways of interactions.

Here, we report results of crystal-isotope stratigraphy studies on several plagioclase phenocrysts in rocks from an eruption ∼200 ky ago and one in 1982 from El Chichón Volcano. Previous work has shown that El Chichón magmas are remarkably similar to one another throughout the entire known span of eruptive activity (276 ka to 1982; Table 1); whole-rock Sr and Nd isotopic compositions plot in a very small compositional field (Rose *et al.*, 1984; McGee *et al.*, 1987; Tilling *et al.*, 1987; Tilling & Arth, 1994; Fig. 1). However, phenocryst phases are in isotopic disequilibrium with their host rock, with their $^{87}Sr/^{86}Sr$ ratios increasing in the order from groundmass to clinopyroxene, apatite, anhydrite, bulk rock and plagioclase (Tilling & Arth, 1994). Crystal-isotope stratigraphy of plagioclase phenocrysts discussed below has revealed systematic core-to-rim decreases in $^{87}Sr/^{86}Sr$ ratios, increases in Sr concentration, large variations in An content and obvious textural discontinuities. Using these isotopic, compositional and textural features, it is our goal to unravel the complex story of disequilibrium evolution of the El Chichón magmatic system.

GEOLOGIC SETTING

El Chichón is a small trachyandesite volcano located in the state of Chiapas in southeastern Mexico (Fig. 2). This volcanic edifice forms part of the Chiapanecan Volcanic Arc, a NW-trending volcanic zone of Pliocene to Recent volcanic centers. It is situated near the junction of the North American plate and the Caribbean and Cocos plates (McGee *et al.*, 1987), which constitutes a region of complex tectonic structure, possibly producing magmatism associated with a subduction and/or a transform fault setting (Duffield *et al.*, 1984). The volcano overlies a Jurassic to Miocene sequence of carbonate, sandstone and evaporite deposits.

El Chichón is a complex of trachyandesitic, plagioclase-porphyritic domes and associated pyroclastic flow and airfall deposits (McGee *et al.*, 1987). In addition to plagioclase, phenocryst phases include hornblende, and lesser amounts of clinopyroxene, biotite, quartz, apatite and anhydrite (observed in fresh samples from the 1982 eruption). The juvenile products of the volcano have essentially maintained the same chemical bulk composition during the past 0·3 My (Rose *et al.*, 1984; McGee *et al.*, 1987; Tilling *et al.*, 1987; Table 1).

El Chichón has erupted at least 11 times within the past 8000 years with repose times of 100–600 years (Tilling *et al.*, 1984; Espíndola *et al.*, 2000). The oldest

Table 1: Chemical and isotopic analyses of pre-1982 and 1982 whole-rock samples illustrating the similarity in chemical composition and diversity in isotopic composition despite ∼276 ky between eruption times

Sample	(Pre-1982) 9	(1982) 5a
SiO_2	57·8	55·7
Al_2O_3	18·2	18·2
Fe_2O_3	6·23	6·37
MgO	2·21	2·21
CaO	7·04	7·92
Na_2O	4·07	4·09
K_2O	2·73	2·72
TiO_2	0·64	0·65
MnO	0·17	0·17
P_2O_5	0·32	0·35
SO_3	0·02	0·65
Total	99·50	99·15
Sr (ppm)	1200	1200
Ba (ppm)	800	800
$^{87}Sr/^{86}Sr$	0·70439	0·70406

Sample 9 is from a lava dome or flow in the wall of the summit crater and dated by K–Ar yielding a whole-rock age of 0·27 ± 0·006 Ma (Duffield *et al.*, 1984). Sample 5a is a bread-crust bomb on top of a pyroclastic flow southwest of the summit (McGee *et al.*, 1987). Major and minor elements of both samples were performed by J. Taggert at US Geological Survey, Denver, using X-ray fluorescence. Trace elements were collected via inductively coupled plasma [from McGee *et al.* (1987)]. Precision of measured $^{87}Sr/^{86}Sr$ is <0·00002 (R. I. Tilling & J. G. Arth, unpublished data, 1993).

rock recovered to date is a trachyandesite yielding a K–Ar whole-rock age of 276 ± 6 ka (Duffield *et al.*, 1984). Additionally, a whole-rock K–Ar age of 209 ± 19 ka (sample 74-26 used in this study) was reported by Damon & Montesinos (1978).

ANALYTICAL PROCEDURES

In this paper, we describe the results of crystal-isotope stratigraphy performed on various crystals in rocks of the 1982 eruption and of one representative of pre-1982 eruption (209 ± 19 ka). We focused on crystals from samples of the 1982 eruption for the following reasons: (1) they chronicle magmatic conditions in the El Chichón magma chamber over the longest extent of time; (2) they have already been well characterized geochemically,
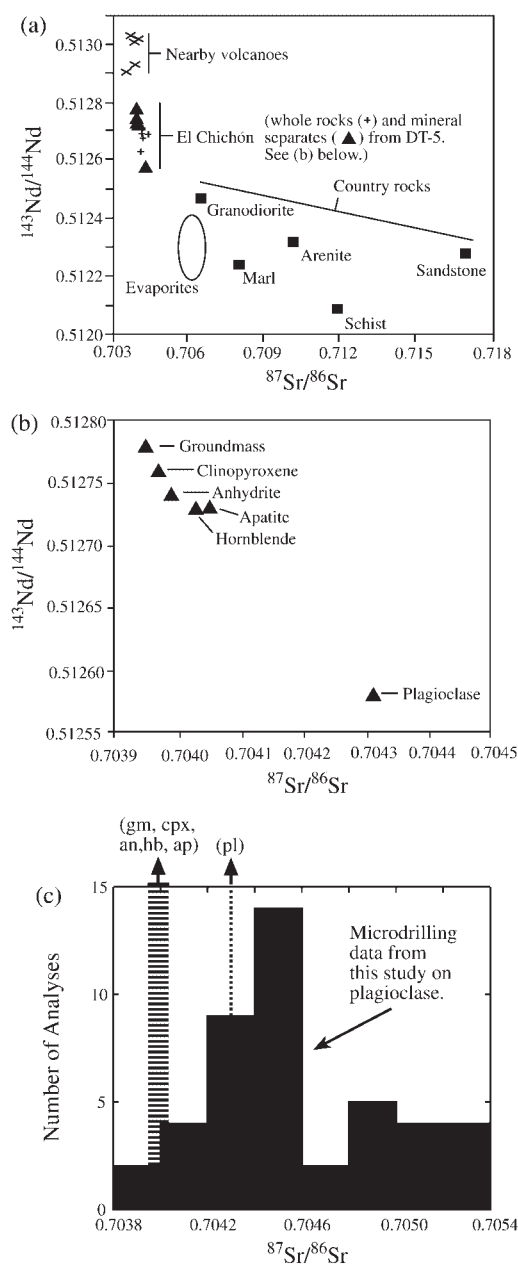
Fig. 1. (a) $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ ratios of El Chichón rocks and mineral separates (Tilling & Arth, 1994), local country rocks and nearby volcanoes (R. I. Tilling & J. G. Arth, unpublished data, 1993). (b) Inset figure of (a) showing the isotopic range of mineral phases from sample DT-5 (Tilling & Arth, 1994). Analysis errors in (a) and (b) are less than symbol size. (c) Diagram of the isotopic range of minerals from DT-5 (gm, groundmass; cpx, clinopyroxene; an, anhydrite; hb, hornblende; ap, apatite; pl, plagioclase; Tilling & Arth, 1994) in comparison with the isotopic range and number of $^{87}Sr/^{86}Sr$ analyses collected from plagioclase microdrilling traverses (11 crystals) in this study.

petrologically and isotopically, including the bulk mineral analyses of different phenocrysts (McGee & Tilling, 1983; Belkin *et al.*, 1984; Luhr *et al.*, 1984; Rose *et al.*, 1984;
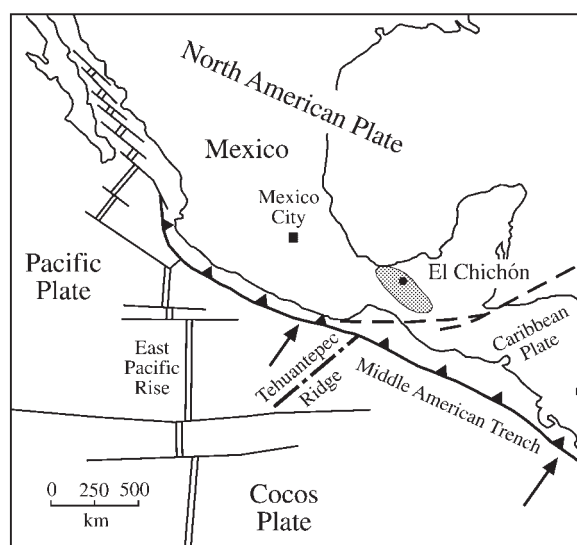

Fig. 2. Location map of El Chichón Volcano, Chiapas, Mexico, and its relationship to the North American, Pacific, Caribbean and Cocos plates. Dashed lines are a transform boundary separating the North American plate from the Caribbean plate. El Chichón lies within the NW-trending Chiapanecan Volcanic Arc (stippled area; after Luhr *et al.*, 1984).

McGee *et al.*, 1987; Tilling & Arth, 1994); (3) they have not undergone any post-eruption alteration processes. One pre-1982 sample was analyzed to determine magmatic conditions in an earlier stage and hence to assess possible changes in the magma system through time. Drill-hole $^{87}Sr/^{86}Sr$ ratios of 11 crystals are listed in Table 2, nine from the 1982 eruption, two from the pre-1982 eruption. Whenever possible, we combined Nomarski differential interference contrast (NDIC) imaging (Anderson, 1983; Pearce & Clark, 1989) and electron-probe traverses with crystal-isotope drill-hole traverses to relate the textural features, major element chemistry and isotopic composition.

NDIC was used to image textures in plagioclase crystals. Electron-microprobe traverses were taken to transect the most growth zones and allow correlation of textural variations with compositional changes. Mineral phases were analyzed with a Cameca CAMEBAX electron microprobe at UCLA, using an accelerating potential of 15 kV, a beam current of 10 nA and count times of 20 s. Corrections for matrix effects were made using the procedures of Bence & Albee (1968) with modifications of Albee & Ray (1970). The micro-strontium isotopic technique we used has been fully described by Davidson & Tepley (1997), Davidson *et al.* (1998) and Tepley *et al.* (1999). Briefly, diamond-tipped drill bits, which range in outside diameter from 0·25 to 0·51 mm, are used to sample the plagioclase crystals at sites selected on the basis of previous NDIC and electron-microprobe work.

Table 2: Crystal-isotope stratigraphy values for plagioclase crystals in eruptive products of El Chichón Volcano

1982 eruptive products

Sample:	DT-5		DT-5a(1)		DT-5a(2)	
	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)
Core	0·705237 ± 23	1627	0·704632 ± 13	1482	0·704573 ± 11	1734
|	0·705290 ± 20	1722	0·704584 ± 11	1948	0·704710 ± 11	1643
to	0·704828 ± 11	1629	0·704574 ± 11	1851	0·704237 ± 11	1758
|	0·704561 ± 14	1794			0·704200 ± 10	2091
Rim	0·704598 ± 11	1941				

Sample:	DT-5a(2) P1		DT-5a(2) P2		DT-5a(3)	
	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)
Core	0·704485 ± 10	1522	0·705357 ± 10	1606	0·704525 ± 11	1622
|	0·704599 ± 11	1733	0·704850 ± 11	1623	0·704162 ± 10	1893
to	0·704599 ± 10	1753			0·704109 ± 15	2067
|	0·704558 ± 10	1697				
Rim	0·704336 ± 9	1939				

Sample:	DT-5a(4)		DT-6(1) P1		DT-6(2)	
	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)
Core	0·704357 ± 17	1639	0·705144 ± 10	1760	0·704952 ± 10	1858
|	0·704391 ± 10	1733	0·705219 ± 11	1714	0·704549 ± 10	1738
|	0·704138 ± 10	2070	0·704806 ± 10	1704	0·704873 ± 10	1753
to	0·704473 ± 13	1597				
|			0·704478 ± 10	1764		
|			0·704338 ± 10	1904		
Rim			0·704213 ± 11	1850		

Pre-1982 eruptive products

Sample:	74-26 P1			74-26 P2		
	$^{87}Sr/^{86}Sr$	Sr (ppm)	Rb (ppm)	$^{87}Sr/^{86}Sr$	Sr (ppm)	Rb (ppm)
Core	0·705184 ± 11	1238	3·8	0·704034 ± 10	2430	25·1
|	0·705038 ± 10	1453	3·7	0·703981 ± 10	1620	24·7
|	0·705090 ± 13	1410	5·0	0·703974 ± 11	1348	33·1
to	0·704599 ± 11	1607	4·7			
|	0·704273 ± 11	1783	11·2			
Rim	0·704314 ± 11	1759	4·8			

Note: Sr and Rb concentrations determined through isotope dilution.

All samples were analyzed using a dynamic peak-switching routine on a multicollector VG Sector thermal ionization mass spectrometer at UCLA. Sr concentrations were determined through spiking and the regular isotope-dilution techniques. All analyses were normalized to $^{86}Sr/^{88}Sr = 0.1194$. The 600 ng NBS 987 standard is routinely analyzed, yielding precisions of the order of 15 ppm, with no significant difference in the measured $^{87}Sr/^{86}Sr$ ratios of the standard (0.710250 ± 15, $n = 16$). Plagioclase crystals in El Chichón lavas typically contain >1200 ppm Sr, and they can be sampled with the smallest available drill bits to easily yield sufficient Sr (e.g. 5–10 ng) for precise analysis. Blanks for the chemical procedure average $\sim$25 pg, which is <0.1% of the Sr being analyzed.

RESULTS
Nomarski interferometry

The crystals of rocks from El Chichón Volcano have a wide variety of textural features represented by dissolution surfaces and zones that record a history of changing magmatic conditions. Dissolution surfaces are characterized by a continuum of textures ranging from rounding of euhedral crystal corners to deep embayments and wavy, irregular sides. Dissolution zones are chaotic regions of irregular boundaries thought to be produced by immersion of a more Na-rich plagioclase into a higher-temperature melt in equilibrium with Ca-rich plagioclase (Tsuchiyama, 1985). Partial dissolution of the crystal produces the sieve-textured appearance consisting of micrometer-scale channels of glass and An-rich plagioclase.

In this section, we examine in depth three different-sized crystals from rocks of the 1982 eruption ($\sim$4·5 mm, $\sim$2·5 mm and $\sim$1 mm; Fig. 3) that are representative of the variety of textures seen in all plagioclase from El Chichón. The largest crystal [DT-6(1) P1; Fig. 3a] has multiple dissolution zones of varied thickness and several areas of relatively smooth topography. In many cases, dissolution features surround or completely truncate interior growth zones. A medium-sized crystal [DT-5a(2) P1; Fig. 3b] is dominated by mostly smooth growth zones, and faceted boundaries are easily distinguished. This crystal lacks the thick and numerous dissolution zones that occur in sample DT-6(1) P1; however, it does have rounding of corners of growth zones, fluctuating growth boundaries, and a few thin dissolution edges or pockets. Sample DT-5a(2) P2 (Fig. 3c), the smallest crystal studied, has textures similar to DT-5a(2) P1 in that regular growth zones are apparent. Closer examination, however, reveals undulating crystal surfaces overlain by faceted growth zones that tend to restore the euhedral outline of the crystal.

Electron-microprobe analysis (EMPA)

As noted in previous studies (Belkin *et al.*, 1984; Luhr *et al.*, 1984; McGee *et al.*, 1987) one of the most distinctive features of the EMPA traverses is that most of the crystals are punctuated by sharp changes in An content. Figure 4 is a compilation of EMPA traverses across some of the crystals examined in this study. Most crystals have sharp jumps in An content of 10–25 mol % at various locations along the traverse. These jumps in An content appear randomly spaced and of variable magnitudes, but some systematic relations are observed. For instance, the larger crystals (>3 mm) vary widely in An content, whereas smaller crystals (<3 mm) have smaller degrees of variation. Only one sample [DT-7(2) P1] has relatively constant An content across the crystal, except for near its rim, where the An content increases. This pattern is in contrast to those of sample DT-7(1) P1 and other larger crystals (>3 mm) that show increases in An content of varying magnitudes, and is similar to smaller crystals (<3 mm) with few or no An spikes. Both crystals from the pre-1982 eruption sample (74-26) have multiple increases in An content of differing magnitudes. Regardless of the varying An contents, the overall pattern is relatively constant. Except for very large increases in An content near the edge of some crystals, which are probably related to eruption-growth kinetics, a baseline of $\sim$An$_{40}$ is generally observed, and consistently reoccurs after large jumps in An content for crystals in rocks of both the pre-1982 and 1982 eruptions.

$^{87}Sr/^{86}Sr$ microanalysis

Invariably, the cores in each of the 11 crystals analyzed have higher $^{87}Sr/^{86}Sr$ ratios than their rims (Table 2). The highest $^{87}Sr/^{86}Sr$ ratio measured in this study is 0.705357 ± 10 from the core of plagioclase sample DT-5a(2) P2. A few of the drill-hole $^{87}Sr/^{86}Sr$ ratios cluster near 0·7052–0·7053, such as in samples DT-5a(2) P2, DT-5, DT-6(1) P1 (all 1982 products) and 74-26 P1 (pre-1982 product). The lowest $^{87}Sr/^{86}Sr$ ratio measured is 0.703974 ± 11 from the rim of sample 74-26 P1. The range in $^{87}Sr/^{86}Sr$ ratios recorded by plagioclase phenocrysts in any hand sample, and even within some crystals, exceeds that for all of the mineral separates reported by Tilling & Arth (1994). This observation is remarkable considering the very limited variation in isotopic composition among the bulk rocks (see Fig. 1). There is an overall correlation between increasing Sr concentrations and decreasing $^{87}Sr/^{86}Sr$ ratios (Fig. 5). Sr concentrations generally increase from core ($\sim$1500 ppm) to rim ($\sim$1800 ppm) although the core Sr concentration of the pre-1982 sample is significantly lower at $\sim$1200 ppm.
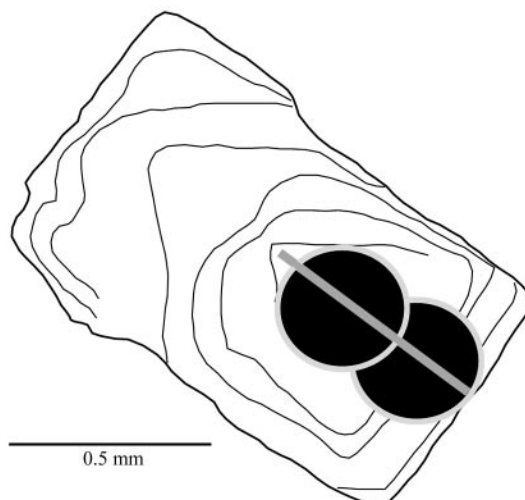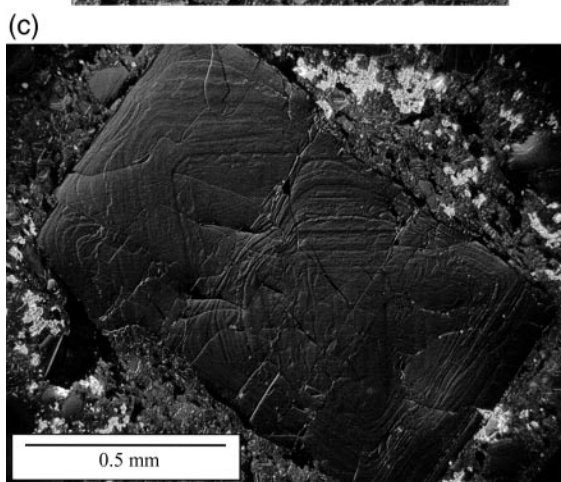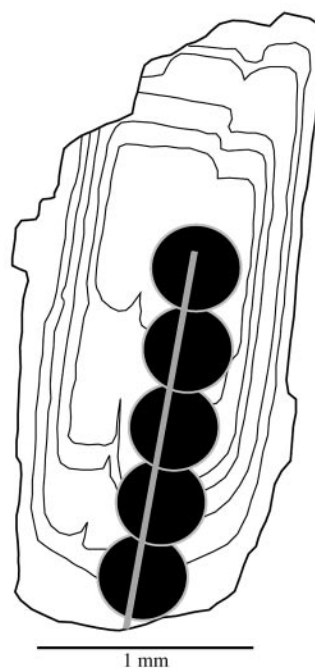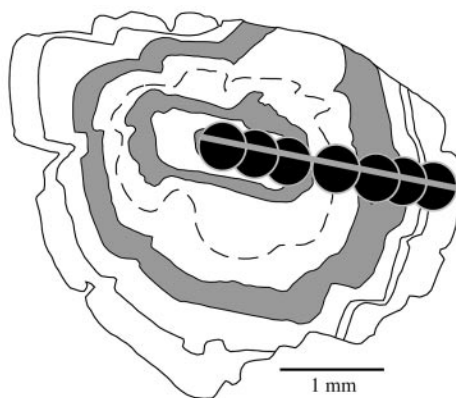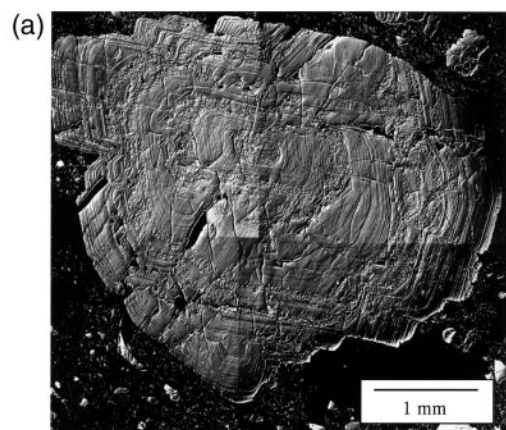
Figure 6 compares the changing $^{87}Sr/^{86}Sr$ ratios and An contents of two crystals from the 1982 eruption and one crystal from a pre-1982 eruption. Sample DT-6(1) P1 (Fig. 6a), ∼4·5 mm in length (see Fig. 3a), has numerous small-scale growth zones exhibited as oscillatory zoning, but it has relatively thick (50–250 μm) dissolution zones that correspond to changes in An contents from ∼An_{35} to >An_{65}. An increase in An content in this region may reflect analysis of micrometer-scale melt channels; however, good stoichiometry on most of the analyses is indicative of Ca-rich plagioclase material. Our attempt to drill micro-Sr holes in either simple, oscillatory-zoned areas or areas on either side of major dissolution zones without transgressing into other areas was largely successful. Although there are numerous fluctuations in An content across the crystal, there is no distinct relationship between the An content and $^{87}Sr/^{86}Sr$ ratios (compare with Fig. 4).

Sample DT-5a(2) P1 is a little longer than 2·5 mm (Fig. 6b) and has many small-scale oscillatory and few obvious dissolution zones (Fig. 3b). The An content has regular oscillations similar to those of DT-6(1) P1, but with a narrower compositional range. The An content varies from An_{35} to approximately An_{50}, except near the outer edge of the crystal, where the An content increases sharply to An_{70}. The $^{87}Sr/^{86}Sr$ ratios throughout the crystal are relatively low compared with those of DT-6(1)P1. The first sample hole (core) has a lower $^{87}Sr/^{86}Sr$ ratio than those for the following three holes, but higher than that for the rim hole. The Sr concentrations generally increase toward the rim from ∼1500 ppm Sr to ∼1900 ppm (Fig. 5). Again, the An content appears uncorrelated with either Sr concentrations or isotope ratios.

Sample 74-26 P1, from an eruption dated at over 200 ky ago, is ∼5 mm in length (Fig. 6c), and has numerous dissolution zones of variable thickness. This crystal has many spikes in An content, most being increases of ∼10–15 mol %, with occasional spikes up to 25 mol %. The total range of An content is similar to that for 1982 eruption crystals, varying between An_{35} and An_{70}. Interestingly, this crystal has a $^{87}Sr/^{86}Sr$ profile similar to that of the 1982 sample DT-6(1) P1. Its highest $^{87}Sr/^{86}Sr$ value is in the core, ∼0·7052, and the ratios decrease to ∼0·7042 near the rim. The Sr concentrations behave similarly: lowest in the core, ∼1200 ppm Sr, and highest near the rim, ∼1750 ppm Sr.

DISCUSSION
Origin of major-element zonation

In common with many other well-documented subduction-related magma systems, the plagioclase phenocrysts have widely fluctuating An contents (Anderson, 1984; Nixon & Pearce, 1987; Tepley *et al.*, 1999). Pearce & Kolisnik (1990) have speculated in general on the ubiquitous compositional zoning and on the textural features commonly seen in volcanic plagioclases. Repeated, fine-scale (zones of 1–10 μm width), and small compositional changes (1–10 mol %) that commonly occur in volcanic plagioclases are thought to reflect near-equilibrium, incremental diffusion-controlled growth (Haase *et al.*, 1980; Pearce & Kolisnik, 1990; Pearce, 1993; Singer *et al.*, 1995). Larger-scale changes (growth zones with thicknesses up to 100 μm), with greater compositional amplitudes (10–25 mol %), are usually associated with well-developed dissolution surfaces, and may reflect large-scale disturbances of the crystallizing system such as magma mixing, magma overturn or significant changes in pressure and volatile content accompanying eruptions (Pearce & Kolisnik, 1990).

Belkin *et al.* (1984) and McGee *et al.* (1987) suggested that the complex zoning and repetitive inclusion-rich zones appear to record systematic fluctuations in the physical and chemical nature of the El Chichón magmatic system during crystal growth. They speculated that fluctuations in the magmatic environment, such as convective stirring or changes in volatile pressure of the system, may account for the repetitive, large compositional variations in the plagioclase crystals. A large increase in volatile content of the system would have the effect of lowering the plagioclase solidus temperature and stabilize a more Ca-rich plagioclase composition. Subsequent release of the volatiles attending eruption would have the effect of supercooling the plagioclase crystals and allow them to re-equilibrate to a lower An content plagioclase composition. The observed multiple and repetitive dissolution and An spikes may then reflect multiple and repetitive eruptions of El Chichón through its eruptive history.

In principle, the textural characteristics and compositional changes described above and reported by Belkin *et al.* (1984) and McGee *et al.* (1987) can be produced by closed-system processes such as convection and periodic eruptions. Open-system processes, such as magma mixing, cannot be *a priori* distinguished simply

Fig. 3. Nomarski differential interference contrast (NDIC) images and interpretative line drawings of representative plagioclase crystals from rocks of El Chichón Volcano. The images demonstrate the variety of equilibrium and disequilibrium textures associated with this complex magmatic system. The line drawings illustrate dissolution surfaces or zones and locations of drill holes (large filled circles) and EMPA traverses (dark gray line from center of crystal to edge). (a) Sample DT-6(1) P1. Stippled areas are dissolution zones. Dashed line outlines pronounced dissolution surface or boundary. (b) Sample DT-5a(2) P1. This sample is characterized by well-developed dissolution or resorption surfaces, as the rounded lines indicate. (c) Sample DT-5a(2) P2. This sample has minor dissolution surfaces. (See text for detailed descriptions.)
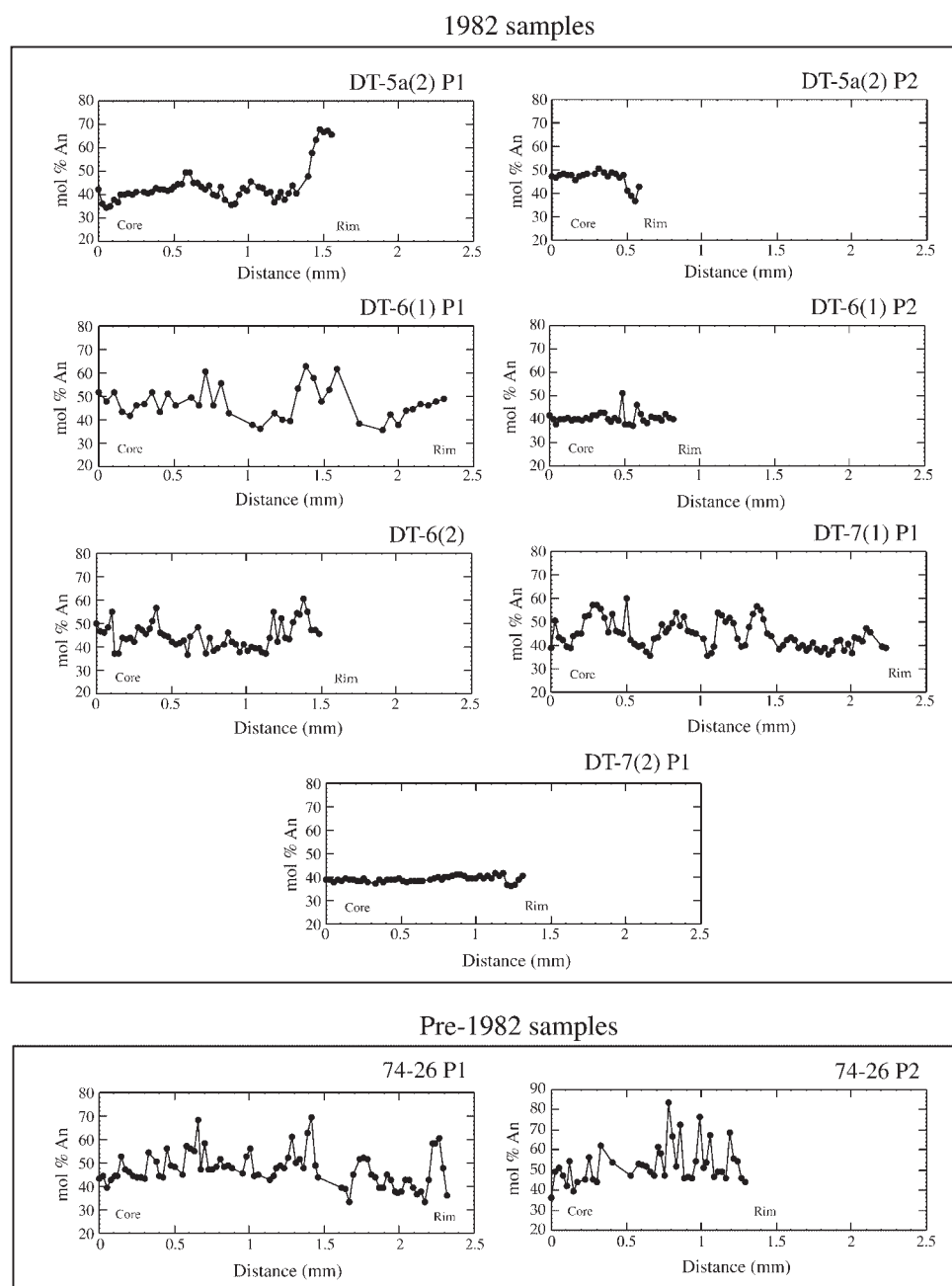
Fig. 4. Electron-microprobe traverses from plagioclase phenocrysts from the 1982 and pre-1982 eruptive products of El Chichón. The traverses are from the approximate core of the crystal to the rim, as noted in the graphs. A common x-scale (0–2·5 mm) is used for easy comparison.

on the basis of textural features among the minerals. However, the changing isotopic ratios across crystals preclude closed-system behavior by itself. We interpret the repetitive large-scale major-element fluctuations in the plagioclase composition to reflect open-system, magma-mixing events. The closed-system processes of convective stirring or changes in volatile pressure of the system may also be important, but only if in response to a magma-mixing event.

Origin of isotopic zonation

Consistent core-to-rim decreases in $^{87}Sr/^{86}Sr$ ratios have been shown for a number of plagioclase phenocrysts of the El Chichón magma system, and these are apparently independent of textural complexity and size. Any explanation for crystal isotopic zonation must also account for (1) isotopic disequilibrium between groundmass (0·7039) and whole-crystal plagioclase phenocrysts
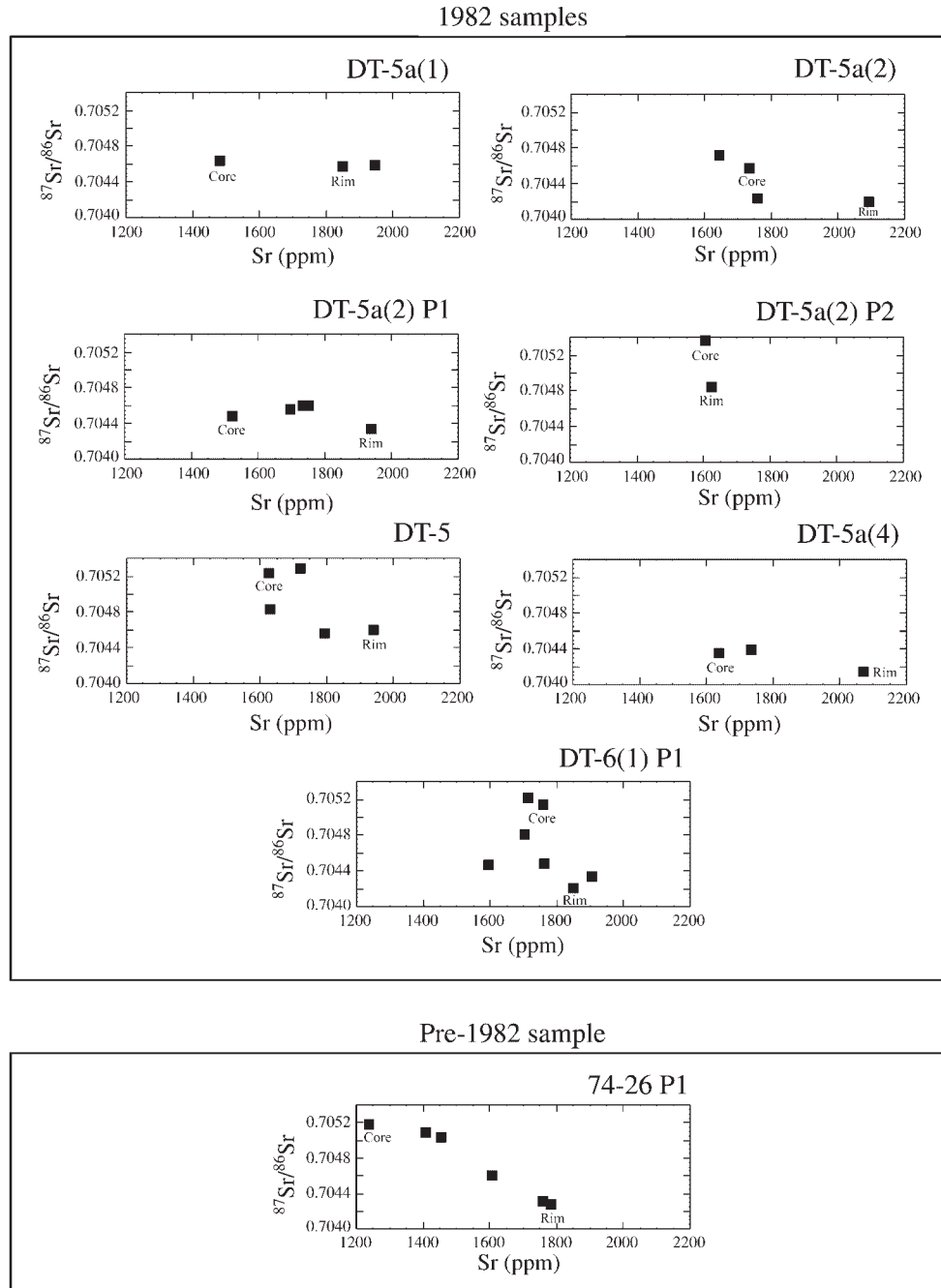
Fig. 5. Plot of $^{87}Sr/^{86}Sr$ ratios vs Sr concentration for microanalyses of traverses in plagioclase phenocrysts. All plagioclase phenocrysts are from rocks of the 1982 eruption, except sample 74-26 P1, which is from an eruption $\sim$ 200 ky ago. The diagram illustrates the general correlation between decreasing $^{87}Sr/^{86}Sr$ ratios and increasing Sr concentrations. Sr concentrations were determined by isotope-dilution analytical techniques.

(0·7042), as well as with other phenocryst phases, and (2) the textural and compositional features developed during phenocryst growth. We offer three possible explanations to account for the isotopic zonation in the plagioclase phenocrysts: (1) growth of phenocrysts in a pre-existing magmatic system with high $^{87}Sr/^{86}Sr$ ratios

(perhaps through contamination) that is periodically recharged by magma(s) with low $^{87}Sr/^{86}Sr$ ratio(s); (2) growth of phenocrysts residing in a high-$^{87}Sr/^{86}Sr$ magma progressively contaminated by country-rock or groundwater with lower $^{87}Sr/^{86}Sr$ ratios; (3) tracer diffusion of Sr from isotopically unzoned plagioclase phenocrysts with

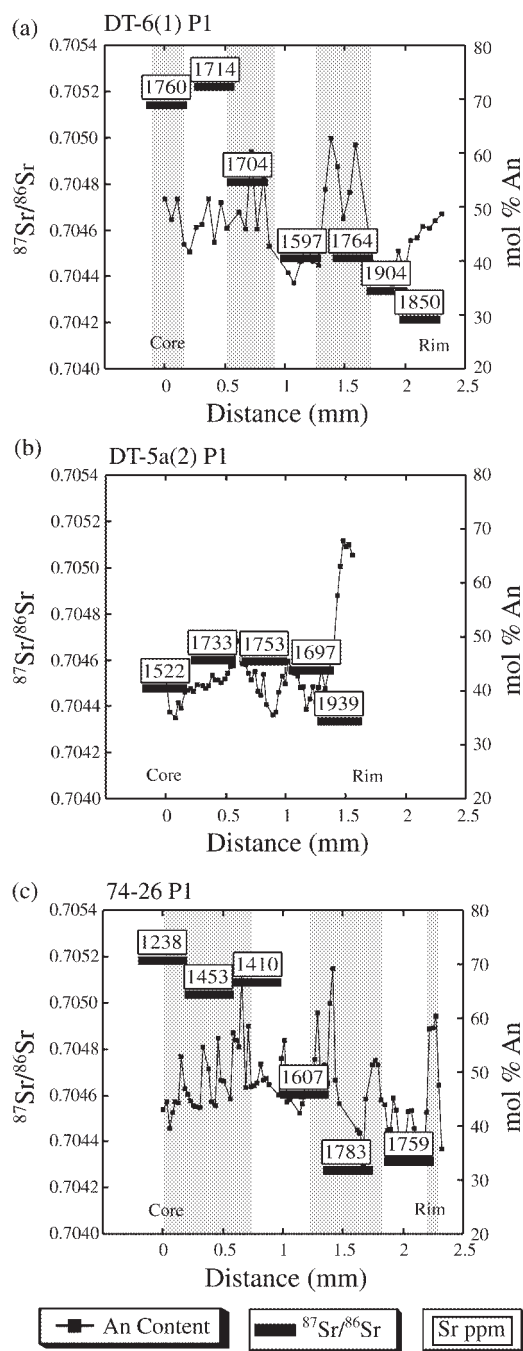**Fig. 6.** Comparison diagram of $^{87}Sr/^{86}Sr$ ratios, Sr concentrations, textural patterns and An content across three representative plagioclase phenocrysts from this study. Stippled bands represent dissolution zones in the crystal. (a) and (b) are from rocks of the 1982 eruption [DT-6(1) P1 and DT-5a(2) P1, respectively], whereas (c) is from rocks of an eruption ~200 ky ago (74-26 P1). The traverses illustrate the general decrease in $^{87}Sr/^{86}Sr$ ratios from core to rim and the compositional traverses punctuated by large Ca-enrichment spikes of plagioclase phenocrysts in this study. Pre-drilled samples in (a) and (b) [DT-6(1) P1 and DT-5a(2) P1] are shown in a Nomarski image in Fig. 3a and 3b, respectively. Uncertainty in reported $^{87}Sr/^{86}Sr$ is given by thickness of black bar; width of bar represents hole diameter.

high $^{87}Sr/^{86}Sr$ ratios immersed in a reservoir of lower $^{87}Sr/^{86}Sr$ magma. At the time of eruption, the liquid as represented by groundmass separates and phenocryst rims had low isotopic ratios, whereas the crystalline phases had higher and variable $^{87}Sr/^{86}Sr$ ratios.

Crystal growth in a periodically recharged magma chamber

We suggest that Sr-isotope variations across the phenocrysts most probably record crystal growth in a magma chamber, to which magma with a lower $^{87}Sr/^{86}Sr$ ratio and a higher Sr concentration was periodically added, possibly resulting in eruption with each addition. Recharge events are recorded in the plagioclase-dissolution surfaces. Major-element variations across crystals attest to rapidly changing magmatic conditions possibly initiated by recharge magmas and maintained by hybrid magmas produced by mixing of the resident and recharge magmas. The high $^{87}Sr/^{86}Sr$ ratios of plagioclase cores, such as in 1982 ejecta, probably represent crystal growth in the early stages of a developing magma chamber when the $^{87}Sr/^{86}Sr$ value of the magma was ~0·7053, perhaps following the maximum influence upon the system by wallrock contamination.

Figure 7a summarizes the general characteristics observed in the plagioclase crystals from El Chichón, and is figuratively a transect from core to rim across a plagioclase crystal. A comparison is drawn between observed representative changes in An content and $^{87}Sr/^{86}Sr$ ratios across plagioclase crystals with time. The An content hovers about a baseline of ~An_{45-40} with periodic spikes to ~An_{75}. We argue that spikes in An content are indicative of changes in the compositional and thermal regimes of a magma chamber periodically recharged with hotter, more mafic magma. Coincident with spikes in An content, dissolution zones or surfaces and postulated recharge events are decreases in $^{87}Sr/^{86}Sr$ ratios from core to rim in the crystals.

The largest phenocrysts analyzed in this study from both the 200 ka eruption and the 1982 eruption have the largest variation in isotopic ratios presumably because, being larger, they had longer crystal-residence times and experienced the maximum range of magmatic conditions (see Table 2). Except for one instance, smaller crystals have lower core $^{87}Sr/^{86}Sr$ ratios and generally have smaller isotopic variation across the crystal. If the hypothesis that intra-crystalline isotopic variations are the result of crystal growth in a progressively recharged magma is correct, these observations suggest that smaller crystals nucleated and grew at a later time in an evolving magma chamber and for only a short time before being erupted. This is illustrated in Fig. 7a, in which the largest phenocrysts crystallize near the peak of crustal contamination. In this manner, the largest crystals experience the longest residence time in a magma chamber
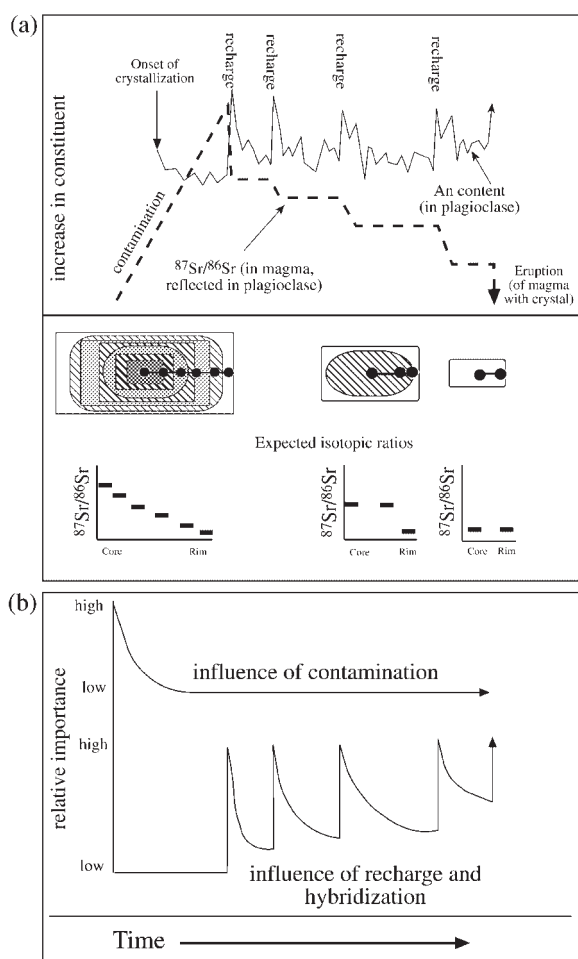
Fig. 7. (a) Relationship with time between contamination and recharge to the changing An contents and $^{87}Sr/^{86}Sr$ ratios across plagioclase phenocrysts. Representative plagioclase crystals of different longevity and their expected $^{87}Sr/^{86}Sr$ ratios are keyed to nucleation and growth at varying times from the maximum influence of contamination to the maximum influence of recharge magmas on the system. (b) Relative importance of contamination and influence of recharge and hybridization with time. This model is representative of the recurring processes in the evolution of one magma chamber with time (see discussion in the text).

and, therefore, record the maximum chemical, isotopic and textural changes. Smaller crystals have progressively less isotopic range (difference between core and rim $^{87}Sr/^{86}Sr$ ratios), shorter residence times and fewer dissolution zones or surfaces.

Figure 7b illustrates the relative importance of crustal contamination and recharge–hybridization processes on the growth of a plagioclase crystal in the system. We postulate that the influence of contamination is high early in the residence time of the larger crystals with relatively low Sr concentrations (e.g. 1200 ppm) and relatively high $^{87}Sr/^{86}Sr$ ratios (e.g. 0·7054). The influence of crustal

contamination during the remaining lifetime of a pla- gioclase crystal is relatively low. This may reflect a greater opportunity for interaction with country rocks when the system is immature, and then progressive insulation of magma by armoring of cumulate phases at the margins of the magma chamber(s). The decrease in $^{87}Sr/^{86}Sr$ ratios may be effected by a significant contribution of dissolved Sr from pre-existing crystals that inevitably have lower $^{87}Sr/^{86}Sr$ ratios than wallrock contaminants. The lowest curve in Fig. 7b suggests that recharge magmas introduced into an existing magma can dra- matically influence the compositional and thermal regime of that pre-existing magma. The influence on $^{87}Sr/^{86}Sr$ ratios can be particularly significant given the contrast in Sr concentrations between recharge magma and low- Sr wallrocks. Spikes in An content show the periodicity of the recharge events, and dissolution zones or surfaces show the effect of changing compositional and thermal parameters. The influence of each recharge event on the texture and composition of any phenocryst decreases as is borne out by the gradually changing isotopic ratios across plagioclase crystals (rather than precipitous isotopic changes) and resumption of constant composition (An content) crystal growth.

This diagram highlights the decoupling between the major-element compositions of plagioclase crystals and the changing isotopic ratios in the crystals. This de- coupling is further illustrated by the constant bulk-rock chemical compositions through time. Accumulation and hybridization of recharge magmas with pre-existing magmas progressively increases the Sr concentration and decreases the $^{87}Sr/^{86}Sr$ ratio of magma in the chamber to the extent that the last liquid from which the plagioclase crystals grows is enriched in Sr and has a low $^{87}Sr/^{86}Sr$ ratio (e.g. 0·7039–0·7040). A complex balance between recharge magmas, assimilation of these magmas, crustal contamination, and fractionation of magmas that yield a characteristic plagioclase composition and bulk whole- rock compositional homogeneity is maintained over time.

Crystal growth in a magma chamber progressively contaminated by a low-$^{87}Sr/^{86}Sr$ assimilant

An alternative explanation is that core-to-rim decreases in $^{87}Sr/^{86}Sr$ ratios reflect growth of phenocrysts in magma with a high $^{87}Sr/^{86}Sr$ initial ratio, which becomes pro- gressively contaminated by an assimilant with a lower $^{87}Sr/^{86}Sr$ ratio. However, all possible contaminants in terms of known basement lithologies have $^{87}Sr/^{86}Sr$ ratios that are distinctly higher than even the plagioclase core with the highest $^{87}Sr/^{86}Sr$ ratio (see Fig. 1). Continued growth of plagioclase crystals in magma differentiating by combined assimilation–fractional crystallization would result in core-to-rim increases in $^{87}Sr/^{86}Sr$ ratios, which we have not observed. Even if a hitherto unrecognized

low-^{87}Sr/^{86}Sr assimilant exists, we would expect assimilation–fractional crystallization to be a gradual process occurring with cooling. Under such conditions, it is unlikely to produce the textural discontinuities in growth and the attendant punctuated changes in major-element composition observed in the plagioclase phenocrysts.

Contamination by groundwater is a possible but unlikely explanation. Observations rule against this scenario because, typically, groundwater may contain only a few ppm of Sr of variable ^{87}Sr/^{86}Sr ratios. It would require huge volumes of groundwater to affect the ^{87}Sr/^{86}Sr of the magmas, as they normally contain very high Sr concentrations ($\sim$900 ppm). Adding large volumes of groundwater to liquid magma is physically implausible (e.g. Taylor & Sheppard, 1986), and in any case, reported δ^{18}O values of 7·2–8·1 (Rye *et al.*, 1984) do not reflect significant shifts of δ^{18}O from the typical magmatic values. Furthermore, there is no reason to believe that groundwater is likely to have ^{87}Sr/^{86}Sr ratios lower than $\sim$0·705.

Diffusion-controlled profiles

It might be argued that tracer diffusion could produce the core-to-rim decreases in ^{87}Sr/^{86}Sr ratios because samples DT-6(1) P1 and 74-26 P1 have isotopic profiles that generally resemble diffusion profiles (Fig. 8). Assuming that a plagioclase phenocryst was homogeneous in Sr-isotopic composition and then immersed in a reservoir with lower ^{87}Sr/^{86}Sr ratios, we modeled the timescale needed to produce the isotopic profile. In Fig. 8, the dotted lines represent modeled diffusion profiles based on Sr tracer diffusion perpendicular to the crystal–melt interface using diffusion coefficients appropriate for An content and temperature of the El Chichón system (Baker, 1989; Cherniak & Watson, 1994; Giletti & Casserly, 1994). The diagram demonstrates that each crystal would be required to remain at high temperatures undisturbed for $\sim$10^5–10^7 years (depending on diffusion coefficient) to produce similar diffusion profiles for plagioclase phenocrysts erupted in 1982 and those erupted $\sim$200 ky ago.

A small-volume, evolving system cannot maintain temperature stability for such long times; this characteristic argues against diffusion as an important process producing the observed isotopic variances. The complexity of crystal textures such as dissolution zones, undulating growth surfaces and growth zones, together with the very sharp spikes in An content, also argues against diffusion as a controlling influence on a crystals' ^{87}Sr/^{86}Sr ratios.

Magma chamber development and crystal residence time

We now address the question of whether the magma system represented by 200 ka rocks was sampled later by
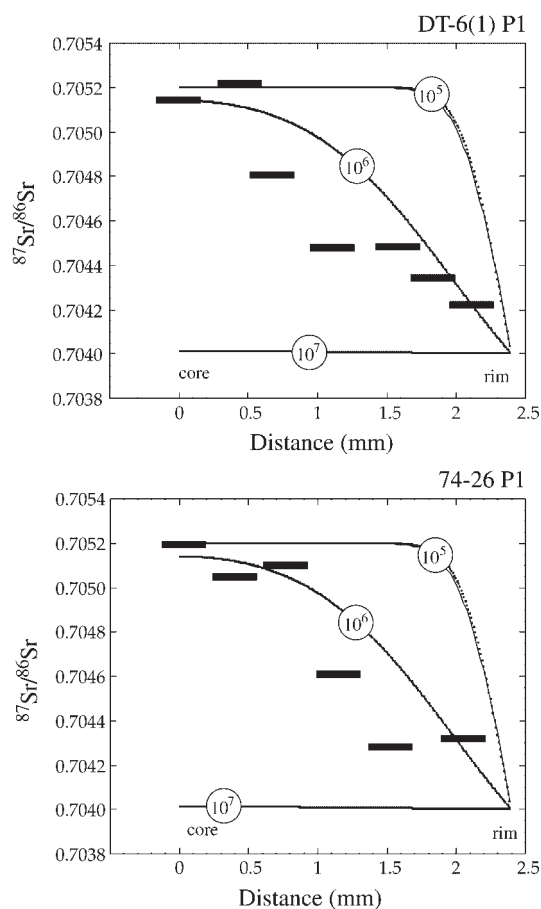

Fig. 8. Modeled diffusion profiles from plagioclase phenocrysts of rocks from the 1982 eruption [DT-6(1) P1] and from an eruption $\sim$200 ky ago (74-26 P1). Uncertainty in reported ^{87}Sr/^{86}Sr is given by thickness of symbol; width of symbol represents hole diameter. Curves labeled in years show solution to equation 6·18 of Crank (1975) for diffusion in a sphere of constant surface concentration. Strontium diffusion coefficient of 10^{-16} cm^2/s at 800°C is used in calculation (Baker, 1989; Cherniak & Watson, 1994; Giletti & Casserly, 1994). Decreasing the diffusion coefficient one order of magnitude decreases the timescales by one order of magnitude.

the 1982 eruption; that is, has a viable (liquid) magma chamber existed beneath El Chichón for 200 ky? A rim ^{87}Sr/^{86}Sr value from the pre-1982 eruption sample 74-26 P2 suggests that by $\sim$200 ka BP, the ^{87}Sr/^{86}Sr ratio of the magma in the system decreased to $\sim$0·7039, the lowest ratio obtained in this study. This observation would require that any crystals nucleating and growing in this magma after 200 ka should record low ^{87}Sr/^{86}Sr ratios in their cores, which we have not observed. The implication of these data is that phenocrysts erupted in 1982 lava must have nucleated and grown in a different magma chamber—albeit perhaps occupying the same general plumbing arrangement.

The hypothesis of different magma chambers for the 1982 and pre-1982 eruptions can be evaluated further

by an assessment of crystal residence times. The number of spikes in An content increases with the size of a crystal (Fig. 4). The general correlation between crystal size and number of An spikes supports the concept that larger crystals have longer residence times and have experienced more events that cause large variations in the An content of the plagioclase phenocryst. The implication is that plagioclase phenocrysts in both the 1982 eruptive products and pre-1982 products represent a range of residence times in the El Chichón magma system and also the similarity indicates similar fluctuations in processes.

On the basis of textural features in plagioclase phenocrysts and eruption frequency, we can assess crystal residence times. Assuming that the periodicity of eruptions (known repose periods are 100–600 years) reflects the minimum frequency of recharge events, and that the recharge events are marked by dissolution surfaces in the plagioclase crystals, then we can calculate maximum crystal growth rates. Using 0·25–0·5 mm as the typical distance between major dissolution surfaces and corresponding An content jumps (see Figs 3 and 4) the average growth rate is $\sim 10^{-11}$ cm/s, similar to growth rates estimated independently for natural systems (10^{-10}–10^{-11} cm/s; Cashman, 1993).

These results also are in accord with calculated residence times using U/Th disequilibria from six phases separated from a trachyandesite from the 1982 eruption. Pickett *et al.* (1993) reported Th isotopic homogeneity in groundmass, plagioclase, hornblende, augite, titanomagnetite and anhydrite, which implies crystal growth within the past few thousand years ($t = 1·3 \pm 2·3$ ky; Pickett *et al.*, 1993) and precludes crystal growth 200 ky ago. These data imply that plagioclase phenocrysts from the 200 ka eruption and from the 1982 eruption were grown at different times in an ephemeral sub-volcanic plumbing system that was capable of producing phenocrysts with similar characteristics over ~ 200 ky of volcanic activity.

Model of contamination and recharge

A model describing this system must account for the isotopic heterogeneity of plagioclase phenocrysts and for these apparently oscillating isotopic conditions over time. The simplest explanation to account for similarities in isotopic range and heterogeneity in plagioclase phenocrysts in rocks from the 200 ka eruption and the 1982 eruption, and the apparently short residence times for phases in the rocks of the 1982 eruption, is the presence of multiple magma chambers, one that produced the 200-ky-old rocks and another one that produced rocks from the 1982 eruption. We interpret the isotopic disequilibrium between phases of rocks of the 1982 eruption and the isotopic profiles of plagioclase phenocrysts to

reflect nucleation and growth of crystals at various times in magma of changing Sr isotopic and chemical composition.

An implication of this explanation is that processes within each magma chamber are similar, and that each one is initiated and maintained, for a finite time, by similar recharge magmas. We assume that mafic recharge magmas with low $^{87}Sr/^{86}Sr$ ratios, similar to primitive magma of nearby volcanoes (Fig. 1a) continually migrate through the crust, reach buoyancy equilibrium, and initiate the contamination–hybridization process anew. We envision either a plexus of magma chambers beneath El Chichón Volcano, some active, some in repose, or a single, irregularly shaped chamber, parts of which are not in good communication with other parts, parts of which are stagnant and isolated, and parts of which are actively feeding eruptions. Such a physical model is similar to those suggested for other intermediate composition, modest-sized magma systems on the basis of detailed petrographic and stratigraphically constrained geochemical observations [such as Ruapehu and Ngauruhoe volcanoes in New Zealand (Gamble *et al.*, 1999; Hobden *et al.*, 1999)].

Despite the complexity of processes postulated to act upon this system, the final product is fairly uniform in time. There is a balance between the diverse processes of recharge, assimilation and fractionation that yield the characteristic trachyandesitic composition. Additionally, although punctuated with spikes of high An content, plagioclase compositions remain relatively constant and buffered against recharge magmas and assimilated wallrock partial melts. Mineral assemblages in isotopic disequilibrium with their groundmass and changing $^{87}Sr/^{86}Sr$ ratios across plagioclase phenocrysts attest to the complexity behind this seemingly simple volcanic system.

An interesting observation is that this system lacks the traditionally obvious magma-mixing characteristics of abundant hybrid magmatic inclusions (e.g. Bacon, 1986; Feeley & Dungan, 1996). Mixing is apparently recorded only in the crystals themselves. The isotopic and textural characteristics of crystals are notably similar to those found in the Chaos Crags magma system, where the distribution of crystals among rhyodacite and variably disaggregated basaltic andesite magmatic inclusions records the hybridization process 'frozen in' an eruption (Tepley *et al.*, 1999). Given the similarities, the main differences between El Chichón and Chaos Crags is the paucity of magmatic inclusions in the former (J. L. Macias, personal communication, 2000). If inclusions were abundant at El Chichón then they have been efficiently disaggregated and hybridized. Magma from the 1982 eruption of El Chichón is known to have 2–4 wt % H_2O (Luhr *et al.*, 1984; McGee *et al.*, 1987). The high water content would reduce the viscosity of the magma, compared with anhydrous magma (Murase

& McBirney, 1973), and may therefore aid in more efficient mixing between the trachyandesite and the basalt.

CONCLUSIONS

In plagioclase phenocrysts of El Chichón, large-magnitude changes in An content, corresponding textural discontinuities, and systematic core-to-rim decreases in $^{87}Sr/^{86}Sr$ ratios reflect growth in a complexly changing thermal and compositional system. We conclude that the El Chichón magmas initially assimilated high-level, high-$^{87}Sr/^{86}Sr$ crust, and evolved subsequently through frequent recharge events to a low-$^{87}Sr/^{86}Sr$, high-Sr magma (Fig. 7). Inferred residence times for these crystals are relatively short (tens of years to a few thousand years). The occurrence of short-residence-time crystals with high-$^{87}Sr/^{86}Sr$ cores and low-$^{87}Sr/^{86}Sr$ rims in rocks erupted 200 ky ago and in 1982 suggests that similar rapid processes of differentiation occur repeatedly over the lifetime of the volcano.

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