Laacher See revisited: High-spatial-resolution zircon dating indicates rapid formation of a zoned magma chamber

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ABSTRACT

High-spatial-resolution (^{230}Th)/(^{238}U) disequilibrium dating reveals rapid zircon crystallization prior to the 12,900 yr B.P. Laacher See (Germany) phonolite eruption. Zircons from Lower Laacher See tephra (LLST) and syenitic subvolcanic nodules share REE and low U/Th characteristics and define an isochron age of 17.1 ± 1.3 ka (1σ). Zircons also show higher initial (^{230}Th)/(^{232}Th)$_0$ and oxygen isotopic disequilibrium compared to their host phonolite. Thus, LLST zircons shortly predate the eruption and yet are not strictly part of the phonolite crystallization sequence. Based on their similarity to zircons in subvolcanic nodules, they are instead interpreted to be scavenged from a precursor syenite intrusion, which implies short (a few k.y.) filling and differentiation timescales for the Laacher See magma chamber. Thorium and oxygen isotopes in zircon as well as evidence for abundant crustal xenocrysts further indicate extensive crustal contamination, which allows for considerably faster basanite-phonolite differentiation than the ~100 k.y. time span previously estimated from bulk U-series data.

Keywords: U-238/Th-230, zircon, phonolite, magma chambers, Allerød.

INTRODUCTION

Understanding timescales of magma accumulation, preeruptive residence, and differentiation is essential for monitoring potentially hazardous volcanoes. Protracted crystallization upon cooling in a shallow magma reservoir, for example, can generate compositional zonation and volatile buildup, thus increasing the potential for violent eruptions (e.g., Wallace and Anderson, 2000). U-series disequilibrium dating has revealed that basaltic magmas in general have shorter residence times and differentiate more rapidly than silicic magmas (Hawkesworth et al., 2000; Reid, 2003; Condomines et al., 2003), but widely differing timescale estimates for basalt-tephrite-phonolite differentiation that range from a few k.y. to several hundred k.y. have been obtained, sometimes even for the same volcanic system (e.g., Canary Islands; Hawkesworth et al., 2000; Lundstrom et al., 2003).

Traditionally, U-series timescale studies have resorted to bulk analysis techniques using combinations of whole-rock, glass, or mineral samples. This is due to low abundances of relevant intermediate daughter isotopes (e.g., ^{230}Th, ^{231}Pa, or ^{226}Ra) that require extensive laboratory extraction and purification. The interpretation of bulk sample data, however, is complex because common processes such as magma mixing or assimilation may cause mixed crystal populations, or mineral compositions can be biased by the presence of actinide-rich accessory mineral inclusions (Condomines et al., 2003). Alternatively, the natural enrichment of U and Th in accessory minerals such as zircon or allanite can be harnessed by high-sensitivity and high-spatial-resolution ion microprobe techniques in order to extract age and compositional information from individual crystals or crystal domains. So far, such studies have concentrated on rhyolitic and rhyodacitic rocks (e.g., Reid et al., 1997; Lowenstern et al., 2000; Charlier et al., 2003; Vazquez and Reid, 2004), but zircon, for example, also occurs in intraplate trachytes and syenites/diorites (Condomines, 1997; Vazquez et al., 2005) or MORB gabbro (Schwartz et al., 2005), thus offering the potential for in situ U-series dating of differentiated mafic systems. Here, a comprehensive U-series, oxygen isotope, and rare earth element (REE) data set is presented for zircons from Laacher See (East Eifel, Germany). High-spatial-resolution results reveal a complex origin of zircon crystals, and urge reinterpretation of protracted magma evolution timescales for Laacher See volcano previously postulated based on bulk mineral analysis.

GEOLOGIC BACKGROUND, PREVIOUS WORK, AND SAMPLING STRATEGY

Laacher See maar volcano is located in the Cenozoic alkaline intraplate volcanic province of central Europe. The 12,900 yr B.P. (van den Bogaard, 1995; Litt et al., 2003) eruption is one of the youngest events in the East Eifel volcanic field (Germany), and with ~6.3 km^3 of erupted magma (dense rock equivalent; Schmincke et al., 1999), it ranks among the most violent recent eruptions in central and southern Europe. Compositional zonation of Laacher See tephra correlates with gradients in preeruptive temperatures and volatile contents, implying that the eruption tapped a vertically stratified magma body (Harms and Schmincke, 2000; Harms et al., 2004). Early-erupted Laacher See phonolite (Lower Laacher See tephra, LLST) is nearly aphyric and more differentiated, whereas pumice compositions from the middle and upper portions of the Laacher See tephra deposits (MLST and ULST, respectively) become progressively more mafic and crystal-rich (Wörner and Schmincke, 1984). LLST and MLST contain abundant crustal xenoliths interpreted as Devonian metasediments, whereas commingled phonolite-basanite lava fragments are present in late-erupted ULST (Wörner and Schmincke, 1984). In addition, juvenile plutonic nodules of mostly syenitic composition were ejected during the middle and later stages of the eruption that are regarded as disintegrated cumulates from the chilled magma chamber carapace (Tait et al., 1989). Rare carbonatitic inclusions have also been described (Liebsch, 1996).

From previous bulk U-series results, Bourdon et al. (1994) argued for a two-stage evolution of the Laacher See magma system: (1) lower crustal basanite-phonolite differentiation over ~100 k.y. based on the decay of the Th isotopic activity ratio (^{230}Th)/(^{232}Th) ≈ 1.05 of a basanitic parent to a ratio of ~0.87 for the least evolved Laacher See phonolite, followed by (2) differentiation in a shallow phonolitic magma chamber by crystallization for ~10–20 k.y. prior to eruption. From single-crystal ^{40}Ar/^{39}Ar data of LLST sanidine phenocrysts, van den Bogaard (1995) postulated crystal recycling from three precursor intrusive events at 127, 55, and 25 ka.

LLST tephra was sampled at the Mendig quarry location ~2 m above the contact to underlying Allerød loess. In their landmark petrologic study of the Laacher See tuff, Wörner and Schmincke (1984) detected zircon only in LLST heavy mineral separates, and consequently no attempt was made to extract zircons from other units. Millimeter-sized zircon crystals, however, occur in vesicles of syenitic subvolcanic nodules from the MLST and ULST. Twenty individual zircons from LLST and four large zircons from three individual subvolcanic nodules were characterized

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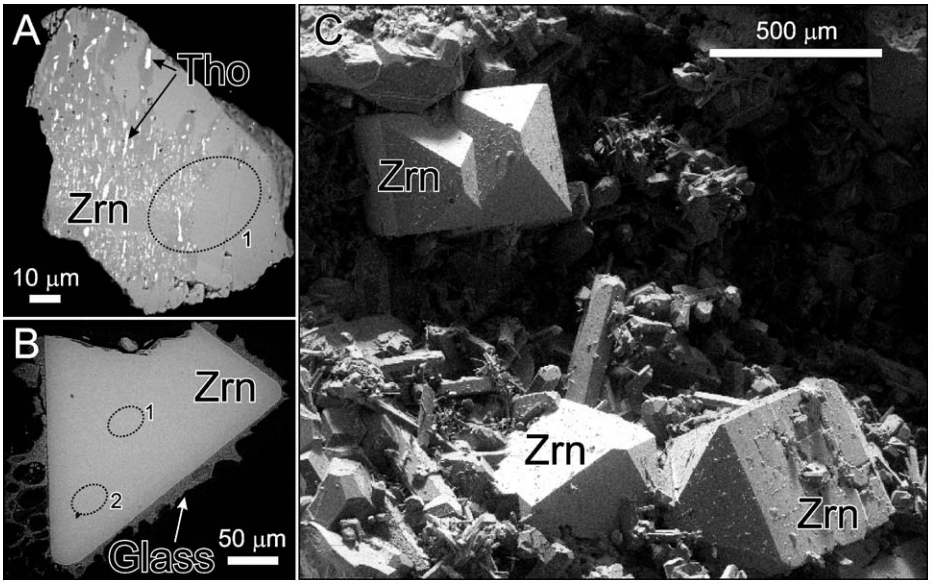

Figure 1. Backscatter electron images of LLST zircon (Zrn) with $ThSiO_4$ (Tho) rods present (A; grain 19) in comparison with homogeneous zircon (B; grain 18). Dotted ovals indicate U-Th ion microprobe spot locations. C: Secondary electron image of vesicle-grown zircon in a syenitic subvolcanic nodule.

by electron beam imaging and analyzed for U-Th (U-Pb) isotopes. A subset was further selected for ion microprobe, oxygen isotopic, and REE analysis (see GSA Data Repository[1] for analytical details, supplementary data tables, and backscatter electron and cathodoluminescence images).

RESULTS: HIGH-SPATIAL-RESOLUTION ANALYSIS OF ZIRCON

Fourteen of the analyzed LLST zircons yielded pre-Quaternary ages that range between ca. 390 Ma (^{206}Pb/^{238}U age) and 2.7 Ga (^{207}Pb/^{206}Pb age; Table DR1; see footnote 1). While most pre-Quaternary zircons lack adherent glass and are interpreted as accidental crystals derived from disseminated country rock fragments, some unequivocally are magma-hosted xenocrysts, based on the presence of adherent glass (Figs. DR1K and DR1L). The remaining six zircons have low U/Th and are characterized in part by extreme Th enrichment (up to ~4 wt%). Some zircons show domains with $ThSiO_4$ rods, tentatively interpreted as thorite exsolution features (Fig. 1A; see also Figs. DR1B, DR1J, DR1O, and DR1P). Vesicle-grown zircons in syenite subvolcanic nodules share these compositional and textural characteristics. Notably, the predicted partitioning for U and Th between phonolite melt and zircon (e.g., Blundy and Wood, 2003) would result in much higher zircon (^{238}U)/(^{232}Th) $\approx$ 5 than observed (0.3–2.5; Table 1), implying that both types of zircon crystallized from an extremely fractionated residual melt or fluid. When combined, U-Th isotope spot analyses of zircons from LLST and subvolcanic nodules yield an isochron age of 17.1 ± 1.3 ka (1σ; mean square of weighted deviates MSWD = 0.53; number of spots n = 22; Fig. 2). This value is indistinguishable from the regression through LLST zircons alone (age: $18.3^{+3.1}_{-3.0}$ ka; MSWD = 0.70; n = 14). While the limited spread of subvolcanic nodule zircons precludes calculation of a precise isochron age, the results nevertheless cluster closely on the LLST zircon isochron. Ion microprobe analysis generally targeted homogeneous zircon domains, and even where thorite-rich domains could not be avoided (e.g., Fig. 1A), U-Th isotopic results were nevertheless consistent.

The range in $\delta^{18}O_{SMOW}$ for LLST zircons (5.3–7.1‰) overlaps with values for zircon from subvolcanic syenite nodules (5.0–5.7‰; Table 1). Zircon-sanidine and zircon-melt oxygen isotopic fractionation factors are both −1.3‰ at 850 °C (Bindeman and Valley, 2003). Published values for LLST bulk mineral and glass separates (Wörner et al., 1987) are heterogeneous for sanidine ($\delta^{18}O_{SMOW}$ = 6.6–8.0‰), but glass values between 7.3 and 7.9‰ suggest oxygen isotopic disequilibrium of up to 1.3‰ between zircon and its eruptive host melt. This is in line with higher zircon initial (^{230}Th)/(^{232}Th)$_0$ = 0.894 ± 0.010 compared to (^{230}Th)/(^{232}Th)$_0$ = 0.863 ± 0.002 obtained from Laacher See glass analyses (Bourdon et al., 1994) (Fig. 2B). Diffusion may have partially equilibrated oxygen isotopes in zircon over several k.y. of magma residence (Bindeman and Valley, 2003), but U and Th diffusion is too sluggish to have altered the

TABLE 1. U-Th AND OXYGEN ISOTOPE RESULTS FOR LAACHER SEE ZIRCONS BY ION MICROPROBE ANALYSIS

Sample grain-spot	(^{230}Th)/(^{232}Th)	(^{238}U)/(^{232}Th)	Concentration		$\delta^{18}O_{SMOW}$* (‰)
			U (ppm)	Th (ppm)	
LS0501 composite pumice					
1-1	0.933 ± 0.024	1.08 ± 0.01	1040	2920	6.4
1-2	0.900 ± 0.022	1.05 ± 0.01	1370	3990	6.6
1-3†	0.918 ± 0.037	1.09 ± 0.03	980	2720	N.D.§
1-4†	0.930 ± 0.038	1.03 ± 0.02	980	2890	N.D.
2-1	0.781 ± 0.024	0.291 ± 0.007	2310	24,130	5.3
2-2	0.820 ± 0.029	0.284 ± 0.007	4130	44,320	N.D.
16-1	1.00 ± 0.04	1.25 ± 0.03	740	1800	6.0
16-2	0.945 ± 0.039	1.14 ± 0.03	960	2570	5.6
17-1	1.08 ± 0.17	1.50 ± 0.04	110	220	7.1
17-2	1.57 ± 0.04	2.51 ± 0.06	40	40	6.8
18-1	0.932 ± 0.036	0.950 ± 0.022	970	3110	N.D.
18-2	0.864 ± 0.028	0.745 ± 0.021	1300	5310	N.D.
19-1	0.936 ± 0.026	1.41 ± 0.03	20580	44,350	N.D.
20-1#	8.88 ± 0.20	9.13 ± 0.49	340	120	6.4
LST sn1-3 syenitic subvolcanic nodules					
1-1	0.766 ± 0.007	0.0228 ± 0.0003	220	28,690	N.D.
1-2	0.765 ± 0.008	0.0200 ± 0.0002	240	36,160	N.D.
2-1	0.773 ± 0.011	0.0223 ± 0.0002	210	28,130	5.7
2-2	0.772 ± 0.009	0.0211 ± 0.0002	230	32,550	5.2
3-1	0.768 ± 0.027	0.0938 ± 0.0026	710	23,020	N.D.
3-2	0.805 ± 0.028	0.0981 ± 0.0023	670	20,740	N.D.
4-1	0.754 ± 0.025	0.0622 ± 0.0015	70	3640	5.4
4-2	0.763 ± 0.024	0.0452 ± 0.0011	280	18,710	5.2
4-3	0.791 ± 0.026	0.0616 ± 0.0015	510	25,230	5.0
4-4	0.766 ± 0.007	0.0228 ± 0.0003	220	28,690	N.D.

Note: Activity calculations using decay constants: λ_{230}: 9.1577 × 10^{-6} a^{-1}; λ_{232}: 4.9475 × 10^{-11} a^{-1}; λ_{238}: 1.55125 × 10^{-10} a^{-1}; for analytical details see GSA Data Repository. All errors 1σ.
*Oxygen isotope analysis after regrinding of ~5 µm and repolishing; uncertainties ± 0.4‰.
†Replicate analysis after regrinding of ~5 µm and repolishing.
§N.D. = not determined.
#Xenocryst (^{207}Pb/^{206}Pb age 1457 ± 44 Ma).

[1]GSA Data Repository item 2006112, analytical details, supplementary data tables, and backscatter electron and cathodoluminescence images, is available online at www.geosociety.org/pubs/ft2006.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

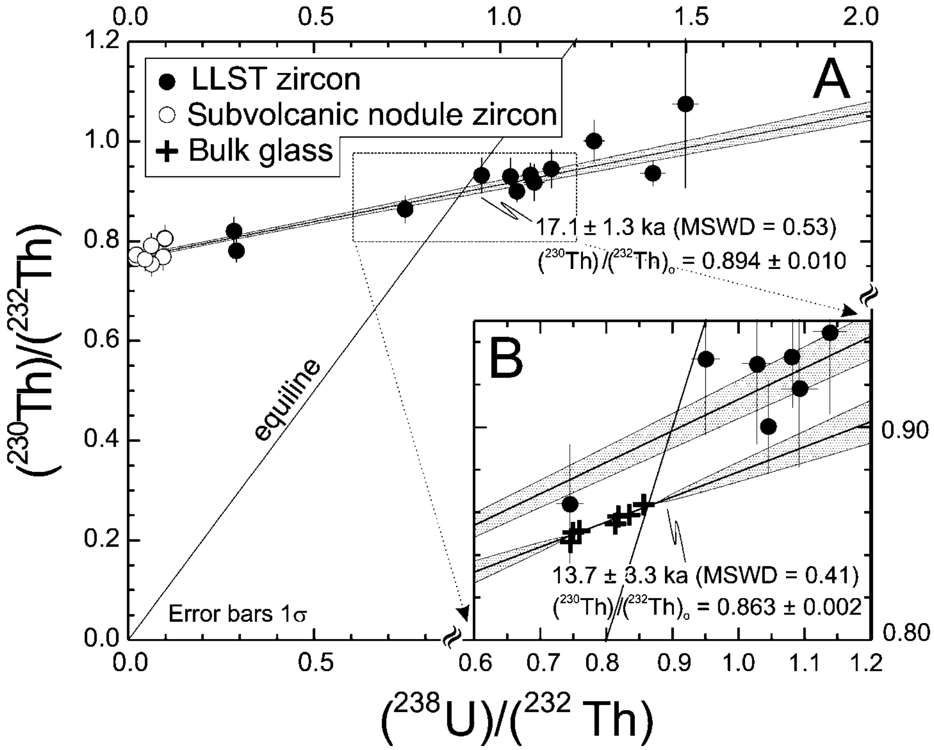

Figure 2. $(^{230}Th)/(^{232}Th)$ versus $(^{238}U)/(^{232}Th)$ diagram for Laacher See zircons including best-fit age and initial $(^{230}Th)/(^{232}Th)_0$ values (A). Xenocryst 20–1 (excluded from regression) and spot 17–2 omitted from plot for clarity. Inset (B) shows close-up with bulk glass separate data for Laacher See phonolitic pumice (Bourdon et al., 1994), recalculated ages, and recalculated $(^{230}Th)/(^{232}Th)_0$. LLST—Lower Laacher See tephra. MSWD—mean square of weighted deviates.

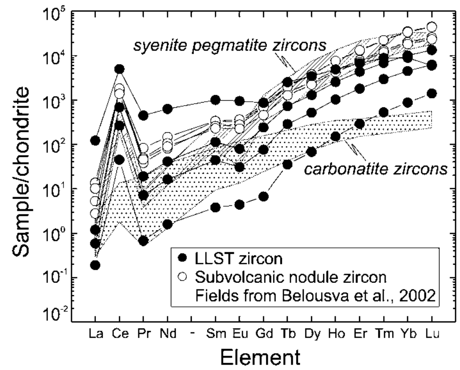

Figure 3. Chondrite-normalized REE patterns for Laacher See zircons. Zircon REE patterns for LLST and subvolcanic nodules are similar and share characteristics of syenite pegmatite zircons (Belousova et al., 2002). LLST—Lower Laacher See tephra.

composition of LLST zircons (Condomines et al., 2003).

LLST and syenite zircons both share REE patterns characterized by heavy rare earth element (HREE) enrichment, prominent positive Ce anomalies (Ce/Ce* = 18–100), and significant negative Eu anomalies (Eu/Eu* = 1.0–0.45). These patterns agree with those described for evolved syenitic zircon (e.g., Belousova et al., 2002), but differ from those of carbonatitic zircons where negative Eu anomalies caused by feldspar fractionation are typically absent (Fig. 3).

DISCUSSION OF ZIRCON PROVENANCE AND CONCLUSIONS

Phase equilibria and petrographic imaging studies (Harms et al., 2004; Ginibre et al., 2004) have previously demonstrated that a considerable fraction of Laacher See phenocrysts, including sanidine, plagioclase, and clinopyroxene, originated outside their host melt. As discussed below, the origin of accessory minerals is equally complex, but high-spatial-resolution U-series and U-Pb zircon geochronology now provides unique age constraints. Two main populations among LLST zircons are readily identified: Paleozoic-Precambrian xenocrysts and late Pleistocene (ca. 17 ka) zircons. While it is conceivable that analytical uncertainties may conceal multiple crystallization events for late Pleistocene zircons, the textural, compositional, and isotopic similarities as well as the low MSWD of the regression are permissive for treating them as a single population. Specifically, there is no evidence for zircon ages equivalent to the older crystallization events reported by van den Bogaard (1995).

High-spatial-resolution U-series data presented here indicate that zircon in LLST and syenite subvolcanic nodules crystallized rapidly within ±1.3 k.y. (1σ) analytical uncertainty, and within a few k.y. before eruption. Previously published bulk mineral-glass U-series ages for Laacher See (Bourdon et al., 1994), in particular for accessory titanite and apatite in LLST and cumulate nodules, yielded ages that were either significantly younger (by ca. 5 ka) or much older (by ca. 12, ca. 22, and ca. 400 ka) than the eruption age. Contamination by secular equilibrium xenocrysts that shifted bulk $(^{230}Th)/(^{238}U)$ closer to the equiline can explain the older ages, whereas heterogeneous initial $(^{230}Th)/(^{232}Th)_0$ for melt and crystals presumably accounts for prohibitively young apparent crystallization ages. To illustrate this effect, geologically unreasonable bulk titanite-melt and apatite-melt model ages were recalculated using the higher $(^{230}Th)/(^{232}Th)_0$ value from the zircon regression as the melt composition (Fig. 2A). This results in model crystallization ages between ca. 12 ± 2 and 15 ± 2 ka. The recalculated ages are thus close to the zircon isochron age and to bulk titanite and apatite crystallization ages for MLST and ULST that show excellent agreement with the eruption age (Bourdon et al., 1994). In the light of these findings, it is warranted to revisit the previously proposed (Bourdon et al., 1994; van den Bogaard, 1995) and widely cited (e.g., Hawkesworth et al., 2000; Condomines et al., 2003) notion of a long-lived Laacher See magma system.

An essential outcome of this study is that ca. 17 ka zircons in LLST pumice and subvolcanic nodules crystallized from a distinct syenitic magma reservoir that was more evolved and had higher $(^{230}Th)/(^{232}Th)_0$ and lower $\delta^{18}O$ compared to their eruptive host melt. Because it is difficult to envisage how an isotopically distinct magma could have bypassed a voluminous, long-lived chamber without homogenization, it appears more plausible that zircons and potentially other crystals were scavenged from a preexisting subvolcanic intrusion, or from marginal apophyses that segregated early from the Laacher See phonolite. To remain isolated from subsequent isotopic modification of the main magma body, such apophyses must have chilled and solidified rapidly. The Th isotopic homogeneity of compositionally heterogeneous phonolite glasses (Bourdon et al., 1994) (Fig. 2B) also requires differentiation and zonation of the Laacher See magma chamber to postdate its isotopic homogenization. In this case, in situ aging can be ruled out because >25 k.y. would be required for the decay of syenite $(^{230}Th)/(^{232}Th) = 0.89$ to the LLST melt value of 0.86, assuming $(^{238}U)/(^{232}Th)$ of 0.75. Instead, assimilation of country rocks with $(^{230}Th)/(^{232}Th)$ of ~0.76 (Bourdon et al., 1994) could explain the decrease in $(^{230}Th)/(^{232}Th)$. In any case, zircon crystallization in the syenite likely constrains shallow phonolite emplacement and/or isotopic homogenization and differentiation to only a few k.y. prior to eruption. This interpretation of rapid crystallization in a shallow magma chamber is supported by bulk mineral-glass isochrons for MLST and ULST pumice of essentially eruption age (Bourdon et al., 1994).

While zircon crystallization ages provide no direct bearing on the duration of lower crustal basanite-phonolite differentiation, the oxygen and Th isotopic evidence as well as the presence of abundant xenocrystic zircons clearly underline the significance of crustal contamination in the Laacher See magma system. Mass balance between basanite ($\delta^{18}O$ = 5.6‰; Wörner et al., 1987) and crust ($\delta^{18}O$ = 11.7‰) implies 20%–40% of crustal contamination in the phonolite melt ($\delta^{18}O$ = 7‰–8‰), which could also account for a significant fraction of the Th isotopic difference between basanite and phonolite. In other words, the previously postulated ~100 k.y. differentiation period based on the assumption of closed-system decay of ^{230}Th to explain the difference in $(^{230}Th)/(^{232}Th)$ between basanitic parent and phonolitic daughter magma (Bourdon et al., 1994) might have been in fact considerably (by 50%–100%) shorter.

In conclusion, high-spatial-resolution U-series dating of accessory minerals has the potential to reveal complex crystal origins in differentiated mafic magma systems that otherwise would go undetected by bulk methods, and may lead to erroneous apparent crystallization ages. In the case of Laacher See, zircons are interpreted to have crystallized in a syenitic intrusion that predates the emplacement and/or isotopic homogenization of the zoned phonolite magma chamber. By this rationale, preeruptive storage and differentiation of the Laacher See phonolite was limited to several k.y. at most, in line with evidence from fluid-dynamic modeling (Wolff et al., 1990) and with rapid differentiation in other intraplate mafic systems (e.g., Tenerife, Canary Islands; Lundstrom et al., 2003; Johansen et al., 2005).

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