

Time constraints on the origin of large volume basalts derived from O-isotope and trace element mineral zoning and U-series disequilibria in the Laki and Grímsvötn volcanic system

Ilya N. Bindeman^{a,*}, Olgeir Sigmarsson^{b,c}, John Eiler^d

^a Department of the Geological Sciences, University of Oregon, Eugene, OR 97403, United States

^b Laboratoire Magmas et Volcans, CNRS-Université Blaise Pascal, 63038 Clermont-Ferrand, France

^c Earth Science Institute, University of Iceland, 101 Reykjavik, Iceland

^d Geological and Planetary Sciences, California Institute of Technology, Pasadena, CA 91125, United States

Received 3 August 2005; received in revised form 2 February 2006; accepted 20 February 2006

Available online 5 April 2006

Editor: V. Courtillot

Abstract

The 1783–1784 AD fissure eruption of Laki (Iceland) produced 15 km³ of homogeneous basaltic lavas and tephra that are characterized by extreme (3‰) ¹⁸O-depletion relative to normal mantle. Basaltic tephra erupted over the last 8 centuries and as late as in November 2004 from the Grímsvötn central volcano, which together with Laki are a part of a single volcanic system, is indistinguishable in $\delta^{18}\text{O}$ from Laki glass. This suggests that all tap a homogeneous and long-lived low- $\delta^{18}\text{O}$ magma reservoir. In contrast, we observe extreme oxygen isotope heterogeneity (2.2–5.2‰) in olivine and plagioclase contained within these lavas and tephra, and disequilibrium mineral-glass oxygen-isotope fractionations. Such low- $\delta^{18}\text{O}_{\text{glass}}$ values, and extreme 3‰ range in $\delta^{18}\text{O}_{\text{olivine}}$ have not been described in any other unaltered basalt.

The energy constrained mass balance calculation involving oxygen isotopes and major element composition calls for an origin of the Laki–Grímsvötn quartz tholeiitic basaltic melts with $\delta^{18}\text{O}=3.1\text{‰}$ by bulk digestion of low- $\delta^{18}\text{O}$ hydrated basaltic crust with $\delta^{18}\text{O}=-4\text{‰}$ to $+1\text{‰}$, rather than magma mixing with ultra-low- $\delta^{18}\text{O}$ silicic melt. The abundant Pleistocene hyaloclastites, which were altered by synglacial meltwaters, can serve as a likely assimilated material for the Grímsvötn magmas.

The (²²⁶Ra/²³⁰Th) activity ratio in Laki lavas and 20th century Grímsvötn tephra is 13% in-excess of secular equilibrium, but products of the 20th century Grímsvötn eruptions have equilibrium (²¹⁰Pb/²²⁶Ra). Modeling of oxygen isotope exchange between disequilibrium phenocrysts and magmas, and these short-lived U-series nuclides yields a coherent age for the Laki–Grímsvötn magma reservoir between 100 and 1000 yrs. We propose the existence of uniquely fingerprinted, low- $\delta^{18}\text{O}$, homogeneous, large volume, and long-lived basaltic reservoir beneath the Laki–Grímsvötn volcanic system that has been kept alive in its position above the center of the Icelandic mantle plume. Melt generation, crustal assimilation, magma storage and homogenization all took place in only a few thousands of years at most.

© 2006 Elsevier B.V. All rights reserved.

Keywords: Laki; oxygen isotopes; diffusion; segregation; U-series disequilibria

* Corresponding author. Tel.: +1 541 346 3817.

E-mail address: bindeman@uoregon.edu (I.N. Bindeman).

1. Introduction

The presence, size, location and longevity of magma reservoirs are key properties of volcanic systems. These variables are constrained by geophysical observations such as seismic tomography [1] of active volcanic systems. Isotopic and trace elemental variations within and between individual crystals in igneous rocks provide valuable insights into timescales of magmatic processes on the order of days to hundreds of years which are unattainable by most other methods [2–4]. Short-lived radionuclides with half-lives comparable to those of magma segregation and differentiation provide complementary time constraints [5–7], and have previously been used to demonstrate surprisingly short timescales of generation and eruption of large volume magma bodies [5]. In this study, we use both approaches to discuss the longevity and origin of the basaltic magma from the Laki–Grímsvötn volcanic system in Iceland (Fig. 1).

The 1783–1784 eruption of Laki was the largest basaltic eruption of the last millennium, and is often compared to large igneous provinces ([8], and references therein). It produced a ‘dry fog’ of sulfate aerosols that severely impacted the Icelandic population and lowered temperatures in the N hemisphere for 2 years [8–11]. This magmatic system remains active today (Fig. 1), but its activity after 1784 is confined to the subglacial Grímsvötn caldera under Vatnajökull glacier, with one exception — the 1996 Gjálp eruption of somewhat more differentiated basaltic icelandite on the northern flank of Grímsvötn central volcano [12,13].

This paper has been motivated by the discovery that not only the 1783–1784 Laki basalt [14,15], but both older and younger tephra from the Grímsvötn are unusually depleted with respect to oxygen isotopes, down to 3‰, demonstrating that this low- $\delta^{18}\text{O}$ value persisted for at least 8 centuries. At the same time, the isotopically homogeneous glass contains mineral populations that exhibit extreme isotope disequilibria, not found elsewhere in any fresh igneous rock. These isotopic disequilibria and zoning allow estimation of the time these phenocrysts spent in contact with the $\delta^{18}\text{O}$ depleted basalt. Moreover, we measured ($^{226}\text{Ra}/^{230}\text{Th}$) and ($^{210}\text{Pb}/^{230}\text{Th}$) activity ratios in a few whole-rock samples in order to assess the timescales of magma formation. The timescales derived from oxygen isotope disequilibria between minerals and melt are compared with those inferred for the melt from ^{210}Pb – ^{226}Ra – ^{230}Th radioactive disequilibria. Finally, we discuss a new model for the origin of low- $\delta^{18}\text{O}$ basalts, such as the one from Laki.

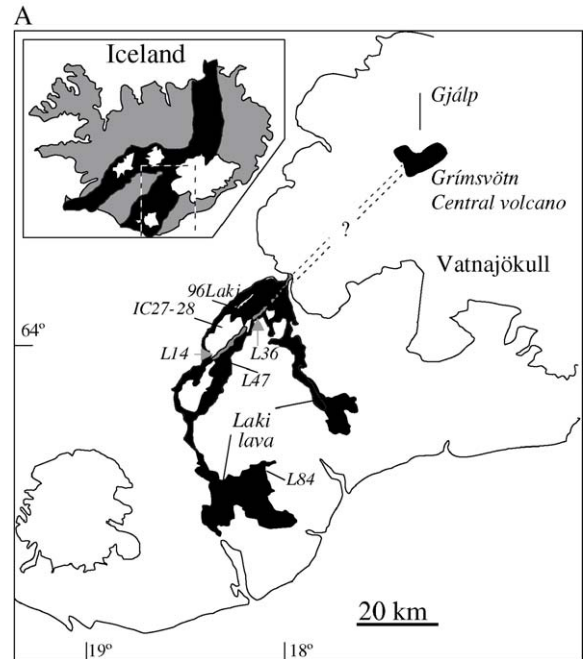


Fig. 1. A. Laki–Grímsvötn volcanic system, the 60 km-long fissures of 1783–1784 eruption extended progressively toward the subglacial Grímsvötn caldera that represents the central volcano of the Laki–Grímsvötn volcanic system. Sample localities for Laki lava and tephra, and hyaloclastites are shown (see Table 1). B. A field photograph of the Laki lava flow with Pleistocene hyaloclastites in the foreground. Notice tuffaceous character of the hyaloclastites.

1.1. Geological and petrological context of Laki–Grímsvötn magma system

The Laki–Grímsvötn magma system is located in the Eastern Volcanic Zone of Iceland, with the Grímsvötn central volcano sitting above the Iceland hotspot (Fig. 1). Frequent, small ($<1\text{ km}^3$) subglacial eruptions occur at Grímsvötn and many of these penetrate the glacier surface and deposit tephra, which records the temporal variations

of the underlying magma system ([16] and references therein). The most important Holocene eruption of this volcanic system is the 1783–1784 Laki fissure eruption that produced 14.7 km³ of basaltic lava and 0.4 km³ of basaltic tephra [17].

The Pleistocene volcanic activity in the area formed the basement rocks that are present under Laki fissures and presumably under the Holocene edifice of Grímsvötn volcano in the form of poorly-dated hyaloclastites [18] that are formed in response to subglacial basalt–ice interaction (Fig. 1B). These are highly fragmented and often permeable tuffaceous materials that form the basement of the Table Mountains such as the well-studied Kistufell, 40 km north of Grímsvötn caldera [19], much of the Grímsvötn itself, and the basement of Laki Fissures (Fig. 1B). The hyaloclastites contain palagonitized basaltic glass with depleted $\delta^{18}\text{O}_{\text{glass}}$ values, but intact phenocrysts ([19]; our data below).

Uniform compositions of basalts from Grímsvötn and Laki suggest a common origin of magma for this volcanic system [14]. However, two mutually exclusive models have been proposed for the connection between Grímsvötn central volcano and Laki fissure system. Large volume fissure eruptions in Iceland may tap rather shallow magma reservoirs located under associated central volcanoes, migrate laterally and erupt far away from the central volcano (e.g., 20). An alternative explanation is that basaltic dikes, parental to fissures, rise directly from homogeneous magma accumulation zones deeper in the crust or crust/mantle boundary and erupt vertically. In this case the deep magma reservoir would be responsible for magmatic behavior of the whole volcanic system ([14,17] and references therein). The association of fissures with central volcanoes (covered by glaciers) is typical for other centers in the Eastern Volcanic Zone of Iceland: where at least 3 other centers: Eldgja-Katla, Veidivotn-Bardarbunga, Raudholar-Thordarhyrna that exhibit similar relationships. Both models require delicate magma plumbing system under each volcanic system, each fingerprinted by the unique and persistent $\delta^{18}\text{O}_{\text{melt}}$ values over time, despite the small distance between the adjacent fissures (such as 9 km between Laki and Eldgja, that have unique $\delta^{18}\text{O}_{\text{melt}}$ values +3.1‰ Laki, and $\pm 4.5\%$ Eldgja [15] and our unpublished results).

The eruptive sequence, petrography, phenocryst chemistry, and isotopic ratios of Laki eruption products have been studied for more than three decades. Compositionally, Laki products are predominantly quartz-tholeiitic basaltic lava (95%) and tephra (~ 5%) ranging in crystal (1–10%) and vesicle contents [9,10,14,17]. Phenocrysts include plagioclase, olivine, and clinopyroxene,

and lack Fe–Ti oxides. Mineral thermometry and crystallization experiments suggest eruption temperatures for Laki lavas and tephra of ~ 1140–1160 °C [10,21,22].

Grímsvötn tephra tend to be nearly aphyric to phenocryst-poor, with only the basaltic icelandites of 1996 Gjalp eruption having almost 5% phenocryst by volume [12,13]. The only compositional difference between Laki and Grímsvötn is slightly higher U and Th concentrations in 20th century tephra from the latter [13].

2. Samples and methods

For the Laki eruption, two samples of lava (96-Laki, and L14), and two samples of tephra (L36 and M4) were studied in detail (Table 1, Appendix). Five tephra samples of the approximate dates 1200, 1450 and 1500 AD were collected from the ablation region of Vatnajökull ice sheet ([16]; and Larsen and Gudmundsson personal communication), 1996, 1998 and 2004 AD from Grímsvötn volcano were collected from air-fall deposits during or soon after the corresponding eruptions. These samples allow the assessment of compositional variability of Grímsvötn magma during the last 8 centuries. Additionally, we collected two samples of Pleistocene hyaloclastites intersected by Laki fissure.

Phenocrysts of olivine, plagioclase, and clinopyroxene were picked up individually from pre-cut thin slabs of rock, this allowed us to record their size, agglomeration, and textural position of these phenocrysts in the rock. Individual grains usually broke during extraction, and the fragments were separated from adjacent glass under a binocular microscope and placed into separate sample vials for oxygen isotope and electron microprobe analyses. In this way we were able to analyze grains individually and in size groups; the latter applies to olivine microphenocrysts (0.05–0.2 mg). Larger crystals (> 1 mg) of olivine and plagioclase were broken into cores and rims, where possible. Additionally, a subset of selected grains > 0.3 mg was mounted into the crystal bond, polished, and analyzed by the electron microprobe; the exact same grains were later analyzed in compositional groups (Fo content of olivine and An content of plagioclase) in order to precisely assess correlation of composition with $\delta^{18}\text{O}$ values.

Oxygen isotope analyses were performed by CO₂ laser fluorination at Caltech [23,24], and the University of Oregon. Analyzed mineral aliquots were typically 0.5–1.5 mg, yielding 8–30 μmol of CO₂; yields were 90–102%, and there is no correlation between yield, sample size, and $\delta^{18}\text{O}$. Minerals and glass samples were pre-treated with 2 Torr of BrF₅

Table 1

Oxygen isotope analyses of glass and individual and bulk phenocrysts in basalts of 1783–4 Laki eruption and preceding and postdating tephra from the subglacial Grímsvötn caldera (Iceland)

Material, composition	$\delta^{18}\text{O}$ (‰)
<i>Sample GR1200, 1200 AD tephra</i>	
Glass	3.07
Glass	2.94
<i>Sample GR1450, 1450 AD tephra</i>	
Glass	3.24
Glass	2.94
<i>Sample GR1505, 1500 AD tephra</i>	
Glass	3.04
Glass	3.05
<i>1783–1784 Lakagigar fissure basaltic eruption</i>	
<i>Sample M4, tephra</i>	
Glass	3.12
Glass	3.17
<i>Sample L36, tephra</i>	
Glass	3.20
Glass	3.11
Olivine, bulk	2.91
Olivine, batch-1, Fo78	3.97
Plag, small crystals	2.84
Plag, 1 large crystal	3.13
Plag, 1 large crystal	3.21
<i>Sample L14, lava</i>	
Glass	3.10
Glass	3.00
Olivine, bulk	2.93
Olivine, Fo74.5, bulk	3.19
Olivine, Fo78, core	3.53
Plag, 1 large crystal	3.37
Clinopyroxene, bulk	2.52
<i>Sample 96-Laki, lava</i>	
Olivine, 1 large crystal	3.18
Olivine #5, Fo75.5, large, core	3.89
Olivine, Fo75.4, small crystals	2.12
Olivine, core	4.71
Olivine, cluster 4-2, small, bulk	2.27
Olivine #6, small	2.54
Olivine #4, Fo85, large, core	4.05
Olivine #4, large, rim	4.06
Olivine #10 small crystals, cluster	4.44
Olivine #7, Fo85, large, core	3.61
Olivine 1+2, Fo74.5, 2 Crystals	3.41
Olivine, bulk	3.06
Olivine, Fo77.9±1.1, bulk	4.81
Olivine, Fo74.9±1.2, bulk	4.38
Olivine, Fo72.0±1.9, bulk	5.23
Plag, An80.5±1.7	3.20
Plag, 1 Crystal, An87, large, core	3.28
Plag, 1 Crystal, slab-shape	3.17
Plag, An75, bulk	2.99
Clinopyroxene, bulk	2.86

Table 1 (continued)

Material, composition	$\delta^{18}\text{O}$ (‰)
<i>Sample 96Gjálp, 1996 AD tephra</i>	
Glass	2.79
Glass	2.61
Plag #1	3.18
Plag #2	4.75
Plag #3	3.41
<i>Sample G1998, 1998 AD tephra</i>	
Glass	3.36
Glass	3.11
<i>Sample G2004, 2004 AD tephra</i>	
Glass	3.12
Glass	3.04
Plag, bulk	3.36
<i>Sample IC-27, Pleistocene hyaloclastites</i>	
Glass	3.26
Glass	3.23
<i>Sample IC-28, Pleistocene hyaloclastites</i>	
Glass	2.78
Glass	3.30
Plag, 5 Crystals	3.80
Olivine #1	4.11
Olivine #2	4.52
Olivine #3	3.47

overnight to remove any surface or water contamination. The CO_2 laser fluorination yielded precise ($\pm 0.05\%$, 1st dev) analyses, used BrF_5 reagent, and conversion to CO_2 gas following fluorination. Two to four garnet standards (GMG-2, 5.75‰), and two to five olivine standards San Carlos olivine (5.35‰) were used in the beginning and during each analytical session, and both $\delta^{18}\text{O}$ and $\Delta^{18}\text{O}$ (garnet-olivine) were monitored to ensure no introduced fractionation as a function of mineral chemistry; day-to-day $\delta^{18}\text{O}$ variations on standards were typically $\pm 0.1\%$, and analyses were corrected using values of olivine standard. Thus, the overall analytical uncertainty on single measurements is better than $\pm 0.1\%$ (1st dev).

Electron microprobe measurements of minerals, glass, and melt inclusions were performed at Caltech and followed standard routines (1 μm , 15 kV, 10 nA); for analyses of feldspar and glass the electron beam was rastered to 6 μm^2 . Analyses are available as the electronic supplement to this paper.

The activity of ^{226}Ra and ^{230}Th was measured by thermal ionization mass spectrometry (TIMS) in Clermont Ferrand as described in Sigmarsson et al. [25], whereas that of ^{210}Pb was analyzed by alpha-spectrometry via its progeny, ^{210}Po .

3. Results

3.1. Mineralogy, mineral compositions, and crystallization modeling

Phenocrysts in tephra and lavas from Laki include plagioclase (0.3 to 1.5 mm), olivine (0.1 to 1 mm), and clinopyroxene (0.2–1 mm). The compositions of groundmass glass and mineral-hosted melt inclusions range from quartz- to olivine-tholeiite, the latter being much less abundant and characteristic of melt inclusions in forsterite-rich olivines ([10,21], this study). It is not straightforward to petrographically distinguish “phenocryst” and “xenocrysts” in Laki volcanics, see discussion below. We consider “phenocryst” to be in compositional equilibrium with the Laki glass which could have been achieved either through protracted residence and exchange, or through initial crystallization from the host quartz-tholeiitic magma. “Xenocryst” are those that contain highly forsteritic ($>F_{080}$ olivine), and anorthitic ($>An_{80}$, plagioclase), those crystals had little time to equilibrate in the host magma prior to its eruption and could have been pre- or syneruptively captured; many xenocrysts brought up to surface during phreatomagmatic explosions for example, but these samples are not considered in this work.

In the samples of this study, plagioclase range from An_{90} to An_{75} , olivine from F_{086} to F_{072} , only very thin rims (1–3 μm) on olivine reach F_{068} , and clinopyroxene is augitic with $Mg\# = 0.79$. Most studied olivines are weakly zoned to unzoned with respect to Fe–Mg, Ni, Mn, and Ca. Plagioclase is zoned and can be classified into two main types: normally-zoned and complexly zoned. The latter includes Ca-rich core and Ca-richer intermediate zone within crystals surrounded by the Ca-poorer rim. Phenocrysts commonly occur as aggregates that suggest olivine–olivine + plagioclase–olivine + plagioclase + augite crystallization sequence. Plagioclase aggregates are often complexly zoned and show evidence of either breakage/deformation during growth, or very complex co-crystallization, synneusis and twinning. These features are consistent with crystallization near chamber’s walls or in the crystal mush (e.g., [26]). Mineral composition, zoning, and texture do not produce an impression of mixing between compositionally diverse magmas, such as water-bearing dacite or rhyolite and basalt, similar to what is observed at Hekla and other localities [24,27,28].

The MELTS [29] program was used to calculate liquid lines of descent of a typical olivine tholeiite composition that is traditionally considered parental to the Icelandic quartz tholeiites (e.g., [30,31]). The

olivine tholeiite composition that we used is similar to that of primitive melt inclusions within F_{086} olivine and An_{85-89} plagioclase xenocrystic cores of Laki phenocrysts [21].

The liquid line of descent matches the observed identity, composition, sequence of appearance, relative proportion of crystallizing minerals ($Plag > Ol > Cpx$), and their compositional trends in Laki and suggests predominantly $Ol-Pl$ (F_{083-75} , An_{82-75}) fractionation. The crystallization modeling is relatively insensitive to the initial small variations in major element contents and intensive parameters, including ~ 1 order of magnitude variations in H_2O content (0.08 to 0.5 wt.%), pressure: 2 to 0.5 kb, and oxygen fugacity: $QFM \pm 1$ log unit. Fig. 2 demonstrates that both equilibrium and fractional crystallization yield satisfactory results, but require pressure small enough to match the sequence of appearance and compositions of phases, as clinopyroxene appears before olivine at greater pressure. Overall, this MELTS model provides robust constraints on phase equilibria that could generate Laki-like compositions at shallow pressures and accounts for variations in major element compositions in Laki glass and glass inclusions; it will be used for the subsequent AFC modeling (see below). However, the model does not account for the observed trace element variations in REE spectra

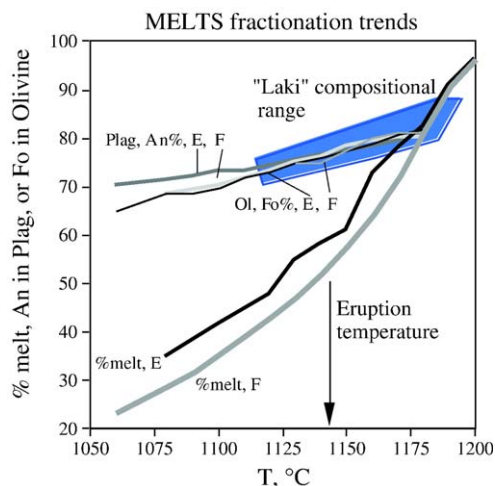


Fig. 2. Modeling of compositional trends of melts and minerals using MELTS algorithm. The initial parameters of 1.5 kb, f_{O_2} between QFM and NNO, and 0.2 wt.% H_2O for a typical olivine tholeiite basaltic magma provide the best match to compositional trends of melts and minerals, interstitial melts and the composition of melt inclusions (see Appendix). Predicted melt and mineral composition showing that 55% of either fractional (F) or equilibrium (E) crystallization of an initial olivine tholeiite can match the compositional ranges of melts and minerals in Laki, and eruption temperature of 1140 °C [10,22].

[21] measured in glass inclusions from phenocrysts and xenocrysts.

Phenocrysts assemblages, percentage of crystallization, and plagioclase and olivine compositions would be most consistent with the eruption temperature of 1160–1170 °C after 25% fractionation; 1 atmosphere equilibrium experiment on Laki compositions [22] showed that plagioclase and olivine appear on the liquidus nearly simultaneously between 1185 and 1161 °C, followed by clinopyroxene, while Fe–Ti oxides appeared only at 1070 °C. The absence of Fe–Ti oxides in Laki volcanics is in agreement with the

estimated emplacement temperature of 1120–1140 °C using Mg in glass geothermometer [10].

3.2. Oxygen isotope results: glass and phenocryst analyses

3.2.1. Glass

Whole-rock and glass samples of Laki lava and tephra are remarkably homogeneous in oxygen isotopic composition (Table 1, Fig. 3). This result corroborates the constancy of chemical and isotopic compositions, determined earlier, including constant $^{230}\text{Th}/^{232}\text{Th}$ and $^{87}\text{Sr}/$

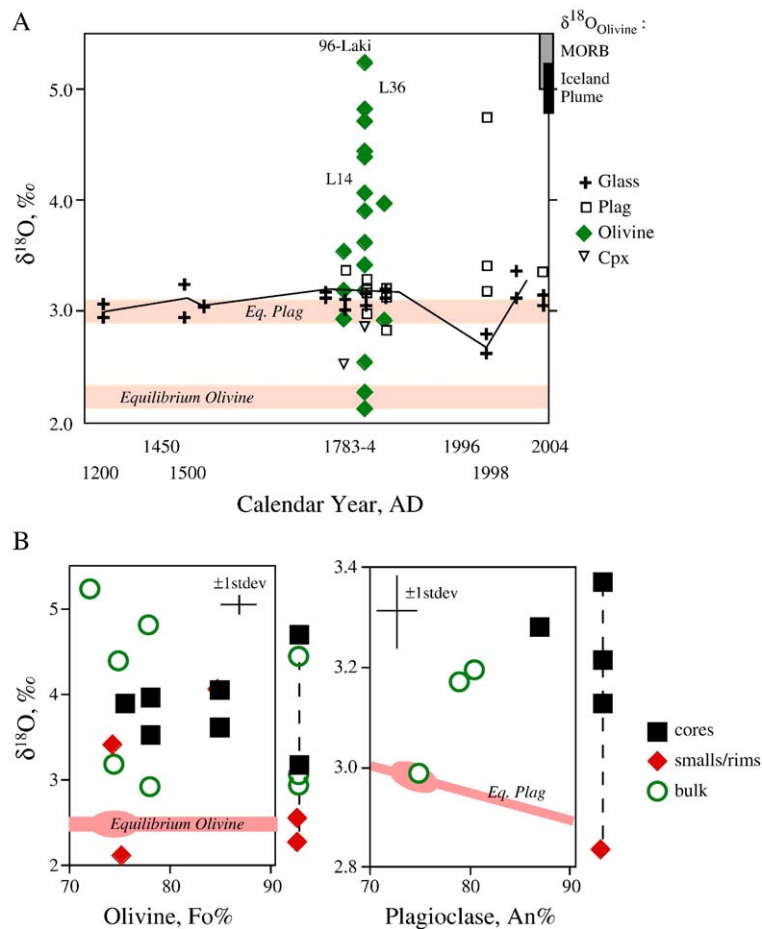


Fig. 3. A: Oxygen isotope evolution of the Laki–Grímsvötn magma system based on historical tephra and products of Laki eruption. There are relatively constant $\delta^{18}\text{O}_{\text{melt}}$ (glass) values through time and extreme diversity in $\delta^{18}\text{O}$ values of phenocrysts. 13th–18th Century AD and 1998 AD tephra are aphyric. The $\delta^{18}\text{O}$ values of An_{75} plagioclase and olivine in equilibrium with $3.1 \pm 0.1\%$ melt are shown. In the 1996 eruption, plagioclase spans far wider range than in Laki, which is interpreted here to indicate shorter residence and later entrainment of feldspars in magma, but possibly the same magma as in 1938, or magma derived thereof, given the exact proximity of the vents [12,13]. B: The $\delta^{18}\text{O}$ values of single crystals vs. their forsterite and anorthite composition and size; equilibrium with melt values are shown by line and oval; most olivines are in compositional but not isotopic equilibrium. Olivine (and plagioclase) shows positive correlation of size and $\delta^{18}\text{O}$. Vertical dashed line on the right of each diagram denotes crystals, that were only selected based on their size.

Table 2
U-series data for Laki and Grímsvötn basalts

Sample	$\left(\frac{^{226}\text{Ra}}{^{230}\text{Th}}\right)$	$\left(\frac{^{210}\text{Pb}}{^{226}\text{Ra}}\right)$	$\frac{^{87}\text{Sr}}{^{86}\text{Sr}}$	U (ppm)	Th (ppm)	$\frac{\text{U}}{\text{Th}}$
<i>Laki (1783–1784 AD)</i>						
L84	1.11		0.70324	0.355	1.16	0.306
L47	1.14		0.70324	0.351	1.14	0.308
<i>Grímsvötn</i>						
G1922	1.12	0.99	0.70321	0.366	1.12	0.327
G1983	1.14	1.02	0.70320	0.400	1.31	0.305

Radium and Th concentrations and Ra isotope ratios were measured by thermal ionization mass spectrometry, whereas ^{210}Pb was analyzed by alpha-counting of its progeny, ^{210}Po . Uncertainties are estimated as 2% for both ratios at the 95% confidence limit. Uranium and Th concentrations and Sr isotope ratios are from Sigmarsson et al. [13] and Sigmarsson et al. [14].

^{86}Sr , which is surprising for the 15 km³ of erupted material ([14,15], Table 2).

Samples of glass from lavas and tephra of the Laki eruption, and the Grímsvötn tephra, have the lowest $\delta^{18}\text{O}$ values ever observed in fresh basalt — +3.1±0.1‰. These values are remarkably homogeneous considering that they represent ~ 800 years of volcanic activity and tens of cubic kilometers of lava and tephra. The overall homogeneity of the Laki–Grímsvötn magmatic products suggests the existence of a unique low- $\delta^{18}\text{O}$ and thoroughly compositionally mixed magma reservoir beneath the Laki–Grímsvötn volcanic system since at least the 12th century. The 1996 Gjalp eruption that happened on the flank of Grímsvötn caldera is represented by somewhat more evolved basaltic icelandites [12,13], and it has the $\delta^{18}\text{O}_{\text{glass}}$ value of 2.7±0.2‰ (Table 1, [13]). This eruption may sample small flank magma chamber where further differentiation occurs.

3.2.2. Crystals

Oxygen isotope and chemical compositions, as well as textural characteristics of minerals examined in this study are summarized in Fig. 3. The remarkable feature of the Laki–Grímsvötn volcanic system, is that whereas the $\delta^{18}\text{O}$ values of basaltic glass from these eruptions is homogeneously low, those of olivine, plagioclase, and clinopyroxene crystals are remarkably variable and exhibit some of the largest disequilibrium mineral–mineral, and mineral–glass fractionations known in fresh igneous rocks (Fig. 3A).

In products of the 1783–1784 Laki eruption, olivines span 3‰. At the high-end, $\delta^{18}\text{O}$ values of 5.2‰ are typical of mantle olivine and most olivines in basalts elsewhere. The low-end values of 2.5–2.3‰ represent the record extreme for fresh igneous olivine, and are in equilibrium with the host lava.

There is a correlation between crystal size and $\delta^{18}\text{O}$ values (Fig. 3B), with larger (smaller) olivines and plagioclase crystals and rims having generally higher (lower) $\delta^{18}\text{O}$ values. The persistence of extreme oxygen isotope heterogeneities within crystals of the Laki and Grímsvötn lavas and tephra requires that these crystals spent relatively short time in contact with their isotopically homogeneous host magmas, based on the diffusion argument below.

3.3. Diffusion modeling

3.3.1. Oxygen diffusion

The oxygen isotope systematics of our data can constrain the period of time crystals spent in contact with melt if we make the seemingly reasonable assumption that during the residence time of normal- $\delta^{18}\text{O}$ “crystal cargo” in low- $\delta^{18}\text{O}$ melt, the crystals undergo oxygen isotopic and trace elemental exchange with adjacent melt, rate-limited by volume diffusion in the crystals. Calculations were made using analytical solution of Crank [32] for diffusion in spheres of different radii. Initial conditions for these calculations are the entrainment of high-Fo olivines (ca Fo₉₀) with high Ni, Mn, and Ca concentrations, as well as normal $\delta^{18}\text{O}$ values of 5.2‰. These olivines, and plagioclase An₈₆ were entrained into more evolved quartz tholeiitic melt held at 1160 °C, which also has $\delta^{18}\text{O}$ value=3.1‰, equivalent to the composition of Laki glass. Such melt should crystallize olivine Fo₇₆ with $\delta^{18}\text{O}$ =2.5‰, and plagioclase An₇₅ with $\delta^{18}\text{O}$ =3.1–3.2‰ in accordance to experiments of Chiba et al. [33]. The diffusion of trace elements and oxygen through crystal lattices of olivine and plagioclase should occur to achieve the new chemical and isotopic equilibrium. This rate of diffusion is the slowest rate-limiting process, thus time can be inferred from the relative progress of exchange. This time spans the interval from the beginning of oxygen exchange upon crystal entrapment to the end of diffusion during eruptive quench.

Because we are able to analyze only small and large crystals by our methods, we tuned up the diffusion model to measure the differences between cores (50% of internal diameter), and rims (50% of external diameter) of small and large crystals, and bulk values for small crystals, and plotted these curves on Fig. 4A. To our advantage, there are two minerals – olivine and plagioclase – which exhibit isotopic zoning. Therefore, the observed $\delta^{18}\text{O}$ range of olivine and plagioclase can be fitted uniquely into these equilibration curves, allowing the diffusion equilibration

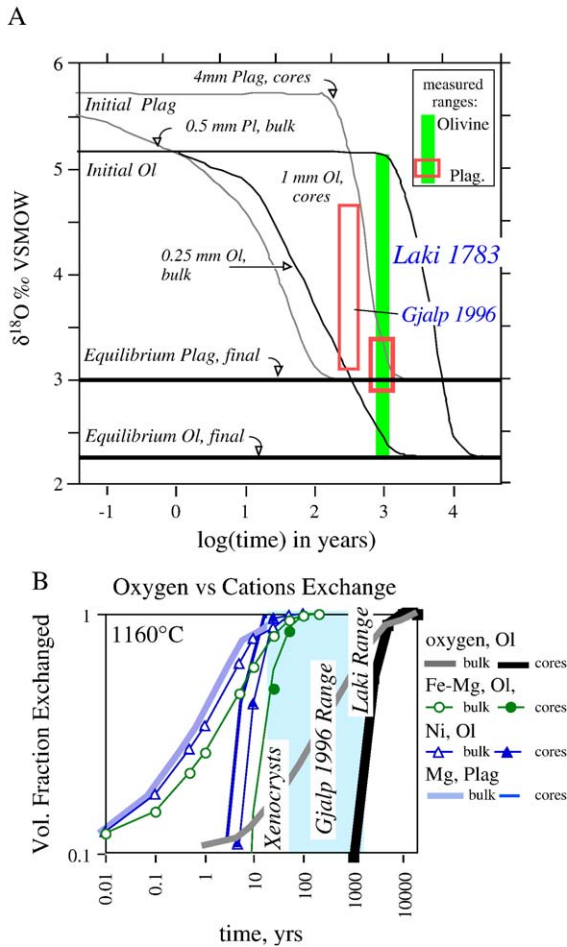


Fig. 4. Diffusion modeling of isotopic (A) and trace elemental (B) equilibration of initially normal- $\delta^{18}\text{O}$ olivine and plagioclase with depleted 3.1‰ basaltic melt of Laki. Calculations were made at 1160 °C using diffusion in a sphere analytical solution [32], and diffusion coefficients of O: ([34] QFM, F_{090}), Ni [39], Fe–Mg [40]; O in plagioclase [36], Mg in plagioclase [41]. The bulk of smaller crystals exchanges faster than the cores of larger crystals, creating heterogeneity within each mineral. Cores are defined as 50% of internal radius of crystals. Rates of diffusion are faster in plagioclase than in olivine, leading to faster equilibration and less variability in plagioclase; this coupled with the largest $\delta^{18}\text{O}$ diversity seen among olivine crystals calls for ~ 1000 yr model diffusive equilibration time. The small volume 1996 eruption has less $\delta^{18}\text{O}$ equilibrated plagioclase, suggesting ~ 200 yrs equilibration time between crystal entrainment and eruption; no olivine is present. (B) Cation exchange in olivine and plagioclase vs. oxygen isotope exchange using bulk diffusion; the inferred time is long enough to equilibrate cations (i.e., > 100 years) but too short to equilibrate oxygen (i.e., < 1000 yr). The majority of analyzed olivine crystals, regardless of size, have Fo equal to $75 \pm 2\%$ with constant Ni concentration (Fig. 3B), at or near the inferred Fe–Mg equilibrium with the groundmass, while Mg in plagioclase exhibits equilibrium partitioning (Fig. 5). Two large F_{086} olivine grains were found in Laki basalts and have higher than equilibrium $\delta^{18}\text{O}$ values of 3.6–4.0‰ (Fig. 3B), and also contain high Ni, Ca, and low Mn; they are surrounded by thin rim of groundmass composition. Such relationship calls for a very short pre-eruptive entrainment of these xenocrystic grains into Laki magma.

time to be forked from both sides, since plagioclase is a fast diffuser of oxygen, and olivine is the slow diffuser [34–37].

Diffusion of oxygen in olivine varies as a function of temperature, oxygen fugacity and forsterite composition [34,35], and that of plagioclase — on anorthite content (e.g., [36,37]). We chose experiments that are the closest to the Laki case: F_{090} , QFM, $T=1160$ °C. For plagioclase, we took a diffusion coefficient that is a factor of two higher than that measured for pure anorthite, a reasonable assumption for An_{86} plagioclase [37].

Timescales on Fig. 4A can be subdivided into three categories: on short timescales of months–years, plagioclase achieved isotopic disequilibrium between cores of large crystals and bulk of small crystals, while olivine has not. On long timescales of more than 1000 years, plagioclase has already achieved equilibrium, while large olivines have not. On intermediate timescales of hundreds of years, both plagioclase and olivine exhibit isotopic zoning, which we observe in Laki, and this timescale is, therefore, the best estimate for the duration of the oxygen exchange process.

The diffusive equilibration hypothesis explains the size-dependence of mineral $\delta^{18}\text{O}$ values, because smaller crystals undergo proportionally faster exchange than larger ones (Figs. 3,4). Moreover, since oxygen diffusion in plagioclase is faster than in olivine [34,36], our model also explains why in any one lava flow (Laki), olivine is significantly more variable in $\delta^{18}\text{O}$ than plagioclase (Fig. 4A). This hypothesis also provides a mechanistic interpretation of the relatively great range in $\delta^{18}\text{O}$ of plagioclase in the 1996 Gjalp eruption — this is the expected result if this small-volume Grímsvötn magma, erupted through the flank of the volcano, underwent relatively brief storage as a crystal–melt mixture before eruption, and the large-volume Laki eruption underwent relatively long storage (Fig. 4A). We estimate, based on a simple diffusion model, constrained by the small plagioclase–melt disequilibria, and the large olivine–melt disequilibria, that the maximum time of oxygen isotopic exchange between minerals and melt in the Laki parent magma was ~ 1000 yrs. This maximum time refers to the duration between entrainment of crystals into melt and eruption of the melt–crystal aggregate.

3.3.2. Major and trace element zoning in olivine and plagioclase

A second constraint on the residence time of crystals in magmas of the Laki/Grímsvötn magmatic system

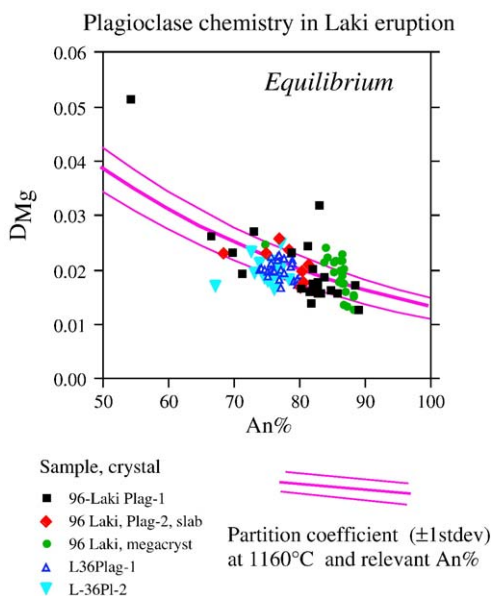


Fig. 5. The equilibrium partitioning relations between large plagioclase phenocrysts and host Laki melt. The D_{Mg} are calculated by dividing measured Mg concentrations within plagioclase by the Mg concentration in the ambient Laki groundmass with MgO=6 wt.%. Lines of partition coefficients as a function of An content of plagioclase and temperature of 1160 ± 50 °C are from Bindeman et al. [38]. Partitioning of Mg in several crystals with variable anorthite content is seen to be largely consistent with equilibrium partitioning and requires 10–100 yrs based on diffusion model of Mg in plagioclase of Costa et al. [3,6], suggesting that at least that much time has elapsed since the beginning of plagioclase-melt exchange and the eruptive quench.

comes from the subdued major and trace element zoning in olivine and plagioclase. Forsterite contents and Ca, Mn, and Ni concentrations in olivine phenocrysts in the Laki lavas and tephra are generally similar to those in groundmass olivines and, in the case of Fo content (76 ± 2 for most grains), are in equilibrium with the host lava. Similarly, Mg contents of plagioclase phenocrysts are in equilibrium partitioning relations, given the dependence of D_{Mg} on anorthite composition (Fig. 5). These data, combined with models of diffusive exchange of cations between phenocrysts and melt (Fig. 4B), suggest that several hundred years elapsed between incorporation of these crystals into Laki magma and eruption.

Rare large olivines in Laki basalt have Fo_{86-90} cores, higher than average Ni, Ca, and lower than average Mn; they are surrounded by thin rims of olivine in equilibrium with the groundmass. These grains have experienced shorter than average residence times in their host magmas (Fig. 3B). Two such grains were analyzed for oxygen isotope composition and found (Fig. 3) to have $\delta^{18}\text{O}$ values of 3.6‰ and 4.0‰ — consistent with only

partial exchange with host melt. Given rapid diffusion of cations in olivine (e.g., [39,40]), the preservation of forsteritic cores suggests short, ca. <10 yrs residence of these crystals (Fig. 4B) in melt, illustrating their xenocrystic nature.

3.4. U-series disequilibria and inferred age of magma

A third line of evidence on the timescales of magma storage and differentiation comes from ^{210}Pb – ^{226}Ra – ^{230}Th disequilibria measured in two lavas from Laki and two 20th century tephra from Grímsvötn (Table 2). The activity ratios of ($^{226}\text{Ra}/^{230}\text{Th}$) in all four of these samples are constant within analytical precision at 1.13 ± 0.01 ($\pm 2\sigma$), or 13% enrichment in ^{226}Ra relative to secular equilibrium with its parent, ^{230}Th . Such excesses of ^{226}Ra are common in basalts from hotspot and mid-ocean ridge environments and are generally interpreted as the result of fractionation of Ra from Th during magma extraction from the mantle source (e.g. [5–7,42]). In principle, assimilation of hydrothermally-altered crust could affect the final ($^{226}\text{Ra}/^{230}\text{Th}$), if recent hydrothermal alteration leads to either ^{226}Ra excesses or deficits. However, the basaltic crust beneath the Laki fissure swarm is Pleistocene and older (e.g., several million years old, [43]) and can be safely assumed to have ^{226}Ra in equilibrium with ^{230}Th , or ($^{226}\text{Ra}/^{230}\text{Th}$)=1. Thus, crustal contamination would lower rather than increase the ^{226}Ra excesses. Therefore, the initial ^{226}Ra -excess in Laki–Grímsvötn basalts was most likely caused by mantle melting and could have been initially higher.

Tephra erupted during the 20th century from Grímsvötn volcano are in ^{210}Pb – ^{226}Ra equilibrium suggesting negligible degassing of either lead or radon. First-order interpretation of the ^{210}Pb – ^{226}Ra equilibrium is that magma storage times exceeded 5 half-lives of ^{210}Pb , i.e. >100 yrs by assuming initial ^{210}Pb deficits like those previously observed in primitive Icelandic basalts [6]. However, the observed excess of ^{226}Ra over ^{230}Th suggests that the time elapsed since the last Ra–Th fractionation is shorter than 5 Ra half lives. The ^{238}U -series disequilibria, therefore, imply a magma storage longer than 100 yrs but shorter than 8000 yrs beneath the Laki–Grímsvötn volcanic system.

4. Discussion and implications

4.1. Phenocryst ages and the age of magma

The timescales of magma storage inferred from U-series equilibria – longer than 100 yrs and less than a few 1000 yrs – are similar to our estimates of the residence

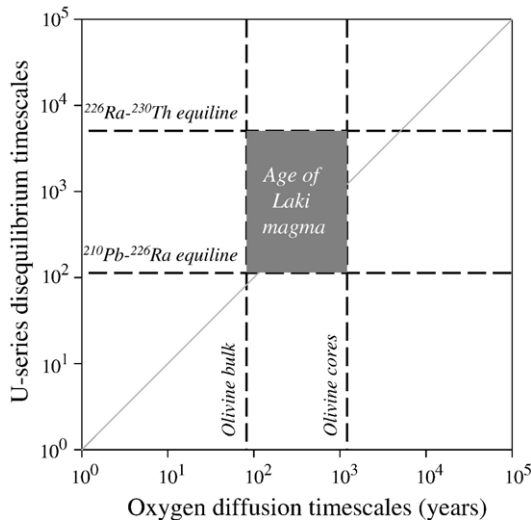


Fig. 6. Comparison of mineral diffusive timescales (Figs. 3–5), and U-series timescales (Table 2), and the inferred age of magma in the Laki–Grímsvötn system, see text for discussion.

times of crystals based on oxygen isotope heterogeneity and cation homogeneity in phenocrysts (Figs. 4,5). Fig. 6 compares these timescales and the ($^{226}\text{Ra}/^{230}\text{Th}$) age of magma. We discuss below petrogenetic mechanisms for the generation of voluminous and homogeneous low- $\delta^{18}\text{O}$ magma with heterogeneous crystal population that persisted for at least 8 centuries.

In order to explain the similarity of phenocryst and melt timescales, we suggest that crystals are formed, or exchanged with their host magma that was undergoing rapid $\delta^{18}\text{O}$ transformation during assimilation. The melt changed its $\delta^{18}\text{O}$ much more rapidly due to faster convective equilibration and faster diffusion of oxygen in melt. In this model, the comparatively short diffusive timescales in phenocrysts and young age of the magma (Fig. 6) are genetically-related.

In principle, the isotopically heterogeneous “crystal cargo” which is present in Laki magma, may represent two sources: normal- $\delta^{18}\text{O}$ cumulates of the initial olivine tholeiite that assimilated the low- $\delta^{18}\text{O}$ crust, or inherited phenocrysts from the assimilated low- $\delta^{18}\text{O}$ source rock, such as Pleistocene hyaloclastites. Prolonged residence time leads to the compositional equilibration of these crystals with the new melt. Results of MELTS modeling show that crystals and evolving residual melt are related by simple crystallization–differentiation, or assimilation–fractional crystallization.

The shorter (ca $\ll 100$ yr) residence times, which were derived from a few compositionally zoned crystals of olivine with higher forsteritic cores (e.g. $> 80\%$ Fig. 3B)

clearly imply that these crystals are “xenocrysts”. Such xenocrystic olivines and plagioclases are observed as free crystal phases and as cores of glomerocrysts, some of which show synneusis. Moreover, crystals of similar composition contain primitive olivine tholeiite melt inclusions and inclusions with relatively flat REE pattern [21]. These crystals may represent the syneruptive entrapment of earlier crystallized and initially isolated cumulates from the same magma system, formed by initial cooling of the mantle derived, normal- $\delta^{18}\text{O}$ olivine tholeiite. The variable REE spectra of the melt inclusions [21] would, in that case, record the cooling history of the olivine tholeiite and the advancement of the crustal contamination process. Indeed, Metrich et al. [21] observed relatively flat REE spectra in olivine with 86% Fo and in a plagioclase with 89% An, similar to those of Recent olivine tholeiites from south Iceland [44], whereas identical LREE-enriched spectra were measured in Fo₇₅ olivine and in the groundmass glass. Next, we address the physical model of assimilation of the low- $\delta^{18}\text{O}$ crust.

4.2. Mass balance calculations

Great oxygen isotope depletions for basalts presented significant mass balance challenges in the past [14,15,45,46]. In order to explain 3‰ depletion of Laki and other basalts from the Grímsvötn magma system, and to constrain isotopic values and proportions of end-members, a series of simple bulk mixing calculations were performed (Fig. 7). Magma mixing with an ultra-low- $\delta^{18}\text{O}$ more silicic melt, or a similar model of bulk assimilation of such siliceous rock, a model which is often inferred for Iceland, is less realistic for Laki or other similarly low- $\delta^{18}\text{O}$ basalts elsewhere (Fig. 7, A1 in Appendix). This assimilant must have $\delta^{18}\text{O}$ in the range of -17‰ to -19‰ to match the $3.1 \pm 0.1\text{‰}$ values of Laki and other tephros from the Grímsvötn magma system. Such an extreme $\delta^{18}\text{O}$ estimate assumes a) $\sim 0\%$ fractionation between water and rock, which is attainable at temperatures of > 500 °C (e.g. [47]), and b) 100% exchange reaction progress. These -17‰ to -19‰ values of the assimilant are lower than the lowest reported $\delta^{18}\text{O}$ value of -12‰ of Iceland, found in drill cuttings into the hydrothermal system of Krafla volcano, and inferred to be in $\delta^{18}\text{O}$ equilibrium with present-day meteoric waters [48]. The extremely low- $\delta^{18}\text{O}$ value of the potential Laki siliceous mixing end-member is, therefore, not attainable using the present day rain or even glacial water from the interior of Vatnajökull glacier (-14‰ , [49]).

In order for the assimilant to be lower in $\delta^{18}\text{O}$, alteration by the synglacial Pleistocene waters would be a better solution. However, there is no solid evidence to

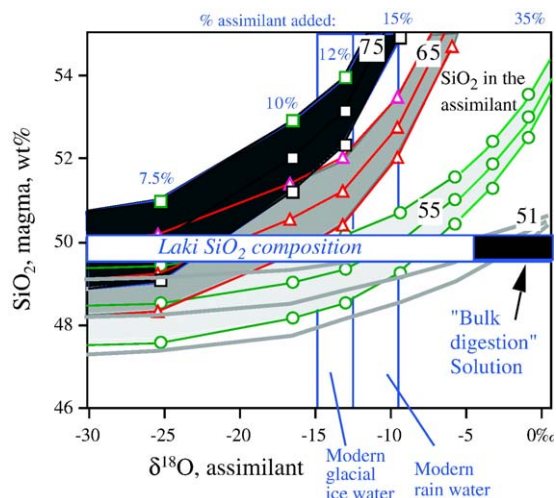


Fig. 7. Mass balance calculation aimed at constraining the possible range of $\delta^{18}\text{O}$ values of the assimilant and its proportion to produce 3.1‰, 50 wt.% SiO_2 Laki basalt. Bulk mixing/digestion of low- $\delta^{18}\text{O}$ assimilant with 75%, 65%, 55%, and 51% SiO_2 (shown on each series of curves) is assumed. The initial basalt was taken to be slightly low- $\delta^{18}\text{O}$ at 5.4‰, i.e. 0.2–0.3‰ lower than average MORB, but perhaps typical of unmodified basalts derived from the Iceland plume (e.g., [50,51]). Families of curves are shown for basaltic magma with initial SiO_2 contents of 47, 48 and 49 wt.%. The fields of $\delta^{18}\text{O}$ values of modern Iceland precipitation, and Vatnajökull ice cap are shown [49]. Notice that the bulk assimilation model is consistent with: 1) minor (<10%) mixing or assimilation of silica rich composition (55–65% of SiO_2) with ultra-low- $\delta^{18}\text{O}$ values of -9‰ to -25‰ ; or 2) more significant, ca 30–35%, assimilation (marked: “bulk digestion” solution) of basaltic–basaltic andesitic crust with 51% SiO_2 . The latter is our preferred interpretation for the origin of basaltic lavas of Laki and Grímsvötn caldera tephra. Notice that this bulk mixing model provides maximum assimilant proportions, and upper end of its $\delta^{18}\text{O}$ value, than any other AFC model which will lead to far faster increase in differentiation parameters, including rapid increase in SiO_2 per increment of assimilation, and would require lower assimilant proportions, and lower $\delta^{18}\text{O}$ of assimilant values.

suggest that the Pleistocene waters were significantly (e.g., by more than a few permil) lower in $\delta^{18}\text{O}$ due to the intraoceanic position of Iceland.

Four other lines of evidence discount mixing with an ultra-low- $\delta^{18}\text{O}$, -19‰ silicic melt: 1) there are no phenocrysts with such ultra-low- $\delta^{18}\text{O}$ values, and 2) no composition in Grímsvötn volcanics is more siliceous than icelanditic (ca 55 wt.% SiO_2), 3) no melt inclusions in phenocrysts are silicic, and 4) there is no textural evidence for mixing of magmas having much different compositions.

Mixing with moderately low- $\delta^{18}\text{O}$, 55 wt.% silica, icelandite melt is a possibility, but such model would still drive the product melt to become more siliceous and evolved than is observed for Laki or Grímsvötn basalts. Thus, low- $\delta^{18}\text{O}$ values in those basalts are best

explained by assimilation of another basic composition. In this case, the assimilant can be constrained to have $\delta^{18}\text{O}$ ranging between -4‰ and $+0.5\text{‰}$ (Fig. 7). These values are much more realistic in view of the estimated bulk of the Icelandic crust (ca. $+1\text{‰}$ to $+4\text{‰}$ [45]), present $\delta^{18}\text{O}$ values of meteoric waters, and moderate (0.1–0.5) water–rock ratios. Therefore, we favor a model of bulk assimilation of hydrothermally altered and hydrated basaltic crust by the heat of cooling and crystallization of hotter basalt, coming from the interior of the Icelandic plume. Such interpretation is getting supporting evidence in other, less ^{18}O -depleted basalts of Iceland [50,51], and may be applicable in other settings.

4.3. Icelandic hyaloclastites — the source of postglacial basaltic lavas?

Given the large proportion of the assimilant, which approach that of “bulk” digestion, or reactive assimilation (e.g., [52]), we search for the available source rocks with basaltic composition and low- $\delta^{18}\text{O}$ values (e.g., Fig. 7). Palagonitized volcanic rocks — those forming hyaloclastitic Table Mountains, and the majority of the >2.6 Ma Pleistocene volcanic record — fit the description well.

First, these rocks form country rocks of the Laki–Grímsvötn magma system. Second, they represent already disaggregated and often permeable material (Fig. 1B). The modification of the isotopic composition of palagonitized material likely happened in two steps: during the syneruptive disaggregation of basalts in contacts with the abundant glacial meltwaters, and during subsequent circulation of synglacial heated meltwaters through this permeable material. Third, hyaloclastites are hydrated, but the whole-rock composition and phenocrysts in them are largely preserved. For example, Breddam [19] presented whole-rock chemical and isotopic parameters for the Pleistocene table mountain Kistufell, that was formed under Vatnajökull glacier prior to deglaciation, ~ 40 km north of the Grímsvötn subglacial volcano. The $\delta^{18}\text{O}$ values of four samples ranged from 4.43‰ to 4.74‰, while the composition was typical olivine tholeiite with unaltered phenocrysts of olivine, clinopyroxene, and plagioclase. We observed palagonitized material in direct contact with Laki fissures (Fig. 1) with values of 2.78–3.3‰ (Table 1), but even lower values are possible based on experimental work on isotope fractionation.

The isotope fractionation between smectite/illite (taken here as proxy for palagonite) and water is

between +14‰ and +16‰ at 100 °C [53,54]. Because the present day $\delta^{18}\text{O}$ value of the Vatnajökul glacial meltwater is -14‰ to -15‰ [49], it is expected that high temperature interaction of +5.5‰ basaltic glass with these waters would generate equilibrium whole-

rock values near 0‰. This constraint is somewhat relaxed toward even lower temperatures, if isotopically lighter by several permil Pleistocene glacial meltwaters were involved. Furthermore, the glass $\delta^{18}\text{O}$ values will be lower than 0‰ if $T > 100$ °C. Temperatures of palagonitization were likely highly variable, as were the water/rock ratios; this will most likely lead to the highly heterogeneous $\delta^{18}\text{O}$ values of palagonite on centimeter scale, but overall $\delta^{18}\text{O}$ values are depleted by several permil.

We further suggest that some or many crystals in Laki magma may represent inherited phenocrysts from these hyaloclastites. Near surface palagonitization of volcanic glass seems to minimally affect phenocrysts, and these phenocrysts may become part of crystal cargo in the Laki magma. Mechanistically, large scale melting/digestion of palagonites, by hot, ca 1320 °C olivine tholeiitic basaltic magma leads to liberation of crystal cargo that sank into the main magma reservoir, get variably recrystallized and (partially) equilibrated with new homogeneous melt.

The model of bulk digestion of buried hyaloclastites is remarkably similar to large scale crustal remelting of tuffaceous intracaldera fills in caldera settings, such as Yellowstone and Timber Mt [2,4]. In silicic tuffs, phenocrysts of quartz and zircon are getting recycled; in rift zones of Iceland, olivine, clinopyroxene, and perhaps plagioclase phenocrysts are getting recycled. In all these environments, wholesale melting/digestion, characterize petrogenetic processes.

4.4. Assimilation-fractional crystallization calculations

Next, we check if the bulk assimilation/digestion model is feasible from the thermal perspective. Thermal modeling uses energy-constrained assimilation-

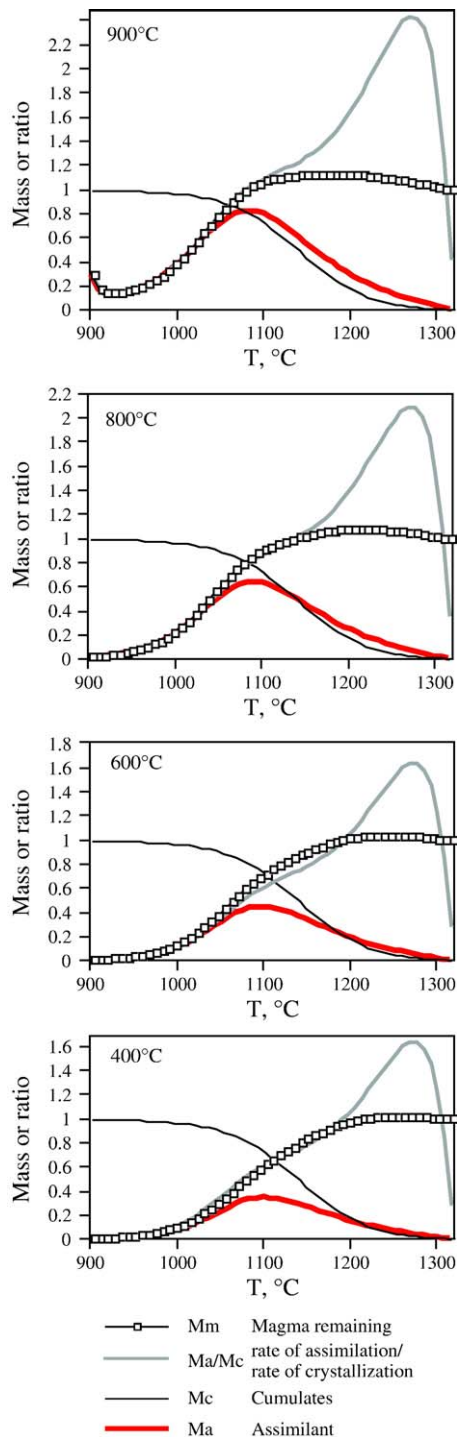


Fig. 8. Energy constrained AFC [55] simulation of a “magmatic digestion” process in which initially hot (1320 °C) and dry olivine tholeiite magma from the center of Icelandic plumehead bulk-digest more evolved, hydrated low- $\delta^{18}\text{O}$ basaltic assimilant with 51 wt.% SiO_2 , which has liquidus and solidus temperatures of 1200 and 900 °C respectively. Total mass of magma, mass of cumulates, mass of assimilant added, and ratio of assimilation/fractionation, are shown for each of the initial temperature of the assimilant. Note that the rate of assimilation/rate of crystallization is the largest in the beginning of the process because of overheat, which explains that the amount of melt initially increases with the progress of assimilation. Higher initial temperatures of country rock assimilant yield larger overall proportion of assimilation, but even assimilation at the initial 400 °C (e.g., buried hyaloclastites) produces significant proportions of assimilation and is capable of explaining the low- $\delta^{18}\text{O}$ of Laki basalt at its eruption temperature of 1160 °C (Fig. 2).

fractional crystallization calculations [55] for a hot olivine tholeiite [56,57] with temperatures of 1300 °C, which assimilates hydrated low- $\delta^{18}\text{O}$ basalt (Fig. 8). At Laki's eruption temperature of 1160 °C, there is about 25–45% cumulate removed and similar amounts of assimilant added using the above AFC model. Such proportions will keep the liquid in the basaltic range, and are in very good agreement with mass balance based on incompatible trace elements and melt inclusions: the resulting quartz tholeiitic basaltic magma became twice K_2O and H_2O -rich, as is observed ([10,21], and Appendix), in slight excess of what would be expected for a parental olivine-tholeiitic basaltic magma fractionation. The 25% to 45% of assimilation would agree well with the $\delta^{18}\text{O}$ value of the assimilant to be -4‰ to $+0.5\text{‰}$. It should be noted, however, that water content of Laki basalt is not well established, but oxide sums of melt inclusion and the residual glass do not suggest exceedingly high (e.g. <2 wt.% H_2O concentrations) ([10,21], this work, Appendix).

For the described digestion process to take place, the assimilating magma should be 1300 °C or hotter. This is permissible in plume environments, because basalts leaving the plume at the crust/mantle boundary have the highest temperatures of all basalts as they come from the hot interior of Earth [56]. The estimates for Iceland plume [57] range from 1390° in the Tertiary to 1350 °C at present, based on olivine–liquidus relationship. Such hot melts leaving mantle may initially contain up to 14 wt.% MgO, which is correlated positively with FeO, but subsequent olivine crystallization rapidly reduces MgO to a more typical 9–10 wt.% with negative correlation with FeO [56]. Concave downward REE patterns in melt inclusions within Fo_{86} olivine in Laki [21] would be consistent with a greater degree of depletion, higher degrees of partial melting, and thus high temperatures (e.g. [58]).

The model of bulk digestion does not have pressure (depth) constrained, and thus can't distinguish between assimilation under Grímsvötn volcano followed by the lateral dike propagation as advocated by Sigurdsson and Sparks [20] vs. magma generation near the Moho, followed by intrusion in the crust as a dike [14,17,43]. While thermally it would be easier to do the latter, MELTS experiments with Laki compositions are consistent with the lower pressure plagioclase–olivine–clinopyroxene assemblage phenocryst appearance sequence and assemblage. Moreover, if synglacially altered hyaloclastites were involved in assimilation, the magma chamber should be shallower, rather than deeper. This idea can be tested by further analysis of both

palgonitized glasses and unaltered minerals in the important hyaloclastite formation of Iceland.

4.5. Physical model for rapid generation of large-volume low- $\delta^{18}\text{O}$ basalts

The low- $\delta^{18}\text{O}$ signature of Laki and Grímsvötn magma system requires digestion of the low- $\delta^{18}\text{O}$ hydrated basaltic crust, and we presented evidence above that the record low- $\delta^{18}\text{O}$ values have been persisting for at least eight centuries. The isotopic disequilibrium in phenocrysts suggests that the initial basaltic magma was normal- to slightly-low in $\delta^{18}\text{O}$, as is recorded by the large, 4.5–5.2‰ olivine phenocrysts (Table 1, Fig. 3), and underwent significant isotopic modification in the low- $\delta^{18}\text{O}$ Icelandic crust on short timescales, possibly in response to assimilation. The same isotopic features and processes may characterize other large volume fissure basalts of Iceland.

Rapid generation of voluminous low- $\delta^{18}\text{O}$ magma reservoirs may be related to the deglaciation. The U-series timescales are clearly “postglacial”, i.e. the excess of ^{226}Ra over ^{230}Th dates generation of a relatively homogeneous magma body after glacial unloading of Iceland $\sim 11,000$ years ago [59,60]. Deglaciation has been proposed to be the cause of sharp increase in asthenospheric melt production and intensification of volcanism by a factor of 20–30 (by volume) in Iceland's northern rift zone and at Reykjanes Peninsula [59–61]. Similar increases might be expected for the center of Iceland's plume, close to which Laki is located.

Bulk digestion warrants large scale crustal recycling; such a model should equally apply to other most $\delta^{18}\text{O}$ depleted basalts of Iceland and elsewhere. It is important to stress that this petrogenetic interpretation is possible because the Icelandic crust is subaerial and is uniquely fingerprinted by the low- $\delta^{18}\text{O}$ values, possibly enhanced by the last glaciation. However, even in areas without such a strong isotopic contrast, similar processes may still apply. For example, the large-scale crustal digestion model for 15 km³ Laki eruption, serves as a model for petrogenesis and emplacement duration of basaltic magmas in the far vaster large igneous provinces, such as Columbia River basalts and the Siberian Traps.

The persistence of a low- $\delta^{18}\text{O}$ signature in the Laki–Grímsvötn magmatic system for at least the past 8 centuries, and as late as 1996–2004, points to the existence of an isotopically fingerprinted and voluminous basaltic reservoir under the largest glacier in Europe, that has significant implications for potential

Laki-size eruptions in the future. The detection of such a large magmatic reservoir should be the goal of high-resolution seismic tomography (e.g., [1]).

Acknowledgements

We thank NSF and the University of Oregon for funding fieldwork and oxygen isotope research, A.T. Anderson, G. Larsen and T. Thordarson for sample donation, the PNRN programme of the French INSU-CNRS and the Icelandic Research Fund for financial support. Reviews by T. Thordarson and Colin MacPherson are acknowledged.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at [doi:10.1016/j.epsl.2006.02.029](https://doi.org/10.1016/j.epsl.2006.02.029).

References

- [1] M. West, W. Menke, M. Tolstoy, S. Webb, R. Sohn, Magma storage beneath axial volcano on the Juan de Fuca mid-ocean ridge, *Nature* 413 (2001) 833–836.
- [2] I.N. Bindeman, J.W. Valley, Low- $\delta^{18}\text{O}$ rhyolites from Yellowstone: magmatic evolution based on analyses of zircons and individual phenocrysts, *J. Petrol.* 42 (2001) 1491–1517.
- [3] F. Costa, S. Chakraborty, R. Dohmen, Diffusion coupling between trace and major elements and a model for calculation of magma residence times using plagioclase, *Geochim. Cosmochim. Acta* 67 (2003) 2189–2200.
- [4] I.N. Bindeman, J.W. Valley, Rapid generation of both high- and low- $\delta^{18}\text{O}$, large-volume silicic magmas at the Timber Mountain/Oasis Valley caldera complex, Nevada, *Geol. Soc. Amer. Bull.* 115 (2003) 581–595.
- [5] M. Condomines, P.J. Gauthier, O. Sigmarsson, Timescales of magma chamber processes and dating volcanic rocks, *Rev. Mineral. Geochem.* 52 (2003) 125–174.
- [6] O. Sigmarsson, Short magma chamber residence time at an Icelandic volcano inferred from U-series disequilibria, *Nature* 382 (1996) 440–442.
- [7] C. Lundstrom, Uranium-series disequilibria in mid-ocean ridge basalts: observations and models of basalt genesis, *Rev. Mineral. Geochem.* 52 (2003) 175–214.
- [8] T. Thordarson, S. Self, Atmospheric and environmental effects of the 1783–1784 Laki eruption: a review and reassessment, *J. Geophys. Res.* 108 (2003) 4011 (NoD1).
- [9] S. Thorarinnsson, The Lakagigar eruption of 1783, *Bull. Volcanol.* 33 (1969) 910–927.
- [10] T. Thordarson, S. Self, N. Óskarsson, T. Hulsebosch, Sulfur, chlorine and fluorine degassing and atmospheric loading by the 1783–1784 AD Laki (Skaftár Fires) eruption in Iceland, *Bull. Volcanol.* 58 (1996) 205–225.
- [11] A.L. Chenet, F. Fluteau, V. Courtillot, Modelling massive sulphate aerosol pollution, following the large 1783 Laki basaltic eruption, *Earth Planet. Sci. Lett.* 236 (2005) 721–731.
- [12] S. Steinthorsson, B.S. Hardarson, R.M. Ellam, G. Larsen, Petrochemistry of the Gjalp-1996 subglacial eruption, Vatnajökull, SE Iceland, *J. Volcanol. Geotherm. Res.* 98 (2000) 79–90.
- [13] O. Sigmarsson, H.R. Karlsson, G. Larsen, The 1996 and 1998 subglacial eruptions beneath the Vatnajökull ice sheet in Iceland: contrasting geochemical and geophysical inferences on magma migration, *Bull. Volcanol.* 61 (2000) 468–476.
- [14] O. Sigmarsson, M. Condomines, K. Gronvold, T. Thordarson, Extreme magma homogeneity in the 1783–84 Lakagigar eruption: origin of a large volume of evolved basalt in Iceland, *Geophys. Res. Lett.* 18 (1991) 2229–2232.
- [15] K. Muehlenbachs, A.T. Anderson, G.E. Sigvaldsson, Low- $\delta^{18}\text{O}$ basalts from Iceland, *Geochim. Cosmochim. Acta* 38 (1974) 577–588.
- [16] G. Larsen, M.T. Gudmundsson, H. Björnsson, Eight centuries of periodic volcanism at the center of the Iceland hotspot revealed by glacier tephrostratigraphy, *Geology* 26 (1998) 943–946.
- [17] T. Thordarson, S. Self, The Laki (Skaftár-Fires) and Grimsvotn eruptions in 1783–1785, *Bull. Volcanol.* 55 (1993) 233–263.
- [18] S.P. Jakobsson, Petrology of recent basalts of the eastern volcanic zone, Iceland, *Acta Nat. Isl.* 26 (1979) 3–103.
- [19] K. Breddam, Kistufell: primitive melt from the Iceland mantle plume, *J. Petrol.* 43 (2002) 345–373.
- [20] H. Sigurdsson, S.R.J. Sparks, Lateral magma flow within rifted Icelandic crust, *Nature* 274 (1978) 126–130.
- [21] N. Metrich, H. Sigurdsson, P.S. Meyer, J.D. Devine, The 1783 Lakagigar eruption in Iceland: geochemistry, CO_2 , and sulphur degassing, *Contrib. Mineral. Petrol.* 107 (1991) 435–447.
- [22] J.D. Bell, D.J. Humphries, Lakagigar fissure eruption. in: *Progress in Experimental Petrology*. National Environmental Rec Council; UK Publ. Series, D2 (1972) 110–112.
- [23] J.M. Eiler, K. Gronvold, N. Kitchen, Oxygen isotope evidence for the origin of chemical variations in lavas from the Theistareykir volcano in Iceland's northern volcanic zone, *Earth Planet. Sci. Lett.* 184 (2000) 269–286.
- [24] J.M. Eiler, Oxygen isotope variations of basaltic lavas and upper mantle rocks, *Rev. Mineral. Geochem.* 43 (2001) 319–364.
- [25] O. Sigmarsson, S. Carn, J.C. Carracedo, Systematics of U-series nuclides in primitive lavas from the 1730–36 eruption on Lanzarote, Canary Island, and implications for the role of garnet pyroxenites during oceanic basalt formation, *Earth Planet. Sci. Lett.* 162 (1998) 137–151.
- [26] C.R. Bacon, J.B. Lowenstem, Late Pleistocene granodiorite source for recycled zircon and phenocrysts in rhyodacite lava at Crater Lake, Oregon, *Earth Planet. Sci. Lett.* 233 (2005) 277–293.
- [27] M. Condomines, K. Gronvold, P.J. Hooker, K. Muehlenbachs, R.K. O'Nions, N. Óskarsson, E.R. Oxburgh, Helium, oxygen, strontium and neodymium isotopic relationships in Icelandic volcanics, *Earth Planet. Sci. Lett.* 66 (1983) 125–136.
- [28] O. Sigmarsson, M. Condomines, S. Fourcade, A detailed Th, Sr, and O isotope study of Hekla: differentiation processes in an Icelandic volcano, *Contrib. Mineral. Petrol.* 112 (1992) 20–34.
- [29] M.S. Ghiorso, R.O. Sack, Chemical mass-transfer in magmatic processes, IV. A revised and internally consistent thermodynamic model for the interpolation and extrapolation of liquid–solid equilibria in magmatic systems at elevated temperatures and pressures, *Contrib. Mineral. Petrol.* 119 (1995) 197–212.
- [30] N. Óskarsson, G.E. Sigvaldason, S. Steinthorsson, A dynamic model of rift zone petrogenesis and the regional petrology of Iceland, *J. Petrol.* 23 (1982) 28–74.
- [31] N. Óskarsson, S. Steinthorsson, G.E. Sigvaldason, Iceland geochemical anomaly: origin, volcanotectonics, chemical

- fractionation and isotope evolution of the crust, *J. Geophys. Res.* 90 (1985) 10,011–10,025.
- [32] J. Crank, *The Mathematics of Diffusion*, 2nd edition, Oxford University Press, 1975. 414 p.
- [33] H. Chiba, T. Chacko, R.N. Clayton, J.R. Goldsmith, Oxygen isotope fractionations involving diopside, forsterite, magnetite, and calcite: application to geothermometry, *Geochim. Cosmochim. Acta* 53 (1989) 2985–2995.
- [34] F.J. Ryerson, W.B. Durham, D.J. Cherniak, Oxygen diffusion in olivine-effect of oxygen fugacity and implications for creep *J. Geophys. Res., Solid Earth* 94 (1989) 4105–4118.
- [35] R. Dohmen, S. Chakraborty, H.W. Becker, Si and O diffusion in olivine and implications for characterizing plastic flow in the mantle, *Geophys. Res. Lett.* 29 (2002), doi:10.1029/2002GRL015480.
- [36] S.C. Elphick, C.M. Graham, P.F. Dennis, An ion microprobe study of anhydrous oxygen diffusion in anorthite — a comparison with hydrothermal data and some geological implications, *Contrib. Mineral. Petrol.* 100 (1988) 490–495.
- [37] J. Brady, Diffusion data for silicate minerals, glasses, and liquids, in: T.J. Ahrens (Ed.), *Mineral Physics and Crystallography. A Handbook of Physical Constants*, AGU Reference Shelf, vol. 2, 1995, pp. 269–290.
- [38] I.N. Bindeman, A.M. Davis, M.J. Drake, An ion microprobe study of plagioclase–basalt partition experiments at natural concentration levels of trace elements, *Geochim. Cosmochim. Acta* 62 (1998) 1175–1192.
- [39] M. Morioka, Cation diffusion in olivine, *Geochim. Cosmochim. Acta* 45 (1981) 1573–1580.
- [40] A.J.G. Jurewicz, E.B. Watson, Cations in olivine. 2. Diffusion in olivine xenocrysts, with applications to petrology and mineral physics, *Contrib. Mineral. Petrol.* 99 (1988) 186–201.
- [41] T. LaTourrette, G.J. Wasserburg, Mg diffusion in anorthite: implications for the formation of early solar system planetesimals, *Earth Planet. Sci. Lett.* 158 (1998) 91–108.
- [42] K.M. Cooper, S.J. Goldstein, K.W.W. Sims, M.T. Murrell, Uranium-series chronology of Gorda Ridge volcanism: new evidence from the 1996 eruption, *Earth Planet. Sci. Lett.* 206 (2003) 459–475.
- [43] G. Palmason, Model of crustal formation in Iceland and application to submarine mid-ocean ridges, in the western N Atlantic region, in: P. Vogt, B.E. Tucholke (Eds.), *The Geology of N. America*, 1986, pp. 87–97.
- [44] P.S. Meyer, H. Sigurdsson, J.G. Schilling, Petrological and geochemical variations along Iceland neovolcanic zones, *J. Geophys. Res., Solid Earth* 90 (1985) 43–72.
- [45] B. Gautason, K. Muehlenbachs, Oxygen isotopic fluxes associated with high-temperature processes in the rift zones of Iceland, *Chem. Geol.* 145 (1998) 275–286.
- [46] J. MacLennan, D. McKenzie, K. Gronvold, N. Shimizu, J.M. Eiler, N. Kitchen, Melt mixing and crystallization under Theistareykir, northeast Iceland, *Geochem. Geophys. Geosys.* 4 (2003) (Art. No. 8624).
- [47] I. Friedman, J.R. O’Neil, Compilation of stable isotope fractionation factors of geochemical interest, Professional Paper, vol. 440-KK, U.S. Geological Survey, 1977.
- [48] K. Hattori, K. Muehlenbachs, Oxygen isotope ratios of the Icelandic crust, *J. Geophys. Res.* 87 (1982) 6559–6565.
- [49] H. Kristmannsdottir, A. Björnsson, S. Pálsson, A.E. Sveinbjörnsdottir, The impact of the 1996 subglacial volcanic eruption in Vatnajökull on the river Jokulsá a Fjöllum, North Iceland, *J. Volcanol. Geotherm. Res.* 92 (1999) 359–372.
- [50] C.G. Macpherson, D.R. Hilton, J.M.D. Day, D. Lowry, K. Gronvold, High-He-3/He-4, depleted mantle and low-delta O-18, recycled oceanic lithosphere in the source of central Iceland magmatism, *Earth Planet. Sci. Lett.* 233 (2005) 411–427.
- [51] P. Burnard, D. Harrison, Argon isotope constraints on modification of oxygen isotopes in Iceland basalts by surficial processes, *Chem. Geol.* 216 (2005) 143–156.
- [52] M.A. Dungan, J. Davidson, Partial assimilative recycling of the mafic plutonic roots of arc volcanoes: an example from the Chilean Andes, *Geology* 32 (2004) 773–776.
- [53] S.M. Savin, M. Lee, Isotopic studies of hydrous phyllosilicates, in: S.W. Bailey (Ed.), *Hydrous Phyllosilicates (Exclusive of Micas)*, *Rev. Mineral.*, vol. 19, 1988, pp. 189–233.
- [54] M. Escande, A. Decarreau, L. Layeyrie, Etude Experimentale de L’exchangeabilite des Isotopes de L’oxygene des Smectites, *C. R. Acad. Sci. Paris* 299 (1984) 707–710.
- [55] F.J. Spera, W.A. Bohron, Energy-constrained open-system magmatic processes I: general model and energy-constrained assimilation-fractional crystallization (EC-AFC) formulation, *J. Petrol.* 42 (2002) 999–1018.
- [56] S.A. Gibson, Major element heterogeneity in Archean to recent mantle plume starting-heads, *Earth Planet. Sci. Lett.* 195 (2002) 59–74.
- [57] C. Herzberg, M.J. O’Hara, Plume-associated ultramafic magmas of Phanerozoic age, *J. Petrol.* 43 (2002) 1857–1883.
- [58] D. McKenzie, R.K. O’Nions, Partial melt distributions from inversion of rare-earth element concentrations, *J. Petrol.* 32 (1991) 1021–1091.
- [59] J. MacLennan, M. Jull, D. McKenzie, L. Slater, K. Gronvold, The link between volcanism and deglaciation in Iceland, *Geochem. Geophys. Geosys.* 3 (2002) (Art. No. 1062).
- [60] L. Slater, M. Jull, D. McKenzie, K. Gronvold, Deglaciation effects on mantle melting under Iceland: results from the northern volcanic zone, *Earth Planet. Sci. Lett.* 164 (1998) 151–164.
- [61] M.A.M. Gee, M.F. Thirlwall, R.N. Taylor, D. Lowry, B.J. Murton, Crustal processes: major controls on Reykjanes Peninsula lava chemistry, SW Iceland, *J. Petrol.* 39 (1998) 819–839.