Pliocene-Quaternary volcanic rocks of NW Armenia: magmatism and lithospheric dynamics within an active orogenic plateau

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Abstract

The Pliocene-Quaternary volcanic rocks of Armenia are a key component of the Arabia-Eurasia collision, representing intense magmatism within the Turkish-Iranian plateau, tens of millions of years after the onset of continental collision. Here we present whole rock elemental and Nd-Sr isotope data from mafic, intermediate, and felsic lava flows and cinder cones in Shirak and Lori provinces, NW Armenia. Magmatism appears to be controlled locally by extension related to major strike-slip faults within the plateau. Major and trace element results show that the three series – valley-filling medium-K alkali basalt flows, ridge-forming andesite to rhyolite flows, and andesitic cinder cones – form a compositional continuum linked by a crystallisation sequence dominated by two pyroxenes, plagioclase and amphibole. There is petrographic and major and trace element evidence for magma mixing *processes and potentially crustal contamination by Mesozoic-early Cenozoic arc-related rocks, which has not significantly affected the isotopic signature. Modelling of the basaltic rocks indicates that they formed by moderate degrees of partial melting (~3-4 %) of an incompatible element enriched, subduction-modified, lithospheric mantle source. Samples have a distinctive high Zr/Hf ratio and high Zr concentrations, which are an intrinsic part of the source or the melting process, and are much more commonly found in ocean island basalts. Regional models for magmatism often argue for whole-scale delamination of the mantle lithosphere beneath Eastern Anatolia and the Lesser Caucasus, but this scenario is hard to reconcile with limited crustal signatures and the apparent lack of asthenospheric components within many studied centres.*

Highlights

- Whole-rock study of Pliocene-Quaternary lavas from the Armenian Highlands
- Compositional range controlled by fractional crystallisation, magma mixing and possible crustal contamination
- Low-degree melting of a shallow metasomatised lithospheric mantle source
- Exploring triggers for <10 Myr increase in magmatic activity in the Arabia-Eurasia collision zone

Keywords

Armenia; geochemistry; petrogenesis; orogenic plateau

1. Introduction

Orogenic plateaux such as the modern Turkish-Iranian, Bolivian Altiplano-Puna and Tibetan plateaus form in response to plate convergence and collision, and represent a primary topographic feature of the continents. In spite of their thickened crust, plateaus are also sites of intense, ultimately mantle-derived magmatism (e.g. Williams et al., 2004; Mo et al., 2007). Such magmatism is often attributed to asthenospheric upwelling following the break-off of the subducted oceanic slab (e.g. Keskin, 2003), or the delamination of the lithosphere inboard of the plate suture (e.g. Kay and Kay, 1993).

The Turkish-Iranian plateau (Fig. 1) is a product of the Cenozoic Arabia-Eurasia collision, and magmatism post-dating initial collision is particularly voluminous from the Late Miocene until the present day, in numerous locations across eastern Turkey, Armenia, and much of Iran (Fig. 1). Erupted products range from mafic to felsic, and sodic to ultrapotassic (Pearce et al., 1990; Karapetian et al., 2001; Davidson et al., 2004; Azizi and Moinevaziri, 2009; Saadat et al., 2011; Saadat and Stern, 2012; Allen et al., 2013). In nearby Georgia and the Greater Caucasus, the most recent magmatism appears to have started slightly earlier, in the Middle Miocene, and continued with some gaps in the record until recent times (Lebedev et al., 2006a,b; 2007; 2008a,b; Adamia et al., 2011).

This plateau-wide ‘recent’ magmatism may be only partly explained by partial melting of asthenospheric or mantle lithosphere sources due to break-off of the southern Neo-Tethys slab (e.g. Keskin, 2003; 2007; Şengör et al., 2008; Dilek et al., 2010; van Hunen and Allen, 2011). Miocene to recent magmatism extends to at least 500 km from the Bitlis-Zagros suture zone, a spatial scale akin to the Cenozoic ‘ignimbrite flare-up’ of the western United States (Johnson, 1991). The wide extent of Miocene-Quaternary magmatism hundreds of kilometres from the suture zone indicates that whole-scale lithospheric delamination (Pearce et al., 1990), or other unrecognised processes may be collectively responsible for magmatism in this region.

To further consider the origin of such magmatism, this paper focuses on Armenia (Fig. 1), where recent volcanism has been under-represented in the international literature. We present whole rock elemental and Nd-Sr isotope data from three series of mafic to felsic Pliocene-Quaternary volcanic rocks in the north of Shirak and west of Lori administrative provinces in the northwest of the country (herein referred to as 'Shirak') (Fig. 2). The local tectonic setting and relationship to magmatism is highlighted, alongside discussion on magmatic evolution, mantle sources and partial melting. We finish by assessing how the results fit with regional geodynamic models.

2. Geological setting, structure, and petrography

2.1. The Turkish-Iranian plateau

The Turkish-Iranian orogenic plateau (Fig. 1) developed following the closure of the Neo-Tethys Ocean (Şengör and Kidd, 1979). The basement of the plateau consists of Mesozoic to Early Cenozoic accretionary belts and arc rocks and also older, Gondwanaland-related micro-continental fragments that all accreted to the southern margin of Eurasia. It is widely assumed that the Neo-Tethys oceanic crust was divided into a northern and a southern segment; the former closed either during the Late Cretaceous (Lordkinpanidze, 1980; Keskin, 2008) or the Paleocene-Early Eocene (Sosson et al., 2010). The two segments of Neo-Tethys were separated by micro-continental fragments such as the South Armenian Block and Tauride-

Anatolide terranes (Sosson et al., 2010). Destruction of the southern segment of Neo-Tethys brought Arabia and Eurasia together along the Bitlis-Zagros suture zone (Fig. 1).

The timing of initial collision between Arabia and Eurasia is debated, although most estimates range between 35 and 20 Ma (Agard et al., 2005; Allen and Armstrong, 2008; Morley et al., 2009; Okay et al., 2010; Ballato et al., 2011; McQuarrie and van Hinsbergen, 2013). Marine carbonates deposited across much of central Iran and eastern Turkey in the Early Miocene indicate that growth of the orogenic plateau is a later phenomenon (Bottrill et al., 2012). Deformation is presently focussed on the plateau margins, from the Greater Caucasus and Alborz in the north to the Zagros in the south, with no active crustal thickening or thinning occurring between (Jackson et al., 1995; Allen et al., 2011). Lithospheric thickness is highly variable, from >200 km in Iran near the Zagros suture, to only 60 km or less in eastern Anatolia (Priestley and McKenzie, 2006; Angus et al., 2006). The current height of the plateau, ~1750 m above sea level, has been attributed to a combination of Late Cenozoic crustal shortening, and also to the detachment of mantle lithosphere and/or subducted Tethyan slabs beneath the plateau, allowing the upwelling of hot, buoyant asthenosphere beneath the region (Keskin, 2003).

The Cenozoic magmatic record of the plateau is divided into several stages (see Dilek et al., 2010; Chiu et al., 2013 for reviews). First is an Eocene ‘flare-up’ of arc magmatism immediately prior to the onset of continental collision, focussed predominantly on the Urumieh-Dokhtar arc in southwest Iran, the Lut Block, Kopeh Dag and Alborz regions of eastern and northern Iran (Verdel et al., 2011), and also in Armenia (Lordkinpanidze et al., 1988). The flare-up has been attributed to back-arc extension (Vincent et al., 2005), an episode of flat subduction (Berberian and Berberian, 1981), perhaps coupled with enhanced slab roll-back (Verdel et al., 2011), or break-off of the northern Neo-Tethyan slab in Armenia and North-Central Turkey (Keskin et al., 2008; Sosson et al., 2010). The second stage is a magmatic 'gap' which comprised some 20-30 Myr of limited magmatic activity between the Eocene and the Late Miocene as continental collision proceeded (Verdel et al., 2011; Richards et al., 2011). The third and final stage is the aforementioned upsurge of mantle-derived volcanism from the Middle to Late Miocene until the present day (Chiu et al., 2013), which forms the basis of this study.

2.2. Basement and structure in Armenia

Armenia, including the Lesser Caucasus mountain range, lies at the northern side of the plateau (Figs. 1, 2). Southern Armenia is underlain by the aforementioned South Armenian Block (SAB), a micro-continental fragment rifted during the early Mesozoic, separating the northern and southern branches of the Neo-Tethyan seaway (Stampfli, 2000). The SAB consists of Proterozoic gneisses, mica schists and amphibolites partially overlain by Devonian to Jurassic sediments, Jurassic and younger ophiolitic material, and Paleocene to Early Oligocene volcanic rocks related to subduction of the southern branch of Neo-Tethys (Rolland et al., 2009). In the north of Armenia, the Armenian Highlands represent the former active continental margin of Eurasia and contain arc and discontinuous ophiolite sequences formed during the closure of the northern branch of the Neo-Tethyan seaway (Adamia et al., 1981; 2011). The largest tract of ophiolitic material in Armenia forms the Sevan-Akera suture zone, a 400 km-long boundary between the SAB and the Mesozoic arc of the Lesser Caucasus to the north. Immediately south of our field area, between Amasia and Stepanavan, is a belt of blueschist-facies mélange (part of the Sevan-Akera ophiolite suite), tectonically overlain by Jurassic to Cretaceous mafic rocks, and two sequences of Cretaceous to Early Oligocene subduction-related volcano-sedimentary rocks (Rolland et al., 2009).

Following the Eocene amalgamation of the Armenian crustal blocks (Rolland et al., 2009), north-directed subduction of the southern Neo-Tethys terminated along the Bitlis-Zagros suture, some 300 km south of Armenia. After the last subduction-related magmatism, the magmatic ‘gap’ in Armenia extended until the Late Miocene (~10 Ma), based on groundmass and mineral K-Ar ages from the oldest volcanic rocks of the Gegham Highlands (Arutyunyan et al., 2007). The most voluminous volcanism is of Pliocene-Quaternary age, covering much of Aragatsotn, Shirak, Kotayk, Gegharkunik, and Syunik provinces, an area >10,000 km^2 (Mitchell and Westaway, 1999; Karapetian et al., 2001; Lebedev et al., 2011) (Fig. 2).

2.3. Pliocene - Quaternary magma series

The geology of NW Armenia was first studied in detail during Soviet times (Kharazyan, 1983). The oldest Pliocene – Quaternary volcanism is represented by poorly-exposed rhyolites and obsidians at Aghvorik (Fig. 2), covered by dolerites which drape much of the lowest topography in Shirak, often part-filling river valleys for tens of kilometers. Relationships with sedimentary deposits dated using mammalian fossils lead authors to conclude that these mafic lavas were of Late Pliocene age (Kharazyan, 1983 and references therein). A K-Ar age determination from dolerite from the Akhurian river basin within our study area gave a result of 2.5 ± 0.2 Ma (Chernyshev et al., 2002). Groundmass K-Ar results from numerous mafic to felsic volcanic rocks across the Javakheti Highlands in southern Georgia gave ages of 4.6 ± 0.2 to 1.54 ± 0.10 Ma (Late Pliocene to Quaternary) (Lebedev et al., 2008a,b). The mafic rocks in Armenia and southern Georgia may have resulted from fissure eruptions (Jrbashyan et al., 1996), but the actual source of these lavas has never been found, and may be buried by younger flows.

On top of the plateau-like topography formed by the mafic flows in Shirak, are hills of intermediate-felsic composition of up to 600 m prominence, including the north-south trending Javakheti, or Kechut, ridge (Fig. 2). Kharazyan (1983) distinguished two units within the Armenian part of the Javakheti ridge. The first unit (Lower Kechut Suite) is said to contain two-pyroxene basaltic andesites, andesites and hornblende andesites which cover the valley series and are thus assumed to be of Early Pleistocene age (Kharazyan, 2005). SHRIMP U-Pb zircon dating of andesitic vent-proximal ash and breccia layers deposited on the eastern flank of the Javakheti ridge in Lori province at the Karakhach archaeological site (Fig. 2) gives maximum eruption ages of 1.94 ± 0.05 to 1.80 ± 0.03 Ma (Presnyakov et al., 2012). This study also noted two Eocene zircons (40-50 Ma) consistent with Eocene arc rocks found to the east of the study area, plus five Proterozoic grains, consistent with an origin in the underthrust SAB. It is uncertain whether or not these old grains were from xenoliths ripped up during explosive volcanism or zircons assimilated during magma ascent. The Javakheti Ridge extends into southern Georgia, where it is higher and has a sharp little denuded topographic profile compared to further south. Groundmass K-Ar dating revealed younger ages of <1 Ma for the Samsari volcanic centre to the north of the Javakheti Ridge (Chernyshev et al., 2006). Kharazyan (1983) also defined an Upper Kechut Suite supposedly containing lavas erupted from cinder cones on the western part of the ridge, and considered to be of Middle Pleistocene age (Kharazyan, 2005). We did not find clear evidence of this unit during our studies. The only other Pliocene-Quaternary volcanic products in this part of Shirak are cinder cones to the west of the Javakheti Ridge around Lake Arpi (Fig. 2), which are estimated to be of Early-Middle Pleistocene age, based on relationships with the mafic rocks and some river terraces (Kharazyan, 1983).

Overall there have been no comprehensive geochronological studies carried out in Shirak using a single reliable technique, unlike those for similar sequences in Georgia (Lebedev et al., 2008a,b). A lack of continuous exposure hampers judgement of the relative age of the different lavas. We have decided to sub-divide the entire Pliocene-Quaternary suite close to the Javakheti Ridge into three components: mafic flows (the dolerites) largely covering the topography developed on the basement (herein termed the *Valley Series*); more evolved flows built up into hills above the mafic flows (*Ridge Series*); and scattered cinder cones (*Cone Series*) (Fig. 2).

2.3.1. Valley Series

The Valley Series consists of mafic flows with a maximum cumulative thickness of 200 m in eastern Lori province (Fig. 2). In the study area, the exposed sequence comprises at most four or five stacked flows up to 40 m thick in total, and directly overlies the Sevan-Akera suture and ophiolite sequence in the Dzoraged gorge and near Amasia (Fig. 2). We have collected samples from these locations, as well as from Lake Arpi and near Tashir in western Lori Province (Fig. 2). Valley Series samples collected from near Lake Arpi, Amasia, and Tashir (Fig. 2), consist of vesicular sub-ophitic dolerites with rare clinopyroxene or optically zoned plagioclase phenocrysts, set in a groundmass of 1-2 mm grain size consisting of clinopyroxene, plagioclase and oxides (see Supplementary Item 1). Clinopyroxene is commonly rimmed or almost totally replaced by red-brown amphibole, and there is occasional interstitial quartz and rare rounded quartz blebs. Some samples contain very rare iddingsite crystals, and the few olivines found in thin section are rounded and <0.5 mm in diameter.

2.3.2. Ridge Series

Magmatism in this series is restricted to intermediate to felsic lavas built up into prominent topographic features. The highest of these is the north to south-trending Javakheti Ridge (Fig. 2), consisting of rounded and glacially eroded peaks reaching ~3100 m above sea level, following a clear north-south trend running north into Georgia (Fig. 2). A second series of hills lie north of the Akhuryan River parallel to the Georgian border, but these are more topographically muted, reaching a maximum elevation of ~2400 m. Rock types from both ridges are almost exclusively andesitic to dacitic flows, with rare black dacites and rhyolitic obsidians (Karapetian et al., 2001). Analysed flows range from basaltic trachyandesites to dacites, and there is much compositional and textural variation. Some of the least evolved samples (<60 wt.% SiO_2) contain 1-3 mm phenocrysts and glomerocrysts of clinopyroxene (rarely orthopyroxene) and plagioclase, set in a flow-banded groundmass of plagioclase, clinopyroxene and Fe-Ti oxides, with accessory apatite and zircon. More evolved samples tend to contain significantly more plagioclase phenocrysts, and several have abundant 1-3 mm euhedral green-brown pleochroic amphiboles, but the presence of amphibole is not ubiquitous even in samples of similar SiO_2 and MgO concentrations. Many plagioclase crystals are sieve-textured and some have corroded margins, usually taken to imply the occurrence of magma mixing processes (Tsuchiyama, 1985; Tepley et al., 1999), and many crystals are also optically zoned. Other evidence for magma mixing is the common occurrence of rounded quartz blebs with dark reaction coronas, and ubiquitous opaque rims on primary hornblende crystals (Tepley et al., 1999; Supplementary Item 1). A black dacite contains a few clinopyroxene glomerocrysts, but mostly consists of a fine-grained groundmass dominated by prismatic to acicular feldspars. The Ridge Series includes the only crustal xenoliths noted from our sampling, at a single site near Darik, north of Lake Arpi (Fig. 2). The xenoliths consist of a coarse-grained groundmass of plagioclase, clinopyroxene, oxide, and rare iddingsite after olivine; and 2-3 mm phenocrysts of clinopyroxene and plagioclase, most consistent with inclusion of material from the Valley Series. A slight chilled margin is observed within the host.

2.3.3. Cone Series

Scattered cinder cones are present in the Akhuryan valley near Lake Arpi (Fig. 2), often tens of metres high and several hundred metres across. We have collected samples from Sepasar, Eznasar, and Kaputkogh cones (Fig. 2). Most are composed of poorly welded unsorted glass or scoria fragments, and more coherent scoria bombs reaching a few tens of centimetres in diameter. The Cone Series compositions mirror the least evolved of the ridge series, the majority of samples containing differing proportions of plagioclase and clinopyroxene as phenocrysts and groundmass (Supplementary Item 1). Bombs from Kaputkogh and Eznasar contain ubiquitous mm-scale rounded quartz xenocrysts, whereas those at Sepasar appear more mafic, and devoid of foreign material.

3. Analytical methods

Samples were powdered in an agate ball mill at Durham University. Major element analysis was conducted on fused glass beads using a PANalytical Axios Advanced X-Ray Fluorescence (XRF) spectrometer at the University of Leicester. Leftover fractions of the powder from XRF analysis were digested using a standard HF and HNO_3 technique prior to trace element analysis. Solutions were run on a Thermo X2 inductively-coupled plasma mass spectrometer (ICP-MS) at the Northern Centre for Isotopic and Elemental Tracing (NCIET) at Durham University. Accuracy, precision, and reproducibility were monitored using blanks, multi-run and within-run duplicates, Re-Rh spike solutions, and five international reference standards. Standard W2 (n = 15) gave first relative standard deviations of 5% or better for most transition metals (excepting 10% for Sc, 12% for Cr, 6% for Ni), the large ion lithophile elements (LILE) and the rare earth elements (REE) (7% for La, 6% for Ce). Elemental results are recorded in Table 1 and Supplementary Item 2, the latter also containing standard results.

Radiogenic Nd and Sr isotope analysis was conducted at NCIET, with column chemistry for elemental pre-concentration based on the method of Dowall et al. (2007). Powders were digested in HF and HNO_3, and solutions run through 1 ml pipettes containing several drops of dilute Sr-spec resin to collect the Sr-bearing fraction. The high field strength element (HFSE)- and rare earth element (REE)-bearing fraction from these columns was run through 10 ml Bio-Rad polypropylene columns containing 1 ml of Bio-Rad AG1-X8 200-400 mesh anion-exchange resin. Neodymium was collected as part of a general rare earth element fraction. Analysis was conducted on a Thermo Neptune Mass Collector ICP-MS. Strontium was run in a single batch during which time blanks averaged 88 pg Sr (n = 6). International reference standard NBS987 gave a mean of $^{87}Sr/^{86}Sr = 0.710263 \pm 0.000012$ (2σ, n = 12), comparable to a preferred value of 0.710240, and providing a minimum uncertainty of 16 ppm (2σ). No correction was applied to the final results. Neodymium was run in two separate batches, with blanks averaging 10 pg Nd (n = 6). During the first run, a combination of the J&M standard and a Sm-doped version gave a mean $^{143}Nd/^{144}Nd = 0.511098 \pm 0.000007$ (2σ, n = 13), and a minimum uncertainty of 13 ppm (2σ). During the second run,

$^{143}Nd/^{144}Nd$ = 0.511100 ± 0.000007 (2σ, n = 13), giving an uncertainty of 14 ppm (2σ). For consistency between the runs, all results were normalised to a preferred value of 0.511110. Results are presented in Table 2.

4. Geochemistry

4.1. Sample freshness

We collected the freshest available samples at each locality, and this is reflected in loss-on-ignition (LOI) values typically <1 wt.%, mostly un-weathered feldspars and mafic minerals, and an overall lack of sericite and chlorite in thin section. Major and trace element data, particularly element vs. SiO_2 plots (see Section 4.2), have trends consistent with magmatic processes, particularly for CaO, MgO, K_2O, and Na_2O, as opposed to the widespread scatter expected during sub-solidus alteration which easily mobilises these elements (e.g. Cann, 1970; Pearce, 1996). $^{87}Sr/^{86}Sr$ isotope results are commonly affected by hydrothermal alteration, but here are depleted with no sign of a trend towards high $^{87}Sr/^{86}Sr$ at constant $^{144}Nd/^{143}Nd$ (Table 2); nor is there a correlation between LOI and isotopic ratios. Furthermore, the samples were erupted in an intra-continental high plateau so have not interacted with high $^{87}Sr/^{86}Sr$ seawater or been exposed to tropical weathering.

4.2. Major and trace element characteristics

4.2.1. Valley Series

The Valley Series lavas (~49-55 wt.% SiO_2) are mostly mildly alkaline trachybasalts based on the total alkali-silica classification (Le Bas et al., 1986; Fig. 3a), and belong to the medium-K series of Peccerillo and Taylor (1976; Fig. 3b). They are evolved, with 4-7 wt.% MgO and low molar Mg# from 46 to 58. Overall the samples have low TiO_2, moderate-high Al_2O_3, and a sodic character (Na_2O/K_2O = 2.7-4.1) (Fig. 4). Trace element abundances and trends can be seen on Figure 5. Also, the lavas have low Sc (<25 ppm), moderate Cr, Ni, and large ion lithophile element (LILE) abundances (e.g. Ba = 280-450 ppm and Sr = 540-720 ppm). Chondrite-normalised (CN) REE abundances are light REE (LREE) enriched (La/Yb_{CN} = 5-9; calc-alkaline), with flat heavy REE (HREE) patterns around 15-20 times chondritic abundances (Fig. 6a). The patterns are split into two groups, one with lower LREE and higher HREE concentrations and vice versa, the patterns crossing over at around Pr-Nd. There are some very slight negative Eu anomalies relative to the MREE. The extended Primitive Mantle-normalised (PMN) plot (Fig. 6b) shows that the samples have spiky LILE patterns (modest positive Ba, Rb, Th, Sr anomalies), prominent negative K, Nb-Ta, and Ti anomalies and positive Zr-Hf anomalies relative to the REE, with super-chondritic Zr/Hf ratios of 47-55.

4.2.2. Ridge Series

The more evolved Ridge Series (~57-68 wt.% SiO_2, 2-5 wt.% MgO, Mg# = 41-55) has a wide range of sub-alkaline compositions from basaltic trachy-andesite through to dacite (Fig. 3a). Samples largely belong to the medium-K series, although two plot in the high-K field (Fig. 3b). They have noticeably lower TiO_2, $Fe_2O_{3(t)}$ and P_2O_5 concentrations compared to the valley series, and are slightly less sodic (Na_2O/K_2O = 1.4-2.5) (Fig. 4). Transition metal abundances are lower than the Valley Series, but the Ridge Series has higher Ba and Zr and lower Sr and Nb concentrations compared to the mafic lavas (Fig. 5). Chondrite-normalised LREE patterns (Fig. 6c) are similar to the Valley Series, again splitting into two groups with higher or lower LREE concentrations. Ridge samples have La/Yb_{CN} from 8 to 18, with a highly fractionated M-HREE distribution such that some samples have a U-shaped pattern. There are small negative Eu anomalies. Primitive Mantle-normalised patterns (Fig. 6d) differ slightly from the Valley Series in having positive Rb spikes, and more pronounced negative Nb-Ta, P and Ti anomalies.

4.2.3. Cone Series

The Cone Series splits into three groups, the most mafic being the two cones at Sepasar (58 wt.% SiO_2, but low Mg# = 43), and the most felsic at Eznasar (62 wt.% SiO_2, Mg# = 52). The large cone at Kaputkogh has intermediate silica content relative to the other cones, although it has the highest Mg# of 54. The cones have higher Nb and Zr concentrations relative to the other series (Fig. 5); REE patterns for the Cone Series are clearly bimodal (Fig. 6e), whilst the Primitive Mantle-normalised plots look similar to the Valley Series (Fig. 6f).

4.3. Radiogenic isotope geochemistry

Most samples span a narrow range from $^{87}Sr/^{86}Sr$ = 0.70416 to 0.70446 and $^{143}Nd/^{144}Nd$ = 0.51280 to 0.51287, giving a range of values in epsilon notation from εNd = +3.1 to +4.6. No age corrections were applied owing to the young age of the rocks. On Figure 7a, there is overlap between samples from the Valley, Ridge, and Cone series, and the samples display only a little isotopic enrichment with major element evolution (Fig. 7b). Overall, the samples lie on the mantle array between bulk silicate earth and depleted MORB mantle, and there is no clear evidence of any trends which might be related to mixing of different mantle end members (e.g. EMI or EMII) or old, isotopically enriched crustal contaminants. The Cone and Ridge Series samples containing quartz xenocrysts are not isotopically different from the other samples.

Regionally, the samples are significantly more depleted than most Pliocene-Quaternary centres in north-west Iran and Mount Damavand (Alborz), and Tendürek volcano in Eastern Anatolia (Fig. 7a). The Iranian volcanic rocks were erupted through thicker lithosphere than beneath Armenia (e.g. Liotard et al., 2008; Kheirkhah et al., 2009; Mirnejad et al., 2010; Davidson et al., unpublished data; Allen et al., 2013). Samples are also more depleted than asthenospheric melts from eastern Iran which have trends towards the EM-II mantle end member (Saadat et al., 2011; Saadat and Stern, 2012). Instead, results are closest to the few analyses conducted on the large stratovolcano, Mount Ararat, close to the Armenian border (Pearce et al., 1990; Kheirkhah et al., 2009), and nearly identical in terms of $^{143}Nd/^{144}Nd$ to six analyses of the ~3.25-2.05 Ma valley series lavas from southern Georgia, and from eight Late Miocene sub-alkaline basalts in central Georgia (Lebedev et al., 2006; 2007). Radiogenic isotope results are not yet widely published from other Armenian Pliocene-Quaternary centres, but Savov et al. (2007) and Lin et al. (2011) reported values from $^{87}Sr/^{86}Sr = 0.7041$ to 0.7051 and $^{143}Nd/^{144}Nd = 0.5128$ to 0.5129 from various locations.

5. Discussion

5.1. Volcano-tectonic interaction

Before considering the petrogenesis of the magma series, we first address the location of volcanic activity with respect to major crustal structures. The internal part of the orogenic plateau is not undergoing contractile deformation (Jackson et al., 1995; Vernant et al., 2004), but internal reorganisation of the plateau during the on-going convergence between Arabia and Eurasia means that Eastern Anatolia, the Lesser Caucasus, and northwest Iran are criss-crossed by numerous active strike-slip fault systems (Rebaï et al., 1993; Koçyiğit et al., 2001). These systems, which often tally with pre-existing crustal discontinuities, have been widely implicated in providing a locus for Quaternary magmatic activity through the production of highly localised pull-apart zones (Dewey et al., 1986; Karakhanian et al., 1997, 2002; Avagyan et al., 2010; Shabanian et al., 2012).

In Armenia, the active Pambak-Sevan-Syunik right-lateral strike-slip fault system is one such structure exploiting the existing discontinuity of the Sevan-Akera suture zone (Fig. 2). All of the magmatism described in this study originated just to the north of the fault zone, and so we propose that this location sat over a region of localised upper crustal extension at the time of magmatism. No active extension is recorded from the Shirak region from earthquake focal mechanisms or geomorphic features. Blanket coverage of most pre-existing fault structures by the Valley Series, and the absence of linear arrays of cinder cones (e.g. Dewey et al., 1986), may have helped obscure this association between faulting and magmatism. The south-eastern termination of the Pambak-Sevan-Syunik fault in the Syunik region is characterised by widespread volcanism in the area south of the fault, with little volcanism to the north (Kharakhanian et al., 2004). The Syunik centres therefore appear to have developed along the complementary trailing imbricate fan (Woodcock and Fischer, 1986) to the Shirak volcanic rocks. The north-south trending volcanism along the Javakheti Ridge is consistent with this idea.

Not all young Armenian centres fit a simple fault-control hypothesis: Aragats volcano, the largest centre in the country, is situated in a region apparently not crossed by presently-active faults (Kharakhanian et al., 2004). It is possible that older faults may be obscured by lava flows, or that some other control on the location of Aragats may apply, such as a local thin-spot in the lithosphere which focussed melting beneath the plateau.

5.2. Fractionation and contamination processes

5.2.1. Fractional crystallisation – minerals involved

No sample from the mafic Valley Series lavas is close to a primary melt (MgO is typically < 7 wt.%), so it is assumed that they have already evolved at depth involving a typical fractionation assemblage of olivine and spinel. In the major element data, clear falling trends for the Valley Series against typical indices of fractionation, such as SiO_2, TiO_2, MgO, and CaO (Fig. 4), corroborate with the observed mineral assemblages in confirming that clino- and orthopyroxene and Fe-Ti oxides were important in Valley Series evolution. Al_2O_3 concentrations cover a narrow range with no trends, indicating that feldspar fractionation was not an important feature of the Valley Series; likewise with P_2O_5 concentrations and apatite. Trace element abundances, such as Ba, Th, and La, display rising trends against SiO_2 in the Valley Series, which confirms their incompatible behaviour (Fig. 5). In the more evolved Ridge and Cone Series, clear falling trends emerge against SiO_2 for Al_2O_3, P_2O_5, Sr, Nb, Zr and La, pointing to addition of plagioclase, along with small proportions of zircon and apatite, to the fractionating assemblage.

The overall pattern of Shirak magmatism can be compared with Eastern Anatolia. Volcanic centres that display either high- or low-Y trends relative to Rb, are associated with fractionation of anhydrous (plagioclase, olivine, pyroxenes, and magnetite), or hydrous assemblages (including amphibole, which is compatible with Y) respectively (Pearce et al., 1990) (Fig. 8). The mafic Valley Series rocks in Shirak follow the same moderate to low-Y trend as the Kars plateau/Mt. Ararat systems, which may indicate amphibole fractionation has taken place at depth, given that amphibole is not seen in any of the Valley Series thin sections. Another indication of amphibole fractionation is the compatible middle to heavy REE (Sm-Lu) showing falling trends as the three series evolve (Fig. 6). The ridge series lavas have a steeply falling trend for Y against Rb, which may reflect increased partition coefficients for both clinopyroxene and amphibole for Y as these minerals fractionate from more evolved rocks (Pearce et al., 1990).

5.2.2. Magma mixing

The petrographic evidence for magma mixing (zoned plagioclases, sieve textures, reaction rims and quartz blebs) also needs to be reconciled with geochemical data. Major element plots, especially TiO_2, CaO and MgO vs. SiO_2 (Fig. 4) have straight line trends which are widely associated with mixing of two compositionally distinct magmas (Langmuir et al., 1978), rather than the curved trends associated with fractional crystallisation. In the field, the key observation is that the most evolved rocks found in the area, the obsidians at Agvorik, underlie the mafic Valley Series, so felsic magmas had already been erupted by the time of mafic magma injection.

5.2.3. Crustal contamination

Many models (e.g. Keskin et al., 1998) consider assimilation-fractional crystallisation (AFC) processes to be important in magma genesis in the plateau. Keskin et al. (1998) argue that recent mafic to felsic samples from Eastern Anatolia have undergone significant amounts of AFC coinciding with enriched $^{87}Sr/^{86}Sr$ ratios up to 0.7065 (Pearce et al., 1990). In Shirak, samples have only a slight variation in isotope ratios relative to SiO_2 (Fig. 7b), and samples containing quartz xenocrysts do not have different isotopic ratios to the other lavas - a feature that should strongly support the hypothesis that fractionation and magma mixing were the dominant processes. Also, preliminary results from elsewhere in Armenia, including the Aragats volcanic system, show little clear isotopic evidence for contamination (Savov et al., 2007; Lin et al., 2011) in spite of Aragats erupting through the SAB basement which has enriched $^{87}Sr/^{86}Sr$ ratios of up to 0.7303 - any contamination should be easily indentified (Bagdasaryan and Gukasyan, 1985). However, along-strike from Shirak at Artvin in eastern Turkey, Eocene rocks have measured $^{143}Nd/^{144}Nd$ of 0.512663 - 0.512854 and $^{87}Sr/^{86}Sr$ of 0.705148 - 0.704233 (Aydinçakir and Şen, in press). These values are very similar to the Shirak lavas (Fig. 7), so partial melting or assimilation of similar Eocene crust may be very difficult to decipher geochemically.

On a Th/Yb vs. Ta/Yb plot (Pearce, 1983) (Fig. 9), the three series form a consistent linear trend sub-parallel to the mantle array, with the felsic rocks having compositions more enriched than those of typical continental crust (Rudnick and Gao, 2003). On this diagram, the fractional crystallisation trend for a typical amphibole-bearing assemblage is shown. The samples plot to the left of the fractionation trend which suggests that AFC processes may be operating - although choice of mineral assemblages and partition coefficients can easily affect the FC trend. AFC modelling (Powell, 1984) using the average composition of the Artvin rocks (Aydinçakir and Şen, in press) quite reasonably reproduces the trend of the Shirak lavas, but the ratio of assimilation to fractionation is high at 0.8. Modelling thus shows that large volumes of isotopically similar material can be incorporated into the Shirak lavas without a significant effect on trace element evolution. Studies of disequilibrium textures and mineral chemistry might better elucidate the processes involved in magma evolution.

5.3. Mantle source and partial melting

5.3.1. Mantle source of the Valley Series

The presence of LILE and HFSE anomalies on normalised plots of mafic rocks (Fig. 6b) are normally taken to indicate a subduction-modified mantle source owing to the retention of HFSE in the slab, and the comparative mobility of the LILE/REE during slab heating and dewatering into the overlying mantle wedge (e.g. Pearce and Peate, 1995). We have already introduced the Th/Yb vs. Ta/Yb diagram (Fig. 9). The alkali basalts from Shirak plot above the mantle array which is commonly taken to indicate the presence of a subduction-modified source. However, they also have very much higher Ta/Yb ratios than many subduction-related rocks. The Shirak lavas are therefore derived from an incompatible element-enriched mantle source, or are derived from a limited degree of melt extraction. As these lavas erupted >20 Myr after the end of Neo-Tethyan subduction, and there is no evidence for a slab at shallow depths beneath Eastern Anatolia and Armenia at the present day (Zor et al., 2008), it is improbable that a normal supra-subduction zone hydrated asthenospheric mantle wedge was involved in the origin of the Shirak lavas. Therefore, the subduction-like characteristics are likely to be derived from a fertile source within the mantle that had been inherited its slab-related geochemical component from earlier Neo-Tethyan subduction. Crustal contamination is unlikely be responsible for the subduction-like characteristics of the Shirak lavas, as LILE and HFSE anomalies are significant in even the most mafic samples.

Some of the trace element characteristics of the valley series may help constrain the mineralogy of the mantle source. Overall, the flat normalised HREE patterns (Fig. 6a) indicate a spinel-facies mantle source at <70 km, unless the degree of partial melting was very high (>25%) in order to completely consume any garnet present at depths of >70 km. This is unlikely given the overall LREE-enriched and Nb-Ta/HREE-enriched trace element patterns (Fig. 6b) which point towards modest degrees of partial melting of an enriched source. Low Sc concentrations (<25 ppm) in all Valley Series samples may indicate residual clinopyroxene, another indicator of a low degree of partial melting, but it is also possible that extensive pyroxene fractionation prior to eruption has resulted in these low values. Intra-LILE variations, including low Ba/Rb (<25) and Rb/Sr ratios (<0.05), do not point towards amphiboles or phlogopite playing an important role during melting (e.g. Furman and Graham, 1999). Melting therefore took place beneath the Armenian crust at depths of ~45-70 km.

One unusual feature of the Shirak lavas is high ocean island basalt (OIB)-like Zr concentrations (~200 ppm) and Zr/Hf ratios (41-52) relative to MOR and arc basalts, the latter having chondritic Zr/Hf ratios of 35-39 (Weaver et al., 1987; David et al., 2000; Pfänder et al., 2007). Nb/Ta ratios range from 16-22 relative to the chondritic ratio of 19.9 (Pfänder et al., 2007). Mafic samples from other nearby centres in the plateau, including Tendürek and Ararat, show similar features (Fig. 10). Lower ratios in the Ridge Series compared to the Valley Series might be explained by contamination with crustal material such as the Eocene basement (Fig. 10). However, the high Zr/Hf ratios of 47-52 in the less evolved Valley Series are a primary feature of the magmas.

There is little correlation between Zr and Zr/Hf ratios (not shown), indicating that zircon accumulation cannot be directly responsible for the high Zr-Zr/Hf character of the Shirak lavas. HFSE fractionation in OIBs may be due to: (1) residual or fractionating clinopyroxene (David et al., 2000); (2) fractionation of Ti-bearing phases such as rutile, ilmenite, and amphibole (Foley et al., 2000; Tiepolo et al., 2001); (3) melting of recycled eclogite or garnet pyroxenite (Pfänder et al., 2007); or (4) the occurrence of carbonate metasomatism (Dupuy et al., 1992). For option (1), fractionation of clinopyroxene only has a modest effect upon Zr/Hf ratios (Pfänder et al., 2000). Our modelling of pure clinopyroxene fractional crystallisation from a starting composition with Zr/Hf and Nb/Ta of primitive mantle shows that unrealistic amounts of fractionation are required to generate the Shirak samples (Fig. 10). In option (2), fractionation of titanate phases such as rutile and ilmenite can generate very high Zr/Hf and Nb/Ta ratios, with $D_{Zr/Hf}$ and $D_{Nb/Ta}$ both <1 (Pfänder et al., 2000); however, titanate fractionation would also strongly reduce overall Nb concentrations, a feature not seen in the Valley Series. Partial melting of garnet-bearing lithologies (option 3) is invoked in many OIB examples (see Pfänder et al., 2007) but can be ruled out here on the basis of flat normalised HREE patterns in the Shirak lavas – these are not OIB-like magmas (Fig. 6b). Where carbonates are invoked in the mantle source (e.g. Dupuy et al., 1992; Hoernle et al., 2002) (option 4), resultant alkaline melts or mantle xenoliths have very high Sr and Ba of >>1000 ppm, and in spite of high Zr/Hf ratios many carbonatites have very low concentrations of these elements (Ionov et al., 1993). This is not the signature of the Shirak samples hence carbonate metasomatism is unlikely in this case. Several studies have shown that amphibole and phlogopite fractionate the HFSE (Moine et al., 2001; Tiepolo et al., 2001; Chakhmouradian, 2006), with Chakhmouradian (2006) demonstrating that low-Ti amphiboles have high Zr/Hf ratios of ~60-200. Hence these minerals can impart high Zr/Hf on a melt; but it is still unclear what the high overall Zr concentrations in the Shirak lavas are caused by - this feature is normally attributed to ancient recycled oceanic crust in OIBs (Weaver, 1991).

5.3.2. Modelling of partial melting

Any model of partial melting conditions for Shirak has to be based on the HREE and HFSE, making the assumption that neither set of elements were transported into the lithospheric mantle source in a slab-derived fluid (Pearce and Peate, 1995). Therefore, we have constructed non-modal batch melting curves using Dy, Yb and Nb (ignoring Zr owing to its anomalous behaviour), in order to constrain the degree of partial melting needed to form the Valley Series. We have taken the approach of Pearce et al. (1990) in assuming that hydrous phases such as amphibole and phlogopite (if present) are completely consumed during melting and do not contribute to the melt model. Given the high Ta/Yb ratios of even the least evolved Valley Series samples (Fig. 9), it is reasonable to compare the melting of depleted MORB mantle (DMM) (Workman and Hart, 2005) with a more incompatible-element enriched source, in this case primitive mantle with 1% bulk continental crust extracted, as used by Fitton and Godard (2004) for the Ontong Java oceanic plateau.

Although we can easily model HREE and HFSE ratios (see below), fractional crystallisation of olivine, spinel, plagioclase and pyroxene versus fractionation of amphibole from primary magma have competing effects on absolute REE and HFSE concentrations. Often, elemental values for basalts in modelling are back-calculated to 9 wt.% MgO to negate the effects of plagioclase and pyroxene crystallisation (Pearce & Parkinson, 1993). However, valley series Dy and Yb concentrations are near-constant in spite of varying MgO and SiO_2 (Fig. 6a), probably due to the competing effects of amphibole and clinopyroxene fractionation, so no realistic back-calculation can be applied. Therefore, we simply attempt to model the elemental ratios of the last-evolved Valley Series lava (7 wt.% MgO) and assume that this best reflects the conditions of partial melting.

Modelling results (Fig. 11) indicate that melting of a garnet peridotite cannot reproduce the compositions of the Valley Series lavas, a finding consistent with the flat normalised HREE patterns (Fig. 6a). Spinel peridotite partial melting curves do intersect the valley series at low degrees of melting, with the DMM melting curve on Figure 11 giving 0.1-0.5% melting. In contrast the more fertile source gives 2-5% melting, which is perhaps more realistic than the tiny proportion of melting required from a DMM source and the difficulties of extracting such a small volume of melt (e.g. Hirth and Kohlstedt, 1995). This spinel peridotite melting outcome is also consistent with geophysical surveys indicating a seismically slow mantle at depths of ~50 km beneath Armenia (e.g. Koulakov et al., 2012).

5.4. Reconciliation with geophysical and geodynamic models

5.4.1. Extent of lithospheric delamination

Debate exists over the extent of lithospheric delamination beneath Eastern Anatolia following the cessation of subduction beneath the Bitlis-Zagros suture (e.g. Keskin, 2003). Seismic surveys indicate a crustal thickness of ~40-45 km, but with significant negative P- and S-wave seismic velocity anomalies beneath, extending from ~50 to ~250 km depth, between eastern Turkey, Armenia, the Black Sea and northwest Iran, concurrent with many Pliocene-Quaternary volcanic centres (e.g. Piromallo and Morelli, 2003; Maggi & Priestley, 2005; Zor et al., 2008; Koulakov et al., 2012). The anomaly has been used by these authors to argue for the presence of hot, perhaps partially molten, asthenosphere, but several authors extend these conclusions to the possibility that there is also no mantle lithosphere 'lid' beneath the Anatolian crust (e.g. Keskin, 2003; Zor et al., 2008) and the Caucasus (Koulakov et al., 2012). In this case, mafic magmatism in Shirak would have to have an asthenospheric source.

There are significant implications for magmatism in this scenario. The impact of hot upwelling asthenosphere on the thickened Lesser Caucasus arc crust should result in extensive lower crustal melting, as observed in the Puna Plateau of the Andes, and the Great Basin Altiplano in the western U.S. (Allmendinger et al., 1997; Babeyko et al., 2002; Best et al., 2009). Going back to our geochemical results, this model of whole-scale lithospheric delamination proposed for the Puna Plateau is incompatible with the observed silica-undersaturated magmatism in Shirak, which bears little evidence for large-scale crustal interaction. We conclude that there is sufficient lithospheric mantle beneath the Armenian crust to act as a thermal barrier between the asthenosphere and crust (Fig. 12), protecting the crust from melting and infiltration by hot asthenospheric melts in the manner described by Babeyko et al. (2002). An asthenospheric source for the Shirak magmas cannot have been influenced by a subducting slab, because subduction processes ended prior to the Miocene. Hence the Shirak magmas would not have subduction-like trace element characteristics, and instead should closely resemble OIB. There are asthenosphere-derived OIB-like lavas without subduction-related geochemical signatures in Eastern Iran (Saadat et al., 2010; Saadat and Stern, 2012) and in the Arabian foreland (Lustrino et al., 2010). The Iranian alkali olivine basalts show trends towards an EMII-like isotope signature (particularly with respect to Pb isotope ratios) (Zindler and Hart, 1986). They also and contain pyroxenite xenoliths from the lithospheric mantle, plagioclase megacrysts of uncertain origin, and some lower crustal gabbroic xenoliths (Saadat and Stern, 2012). These lavas are distinct in terms of xenolith content, trace element signatures and isotope geochemistry from those erupted in Shirak.

5.4.2. Geodynamic model

Our geodynamic model is presented schematically in Figure 12. We propose that, upon the termination of Neo-Tethyan subduction along the Bitlis-Zagros suture during the Oligocene, Armenia lay in a continental back-arc position relative to the former subducting slab and mantle wedge. Modelling studies have suggested that an old slab may be able to persist or 'stall' in the upper mantle without breaking off for up to 20 Myr after terminal collision (van Hunen and Allen, 2011). Delayed break-off of the Neo-Tethyan slab from Arabia beneath Eurasia may thus be responsible for the upsurge in magmatism since 10 Ma, and particularly in the Pliocene-Quaternary, across the orogenic plateau (e.g. Keskin, 2003), concurrent with the influx of hot asthenosphere into the region the slab once occupied. In Eastern Anatolia, the region immediately above the detached slab might lack mantle lithosphere, and asthenospheric and crustal melting would combine to produce arc-like magmas (Fig. 12) (Keskin, 2003). However, as we have already discussed, it is improbable that whole-scale lithospheric mantle delamination occurred beneath Armenia because we do not see attendant whole-scale lower crustal melting. The former asthenospheric mantle wedge of the Neo-Tethyan arc system would be refrigerated by the presence of a stalled slab, and rapidly converted into lithospheric mantle over the 15-25 Myr following terminal collision (c.f. Holt et al., 2010). This depleted lithospheric mantle could be stable and buoyant enough to be at least partially preserved following the eventual detachment of the underlying oceanic slab, whilst the aforementioned influx of convecting asthenosphere would trigger partial melting in the overlying lithospheric mantle, as well as providing a thermal support for the orogenic plateau (Fig. 12).

Another potentially important consideration is that, although much of the LILE and LREE budget of the southern Neo-Tethyan slab may have been delivered to the lithosphere before continental collision, a stalled slab and associated sediments would continue dewatering before break-off. This would contribute to the subduction-like signature on the mantle frozen-in beneath Armenia. Other Pliocene-Quaternary centres in the collision zone, such as the foreland volcanic system at Karacadağ, may result from asthenospheric melting beneath thin spots in the lithosphere (Lustrino et al., 2010; Ekici et al., 2012). Mantle-derived volcanism is also apparent even in the 50+ km thick crust of the Elbrus region of the Greater Caucasus (Lebedev et al., 2006b; Koulakov et al., 2012), and it is here that magmatism may be related to melting during collisional thickening of lithospheric mantle and the breakdown of hydrous mineral phases such as micas and amphiboles (e.g. Pearce et al., 1990; Allen et al., 2013) or to asthenospheric upwelling during lithospheric dripping (see Sosson et al., 2010) (Fig. 12).

6. Conclusions

- Mafic and more evolved Pliocene-Quaternary lavas in Shirak, NW Armenia, were emplaced through a former continental margin arc sequence as a result of localised extensional tectonics within the present-day Arabia-Eurasia collision zone.
- Magmas evolved from mafic through to dacitic compositions by fractional crystallisation dominated by pyroxene, amphibole and plagioclase; and although evolved samples contain quartz xenocrysts, none preserves clear isotopic evidence for

large-scale crustal assimilation - magma mixing appears to be the dominant petrogenetic process. We conclude that if assimilation did occur, it was of local arc-related crust of a similar isotopic composition to the Shirak melts.

- The least-evolved magmas preserve trace element evidence for derivation by moderate degrees of melting (~3-4%) from a shallow, spinel-facies lithospheric mantle source with an inherited subduction component probably related to earlier Tethyan subduction processes. They contain high Zr concentrations and high Zr/Hf ratios which are an intrinsic feature of the source or partial melting process.
- The presence of lithospheric mantle beneath Armenia is a requirement for geodynamic models of the region in order to prevent the occurrence of whole-scale lower crustal melting.

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Table captions

Table 1. Major and trace element data for selected samples from the valley, ridge, and cone series, Shirak. LOI = loss-on-ignition.

Table 2. Measured Nd and Sr isotope compositions of the valley, ridge, and cone series, Shirak.

Supplementary Items

Item 1. Selected photomicrographs of samples from the valley, ridge, and cone series, Shirak.

Item 2. Complete whole rock major and trace element data for the Shirak lavas, including trace element standards.

Figures

Figure 1. Map of the Turkish- Iranian plateau with shaded digital topography, showing locations of Pliocene-Quaternary volcanic centres (cones) and the study area (rectangle).

Figure 2. Digital topography map of the study region with geological features simplified from work by Khachatur Meliksetian, Gevorg Navasardyan and Sergey Karapetyan of the Institute of Geology of the National Academy of Sciences of Armenia, plus outline map of Armenia showing administrative boundaries and regional coverage of Late Miocene-Quaternary magmatic products.

Figure 3. (a) Total alkali-silica classification (Le Bas et al., 1986) and (b) K_2O vs. SiO_2 classification (Peccerillo and Taylor, 1976).

Figure 4. Major element variation diagrams for the Shirak lavas.

Figure 5. Minor and trace element variation in the Shirak lavas.

Figure 6. Rare earth element and extended trace element normalised plots. Chondrite normalisation values from McDonough and Sun (1995) and Primitive Mantle and OIB values from Sun and McDonough (1989).

Figure 7. (a) Nd-Sr isotope plot for Shirak lavas, compared to mafic centres within the collision zone. Pliocene-Pleistocene valley series in southern Georgia and Late Miocene mafic lavas from the Elbrus region of Southern Russia - Lebedev et al. (2007; 2010); NW

Iran minor centres, Tendurek, Ararat, Kurkistan - Kheirkhah et al. (2009), Allen et al. (2013); Damavand - Davidson et al. (unpublished data), Mirnejad et al. (2010); Artvin, Eastern Turkey - Aydiçakir and Şen, in press). Mantle end members and array - Zindler & Hart (1986). (b) Variation of Nd and Sr isotopes as a function of magmatic evolution.

Figure 8. Fractional crystallisation (FC) trends within the Shirak lavas (symbols as per previous diagrams). Data and FC vectors for basic to acidic rocks for Eastern Anatolia are from Pearce et al. (1990). pl = plagioclase; o = olivine; opx = orthopyroxene; cpx = clinopyroxene; hb = hornblende; gnt = garnet.

Figure 9. Th/Yb vs. Ta/Yb diagram (Pearce, 1983) for the Shirak lavas, showing an FC vector for a hydrous assemblage, taking into account increasing partition coefficients during magmatic evolution (after Keskin et al., 1998), and an AFC vector as described on the figure. Eocene rocks from Eastern Turkey, likely to be similar to those directly underlying the Shirak lava series, are plotted (Aydinçakir and Şen, in press). Crust (UCC: upper continental crust; MCC: middle crust; LCC: lower crust; BCC: bulk continental crust, all from Rudnick and Gao, 2003). Active margins - Pearce (1983). See text for discussion.

Figure 10. Zr/Hf vs. Nb/Ta plot for Shirak lavas and selected mafic Pliocene-Quaternary lavas from the Arabia-Eurasia collision. Ararat and Tendürek are from lithospheric mantle sources, Karacadağ in southern Turkey has an OIB source. Sources: Tendürek, Ararat – Kheirkhah et al. (2009); Karacadağ - Sen et al. (2004), Lustrino et al. (2010); clinopyroxene fractionation based on partition coefficients of Pagé et al. (2009); other references as per Figure 9.

Figure 11. Non-modal batch partial melting models for the valley series lavas using depleted MORB mantle (Workman and Hart, 2005) and incompatible element enriched oceanic plateau (Fitton and Godard, 2004) sources. Source modes: spinel lherzolite - ol = 0.578, opx = 0.27, cpx = 0.119, sp = 0.033; garnet lherzolite - 0.598, 0.211, 0.076, gnt = 0.115. Melt modes: spinel lherzolite - 0.1, 0.27, 0.5, sp = 0.13; garnet lherzolite - 0.05, 0.2, 0.3, gnt = 0.45. Partition coefficients are from the GERM Partition Coefficient Database (http://earthref.org/KDD).

Figure 12. A schematic cross-section through the present-day Arabia-Eurasia collision zone highlighting potential processes involved in Pliocene-Quaternary magmatism. Hatchings represent regions of partial melting. Crustal thicknesses estimated from Zor et al. (2008).

Pliocene-Quaternary volcanic rocks of NW Armenia: magmatism and lithospheric dynamics within an active orogenic plateau

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Abstract

The Pliocene-Quaternary volcanic rocks of Armenia are a key component of the Arabia-Eurasia collision, representing intense magmatism within the Turkish-Iranian plateau, tens of millions of years after the onset of continental collision. Here we present whole rock elemental and Nd-Sr isotope data from mafic, intermediate, and felsic lava flows and cinder cones in Shirak and Lori Provinces, NW Armenia. Magmatism appears to be controlled locally by extension related to major strike-slip faults within the plateau. Major and trace element results show that the three series – valley-filling medium-K alkali basalt flows, ridge-forming andesite to rhyolite flows, and andesitic cinder cones – form a compositional continuum linked by a crystallisation sequence dominated by two pyroxenes, plagioclase and amphibole. There is ~~some~~ petrographic and major and trace element evidence for magma *mixing processes and potentially crustal contamination by Mesozoic-early Cenozoic arc-related rocks, which has not significantly affected the isotopic signature. Modelling of the basaltic rocks indicates that they formed by moderate degrees of partial melting (~3-4 %) of an incompatible element enriched, subduction-modified, lithospheric mantle source. Samples have a distinctive high Zr/Hf ratio and high Zr concentrations, which are an intrinsic part of the source or the melting process, and are much more commonly found in ocean island basalts. Regional models for magmatism ~~commonly~~ often argue for whole-scale delamination of the mantle lithosphere beneath Eastern Anatolia and the Lesser Caucasus, but this scenario is hard to reconcile with limited crustal signatures and the apparent lack of asthenospheric components within many studied centres.*

Highlights

- Whole-rock study of Pliocene-Quaternary lavas from the Armenian Highlands
- ~~Range of alkali basalts through to rhyolites~~Compositional range controlled by fractional crystallisation, magma mixing and possible ~~limited~~ crustal contamination
- Low-degree melting of a shallow metasomatised lithospheric mantle source
- Exploring triggers for <10 Myr increase in magmatic activity in ~~Armenia and elsewhere in~~ the Arabia-Eurasia collision zone

Keywords

Armenia; geochemistry; petrogenesis; orogenic plateau

1. Introduction

Orogenic plateaus such as the modern Turkish-Iranian, Bolivian Altiplano-Puna and Tibetan plateaus form in response to plate convergence and collision, and represent a primary topographic feature of the continents. In spite of their thickened crust and compression tectonics, plateaus are also sites of intense, ultimately mantle-derived magmatism (e.g. Williams et al., 2004; Mo et al., 2007). Such magmatism is often attributed to asthenospheric upwelling following the break-off of the subducting oceanic slab (e.g. Keskin, 2003), or the delamination of the lithosphere inboard of the plate suture (e.g. Kay and Kay, 1993).

The Turkish-Iranian plateau (Fig. 1) is a product of the Cenozoic Arabia-Eurasia collision, and magmatism post-dating initial collision is particularly voluminous from the Late Miocene until the present day, in numerous locations across eastern Turkey, Armenia, and much of Iran (Fig. 1). Erupted products~~,~~ range~~ing in composition~~ from mafic to felsic, and sodic to ultrapotassic (Pearce et al., 1990; Karapetian et al., 2001; Davidson et al., 2004; Azizi and Moinevaziri, 2009; Saadat et al., 2011; Saadat and Stern, 2012; Allen et al., 2013). In nearby Georgia and the Greater Caucasus, the most recent magmatism appears to have started slightly earlier, in the Middle Miocene, and continued with some gaps in the record until recent times (Lebedev et al., 2006a,b; 2007; 2008a,b; Adamia et al., 2011).

This plateau-wide ‘recent’ magmatism may be only partly explained by partial melting of asthenospheric ~~and~~ or mantle lithosphere sources ~~resultant from~~due to ~~slab~~ break-off of the southern Neo-Tethys slab (e.g. Keskin, 2003; 2007; Şengör et al., 2008; Dilek et al., 2010; van Hunen and Allen, 2011). ~~Both the Eocene flare-up and~~ Miocene to recent magmatism extends to at least 500 km from ~~their respective~~the Bitlis-Zagros suture zones, ~~on~~ a spatial scale akin to the Cenozoic ‘ignimbrite flare-up’ of the western United States

(Johnson, 1991)~~, with a remarkable co-incidence between the locations of the Eocene and recent magmatism (Verdel et al., 2011)~~. The wide extent of Miocene-Quaternary magmatism hundreds of kilometres from the suture zone indicates that ~~slab break-off is not the sole mantle melting trigger. W~~whole-scale lithospheric delamination (Pearce et al., 1990), ~~on-going under-thrusting of Arabian crust (Allen et al., in press),~~ or other unrecognised processes may be collectively responsible for magmatism in this region.

To further consider the origin of such magmatism, ~~we turn our attention~~this paper focuses on ~~to~~ Armenia (Fig. 1), where ~~the geochemistry of~~ recent volcanism has been under-represented in the international literature ~~(Karapetian et al., 2001; Savov et al., 2007; Lin et al., 2011)~~. We present whole rock elemental and Nd-Sr isotope data from three series of mafic to felsic Pliocene-Quaternary volcanic rocks in the north of Shirak and west of Lori administrative provinces in the northwest of the country (herein referred to ~~simply~~ as 'Shirak') (Fig. 2). The local tectonic setting and relationship to magmatism is highlighted, alongside discussion on magmatic evolution~~fractional crystallisation, crustal contamination~~, mantle sources~~,~~ and partial melting. We finish by assessing how the results fit with regional geodynamic models.

2. Geological setting, structure, and petrography

2.1. The Turkish-Iranian plateau

The Turkish-Armenian-Iranian orogenic plateau (Fig. 1)~~, itself a product of the Arabia-Eurasia continental collision, and~~ developed following the closure of the Neo-Tethys Ocean (Şengör and Kidd, 1979). The basement of the plateau consists of Mesozoic to Early

Cenozoic accretionary belts and arc rocks and also older, Gondwanaland-related micro-continental fragments that all accreted to the southern margin of Eurasia. It is widely assumed that ~~in the region of study,~~ the Neo-Tethys oceanic crust~~Ocean~~ was divided into a northern and a southern segment, the former closed ~~by collisions marked by the Sevan–Akera ophiolite suite (Fig. 2)~~ either during the Late Cretaceous (Lordkinpanidze, 1980; Keskin, 2008) or the Paleocene-Early Eocene (Sosson et al., 2010). The ~~northern and southern~~two segments of Neo-Tethys were separated by ~~rifted~~ micro-continental fragments such as the South Armenian Block and Tauride-Anatolide terranes (Sosson et al., 2010). Destruction of the southern segment of Neo-Tethys brought Arabia and Eurasia together along the Bitlis-Zagros suture zone (Fig. 1).

The timing of initial collision between Arabia and Eurasia is debated, although ~~many~~ most estimates range between 35 and 20 Ma (Agard et al., 2005; Allen and Armstrong, 2008; Morley et al., 2009; Okay et al., 2010; Ballato et al., 2011; McQuarrie and van Hinsbergen, 2013). ~~Furthermore, the timing of plateau growth from this point on is uncertain.~~ Marine carbonates ~~were~~ deposited across much of central Iran and eastern Turkey in the Early Miocene~~,~~ ~~indicating~~ indicate that ~~surface uplift~~growth of the orogenic plateau is a later phenomenon (Bottrill et al., 2012). Deformation is ~~at~~ presently focussed on the plateau margins~~ of the plateau~~, from the Greater Caucasus and Alborz in the north to the Zagros in the south, with no active crustal thickening or thinning occurring between (Jackson et al., 1995; Allen et al., 2011). Lithospheric thickness is highly variable, from >200 km in Iran near the Zagros suture, to only 60 km or less in eastern Anatolia (Priestley and McKenzie, 2006; Angus et al., 2006). The current height of the plateau, ~1750 m above sea level, has been attributed to a combination of Late Cenozoic crustal shortening, and also to the detachment of mantle lithosphere and/or subducted Tethyan slabs beneath the plateau, allowing the upwelling of hot, buoyant asthenosphere beneath the region (Keskin, 2003).

~~Plateau elevations in northern Armenia are approximately 2 km above sea level at present. Understanding the association between the growth of the plateau and partial melting processes, through interpreting the geochemical patterns of plateau magmatism, can provide key constraints on how the plateau has evolved through time, and give insights into orogenic plateaux in general.~~ The Cenozoic magmatic record ~~within the region of the present plateau~~of the plateau is divided into several stages (see Dilek et al., 2010; Chiu et al., 2013 for reviews~~a review~~). First is an Eocene ‘flare-up’ of arc magmatism immediately prior to the onset of continental collision, focussed predominantly on the Urumieh-Dokhtar arc in southwest Iran, the Lut Block, Kopeh Dag and Alborz regions of eastern and northern Iran (Verdel et al., 2011), and also in Armenia (Lordkinpanidze et al., 1988). The flare-up has been ~~variously~~ attributed to back-arc extension ~~behind the Iranian margin~~ (Vincent et al., 2005), an episode of flat subduction (Berberian and Berberian, 1981), perhaps coupled with enhanced slab roll-back (Verdel et al., 2011), or break-off of the northern Neo-Tethyan slab in Armenia and North-Central Turkey (Keskin et al., 2008; Sosson et al., 2010). The second stage is a magmatic ‘gap’ which comprised some 20-30 Myr of ~~very~~ limited magmatic activity between the Eocene and the Late Miocene as continental collision proceeded (Verdel et al., 2011; Richards et al., 2011). The third and final stage is the ~~resumption~~ aforementioned upsurge of ~~ultimately~~ mantle-derived volcanism from the ~~Late~~ Middle to Late Miocene until the present day (Chiu et al., 2013), ~~in numerous locations~~ which forms the basis of this study.

2.2~~1~~. Basement and structure in Armenia

Armenia, including the Lesser Caucasus mountain range, lies at the northern side of the ~~Turkish-Iranian orogenic~~ plateau (Figs. 1, 2). Southern Armenia is underlain by the aforementioned South Armenian Block (SAB), a micro-continental fragment rifted during the early Mesozoic, separating the ~~During the early Mesozoic, the~~ northern and southern branches of the Neo-Tethyan seaway ~~opened between Africa/Arabia and Eurasia, separated by micro-continental fragments of the former Gondawana supercontinent~~ (Stampfli, 2000). ~~Much of southern Armenia is one such fragment, known as the South Armenian Block (SAB), which may link to the Tauride and Anatolide terranes to the west (Sosson et al., 2010). Where exposed beneath Pliocene-Quaternary rocks, t~~The SAB consists of Proterozoic gneisses, mica schists~~,~~ and amphibolites partially overlain by Devonian to Jurassic sediments, Jurassic and younger ophiolitic material, and Paleocene to Early Oligocene volcanic rocks related to subduction of the southern branch of Neo-Tethys (Rolland et al., 2009). ~~The~~ In the north of Armenia, the Armenian Highlands represent the former active continental margin of Eurasia and contain~~s island~~ arc and discontinuous ophiolite sequences formed during the closure of the northern branch of the Neo-Tethyan seaway (Adamia et al., 1981; 2011). ~~There is significant debate over the mode of formation and location within the Neo-Tethyan realm of each ophiolite fragment (Sosson et al., 2010).~~ The largest tract of ophiolitic material in Armenia forms the Sevan-Akera suture zone, a 400 km-long boundary between the SAB and the Mesozoic arc of the Lesser Caucasus to the north. Immediately south of our field area, between Amasia and Stepanavan, is a belt of blueschist-facies mélange (part of the Sevan-Akera ophiolite suite), tectonically overlain by Jurassic to Cretaceous mafic ~~oceanic~~ rocks, ~~which are in turn overlain by~~and two sequences of Cretaceous to Early Oligocene subduction-related volcano-sedimentary rocks (Rolland et al., 2009). ~~^{40}Ar/^{39}Ar phengite ages from the blueschist suggest metamorphism took place during northwards Tethyan subduction in the Late Cretaceous (~95-91 Ma), followed by retrogression which started during collision~~

~~of the SAB with the Lesser Caucasus island arc at ~74-71 Ma, and continued until terminal collision with the Eurasian margin during the Eocene (Rolland et al., 2009).~~

Following the Eocene amalgamation of the Armenian crustal blocks (Rolland et al., 2009), north-directed subduction of the southern ~~branch of~~ Neo-Tethys terminated along the Bitlis-Zagros suture, ~~(~~some 300 km south of Armenia~~)~~. After the last ~~Eocene-Oligocene~~ subduction-related magmatism, the magmatic 'gap' in Armenia extended until the Late Miocene (~~~<~~10 Ma), based on groundmass and mineral K-Ar ~~^{40}K-^{40}Ar~~ ages from the oldest volcanic rocks of the Gegham Highlands (Arutyunyan et al., 2007). The most voluminous volcanism is ~~however~~ of Pliocene-Quaternary age, covering much of Aragatsotn, Shirak, Kotayk, Gegharkunik, and Syunik provinces, an area >10,000 km^2 (Mitchell and Westaway, 1999; Karapetian et al., 2001; Lebedev et al., 2011) (Fig. 2). ~~If interpretations of crustal structure are correct, the magmas from Shirak in this study may have passed through a complex series of interwoven crustal units including SAB basement, Jurassic arc material, Jurassic-Early Oligocene arc rocks, flysch, and limestone (Sosson et al., 2010).~~

2.3~~2~~. Pliocene - Quaternary magma series

~~Much of t~~The geology of NW Armenia was first studied in detail during Soviet times (Kharazyan, 1983). The oldest Pliocene – Quaternary volcanism is represented by poorly-exposed rhyolites and obsidians at Aghvorik (Fig. 2), covered by ~~. These felsic rocks are covered by Upper Pliocene~~ dolerites ~~(see below). Aside from these felsic rocks, much of the lowest topography in Shirak is covered by mafic rocks, traditionally named as doleritic basalts or Pliocene plateau basalts in Armenian geological literature~~which drape much of the lowest topography in Shirak, often part-filling river valleys for tens of kilometers. ~~These~~

~~flows form a stratigraphic horizon across much of northern and central Armenia, due to presence of extended lava flows cropping out on plateaus and in cross-sections in many of the river valleys.~~ ~~Some r~~Relationships with sedimentary deposits dated using mammalian fossils lead authors to conclude that these mafic lavas were of Late Pliocene age (Kharazyan, 1983 and references therein). A ~~^{40}K-^{40}Ar~~ K-Ar age determination from dolerite from the Akhurian river basin within our study area gave a result of 2.5 ± 0.2 Ma (Chernyshev et al., 2002). Groundmass K-Ar ~~^{40}K-^{40}Ar~~ results from ~~a wide range of~~numerous mafic to felsic volcanic rocks across the Javakheti Highlands in southern Georgia gave ages of 4.6 ± 0.2 to 1.54 ± 0.10 Ma (Late Pliocene to Quaternary) (Lebedev et al., 2008a,b). The mafic rocks in Armenia and southern Georgia ~~have been described as resulting~~may have resulted from fissure eruptions (Jrbashyan et al., 1996), but the actual source of these lavas has never been found, and may be buried by younger~~, more evolved~~ flows.

~~Also present~~On top of the plateau-like topography formed by the mafic flows in Shirak, are ~~mountainous~~ hills~~areas~~ of intermediate-felsic composition ~~extending~~ of up to 600 m prominence ~~above the plateau-like topography formed by the mafic flows~~, including the ~~prominent~~ north-south trending Javakheti, or Kechut, ridge (Fig. 2). ~~Prior to any radiometric age determinations,~~ Kharazyan (1983) distinguished two ~~age~~ units within the Armenian part of the Javakheti ridge. The first unit (Lower Kechut Suite) ~~is presented by~~is said to contain two-pyroxene basaltic andesites, andesites and hornblende andesites which cover the valley series and are thus assumed to be of Early Pleistocene age (Kharazyan, 2005). SHRIMP U-Pb zircon dating of andesitic vent-proximal ash and breccia layers deposited on the eastern flank of the Javakheti ridge in Lori province at the Karakhach archaeological site (Fig. 2) ~~has~~ gives ~~confirmed~~ maximum eruption ages of 1.94 ± 0.05 to 1.80 ± 0.03 Ma~~, giving a maximum eruption age~~ (Presnyakov et al., 2012). This study also noted ~~several older grains, two of Eocene age~~two Eocene zircons (40-50 Ma) consistent with Eocene arc rocks found to the east of the study area, plus five Proterozoic grains, consistent with an origin in the underthrust SAB~~ (e.g. Rolland et al., 2009)~~. It is uncertain whether or not these old grains were from xenoliths ripped up during explosive volcanism or zircons assimilated during magma ascent. The Javakheti Ridge extends into southern Georgia, where it is higher and has a sharp little denuded topographic profile compared to further south. Groundmass ~~^{40}K-^{39}Ar~~ K-Ar dating revealed younger ages of ~~<~~<1 Ma for the Samsari volcanic centre ~~which lies~~ to the north of the Javakheti Ridge (Chernyshev et al., 2006). Kharazyan (1983) also defined an Upper Kechut Suite ~~on the Javakheti Ridge,~~ supposedly containing lavas erupted from cinder cones on the western part of the ridge ~~ridge covering eroded lavas and pyroclastic material of the Lower Kechut Suite~~, and considered to be of Middle Pleistocene age (Kharazyan, 2005). We did not find clear evidence of this unit during our studies. The only other Pliocene-Quaternary volcanic products in this part of Shirak are cinder cones to the west of the Javakheti Ridge around Lake Arpi (Fig. 2), which are estimated to be of ~~Lower~~Early-Middle Pleistocene age, based on relationships with ~~valley series~~the mafic rocks and some river terraces (Kharazyan, 1983).

Overall ~~it is evident that~~ there have been no comprehensive geochronological studies carried out in ~~Armenia~~ Shirak using a single reliable technique, unlike those ~~produced~~ for similar sequences in Georgia (Lebedev et al., 2008a,b). ~~Furthermore there is a~~A lack of continuous exposure ~~, which~~ hampers judgement of the relative age of the different lavas. ~~Hence, w~~We have decided to ~~simplify the lavas, and~~ sub-divide the entire Pliocene-Quaternary suite close to the Javakheti Ridge into three ~~main~~ components: mafic flows (the dolerites) largely covering the topography developed on the ~~pre-existing Sevan-Akera suture~~basement (herein termed the *Valley Series*); more evolved flows built up into hills

~~rising up to 600 m~~ above the mafic flows (*Ridge Series*); and scattered cinder cones (*Cone Series*) (Fig. 2).

2.3~~2~~.1. Valley Series

The Valley Series consists of mafic flows ~~which reach~~with a maximum cumulative thickness of 200 m in eastern Lori province (Fig. 2). In the study area, the exposed sequence ~~usually~~ comprises at most four or five stacked flows up to 40 m thick in total, and directly overlies the Sevan-Akera suture and ophiolite sequence in the Dzoraged gorge and near Amasia (Fig. 2). We have collected samples from these locations, as well as from Lake Arpi and near Tashir in western Lori Province (Fig. 2). ~~Jrbashyan et al. (1996) suggested that these flows are likely to have been fissure-fed, and flowed from the north, before following the palaeo-topography of the Debed and Akhuryan river valleys to the east and south.~~ Valley Series samples collected from near Lake Arpi, Amasia, and Tashir (Fig. 2), consist of vesicular sub-ophitic dolerites with rare clinopyroxene or optically zoned plagioclase phenocrysts, set in a groundmass of 1-2 mm grain size consisting of clinopyroxene, plagioclase~~,~~ and oxides (see Supplementary Item 1). Clinopyroxene is commonly rimmed or almost totally replaced by red-brown amphibole, and there is occasional interstitial quartz and rare rounded quartz blebs. ~~None of the samples contain any fresh olivine~~Some samples contain very rare iddingsite crystals, and the few olivines found in thin section are rounded and <0.5 mm in diameter.~~although a number do have very rare 1-2 mm bright red cubic crystals, which may be iddingsite replacing olivine. The vesicles in the selected samples do not contain secondary infill.~~

2.3~~2~~.2. Ridge Series

Magmatism in this series is restricted to ~~more evolved~~ intermediate to felsic lavas ~~which have~~ built up into prominent topographic features. The highest of these is the north to south-trending Javakheti Ridge (Fig. 2), consisting of ~~multiple~~ rounded and glacially eroded ~~centres~~ peaks reaching ~3100 m above sea level, following a clear north-south trend running north into Georgia (Fig. 2). A second series of hills ~~and ridges~~ lie north of the Akhuryan River parallel to the Georgian border, but these are more topographically muted, reaching a maximum elevation of ~2400 m. Rock types from both ridges are almost exclusively ~~flows of~~ andesitic to dacitic ~~composition~~flows, with ~~some~~ rare black dacites and rhyolitic obsidians (Karapetian et al., 2001). ~~The flows that we have analysed~~Analysed flows range from ~~evolved~~ basaltic trachyandesites to dacites, ~~but~~and ~~they display~~there is much compositional and textural variation. Some of the least evolved samples (<60 wt.% SiO_2) contain 1-3 mm phenocrysts and glomerocrysts of clinopyroxene (rarely orthopyroxene) and ~~some~~ plagioclase, set in a flow-banded groundmass of plagioclase, clinopyroxene and Fe-Ti oxides, with accessory apatite and zircon. More evolved samples tend to contain significantly more plagioclase phenocrysts, and several have abundant 1-3 mm euhedral green-brown pleochroic amphiboles, but the presence of amphibole is not ubiquitous even in samples ~~containing~~ of similar SiO_2 and MgO concentrations. Many plagioclase crystals ~~in either clinopyroxene- or amphibole-dominated suites~~ are sieve-textured and some have corroded margins, ~~implying~~ usually taken to imply ~~either~~ the occurrence of magma mixing processes (Tsuchiyama, 1985; Tepley et al., 1999), ~~or rapid decompression during eruption (Nelson and Montana, 1992);~~ and many crystals are also optically zoned. Other evidence for magma mixing is the common occurrence of rounded quartz blebs with dark reaction coronas, and ubiquitous opaque rims on primary hornblende crystals (Tepley et al., 1999; Supplementary Item 1). ~~The~~ A black dacite contains a few clinopyroxene glomerocrysts, but mostly consists of a fine-grained groundmass dominated by prismatic to acicular feldspars.~~ Two photomicrographs from the Ridge Series are in Supplementary Item 1.~~ The Ridge Series include~~d~~s the only ~~polycrystalline~~ crustal xenoliths noted from our sampling, ~~in a single sample from~~at a single site near Darik, north of Lake Arpi (Fig. 2). The xenoliths consist of a coarse-grained groundmass of plagioclase, clinopyroxene, oxide, and rare iddingsite after olivine; and 2-3 mm phenocrysts of clinopyroxene and plagioclase, most consistent with inclusion of material from the Valley Series. A slight chilled margin is observed within the host.~~ Based on petrographic similarities, we suggest that this xenolith has been derived from the Valley Series flows immediately underlying the ridge, and not the deeper crust. The host itself also contains some sub-rounded quartz xenocrysts (see Supplementary Item 1) and degraded plagioclase crystals with a speckled appearance, sometimes rimmed by new growth. The presence of quartz xenocrysts with reaction rims is taken to indicate assimilation of host rocks~~.

2.3~~2~~.3. Cone Series

Scattered cinder cones are present in the Akhuryan valley near Lake Arpi (Fig. 2), often tens of metres high and~~ reaching~~ several hundred metres across. ~~A notable lack of erosion of these cones suggests they represent the youngest volcanism in the area.~~ We have collected samples from Sepasar, Eznasar, and Kaputkogh cones (Fig. 2). Most are composed of poorly welded unsorted glass~~y~~ or scoria~~ceous~~ fragments~~ of various sizes~~, and more coherent scoria bombs reaching a few tens of centimetres in diameter. The Cone Series compositions mirror the least evolved of the ridge series, the majority of samples containing differing proportions of plagioclase and clinopyroxene as phenocrysts and groundmass (Supplementary Item 1). Bombs from Kaputkogh and Eznasar contain ubiquitous mm-scale rounded quartz xenocrysts, whereas those at Sepasar appear more mafic, and devoid of foreign material.~~. A photomicrograph of a typical Cone Series sample is available in Supplementary Item 1.~~

3. Analytical methods

Samples were powdered in an agate ball mill at Durham University. Major element analysis was conducted on fused glass beads using a PANalytical Axios Advanced X-Ray Fluorescence (XRF) spectrometer at the University of Leicester. Leftover fractions of the powder from XRF analysis were digested using a standard HF and HNO_3 technique prior to trace element analysis. Solutions were run on a Thermo X2 inductively-coupled plasma mass spectrometer (ICP-MS) at the Northern Centre for Isotopic and Elemental Tracing (NCIET) at Durham University. Accuracy, precision, and reproducibility were monitored using blanks, multi-run and within-run duplicates, Re-Rh spike solutions, and five international reference standards. Standard W2 (n = 15) gave first relative standard deviations of 5% or better for most transition metals (excepting 10% for Sc, 12% for Cr, 6% for Ni), the large ion lithophile elements (LILE) and the rare earth elements (REE) (7% for La, 6% for Ce). Elemental results are recorded in Table 1 and Supplementary Item 2, the latter also containing standard results.

Radiogenic Nd and Sr isotope analysis was conducted at NCIET, with column chemistry for elemental pre-concentration based on the method of Dowall et al. (2007). Powders were digested in HF and HNO_3, and solutions run through 1 ml pipettes containing several drops of dilute Sr-spec resin to collect the Sr-bearing fraction. The high field strength element (HFSE)- and rare earth element (REE)-bearing fraction from these columns was run through 10 ml Bio-Rad polypropylene columns containing 1 ml of Bio-Rad AG1-X8 200-400 mesh anion-exchange resin. Neodymium was collected as part of a general rare earth element fraction. Analysis was conducted on a Thermo Neptune Mass Collector ICP-MS. Strontium was run in a single batch during which time blanks averaged 88 pg Sr (n = 6). International reference standard NBS987 gave a mean of $^{87}Sr/^{86}Sr$ = 0.710263 ± 0.000012 (2σ, n = 12), comparable to a preferred value of 0.710240, and providing a minimum uncertainty of 16 ppm (2σ). No correction was applied to the final results. Neodymium was run in two separate batches, with blanks averaging 10 pg Nd (n = 6). During the first run, a combination of the J&M standard and a Sm-doped version gave a mean $^{143}Nd/^{144}Nd$ = 0.511098 ± 0.000007 (2σ, n = 13), and a minimum uncertainty of 13 ppm (2σ). During the second run, $^{143}Nd/^{144}Nd$ = 0.511100 ± 0.000007 (2σ, n = 13), giving an uncertainty of 14 ppm (2σ). For consistency between the runs, all results were normalised to a preferred value of 0.511110. Results are presented in Table 2.

4. Geochemistry

4.1. Sample freshness

We collected the freshest available samples at each locality, and this is reflected in loss-on-ignition (LOI) values typically <1 wt.%, mostly un-weathered feldspars and mafic minerals, and an overall lack of sericite and chlorite in thin section. Major and trace element data, particularly element vs. SiO_2 plots (see Section 4.2), have trends consistent with magmatic processes, particularly for CaO, MgO, K_2O, and Na_2O, as opposed to the widespread scatter expected during sub-solidus alteration which easily mobilises these elements (e.g. Cann, 1970; Pearce, 1996). $^{87}Sr/^{86}Sr$ isotope results are commonly affected by hydrothermal alteration, but here are depleted with no sign of a trend towards high $^{87}Sr/^{86}Sr$ at constant $^{144}Nd/^{143}Nd$ (Table 2); nor is there a correlation between LOI and isotopic ratios.

Furthermore, the samples were erupted in an intra-continental high plateau so have not interacted with high $^{87}Sr/^{86}Sr$ seawater or been exposed to tropical weathering.

4.2. Major and trace element characteristics

4.2.1. Valley Series

The Valley Series lavas (~49-55 wt.% SiO_2) are mostly mildly alkaline trachybasalts based on the total alkali-silica classification (Le Bas et al., 1986; Fig. 3a), and belong to the medium-K series of Peccerillo and Taylor (1976; Fig. 3b). They are evolved, with 4-7 wt.% MgO and low molar Mg# from 46 to 58. Overall the samples have low TiO_2, moderate-high Al_2O_3, and a sodic character (Na_2O/K_2O = 2.7-4.1) (Fig. 4). Trace element abundances and trends can be seen on Figure 5. Also, the lavas have low Sc (<25 ppm), moderate Cr, Ni, and large ion lithophile element (LILE) abundances (e.g. Ba = 280-450 ppm and Sr = 540-720 ppm). ~~Of the high field strength elements (HFSE), they have considerable enrichment in Zr (170-210 ppm), and super-chondritic Zr/Hf (47-52), with Nb/Ta (17-22).~~ Chondrite-normalised (CN) ~~Rare Earth Element (REE)~~REE abundances are light REE (LREE) enriched (La/Yb_{CN} = 5-9; calc-alkaline), with flat heavy REE (HREE) patterns around 15-20 times chondritic abundances (Fig. 6a). The patterns are split into two groups, one with lower LREE and higher HREE concentrations and vice versa, the patterns crossing over at around Pr-Nd. There are some very slight negative Eu anomalies relative to the ~~middle REE (~~MREE~~)~~. The extended Primitive Mantle-normalised (PMN) plot (Fig. 6b) shows that the samples have spiky LILE patterns (modest positive Ba, Rb, Th, Sr anomalies), prominent negative K, Nb-Ta, and Ti anomalies and positive Zr-Hf anomalies relative to the REE, with super-chondritic Zr/Hf ratios of 47-55.

4.2.2. Ridge Series

The more evolved Ridge Series (~57-68 wt.% SiO_2, 2-5 wt.% MgO, Mg# = 41-55) has a wide range of sub-alkaline compositions from basaltic trachy-andesite through to dacite (Fig. 3a). Samples largely belong to the medium-K series, although two plot in the high-K field (Fig. 3b). They have noticeably lower TiO_2, $Fe_2O_{3(t)}$ and P_2O_5 concentrations compared to the valley series, and are slightly less sodic (Na_2O/K_2O = 1.4-2.5) (Fig. 4). Transition metal abundances are lower than the Valley Series, but the Ridge Series has higher Ba and Zr and lower Sr and Nb concentrations compared to the mafic lavas (Fig. 5). Chondrite-normalised LREE patterns (Fig. 6c) are similar to the Valley Series, again splitting into two groups with higher or lower LREE concentrations. Ridge samples have La/Yb_{CN} from 8 to 18, with a highly fractionated M-HREE distribution such that some samples have a U-shaped pattern. There are small negative Eu anomalies. Primitive Mantle-normalised patterns (Fig. 6d) differ slightly from the Valley Series in having positive Rb spikes, and more pronounced negative Nb-Ta, P and Ti anomalies.

4.2.3. Cone Series

The Cone Series splits into three groups, the most mafic being the two cones at Sepasar (58 wt.% SiO_2, but low Mg# = 43), and the most felsic at Eznasar (62 wt.% SiO_2, Mg# = 52). The large cone at Kaputkogh has intermediate silica content relative to the other cones, although it has the highest Mg# of 54. The ~~inverted relationship between Mg# and silica content may be derived from the most primitive magmas happening to be contaminated by relatively higher proportions of associated felsic magmas or crustal components during ascent. This~~

~~relationship means that the Cone Series do not form a significant part of the discussion on overall fractionation and contamination trends in Section 5.2. Finally, t~~The cones have higher Nb and Zr concentrations relative to the other series (Fig. 5); REE patterns for the Cone Series are clearly bimodal (Fig. 6e), whilst the Primitive Mantle-normalised plots look similar to the Valley Series (Fig. 6f).

4.3. Radiogenic isotope geochemistry

Most samples span a narrow range from $^{87}Sr/^{86}Sr$ = 0.70416 to 0.70446 and $^{143}Nd/^{144}Nd$ = 0.51280 to 0.51287, giving a range of values in epsilon notation from εNd = +3.1 to +4.6. No age corrections were applied owing to the young age of the rocks. On Figure 7a, there is overlap between samples from the Valley, Ridge, and Cone series, and the samples display only a little isotopic enrichment with major element evolution (Fig. 7b). Overall, the samples lie on the mantle array between bulk silicate earth and depleted MORB mantle, and there is no clear evidence of any trends which might be related to mixing of different mantle end members (e.g. EMI or EMII) or old, isotopically enriched crustal contaminants. The ~~cones~~ Cone and ~~Valley~~ Ridge Series samples ~~with~~ containing quartz xenocrysts are not isotopically different from the other samples.

Regionally, the samples are significantly more depleted than most Pliocene-Quaternary centres in north-west Iran and Mount Damavand (Alborz), and Tendürek volcano in Eastern Anatolia (Fig. 7a). The Iranian volcanic rocks were erupted through thicker lithosphere than beneath Armenia (e.g. Liotard et al., 2008; Kheirkhah et al., 2009; Mirnejad et al., 2010; Davidson et al., unpublished data; Allen et al., ~~in press~~2013). Samples are also more depleted than asthenospheric melts from eastern Iran which have trends towards the

EM-II mantle end member (Saadat et al., 2011; Saadat and Stern, 2012). Instead, results are closest to the few analyses conducted on the large stratovolcano, Mount Ararat, close to the Armenian border (Pearce et al., 1990; Kheirkhah et al., 2009), and nearly identical in terms of $^{143}Nd/^{144}Nd$ to six analyses of the ~3.25-2.05 Ma valley series lavas from southern Georgia, and from eight Late Miocene sub-alkaline basalts in central Georgia (Lebedev et al., 2006; 2007). Radiogenic isotope results are not yet widely published from other Armenian Pliocene-Quaternary centres, but Savov et al. (2007) and Lin et al. (2011) reported values from $^{87}Sr/^{86}Sr$ = 0.7041 to 0.7051 and $^{143}Nd/^{144}Nd$ = 0.5128 to 0.5129 from ~~across much of the country~~various locations.~~ This is a similar narrow range of results compared to our study, except for some $^{87}Sr/^{86}Sr$ values extending to more enriched compositions outside of the mantle array. The overall lack of enriched, continental crust-like values and crustal or mantle xenoliths is an interesting feature of magmatism across the orogenic plateau in spite of varied basement compositions.~~

5. Discussion

5.1. Volcano-tectonic interaction

Before considering the petrogenesis of the magma series, we first address the location of volcanic activity with respect to major crustal structures. The internal part of the orogenic plateau is not undergoing contractile deformation (Jackson et al., 1995; Vernant et al., 2004), but internal reorganisation of the plateau during the on-going convergence between Arabia and Eurasia means that Eastern Anatolia, the Lesser Caucasus, and northwest Iran are criss-crossed by numerous active strike-slip fault systems (Rebaï et al., 1993; Koçyiğit et al., 2001). These systems, which often tally with pre-existing crustal discontinuities, have been widely implicated in providing a locus for Quaternary magmatic activity through the production of highly localised pull-apart zones (Dewey et al., 1986; Karakhanian et al., 1997, 2002; Avagyan et al., 2010; Shabanian et al., 2012).

In Armenia, the active Pambak-Sevan-Syunik right-lateral strike-slip fault system is one such structure exploiting the existing discontinuity of the Sevan-Akera suture zone (Fig. 2). All of the magmatism described in this study originated just to the north of the fault zone, and so we propose that this location sat over a region of localised upper crustal extension at the time of magmatism. No active extension is recorded from the Shirak region from earthquake focal mechanisms or geomorphic features. Blanket coverage of most pre-existing fault structures by the Valley Series, and the absence of linear arrays of cinder cones (e.g. Dewey et al., 1986), may have helped obscure this association between faulting and magmatism. The south-eastern termination of the Pambak-Sevan-Syunik fault in the Syunik region is characterised by widespread volcanism in the area south of the fault, with little volcanism to the north (Kharakhanian et al., 2004). The Syunik centres therefore appear to have developed along the complementary trailing imbricate fan (Woodcock and Fischer, 1986) to the Shirak volcanic rocks. The north-south trending volcanism along the Javakheti Ridge is consistent with this idea.

Not all young Armenian centres fit a simple fault-control hypothesis: Aragats volcano, the largest centre in the country, is situated in a region apparently not crossed by presently-active faults (Kharakhanian et al., 2004). It is possible that older faults may be obscured by lava flows, or that some other control on the location of Aragats may apply, such as a local thin-spot in the lithosphere which focussed melting beneath the plateau.

5.2. Fractionation and contamination processes

5.2.1. Fractional crystallisation – minerals involved

No sample ~~of~~ from the mafic Valley Series lavas is close to a primary melt (MgO is typically < 7 wt.%), so it is assumed that they have already evolved at depth involving a typical fractionation assemblage of olivine and spinel. In the major element data, clear falling trends for the Valley Series against typical indices of fractionation, such as SiO_2, TiO_2, MgO, and CaO (Fig. 4), corroborate with the observed mineral assemblages in confirming that clino- and orthopyroxene and Fe-Ti oxides were important in Valley Series evolution. Al_2O_3 concentrations cover a narrow range with no trends, indicating that feldspar fractionation was not an important feature of the Valley Series; likewise with P_2O_5 concentrations and apatite. Trace element abundances, such as Ba, Th, and La, display rising trends against SiO_2 in the Valley Series, which confirms their incompatible behaviour (Fig. 5). In the more evolved Ridge and Cone Series, clear falling trends emerge against SiO_2 for Al_2O_3, P_2O_5, Sr, Nb, Zr and La, pointing to addition of plagioclase, along with small proportions of zircon and apatite, to the fractionating assemblage.

The overall pattern of Shirak magmatism can be compared with Eastern Anatolia. Volcanic centres that display either high- or low-Y trends relative to Rb, are associated with fractionation of anhydrous (plagioclase, olivine, pyroxenes, and magnetite), or hydrous assemblages (including amphibole, which is compatible with Y) respectively (Pearce et al., 1990) (Fig. 8). The mafic Valley Series rocks in Shirak follow the same moderate to low-Y trend as the Kars plateau/Mt. Ararat systems, which may indicate amphibole fractionation has taken place at depth, given that amphibole is not seen in any of the Valley Series thin sections. Another indication of amphibole fractionation is the compatible middle to heavy REE (Sm-Lu) showing falling trends as the three series evolve (Fig. 6). The ridge series lavas have a steeply falling trend for Y against Rb, which may reflect increased partition coefficients for both clinopyroxene and amphibole for Y as these minerals fractionate from more evolved rocks (Pearce et al., 1990).

5.2.2. Magma mixing

The petrographic evidence for magma mixing (zoned plagioclases, sieve textures, reaction rims and quartz blebs) also needs to be reconciled with geochemical data. Major element plots, especially TiO_2, CaO and MgO vs. SiO_2 (Fig. 4) have straight line trends which are widely associated with mixing of two compositionally distinct magmas (Langmuir et al., 1978), rather than the curved trends associated with fractional crystallisation. In the field, the key observation is that the most evolved rocks found in the area, the obsidians at Agvorik, underlie the mafic Valley Series, so felsic magmas had already been erupted by the time of mafic magma injection.

5.2.3~~2~~. Crustal contamination

Many models (e.g. Keskin et al., 1998) consider assimilation-fractional crystallisation (AFC) processes to be important in magma genesis in the plateau. ~~The extent to which crustal contamination has affected primary magma compositions may vary widely across the collision zone.~~ Keskin et al. (1998) argue~~s~~ that recent mafic to felsic samples from Eastern Anatolia have undergone significant amounts of ~~combined assimilation-fractional crystallisation (AFC)~~AFC coinciding with enriched $^{87}Sr/^{86}Sr$ ratios up to 0.7065 (Pearce et al., 1990). In Shirak, ~~there is~~samples have only ~~very~~ a slight variation in isotope ~~signatures~~ ratios relative to ~~silica concentration~~SiO_2 (Fig. 7b)~~ in the Valley and Ridge Series~~, and samples containing quartz xenocrysts ~~have neither higher $^{87}Sr/^{86}Sr$ nor lower $^{143}Nd/^{144}Nd$ ratios than~~do not have different isotopic ratios to the other lavas - a feature that should strongly support the hypothesis that fractionation and magma mixing were the dominant processes. Also, preliminary results from elsewhere in Armenia, including the Aragats volcanic system, show little clear isotopic evidence for contamination (Savov et al., 2007; Lin et al., 2011) in spite of Aragats erupting through the SAB basement which has enriched $^{87}Sr/^{86}Sr$ ratios of up to 0.7303 - any contamination should be easily indentified (Bagdasaryan and Gukasyan, 1985). However, along-strike from Shirak at Artvin in eastern Turkey, Eocene rocks have measured $^{143}Nd/^{144}Nd$ of 0.512663 - 0.512854 and $^{87}Sr/^{86}Sr$ of 0.705148 - 0.704233 (Aydinçakir and Şen, in press). These values are very similar to the Shirak lavas (Fig. 7), so partial melting or assimilation of similar Eocene crust may be very difficult to decipher geochemically.

~~In terms of trace element ratios, o~~On a Th/Yb vs. Ta/Yb plot (Pearce, 1983) (Fig. 9), the ~~most mafic Valley Series lavas have enriched Ta/Yb ratios of 2.9-4.7, pointing to lower degrees of melting or a more fertile source than expected for mid-ocean ridge basalts (MORB) (Ta/Yb = 0.9; Sun and McDonough, 1989). The lavas also lie above the mantle array in the calc-alkaline field, indicative of a small subduction-related component (see Section 5.3). The~~ three series form a consistent linear trend sub-parallel to the mantle array, with the felsic rocks having compositions more enriched than those of typical continental ~~upper~~ crust (Rudnick and Gao, 2003). On this diagram, the fractional crystallisation trend for a typical amphibole-bearing assemblage is shown. ~~An important consideration is that t~~The samples plot ~~both~~ to the left of the fractionation trend ~~and left of a direct mixing line with~~

~~upper continental crust (Taylor and McLennan, 1985). This result~~which suggests that AFC processes may be operating - although choice of mineral assemblages and partition coefficients can easily affect the FC trend. AFC modelling (Powell, 1984) using the average composition of the ~~Eocene Turkish~~ Artvin rocks (Aydinçakir and Şen, in press) quite reasonably reproduces the trend of the Shirak lavas, but the ~~and a~~ ratio of assimilation to fractionation is high at= 0.8. ~~Results of modelling shown on Figure 9 indicate that this significant level of assimilation may produce a trace element trend more compatible with the composition of the Shirak rocks than attainable by pure FC processes. Unfortunately, isotopic analyses are not available for these Turkish rocks~~. Modelling thus shows that large volumes of isotopically similar material can be incorporated into the Shirak lavas without a significant effect on trace element evolution. Studies of disequilibrium textures and mineral chemistry might better elucidate the processes involved in magma evolution.

5.3. Mantle source and partial melting

5.3.1. ~~Overall nature of the m~~Mantle source of the ~~Shirak lavas~~Valley Series

The presence of LILE and HFSE anomalies on normalised plots of mafic rocks (Fig. 6b) are normally taken to indicate a subduction-modified mantle source owing to the retention of HFSE in the slab, and the comparative mobility of the LILE/REE during slab heating and dewatering into the overlying mantle wedge (e.g. Pearce and Peate, 1995). We have already introduced the Th/Yb vs. Ta/Yb diagram (Fig. 9). The alkali basalts from Shirak plot above the mantle array which is commonly taken to indicate the presence of a subduction-modified source. However, they also have very much higher Ta/Yb ratios than many subduction-related rocks. ~~We take this to indicate that t~~The Shirak lavas are therefore derived from an incompatible element-enriched mantle source, or ~~one that has undergone a~~are derived from a limited degree of melt extraction. As these lavas erupted >20 Myr after the end of Neo-Tethyan subduction, and there is no evidence for a slab at shallow depths beneath Eastern Anatolia and Armenia at the present day (Zor et al., 2008), it is improbable that a normal supra-subduction zone hydrated asthenospheric mantle wedge was involved in the origin of the Shirak lavas. Therefore, the subduction-like characteristics are likely to be derived from a fertile source within the mantle that had been inherited its slab-related geochemical component from earlier Neo-Tethyan subduction~~ processes~~. Crustal contamination is unlikely be responsible for the subduction-like characteristics of the Shirak lavas, as LILE and HFSE anomalies are significant in even the most mafic samples.

Some of the trace element characteristics of the valley series may help constrain the mineralogy of the mantle source. Overall, the flat normalised HREE patterns (Fig. 6a) indicate a spinel-facies mantle source at <70 km, unless the degree of partial melting was very high (>25%) in order to completely consume any garnet present at depths of >70 km. This is unlikely given the overall LREE-enriched and Nb-Ta/HREE-enriched trace element patterns (Fig. 6b) which point towards modest degrees of partial melting of an enriched source. Low Sc concentrations (<25 ppm) in all Valley Series samples may indicate residual clinopyroxene, another indicator of a low degree of partial melting, but it is also possible that extensive pyroxene fractionation prior to eruption has resulted in these low values. Intra-LILE variations, including low Ba/Rb (<25) and Rb/Sr ratios (<0.05), do not point towards amphiboles or phlogopite playing an important role during melting (e.g. Furman and Graham, 1999). Melting therefore took place beneath the Armenian crust at depths of ~45-70 km.

One unusual feature of the Shirak lavas is high ocean island basalt (OIB)-like Zr concentrations (~200 ppm) and Zr/Hf ratios (41-52) relative to MOR and arc basalts, the latter having chondritic Zr/Hf ratios of 35-39 (Weaver et al., 1987; David et al., 2000; Pfänder et al., 2007). Nb/Ta ratios range from 16-22 relative to the chondritic ratio of 19.9 (Pfänder et al., 2007). Mafic samples from other nearby centres in the plateau, including Tendürek and Ararat, show similar features (Fig. 10). Lower ratios in the Ridge Series compared to the Valley Series might~~can~~ be explained by contamination with crustal material such as the Eocene basement (Fig. 10). However, the high Zr/Hf ratios of 47-52 in the less evolved Valley Series are a primary feature of the magmas.

There is little correlation between Zr and Zr/Hf ratios (not shown), indicating that zircon accumulation cannot be directly responsible for the high Zr-Zr/Hf character of the Shirak lavas. HFSE fractionation in OIBs may be due to: (1) residual or fractionating clinopyroxene (David et al., 2000); (2) fractionation of Ti-bearing phases such as rutile, ilmenite, and amphibole (Foley et al., 2000; Tiepolo et al., 2001); (3) melting of recycled eclogite or garnet pyroxenite (Pfänder et al., 2007); or (4) the occurrence of carbonate metasomatism (Dupuy et al., 1992). For option (1), fractionation of clinopyroxene only has a modest effect upon Zr/Hf ratios (Pfänder et al., 2000). Our modelling of pure clinopyroxene fractional crystallisation from a starting composition with Zr/Hf and Nb/Ta of primitive mantle shows that unrealistic amounts of ~~clinopyroxene~~ fractionation are required to generate the Shirak samples (Fig. 10). In option (2), fractionation of titanate phases such as rutile and ilmenite can generate very high Zr/Hf and Nb/Ta ratios, with $D_{Zr/Hf}$ and $D_{Nb/Ta}$ both <1 (Pfänder et al., 2000); however, titanate fractionation would also strongly reduce overall Nb concentrations, a feature not seen in the Valley Series. Partial melting of garnet-bearing lithologies (option 3) is invoked in many OIB examples (see Pfänder et al., 2007) but can be ruled out here on the basis of flat normalised HREE patterns in the Shirak lavas – these are not OIB-like magmas (Fig. 6b). Where carbonates are invoked in the mantle source (e.g. Dupuy et al., 1992; Hoernle et al., 2002) (option 4), resultant alkaline melts or mantle xenoliths have very high Sr and Ba of >>1000 ppm, and in spite of high Zr/Hf ratios many carbonatites ~~have very low overall concentrations of these two~~have very low concentrations of these elements (Ionov et al., 1993). This is not the signature of the Shirak samples hence carbonate metasomatism is unlikely in this case. Several studies have shown that amphibole and phlogopite fractionate the HFSE (Moine et al., 2001; Tiepolo et al., 2001; Chakhmouradian, 2006), with Chakhmouradian (2006) demonstrating that low-Ti amphiboles have high Zr/Hf ratios of ~60-200. Hence these minerals can impart high Zr/Hf on a melt; but it is still unclear what the high overall Zr concentrations in the Shirak lavas are caused by - this feature is normally attributed to ancient recycled oceanic crust in OIBs (Weaver, 1991).

5.3.2. Modelling of partial melting

Any model of partial melting conditions for Shirak has to be based on the HREE and HFSE, making the assumption that neither set of elements were transported into the lithospheric mantle source in a slab-derived fluid (Pearce and Peate, 1995). Therefore, we have constructed non-modal batch melting curves using Dy, Yb and Nb (ignoring Zr owing to its anomalous behaviour), in order to constrain the degree of partial melting needed to form the Valley Series. We have taken the approach of Pearce et al. (1990) in assuming that hydrous phases such as amphibole and phlogopite (if present) are completely consumed during melting and do not contribute to the melt model. Given the high Ta/Yb ratios of even the least evolved Valley Series samples (Fig. 9), it is reasonable to compare the melting of depleted MORB mantle (DMM) (Workman and Hart, 2005) with a more incompatible-element enriched source, in this case primitive mantle with 1% bulk continental crust extracted, as used by Fitton and Godard (2004) for the Ontong Java oceanic plateau.

Although we can easily model HREE and HFSE ratios (see below), fractional crystallisation of olivine, spinel, plagioclase and pyroxene versus fractionation of amphibole from primary magma have competing effects on absolute REE and HFSE concentrations. Often, elemental values for basalts in modelling are back-calculated to 9 wt.% MgO to negate the effects of plagioclase and pyroxene crystallisation (Pearce & Parkinson, 1993). However, valley series Dy and Yb concentrations are near-constant in spite of varying MgO and SiO_2 (Fig. 6a), probably due to the competing effects of amphibole and clinopyroxene fractionation, so no realistic back-calculation can be applied. Therefore, we simply attempt to model the elemental ratios of the last-evolved Valley Series lava (7 wt.% MgO) and assume that this best reflects the conditions of partial melting.

Modelling results (Fig. 11) indicate that melting of a garnet peridotite cannot reproduce the compositions of the Valley Series lavas, a finding consistent with the flat normalised HREE patterns (Fig. 6a). Spinel peridotite partial melting curves do intersect the valley series at low degrees of melting, with the DMM melting curve on Figure 11 giving 0.1-0.5% melting. In contrast the more fertile source gives 2-5% melting, which is perhaps more realistic than the tiny proportion of melting required from a DMM source and the difficulties of extracting such a small volume of melt (e.g. Hirth and Kohlstedt, 1995). This spinel peridotite melting outcome is also consistent with geophysical surveys indicating a seismically slow~~, probably partially molten~~ mantle at depths of ~50 km beneath Armenia (e.g. Koulakov et al., 2012).

5.4. Reconciliation with geophysical and geodynamic models

5.4.1. Extent of lithospheric delamination

Debate exists over the extent of lithospheric delamination beneath Eastern Anatolia following the cessation of subduction beneath the Bitlis-Zagros suture (e.g. Keskin, 2003). Seismic surveys indicate a crustal thickness of ~40-45 km, but with significant negative P- and S-wave seismic velocity anomalies beneath, extending from ~50 to ~250 km depth, between eastern Turkey, Armenia, the Black Sea and northwest Iran, concurrent with many Pliocene-Quaternary volcanic centres (e.g. Piromallo and Morelli, 2003; Maggi & Priestley, 2005; Zor et al., 2008; Koulakov et al., 2012). The anomaly has been used by these authors to argue for the presence of hot, perhaps partially molten, asthenosphere, but several authors extend these conclusions to the possibility that there is also no mantle lithosphere 'lid' beneath the Anatolian crust (e.g. Keskin, 2003; Zor et al., 2008) and the Caucasus (Koulakov et al., 2012). In this case, mafic magmatism in Shirak would have to have an asthenospheric source.

There are significant implications for magmatism in this scenario. The impact of hot upwelling asthenosphere on the thickened Lesser Caucasus arc crust should result in extensive lower crustal melting, as observed in the Puna Plateau of the Andes, and the Great Basin Altiplano in the western U.S. (Allmendinger et al., 1997; Babeyko et al., 2002; Best et al., 2009). Going back to our geochemical results, this model of whole-scale lithospheric delamination proposed for the Puna Plateau is incompatible with the observed silica-undersaturated magmatism in Shirak, which bears little evidence for large-scale crustal interaction. We conclude that there is sufficient lithospheric mantle beneath the Armenian crust to act as a thermal barrier between the asthenosphere and crust (Fig. 12), protecting the crust from melting and infiltration by hot asthenospheric melts in the manner described by Babeyko et al. (2002). A~~Furthermore, a~~n asthenospheric source for the Shirak magmas ~~would not~~cannot have been influenced by a subducting slab, because subduction processes ended ~~beneath the region~~ prior to the Miocene. Hence the Shirak magmas would not have subduction-like trace element characteristics, and instead should closely resemble OIB. There are asthenosphere-derived OIB-like lavas without subduction-related geochemical signatures in Eastern Iran (Saadat et al., 2010; Saadat and Stern, 2012) and in the Arabian foreland (Lustrino et al., 2010). The Iranian alkali olivine basalts show trends towards an EMII-like isotope signature (particularly with respect to Pb isotope ratios) (Zindler and Hart, 1986). They also and contain pyroxenite xenoliths from the lithospheric mantle, plagioclase megacrysts of uncertain origin, and some lower crustal gabbroic xenoliths (Saadat and Stern, 2012). These lavas are ~~clearly~~ distinct in terms of xenolith content, trace element signatures and isotope geochemistry from those erupted in Shirak.

5.4.2. Geodynamic model

Our geodynamic model is presented schematically in Figure 12. We propose that, upon the termination of Neo-Tethyan subduction along the Bitlis-Zagros suture during the Oligocene, Armenia lay in a continental back-arc position relative to the former subducting slab and mantle wedge. Modelling studies have suggested that an old slab may be able to persist or ‘stall’ in the upper mantle without breaking off for up to 20 Myr after terminal collision (van Hunen and Allen, 2011). Delayed break-off of the Neo-Tethyan slab from Arabia beneath Eurasia may thus be responsible for the upsurge in magmatism since 10 Ma, and particularly in the Pliocene-Quaternary, across the orogenic plateau (e.g. Keskin, 2003), concurrent with the influx of hot asthenosphere into the region the slab once occupied. In Eastern Anatolia, the region immediately above the detached slab might lack mantle lithosphere, and asthenospheric and crustal melting would combine to produce arc-like magmas (Fig. 12) (Keskin, 2003). However, as we have already discussed, it is improbable that whole-scale lithospheric mantle delamination occurred beneath Armenia because we do not see attendant whole-scale lower crustal melting. The former asthenospheric mantle wedge of the Neo-Tethyan arc system would be refrigerated by the presence of a stalled slab, and rapidly converted into lithospheric mantle over the 15-25 Myr following terminal collision (c.f. Holt et al., 2010). This depleted lithospheric mantle could be stable and buoyant enough to be at least partially preserved following the eventual detachment of the underlying oceanic slab, whilst the aforementioned influx of convecting asthenosphere would trigger partial melting in the overlying lithospheric mantle, as well as providing a thermal support for the orogenic plateau (Fig. 12).

Another potentially important consideration is that, although much of the LILE and LREE budget of the southern Neo-Tethyan slab may have been delivered to the lithosphere before continental collision, a stalled slab and associated sediments would continue dewatering before break-off. This would contribute to the subduction-like signature on the mantle frozen-in beneath Armenia. Other Pliocene-Quaternary centres in the collision zone, such as the foreland volcanic system at Karacadağ, may result from asthenospheric melting beneath thin spots in the lithosphere (Lustrino et al., 2010; Ekici et al., 2012). Mantle-derived volcanism is also apparent even in the 50+ km thick crust of the Elbrus region of the Greater Caucasus (Lebedev et al., 2006b; Koulakov et al., 2012), and it is here that magmatism may be related to melting during collisional thickening of lithospheric mantle and the breakdown of hydrous mineral phases such as micas and amphiboles (e.g. Pearce et al., 1990; Allen et al., ~~in press~~2013) or to asthenospheric upwelling during lithospheric dripping (see Sosson et al., 2010) (Fig. 12)~~ - further work is required to distinguish these hypotheses~~.

6. Conclusions

- Mafic and more evolved Pliocene-Quaternary lavas in Shirak, NW Armenia, were emplaced through a former continental margin arc sequence as a result of localised extensional tectonics within the present-day Arabia-Eurasia collision zone.
- Magmas evolved from mafic through to dacitic compositions by fractional crystallisation dominated by pyroxene, amphibole and plagioclase; and although evolved samples contain quartz xenocrysts, none preserves clear isotopic evidence for large-scale crustal assimilation - magma mixing appears to be the dominant petrogenetic process. We conclude that if assimilation did occur, it was of local ~~Mesozoic to Early Cenozoic~~ arc-related crust of a similar isotopic composition to the ~~primary~~ Shirak melts.
- The least-evolved magmas preserve trace element evidence for derivation by moderate degrees of melting (~3-4%) from a shallow, spinel-facies lithospheric mantle source with an inherited subduction component probably related to earlier Tethyan subduction processes. T~~Unusually, t~~hey contain high Zr concentrations and high Zr/Hf ratios which ~~cannot be explained by fractional crystallisation processes, and~~ are an intrinsic feature of the source or partial melting process.
- ~~L~~The presence of lithospheric mantle beneath Armenia is a requirement for geodynamic models of the region~~,~~ in order to prevent the occurrence of whole-scale lower crustal melting.

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Table captions

Table 1. Major and trace element data for selected samples from the valley, ridge, and cone series, Shirak. LOI = loss-on-ignition.

Table 2. Measured Nd and Sr isotope compositions of the valley, ridge, and cone series, Shirak.

Supplementary Items

Item 1. Selected photomicrographs of samples from the valley, ridge, and cone series, Shirak.

Item 2. Complete whole rock major and trace element data for the Shirak lavas, including trace element standards.

Figures

Figure 1. Map of the Turkish- Iranian plateau with shaded digital topography, showing locations of Pliocene-Quaternary volcanic centres (cones) and the study area (rectangle).

Figure 2. Digital topography map of the study region with geological features simplified from work by Khachatur Meliksetian, Gevorg Navasardyan and Sergey Karapetyan of the Institute of Geology of the National Academy of Sciences of Armenia, plus outline map of Armenia showing administrative boundaries and regional coverage of Late Miocene-Quaternary magmatic products.

Figure 3. (a) Total alkali-silica classification (Le Bas et al., 1986) and (b) K_2O vs. SiO_2 classification (Peccerillo and Taylor, 1976).

Figure 4. Major element variation diagrams for the Shirak lavas.

Figure 5. Minor and trace element variation in the Shirak lavas.

Figure 6. Rare earth element and extended trace element normalised plots. Chondrite normalisation values from McDonough and Sun (1995) and Primitive Mantle and OIB values from Sun and McDonough (1989).

Figure 7. (a) Nd-Sr isotope plot for Shirak lavas, compared to mafic centres within the collision zone. Pliocene-Pleistocene valley series in southern Georgia and Late Miocene mafic lavas from the Elbrus region of Southern Russia - Lebedev et al. (2007; 2010); NW

Iran minor centres, Tendurek, Ararat, Kurkistan - Kheirkhah et al. (2009), Allen et al. (~~in press~~2013); Damavand - Davidson et al. (unpublished data), Mirnejad et al. (2010); Artvin, Eastern Turkey - Aydiçakir and Şen, in press). Mantle end members and array - Zindler & Hart (1986). (b) Variation of Nd and Sr isotopes as a function of magmatic evolution.

Figure 8. Fractional crystallisation (FC) trends within the Shirak lavas (symbols as per previous diagrams). Data and FC vectors for basic to acidic rocks for Eastern Anatolia are from Pearce et al. (1990). pl = plagioclase; o = olivine; opx = orthopyroxene; cpx = clinopyroxene; hb = hornblende; gnt = garnet.

Figure 9. Th/Yb vs. Ta/Yb diagram (Pearce, 1983) for the Shirak lavas, showing a ~~fractional crystallisation (FC)~~an FC vector for a hydrous assemblage, taking into account increasing partition coefficients during magmatic evolution (after Keskin et al., 1998), and an ~~assimilation FC~~AFC vector as described on the figure. Eocene rocks from ~~North-Central~~Eastern Turkey, likely to be similar to those directly underlying the Shirak lava series, are plotted (~~Keskin et al., 2008~~Aydinçakir and Şen, in press). ~~Upper crust – Taylor and McLennan (1985),~~Crust (UCC: upper continental crust; MCC: middle crust; LCC: lower crust; BCC: bulk continental crust, all from Rudnick and Gao, 2003). A~~a~~ctive margins - Pearce (1983). See text for discussion.

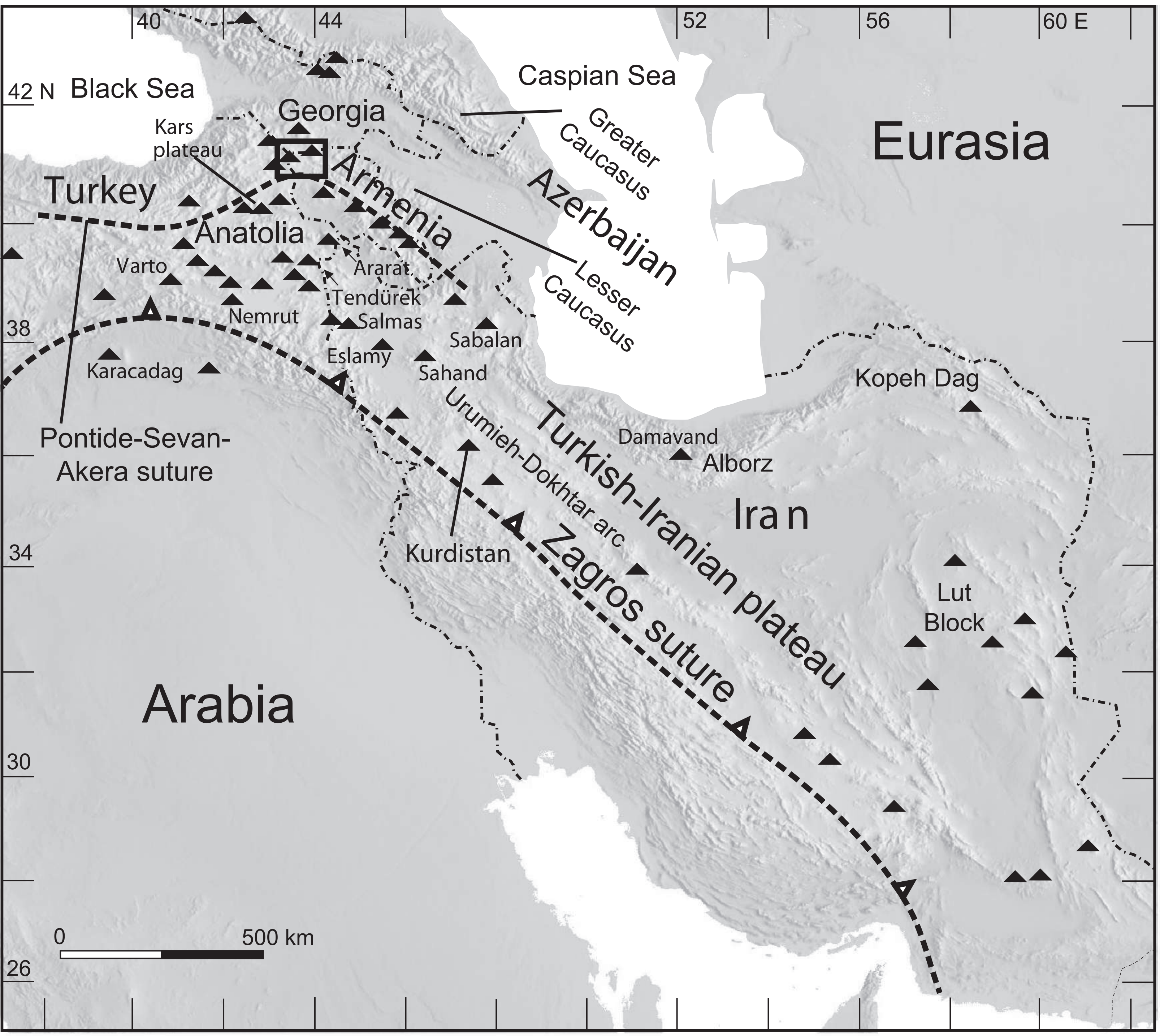
Figure 10. Zr/Hf vs. Nb/Ta plot for Shirak lavas and selected mafic Pliocene-Quaternary lavas from the Arabia-Eurasia collision. Ararat and Tendürek are from lithospheric mantle sources, Karacadağ in southern Turkey has an OIB source. Sources: ~~Eocene arc rocks – Keskin et al. (2008);~~ Tendürek, Ararat – Kheirkhah et al. (2009); Karacadağ - Sen et al.

(2004), Lustrino et al. (2010); clinopyroxene fractionation based on partition coefficients of Pagé et al. (2009); other references as per Figure 9.~~.~~

Figure 11. Non-modal batch partial melting models for the valley series lavas using depleted MORB mantle (Workman and Hart, 2005) and incompatible element enriched oceanic plateau (Fitton and Godard, 2004) sources. Source modes: spinel lherzolite - ol = 0.578, opx = 0.27, cpx = 0.119, sp = 0.033; garnet lherzolite - 0.598, 0.211, 0.076, gnt = 0.115. Melt modes: spinel lherzolite - 0.1, 0.27, 0.5, sp = 0.13; garnet lherzolite - 0.05, 0.2, 0.3, gnt = 0.45. Partition coefficients are from the GERM Partition Coefficient Database (http://earthref.org/KDD).

Figure 12. A schematic cross-section through the present-day Arabia-Eurasia collision zone highlighting potential processes involved in Pliocene-Quaternary magmatism. Hatchings represent regions of partial melting. Crustal thicknesses estimated from Zor et al. (2008).

Figure 1



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Turkey
Armenia
Azerbaijan
Greater
Caucasus
Lesser
Caucasus
Eurasia
Anatolia
Varto
Ararat
Tendürek
Nemrut
Salmas
Sabalan
Eslamy
Sahand
Karacadag
Kopeh Dag
Pontide-Sevan-
Akera suture
Urumieh-Dokhtar arc
Turkish-Iranian plateau
Damavand
Alborz
Iran
Kurdistan
Zagros suture
Lut
Block
Arabia
0
500 km

Figure 2
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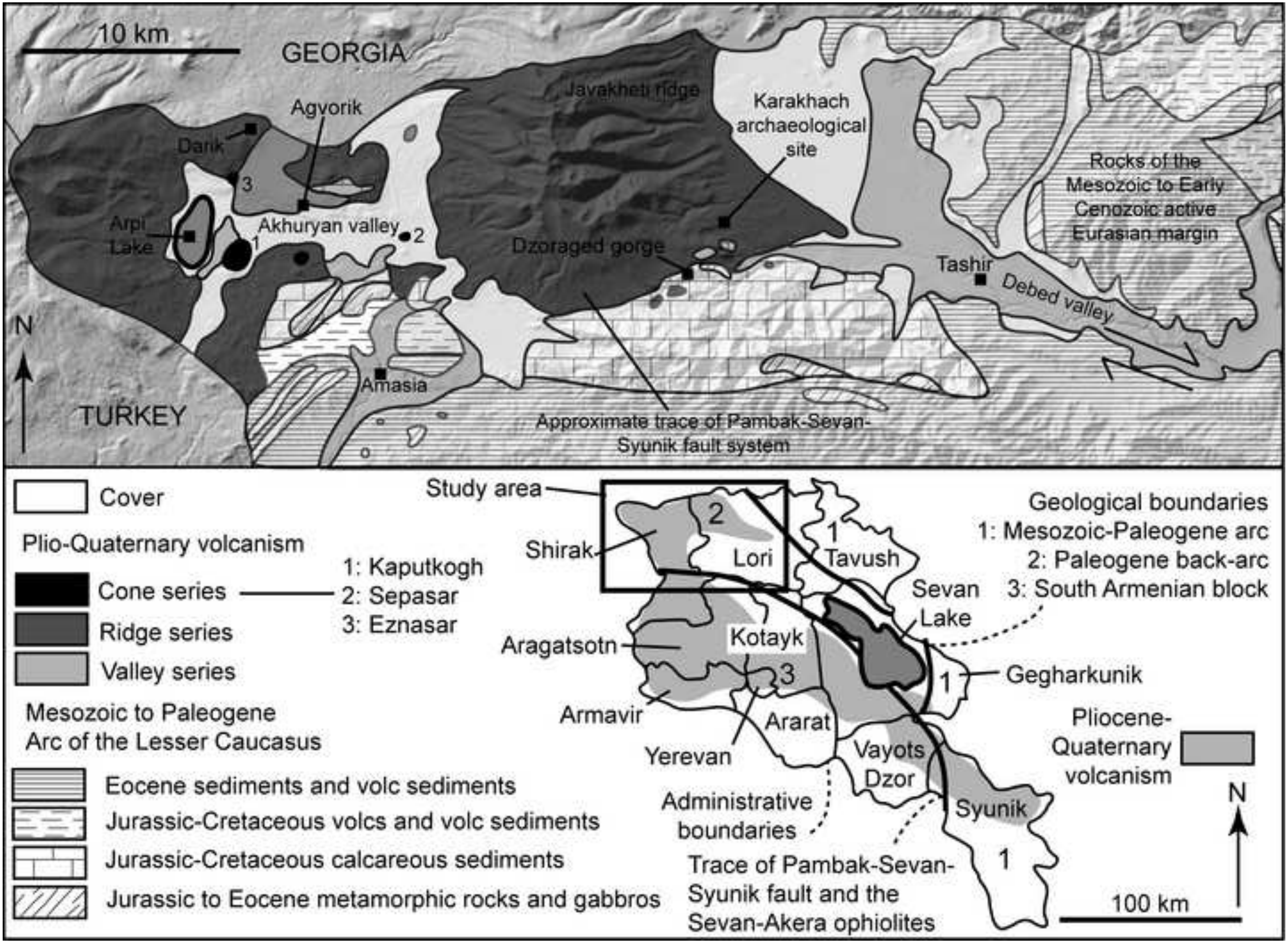


Figure 3

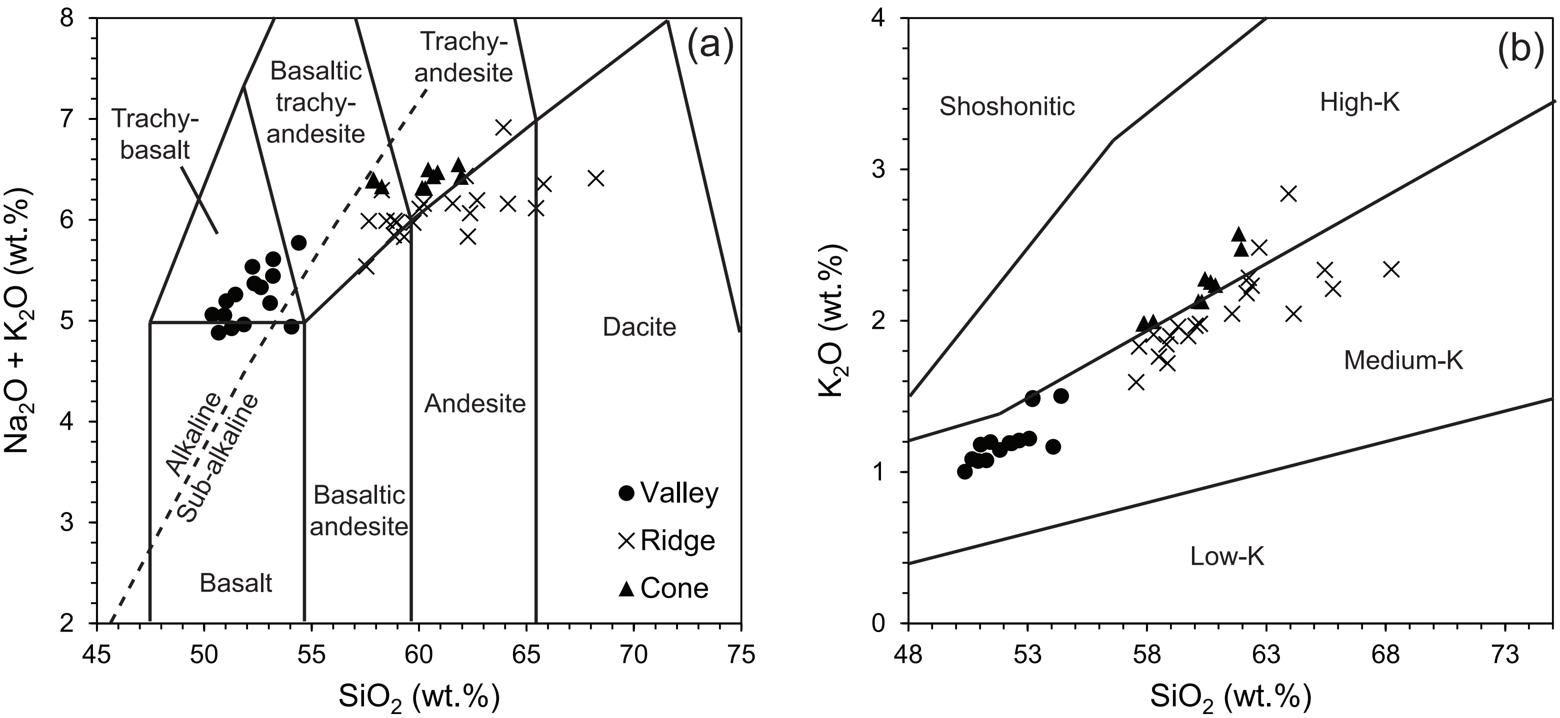
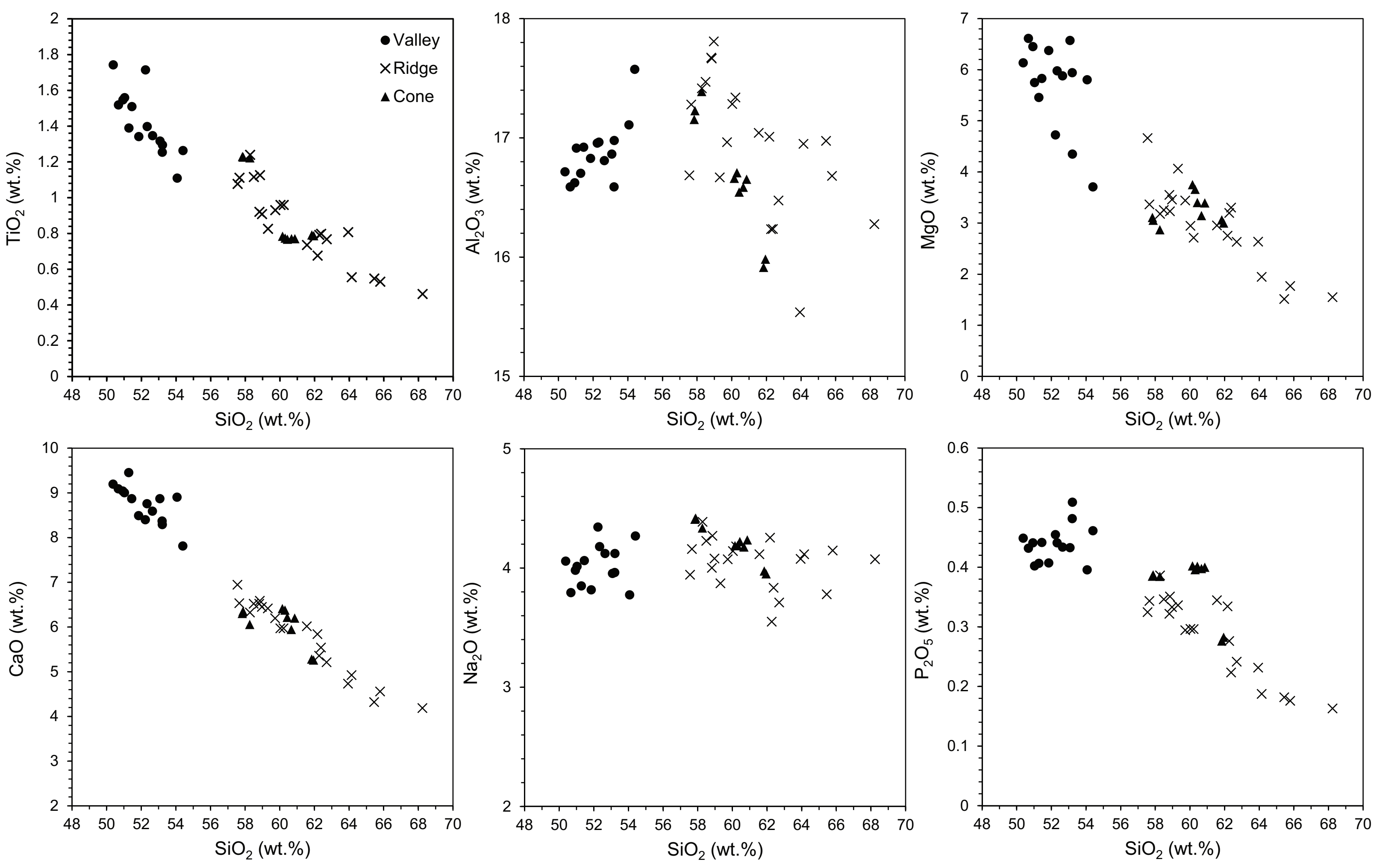

(a)
Trachy-
basalt
Basaltic
trachy-
andesite
Trachy-
andesite
Dacite
Andesite
Basaltic
andesite
Basalt
Alkaline
Sub-alkaline
Valley
Ridge
Cone
$Na_2O + K_2O$ (wt.%)
SiO_2 (wt.%)
(b)
Shoshonitic
High-K
Medium-K
Low-K
K_2O (wt.%)
SiO_2 (wt.%)

Figure 4



Valley
Ridge
Cone
TiO2 (wt.%)
Al2O3 (wt.%)
MgO (wt.%)
CaO (wt.%)
Na2O (wt.%)
P2O5 (wt.%)
SiO2 (wt.%)

Figure 5

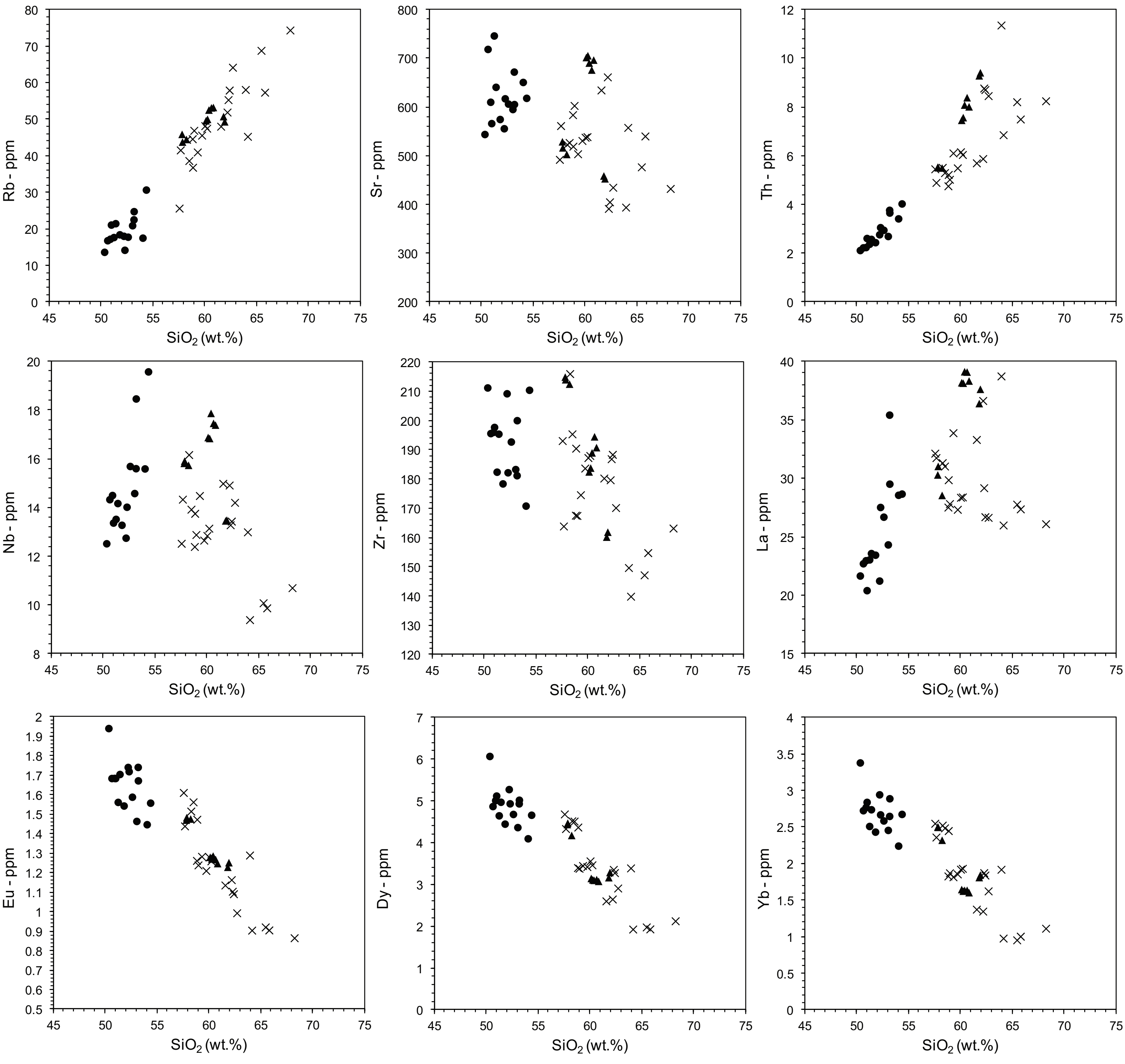

Rb - ppm
Sr - ppm
Th - ppm
Nb - ppm
Zr - ppm
La - ppm
Eu - ppm
Dy - ppm
Yb - ppm
SiO2 (wt.%)

Figure 6

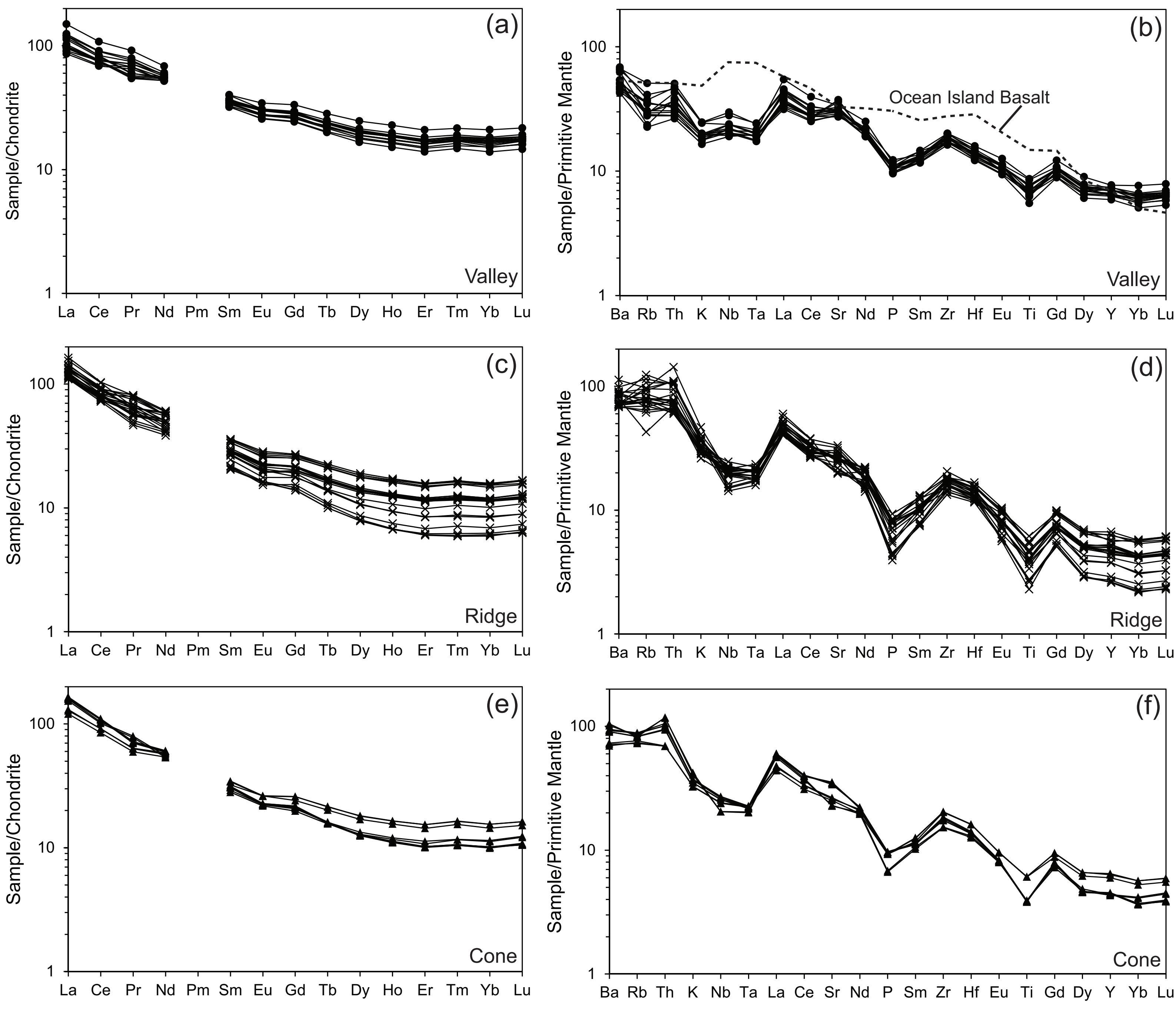
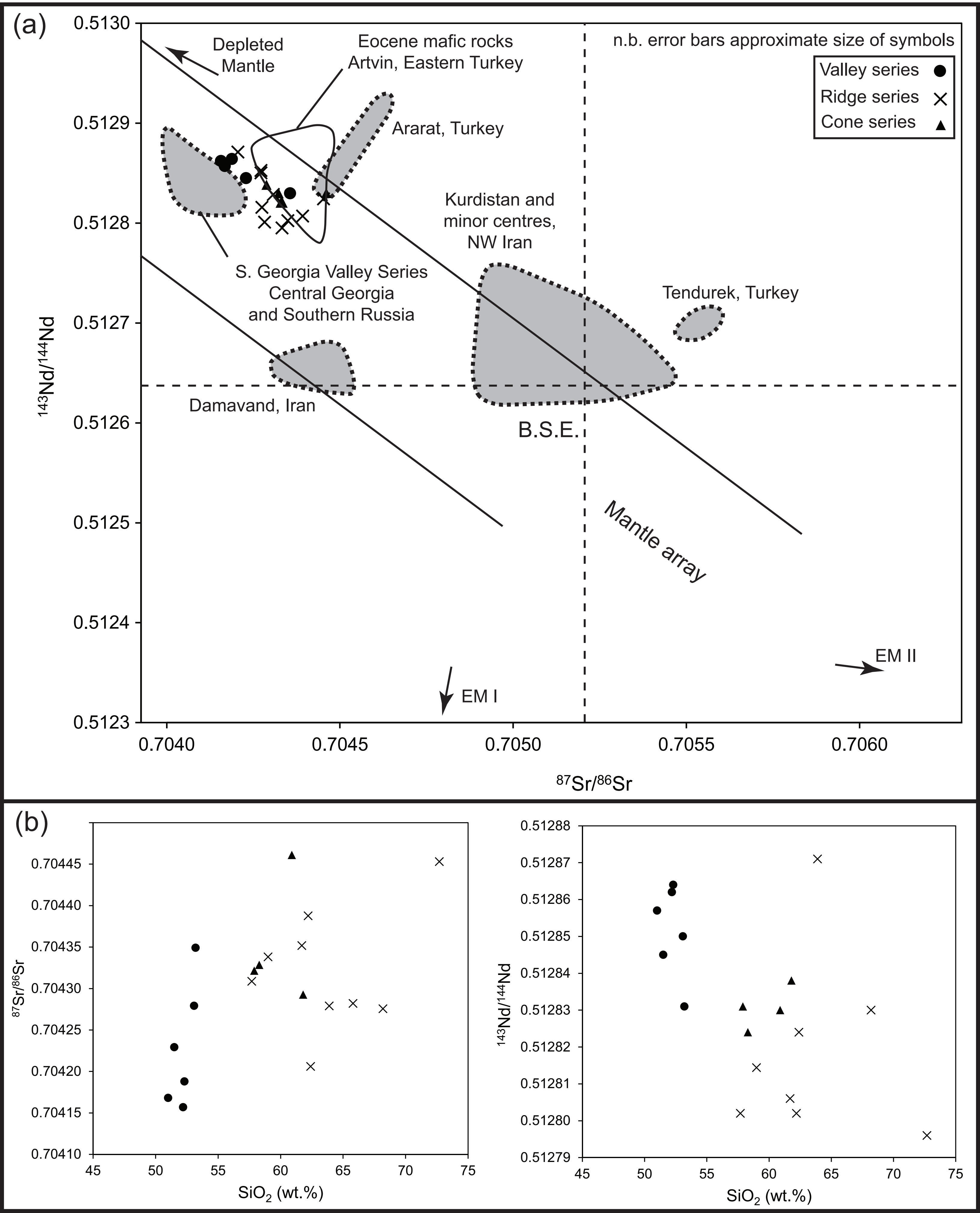
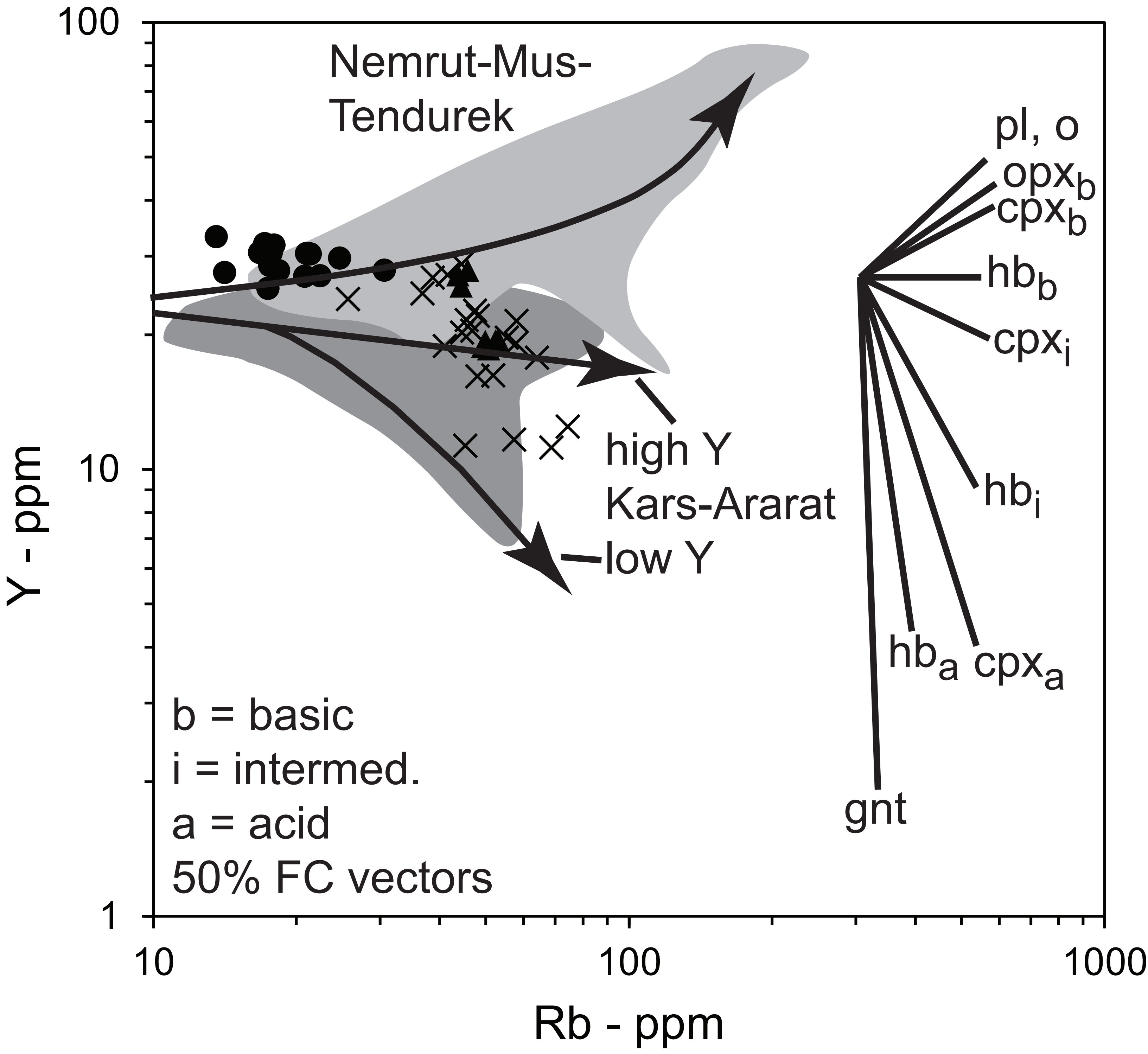

(a)
Sample/Chondrite
Valley
La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
(b)
Sample/Primitive Mantle
Ocean Island Basalt
Valley
Ba Rb Th K Nb Ta La Ce Sr Nd P Sm Zr Hf Eu Ti Gd Dy Y Yb Lu
(c)
Sample/Chondrite
Ridge
La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
(d)
Sample/Primitive Mantle
Ridge
Ba Rb Th K Nb Ta La Ce Sr Nd P Sm Zr Hf Eu Ti Gd Dy Y Yb Lu
(e)
Sample/Chondrite
Cone
La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
(f)
Sample/Primitive Mantle
Cone
Ba Rb Th K Nb Ta La Ce Sr Nd P Sm Zr Hf Eu Ti Gd Dy Y Yb Lu
1
10
100

Figure 7



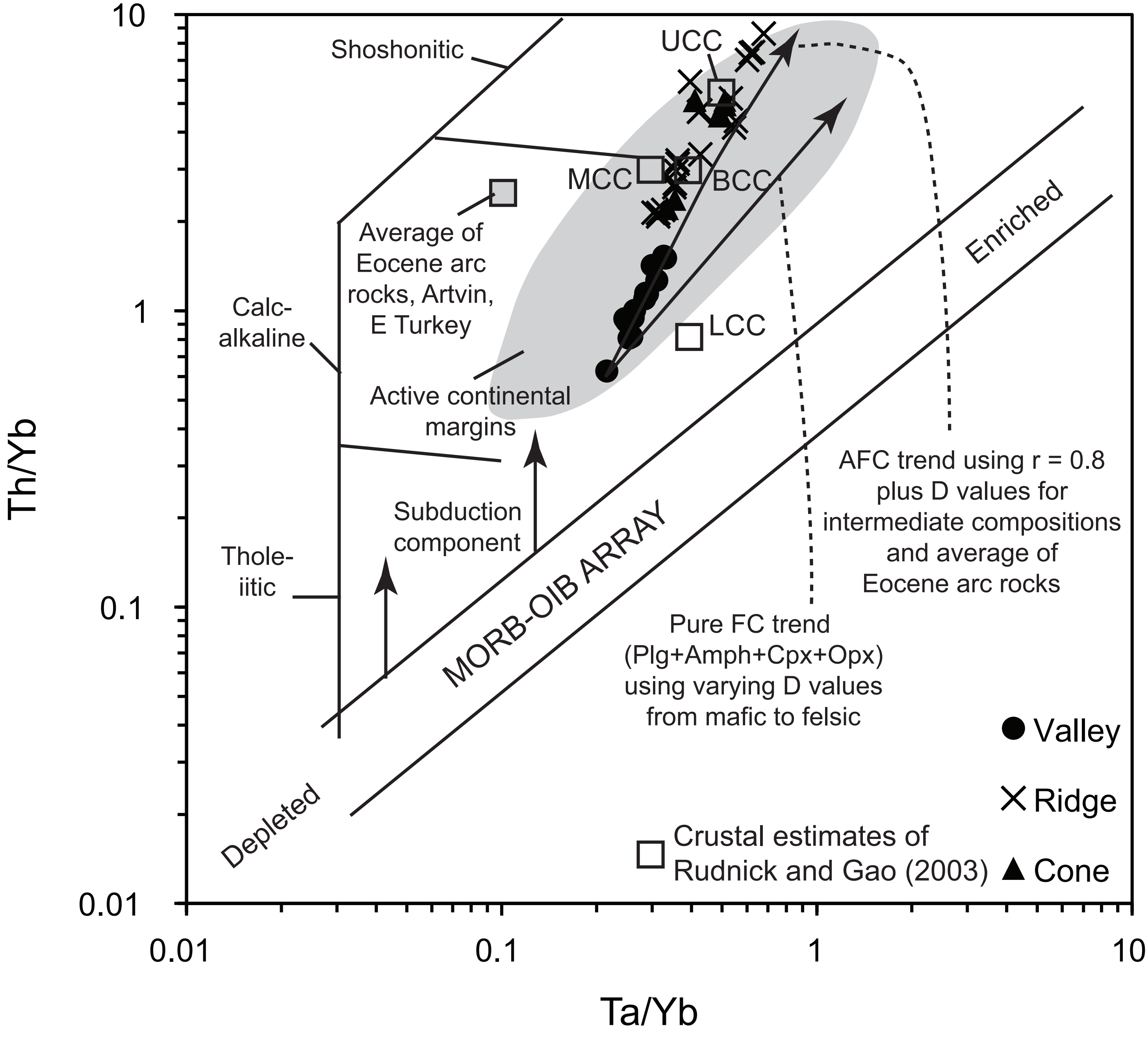
(a)
Depleted Mantle
Eocene mafic rocks Artvin, Eastern Turkey
n.b. error bars approximate size of symbols
Valley series
Ridge series
Cone series
Ararat, Turkey
Kurdistan and minor centres, NW Iran
S. Georgia Valley Series Central Georgia and Southern Russia
Tendurek, Turkey
Damavand, Iran
B.S.E.
Mantle array
EM I
EM II
$^{143}Nd/^{144}Nd$
$^{87}Sr/^{86}Sr$
0.5130
0.5129
0.5128
0.5127
0.5126
0.5125
0.5124
0.5123
0.7040
0.7045
0.7050
0.7055
0.7060
(b)
$^{87}Sr/^{86}Sr$
0.70445
0.70440
0.70435
0.70430
0.70425
0.70420
0.70415
0.70410
$^{143}Nd/^{144}Nd$
0.51288
0.51287
0.51286
0.51285
0.51284
0.51283
0.51282
0.51281
0.51280
0.51279
45
50
55
60
65
70
75
SiO_2 (wt.%)

Figure 8



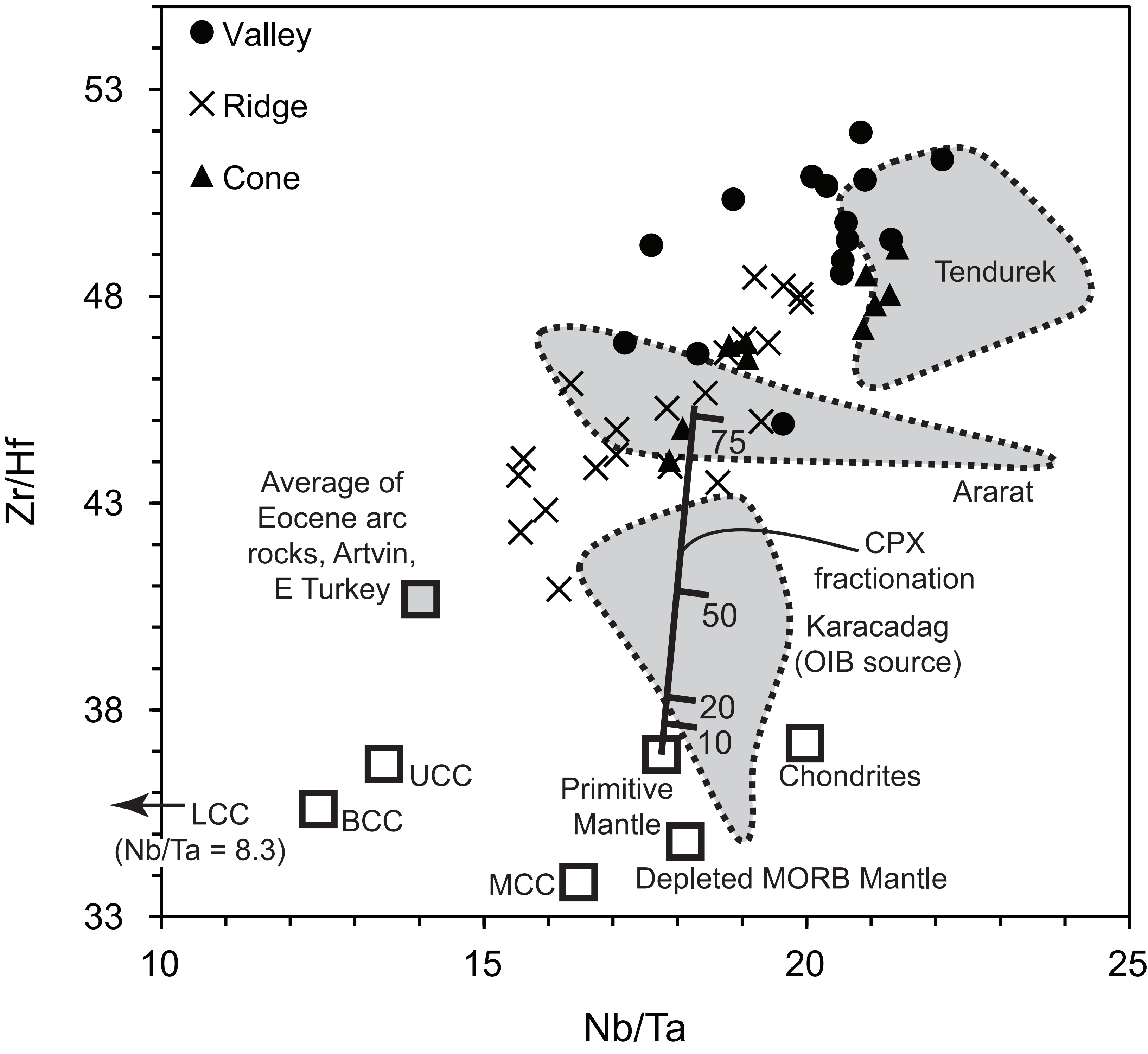
Nemrut-Mus-
Tendurek
high Y
Kars-Ararat
low Y
pl, o
opx b
cpx b
hb b
cpx i
hb i
hb a
cpx a
gnt
b = basic
i = intermed.
a = acid
50% FC vectors
100
10
1
10
100
1000
Y - ppm
Rb - ppm

Figure 9



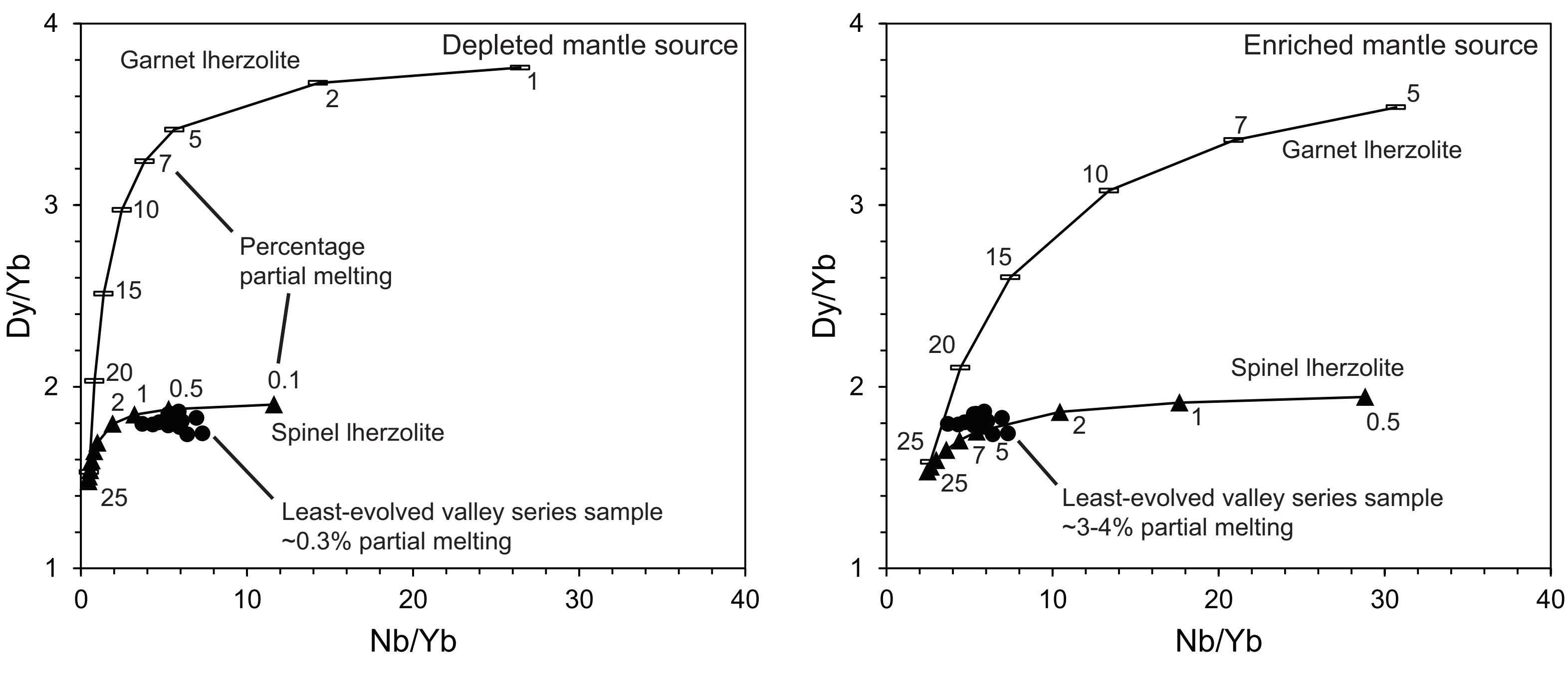
10
1
0.1
0.01
0.01
0.1
1
10
Th/Yb
Ta/Yb
Shoshonitic
UCC
MCC
BCC
LCC
Average of
Eocene arc
rocks, Artvin,
E Turkey
Calc-
alkaline
Active continental
margins
Subduction
component
Thole-
iitic
MORB-OIB ARRAY
Enriched
Depleted
AFC trend using r = 0.8
plus D values for
intermediate compositions
and average of
Eocene arc rocks
Pure FC trend
(Plg+Amph+Cpx+Opx)
using varying D values
from mafic to felsic
Valley
Ridge
Cone
Crustal estimates of
Rudnick and Gao (2003)

Figure 10



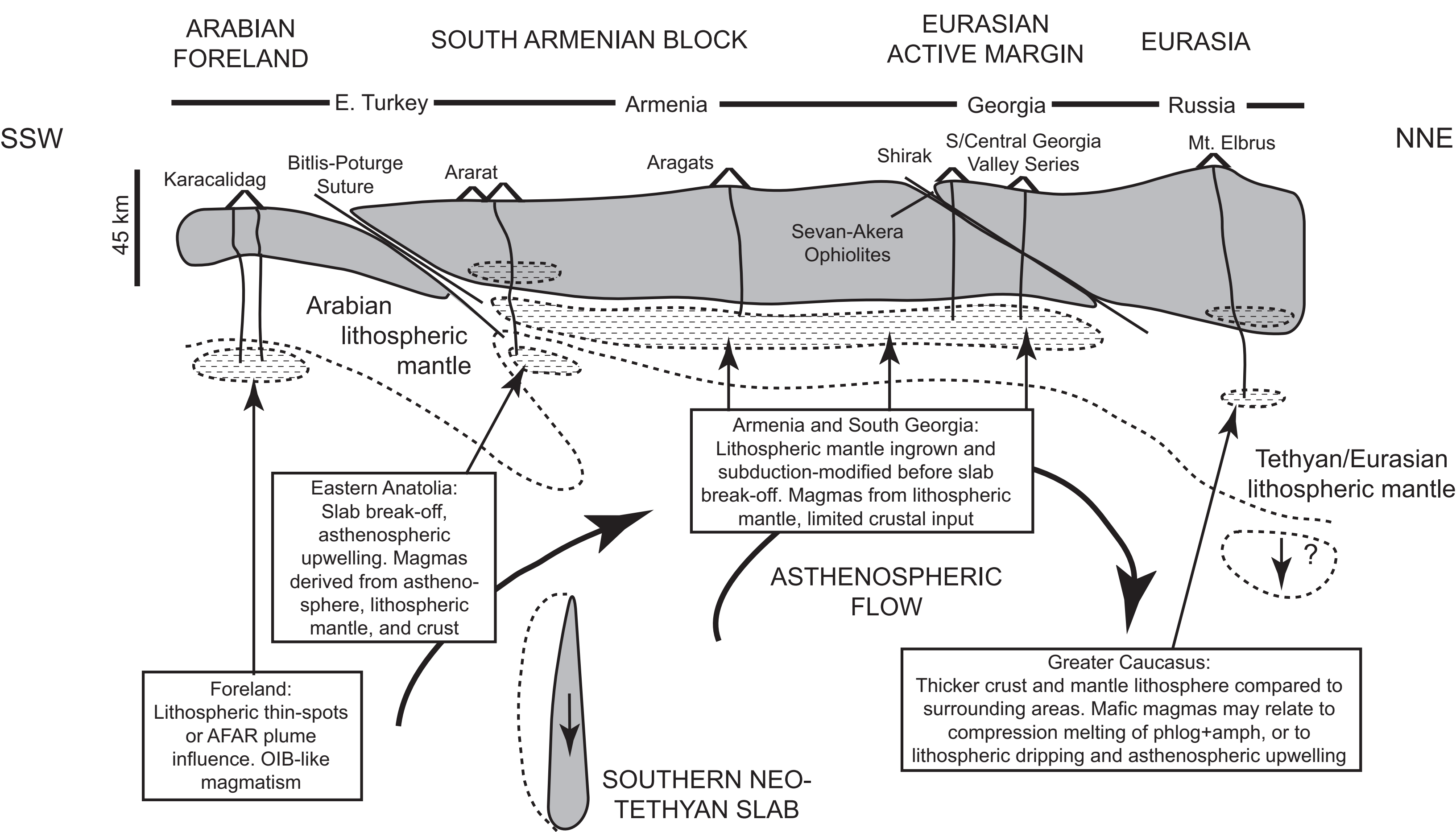
Valley
Ridge
Cone
Zr/Hf
Nb/Ta
53
48
43
38
33
10
15
20
25
Tendurek
Ararat
75
50
20
10
CPX
fractionation
Karacadag
(OIB source)
Average of
Eocene arc
rocks, Artvin,
E Turkey
UCC
LCC
(Nb/Ta = 8.3)
BCC
MCC
Primitive
Mantle
Chondrites
Depleted MORB Mantle

Figure 11



Depleted mantle source
Garnet lherzolite
Percentage
partial melting
Spinel lherzolite
Least-evolved valley series sample
~0.3% partial melting
Dy/Yb
Nb/Yb
Enriched mantle source
Garnet lherzolite
Spinel lherzolite
Least-evolved valley series sample
~3-4% partial melting

Figure 12



ARABIAN
FORELAND
SOUTH ARMENIAN BLOCK
EURASIAN
ACTIVE MARGIN
EURASIA
E. Turkey
Armenia
Georgia
Russia
SSW
NNE
Karacalidag
Bitlis-Poturge
Suture
Ararat
Aragats
Shirak
S/Central Georgia
Valley Series
Mt. Elbrus
45 km
Sevan-Akera
Ophiolites
Arabian
lithospheric
mantle
Armenia and South Georgia:
Lithospheric mantle ingrown and
subduction-modified before slab
break-off. Magmas from lithospheric
mantle, limited crustal input
Tethyan/Eurasian
lithospheric mantle
Eastern Anatolia:
Slab break-off,
asthenospheric
upwelling. Magmas
derived from astheno-
sphere, lithospheric
mantle, and crust
ASTHENOSPHERIC
FLOW
?
Foreland:
Lithospheric thin-spots
or AFAR plume
influence. OIB-like
magmatism
Greater Caucasus:
Thicker crust and mantle lithosphere compared to
surrounding areas. Mafic magmas may relate to
compression melting of phlog+amph, or to
lithospheric dripping and asthenospheric upwelling
SOUTHERN NEO-
TETHYAN SLAB

Table 1. Major and trace element data for selected samples from the valley (v), ridge (r), and cone (c) series, Shirak.

Number	S14.4	S14.5	S19.1	S25.2	S26.2	S28.1	S29.1	S4.2	S5.1	S6.1	S9.2	S21.1	S23.1	S30.2	S30.3	S1.1	S2.1	S7.3	S20.2
Series	V	V	V	V	V	V	V	R	R	R	R	R	R	R	R	C	C	C	C
SiO_2	50.68	53.21	52.33	53.08	52.24	51.45	51.03	57.67	51.57	52.18	65.80	63.94	62.39	58.97	68.23	57.88	58.27	60.68	61.84
TiO_2	1.52	1.25	1.40	1.32	1.71	1.51	1.56	1.11	0.73	0.68	0.53	0.81	0.80	0.91	0.46	1.23	1.22	0.77	0.79
Al_2O_3	16.59	16.59	16.96	16.86	16.96	16.92	16.91	17.28	17.04	17.01	16.68	15.54	16.23	17.81	16.28	17.23	17.39	16.65	15.91
Fe_2O_3(T)	10.20	9.03	9.27	9.58	10.43	10.16	10.52	7.35	5.56	5.25	4.07	5.40	5.55	6.63	3.56	7.76	7.85	5.82	5.70
MnO	0.16	0.15	0.15	0.15	0.16	0.18	0.17	0.12	0.09	0.09	0.07	0.10	0.09	0.11	0.06	0.11	0.10	0.10	0.10
MgO	6.61	5.94	5.98	6.57	4.72	5.83	5.75	3.36	2.95	2.75	1.77	2.63	3.30	3.46	1.55	3.05	2.87	3.39	3.06
CaO	9.09	8.37	8.75	8.87	8.40	8.87	9.00	6.53	6.02	5.84	4.56	4.73	5.54	6.45	4.19	6.35	6.06	6.20	5.28
Na_2O	3.79	3.96	4.12	3.95	4.34	4.06	4.01	4.16	4.11	4.25	4.15	4.07	3.83	4.08	4.07	4.42	4.33	4.24	3.97
K_2O	1.09	1.48	1.19	1.22	1.19	1.20	1.18	1.83	2.05	2.18	2.21	2.84	2.23	1.90	2.34	1.94	2.00	2.24	2.58
P_2O_5	0.43	0.48	0.44	0.43	0.46	0.44	0.40	0.34	0.35	0.33	0.18	0.23	0.22	0.33	0.16	0.39	0.39	0.40	0.28
LOI	0.73	-0.30	-0.14	-0.23	-0.09	0.05	-0.14	-0.07	0.45	-0.06	0.63	0.17	0.74	0.09	0.00	-0.06	0.11	-0.11	0.84
Total	100.89	100.16	100.52	101.81	100.56	100.67	100.40	99.68	100.92	100.50	100.64	100.47	100.94	100.74	100.91	100.34	100.58	100.55	100.37
Sc	15.6	23.1	19.3	20.3	21.7	22.1	22.2	bd	bd	bd	bd	bd	12.7	15.0	bd	bd	bd	bd	11.5
V	167	179	164	169	188	185	184	131	103	100	71	85	105	142	68	147	140	112	101
Cr	154	162	162	175	131	153	129	bd	45	44	bd	38	72	bd	bd	bd	bd	73	47
Co	37.6	33.0	36.5	36.5	35.0	41.4	42.2	22.9	16.1	15.3	9.8	18.9	19.0	23.1	9.5	20.6	20.3	18.1	17.6
Ni	106.8	113.8	113.5	125.6	65.4	106.3	101.5	bd	16.6	bd	bd	47.2	52.1	32.9	bd	bd	bd	39.2	36.7
Rb	16.7	22.5	14.1	20.8	17.9	21.4	21.0	41.6	48.1	51.9	57.4	58.1	57.9	46.9	74.4	43.7	44.4	53.2	50.7
Sr	718	671	617	595	555	640	566	562	635	662	540	395	405	603	433	516	503	696	458
Y	30.5	27.1	27.6	27.0	31.7	30.3	30.4	27.2	16.1	16.2	11.7	21.5	19.2	20.4	12.5	27.2	25.7	19.2	18.6
Zr	195.5	181.1	182.1	183.2	209.1	195.3	197.6	163.9	180.3	179.7	154.8	149.7	188.4	167.5	163.2	213.9	212.4	190.7	160.1
Nb	14.3	15.6	14.0	14.6	12.7	14.2	13.4	14.3	15.0	14.9	9.9	13.0	13.4	12.9	10.7	15.9	15.7	17.4	13.5
Ba	329	452	356	340	288	334	312	449	556	599	589	742	471	491	545	474	460	609	670
Hf	3.9	4.0	3.9	3.5	4.3	3.8	3.9	3.7	3.8	3.8	3.5	3.3	4.3	3.6	3.7	4.6	4.6	3.9	3.6
Ta	0.7	0.8	0.8	0.7	0.7	0.7	0.7	0.7	0.8	0.8	0.6	0.8	0.8	0.7	0.7	0.8	0.8	0.8	0.7
Pb	4.6	6.6	5.5	5.1	4.3	4.6	4.7	8.0	9.0	9.6	11.6	10.6	8.9	8.3	11.1	8.7	8.0	11.8	9.9
Th	2.2	3.8	3.1	2.7	2.8	2.6	2.6	4.9	5.7	5.9	7.5	11.4	8.7	5.0	8.3	5.5	5.5	8.0	9.3
U	0.7	0.9	0.3	0.8	0.3	0.8	0.6	1.2	1.3	1.4	1.9	2.0	2.2	1.3	2.2	1.5	1.4	1.8	1.7
La	22.7	35.4	27.5	24.3	21.2	23.6	20.4	31.8	33.3	36.6	27.4	38.7	26.7	27.8	26.1	31.0	28.5	38.3	26.4
Ce	45.6	66.2	51.1	47.1	43.0	47.0	42.1	54.6	58.1	62.7	46.3	63.6	48.2	50.9	46.0	55.8	52.1	66.3	62.0
Pr	5.2	8.5	6.9	6.3	6.3	6.6	6.0	5.9	5.7	6.1	4.5	7.5	5.9	6.4	5.5	5.9	5.5	6.6	7.2
Nd	24.7	31.3	26.8	23.7	25.1	26.0	23.9	26.6	23.3	25.2	18.6	25.4	20.7	22.7	18.7	26.7	24.7	27.7	24.5
Sm	5.3	5.9	5.3	4.7	5.4	5.4	5.2	5.0	3.9	4.2	3.1	4.3	3.9	4.2	3.2	5.1	4.8	4.5	4.2
Eu	1.7	1.7	1.7	1.5	1.7	1.7	1.7	1.4	1.1	1.2	0.9	1.3	1.1	1.2	0.9	1.5	1.5	1.3	1.2
Gd	5.6	5.8	5.6	4.9	5.9	5.7	5.7	5.0	3.6	3.8	2.8	4.2	3.9	4.3	3.1	5.2	4.8	4.3	3.9
Tb	0.8	0.9	0.9	0.7	0.9	0.9	0.9	0.8	0.5	0.5	0.4	0.6	0.6	0.6	0.4	0.8	0.7	0.6	0.6
Dy	4.9	4.9	4.9	4.3	5.3	5.0	5.1	4.3	2.6	2.7	1.9	3.4	3.3	3.4	2.1	4.5	4.2	3.1	3.2
Ho	1.0	1.0	1.0	0.9	1.1	1.0	1.0	0.9	0.5	0.5	0.4	0.7	0.7	0.7	0.4	0.9	0.9	0.6	0.6
Er	2.7	2.7	2.7	2.4	2.9	2.7	2.8	2.4	1.4	1.4	1.0	0.9	1.8	1.8	1.1	2.5	2.3	1.6	1.7
Tm	0.4	0.4	0.4	0.4	0.5	0.4	0.5	0.4	0.2	0.2	0.2	0.3	0.3	0.3	0.2	0.4	0.4	0.3	0.3
Yb	2.7	2.6	2.7	2.5	2.9	2.7	2.8	2.4	1.4	1.4	1.0	1.9	1.8	1.9	1.1	2.5	2.3	1.6	1.8
Lu	0.4	0.4	0.4	0.4	0.4	0.4	0.5	0.4	0.2	0.2	0.2	0.3	0.3	0.3	0.2	0.4	0.4	0.3	0.3

LOI = loss on ignition; b.d. = below detection

Table 2
Click here to download Table: Neilletal Table2.docx

Table 2. Measured Nd and Sr isotope compositions of the valley, ridge, and cone series, Shirak.

	$^{143}Nd/^{144}Nd$	± 1σ	εNd	± 1σ	$^{87}Sr/^{86}Sr$	± 1σ
Valley series						
S14.5	0.512831	0.000004	+3.76	0.08	0.704349	0.000009
S19.1	0.512864	0.000005	+4.41	0.10	0.704188	0.000007
S25.2	0.512850	0.000006	+4.14	0.12	0.704279	0.000007
S26.2	0.512862	0.000006	+4.37	0.12	0.704157	0.000008
S28.1	0.512845	0.000005	+4.04	0.10	0.704229	0.000008
S29.1	0.512857	0.000005	+4.27	0.10	0.704168	0.000007
Ridge series						
S4.2	0.512830	0.000007	+3.75	0.14	0.704309	0.000007
S5.1	0.512802	0.000007	+3.20	0.14	0.704352	0.000008
S6.1	0.512806	0.000005	+3.28	0.10	0.704388	0.000010
S9.2	0.512802	0.000005	+3.20	0.09	0.704282	0.000008
S21.1	0.512848	0.000004	+4.10	0.08	0.704279	0.000006
S23.1	0512871	0.000005	+4.55	0.10	0.704206	0.000007
S30.2	0.512796	0.000005	+3.08	0.10	0.704338	0.000010
S30.3	0.512814	0.000005	+3.44	0.10	0.704276	0.000007
Cone series						
S1.1	0.512831	0.000005	+3.76	0.10	0.704321	0.000007
S2.1	0.512824	0.000005	+3.63	0.10	0.704329	0.000007
S7.3	0.512830	0.000003	+3.75	0.06	0.704461	0.000007
S20.2	0.512838	0.000004	+3.90	0.09	0.704293	0.000007

Supplementary Item 2

Click here to download Background dataset for online publication only: Neilletal Shirak SUP 2.xlsx