A Volcano Rekindled: The Renewed Eruption of Mount St. Helens, 2004–2006
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Chapter 36

^{238}U-^{230}Th-^{226}Ra Disequilibria in Dacite and Plagioclase from the 2004–2005 Eruption of Mount St. Helens

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Abstract

Uranium-series disequilibria in whole-rock samples and mineral separates provide unique insights into the time scales and processes of magma mixing, storage, and crystallization. We present ^{238}U-^{230}Th-^{226}Ra data for whole-rock dacite and gouge samples and for plagioclase separated from two dacite samples, all erupted from Mount St. Helens between October 2004 and April 2005. We also present new ^{238}U-^{230}Th disequilibria for a suite of four reference samples from the 1980–86 eruption of Mount St. Helens. We use the U-series data to evaluate the origin of the 2004–5 magma, its relation to the 1980–86 magma, and the relation of 2004–5 phenocrysts to their host magmas. Dacite samples from 2004–5 show variable (^{230}Th)/(^{238}U), ranging from ^{238}U-enriched to ^{230}Th-enriched. (^{230}Th)/(^{232}Th) ratios in 2004–5 dacite and gouge samples do not vary outside of analytical error and are within the range of (^{230}Th)/(^{232}Th) measured for the 1980s reference suite. However, (^{230}Th)/(^{232}Th) for plagioclase separates for dome samples erupted during October and November 2004 are significantly different from corresponding whole-rock values, which suggests that a large fraction (>30 percent) of crystals in each sample are foreign to the host liquid. Furthermore, plagioclase in the two 2004 samples have U-series characteristics distinct from each other and from plagioclase in dacite erupted in 1982, indicating that (1) the current eruption must include a component of crystals (and potentially associated magma) that were not sampled by the 1980–86 eruption, and (2) dacite magmas erupted only a month apart in 2004 contain different populations of crystals, indicating that this foreign component is highly heterogeneous within the 2004–5 magma reservoir.

Introduction

After an 18-year hiatus, Mount St. Helens erupted in October 2004, beginning a period of dacite dome extrusion that produced more than 70 million cubic meters of crystal-rich, gas-poor dacite during the first 18 months of eruption (Pallister and others, this volume, chap. 30). Extrusion continues at the time of this writing (2007). The dome has erupted as essentially a solid plug, extruding a series of spines mantled by meter-thick fault gouge (for example, Pallister and others, this volume, chap. 30; Cashman and others, this volume, chap. 19). Samples have been collected by the staff at the U.S. Geological Survey Cascades Volcano Observatory and distributed to a group of ~25 investigators working on various petrologic and geochemical investigations aimed at understanding the origin and characteristics of the newly erupted material and the relation of the currently erupting magma to that erupted in the 1980–86 eruptive cycle. Major- and trace-element compositions of the 2004–5 dacite whole-rock samples are similar to those of the 1980–86 dacite dome (for example, Pallister and others, 2005; and this volume, chap. 30), suggesting the possibility that the current eruption is tapping magma that remained in the reservoir after the 1980s. In this chapter, we present ^{238}U-^{230}Th-^{226}Ra data for a suite of samples from the first six months (October 2004–April 2005) of the ongoing eruption of Mount St. Helens, and ^{238}U-^{230}Th data for a suite of reference samples from the 1980–86 eruption. We focus here on whole-rock data for dacite dome and gouge samples for the 2004–5 eruption; we also present data for plagioclase separated from two dacite samples. These data complement other analyses by the Mount St. Helens petrology working group presented in this volume. We use these data to address the following questions: (1) Is the current eruption fed by a new batch of magma previously unseen at Mount St. Helens, and/or is this eruption tapping a remnant magma body from the 1980–86 eruption? (2) What is the relation of plagioclase phenocrysts to the host magmas in the 2004 dome samples?

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Background: U-Series Disequilibria in Older Mount St. Helens Lavas

U-series Disequilibria in Volcanic Rocks

U-series disequilibria in volcanic rocks have been used for decades as a tool for understanding magmatic processes (see reviews by Bourdon and others, 2003; Ivanovich and Harmon, 1992, and references therein). Disequilibria measured in whole-rock, glass, and mineral separates can be used both as tracers of chemically distinct magmas or crystals (for example, Cooper and Reid, 2003; Turner and others, 2003c) and to provide temporal and chemical information about magmatic processes ranging from melt generation (Bourdon and Sims, 2003; Lundstrom, 2003; Sims and others, 2003; Sims and others, 2002; Turner and others, 2003a) to magma residence and differentiation within the crust (Blake and Rogers, 2005; Condomines and others, 2003; Cooper and others, 2001; Hawkesworth and others, 2004; Reid, 2003; Reid and others, 1997; Rogers and others, 2004; Turner and others, 2003b; Turner and others, 2003c; Vazquez and Reid, 2004) to degassing and related crystal growth (Berlo and others, 2004; Reagan and others, this volume, chap. 37; Turner and others, 2004). A unique aspect of U-series studies is that they allow connection between chemical variations within crystals and absolute ages (for example, Vazquez and Reid, 2004).

U-series studies of magmatic processes are based on the fact that ^{238}U decays to ^{206}Pb through a series of intermediate daughter isotopes with half-lives ranging from less than one second to as much as 0.245 m.y. (^{234}U; Cheng and others, 2000). Any U-bearing system, given sufficient time as a closed system, will attain a state of radioactive or secular equilibrium where the rate of disintegration of each of the nuclides in the chain is the same. Magmatic processes, such as melt generation, crystallization, and degassing, can fractionate the different nuclides in the chain, creating disequilibria between individual parent-daughter pairs. Such disequilibria will return to radioactive equilibrium at a rate dictated by the half-life of the daughter isotope; thus the degree of disequilibrium preserved in erupted lavas contains information about the time since the last fractionation event, and each parent-daughter pair is sensitive to a different time scale. In particular, ^{230}Th-^{238}U disequilibria can record events that occurred within the past ~10,000 years to ~350,000 years, whereas ^{230}Th-^{226}Ra disequilibria are sensitive to time scales of a few hundred years to ~10,000 years. This range of hundreds to hundreds of thousands of years is commensurate with time scales of magma differentiation and storage in many systems (for example, recent reviews by Condomines and others, 2003; Hawkesworth and others, 2004; Reid, 2003; Zellmer and others, 2005), including that of older Mount St. Helens lavas (Cooper and Reid, 2003; Volpe and Hammond, 1991).

U-series Disequilibria in Older Mount St. Helens Lavas

U-series data in older Mount St. Helens lavas have been discussed previously (Bennett and others, 1982; Cooper and Reid, 2003; Volpe and Hammond, 1991), which provides an excellent context for our analyses of the 2004–5 samples. Volpe and Hammond (1991) analyzed ^{238}U-^{230}Th-^{226}Ra disequilibria in whole-rock samples, groundmass, and mineral separates for seven lavas erupted from Mount St. Helens within the past ~2,000 years (figs. 1*A*, 2*A*). Data for whole-rock and groundmass samples span an analytically significant range of ^{230}Th/^{232}Th ratios (fig. 1*A*), indicating that the different magmas had different source regions (Volpe and Hammond, 1991). These samples are also significantly enriched in ^{226}Ra compared to radioactive equilibrium with ^{230}Th (that is, (^{226}Ra)/(^{230}Th) > 1, where by convention parentheses around the chemical symbols represent activities) with (^{226}Ra)/(^{230}Th) ranging from 1.07 in an andesite of Castle Creek age (MSH 90-4) to 1.55 in a sample of dacite erupted in 1982 (MSH 90-9; fig. 2*A*). The preservation of these ^{226}Ra excesses requires that the time between the last fractionation event (likely to be melt generation) and eruption was less than about 10,000 years.

Volpe and Hammond's (1991) ^{238}U-^{230}Th data for mineral separates and whole-rock samples define linear arrays on a ^{238}U-^{230}Th isochron diagram, with slopes corresponding to apparent ages of 2–6 ka for most samples (fig. 1A). In contrast, two samples of Castle Creek age (erupted ~2 ka) yield apparent ages of 34–27 ka (Volpe and Hammond, 1991). At the same time, all of the mineral separates preserve ^{226}Ra-^{230}Th disequilibria indicating fractionation of Ra from Th within the past 10,000 years (fig. 2*A*), which is in conflict with the ^{230}Th-^{238}U apparent ages for Castle Creek samples. In detail, the ^{226}Ra-^{230}Th data for minerals and whole-rock analyses of the same samples do not define linear arrays on a Ba-normalized ^{226}Ra-^{230}Th isochron diagram (fig. 2*A*), which Volpe and Hammond (1991) interpreted as evidence of addition of a ^{226}Ra-enriched fluid to the liquid after crystallization of the minerals. Cooper and Reid (2003) reexamined Volpe and Hammond's data by using a new method of calculating ^{226}Ra-^{230}Th ages that accounts for impurities in the mineral separates and for differences in partitioning behavior of Ra and Ba. After applying this method, all of the Mount St. Helens data can be explained by crystallization and aging alone, without requiring late-stage addition of Ra to the system. All of the revised ^{226}Ra-^{230}Th ages are consistent with recent (less than a few thousand years) crystal growth, and, with the exception of the two Castle Creek samples, are concordant with ^{230}Th-^{238}U ages. ^{226}Ra-^{230}Th and ^{230}Th-^{238}U disequilibria return to secular equilibrium on different time scales; thus average ages for bulk separates with a protracted growth history will weight the old and young components differently for the two parent-daughter pairs, resulting in different average ages. Cooper and Reid (2003) interpreted the discordant ^{226}Ra-^{230}Th and ^{230}Th-^{238}U ages in the Castle Creek samples to reflect multiple phases of crystallization within the bulk mineral separate, either crystal populations of differing ages or old cores with younger overgrowths. In addition, plagioclase in the 1982 dacite (MSH90-9) has anomalously high Ra/Ba compared to what would be expected for equilibrium partitioning, which may reflect crystallization that was rapid enough to prevent chemical equilibrium between liquid and crystals (Cooper and Reid, 2003). These observations would predict that if the 2004–5 magmas were simply remnant magma from the 1980–86 eruption, then (1) whole-rock isotopic compositions (including (^{230}Th)/(^{232}Th)) would be identical in 2004–5 and 1980–86 samples, and (2) crystals within the 2004–5 dacite would have ^{238}U-^{230}Th-^{226}Ra characteristics like those in crystals erupted in the 1980s. Our new data for 2004–5 samples provide a test of these predictions.

Analytical Methods

Chemical separation of U-series elements and isotopic analysis by multicollector inductively coupled plasma mass spectrometry (MC-ICP-MS) were conducted at the University of Washington. Mass-spectrometry protocols and results for rock and solution standards are detailed in appendix 1. Chemical separation procedures were modified from published procedures (Goldstein and Stirling, 2003; Pietruszka and others, 2002) and are also detailed in the appendix. Briefly, the elements of interest were separated from the rock or mineral matrix using ion-exchange chromatography, conducted in class-100 laboratory facilities (low-particulate facilities, designed to minimize trace-metal contamination). Rock chips or mineral separates (0.5–1.5 g) were dissolved in concentrated hydrofluoric+nitric acids, followed by evaporation and dissolution in hydrochloric acid. Resulting sample solutions were split into three aliquots:

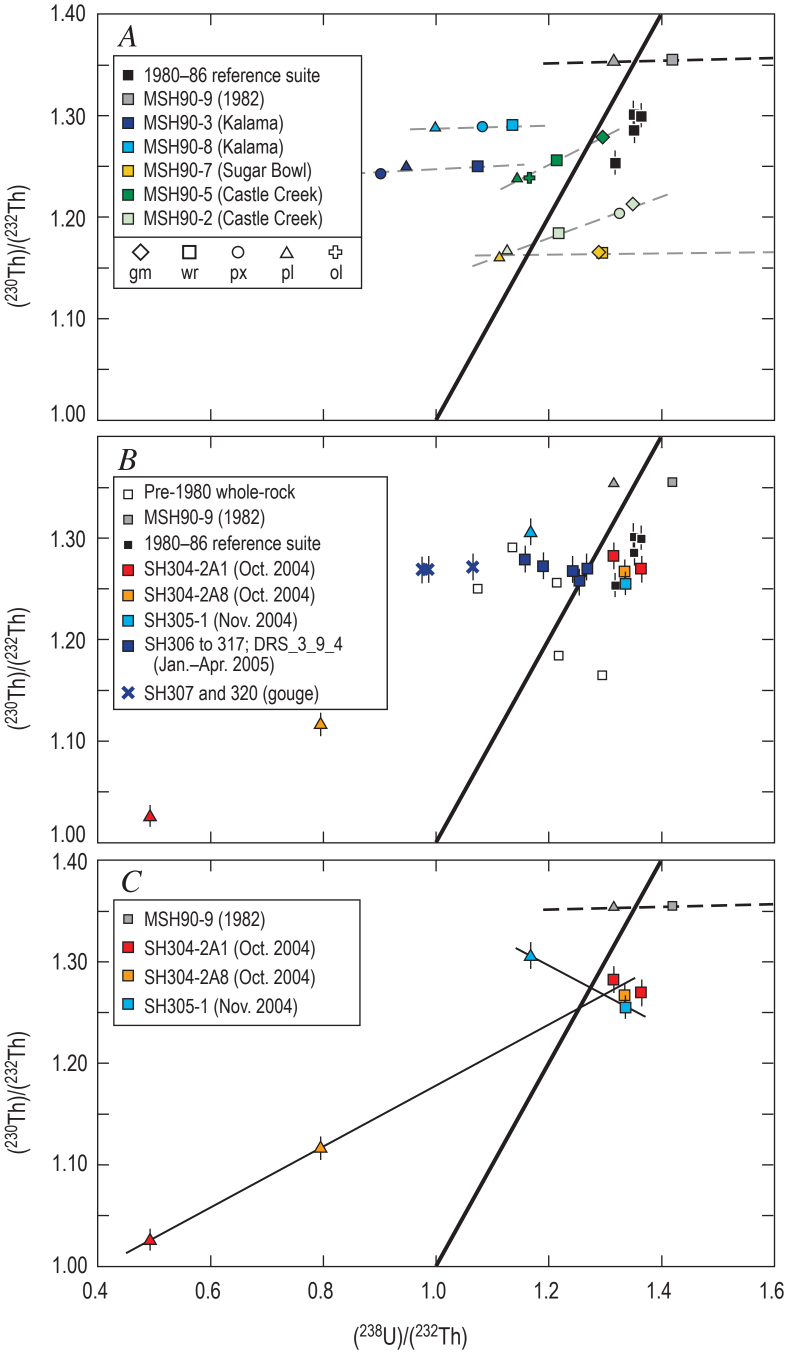

Figure 1. ^{238}U-^{230}Th isochron diagrams for Mount St. Helens samples. Solid bold line with slope 1 on all panels is the equiline (representing radioactive equilibrium). Thin solid and dashed lines are fits to the mineral-separate data for each sample. *A*, Pre-2004 samples. Black squares are analyses of 1980–86 samples from this study; error bars are shown for ±1-percent uncertainties. “MSH90-X” data are from Volpe and Hammond (1991); error bars for those samples are omitted for clarity but are similar in size to those shown for the 1980–86 reference-suite samples. Symbols for different phases are as follows: diamonds, groundmass (gm); squares, whole-rock (wr); circles, pyroxene (px); triangles, plagioclase (pl); cross, olivine (ol). *B*, Data for 1980–2005 samples. Shown for reference are pre-1980 samples (whole-rock data only; open squares) and 1982 dacite (MSH90-9; gray symbols; from Volpe and Hammond, 1991). Error bars for 2004–5 samples are shown only where larger than plotted symbol. Symbols for plagioclase and whole-rock data as in panel *A*; gouge samples indicated by “x” symbols. *C*, Whole-rock data and plagioclase separates for October 2004 dome (SH304-2A) and November 2004 dome (SH305-1). The 1982 dacite sample is shown for reference (Volpe and Hammond, 1991). Symbols as in panels *A* and *B*.

one small aliquot was spiked with ^{233}U and ^{229}Th for U and Th concentration measurement by isotope dilution, a second small aliquot was spiked with ^{135}Ba for Ba concentration measurement by isotope dilution, and the largest aliquot was spiked with ^{228}Ra and used for measurement of Ra concentration by isotope dilution and U and Th isotopic compositions. Uranium and thorium (both for measurement of isotopic composition and for isotope dilution) were separated from the rock matrix and from each other using Eichrom TRU™ resin. Radium was subsequently separated from the rock matrix washed from the TRU column of the Th and U isotopic-composition aliquot using cation-exchange resin and was purified using Eichrom Sr-spec™resin and a final cation-exchange microcolumn. Isotopic ratios were measured using a Nu Plasma MC-ICP-MS equipped with three ion counters and an energy filter on the high-mass ion counter for increased abundance sensitivity.

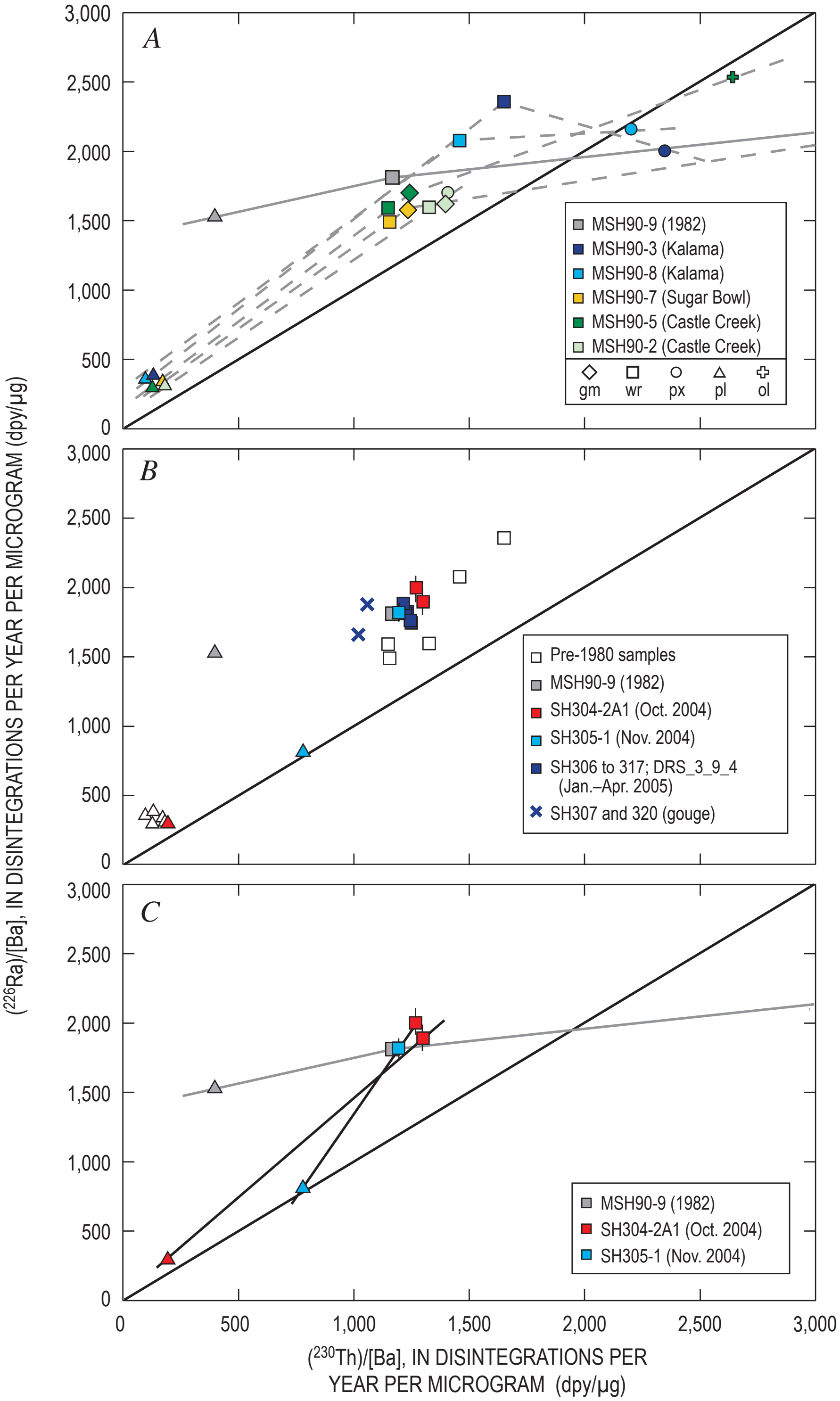

Figure 2. ^{226}Ra-^{230}Th-Ba isochron diagrams for Mount St. Helens samples. Solid line with slope 1 (the equiline, representing radioactive equilibrium) is shown on all panels. *A*, Pre-2004 samples. Symbols as in figure 1 and shown in the legend. *B*, Data for 1980–2005 samples. Pre-1980 samples (whole-rock and plagioclase data only; open symbols) are shown for reference. Error bars are shown where they are larger than the size of the symbols. *C*, Whole-rock data and plagioclase separates for October 2004 dome (SH304-2A) and November 2004 dome (SH305-1). The 1982 dacite sample (Volpe and Hammond, 1991) is shown for reference (gray symbols). Symbols as in panels *A* and *B*.

Results

^{238}U-^{230}Th Disequilibria

2004–2005 Dome Samples

^{230}Th-^{238}U disequilibria for 2004–5 Mount St. Helens dome dacites are within the range of those measured in older Mount St. Helens lavas (Volpe and Hammond, 1991; table 1, fig. 1*B*). (^{230}Th)/(^{232}Th) ratios measured in 2004–5 whole-rock dacite samples span a range of ~2 percent (from 1.257 to 1.282), analytically indistinguishable from each other given our reproducibility of ±1 percent. The dome samples show some variation in U/Th ratio, ((^{238}U)/(^{232}Th) = 1.159–1.366), resulting in a variation in ^{230}Th-^{238}U disequilibria from ^{238}U-enriched ((^{230}Th)/(^{238}U) = 0.929) to ^{230}Th-enriched ((^{230}Th)/(^{238}U) = 1.103). The majority of arc lavas are ^{238}U enriched, which is generally attributed to the influence of subduction-zone fluids, but ^{230}Th-enriched lavas have been observed in some arc settings (Turner and others, 2003a). It is also possible, given the presence of zircon in a sample of 2004–6 dome material (C. Miller, written commun., 2006), that some of the variation in U/Th ratios is due to differences in percentage of zircon and/or incomplete dissolution of zircon in different samples. No systematic decrease in U/Th ratio or ^{230}Th-^{238}U disequilibria over time is observed (not shown), although the two earliest samples (October–November 2004 lava spines) have the highest U/Th ratios and are the only samples that are ^{238}U-enriched.

1980–1986 Reference Suite

The four reference samples from the 1980–86 eruption have thorium isotopic compositions that overlap ^{230}Th/^{232}Th measured in the 2004–5 samples (fig. 1*B*). Three samples (SH32, SH131, and SH157, erupted in 1980, 1982, and 1984, respectively) have ^{230}Th/^{232}Th ratios at the high end of those measured in 2004–5 dome samples, though they overlap within measurement error. The 1986 sample has (^{230}Th)/(^{232}Th) ~4 percent lower than the other three 1980s samples, but it is within error of the lowest (^{230}Th)/(^{232}Th) ratio measured in 2004–5 dome samples. Our measurement of ^{230}Th-^{238}U disequilibria in SH131 (1982 dome) is not within error of the measurement of the 1982 dome sample (MSH90-9) reported by Volpe and Hammond (1991). This discrepancy could be due to thorium isotopic heterogeneity within the 1982 dome, or it could indicate that one of the measurements is in error. Thorium and U concentrations and Th/U ratios in the two samples of the 1982 dome are similar but not identical, which may support the former interpretation. For the purposes of this paper, we will focus on comparisons of 2004–5 samples with our measurements of the 1980s reference suite because they were performed in the same lab using the same methods.

Plagioclase Separates for 2004 Dome Samples

Plagioclase separates from two subsamples of dome rock erupted in October 2004 (SH304-2A1 and SH304-2A8) have U/Th ratios significantly lower than the whole-rock values for the same samples, qualitatively consistent with relative partitioning of U and Th in plagioclase (Blundy and Wood, 2003). ^{230}Th/^{232}Th ratios in the plagioclase separates are also low compared to the corresponding whole-rock values (table 1; fig. 1), and both plagioclase separates have large ^{230}Th-enrichments ((^{230}Th)/(^{238}U) = 2.087 and 1.404, respectively). A plagioclase separate from the dome erupted in November 2004 (SH305-1) is also ^{230}Th enriched ((^{230}Th)/(^{238}U) = 1.117), with U/Th ratio lower than the corresponding whole-rock measurement but higher than that in plagioclase from SH304-2A. The ^{232}Th/^{230}Th ratios in plagioclase and whole-rock samples of SH305-1 differ by about 4 percent, outside of analytical uncertainty. The U/Th ratios measured in plagioclase separates for the October 2004 dome (SH304-2A1 and SH304-2A8) are significantly lower than U/Th ratios measured by Volpe and Hammond (1991) in plagioclase separated from older Mount St. Helens samples, whereas plagioclase in the November 2004 dome sample (SH305-1) has U/Th ratio similar to those measured in plagioclase by Volpe and Hammond (1991). These differences in U/Th most likely reflect differences in the percentage or nature of impurities (groundmass and/or inclusions of glass or other mineral phases) in the plagioclase separates, although some part of the variation may reflect differences in U/Th of the magmas from which they crystallized.

2004–2005 Gouge Samples

The (^{230}Th)/(^{232}Th) ratios measured in gouge samples (SH307-2A and SH320-1) are within error of those measured in the 2004–5 dacites, although (^{238}U)/(^{232}Th) ratios are significantly lower than those measured in dome samples. Concentrations of both uranium and thorium are lower in gouge samples than in dacite samples (table 1). Sample SH320-1 also has lower barium and radium concentrations than do dome samples, although SH307-2A has barium and radium concentrations within the range of dome whole-rock samples.

^{226}Ra-^{230}Th Disequilibria

All whole-rock dome and gouge samples are enriched in ^{226}Ra relative to equilibrium with ^{230}Th, with ^{226}Ra excesses of 40–55 percent for whole-rock samples (similar to those measured by Volpe and Hammond (1991) for older whole-rock samples) and 60–77 percent for gouge samples (table 1; fig. 2*B*). Gouge samples have (^{230}Th)/[Ba] ratios lower than those of dacite dome samples, but broadly similar (^{226}Ra)/[Ba] ratios (fig. 2). (Brackets around the chemical symbol, by convention, indicate concentration.)

A bulk plagioclase separate from the October 2004 dome sample (SH304-2A1) has ^{226}Ra-^{230}Th-Ba characteristics similar to plagioclase separates from older Mount St. Helens samples, but it has lower (^{226}Ra)/[Ba] and (^{230}Th)/[Ba] than Volpe and Hammond's (1991) measurements of plagioclase in the 1982 dome (MSH90-9; fig. 2). The plagioclase separate for the November 2004 dome (SH305-1), in contrast, is within error of radioactive equilibrium ((^{226}Ra)/(^{230}Th) = 1.039), with significantly higher (^{230}Th)/[Ba] than any of the other Mount St. Helens plagioclase separates.

Discussion

Origin of the 2004–2005 Dacite: Remnant Magma from 1980–1986?

Whole-Rock Data

Major- and trace-element composition of the 2004–5 dacite whole-rock samples are similar to those of the 1980–86 dacite dome (for example, Pallister and others, 2005; this volume, chap. 30), suggesting the possibility that the cur-

Table 1. Concentrations, isotopic compositions, and activity ratios measured in Mount St. Helens samples.

[See appendix 1 for analytical details. WR, whole-rock; Pl, plagioclase. For duplicate sample names, #1 or #2 indicate chemical separation and analysis of splits of the same solution; "replicate" indicates separate dissolution and analysis of splits of the same rock or mineral separate. Accuracy and reproducibility of measurements was <0.5 percent for Th and U concentrations, <2 percent for Ba concentrations, <1 percent for ^{230}Th/^{232}Th, and <0.6 percent for ^{234}U/^{238}U (see appendix 1 for details). Uncertainties in Ra concentrations were limited by in-run errors during mass spectrometry, which are quoted here as 1 standard error (SE) of the mean (relative). Decay constants used in calculations were $\lambda_{234}=2.82629\times10^{-6}$ (Cheng and others, 2000), $\lambda_{230}=9.158\times10^{-6}$ (Cheng and others, 2000), $\lambda_{232}=4.933\times10^{-11}$, $\lambda_{238}=1.551\times10^{-10}$, and $\lambda_{226}=4.332\times10^{-4}$. Dashes indicate no data.]

Sample No.	Eruption date	Ba, in ppm	Ra, in fg/g	Ra %SE	Th, in ppm	U, in ppm	^{230}Th/ ^{232}Th (10^{-6})	(^{234}U)/ (^{238}U)	(^{238}U)/ (^{232}Th)	(^{230}Th)/ (^{232}Th)	(^{230}Th)/ (^{238}U)	(^{226}Ra)/ (^{230}Th)	(^{230}Th)/ [Ba], in dpy/µg	(^{226}Ra)/ [Ba], in dpy/µg
1980–1986 samples														
SH52	1980	--	--	--	2.049	0.911	7.010	--	1.353	1.301	0.962	--	--	--
SH131	1982	--	--	--	1.974	0.885	7.002	--	1.364	1.300	0.953	--	--	--
SH157	1984	--	--	--	2.390	1.063	6.928	--	1.353	1.286	0.950	--	--	--
SH226	1986	--	--	--	2.496	1.084	6.752	--	1.322	1.253	0.948	--	--	--
2004–2005 samples														
SH304-2A1 WR #1	10/18/04	338.6	555.0[1]	0.61	2.685	1.162	6.906	1.002	1.317	1.282	0.974	1.454	1,301	1,892
SH304-2A1 WR #2	10/18/04	332.2	575.1[2]	2.4	2.598	1.166	6.839	1.002	1.366	1.270	0.930	1.572	1,271	1,998
SH304-2A1 Pl #1	10/18/04	117.9	30.1[1]	1.6	0.175	0.028	5.529	1.004	0.492	1.026	2.087	1.514	195	295
SH304-2A1 Pl #2	10/18/04	116.4	--	--	--	0.029	5.528	1.001	--	1.026	--	--	--	--
SH304-2A8 WR	10/18/04	327.8	--	--	2.581	1.134	6.826	1.001	1.337	1.267	0.948	--	1,277	--
SH304-2A8 Pl	10/18/04	118.1	--	--	0.198	0.052	6.015	1.001	0.795	1.117	1.404	--	240	--
SH305-1 WR	11/20/04	330.9	521.2[1]	1.8	2.585	1.131	--	1.005	--	--	--	--	--	--
SH305-1 WR replicate	11/20/04	--			2.465	1.083	6.770	--	1.338	1.257	0.939	1.517[4]	1,198	1,818 [4]
SH305-1 Pl	11/20/04	113.4	79.8[1]	1.3	0.531	0.204	7.034	1.005	1.169	1.306	1.117	1.039	782	813
SH306	12/15/04	329.6	538.0[2]	0.43	2.464	0.965	6.854[3]	1.002	1.191	1.272	1.068	1.547	1,218	1,884
SH307-2A gouge		315.9	514.0[2]	1.4	2.065	0.663	6.836	1.003	0.977	1.269	1.299	1.768	1,062	1,878
SH307-2A gouge replicate		332.6	--	--	2.137	0.693	--	--	0.987	--	--	--	--	--
SH311-1B WR	01/16/05	327.9	496.3[1]	2.4	2.504	0.953	6.888	1.007	1.159	1.279	1.104	1.397	1,250	1,747
MSH05DRS_3_9_4 WR	03/08/05	326.9	515.0[2]	1.1	2.513	1.036	6.776	1.004	1.255	1.258	1.002	1.469	1,238	1,818
SH316-1A WR	04/15/05	331.5	505.7[2]	0.44	2.544	1.061	6.842	1.002	1.269	1.270	1.001	1.411	1,248	1,761
SH317-1A WR	05/01/05	331.5	520.2[2]	0.49	2.504	1.023	6.830	1.000	1.244	1.268	1.019	1.477	1,226	1,811
SH320-1 gouge		283.2	406.0[2]	1.2	1.783	0.625	6.849	1.000	1.066	1.271	1.193	1.615	1,025	1,655
Rock standards														
TML		--	3627 [1]	0.19	31.780	11.201	5.763	--	1.073	1.070	0.997	0.962	--	--
TML replicate		--	3707 [2]	1.3	31.119	10.944	5.806	1.002[5]	1.070	1.078	1.007	0.996	--	--
BHVO-1		--	163.3[2]	1.1	1.214	0.421	5.784	1.003[5]	1.057	1.074	1.016	1.130	--	--
BHVO-1 replicate		--	--	--	1.213	0.421	--	--	1.056	--	--	--	--	--
BCR-2		672.8	--	--	5.879	1.701	4.745	--	0.880	0.881	1.001	--	--	--

[1] Ra analyses by static, multi-ion-counting routine, as described in appendix 1.

[2] Ra analyses by dynamic routine using only IC0, as described in appendix 1.

[3] Average of two measurements of the same solution during the same day. Individual measurements differed by 0.9 percent.

[4] Calculated using Ra concentration measured for SH305-1 WR.

[5] (^{234}U)/(^{238}U) of rock standards represents the mean of eight measurements of the same solution over four analytical sessions. Relative standard deviation of replicate measurements was 0.17 percent for sample TML and 0.24 percent for BHVO-1.

rent eruption is tapping magma that remained in the reservoir after the 1980s. If the current eruption is tapping magma that is either unmodified from the 1980s or has been modified only by fractional crystallization, one prediction would be that isotopic compositions of the 1980–86 dacite would be identical to those in the 2004–5 dacite. In the case that the two magmas are related by cooling and fractional crystallization, but without significant influence of open-system processes such as wall-rock assimilation or mixing with compositionally distinct batches of magma, some variation in trace-element concentrations of the residual liquid would also be predicted. Kent and others (this volume, chap. 35) propose a model in which the 1980s and 2004–5 magmas are related by fractional crystallization that occurred primarily by chemical isolation of grain interiors from the host magma, without volumetrically significant gain or loss of plagioclase crystals, but with a small volume of an additional crystal component added to the magma through disaggregation of xenoliths. In this scenario, because crystals are not lost from or appreciably added to the bulk magma, whole-rock trace-element ratios would be unmodified during crystallization. In detail, however, a simple model of fractional crystallization would predict higher concentrations of incompatible elements (such as Ra, Th, U, and Ba) in the 2004–5 samples compared to the 1980–86 samples. Although Ra, Th, and U concentrations are indeed higher in the 2004–5 samples (table 1; Volpe and Hammond, 1991), Ba concentrations are lower, which suggests that a more complex explanation is necessary.

Nevertheless, the similarities between magmas erupted during the 1980s and the 2004–5 dacite (for example, major- and trace-element data in Pallister and others, this volume, chap. 30) suggest that the 2004–5 eruption is tapping at least some component of magma that remained in the reservoir after the 1980–86 eruption. The U-series data are consistent with this hypothesis but require some additional component(s) in the plagioclase populations (see below). Thorium isotopic compositions that we measured in whole-rock samples from 1980–86 samples span a small, but analytically significant, range (^{230}Th/^{232}Th ratios vary by ~4 percent), overlapping the thorium isotopic compositions that we measured in the 2004–5 samples, with (^{238}U)/(^{232}Th) similar to the October–November 2004 dome (SH304-2A1 and SH305-1; fig. 1*B*). Later-erupted 2004–5 samples have lower (^{238}U)/(^{232}Th), with no change in (^{230}Th)/(^{232}Th), suggesting that the 2004–5 magma has some trace-element heterogeneity, but that different magmas erupted are all closely related. ^{226}Ra-^{232}Th disequilibria are also similar in all 2004–5 and 1982 dacites, consistent with the similarity in thorium isotopic compositions. Interestingly, there appears to have been a slight decrease in ^{230}Th/^{232}Th ratios during the course of the 1980–86 eruption (fig. 1; table 1), suggesting that at least two magmas with different origins were involved in that eruption (and perhaps more, if the much higher ^{230}Th/^{232}Th measured in a 1982 dacite sample by Volpe and Hammond (1991) also reflects compositional heterogeneity).

Volpe and Hammond's (1991) measurements of (^{226}Ra)/[Ba] and (^{230}Th)/[Ba] in their sample of 1982 dacite do not overlap with our measurements of 2004–5 samples (fig. 2*B*). Whether there is significant heterogeneity within the 1980–86 and 2004–5 suites, as suggested above for ^{230}Th-^{238}U disequilibria, cannot be assessed at this time because we do not yet have ^{226}Ra data for the 1980–86 reference suite. However, the 2004–5 samples fall in a relatively tight cluster with the exception of the October 2004 dome (SH304-2A1), which has similar (^{226}Ra)/[Ba] but slightly higher (^{230}Th)/[Ba], possibly due to more plagioclase fractionation from this sample than seen in the other dacites.

Plagioclase Separates

When crystal compositions are considered in addition to whole-rock compositions, the 2004–5 magma must contain a component not observed in the 1980–86 magmas. The 2004–5 magmas contain plagioclase with compositions unlike those in older Mount St. Helens samples, which would require addition of crystals to a putative remnant 1980–86 magma. This is most dramatically illustrated by ^{230}Th/^{232}Th measured in plagioclase separated from the October 2004 dome (SH304-2A1), which is substantially lower than ^{230}Th/^{232}Th measured in any recently erupted samples of Mount St. Helens lavas (figs. 1*B*, *C*). Whether this lower ^{230}Th/^{232}Th ratio is attributed to an old component in the plagioclase separate or to the addition of a population of plagioclase unrelated to the host magma, the low-^{230}Th/^{232}Th component is not present to any significant degree in the plagioclase separate from the 1982 dacite measured by Volpe and Hammond (1991). Plagioclase in the November 2004 dome (SH305-1) has ^{230}Th/^{232}Th higher than the whole rock, also consistent with a plagioclase component that is foreign to the host liquid, although the foreign crystal component must be of a significantly different composition than that in SH304-2A1. In contrast to differences in their ^{230}Th-^{238}U signatures, the ^{226}Ra-^{230}Th-Ba composition of plagioclase in the October 2004 dome (SH304-2A1) is similar to plagioclase in samples erupted before 1980, but distinct from plagioclase in the 1982 dome sample (MSH90-9; fig. 2). In addition, the ^{226}Ra-^{230}Th-Ba signature of plagioclase of the November 2004 dome (SH305-1) is different from all measurements of plagioclase in older Mount St. Helens samples, including MSH90-9 (Volpe and Hammond, 1991) and SH-04-2A1 (fig. 2).

The presence of a foreign plagioclase component in the 2004–5 lavas is supported by Pb isotopic heterogeneity between plagioclase crystals and groundmass (Kent and others, this volume, chap. 35), by high Al in amphibole crystals that is not in equilibrium with the host liquid at pressures <300 MPa (Rutherford and Devine, this volume, chap. 31), by variable pressure-temperature conditions of crystallization of amphibole (Thornber and others, 2005), and by textures and zoning in plagioclase in the 2004–5 dacite that are distinct from the patterns observed in 1980–86 dacites (Streck and others, this volume, chap. 34). Therefore, the 2004–5 eruption clearly samples multiple crystal populations and, potentially, some associated liquid fractions that were not present in the eruptive products from 1980–86. However, given the similarity in whole-rock U-series compositions of the dacites, any liquid fraction associated with the crystals must be volumetrically minor, suggesting that the crystals were incorporated largely as a crystal mush or cumulate.

One potential scenario to account for these observations is that the 2004–5 magma is dominated volumetrically by remnant magma from 1980–86 but that it incorporated disaggregated crystal mush from within the reservoir system during the 18-year hiatus between eruptions, consistent with the presence of gabbroic inclusions within many of the 2004–5 dacite samples. In this case, the crystal mush must be genetically distinct from the 1980–86 magmas in order to explain the difference between (^{230}Th)/(^{232}Th) measured in the 2004–5 plagioclase separates and that measured in 1980–86 and 2004–5 whole-rock samples. It is also possible that the 2004–5 eruption is at least partially tapping new magma introduced into the reservoir system, perhaps associated with some of the deep earthquake swarms between 1986 and 2004 (Moran and others, 2005). Considering the similarity in major- and trace-element compositions of the 2004–5 dacite to dacites erupted earlier at Mount St. Helens, this new component may have originated through partial melting in the lower crust (see, for example, Pallister and others, 1992; Smith and Leeman, 1987). The range in (^{230}Th)/(^{238}U) and (^{226}Ra)/(^{230}Th) ratios observed in the 2004–5 samples is within the predicted range for incongruent dehydration melting of lower crustal amphibolite (Dufek and Cooper, 2005). However, we cannot rule out an origin through fractional crystallization of a garnet-bearing assemblage from a mantle-derived magma at lower crustal depths, as argued by Blundy and others (this volume, chap. 33). Finally, it is possible that our plagioclase separates contain a component of plagioclase from disaggregated xenoliths, either incorporated from Tertiary basement rocks or from the plutonic roots of the Mount St. Helens reservoir system (Kent and others, this volume, chap. 35; Pallister and others, this volume, chap. 30). In any case, the preservation of significant ^{226}Ra excesses in all of the 2004–5 whole-rock samples indicates that the time between melt generation and eruption for the bulk of the magma was short, less than one or two half-lives of ^{226}Ra (that is, less than a few thousand years).

Plagioclase in 2004–2005 Dacite: Effects of Impurities and Crystal Ages

Plagioclase in October 2004 Dome Samples

Plagioclase separates for the October 2004 dome subsamples (SH304-2A1 and SH304-2A8) have strikingly different thorium isotopic compositions than any of the 2004–5 whole-rock samples (fig. 1). Taken at face value, the slope of the two-point plagioclase-whole-rock isochron would correspond to an apparent crystallization age of ~42 ka. However, this apparent age is inconsistent with the preservation of ^{226}Ra excess measured in the same separates, precluding a simple history of crystal growth over a short time interval (less than a few hundred years) that occurred tens of thousands of years ago. Instead, this anomalously old apparent age likely indicates that the plagioclase separates include some percentage of older and/or foreign crystals in addition to plagioclase crystallized recently from the host liquid.

We estimate, on the basis of examination with binocular microscope, that the bulk plagioclase separate for SH304-2A1 is >95 percent pure. However, there are a number of components that could be present in small quantities in the bulk plagioclase separate, and the data plotted in figure 1 reflect the mixture of all of these; thus, in order to discriminate the effects of plagioclase aging from the effects of impurities, we must consider the U-series signature of each potential contributor. The bulk plagioclase separate could comprise a combination of plagioclase crystallized from the host liquid (at or near the time of eruption, and/or during storage in the crustal reservoir system); plagioclase that is foreign to the host liquid (either as a separate population of plagioclase crystals and/or as cores of crystals with multiple growth stages); groundmass adhering to the outside of plagioclase grains; and microscopic inclusions of melt or other phases within the plagioclase crystals. These in turn could include small amounts of the other major crystallizing phases, for example, amphibole, pyroxene, and Fe-Ti oxides (Pallister and others, this volume, chap. 30; Rutherford and Devine, this volume, chap. 31), and small mass fractions of accessory phases: apatite and zircon (C. Miller, written commun., 2006). Concentrations of Th, U, and Ra in pyroxene or Fe-Ti oxides will be low enough that a few percent of each would not significantly affect the ^{238}U-^{230}Th-^{226}Ra systematics of the bulk plagioclase separate (see also Blundy and Wood, 2003; Cooper and others, 2001); therefore, we will not consider them further. We discuss the U-series signatures and effects of each of the other components below.

Zero-Age Plagioclase

Plagioclase textures and rim compositions that are in equilibrium with the host liquid indicate that some fraction of plagioclase in each of the dome samples grew within a short time of eruption (for example, Pallister and others, this volume, chap. 30; Streck and others, this volume, chap. 34), at least partly as a result of decompression-induced crystallization that occurred during ascent of the 2004–5 magma (Blundy and others, this volume, chap. 33; Pallister and others, this volume, chap. 30). In addition, the lack of europium anomalies in REE patterns for whole-rock samples indicates that gain or loss of plagioclase to the magma must have been volumetrically minor (Blundy and others, this volume, chap. 33; Kent and others, this volume, chap. 35; Pallister and others, this volume, chap. 30), far less than the ~40 percent by volume of plagioclase crystals present in the magmas. In addition, plagioclase separates for 2004–5 samples have ^{210}Pb/^{226}Ra ratios that are not in radioactive equilibrium, indicating that some component of the plagioclase grew within decades of eruption (Reagan, 2005; Reagan and others, this volume, chap. 37). Therefore, the plagioclase separates certainly contain some plagioclase that crystallized from the host liquid at effectively zero age with respect to ^{238}U-^{230}Th-^{226}Ra disequilibria (that is, less than ~10 ka for ^{238}U-^{230}Th and less than a few hundred years for ^{230}Th-^{226}Ra). Such zero-age plagioclase would have ^{230}Th/^{232}Th equal to that in the host liquid, and ^{238}U/^{232}Th consistent with partitioning of Th and U between plagioclase and liquid at low pressure at ~850°C (based on oxide thermometry; Pallister and others, this volume, chap. 30; Rutherford and Devine, this volume, chap. 31). Blundy and Wood (2003) estimate D_U/D_{Th} in plagioclase to be ~0.18 at 900°C for An_{40}, similar to the temperatures and compositions appropriate for 2004–5 Mount St. Helens lavas. Using these data, we calculate (^{238}U)/(^{232}Th) in plagioclase in equilibrium with the October 2004 dome sample to be ~0.24 (fig. 3, table 2). Absolute values for D_U and D_{Th} are less well constrained, but by using $D_U = 6.0\pm3.7\times10^{-4}$ (measured in An_{58} plagioclase at 950°C; Blundy and Wood, 2003), we estimate Th and U concentrations of roughly 1–2 ppb in pure plagioclase (table 2). These calculations provide minimum estimates of Th and U concentrations in plagioclase in equilibrium with the liquid fraction of the magma, as the whole rock contains ~30–40 percent crystals and Th and U are incompatible in all of the major crystallizing phases. However, the Th/U ratio will likely be similar in the liquid and whole rock, and calculated (^{238}U)/(^{232}Th) is likely more robust to changes in mineral mode than are concentrations of U or Th. The bulk plagioclase separate, therefore, must contain some phase with (^{238}U)/(^{232}Th) higher than ~0.24 and (^{230}Th)/(^{232}Th) lower than that measured in the bulk separate (1.026).

Using the elastic-strain model and the latest fitting parameters for plagioclase (Blundy and Wood, 2003), we calculate (^{226}Ra)/[Ba] in plagioclase in equilibrium with a liquid of the October 2004 whole-rock composition to be ~330 dpy/µg (An_{40} at 850°C), and (^{230}Th)/[Ba] of ~1–2 dpy/µg (disintegrations per year per microgram). Calculated (^{226}Ra)/[Ba] is slightly higher than that measured in the bulk mineral separate, which could reflect some aging of the plagioclase since crystallization (as much as ~500 years), and/or the effects of other, low-Ra/Ba phases in the bulk separate. Th/Ba ratios could be significantly different in melt and whole-rock, considering that Ba is several orders of magnitude less incompatible than Th in plagioclase and amphibole, which dominate the crystal mode. Th/Ba in the liquid will therefore be higher than that in the whole-rock, leading to higher (^{230}Th)/[Ba] in plagioclase in equilibrium with the liquid. This is consistent with ion microprobe measurements of Th and Ba in plagioclase in older Mount St. Helens samples, where (^{230}Th)/[Ba] ratios were 10–50 for the 1982 dome and ~40–75 for older samples (Cooper and Reid, 2003). We estimate that (^{230}Th)/[Ba] in plagioclase crystallizing from the October 2004 magma is likely ~10–100, lower than the ratio of ~200 in the bulk separate.

Other Populations of Plagioclase

In addition to zero-age plagioclase crystallized from the host liquid, the bulk plagioclase separate may contain some older plagioclase, which would have lower (^{226}Ra)/[Ba] and (if older than ~5–10 ka) lower (^{230}Th)/(^{232}Th) than zero-age plagioclase. There are three possible origins for older crystals in the October 2004 dome plagioclase separate: (1) crystals that precipitated from the host magma and subsequently aged; (2) plagioclase crystallized from a different magma within the Mount St. Helens system (“antecrysts” in the terminology of W. Hildreth, as cited in Charlier and others (2005)); or (3) crystals incorporated from wall rocks unrelated to the Mount St. Helens magmatic system (xenocrysts). Gabbroic to dioritic xenoliths have been observed both in the 1980s eruptive products (Heliker, 1995) and in the current eruption (Kent and others, this volume, chap. 35; Pallister and others, this volume, chap. 30). Note that these are termed “inclusions” by Kent and others (this volume) and Pallister and others (this volume), but we use the term “xenoliths” instead to avoid confusion with microscopic inclusions within plagioclase crystals. Pallister and others (this volume, chap. 30) estimate the abundance of gabbroic to dioritic xenoliths in the 2004–6 dacite at 1–5 percent. They report ion microprobe U-Pb ages of 25 Ma for zircon separated from xenoliths in the 1980–86 lava dome, suggesting that the zircons are samples of Tertiary intrusions unrelated to the Mount St. Helens magmatic system. Kent and others (this volume, chap. 35) interpret the disaggregated xenoliths within the 2004–5 dome to have a similar origin. However, no xenoliths from the 2004–5 dome have been dated and they could, in theory, be of any age as old as the age of the basement rocks. In the case of Mount St. Helens, antecrysts could have ages as old as ~300 ka (Clynne and others, this volume, chap. 28), but older crystals (ranging back to Tertiary in age) would be xenocrysts. Regardless of origin, old plagioclase would likely have low (^{230}Th)/(^{232}Th) and low (^{238}U)/(^{232}Th), which makes it the only one of the likely components in the bulk plagioclase separate that could explain the low (^{230}Th)/(^{232}Th) measured in that separate (fig. 3*A*).

If the older plagioclase crystallized from the host liquid, it would have (^{238}U)/(^{232}Th) like that in zero-age plagioclase but with lower (^{230}Th)/(^{232}Th). Assuming that thorium concentrations are the same in old and young populations, we calculate that (^{230}Th)/(^{232}Th) of the bulk separate could be produced by a mixture of ~70 percent zero-age plagioclase with ~30 percent plagioclase in ^{238}U-^{230}Th radioactive equilibrium. The percentage of old crystals in the mixture would be higher if they were not in radioactive equilibrium; for example, a mix with 200-ka crystals would require ~40 percent old crystals, whereas a mix with ~100-ka crystals would require ~50 percent old crystals. Conversely, the percentage of old crystals may be lower if the thorium concentrations in the old crystals are higher, as suggested by the trace-element data for plagioclase interpreted by Kent and others (this volume, chap. 35) as disaggregated gabbroic xenoliths. For example, if the old crystals had thorium concentrations twice as high as the zero-age crystals, which may be reasonable given the factor of 2–4 enrichment in incompatible elements, such as Pb and La, relative to the zero-age crystals (Kent and others, this volume, chap. 35), the percentage of old crystals required would

Table 2. Calculated compositions of mineral phases in chemical equilibrium with Mount St. Helens sample SH304-2A1.

Phase	Ba, in ppm	Ra, in fg/g	Th, in ppm	U, in ppm	(^{238}U)/(^{232}Th)	(^{230}Th)/(^{232}Th)[1]	(^{230}Th)/(^{238}U)	(^{230}Th)/[Ba], in dpy/µg	(^{226}Ra)/[Ba], in dpy/µg
Zero-age plagioclase	193	56	0.001	0.001	0.24–0.41	1.270	3.10–5.29	1.1 [2]	332
Zero-age amphibole	34	4.4	0.010	0.005	1.37	1.270	0.929	51	154
Zero-age apatite (L)[3]	100	no data	4.57	2.12	1.41	1.270	0.899	7.5×10^3	0 [4]
Zero-age apatite (M&S)[3]	149	no data	4.16	3.03	2.22	1.270	0.572	4.5×10^3	0 [4]
Zero age zircon	1.3	0.0	43.3	117	8.20	1.270	0.155	5.3×10^6	0 [4]
2000 yr apatite (L)	100	373	4.57	2.12	1.41	1.272	0.901	7.5×10^3	4.3×10^3
2000 yr apatite (M&S)	149	226	4.16	3.03	2.22	1.287	0.580	4.6×10^3	2.6×10^3
2000 yr zircon	1.3	3.5×10^3 [5]	43.3	117	8.20	1.395	0.170	5.8×10^6	3.1×10^6

[1] (^{230}Th)/(^{232}Th) for zero-age phases assumed to be equal to that in whole rock.

[2] (^{230}Th)/Ba for zero-age plagioclase calculated from whole-rock composition; actual value may be somewhat higher in plagioclase in equilibrium with liquid (see text for discussion).

[3] Calculated using partition coefficients of Luhr and others, 1984 (L), and Mahood and Stimac, 1990 (M&S), respectively.

[4] (^{226}Ra)/Ba assumed to be zero for apatite and zircon at time of crystallization; this assumption makes little difference in the values calculated for older crystals.

[5] Ra concentration in 2,000-yr-old zircon calculated by assuming constant (^{230}Th), which gives a minimum value; because of the high U/Th ratio in zircon, ^{230}Th activity increases by ~10 percent over 2,000 years.

decrease by a factor of two. Kent and others (this volume, chap. 35) interpreted the lack of Eu anomalies in REE patterns in the whole-rock data for the 2004–5 dacite to indicate that only a small percentage of plagioclase can have been added to or lost from the magma, which would suggest that any older crystals present are near radioactive equilibrium (that is, hundreds of thousands of years old) and/or that they have high concentrations of thorium and presumably other trace elements. However, given the uncertainties in the thorium concentrations of different potential populations of plagioclase, coupled with the possibility that other phases are present in the bulk separates, we cannot uniquely determine whether the old crystals are older than ~300 ka and therefore must be xenocrystic.

Any old-plagioclase component, regardless of origin, with (^{230}Th)/(^{232}Th) lower than the bulk plagioclase separate is likely to be older than ~10 ka and therefore the ^{230}Th-^{226}Ra parent-daughter pair will be in radioactive equilibrium. If we assume that plagioclase in the bulk separate is a mixture of (1) plagioclase rims or crystals that grew immediately before or during eruption in chemical equilibrium with the host liquid and (2) an old plagioclase component (cores or crystals) that is >10 ka and therefore in ^{226}Ra-^{230}Th equilibrium, and if we further assume that both components have the same thorium and barium concentrations and (^{230}Th)/[Ba] ratios, we calculate that the bulk separate contains 25–40 percent old plagioclase (where the range encompasses variations in crystallization temperature from 850°C to 900°C and plagioclase from An_{50} to An_{40}). If the old component is <10 ka, or if the young component has (^{226}Ra)/[Ba] higher than predicted by equilibrium (a potential consequence of rapid crystallization (Cooper and Reid, 2003)), the proportion of old crystals would be higher.

However, such calculations are sensitive to assumptions about (^{230}Th)/[Ba] and barium concentrations in each population of plagioclase, and the mixing proportions are therefore only broadly constrained by this analysis. An additional complication is that radium concentrations in the bulk plagioclase separate will be sensitive to the effects of inclusions of other phases (for example, zircon, allanite, and groundmass) within the separate. Therefore, we do not attempt to interpret the percentage of old plagioclase crystals derived from ^{230}Th-^{226}Ra-Ba relations, except to note that the possible range in the percentage of old crystals is similar to the range that we calculated using thorium isotopic compositions.

Groundmass or Melt Inclusions

In addition to zero-age plagioclase and older plagioclase, some amount of groundmass or melt is likely to be present in the plagioclase separate as adherents to the outside of grains and/or as melt inclusions within plagioclase. The colinearity of the two plagioclase separates for subsamples of the October 2004 dome (SH304-2A1 and SH304-2A8) with the whole-rock data (fig. 1), together with higher concentrations of thorium, uranium, and barium in sample 2A8 compared to 2A1, suggests that SH304-2A8 plagioclase contains a higher percentage of included groundmass or melt inclusions. The difference in concentrations of thorium, barium, and uranium between the two subsamples is consistent with the presence of 1–2 percent more groundmass (with the composition of the whole rock) in SH304-2A8 plagioclase than in the separate from SH304-2A1, and it is possible that SH304-2A8 plagioclase itself contains a few percent by mass of groundmass.

The presence of groundmass will not affect the slope of the two-point plagioclase-whole rock ^{238}U-^{230}Th isochron and therefore will not change the apparent age of the plagioclase separate (~42 ka). As demonstrated by a comparison of the two plagioclase separates for the October dome samples, the presence of groundmass or melt inclusions will shift the bulk separate toward higher ^{230}Th/^{232}Th and ^{238}U/^{232}Th, and therefore the pure plagioclase will have ^{230}Th/^{232}Th and ^{238}U/^{232}Th even lower than those measured in the bulk separate (fig. 3). The presence of groundmass would also lead to higher (^{226}Ra)/[Ba] and (^{230}Th)/[Ba] in the bulk plagioclase separate compared to that in pure, zero-age plagioclase. This presence of groundmass is qualitatively consistent with the observations, but a mixing line on a ^{230}Th-^{226}Ra-Ba isochron diagram between zero-age plagioclase and the whole rock does not pass through the bulk separate (fig. 3) indicating that some other phase(s) must be present in the bulk separate.

Amphibole

Thorium and uranium are moderately incompatible in amphibole (LaTourrette and others, 1995; Tiepolo and others, 2000), and although the absolute value of the partition coefficients vary with SiO_2, the partitioning behavior of U is similar to that of Th in all compositions. Therefore, amphibole does not appreciably fractionate U from Th (Blundy and Wood, 2003, and references therein), and the effect of amphibole on the ^{238}U-^{230}Th systematics of the plagioclase separate will be similar to, but of a much smaller magnitude than, that of groundmass (fig. 3*A*).

Radium and barium are incompatible in amphibole, and D_{Ra}/D_{Ba} in amphibole is ~0.08 regardless of crystal composition (Blundy and Wood, 2003). Absolute values of D_{Ba} range from 0.10 to 0.72, similar to the range of D_{Ba} in plagioclase. Therefore, at the time of crystallization, amphibole would have (^{226}Ra)/[Ba] slightly lower than that in coexisting plagioclase, with (^{230}Th)/[Ba] similar to, or slightly higher than, that in plagioclase (fig. 3*B*). However, because of the

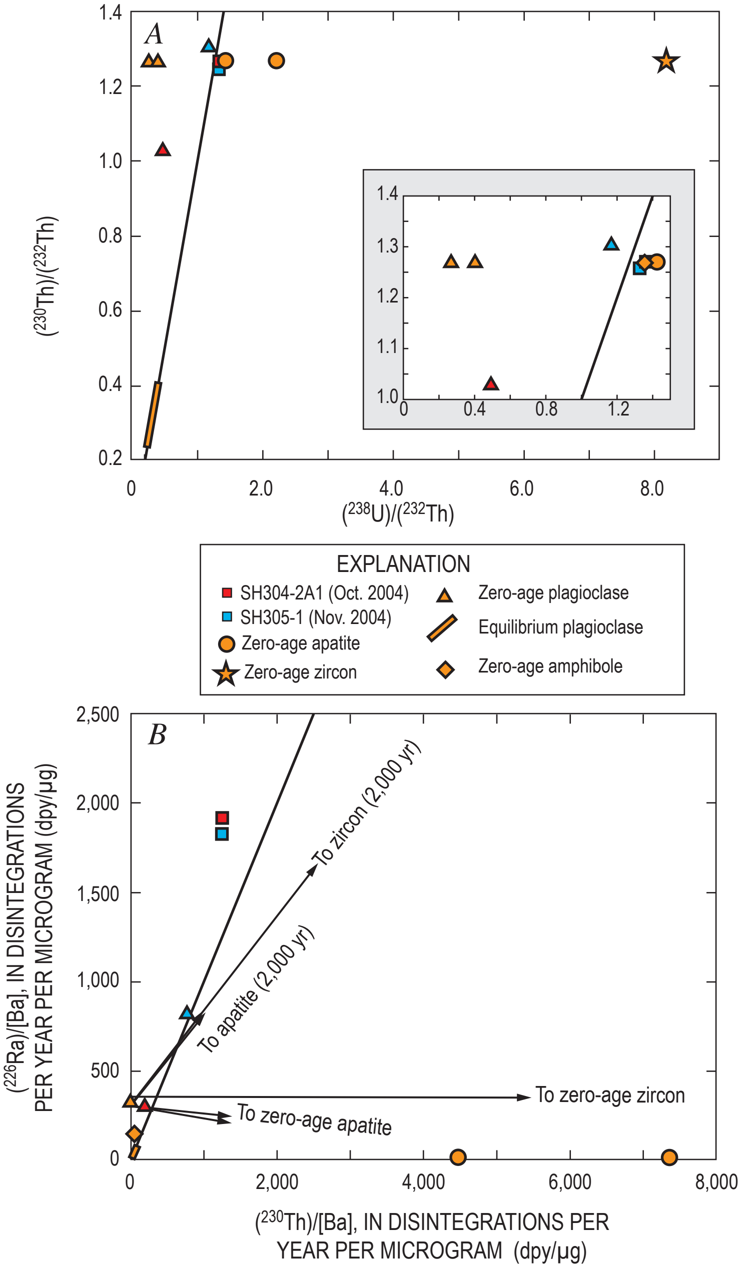

Figure 3. *A*, ^{238}U-^{230}Th; *B*, ^{230}Th-^{226}Ra isochron diagrams illustrating the effects of different types of impurities in the bulk plagioclase separates. Orange symbols are calculated values for zero-age apatite, zircon, plagioclase, and amphibole, in chemical equilibrium with October 2004 whole-rock (SH304-2A1) and plagioclase in radioactive equilibrium; for calculated values see table 2. The explanation applies to both panels; multiple symbols are shown for zero-age plagioclase and apatite where a range of partition coefficients are considered. The inset in panel *A* shows detail of upper-left corner of the larger diagram. Mixing trajectories on both diagrams will be straight lines; arrows shown in panel *B* illustrate mixing lines between zero-age plagioclase (orange triangle) and either zero-age or 2,000-year-old apatite and zircon. Length of arrows schematically indicates that (because of its higher concentrations of Th and U) zircon will have more leverage on the composition of the bulk plagioclase separate than will apatite.

similarity of the magnitude of radium and barium partition coefficients in amphibole and plagioclase, the effects of a few percent amphibole on the ^{226}Ra-^{230}Th-Ba systematics of the bulk plagioclase separate would be negligible.

Apatite and Zircon

Thorium and uranium are compatible in apatite and zircon, which results in orders of magnitude difference between thorium or uranium concentrations in plagioclase and in these accessory phases. Therefore, even small mass fractions of apatite and zircon could have significant effects on ^{238}U-^{230}Th-^{226}Ra disequilibria in the bulk plagioclase separate.

No experimental data exist for partition coefficients of the U-series nuclides in apatite or zircon. However, studies of coexisting apatite and glass in natural systems yield D_{Th}=1.6–1.7, with uranium somewhat more compatible (D_U=1.8–2.6; Luhr and others, 1984; Mahood and Stimac, 1990). Although some caution is warranted because these data were collected from trachytic to pantelleritic samples, the relative partitioning behavior when comparing two elements should be broadly similar in other compositions. This will lead to (^{238}U)/(^{232}Th) in apatite that is higher than that in the coexisting liquid at the time of crystallization (fig. 3*A*). On the basis of microbeam studies of natural systems, partition coefficients for thorium and uranium in zircon are higher than those for apatite, and D_U/D_{Th} will also be higher than in apatite (D_U ~100 and D_U/D_{Th} ~6; Blundy and Wood, 2003). Ion microprobe measurements of ^{238}U-^{230}Th disequilibria in magmatic zircon crystals confirm that (^{238}U)/(^{232}Th) is high, although the range of (^{238}U)/(^{232}Th) (~2 to >10, compared to ~1 for coexisting glass; for example, Bacon and Lowenstern, 2005; Charlier and others, 2003; Reid and others, 1997) is typically greater than predicted by equilibrium partitioning and may reflect local effects (for example, Bacon and Lowenstern, 2005). On the basis of partitioning data and previous measurements of magmatic zircon, (^{238}U)/(^{232}Th) in zircon coexisting with the October 2004 magma will likely be higher by a factor of 2–6 than that in the whole rock, and therefore zircon or apatite inclusions would elevate (^{238}U)/(^{232}Th) of the bulk plagioclase separate compared to that in pure plagioclase. The effect on (^{230}Th)/(^{232}Th) will depend on the age of the accessory phases; zero-age zircon or apatite will not change (^{230}Th)/(^{232}Th) compared to zero-age plagioclase, but because of their high (^{238}U)/(^{232}Th) ratios, both zircon and apatite will rapidly evolve to higher (^{230}Th)/(^{232}Th) with time. Therefore, addition of older zircon or apatite will increase (^{230}Th)/(^{232}Th) in the bulk separate. The only way that addition of zircon or apatite could explain the low (^{230}Th)/(^{232}Th) in the bulk separate would be if they were xenocrystic and had crystallized from a magma which itself had unusually low (^{230}Th)/(^{232}Th).

In detail, this scenario is difficult to reconcile with the position of the bulk plagioclase on a ^{238}U-^{230}Th isochron diagram (fig. 3*A*). Global compilations of U-series disequilibria in volcanic rocks (for example, Lundstrom, 2003, and references therein) show that all samples measured to date have (^{230}Th)/(^{232}Th) ratios above ~0.5. The high D_U/D_{Th} (and correspondingly high (^{238}U)/(^{232}Th)) for apatite and zircon means that even if they crystallized from a magma with (^{230}Th)/(^{232}Th) of approximately 0.5, a mixing line between pure plagioclase and these accessory phases would not pass through the point for the bulk plagioclase separate. Therefore, an additional phase or phases with low (^{230}Th)/(^{232}Th) and low (^{238}U)/(^{232}Th) (for example, old plagioclase crystals, as argued above) must be present in the bulk plagioclase separate. Even if apatite or zircon are present, they must make up a small percentage of the bulk plagioclase separate. A maximum of 1.4 percent or 0.025 percent (by weight) of apatite or zircon, respectively, could be present if we assume that all of the uranium in the bulk plagioclase separate resides in apatite or zircon; the presence of other impurities (such as groundmass) would decrease the allowable percentage of these phases.

Although apatite or zircon alone cannot be controlling the ^{238}U-^{230}Th disequilibria in the bulk plagioclase separate, small mass fractions of these accessory phases may be present and could influence the ^{230}Th-^{226}Ra disequilibria of the bulk separate. Barium is incompatible in both apatite (D_{Ba} <0.3; Luhr and others, 1984) and zircon (D_{Ba} = 0.003–0.005; Blundy and Wood, 2003), and Blundy and Wood (2003) estimate that D_{Ra} in zircon will be ~10^{-6}. Radium partitioning in apatite has not been measured or estimated, but it is likely to be more incompatible than barium, and both phases will likely have very low (^{226}Ra)/[Ba] at the time of crystallization, effectively zero for zircon. However, considering that thorium is compatible in apatite and zircon, both phases will have extremely high (^{230}Th)/[Ba] compared to coexisting plagioclase (or liquid), and they will evolve rapidly to high (^{226}Ra)/[Ba]. Mixing trajectories for zero-age plagioclase with zircon and apatite are shown on figure 3*B*; contamination with zero-age apatite could reproduce the low (^{226}Ra)/[Ba] of the plagioclase separate. Older apatite or zircon produce mixing trajectories that have slopes too shallow to pass through the data for the bulk plagioclase separate. The interpretation that apatite and zircon are not the dominant control on the ^{226}Ra-^{230}Th-Ba systematics of plagioclase is consistent with the ^{238}U-^{230}Th data, where apatite or zircon in small amounts could contribute to the U-series budget of the bulk plagioclase separate, but an additional component (in addition to zero-age plagioclase and zircon/apatite) is required to explain the data.

^{230}Th-^{226}Ra Age of October 2004 Plagioclase

The bulk plagioclase separate for the October 2004 dome (SH305-1) has (^{226}Ra)/(^{230}Th)>1, indicating that at least some mass fraction of the crystals measured are significantly younger than 10 ka. However, curves for the evolution of melt in equilibrium with plagioclase and curves for the evolution of the whole rock on a (^{226}Ra)/[Ba] evolution diagram for SH304-2A1 do not intersect, even when considering a range of preeruptive temperatures and plagioclase compositions and the potential effects of melt inclusions in the bulk separate (fig. 4). This pattern of data on an evolution diagram could be explained if the melt from which (at least some of) the plagioclase precipitated had significantly lower (^{226}Ra)/[Ba] than the SH304-2A1 whole rock, but such an explanation would require (^{226}Ra)/[Ba] < ~900 dpy/µg in this melt (if we assume some impurities in the bulk separate), lower than that observed in any recent Mount St. Helens lavas (see fig. 2). It is more likely that the plagioclase history and the effects of multiple types of impurities are too complex to allow a meaningful age determination by this method.

Composition of the October 2004 Plagioclase Separate

On the basis of the mixing relations outlined above, the plagioclase separate for the October 2004 dome is composed of at least three components: (1) zero-age plagioclase crystals, (2) older plagioclase crystals (whether xenocrysts or antecrysts), and (3) some groundmass or melt inclusions and/or a minute mass fraction of apatite or zircon. Groundmass, apatite, and zircon will act in a similar way on the ^{238}U-^{230}Th disequilibria, shifting the bulk plagioclase separate toward high (^{238}U)/(^{232}Th) and high (^{230}Th)/(^{232}Th) ratios. The volume of crystals must be dominated by young plagioclase, but there must also be some component of older plagioclase with low (^{230}Th)/(^{232}Th) in order to explain the low (^{230}Th)/(^{232}Th) measured in the bulk separate. The exact age of these foreign crystals cannot be uniquely determined; therefore, they may represent xenocrysts or antecrysts.

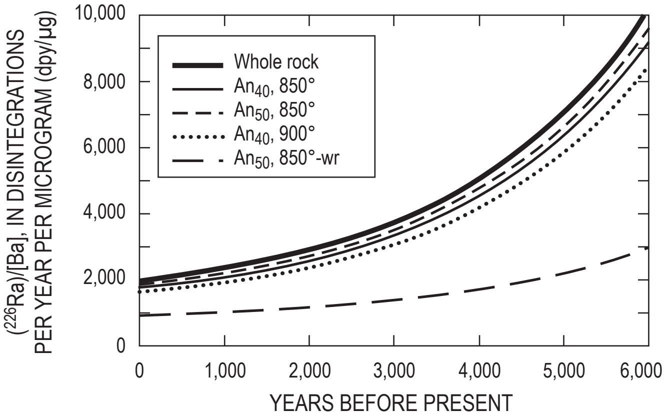

Figure 4. ^{226}Ra evolution diagram for October 2004 dacite (SH304-2A1). Curves shown are for evolution of (^{226}Ra)/[Ba] over time in whole rock (bold line) and for melt in equilibrium with plagioclase with composition of An_{40}–An_{50} at temperatures of 850–900°C. In a case where plagioclase crystallized rapidly within the past ~10,000 years in equilibrium with a liquid having the composition of the whole-rock, the intersection of curves for melt in equilibrium with plagioclase and whole rock would indicate the crystallization age (see Cooper and others, 2001, for details of the technique). The fact that curves for melt in equilibrium with plagioclase in SH304-2A1 do not intersect the whole-rock curve indicates that the bulk plagioclase separate and melt were not in equilibrium, even considering a range of potential crystallization temperatures and compositions of plagioclase. Correcting the plagioclase composition for the effects of having impurities (groundmass, zircon, or apatite) in the bulk separate makes the disequilibrium between melt and plagioclase more extreme. An example is shown by the long-dash curve, calculated assuming 3 percent whole-rock in the bulk separate, which is broadly consistent with concentrations of Th in the plagioclase being below detection limit for laser-ablation ICP-MS analyses (Kent and others, this volume, chap. 35). The lack of equilibrium between plagioclase bulk separate and the likely melt composition suggests that at least some of the crystals are foreign to the host liquid (see text for discussion).

Plagioclase in November 2004 Dacite

In contrast to the low (^{230}Th)/(^{232}Th) measured in the October 2004 plagioclase separate, the (^{230}Th)/(^{232}Th) ratio measured in plagioclase separated from the November 2004 dome sample (SH305-1) is approximately 4 percent higher than that in the host magma. As a result, a two-point plagioclase-whole-rock isochron has a negative slope, yielding a negative apparent age. This observation is inconsistent with crystallization of plagioclase solely from the SH305-1 host liquid, but (as with plagioclase in the October 2004 dome sample) it is consistent with the presence of impurities in the bulk plagioclase separate, which may include an antecrystic or xenocrystic plagioclase component. Interestingly, this foreign component must have higher (^{230}Th)/(^{232}Th) than zero-age plagioclase and, therefore, is compositionally distinct from that present in the October 2004 dome sample, despite the fact that the two dome samples were erupted only a month apart. The plagioclase separate for the November 2005 dome (SH305-1) also has higher (^{226}Ra)/[Ba] and (^{230}Th)/[Ba] than other plagioclase from Mount St. Helens samples (with the exception of Volpe and Hammond's (1991) measurement of MSH90-9; fig. 2), and those ratios are higher than those which would be in equilibrium with the host magma. As argued above for the October 2004 dome, it is likely that the bulk plagioclase separate for the November 2004 dome contains some zero-age plagioclase, and in addition, the separate could contain older plagioclase, zircon, apatite, amphibole, or groundmass in small percentages. The whole-rock October 2004 and November 2004 dome samples have similar U-series characteristics; therefore, the effects of zero-age impurities in the bulk plagioclase separate for the November 2004 dome would be similar to those shown in figure 3 for the October 2004 dome. In the case of the November 2004 plagioclase, the combined ^{238}U-^{230}Th-^{226}Ra-Ba data for the bulk separate are consistent with the presence of a small percentage of apatite, zircon and/or melt inclusions (fig. 3). The presence of less than 1 percent apatite or zircon (±melt) inclusions within the plagioclase separate could explain high (^{238}U)/(^{232}Th), (^{230}Th)/[Ba], and thorium concentrations in the bulk separate; and if the zircon or apatite were a few thousand years old, it could also explain the high (^{230}Th)/(^{232}Th) and (^{226}Ra)/[Ba]. The high (^{230}Th)/(^{232}Th) and (^{226}Ra)/[Ba] could still be consistent with the presence of some old plagioclase in the bulk separate, if offset by the presence of zircon or apatite, but the mixing proportions are difficult to determine because of the complexity of the data. Given the negative apparent ^{230}Th-^{238}U age and the near-equilibrium ^{226}Ra-^{230}Th-Ba data for the SH305-1 plagioclase separate, it is difficult to constrain the age of crystallization of any old component of plagioclase that may be in the bulk separate.

Implications for Magma Reservoir Processes

Age considerations aside, it is striking that plagioclase separates from the October and November 2004 dome samples have quite different U-series characteristics, despite having erupted only one month apart. Evidently, these two magmas entrained different populations of crystals, implying significant heterogeneity within the reservoir system or different transport paths intersecting different resident crystals.

In the case of the October 2004 dome, we can quantify the proportion of old and young crystals, but we cannot uniquely determine the age (and therefore the origin) of the old component. However, we speculate that the percentage of old crystals required (~15–50 percent of the plagioclase separate, depending on Th concentrations and age of the old component) might be higher than is easily explained by disaggregation of xenoliths that are present only in small amounts (1–5 percent of the bulk sample). Considering that 2004–5 dacites have a total of 30–40 percent plagioclase by volume, the assumption that all old crystals represent disaggregated xenoliths would imply that ~5–20 percent of the volume of the whole-rock dacite is composed of these xenocrysts. This, in turn, would imply that at least 1–5 times the volume of xenoliths that are observed has been completely disaggregated and incorporated into the dacite, which would likely produce some trace-element signature of plagioclase addition. Furthermore, we suggest that incorporation of plagioclase from a partially molten crystal mush perhaps may be physically easier than disaggregation of completely solidified xenoliths, and that an antecrystic origin is likely for at least some of the old crystals within the bulk separate. It is likely that there are at least three plagioclase components within the October 2004 dome sample: (1) zero-age plagioclase, (2) antecrystic plagioclase disaggregated from crystal mush, and (3) xenocrystic plagioclase disaggregated from gabbroic xenoliths. The age and origin of the old component(s) may be better constrained by analysis of different size fractions of plagioclase within the dome samples (which could preferentially sample different crystal populations), and work is currently underway to analyze different size fractions of plagioclase in SH304-2A1.

We have found evidence for a xenocrystic or antecrystic component in plagioclase separates in both of the samples from the 2004–5 eruption for which U-series disequilibria in plagioclase have been measured, as well as in at least two of the older Mount St. Helens lavas of Castle Creek age (Cooper and Reid, 2003). Thus, entrainment and disaggregation of crystal mush or xenoliths appears to be a common, if not ubiquitous, process in the Mount St. Helens reservoir system. Furthermore, the heterogeneity of this foreign component within different samples from the 2004–5 eruption suggests that the reservoir system feeding the current eruption is complex and not well mixed.

U-Series Disequilibria in Gouge

The exterior of the Mount St. Helens dome is coated with meter-thick fault gouge, which appears to consist mainly of fragments of dacite dome, as large as several centimeters, within a matrix of finer fragments (Cashman and others, this volume, chap. 19). Gouge samples have ^{230}Th/^{232}Th indistinguishable from that of dacite dome samples (fig. 1*B*), consistent with an origin for the gouge as being dominated by fragmented juvenile dacite. However, lower U/Th and lower U and Th concentrations in gouge compared to dome samples suggests either loss of U and Th (with preferential loss of U) from the gouge material or dilution of dacite fragments within the gouge with some material lower in U and Th concentrations than dome rocks. Similarly, gouge samples have lower (^{230}Th)/[Ba] than dome samples, but they have (^{226}Ra)/[Ba] similar to dome samples (fig. 2*B*), suggesting loss or dilution of Th rather than gain of Ba. Fluxing of gouge with a vapor or fluid could modify trace-element composition of the gouge either by precipitation of phases from the fluid or by leaching of fluid-mobile elements from gouge material. Fluids would be expected to carry U, Ra, and Ba, but not Th; thus, addition of a fluid-derived phase would be expected to add U, Ra, and Ba to the gouge rather than to decrease concentrations relative to the dacite. Leaching of dacite by a fluid could remove U, Ra, and Ba but could not explain the lower concentrations of Th in gouge. Therefore, the observations are most consistent with the presence of a component in the gouge (as yet unidentified) that has low concentrations of U and Th (and, in the case of SH320-1, low concentrations of Ra and Ba) compared to dome dacite.

Conclusions

Juvenile material from the 1980–86 eruption shows heterogeneity in ^{230}Th/^{232}Th ratios. The total range in ^{230}Th/^{232}Th measured in 1980–86 samples overlaps but is larger than the range of ^{230}Th/^{232}Th ratios measured in 2004–5 dacite and dome samples. However, the differences in crystal compositions between 1980s and 2004–5 samples indicates substantial heterogeneity of plagioclase crystals sampled during the two eruptions and even within a short time period during the 2004–5 eruption. This heterogeneity precludes an origin for the 2004–5 magma solely by remobilization of remnant magma from the 1980–86 eruptive cycle. Instead, the U-series data are consistent with a model in which the 2004–5 magma represents a mixture of some remnant 1980s magma with plagioclase crystals (and perhaps associated magma) having an isotopic composition previously unidentified in the erupted products of Mount St. Helens. The U-series data are consistent with an origin for the 2004–5 magma by partial melting of the lower crust, as has been proposed previously for Mount St. Helens dacites. The preservation of significant ^{226}Ra excesses in all measured whole-rock samples of the 2004–5 eruption indicates that the magma was generated recently, likely within the past few thousand years.

In detail, plagioclase separates for October 2004 and November 2004 dome samples (SH304-2A1 and SH305-1) have different U-series characteristics. This suggests that the two dacites, erupted only about a month apart, sample different populations of plagioclase crystals and/or different types of impurities within the bulk plagioclase separates. Plagioclase in both samples is also different in ^{230}Th-^{238}U disequilibria from that measured in older Mount St. Helens samples. Because of the complexities within the crystal populations sampled in the 2004–5 dacites, exact ages for plagioclase cannot be calculated. However, the preservation of ^{226}Ra excess in plagioclase from the October 2004 dome (SH304-2A1) requires that some mass fraction is younger than ~10 ka, and other petrologic constraints suggest that at least some crystallization occurred shortly before or during magma ascent. Simple mass balance calculations suggest that the percentage of a zero-age component is ~60–80 percent, although it could be lower if the older plagioclase component is <10 ka, and/or if the young component has (^{226}Ra)/[Ba] higher than predicted in equilibrium with the whole rock. The bulk plagioclase separate for the November 2004 dome (SH305-1) is within error of ^{226}Ra-^{230}Th equilibrium, and a young component (<10 ka) is not required by the ^{226}Ra-^{230}Th data. However, neither is it precluded by the data, and any older plagioclase is likely less than ~20 ka, if it crystallized from a magma with ^{230}Th/^{232}Th similar to that measured in Mount St. Helens whole-rock samples.

The combined whole-rock and plagioclase data for the 1980–86 and 2004–5 eruptions suggest that the crystal populations sampled by Mount St. Helens magmas commonly include a recycled component (whether antecrystic or xenocrystic) and that the magma-storage region beneath Mount St. Helens is complex, preserving chemical distinctions between magmas erupted only a short time apart. Thus, the crystals may contain a longer record of processes within the magma reservoir beneath Mount St. Helens than do the liquid fractions of these magmas. Detailed studies of the crystals may therefore provide insights into the temporal and spatial distribution of magmas and crystal mushes within the reservoir system.

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Appendix 1. Analytical Methods

Chemical Separations

Procedures for chemical separation were modified from those described by Goldstein and Stirling (2003) and Pietruszka and others (2002). Plagioclase separates were prepared using standard magnetic separation followed by hand-picking using a binocular microscope. Samples (0.5–1.5 g) of mineral separates and whole rocks were cleaned before dissolution using successive baths of distilled acetone, deionized, reverse-osmosis (RO) water, 0.1N HCl + 2 percent H_2O_2, RO water, and distilled acetone. Samples were dissolved in closed Savillex beakers on a hotplate using a 3:1 mixture of concentrated HF:HNO_3. After dissolution, perchloric acid and saturated boric acid were added and samples were evaporated. Samples were treated a second time with perchloric and boric acids, evaporated again, dissolved in 3 N HCl + saturated boric acid, and centrifuged. The solution was decanted, and any residue was dissolved in additional 3N HCl + boric acid (in the proportions 100 mL HCl:5 mL saturated boric acid); this process was repeated until no residue was visible after centrifuging. Sample solutions were split into three aliquots: one small aliquot (corresponding to ~10–100 mg of rock or mineral) was spiked with ^{233}U and ^{229}Th for U and Th concentration measurement by isotope dilution, a second small aliquot (corresponding to ~5–10 mg of rock or mineral) was spiked with ^{135}Ba for Ba concentration measurement by isotope dilution, and the largest aliquot (generally the remainder of the solution, corresponding to ~0.5–1.5 g of rock or mineral) was spiked with ^{228}Ra and used for measurement of Ra concentration by isotope dilution and of U and Th isotopic compositions. All resins used in the chemical separations were precleaned with nitric and hydrochloric acids and RO water, and they were also cleaned and conditioned on the column with the acids used for sample loading and elution.

For the U-Th-Ra aliquot, radium (with major and most trace elements) was separated from Th and U using a column packed with 0.5 mL of Eichrom “pre-filter” (inert beads) and 1–2 mL of Eichrom TRU resin. Samples were loaded in 2–8 column volumes (cv) of 7M HNO_3; the load, together with subsequent washes with 2 cv of 7M HNO_3 and 10 cv of 1.5M HNO_3, was collected and saved for further purification of Ra. This was followed by a wash of 8 cv 3N HCl, which was discarded. Th was eluted from the column in 8 cv 0.2N HCl, followed by U elution from the column in 8 cv 0.1N HCl + 0.05N HF. A second pass through a small TRU column (0.25 mL pre-filter plus 0.5 mL TRU resin) was necessary to remove residual Th from the U fraction.

The thorium and uranium isotope dilution aliquots were spiked with ~1 ng each of ^{229}Th and ^{233}U tracers. Spiked samples were equilibrated by heating for ~48 hours followed by evaporation. Some samples (1980s samples) were treated with 1–2 drops of perchloric acid during equilibration in order to aid sample-spike equilibration. Thorium and uranium were separated using a column loaded with 0.25 mL Eichrom prefilter and 0.5 mL TRU resin. A single pass through the column was generally sufficient to separate U and Th from the matrix.

The radium fraction from the TRU column was dried and redissolved in 1N HCl. Radium was separated from the major elements and most trace elements using a 5–10 mL column loaded with cation-exchange resin (Dowex AG 50-X8). After loading in 1–3 cv 1N HCl, the column was washed with 0–2 cv 1N HCl (adjusted so that the total load + wash was 3 cv), 5 cv 2.0N HCl, 1.5 cv 2.5N HCl, and 1.5 cv 3.0N HCl. The Ra (+Ba) fraction was eluted from the column in 8 cv 6N HCl. For samples larger than 0.5 g, a second, 0.5–1 mL cation-exchange column was run following the same procedure. Radium was separated from barium using a Teflon column packed with 0.45 mL Eichrom Sr-spec resin. The sample was dissolved in 2 cv 2.5N HNO_3 for loading on the column. After loading, Ba was washed from the column with 0.75 cv 2.5N HNO_3 and Ra was eluted from the column in 6 cv 2.5N HNO_3. This column procedure was repeated an additional one to two times, followed by passing the Ra cut through a column loaded with 0.5 mL of Eichrom prefilter before evaporating. A final cation column (0.25 mL) was run 1–4 days prior to mass spectrometry in order to separate ^{228}Th from ^{228}Ra. The sample was loaded on this column in 1 cv 6N HCl; radium was eluted in this fraction plus an additional 6 cv of 6N HCl while Th remained on the column.

The barium isotope dilution aliquot was spiked with a ^{135}Ba-enriched tracer, equilibrated on a hotplate for at least 48 hours, and dried. Barium was separated using a column loaded with 0.65 mL of cation exchange resin (Dowex AG 50-X8). The sample was loaded on the column in 0.65 mL 1.5N HCl (1 cv), followed by washes with 1.5N and 2N HCl. Barium was eluted in 2.5 N HNO_3.

Total-process blanks during the time that the Mount St. Helens samples were run were <20 pg Th and <15 pg U for the large (isotopic composition) aliquots, and <4 pg Th and <6 pg U for the small (isotope dilution) aliquots (compared to typical sample sizes of 0.1–2 µg Th and 0.02–1 µg U in the isotopic composition aliquots and 10–50 ng Th and 2–20 ng U in isotope dilution aliquots). Barium blanks were 5–10 ng, compared to typical sample sizes of 1–2 µg Ba. Radium blanks were always below the limit of detection (~0.1 fg), and sample sizes were a few tens of femtograms Ra for minerals and 200–500 fg Ra for whole-rock samples. Blanks for each measurement were subtracted before calculation of concentrations; in most cases these had a negligible effect on calculated concentrations, but there were small corrections to Ba concentrations.

Mass Spectrometery

All measurements of isotopic ratios for chemically separated samples were performed with a Nu Plasma multicollector-ICP-MS (MC-ICP-MS) at the University of Washington.

This instrument is equipped with three ion counters, one of which (IC0; the high-mass ion counter) is equipped with an energy filter to reduce tailing effects of major peaks. All analyses with the exception of Ba measurements were performed using a desolvating nebulizer (dry plasma), which improves sensitivity by a factor of ~10 compared to use of a peristaltic pump (wet plasma). Ba abundances were high enough that the desolvating nebulizer was unnecessary, and samples were introduced as a wet plasma.

Uranium Isotopic Composition Measurements

^{234}U/^{238}U ratios were measured using a static analysis routine, with ^{238}U measured on a Faraday cup and ^{234}U measured on an ion counter. Instrumental mass bias and ion counter-faraday gain were calibrated using NIST uranium standard CRM-112A with reference to the accepted ^{238}U/^{235}U and ^{234}U/^{238}U ratios of 137.88 and 5.286×10^{-5}, respectively (Cheng and others, 2000). Rock and solution standards analyzed as unknowns were interspersed with unknowns during a day of analysis, and re-calibrations of mass bias and gain were performed every third or fourth sample. Rock standards known to be in radioactive equilibrium reproducibly yielded (^{234}U)/(^{238}U) ratios within 4–6 per mil of 1.000 (see table 1), which we therefore consider to be our analytical uncertainty for unknowns.

Thorium Isotopic Composition Measurements

^{230}Th/^{232}Th ratios were measured using a static analysis routine with ^{232}Th measured on a Faraday cup and ^{230}Th measured on the high-mass ion counter equipped with an energy filter (IC0). At the beginning of each day of analysis of ^{230}Th/^{232}Th ratios, the voltage for the energy filter was tuned in order to optimize the abundance sensitivity, and the instrumental mass bias and ion counter-faraday gain were calibrated using NIST uranium standard CRM-112A. Abundance sensitivity was measured at 400–500 ppb at one AMU during the period of analyses. Correction for tailing of ^{232}Th into the ^{230}Th mass range was done using an exponential fit to the tail, which was measured each day using a Th solution standard (Th ‘U’ or WUN-1). Solution standards for Th measured as unknowns (Th ‘U’, WUN-1 (sometimes called ZSR), and IRMM-035) were generally within 1 percent of accepted or certified values (table 1). Rock standards (TML, BHVO-1, BCR-2), which went through the same chemical separation procedure as unknowns and were analyzed as unknowns, yielded (^{230}Th)/(^{238}U) within 1 percent of radioactive equilibrium, or within error of previously measured values (table 1). Therefore, we consider our measurement accuracy and reproducibility on unknowns to be ~1 percent.

Ba Concentration Measurements by Isotope Dilution

Isotopic compositions of sample-spike mixtures were measured using wet plasma and a static routine where all peaks were measured using Faraday cups. Ratios were normalized to the natural ratio of ^{136}Ba/^{138}Ba (=0.10954) to correct for instrumental mass bias. Normalized ^{137}Ba/^{138}Ba ratios were monitored, and they agreed with the natural ratio to within <0.5 percent for samples. NIST SRM 3104a (barium normal) was run as an unknown throughout the day and always yielded ^{137}Ba/^{138}Ba and ^{135}Ba/^{138}Ba ratios that were within 0.12 percent and 0.65 percent of the natural ratios, respectively. The barium spike concentration was calibrated with reference to NIST SRM 3104a, which has concentration certified to 0.3 percent at the 95-percent confidence level. Replicate spike calibration measurements agreed to within 0.3 percent. Measurement of USGS standard BCR-2 agreed with the accepted value within 1.6 percent and replicates of the same unknowns agreed within 2 percent. Therefore, measurement precision is better than ~0.5 percent, whereas reproducibility and accuracy is better than 2 percent.

U and Th Concentrations by Isotope Dilution

^{233}U/^{238}U and ^{229}Th/^{232}Th ratios were analyzed separately using static routines on Faraday cups. Mass bias was calibrated before analysis and several times over the course of a day of measurements using NIST CRM112A. ^{233}U and ^{229}Th spike concentrations were calibrated using Claritas™ Th and U standard solutions, certified to 0.5 percent, and calculations of concentrations in unknowns included corrections for total-process chemical blanks and evaporation corrections for the spikes. Accuracy was checked by analysis of rock standards BHVO-1, BCR-2 and TML. Our measurements of Th and U concentrations and (^{230}Th)/(^{238}U) of BHVO-1 agree within ~1 percent with published measurements of KIL1919 (collected from the same sample locality as BHVO; Pietruszka and others 2002); both are within error of the recommended value for Th concentration of 1.2±0.3 ppm for USGS standard BHVO-2 and are likely within error of the recommended value for Th concentration in BHVO-1 (of 1.1 ppm), although no error is reported for this value. Certified concentrations of U in BHVO-1 or BHVO-2 are not available. Rock standard TML is known to be slightly heterogeneous in Th and U concentration but to be in radioactive equilibrium; BCR-2 is a sample of a Columbia River Basalt flow erupted before 6 Ma and, therefore, should also be in radioactive equilibrium. Our analyses of TML and of BCR-2 yielded (^{230}Th)/(^{238}U) ratios within ~1 percent of equilibrium; therefore, the combined errors on measured U/Th and ^{230}Th/^{232}Th ratios must be better than 1 percent. Thus, accuracy of our measurements of U and Th concentrations must also be better than 1 percent.

Ra Concentrations by Isotope Dilution

^{226}Ra/^{228}Ra measurements were made using two different routines during this study. ^{226}Ra/^{228}Ra ratios in the first samples run (including SH304-2A1 whole-rock and plagioclase, SH305-1 whole-rock and plagioclase, and SH311-1B whole-rock) were measured using a static analysis routine in which

^{228}Ra and ^{226}Ra peaks were measured on ion counters IC0 and IC1, respectively. Mass bias and relative ion counter gains were calibrated using NIST CRM112A. After the initial group of samples had high background counts at mass 225.5, we began scanning all samples in the mass range 224.5–229.5 for interferences and found that backgrounds measured at masses 225.5 and 227.5 were significantly lower when using IC0 (which is equipped with an energy filter) than when using IC1. When using IC0, small peaks of ~10 cps are present in some regions of the mass range, but interferences and/or instrumental background at the half-masses used for background measurements were negligible (tenths of counts per second to a few cps, compared to count rates on peaks of hundreds of cps to thousands of cps for most samples). Furthermore, the ion counter gain for IC0 was more stable than that for IC1, so we switched to a dynamic routine where both peaks were measured on IC0. This also has the advantage that ion counter gain calibrations are less critical as long as gains are stable over the time scale of a sample run (~10–15 minutes). The disadvantages of this approach are that the total counting time for each peak is reduced and that the analyses are susceptible to intensity variations due to instability of the plasma; the latter effect was minimized by adding a small amount (a few tens of picograms) of ^{232}Th (as a solution prepared from Ames Th metal) to each unknown and normalizing Ra count rates to the intensity of ^{232}Th measured on a Faraday cup during the same cycle. This analysis routine produces better in-run precision (~0.5–1 percent SE, for peaks of ~200–3000 cps) than the initial routine (~2 percent SE). Concentration of ^{228}Ra in the spike was calibrated with respect to NIST SRM 4965, which has Ra concentration certified to 1.23 percent (3σ). (^{226}Ra)/(^{230}Th) measured in rock standard TML differs from equilibrium by less than 1 percent, and (^{226}Ra)/(^{230}Th) in USGS standard BHVO-1 is within ~2 percent of the previously measured value (Pietruszka and others, 2002). Taking these measurements and the in-run precision into account, we consider our overall uncertainty at the 2σ level to be ~3–5 percent for the early-run samples, and ~1–2 percent for the later samples.