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Magma dynamics of ancient Mt. Etna inferred from clinopyroxene isotopic and trace element systematics

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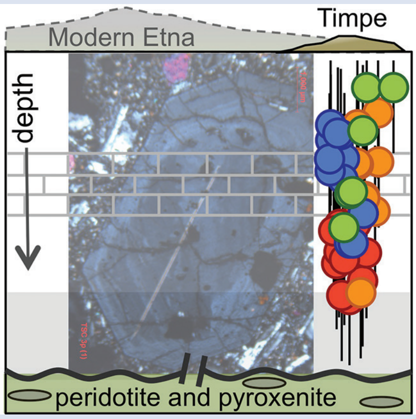
Abstract



Dynamic magmatic processes driving volcanic eruptions, including melting, fractionation, and assimilation, provide critical insights into plumbing systems supporting long-lived magmatism. Here we describe an approach combining *in situ* elemental analyses in clinopyroxene phenocrysts, integrated thermobarometry models, and bulk crystalline Hf, Nd, and Pb isotopic studies to reconstruct a key period of ancient eruptions of Mount Etna (Sicily), Europe's largest, most active volcano. Trace element signatures recorded in clinopyroxene from 220 to 100 ka are consistent with derivation from a heterogeneous mantle of hydrated peridotite and ~10 % pyroxenite, also consistent with sources feeding recent Etna eruptions. Isotopic data from Mount Etna alkaline lava clinopyroxene, crystallised between 0.5 and 0.2 GPa, insignificantly vary from whole rock values, ruling out substantive assimilation of material during magma ascent from the onset of clinopyroxene fractionation through the mid-crust, storage, and eruption. Together, our results suggest that varying contributions of well-mixed hydrated peridotite and pyroxenite melts have been consistent features of magma assembly beneath Mt. Etna since the development of ancient alkaline centres.

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Introduction

Volcanism began at Mount Etna, Europe's largest and most active volcano, at ~0.5 Ma (Gillot *et al.*, 1994), with ancient lavas now exposed around the perimeter of the modern-day edifice. Tholeiitic lavas were overlain by transitional and alkaline sequences starting at ~230 ka (Gillot *et al.*, 1994; Branca and Del Carlo, 2004). Mt. Etna sits on the northern edge of the African plate at the European-African collision zone and the western hinge of escarpments dividing it from where the Ionian slab descends beneath the Aeolian arc (Fig. S-1, Supplementary Information). Volcanism has been attributed to the manifestation of mantle upwelling independent of, or in response to, a slab tear (*e.g.,* Gasperini *et al.*, 2002), subduction-related fluid-triggered melting (*e.g.,* Armienti *et al.*, 2007 and references therein) or enhanced decompression melting resulting from convective anomalies (Gvirtzman and Nur, 1999; Schellart, 2010).

Magmatic products of the early Etna centres, including those of the ancient alkali centres active at ~200–100 ka, bear mantle-derived isotopic signatures consistent with contributions from both enriched and depleted source components (Marty *et al.*, 1994; Tanguy *et al.*, 1997). Though more recent Etna volcanic products exhibit distinctive signs of assimilation in the form of elevated Sr isotopic values and large ion lithophile element enrichments (Tonarini *et al.*, 1995), the degree to which crustal contamination influenced early alkaline products is uncertain. Similarly, magmatic processes between mantle melting regions and shallow reservoirs supplying volcanic activity remain enigmatic. In this study, we combine clinopyroxene (cpx) barometry, trace element concentrations, and Pb, Hf, and Nd mineral-whole rock (WR) isotopic (dis)equilibria to constrain source compositions and differentiation depths of magmas feeding lavas erupted at Timpe Santa Caterina (TSC). The advantage of employing these three isotopic systems together lies in the coupling of the slowly diffusing Hf and Nd with the more rapidly diffusing Pb, thereby providing the potential to infer magma assembly processes prior to eruption during this early period.

Sample Selection and Analytical Methods

Lavas at TSC encompass the whole ancient alkaline magmatism period at Etna, from 220 ka near sea level to likely <100 ka exposed atop the sea cliff (Gillot *et al.*, 1994). Early trachybasaltic and basaltic flows, TSC-2 and TSC-3, are overlain by more alkalic basanites and phonotephritic lavas (TSC-7 and TSC-9). Flows selected for this study contain the most

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abundant large cpx from the TSC suite (Fig. S-2, Supplementary Information). Major and trace element and isotopic analytical details and data are provided in Tables S-1 to S-5 (Supplementary Information).

Results and Discussion

Barometry. Observed cpx phenocrysts (>2 mm, large relative to other TSC lava phases) coupled with theoretical modelling of Etna compositions indicate early cpx crystallisation; hence cpx holds a potential record of pre-eruptive magma assembly processes (Armienti *et al.*, 2009). Crystallisation temperatures and pressures, solved iteratively using a single-cpx thermometer and single-cpx barometer for hydrous systems (respectively, Eqs. 32d and 32b in Putirka, 2008), yielded temperatures of 1060–1175 °C and an average pressure of 0.34 ± 0.16 GPa (Fig. 1a,b). Thermobarometric model accuracy was evaluated using a literature dataset of >100 experimentally coexisting cpx-liquid pairs over a compositional range bracketing TSC lavas and cpx compositions (*cf.* Supplementary Information Table S-6 for equations, ranges, references, and selection criteria). As noted by Mollo *et al.* (2010), single-cpx barometers can outperform liquid-based models for volatile-rich alkaline compositions. The single-cpx barometer for hydrous systems yields an average uncertainty of 0.17 *versus* 0.28 GPa for the cpx-liquid model of Putirka *et al.* (2003) for the compiled experiments, placing a lower bound of pressures recorded by TSC cpx at below 0.8 GPa, within the uppermost lithospheric mantle.

Crystallisation of TSC-2 and TSC-7 cpx generally occurred at depths centred around 0.5 GPa and 0.2–3 GPa, respectively (Fig. 1a), suggesting specific magma reservoir locations near the crystalline basement-granulite boundary and within the carbonate platform beneath Etna. More continuous polybaric crystallisation is apparent in TSC-3 and TSC-9. Combined with previous work on Etna lavas (see Supplementary Information), thermobarometry indicates that the bulk of ancient clinopyroxene phenocrysts crystallised between 0.5 and 0.2 GPa (Fig. 1b).

Heterogeneous mantle sources for ancient Etna. Clinopyroxene trace element concentrations, when coupled with single-cpx barometry pressure estimates, place constraints on magma source compositions and crustal mixing depths. Cerium, incompatible in all major TSC phases, functions as a fractionation proxy and indicator of magma evolution. Two distinct crystallisation paths are apparent in TSC cpx: trends characterised by high Y/La (TSC-2, TSC-7) and low Y/La (TSC-3, TSC-9) when linked with Ce (Fig. 1c). Clinopyroxene from the 2001 eruption also follow the low-Y/La trend, as do other known historic and recent Etna cpx (Viccaro *et al.*, 2006). Scarlato *et al.* (2014) have documented preferential HREE incorporation into cpx relative to LREE as a function of cooling rate, but in TSC phenocrysts, HREE-like Y has either negative or no correlation with major element chemistry associated with elevated cooling rates (*e.g.*, Na, Al^{IV}, and Ti). Accordingly, we interpret the Y/La-Ce trends to reflect source characteristics beneath Etna over time rather than being a feature of crystallisation conditions.

Clinopyroxene grains record existence of magmas beneath Etna deriving from melting of both pyroxenitic and peridotitic mantle components. The source characterisation enabled by analysis of Y/La-Ce trends in Etna TSC cpx can also be used to evaluate the composition of cpx in pyroxenite and peridotite xenoliths from the nearby Hyblean Plateau (Fig. 1c). Clinopyroxene from Hyblean pyroxenite xenoliths plot along the high-Y/La TSC trend (Fig. 1c), which is reproduced with a primary melt generated by a heterogeneous source of 10 % dry pyroxenite and 90 % hydrated peridotite in which ~10 % of each lithology melts and mixes at 1.5 GPa. Figure S-3 shows hypothetical source compositions with up to 20 % pyroxenite to constrain model sensitivity (Supplementary Information). These lithologies, similar to those determined by Correale *et al.* (2014) modelling trace element systematics in primitive Etna WR samples <15 ka, are distinct from peridotitic cpx from the nearby Hyblean plateau that fall below the low-Y/La trend. Low Y/La in cpx may result from either a hydrated peridotite source or a more evolved melt of the mixed pyroxenite source following apatite saturation.

Isotopic (dis)equilibria. Most Etna mineral-WR pair isotopic work has focused on the Sr and Nd systems in recent lavas (*e.g.*, Tonarini *et al.*, 1995; Armienti *et al.*, 2007), which generally exhibit more radiogenic Sr and less radiogenic Nd than ancient lavas. Within recent eruptive episodes, marked increases in WR $^{87}Sr/^{86}Sr$ are often accompanied by $^{87}Sr/^{86}Sr$ WR-cpx disequilibria (*e.g.*, 0.70348 cpx core values accompanied by 0.70362 WR values in 2001 eruptives; Armienti *et al.*, 2007).

Our approach employing coupled Hf, Nd, and Pb isotopic signatures in ancient volcanics brings three distinct chemical affinities to bear on determining magma assembly, as recorded in cpx trace elements, at depths constrained by thermobarometry. As refractory elements diffusing slowly in clinopyroxene (*cf.* Van Orman *et al.*, 2001), Nd and Hf may be expected to retain isotopic signatures from early crystallisation depths and exhibit large isotopic disequilibria with hosting magmas subject to mixing with recharging, or assimilating magmas, carrying isotopically distinctive compositions immediately prior to eruption. In contrast, Pb diffuses relatively rapidly, making Pb isotope systematics an especially promising approach for placing constraints on magma residence times within the crust.

Since each separate cpx analysis represents digestion of multiple grains likely crystallised at different depths, reported isotopic values reflect an average over the polybaric cpx crystallisation history. However, sluggish Nd and Hf re-equilibration will manifest itself as WR-cpx disequilibria in cases of late-stage incorporation of any volumetrically significant isotopically distinct magma during the final stages of magma assembly.

Neodymium and Hf isotopic compositions (Fig. 2a) of TSC cpx and WR demonstrate they are insignificantly distinctive at the 2σ level. However, it is notable that all cpx have slightly more enriched Nd isotopic signatures that trend toward those of continental values. This could result from a recharge process of fresher mantle-derived material that drives eruption. Late-stage shallow contamination, by contrast, would impart enriched crustal signatures to the WR, presumably after cpx phenocryst formation. Though Hf and Nd isotopic data for sedimentary units directly beneath Etna are unavailable for comparison with cpx and WR values, Sicilian beach sand ε_{Nd} derived from the western extension of sedimentary units underlying Etna and crustal rocks of south and central Italy are all considerably more enriched (Fig. 2a; ε_{Nd} -10.3 to -16.0, Conticelli *et al.*, 2002; Brems *et al.*, 2013). Such large differences make it unlikely that crustal sediments contributed to the Hf and Nd isotopic compositions observed in TSC cpx and WR materials. Rather, we infer that the isotopic signatures of these magmas were locked in at pressures corresponding, at minimum, to early cpx crystallisation at mid-crustal pressures of 0.5–0.2 GPa.

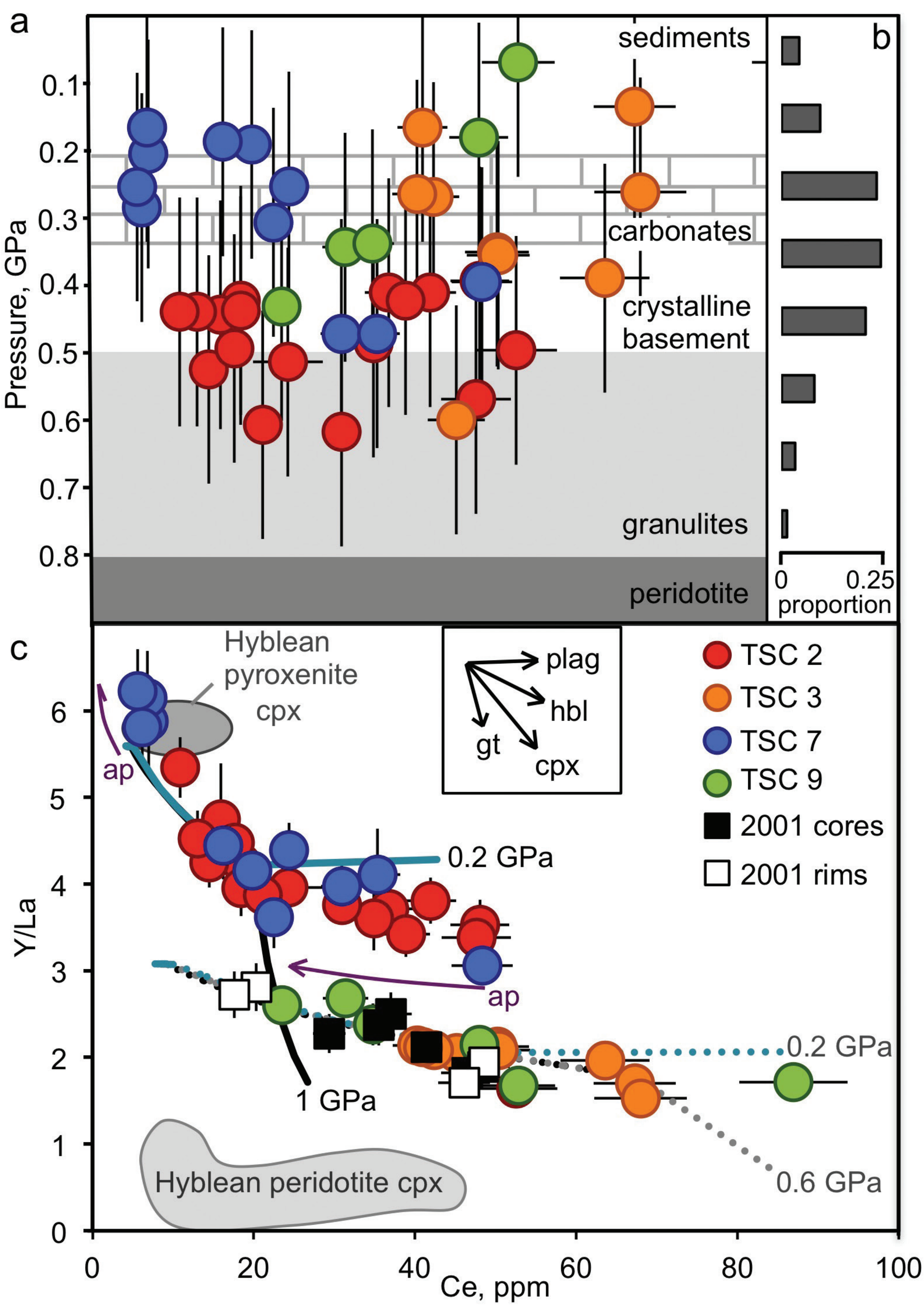

Figure 1 **(a)** Ce contents of TSC cpx as a function of single-cpx pressure estimates (1σ uncertainty) superimposed on Etna stratigraphy (after Spilliaert *et al.*, 2006). **(b)** Proportions of ancient Etna barometry from this study and previous work (*cf.* Supplementary Information, n = 287). **(c)** TSC cpx and 2001 eruption cpx (Viccaro *et al.*, 2006) shown with Hyblean pyroxenite and peridotite cpx fields (Correale *et al.*, 2012, and references therein). Isobaric cpx fractionation modelling for peridotite melt (solid lines) and pyroxenite melt (dashed lines) at 1.0 (black), 0.6 (grey), and 0.2 (blue) GPa performed using alphaMELTS (Smith and Asimow, 2005); conditions described in Supplementary Information. Fractionation of apatite, well known to incorporate REEs, is modelled in purple using the partitioning of Prowatke and Klemme (2006). Ol+cpx±opx+sp is present at the start of both trends, though olivine drops out at T < ~1100 °C for pyroxenite melt.

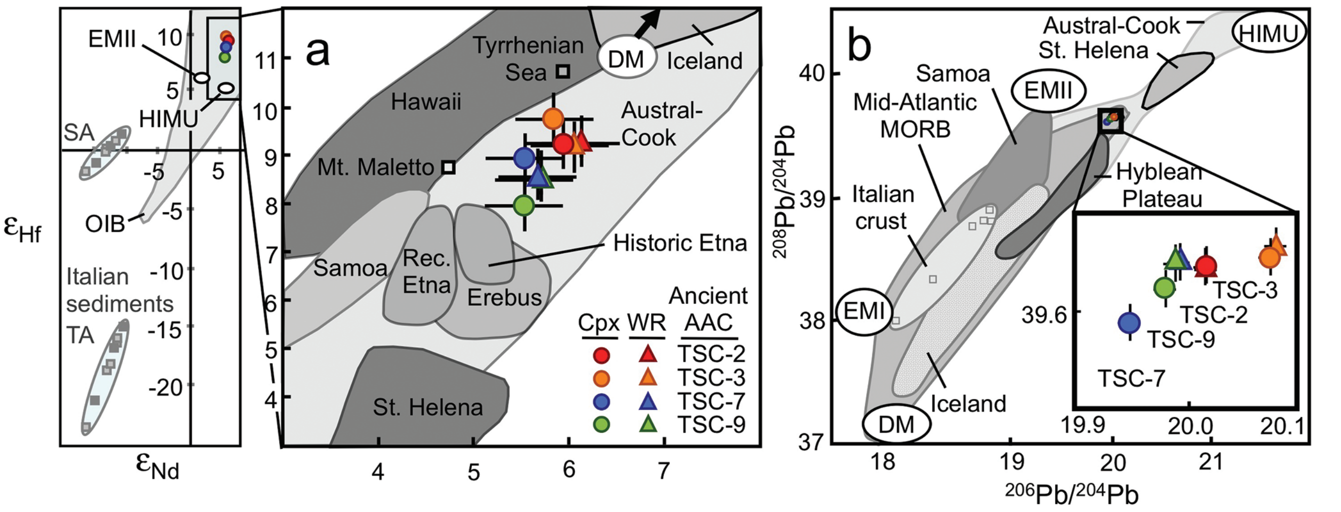

Figure 2 Ancient Etna cpx and WR data. **(a)** ε_{Hf} *vs.* ε_{Nd} for recent and historic Etna and the Mediterranean region. Historic Etna, mid-ocean ridge basalt (MORB) and ocean island basalt (OIB) fields from Lassiter *et al.*, 2003; Stracke *et al.*, 2003; Gaffney *et al.*, 2004; Huang *et al.*, 2005; Xu *et al.*, 2007; Sims *et al.*, 2008; Blichert-Toft and Albarède, 2009; Yamasaki *et al.*, 2009; Garcia *et al.*, 2010; Peate *et al.*, 2010; Chekol *et al.*, 2011; Salters *et al.*, 2011; Viccaro *et al.*, 2011); mantle components from Zindler and Hart, 1986; Salters and White, 1998; Workman *et al.*, 2004; Stracke *et al.*, 2005; Workman and Hart, 2005. Hafnium isotopic values for Italian sediments (Conticelli *et al.*, 2002; Brems *et al.*, 2013) are calculated from Nd isotopic data and both cases following the seawater array (SA) and the terrestrial array (TA) of Vervoort *et al.* (2011) are shown. **(b)** $^{208}Pb/^{204}Pb$ *vs.* $^{206}Pb/^{204}Pb$ shown with OIB and mid-ocean ridge basalt (MORB) fields, historic Etna (Viccaro and Cristofolini, 2008) and Hyblean Plateau field from Trua *et al.* (1998). Italian crustal values from Conticelli *et al.* (2002). External reproducibility is conservatively set at 0.01 for $^{206}Pb/^{204}Pb$ and 0.02 for $^{208}Pb/^{204}Pb$.

Constraints on mantle mixing processes. In spite of barometric model uncertainties, sites of cpx crystallisation (shallow crust *vs.* lower crust/mantle) can be readily distinguished by the barometry and thus provide meaningful stratigraphic context for cpx isotopic values. The lack of significant disequilibrium can be explained by magma sources feeding Etna during the period of ancient alkaline eruptive activity being either broadly isotopically homogeneous or well mixed before eruption. The few reported WR-cpx pairs from 15–30 ka (Valle del Bove sequence; D'Orazio *et al.*, 1997) show corresponding Sr and Nd isotopic equilibria (isotopic differences <0.00002 and <0.00001, respectively) and results here extend this phenomenon back an additional 200 ka.

The interpretation of limited mixing is further supported by the observed equilibrium in three of the TSC lavas between cpx and WR Pb isotopic signatures (Fig. 2b). Only one WR-cpx pair (TSC-7) exhibits isotopic disequilibrium in the Pb isotope system just outside the range of external reproducibility. Limited crustal storage time implied by Pb isotopic cpx-WR equilibria also bolsters trace element records of crystallisation from heterogeneously sourced magmas being largely preserved in this system. Trace element modelling of sources is particularly valuable in cases where source isotopic signatures are relatively well homogenised.

The restricted isotopic range of TSC cpx and WR values contrasts sharply with the variety of Pb isotopic signatures observed for plagioclase-rich and magnetic splits of a finer-grained 260 ka Etna tholeiite (SdV-1) reported by Bryce and DePaolo (2004) and olivine-hosted melt inclusions from recent (2002) eruptions (Rose-Koga *et al.*, 2012). Possible explanations include that these lavas may sample geographically different magma supplies or derive from magmas experiencing additional mixing immediately prior to eruption, as inferred from olivine in recent lavas (Kahl *et al.*, 2011). Variable Pb isotopic compositions in olivine-hosted melt inclusions could signify that minute amounts of isotopically distinct melts are simply insufficiently abundant to change the "deep", dominant isotopic signal locked into cpx.

Lack of Hf-Nd-Pb isotopic disequilibria in ancient TSC lavas between cpx-WR pairs indicates that any mixing of isotopically distinct magmas supplying ancient Etna eruptions occurred at depths preceding cpx crystallisation. Melts then rose to the surface without significant assimilation in (and associated heat exchange with) shallow reservoirs.

Conclusions

Thermobarometrically controlled elemental and isotopic analyses of clinopyroxene provide a means to reconstruct ancient magma assembly processes at Mt. Etna. Single-crystal cpx barometry places most phenocryst crystallisation within the mid-crust and permits distinction between deep and shallow processes when coupled with trace element and isotopic data. *In situ* trace element data from cpx allow for the assessment of pyroxenite *vs.* peridotite contributions to Etna magmas. Chemical signatures apparent in these ancient lavas as well as in modern products suggest that hydrated peridotite has been an important component of the magma source region over the history of this volcano. The present dataset further supports the interpretation that observed isotopic systematics in ancient Etna lavas resulted from mixing between depleted and enriched mantle sources, with volatile-bearing peridotite and pyroxenite components preferentially melting to generate volatile-rich ancient alkaline volcanism. Hf-Nd-Pb isotopic equilibria between TSC WR and cpx are consistent with a model of an ancient Etna plumbing system wherein melts were homogenised below mid-crustal depths and then rapidly transported to the surface without substantial assimilation of crustal material at pressures lower than 0.5 GPa. More extensive combinations of bulk isotopic stratigraphy with mineral barometric and trace element modelling as applied here are expected to afford opportunities to reconstruct the longevity of magmatic plumbing systems and deconvolve distinctive magma source regions feeding Mt. Etna through time.

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Additional Information

Supplementary Information accompanies this letter at www.geochemicalperspectivesletters.org/article1735

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