

Origin of 'ultra'-rocks

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INTRODUCTION

After decades of studies, it seems generally accepted that the Earth's mantle cannot be characterized by a homogeneous composition, either from a mineralogical, geochemical and isotopic point of view. It would rather be affected by re-fertilization and metasomatic processes in addition to continuous melt extraction, mostly along mid oceanic ridges. Indeed, the constant subduction and the recycling of crustal material, either continental and oceanic, necessarily causes selective enrichment of the previously depleted peridotitic mantle, resulting in an irreversible chemical and mineralogical heterogeneity (Stracke, 2021). This is clearly reflected in the variegated igneous lithologies characterised by a wide chemical compositional range. In particular, partial melting of veined lithospheric mantle sources may explain the presence of exotic liquids, whose formation cannot be ascribed to a classical four-phase mantle assemblage (Foley, 1992). Indeed, Davis et al. (2011) demonstrated that low degrees partial melting of a volatile-free Iherzolite source, which did not undergo any metasomatic events, cannot lead to the generation of melts with total alkali content higher than 3-4 wt.%.

Despite the rocks produced by these exotic melts have always attracted the attention of the scientific community, many rare lithologies continue to be a matter of discussion, and the characterisation of their mantle sources, and even their classification and nomenclature continue to be poorly constrained. A detailed and systematic study of these debated rocks might allow reaching a better knowledge of the mantle conditions, i.e., its thermal and chemical state, and of the processes responsible for their formation.

The goal of this thesis is to gain a deeper understanding of different groups of exotic and peculiar lithologies, mainly ultrabasic to basic, ultramafic and/or ultra-alkaline in composition, through a multidisciplinary investigation, based on petrographic, mineral chemical (SEM-EMP analyses), whole-rock geochemical data, together

with isotopic analyses (either on radiogenic systematics, as Sr-Nd-P, plus conventional, as C and O, and unconventional stable isotopes, as B) and petrological experiments at different pressure (from 1 to 5 GPa) and temperature (from sub-solidus to supra-liquidus) conditions.

The exotic samples investigated in this study have been collected from many sampling sites in different continental settings. They have been chosen by selecting the most variable petrographic, geochemical and isotopic differences, with the aim of performing detailed comparisons to highlight similarities and differences helpful to constrain the mantle sources and the processes involved in their formation.

The three main rock-types that have been accurately analysed are carbonatites, ultramafic lamprophyres and kamafugites. In particular, carbonatite samples come from Uganda (Toro Ankole Volcanic Province or TA) and from western Germany (Eifel Volcanic Field or EVF); ultramafic lamprophyres are from eastern Antarctica (Beaver Lake); and kamafugite from south-eastern Brazil (Alto Paranaíba Igneous Province or APIP), central Italy (Intra-Apennine Province or IAP) and Uganda (TA). Together with these samples, minor melilitites (Montefiascone Volcanic Complex, central Italy), leucitite and nephelinite (TA) have also been studied.

CARBONATITES

Carbonatites are igneous rocks containing more than 50% of primary carbonates and less than 20% of SiO₂ (Le Maître, 2002).

Carbonatites have been largely reported in the Cenozoic volcanism of Germany, as in the Eifel Volcanic Field (EVF) and in the Kaiserstuhl Volcanic Field (e.g., Schmitt et al., 2010). EVF comprises a western and an eastern district, almost parallel to each other, counting ~340 emission centres (mostly scoria cones, plus rarer lava flows and maars). EVF eastern areas display prevalently evolved terms (as phonolite and syenite), with less common carbonatites and mafic lithologies (e.g., Schmincke,

2007; Schmitt et al., 2010). Here, the Laacher See volcano showed mostly explosive and caldera-forming eruptions, coupled with the emplacement of a very limited volume of potassic lava flows (Schmincke, 2007).

The pyroclastic deposits, with phonolite composition, host a heterogeneous cargo of plutonic ejecta. The most common among them show mafic to felsic compositions, while < 5% is carbonate-bearing. On the other hand, the EVF western district shows basic to ultrabasic and mafic compositions, rarer evolved rocks, and only one occurrence of carbonatites.

The study has been mainly focused on plutonic ejecta, with a variable carbonate content (from sövite to syenite-carbonatite mixed samples; Fig. 1), coming from the Laacher See volcano. A subordinate set of leucitite samples has also been studied to highlight an eventual genetic link. Geochemical compositions vary from almost silica-free and CaO-rich rocks, up to strongly ultrabasic (moderately enriched in CaO) or to silica-rich (and CaO-poor) terms.

All the collected data suggest an evolution process for the Eifel melts, mainly constituted by two stages. Evolution started from a parental magma with a composition

similar to an olivine-melilitite (as those found in the near volcanic provinces) or leucitite (as those found in the Eifel), through a prolonged fractionation of olivine, clinopyroxene and/or melilitite, plus oxides in the latest stage (Fig. 2).

Such fractional crystallization would have allowed the melts to reach a phonolite composition, then causing an unmixing process with the production of a melt enriched in SiO₂ but poor in CaO and CO₂ (crystallizing syenite) and a melt poor in SiO₂ and enriched in CaO and CO₂ (crystallizing alvikites and sövites; Fig. 2).

The large chemical compositional range of the studied rocks would be the results of either silico-carbonatite melts (produced by minor carbonatite liquids trapped in the silicate aliquot; Fig. 1) or mechanical mixing of different percentage of silicate (as syenite) and carbonatite rocks.

ULTRAMAFIC LAMPROPHYRES

The term ultramafic lamprophyres (UML) was coined in 1986 by Nicholas Rock to indicate ultramafic, potassic to ultrapotassic melanocratic to holomelanocratic silicaundersaturated compositions, containing phenocrysts and

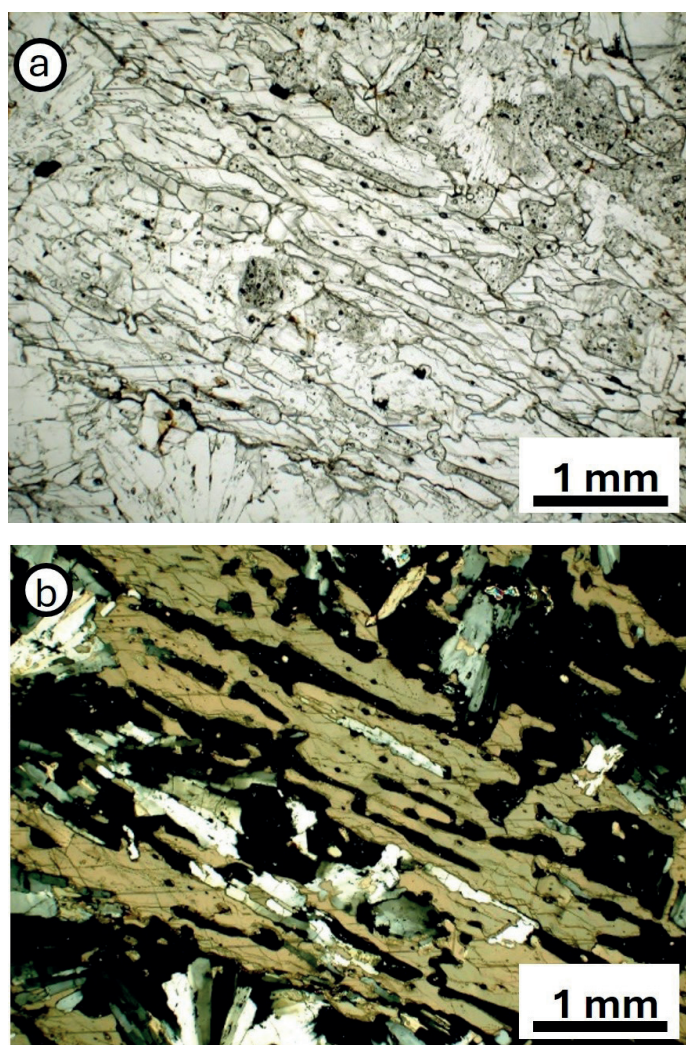


Figure 1 optical microscope micrographs of a calcite-bearing nosean syenite from Laacher See volcano a) plane polarized light; b) crossed polarized light.

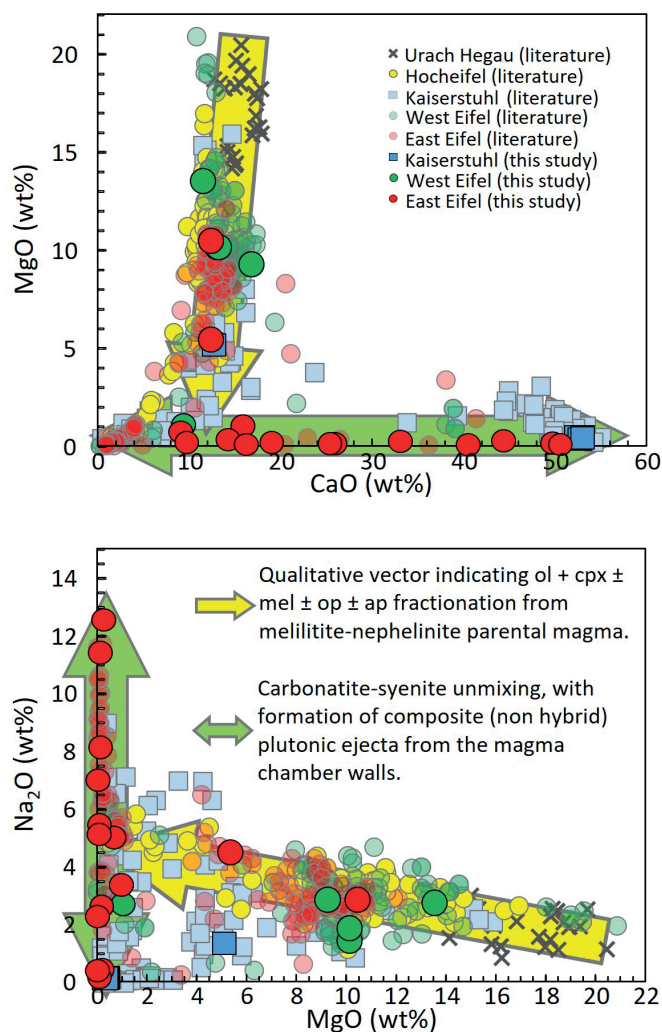


Figure 2 MgO vs. CaO and Na₂O vs. MgO Harker diagrams for EVF.

macrophenocrysts of olivine and phlogopite. UML represent another exotic igneous lithology with a still unclear classification, as the current IUGS classification includes only two UML types, grouped with the melilite-bearing rock (e.g., Le Maitre, 2002; Tappe et al., 2005).

Beaver Lake area is located along the Prince Charles Mountains, which mainly represent the East Antarctic Shield, Precambrian in age (e.g., Foley et al., 2006). The Beaver Lake area is part of the western flank of the Lambert-Amery rift, developed from Late Carboniferous during Mesozoic to Cenozoic (Foley et al., 2002; 2006).

The Beaver Lake UML products, emplaced in the Cretaceous (110-117 Ma), are represented by one dyke, two plugs (named the Konus and Novoe plugs) and two sills (named Radok-1 and Radok-2; Foley et al., 2002).

Petrographic and SEM-EDS analyses have been performed on three samples from the dyke, three from the plugs (two from the Konus Plug and one from the Novoe Plug) and four from the Radok-1 Sill. Major oxides and trace element data have been acquired for five of the most representative and less altered samples.

All samples (Fig. 3) are inequigranular and with $M > 90\%$, containing phenocrysts of olivine and microphe-nocryst or intergranular phlogopite, coupled with carbonate patches (here interpreted as a primary feature of the samples as they contain primary silicate phases). Therefore, samples must be classified as UML, and more specifically alnöite, as they also are melilite-bearing.

Among the analysed rocks, sill samples are potassic to ultrapotassic (K_2O/Na_2O ranging from 1.8 to 2.7), whereas the dike and the plug samples belong to the transitional series, having $K_2O/Na_2O < 1$. Although the K_2O/Na_2O ratios of the samples may have been affected by a post-emplacement alteration process, the sill samples are the most enriched in phlogopite, and potentially actually potassic to ultrapotassic, whereas dyke and plug samples show a phlogopite amount $< 5\%$ and probably an original much lower K_2O content.

The homogeneous petrographic and geochemical features of the samples point to similar petrogenesis, also supported by the spatial and temporal close relation. Minor differences recorded among the samples may be easily related to variable degree of partial melting.

The petrographic evidence, i.e., the presence of primary carbonate and the quite abundant K_2O -rich (as phlogopite) and Na_2O -rich phases (as nepheline; Fig. 3), together with the geochemical data, i.e., the very low silica content (33.0-35.9 wt.%) and the moderately high alkali content ($K_2O + Na_2O = 4.9$ -5.7 wt.%), may point to a source that is different from the classical peridotite mantle. It is generally accepted that the generation of melilite-bearing magmas is related to re-fertilized garnet-bearing peridotite, which has melted under vola-

tile-rich conditions (H_2O and, especially, CO_2 ; Dasgupta et al., 2006). Usually, the phase invoked to host hydrogen in the upper mantle is pargasite amphibole, which is stable up to a pressure of ~ 3 GPa (Foley & Pinter, 2018). As the depth of the Beaver Lake UML melts formation has been modelled to be higher than the spinel/garnet transition zone, at about 130 km (e.g., Foley et al., 2002), the presence of amphibole can be excluded. Instead, as phlogopite may be stable until 9-12 GPa (e.g., Pinter and Foley, 2018), it is therefore the best candidate to be in the UML mantle sources. About 15-30% of phlogopite would also guarantee the modest enrichment in K_2O content (1.5-3.3 wt.%) of the UML.

However, it is true that in two samples (dyke and plug rocks), Na_2O budget is higher than K_2O , and without amphibole to provide such element, clinopyroxene must be responsible for it. High modal contents of clinopyroxene in the mantle can be stabilized at the expense of orthopyroxene due to the infiltration of mobile carbonatite liquids in the wall-rocks (Yaxley et al., 1991). Sodic diopside can be produced by Na-rich dolomite liquids. Other than the presence of clinopyroxene, carbonatite metasomatism would guarantee the absence of orthopyroxene and the consequent low silica content of the melts, but also the CO_2 -rich conditions of the system.

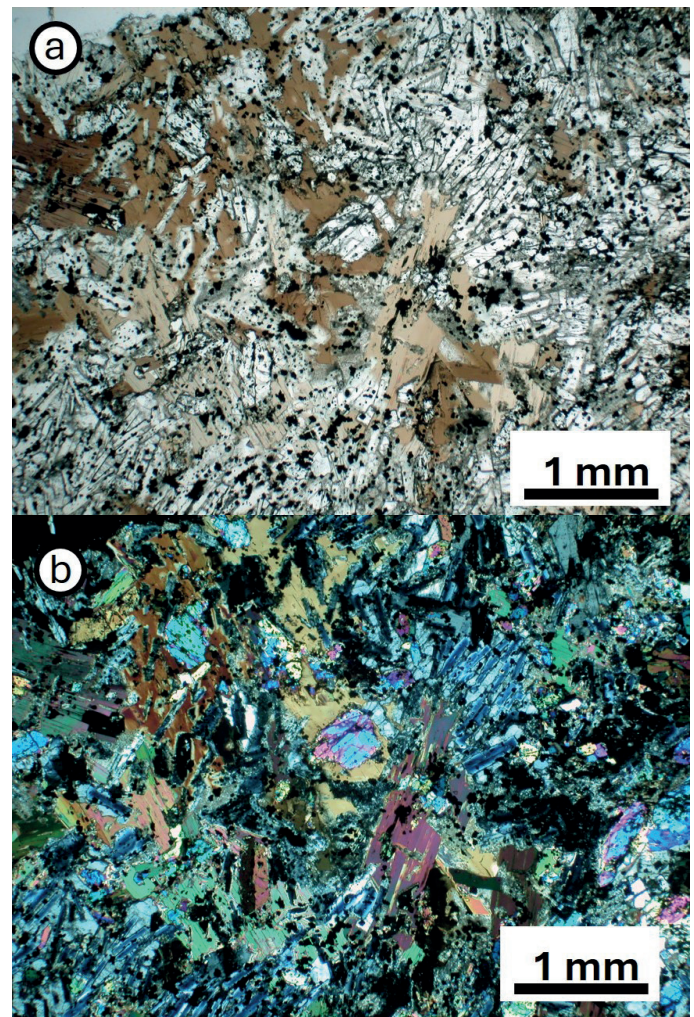


Figure 3 optical microscope micrographs of a UML from Beaver Lake, Radok-1 sill. a) plane polarized light; b) crossed polarized light.

Crustal recycling in the mantle beneath Beaver Lake area, as a consequence of the Antarctica and India collision (~1.1 Ga), could be responsible for the formation of metasomatic liquid in the UML sources. The formation of the Lambert-Amery Rift seems to be necessary both in its earlier stage for the production of exotic metasomes (phlogopite, clinopyroxene and carbonate) in the mantle and later for the melting events that generated the UML. Partial melting would have started from the metasomatic veins, which are characterized by a lower liquidus temperature, and eventually also involved the surrounding peridotite mantle.

KAMAFUGITES

The acronym kamafugite was coined by Sahama (1974), to refer to volcanic products with a silica-poor and alkali-rich (and with $K/Al > 1$) nature. The term kamafugite results from the initials of the names of three types cropping out in south-western Uganda, in Toro Ankole, along the western branch of the East African Rift System (i.e., KAtungite, MAFurite and UGandite).

Today kamafugite can be classified either according to its mineral (Le Maitre, 2002) or geochemical composition

(Foley et al., 1987). Following the IUGS guidelines (Le Maitre, 2002), volcanic rocks characterized by the presence of primary kalsilite should be named kamafugite. Kamafugite rocks also represent the second group of ultrapotassic rocks (igneous products with K_2O and $MgO > 3$ wt.%; $K_2O/Na_2O > 2$), according to the classification of Foley et al. (1987). Kamafugite should be characterized by low SiO_2 content (< 46 wt.%), coupled with low Al_2O_3 and Na_2O , and strong CaO enrichment. The $Mg\#$ [$Mg\# = Mg/(Mg+Fe)$] should be > 60 (Foley et al., 1987).

Notwithstanding the existence of these two indications, the classification of kamafugites is still debated (e.g., Lustrino et al., 2020). The diagrams used by Foley et al. (1987) to discriminate the three ultrapotassic groups appear less appropriate in light of the new data that have been produced in the last 30-35 years. The IUGS guidelines have been recently questioned by Oliveira et al. (2022), highlighting the still confuse framework.

Until today, kamafugite rocks have been found in three worldwide localities only:

- Uganda, along the western branch of the East African Rift, and in particular in the northernmost and youngest volcanic province, Toro Ankole (TA; Uganda; Rosenthal et al., 2009). In TA, the volcanic activity started about 188 ka, and emplaced principally pyroclastic deposits, coupled with rarer lavas, often showing extreme compositions, as carbonatites, foidites, melilitites and kamafugites (Rosenthal et al., 2009; Innocenzi et al., 2024). These products embed exotic nodules, ultramafic in nature (from clinopyroxenite to glimmerite, with minor accessory phases as Ti-magnetite, apatite, perovskite and carbonate), interpreted either as mantle xenoliths or cumulate;

- Central Italy, in the Intra-Apennine Province (IAP), in San Venanzo and Cupaello (Lustrino et al., 2020). IAP (0.2-0.8 Ma) is an igneous district situated in the central Apennine Chain and composed of small monogenic to polygenic emission centres, probably developed as a consequence of the extensional faults of Plio-Pleistocene age. San Venanzo and Cupaello represent the type locality for two kamafugite endmembers (venanzite and coppaelite);

- South-eastern Brazil, in the Alto Paranaíba and Goiás Alkaline Province (Brod et al., 2005; Melluso et al., 2008). In particular, APIP is a Late Cretaceous and K_2O -rich province, probably related to an extensional tectonic regime. Intrusive bodies, pyroclastic products and lavas have been emplaced along the Neoproterozoic Brasília belt, with a composition varying between carbonatite, kimberlite, kamafugite, lamproite, dunite and alkali pyroxenite.

To these, we should probably add the Montefiascone Volcanic Complex (0.3-0.2 Ma), part of the Vulsini Vol-

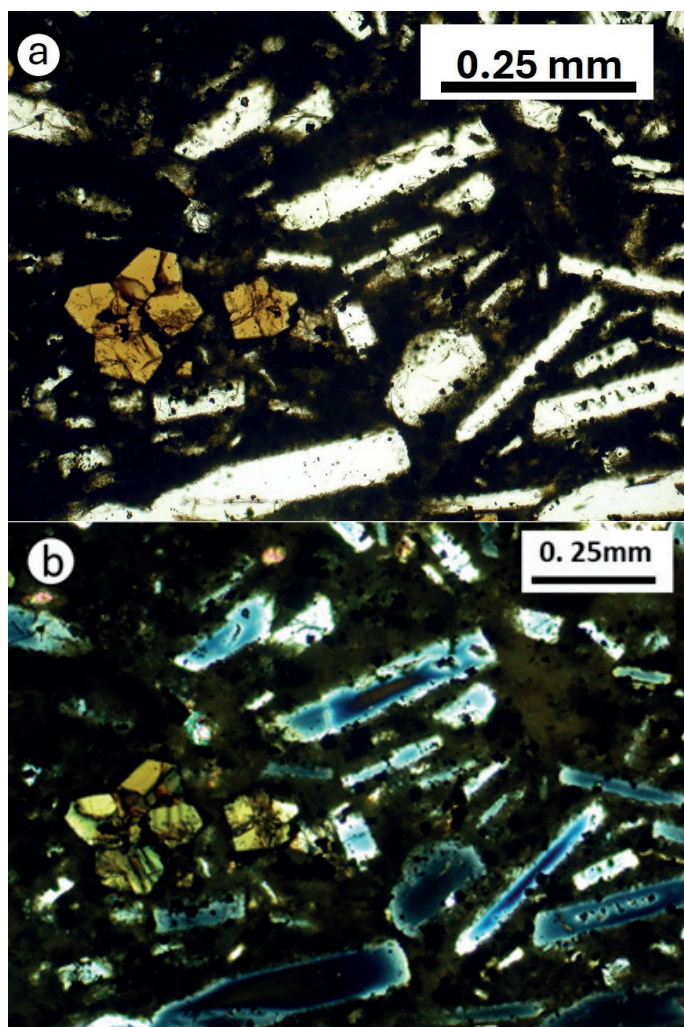


Figure 4 optical microscope micrographs of a porphyritic lava with euhedral microphenocrysts of melilite and perovskite, in a hypocrystalline altered groundmass. a) plane polarized light; b) crossed polarized light.

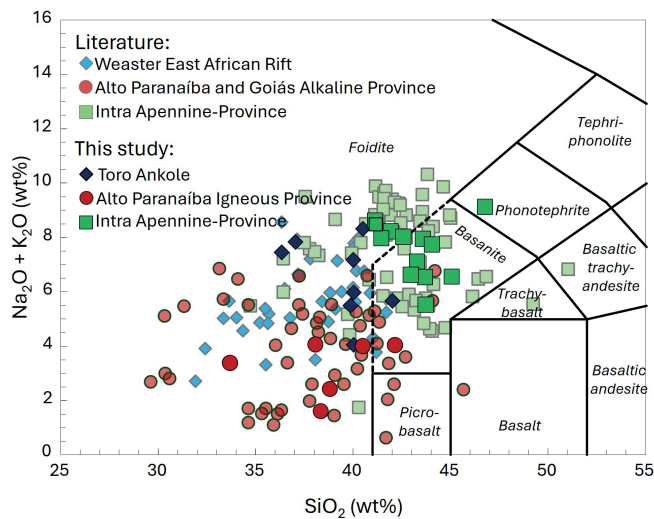


Figure 5 TAS diagram for the TA, APIP and IAP kamafugites, either from this study (bold symbols) and literature data.

canic District and the Roman Comagmatic Region (di Battistini et al., 2001). In this thesis, EMP analyses have been carried out on lava samples previously classified as leucite melilitite (di Battistini et al., 2001), highlighting the anhedral kalsilite crystals in their groundmass. Therefore, these samples should be reclassified as kamafugite, following the IUGS guidelines.

Kamafugites from the three provinces show similar parageneses, mainly differing in their olivine content, which ranges from less than 1% in Cupaello lavas (IAP) to 25% (San Venanzo products, IAP). Kamafugite samples are ultrabasic to basic, with a silica content of 33.7–46.8 wt.%, and show a variable enrichment in CaO (= 4.6–16.1 wt.%) and $\text{Na}_2\text{O} + \text{K}_2\text{O}$ (= 1.6–9.1 wt.%; Fig. 5). Moreover, IAP and TA kamafugite have a potassic or ultrapotassic affinity ($\text{K}_2\text{O}/\text{Na}_2\text{O}$ ranging from 1.8 to 9.0 in TA and from 7.3 to 24.1 in IAP). The $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratio for APIP rocks is much lower (0.4–2.7).

While TA and APIP kamafugites share comparable budget for major oxides (e.g., Fig. 5) and trace elements (i.e., high TiO_2 , from 3.6 to 5.8 wt.% in TA and from 3.6 to 6.4 wt.% in APIP, and MgO, from 6.4 to 22.2 wt.% in TA and from 13.5 to 20.5 wt.% in APIP), IAP counterparts often display different geochemical features (as lower TiO_2 = 0.7–1.1 wt.% and MgO = 9.9–14.0 wt.%). Kamafugite lavas from Uganda and Brazil also exhibit similar trace element enrichments and overlapping patterns on the Primitive mantle normalized diagrams, with negative peaks for K and P. On the other hand, Italian variants have deep trough for Nb-Ta, P, Ti and Eu, coupled with positive anomalies for Th, U, and Pb.

Radiogenic isotope analyses highlighted enriched values for all the provinces. $^{87}\text{Sr}/^{86}\text{Sr}$ isotopes ratios for TA and APIP, once again, is overlapping (~0.7056 and 0.7051–0.7057, respectively). IAP samples show the most radiogenic Sr values (0.7105–0.7113).

$^{143}\text{Nd}/^{144}\text{Nd}$ values are generally lower in Brazilian kamafugite (0.51213–0.51227) than in Ugandans

(~0.512531), probably due to the older age of metasomatism. Again, the lowest values are related to IAP products (0.51200–0.51210). Pb isotope ratios are also slightly different ($^{206}\text{Pb}/^{204}\text{Pb}$ ~19.40 in TA, 17.59–19.89 in APIP and 18.75–17.76).

$\delta^{11}\text{B}$ analyses have been conducted on IAP and TA lavas, as B is considered an effective tracker for mantle heterogeneities, and in particular crustal recycling (e.g., Agostini et al., 2021). $\delta^{11}\text{B}$ for TA kamafugites match well with mantle typical values (MORB from -6 to -8‰; OIB = -5 to -10‰; Agostini et al., 2021 and references therein) but the B ratios of the related lithologies, as glimmerite and clinopyroxenite nodules and carbonatites, reaches positive values, up to 6.6‰, highlighting altered oceanic crust \pm marine limestone in the mantle source. IAP $\delta^{11}\text{B}$ (Cupaello = -7.6 to -7.9 ‰ and San Venanzo = -7.5 to -8.8‰) mainly overlap with the OIB range, with slightly lighter values that may point to sedimentary input in the mantle underlying central Italy.

The data suggest a metasomatized source involving carbonate, with similar mantle parageneses (phlogopite, clinopyroxene), suggested by the samples affinities. The heterogeneities highlight variable contributions to the melt from clinopyroxene and phlogopite in the three provinces, and different amounts of accessory phases, such as apatite and Ti/Fe-bearing opaques, that play a key role for melt compositions. Brazilian kamafugites require a slightly different source, with stronger contribution of olivine from the former lherzolitic mantle and/or a higher degree of partial melting.

Experimental study

As petrological studies provide further evidence to be combined with data from the natural samples, a set of near-liquidus experiments and a set of partial melting experiments on kamafugite have been carried out.

Near-liquidus experiments were carried out on four of the most representative and less altered kamafugite samples (one from TA, one from APIP and two from IAP, both from San Venanzo and Cupaello). Minerals resulting from the experimental runs should be in equilibrium with the melts and should represent the main phases of the kamafugite sources. Indeed, chosen samples have Mg# > 60 (considered the reference value for primitive exotic melts; Foley, 1999) and show no fractionation trend among the studied compositions. Moreover, petrography highlighted no or very few xenoliths and cumulate. Therefore, they represent the best approximation of the real melts.

Experiments support the idea of quite similar mantle sources for the three volcanic provinces, with a paragenesis different from the classical peridotite. A total lack of orthopyroxene has been recorded, while phlogopite

and clinopyroxene are abundant in all four experimental runs.

Compositional heterogeneities recorded among the worldwide kamafugites are reflected in the experimental results, which highlight a variable modal amount of phlogopite and clinopyroxene in the different mantle sources. For TA and APIP, the source composition is probably a phlogopite-rich (TA = 30-40% and APIP = 20-30%) clinopyroxenite, while for IAP a glimmerite hosting 40-50% of clinopyroxene. Olivine is found as a liquidus phase only in APIP (10-20%) and SAV (10%) runs, justifying the higher MgO contents.

Experimental results also outlined variable abundances of different accessory phases in the four samples. SAV runs show no minor phases, whereas in all the other experiments, apatite and Ti- and Fe-rich oxides are quite common. The mantle underlying Toro Ankole is probably the most enriched in accessory phases (3% of apatite, 4% of titanite and 5% Ti-magnetite). APIP mantle source probably hosts 2% of apatite and 5% of ilmenite (or Ti-magnetite).

Starting from these results, a possible source paragenesis has been reconstructed for the three provinces. The chosen compositions are: a phlogopite-bearing clinopyroxenite (with apatite, ilmenite, magnetite and titanite) for TA; an olivine clinopyroxenite (with phlogopite, apatite and ilmenite) for APIP; and to a clinopyroxene-bearing glimmerite (with apatite and magnetite) for IAP. Partial melting experiments were carried out at 2.7 and 5 GPa, reconstructing the conditions of the lithospheric mantles underlying the studied volcanic provinces, to test if the resulting melts actually match with the natural kamafugite. The resulting glass composition is influenced by the degree of partial melting (as temperature ranges from 1200 to 1550°C) and by the percentage of each phase that contributes to the melt.

Experimental glasses, for a partial melting degree from 20-30 to 70% (temperature = 1350 and 1400°C at 2.7 GPa, and = 1450-1550°C at 5 GPa), show an ultrabasic to basic, potassic to ultrapotassic and CaO-rich composition. MgO is generally quite high (up to 16.5 wt.%).

There is a quite good overlap between the glasses and the natural kamafugites compositions, in particular for Brazilian and Ugandan lavas.

Experiments demonstrated that slightly different degrees of partial melting of a clinopyroxenite (\pm olivine), variably enriched in phlogopite and accessory minerals (i.e., apatite, ilmenite and titanite) could represent the mantle source paragenesis of kamafugite magmas, also justifying the different features of the worldwide localities kamafugites.

MAIN IMPLICATIONS

All the analyses that have been carried out highlight the exotic nature of the samples. Petrography and mineral chemistry point out the presence of rare phases, as primary carbonate (found not only in the carbonatite samples, but also in the groundmass of kamafugites, ultramafic lamprophyres and melilitites), kalsilite, melilite and perovskite (diffuse in kamafugite, ultramafic lamprophyres and melilitite samples). Whole-rock geochemistry indicates strongly ultrabasic (carbonatites), ultrabasic (Ugandan and Brazilian kamafugite, ultramafic lamprophyres) and basic (Italian kamafugites) compositions, mostly characterized by a potassic to ultrapotassic affinity (K_2O up to 8.36 wt.% in Italian kamafugites, $K_2O/Na_2O < 24$). Moreover, all samples show a variable, but often marked, enrichment in trace elements. Also, the isotopic data indicates strongly radiogenic $^{87}Sr/^{86}Sr$ and $^{206}Pb/^{204}Pb$, coupled with low values of $^{143}Nd/^{144}Nd$ isotopic ratios. Only two carbonatites represent an exception, having $^{87}Sr/^{86}Sr < BSE$ (present-day Bulk Silicate Earth). $\delta^{11}B$ values roughly fall in the mantle range (-8.3 to -3.3‰; Agostini et al., 2021), except for a few Ugandan rocks, having heavier values ($\delta^{11}B = -1.9$ to 6.6‰).

Even if samples cover a large compositional spectrum, multi-stage refertilization events must have occurred and must have strongly modified all the lithospheric mantle sources. Recycling of various lithologies, as marine limestone, caused by more or less old subductions, could explain either the carbonated metasomatism, necessary in all the analysed volcanic districts, but also the phlogopite and diopsidic clinopyroxene-rich veins, necessary in the formations of kamafugites, ultramafic lamprophyres and melilitites.

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