Magmatic constraints on geodynamic models of subduction in the East Carpathians, Romania

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Abstract

The East Carpathian volcanic arc is the youngest region of calc-alkaline magmatic activity in Eastern Europe. A general age progression of the onset and cessation of magmatic activity occurs along the East Carpathian arc from older volcanic structures (ca. 12 Ma) in the NW to the youngest (<1 Ma) in the SE. Magmatism continued into the Plio-Pleistocene, significantly later than the end of basin closure and the onset of continental collision along the Inner Carpathian arc that is thought to have taken place during the Miocene (9–5 Ma). Migration of magmatic activity from NW to SE along the arc may be explained by a corresponding migration of the magma-generating zone at mantle depths. Major and trace element characteristics of the erupted products are typical of subduction-related magmas and suggest an input of fluids from a dehydrating subducting slab into their mantle source region. Subduction of a narrow oceanic basin is considered to be the most probable cause of the East Carpathian magmatism and its migration. As thick continental crust began to enter the northern part of the trench at around 9 Ma, slab breakoff began although subduction of the detached slab continued at depth. As breakoff progressed from north to south, a rupture or tear propagated along the slab, causing termination of volcanism as the slab sank out of the magma-generation zone. Breakoff of the slab occurred at progressively shallower levels, southward along the arc, causing the volume of erupted arc magmas to diminish. Some unusual geological features at the southern end of the volcanic arc (e.g. contemporaneous eruption of alkaline and calc-alkaline magmas; extreme enrichment in K and other large ion lithophile elements in the arc magmas) may be accounted for by asthenospheric mantle upwelling into the void left behind by the broken slab and increased efficiency of dehydration of the remnants of the slab under the higher thermal regime. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: East Carpathians; subduction; magmatism; slab breakoff; slab window

1. Introduction

Previous studies of the evolution of the Carpatho-Pannonian region have indicated that a large basin, probably floored by oceanic crust, lay in the area of the present-day Carpathians and was consumed by subduction during Tertiary times (Burchfiel, 1976; Csontos, 1995). Collision was accompanied by the formation of large thrust belts composed of flysch sediments (Sândulescu, 1984) and the eruption of large volumes of calc-alkaline magma along the northern and eastern edges of the Al-
Fig. 1. Map showing main tectonic units in the east of the Carpatho-Pannonian region. Calc-alkaline magmatism along the East Carpathian arc is divided into the following areas: Oas = Oas Mountains; Gut = Guti Mountains; Tib = Tibles Mountains; Tga = Toroiaga Mountains; Rod = Rodna Mountains; Barg = Bargau Mountains; Cal = Calimani Mountains; Gur = Gurghiu Mountains; NH = North Harghita Mountains; SI = South Harghita Mountains. Alkaline magmatism occurs in the eastern Transylvanian Basin (ETB). Inset map shows the location of the Tisza-Dacia (TDM) and Alcapa (AM) microplates.

capa and Tisza-Dacia microplates (Szabó et al., 1992).

The East Carpathian arc lies along the eastern rim of the Tisza–Dacia microplate, to the east of the Transylvanian Basin (Fig. 1; Seghedi et al., 1995). The arc includes a 160-km-long chain of calc-alkaline volcanoes, which form the Calimani, Gurghiu and Harghita Mountains (herein referred to as the CGH arc). Calc-alkaline magmatism occurred from late Miocene to Quaternary times and is the youngest arc volcanism in the Carpatho-Pannonian region (Pécskay et al., 1995a). Magmatic activity migrated from NW to SE along the strike of the arc in this area and was accompanied by a general decrease in the volume of magma erupted within each volcanic centre (Szakács and Seghedi, 1995). Small volumes of mafic alkaline magmas were erupted close to the southern end of the CGH volcanic arc in the eastern Transylvanian Basin (ETB) and were contemporaneous with the final stages of calc-alkaline activity in the CGH arc (Seghedi and Szakács, 1994; Downes et al., 1995). The cessation of active volcanism in this area during the Quaternary coincided with the end of magmatic activity in the region as a whole. Seismic activity continues to the present day in the Vrancea Zone, south east of the CGH arc, although no magmatism is associated with this active tectonism. Earthquakes in the Vrancea Zone define a steeply dipping Benioff zone, which gives an indication of the geometry of the final stages of subduction (Oncescu et al., 1984).

The dynamics of Neogene–Quaternary subduction remain poorly constrained for the East Carpathian region. The most notable problems are:
(a) the relationship between subduction and volcanism, (b) the nature of the subducted lithosphere, (c) the timing and nature of subduction, and (d) the role (if any) of slab breakoff following the cessation of plate convergence. Significantly, the arc magmatism continued long after the main period of convergence related to subduction had ceased.

This paper will synthesise geochemical and geodynamic data to establish constraints on the processes responsible for formation and migration of the calc-alkaline magmatism in the East Carpathians during the Neogene and Quaternary. Our rigorous investigation of magmatism in the East Carpathian arc yields a greater understanding of the style and timing of subduction over the last 10-15 Ma. We believe that the time-gap between cessation of convergence and the magmatic activity is a consequence of subduction of a narrow ocean basin. Furthermore, we consider that the contemporaneity of the calc-alkaline arc volcanism and alkaline magmatism is a result of asthenospheric mantle upwelling into a slab window, as slab breakoff progressed and the eruption of high-K and shoshonitic magmas at the southern end of the arc is due to an increased contribution of fluid from the dehydrating slab.

2. Geological and geophysical evidence for subduction

Palaeomagnetic, sedimentological and geophysical studies indicate that collision took place between NE Europe and the Intra-Carpathian area during Cenozoic times (Royden and Báldi, 1988; Ziegler, 1988; Royden and Burchfiel, 1989; Csontos et al., 1992; Kovács et al., 1994; Tari, 1994; Csontos, 1995). The geodynamic driving forces behind the movement of microplates in the region are thought to be slab-pull and slab rollback along the Carpathian subduction zone (Royden, 1993) and lateral extrusion caused by continuous convergence in the Alps (Ratschbacher et al., 1991). The main Neogene tectonic events in the East Carpathians are summarised in Fig. 2. Here we attempt (a) to constrain the timing and rate of Tertiary subduction and (b) to estimate the extent and nature of the subducted lithosphere.

Palaeomagnetic data indicate that major rotations of the Alcapa and Tisza–Dacia microplates took place in the Late Palaeogene to Middle Miocene during the main period of Tertiary convergence (Balla, 1987; Márton et al., 1992; Patrascu et al., 1994; Márton and Fodor, 1995). A 40° clockwise rotation of the Tisza–Dacia microplate was accompanied by
closure of a basin which lay to the north and east of the Tisza–Dacia microplate and was followed by relatively minor crustal shortening and continental collision (Lillie et al., 1994). In the Late Miocene (9–5.3 Ma) an E–W compressional event occurred across the entire Carpatho-Pannonian region (Perresson and Decker, 1997), which has been interpreted as reflecting the arrival of thick and buoyant continental crust at the trench and its resistance to subduction. The youngest E-directed thrusts in the East Carpathians terminated at approximately 7.1 Ma (Perresson and Decker, 1997). Plio-Quaternary (3–0.5 Ma) deformation was less intense and limited to the extreme SE Carpathians (Hippolyte and Sandulescu, 1996).

The width of the basin that was subducted during the Neogene can be estimated from the reconstruction of balanced cross-sections for the East Carpathian Neogene flysch sediments (Roure et al., 1993; Roca et al., 1995). For a section taken through the East Carpathians, a total of 130 km of shortening can be divided into 108 km Mid-Miocene (~15 Ma) subduction-related episode and 22 km during Plio-Quaternary post-collisional deformation. Assuming that the rate of basin closure is closely related to rate of plate convergence, a convergence rate of approximately 2.5 cm yr⁻¹ was estimated from Ukrainian Miocene flysch sections by Roca et al. (1995). A similar rate may be envisaged for the convergence rate across the CGH arc but a slowdown may have occurred towards the end of subduction and onset of collision as continental crust arrived at the trench.

The Vrancea zone at the SE end of the East Carpathian arc (Fig. 1) is the site of Neotectonic intermediate and shallow focus seismic activity (Roman, 1970; Horváth, 1988; Trifu and Radulian, 1991; Radulian and Popa, 1996). A slab of sinking lithosphere appears to dip at 58° to the NW (Oncescu et al., 1984). Earthquake epicentres occur down to a depth of 220 km and are separated from shallow crustal epicentres by a seismic gap between 40 and 60 km that may represent a zone of reduced viscosity where breakoff of the slab has taken place (Fuchs et al., 1979). The shallow depth of the gap is typical for breakoff under very slow (<0.2 cm yr⁻¹) plate convergence velocities (Davies and von Blankenburg, 1995). Such slow convergence velocities may be appropriate for the end of subduction in the CGH arc. The sinking lithosphere may represent a final relic of the plate that was subducted beneath the East Carpathians, now detached from the remainder of the subducted oceanic crust to the north, which has sunk into the deeper mantle.

Balla (1987), Csontos et al. (1992) and many other authors have proposed that SW- and W-directed subduction took place along the whole Carpathian arc during Miocene times. However, an alternative model proposed by Linzer (1996) suggests that the subducted basin was attached to the Moesian platform and was subducted to the NW rather than being attached to the East European Platform and subducted to the west. Linzer (1996) also suggested that the slab underwent extensive roll-back to the SE and that the lithospheric slab now imaged in the Vrancea tectonic zone is the subducted oceanic slab that caused the magmatism of the CGH arc.

The subducted basin may have been floored by oceanic, transitional or continental crust (Burchfiel, 1976; Csontos et al., 1992). Many previous studies have supported the concept of subduction of oceanic lithosphere in the East Carpathians (Rădulescu and Sandulescu, 1973; Balla, 1987; Sandulescu, 1988; Royden and Burchfiel, 1989; Csontos et al., 1992; Linzer, 1996). Oceanic subduction is suggested by a lack of major crustal thickening, minor post-collisional shortening of less than 50 km, an absence of metamorphism in the accretionary flysch sediments and a well preserved foredeep filled with fine-grained pelagic sediments. Subduction of oceanic crust is required to explain the lack of significant collisional features between the Tisza–Dacia microplate and the European foreland, such as those seen in the Eastern Alps (Royden, 1993). Thinned continental crust may have entered the system in the later stages of subduction during the Miocene which slowed down and eventually halted the convergence in late Miocene times (Bleahu et al., 1973; Csontos et al., 1992; Decker and Peresson, 1996; Perresson and Decker, 1997). Continental crust clearly entered the subduction zone to the NW as underthrust continental crust has been imaged by deep seismic surveys in the West Carpathians (Tomek, 1993; Tomek and Hall, 1993).

The postulated oceanic crust may have formed the basement to large flysch basins that were open in the Outer Carpathian area during the Late Cretaceous and that are now represented by the thrustsed flysch sediments of the paleo-accretionary wedge (Fig. 2;
Table 1

Isotopic characteristics of potential magmatic sources and crustal contaminants in the East Carpathian area

<table>
<thead>
<tr>
<th></th>
<th>Parental CGH arc (Calimani–N. Harghita)</th>
<th>Parental southern CGH arc (S. Harghita)</th>
<th>ETB alkaline magmas</th>
<th>ETB mantle xenoliths</th>
<th>Tisza-Dacia crystalline upper crust</th>
<th>Outer Carpathian Cretaceous and Neogene flysch sediments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{87}\text{Sr}/^{86}\text{Sr}$</td>
<td>0.7050–0.7058</td>
<td>0.7045–0.7050</td>
<td>0.7037–0.7045</td>
<td>0.7019–0.7044</td>
<td>0.7094–0.7362</td>
<td>0.7078–0.7501</td>
</tr>
<tr>
<td>$^{143}\text{Nd}/^{144}\text{Nd}$</td>
<td>0.51280–0.51265</td>
<td>0.51275–0.51255</td>
<td>0.51285–0.51273</td>
<td>0.51355–0.51271</td>
<td>0.51235–0.51196</td>
<td>0.51226–0.51213</td>
</tr>
<tr>
<td>$^{206}\text{Pb}/^{204}\text{Pb}$</td>
<td>18.75–18.85</td>
<td>18.55–18.65</td>
<td>18.60–18.68</td>
<td>15.63–15.66</td>
<td>15.44–15.63</td>
<td>15.65–15.69</td>
</tr>
<tr>
<td>$\delta^{18}\text{O}$</td>
<td>&lt;6‰</td>
<td>&lt;7‰</td>
<td>&lt;6‰</td>
<td>&lt;6‰</td>
<td>&lt;13‰</td>
<td>&gt;13‰</td>
</tr>
</tbody>
</table>

Data sources are: CGH arc magmas: Mason et al. (1996); ETB alkaline magmas: Downes et al. (1995); ETB mantle xenoliths (Vaselli et al., 1995; Rosenbaum et al., 1997); Tisza-Dacia crystalline upper crust and Outer Carpathian Cretaceous and Neogene flysch sediments (Mason et al., 1996). $\delta^{18}\text{O}$ data for ETB alkaline magmas and ETB mantle xenoliths is from Mason (1995).

Winkler and Slaczka, 1992; Roca et al., 1995). These basins served as a zone of weakened lithosphere into which the Alcapa and Tisza–Dacia microplates could indent during the Miocene collision. This area, prior to deformation, has been estimated to be up to 500 km across (Csontos, 1995). However, direct evidence for the existence of oceanic crust in this region during the Neogene is limited (Pană and Erdmer, 1996). Ophiolitic thrust slices (up to several km across) have been identified in parts of the outer Carpathian flysch but their detailed origin is not fully understood (Sândulescu, 1988) and some have been interpreted as intraplate basalts (Russo–Sândulescu and Bratosin, 1985). The sediments that form the Outer Carpathian flysch may be similar to the sedimentary veneer that could have been subducted with the downgoing oceanic lithosphere. Fluids derived from these sediments may have added to the metasomatising fluids derived from the prograde metamorphism of the subducting oceanic crust and would carry a distinctive trace element and isotopic signature. The geochemical characteristics of these representative flysch sediments have been previously determined (Mason et al., 1996) and isotopic data are given in Table 1.

3. Magmatic geochronology

Three stages of Neogene magmatic activity have been recognised in the eastern part of the Carpatho-Pannonian region (Szabó et al., 1992; Pécskay et al., 1995a). Acidic tuffs and ignimbrites were initially erupted in the northern part of the Transylvanian Basin during a period of mainly acidic volcanism in the Early Miocene. This was followed by Middle Miocene to Pliocene, mainly intermediate, calc-alkaline strato-volcanism along the East Carpathian arc, including the Calimani, Gurghiu and Harghita areas (CGH arc). Finally, Plio-Pleistocene alkaline magmas were erupted behind the arc in the eastern Transylvanian Basin (ETB). Szabó et al. (1992) considered that subduction controlled both the initial burst of acidic volcanism and the main period of calc-alkaline volcanism, whereas the alkaline magmas were a result of the upwelling of asthenospheric mantle in response to lithospheric extension across the Pannonian Basin.

The main period of extrusive calc-alkaline volcanic activity in the CGH arc took place between the Late Miocene and the Pleistocene (~12 Ma to ~0.2 Ma; Fig. 3), following Mid-Miocene extrusive activity in the Oas and Gutii Mountains to the north (Peltz et al., 1985; Pécskay et al., 1995b). Previous studies have described a general age progression in calc-alkaline magmas along the entire Inner Carpathian arc, from older structures in the north and west to younger volcanics in the south east (Póka, 1988; Szabó et al., 1992). However, a recent compilation of internally consistent K–Ar age data for the Neogene Carpathian magmatism suggests that the age progression is only significant in the CGH segment of the East Carpathian arc (Pécskay et al., 1995a, Szakács and Seghedi, 1996), where the rate of migration was approximately 18 km Ma⁻¹ (equivalent to ~2 cm year⁻¹).

Magmatism started in the northern Calimani area at approximately 12 Ma with the emplacement of in-
Fig. 3. K–Ar geochronology of magmatic activity in the East Carpathian arc and Transylvanian Basin. (a) Approximate age span of magmatism in each segment of the arc, after Pécskay et al. (1995a). Notation for arc magmatism as in Fig. 1. (b) Age vs. distance for calc-alkaline magmas in the CGH arc. Ages are taken from Pécskay et al. (1995b). Distances along the arc axis are measured from the most northerly area of calc-alkaline volcanism in the Calimani Mountains. The curve drawn through the data is a third-order polynomial regression line.

Intermediate intrusions followed by eruption of small volumes of dacitic magma (Fig. 3). Activity in Calimani continued until the mid–late Pontian (6.8 Ma), resulting in the build up of a large volcanic edifice. Immediately to the south, in the northern part of the Gurghiu Mountains, volcanic activity was partly contemporaneous with that in Calimani but continued later into the Pontian. Other Gurghiu volcanic centres are quite closely related to one another in time and their products overlap each other in a complex mix of lava flows and volcaniclastics. The final activity in the Gurghiu centres occurred in the Dacian, significantly later than the termination of magmatism in Calimani.
Volcanism in the Harghita area shows a well-defined north–south age progression (Fig. 3). At the Pontian/Dacian boundary (6–5.5 Ma) all of the centres in North Harghita were erupting contemporaneously. However, activity in each volcanic centre generally ceased at a progressively later date from north to south. Therefore, the focus of magmatism continued to progress southwards with time. The timing of the volcanism in the South Harghita area shows the most distinct north–south geographical progression, from late Dacian to Quaternary (Szakács et al., 1993). Lavas and intrusions within each volcanic centre have a narrow age range (typically ±1 Ma) reflecting a rapid southward migration of magmatic activity. The youngest K/Ar age reported is 0.20 Ma and 14C dating carried out on charcoal fragments from pyroclastic deposits in the same volcanic centre yielded ages of 10,700 ± 180 yr B.P. (Juvigne et al., 1994) and 35,000–42,000 yr B.P. (Moriya et al., 1996). Alkaline magmas were erupted in the eastern Transylvanian Basin between 2.5 and 0.7 Ma (Seghedi and Szakács, 1994; Downes et al., 1995), only 40 km to the west of the main CGH arc where calc-alkaline magmas were being erupted contemporaneously.

4. Magma geochemistry

4.1. Identification of mantle sources

Neogene–Quaternary magmatism in the East Carpathian arc consists of medium-K calc-alkaline to high-K calc-alkaline and shoshonitic compositions (Seghedi et al., 1987; Mason, 1995; Mason et al., 1996) whose major element geochemistries are typical of these classifications (Mason et al., 1995). In this section, we will use isotope data (Table 1) to constrain the mantle source of these and the closely-related alkaline magmas, as different sources (e.g. deep asthenosphere, fluid-metasomatised mantle wedge, shallow mantle lithosphere) have different isotopic characteristics.

Most of the calc-alkaline magmas in the CGH arc show isotopic evidence of having assimilated large amounts of continental crustal material as they passed through the lithosphere (Mason et al., 1996). The magmas have δ18O values that are much higher than those of normal mantle-derived magmas, and they show offsets to high 87Sr/86Sr, low 143Nd/144Nd and radiogenic Pb isotope ratios (Fig. 4). Possible crustal contaminants include crystalline metamorphic basement rocks and terrigenous flysch sediments for which the isotopic (Table 1) and trace element characteristics have been previously determined (Mason et al., 1996). This crustal contamination masks the source characteristics of the calc-alkaline magmas and can cause difficulties in geodynamic interpretations. To some extent, the effects of crustal assimilation can be filtered out using radiogenic (Sr–Nd–Pb) and stable (δ18O) isotope signatures to identify those calc-alkaline magmas that have been least affected (Mason et al., 1996). However, it is impossible to rule out the effects of assimilation completely, as most of the magmas are also highly fractionated, and primitive basic magmas (i.e. with MgO > 6%; high Ni and high Cr) are absent from the arc suite.

The most basic and least crustally contaminated magmas in the Calimani, Gurghiu and Northern Harghita parts of the arc are rare basalts and basaltic andesites which have homogeneous Sr–Nd–Pb isotopic compositions (Table 1; Fig. 4) which probably reflect a common mantle source composition. This is considered to be the metasomatised mantle wedge situated above the subducting slab. However, significant isotopic differences are seen in the least contaminated basaltic andesites from the South Harghita area (Fig. 4) which have lower 87Sr/86Sr and 206Pb/204Pb ratios (Table 1). These differences most probably reflect variations in the isotopic composition of the mantle source (Mason et al., 1996).

Downes et al. (1995) showed that the contemporaneous ETB alkaline intraplate magmas erupted near to the South Harghita area show geochemical similarities to the compositions of ocean island basalts (OIB). This alkaline magmatism is thought to be derived directly from the asthenospheric mantle beneath the East Carpathians and is similar to that seen in other parts of the Carpatho-Pannonian region (Embery-Isztin et al., 1993). Crustal contamination occurred to a much smaller degree in the alkaline basaltic andesites which have lower 87Sr/86Sr and 206Pb/204Pb ratios (Table 1). These differences most probably reflect variations in the isotopic composition of the mantle source (Mason et al., 1996).
Fig. 4. Identification of the main isotopic components in the East Carpathian area. (a) Sr vs. Nd isotope diagram showing data for the ETB mantle xenoliths (Vaselli et al., 1995), ETB alkaline magmas (Downes et al., 1995), CGH calc-alkaline magmas, upper crust and flysch sediments (Mason et al., 1996). (b) Pb- isotope diagram using data from the above sources with the addition of ETB mantle xenolith data (Rosenbaum et al., 1997). The northern hemisphere reference line (NHRL) of Hart (1984) is shown for illustration.
related to asthenospheric mantle upwelling of the type which also forms OIB (Hoernle et al., 1995). They have mantle-like δ18O (Dobosi et al., 1998 in press), unradiogenic Sr and Pb isotope ratios and high 143Nd/144Nd (Downes et al., 1995; Rosenbaum et al., 1997; Table 1). Their isotopic compositions reflect that of the asthenosphere, another possible mantle source. However, compared with a global average of OIB, the alkaline magmas have slightly higher large ion lithophile element (LILE) and lower high field strength element (HFSE) concentrations, features that resemble slightly the arc magmas and suggest that the source of the ETB magmas may have been slightly contaminated by slab-derived fluids (Downes et al., 1995). It is significant that the ETB alkali basalts and the primitive arc magmas from the South Harghita area are isotopically quite similar, and may therefore share a common asthenospheric mantle source component (Fig. 1).

Another possible mantle source for the CGH arc magmatism is the shallow lithospheric mantle, which has been investigated via the abundant ultramafic xenoliths in the ETB alkaline magmas (Vaselli et al., 1995). The xenoliths are cut by amphibole veins that were formed by infiltration metasomatism by alkaline magmas that were derived from a subduction-modified mantle source (Rosenbaum et al., 1997). However, the majority of the mantle xenoliths are highly depleted. The xenoliths are unlikely to represent the composition of the lithospheric mantle directly above the subducted slab (Vaselli et al., 1995), but their composition suggests that the bulk of the mantle lithosphere was not metasomatized by subduction-related fluids. The peridotitic xenoliths are characteristic of the depleted mantle in their Sr, Nd and Pb isotope ratios (Table 1; Fig. 4; Vaselli et al., 1995; Rosenbaum et al., 1997) and rare earth element patterns and their isotopic ratios do not overlap those of the CGH primitive arc magmas. Thus we consider that the lithospheric mantle did not contribute to magmatogenesis as it is too depleted to generate either the calc-alkaline or alkaline magmas.

4.2. Variations in trace element geochemistry of calc-alkaline magmas along the CGH arc

Groups of incompatible trace elements behave differently depending on their ionic radius and charge. Large ion lithophile elements (LILE) such as Rb and Ba are soluble in water-rich fluids (e.g. fluids formed by dehydration of a subducting slab). High field strength elements (HFSE) such as Zr and Nb are strongly depleted in subduction-related magmas but are enriched in asthenosphere-derived OIB. The rare earth elements (REE) generally behave coherently but certain minerals have a preference for individual REE — e.g. plagioclase takes up Eu²⁺ and garnet takes up the heavy REE (Dy to Lu). Thus trace elements can be used to fingerprint different processes which have occurred in magmatogenesis.

Significant variations in incompatible element characteristics are observed in calc-alkaline magmas erupted along the strike of the CGH arc from the older centres in the NW to the youngest volcanic centres in the SE. Crustal contamination is very widespread throughout the calc-alkaline magmas and there are very few uncontaminated samples in only a limited number of volcanic centres (Mason et al., 1996). Because of this, we cannot use the database that has been screened isotopically to minimise the effects of crustal contamination to investigate along-arc variations in the mantle source of the magmas. Consequently some of the variation in incompatible trace element ratios within and between the CGH volcanic centres must be due to crustal contamination.

All CGH volcanic rocks have high LILE/HFSE ratios (e.g. K/Zr, Ba/Zr, Ba/Nb and Th/Zr) and typically closely resemble subduction-related magma suites from elsewhere in the world, e.g. the Aegean, the Aeolian arc and the Andes (Fig. 5). Samples from the Calimani area show a large range in LILE/HFSE that spans the variation for much of the arc (Fig. 6a) due to the combined effect of crustal assimilation and fractional crystallisation (Mason et al., 1996). In the volcanics of the Gurghiu arc segment, LILE/HFSE ratios are closer to those of normal mid-ocean ridge basalt (N-MORB) and they increase to values which are higher than normal island arc compositions in the South Harghita area. Crustal contamination alone cannot account for this strong LILE-enrichment in the South Harghita volcanics as LILE/HFSE ratios in the volcanics are higher than those of the analysed potential crustal contaminants. LILE/HFSE ratios may also have been affected by the addition of sediment to the mantle source through subduction.
It is not possible to produce such an increase in LILE/HFSE if subducted sedimentary material had the composition of sediments that are present in the Cretaceous and Neogene flysch sediments (Fig. 6a; Mason et al., 1996). However, fluids derived from dehydration of such sediments would be even more enriched in LILE and can account for the observed increase. Therefore we conclude that the extreme LILE/HFSE ratios are due to an increase in contribution of the fluid flux from the subducting slab.

LILE/LILE ratios such as K/Rb, Rb/Ba (Fig. 6b) are sensitive both to variations in small degrees of partial melting and to variations in the composition of metasomatizing slab-derived fluids or contaminating crust (Ellam and Hawkesworth, 1988). Extreme amounts of fractional crystallisation are required to produce significant variations in for example K/Rb, Rb/Ba and Ba/Th ratios. The partition coefficient for Rb is an order of magnitude lower than that for Ba in the source region of most arcs (Ellam and Hawkesworth, 1988). Smaller degrees of melting of a relatively Rb-poor mantle source would increase Rb/Ba (Ellam and Hawkesworth, 1988) but in the South Harghita samples, K/Rb increases slightly but Rb/Ba is reduced (Fig. 6b), which suggests that the unusual chemistry is not controlled by melt extraction processes. The slight deviations in the LILE/LILE ratios of the South Harghita magmas from those in the rest of the CGH arc are probably due to an enrichment in K and Ba. This may have been introduced by a greater flux of LILE in fluids from the subducting slab or by assimilation of LILE-rich crust. However, Ba and K concentrations are not exceptionally high in potential crustal contaminants and therefore the LILE variation is most likely due to a greater influx of fluids from the slab during the final stages of magmatism.

The LILE/LREE ratios Ba/La, Sr/Nd and Pb/Nd exhibit a spectacular increase in the South Harghita magmas (e.g. Ba/La: Fig. 6c), where they are several times higher in the youngest volcanic centre than in magmas from the Gurghiu and North Harghita. Ba/La and Pb/Nd increase slightly from the Gurghiu volcanics to the North Harghita and then rapidly in the South Harghita centres. Sr/Nd is quite consistent for most of the CGH arc and increases only in the extreme south. LILE/LREE ratios in most of the arc magmas overlap the fields of potential local crustal contaminants and the flysch sediments, but Ba/La and Sr/Nd ratios in the South Harghita volcanics are higher than those found in the local crust. This also supports the explanation that these anomalous features are due to an increase in slab-derived fluid flux in the mantle source.

Variations in HFSE/HFSE (e.g. Zr/Nb; Fig. 6d) in the CGH volcanics are small but significant. A steady decrease in Zr/Nb southwards along the CGH arc is a reflection of an increase in Nb content of the
Fig. 6. (a) Variation in the LILE/HFSE ratio Ba/Zr between CGH arc volcanic centres. Data, taken from Mason (1995) and Mason et al. (1996), are plotted at the axis position of each volcanic centre relative to the most northerly part of the Calimani Mountains. Lines within each diagram cover the total variation within each volcano and filled squares show the value for samples with 57% SiO₂, the composition of the most typical andesites. A comparison is made with the local upper crustal metasediments (UCM) and local Cretaceous and Neogene terrigenous flysch sediments (FS) (Mason et al., 1996), average N-MORB and average ocean island basalt (OIB) (Sun and McDonough, 1989). (b) Variation in LILE/LILE ratios between CGH arc volcanic centres. Data sources and legend as in (a). (c) Variation in the LILE/LREE ratio Ba/La, and (d) the HFSE/HFSE ratio Zr/Nb between CGH arc volcanic centres. Data sources and legend as in (a).
magnas. This cannot be due to crustal contamination as most crustal rocks have low Nb contents and the Zr/Nb ratios of the most likely contaminants in the local crystalline basement or the flysch sediments are higher than those of the South Harghita magnas. Typical asthenosphere-derived ocean island basalts (OIB) have low Zr/Nb (Sun and McDonough, 1989; Fig. 6d) and the low ratios seen in South Harghita magnas may reflect a component from a more OIB-like asthenospheric source. The South Harghita calc-alkaline magnas with lower Zr/Nb ratios are spatially and temporally close to the ETB alkaline magmatism. Thus, in addition to an increased fluid flux, upwelling of an OIB-source asthenospheric mantle component appears to have affected the magmatism at the southern end of the CGH arc.

Although incompatible trace element variations are frequently decoupled from radiogenic isotope characteristics in oceanic and continental arcs (e.g. Kay, 1980), some correlations exist between radiogenic isotopes and certain key trace element ratios in the CGH arc, suggesting control by a common process. Variations to lower $^{206}\text{Pb}/^{204}\text{Pb}$ correlate with increasing LILE/HFSE ratios, decreasing Zr/Nb and increasing Ba/La (Fig. 7) along the arc from NW to SE. This is not accompanied by any systematic variation in $\delta^{18}\text{O}$ (Fig. 7), ruling out crustal contamination as a controlling process for the trace element variation. Increasing LILE/HFSE and LILE/LREE ratios in the South Harghita magnas reflect the increase in LILE which are highly mobile in fluids. Sr and Pb (members of the LILE group) would have distinctive isotopic characteristics within infiltrating fluids derived from the subducting slab and hence fluid metasomatism of the mantle source may control $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios.

The ratio of LREE to HREE varies from mildly LREE-enriched for the bulk of the CGH arc rocks (basalts and andesites with Ce/Yb$_N$ = 2–6) to highly LREE-enriched for some of the Southern Harghita rocks (high-K andesites with Ce/Yb$_N$ = 19–39; Fig. 8a). Although both crustal contamination and fractional crystallisation could increase Ce/Yb$_N$, they cannot account for LREE-enrichment in the South Harghita magnas, as potential contaminants are not sufficiently enriched in LREE (Fig. 8b) and the volcanics have negligible Eu anomalies (Eu/Eu* ~1), precluding extensive plagioclase fractionation in crustal magma chambers. Increases in LREE/HREE in South Harghita magnas closely mirror the increases in LILE/HREE (e.g. Ba/Yb and Sr/Y), suggesting that the differences in trace element ratios are controlled by the influence of the same process, i.e. an increase in subduction zone fluids.

However, South Harghita magnas not only have higher LREE concentrations, but also have lower

![Fig. 7. Variation of the LILE/LREE ratio Ba/La with $^{206}\text{Pb}/^{204}\text{Pb}$ in samples from the south of the CGH arc. $\delta^{18}\text{O}$ determined on separated mineral phenocrysts (Mason et al., 1996) is shown for each sample from the Southern Harghita area.](image-url)
HREE concentrations than all other CGH magmas. The HREE in the South Harghita magmas are also lower than in the potential crustal contaminants and are lower than in the ETB alkaline magmas (Fig. 8). Low HREE may be due to retention of the HREE in a mineral such as garnet during melting (Peccerillo and Taylor, 1976). Garnet is not usually present in either the mantle or the subducting slab at the depths at which melting occurs in subduction zones, although garnet may occur when the thermal regime is hot enough for eclogite to be formed in the slab (Defant and Drummond, 1990). However, this requires unusual geodynamic conditions such as the subduction of hot, young (<5 Ma) oceanic crust (Defant and Drummond, 1990) or extremely high (>100 MPa) shear stresses in the subducting crust (Peacock et al., 1994). Such conditions are not thought to have been present in the East Carpathians, where the subducted oceanic crust was probably old and cold (as indicated by the thick flysch sediments which were deposited on it), and where shear stress is estimated to have been only 40 MPa (Demetrescu and Andreescu, 1994).

In summary, we have identified three processes that have influenced the magma of the East Carpathian arc: crustal contamination, metasomatism of the mantle wedge by fluid driven off the subducting slab, and an OIB-related asthenospheric mantle upwelling. All magmas in the CGH arc show evidence of the first two processes to varying degrees, however the South Harghita magmas have an anomalous geochemistry compared with the rest of the arc. Their Unusual Pb–Sr–Nd isotope ratios are similar to those of the ETB alkaline magmas and this is closely associated with a drop in Zr/Nb towards values more typical of alkaline (OIB-like) melts. The LILE and LREE enrichment suggests a stronger influx of fluids from the subducted slab, which also affected Pb-isotope ratios, whereas the HREE-depletion implies the presence of garnet in the source.

5. Discussion

5.1. Relationship between subduction and volcanism

Previous studies of the chronology of thrusting in the East Carpathian Flysch zone (e.g. Sandulescu, 1988; Roure et al., 1993) and palaeomagnetic data for the Carpatho-Pannonian region (Balla, 1987; Patrascu et al., 1994) show that there is a significant time-lag between the main period of Neogene shortening (17–11 Ma) when consumption of the proposed oceanic crust must have occurred and the
episode of calc-alkaline magmatism in the CGH arc (12–0.5 Ma). Thus it is necessary to explain the onset and continuation of magma generation long after the main period of subduction at the surface had passed.

Subduction-related magmatism originates by dehydration of the subducting lithospheric slab at depths between 80 and 120 km (Gill, 1981; Sekine and Wyllie, 1982), causing fluids to flux into the overlying mantle wedge. The geochemical data discussed above lead us to propose that the mafic precursors to the CGH calc-alkaline arc magmas were produced by partial melting of subduction-metasomatized mantle immediately above the postulated subducting slab. The chemical and isotopic composition of the mantle source was quite uniform except in the southern extremity of the CGH arc where a greater degree of fluid metasomatism may have occurred, accompanied by the possible infiltration of more OIB-like asthenospheric melts.

We propose that a narrow lithospheric slab sank through the 80–120 km deep magma-generating zone into the asthenospheric mantle during Neogene convergence along the East Carpathian arc. As the slab passed through this ‘window’, the mantle wedge was metasomatized by slab-derived LILE-rich fluids and melted to produce the parental magmas to the calc-alkaline lavas. Eruptive products may not have appeared at the surface until 1–2 Ma after the onset of dehydration and melting, due to the time required for establishment of magmatic pathways through the overlying continental lithosphere. The early–mid Miocene acidic magmatism in the north of the Transylvanian Basin (Szabó et al., 1992) may have been caused by crustal melting resulting from ponding of early phases of deeper mafic magmatism within the continental crust.

The relative timing of Miocene subduction and magmatism was also controlled by the width of the consumed ocean basin. If the basin were very narrow, subducted oceanic crust would not reach magma-generating depths before continental crust entered the subduction zone. A narrow basin would also only allow slab dehydration to occur in the magma-generating window for a short period of time. This agrees with the 130 km of Neogene shortening observed in the East Carpathian flysch belt (Roca et al., 1995).

The timing of subduction and subsequent mantle wedge metasomatism is critical to the timing of magmatism in the overlying volcanic arc. A lack of significant uplift and compression across the two colliding plates suggests that the rate of subduction quite closely matched the rate of convergence, estimated as 2.5 cm yr⁻¹ in the East Carpathians (Roca et al., 1995). If subduction were continuous, it would take 5–7 Ma for subducted lithosphere to reach the magma-generating depth of 80–120 km, assuming a subduction angle of 60° (as seen in the Vrancea zone). Decreasing the subduction rate or decreasing the angle of subduction increases the amount of time required for lithosphere to reach the magma-generating window. For example, a slow subduction rate of 0.5 cm yr⁻¹ at 60° would delay the onset of magmatism for 18–28 Ma after the initiation of slab descent at the surface.

Thus the observed time delay of 5–10 Ma between the main period of Miocene shortening and initial volcanism in the Late Miocene may be explained by the time required for a narrow slab of subducted lithosphere to reach depths of 80–120 km where dehydration, metasomatism and mantle partial melting occur and calc-alkaline magmas could be formed.

5.2. Breakoff of the subducted slab

The surface manifestation of subduction along the East Carpathians ceased as the main period of E–W directed thrusting in the flysch sediments came to an end. The transition from E–W compression across the East Carpathians to N–S compression in localised parts of the foredeep coincided with the main period of Pliocene volcanism in the CGH arc. Melt generation was still taking place at depth and arc volcanism continued long after collision had occurred. However, the duration of magmatic activity in any one part of the arc was brief and the length of time each volcanic centre was active decreased along the arc, as did the volume of erupted magma. To account for these observations we postulate that post-collisional slab breakoff took place in the subducted slab at mantle depths.

Slab breakoff can occur when light continental lithosphere follows dense oceanic lithosphere into a subduction zone (Spakman et al., 1988; Davies and
The buoyancy of the continental crust and downward pull of the dense oceanic slab cause extension across the continental–oceanic transition leading to tearing in the subducting lithosphere. The oceanic part eventually falls away into the deeper mantle and the resulting gap is filled by hot upwelling asthenosphere. Breakoff may take 5–10 Ma to occur and it may take another 2–10 Ma for the heat front to rise sufficiently to cause conductive melting of the overlying mantle wedge, possibly leading to magmatism at or near the surface (von Blanckenburg and Davies, 1995). The depth at which breakoff occurs is dependent upon the velocity of plate convergence, with slow subduction rates leading to shallower slab breakoff (Davies and von Blanckenburg, 1995).

Once breakoff has been triggered in the subducting lithosphere, it will tend to propagate laterally along the strike of the slab, particularly under the conditions of a low convergence rate (Yoshioka and Wortel, 1995). The downward pull on the detached part of the slab is transferred to the adjacent segment that is still attached to the surface, and a tear or rupture will migrate along the slab. This may occur at a rate of at least 10 km Ma\(^{-1}\) (1 cm yr\(^{-1}\)) depending upon the viscosity of the slab and the surrounding mantle material and the convergence rate (Yoshioka and Wortel, 1995). Lateral migration of slab detachment has been imaged by seismic tomography in the Hellenic arc and the Apennines/Tyrrhenian system (Wortel and Spakman, 1992), where slab breakoff occurred at depths between 100 and 300 km. Slab breakoff is also thought to have occurred in the Alps (von Blanckenburg and Davies, 1995) and has been suggested as an important tectonic process in the East Carpathian arc (Wortel and Spakman, 1993; Balintoni et al., 1998 in press).

In the East Carpathians, the relatively fast inferred subduction rate during the Miocene (>2 cm yr\(^{-1}\)) would suggest that slab breakoff would occur at depths >150 km, using the model of Davies and von Blanckenburg (1995). The general lack of alkali magmatism related to asthenospheric upwelling along the main part of the arc also suggests that the breakoff occurred at depths >50 km. If the convergence rate slowed as buoyant continental crust entered the subduction zone, slab breakoff would have migrated to shallower depths. Slab breakoff has been suggested at very shallow depths (40–60 km) beneath the Vrancea zone (Fuchs et al., 1979), where a seismic gap occurs between intermediate and shallow-level earthquakes.

It is likely that slab breakoff was initiated as the down-going subducted lithosphere was still dehydrating. If breakoff occurred at depths >120 km it would not have directly affected the melt generation zone. However, breakoff at depth would cause the slab pull force to cease, causing cessation of magma generation as the remaining attached slab would no longer be subducting. If breakoff occurred at shallower depths then magmatism may have continued as sinking lithosphere continued to pass through the magma-generating window. We suggest that slab breakoff occurred at depths greater than the magma-generation window in the north of the CGH arc and at much shallower depths beneath the South Harghita area and the Vrancea zone.

5.3. Migration of magmatic activity along the arc

Linzer (1996) explained the migration of the locus of calc-alkaline magmatic activity from NW to SE along the CGH arc as being related to the rollback of a small subducted slab beneath the East Carpathians to the Vrancea Zone. Rollback on the order of 600 km at a rate of 3.75 cm yr\(^{-1}\) beneath the Transylvanian Basin was suggested. We find the lack of extension in the immediate back-arc region (i.e. the Transylvanian Basin) surprising in a model of such rapid rollback. Furthermore this model ignores the possibility of subduction further north that produced the strongly thrusted flysch sediment belt (e.g. Sândulescu, 1984).

Sperner (1996) suggested that the down-going lithosphere in the Carpathian subduction zone broke up into small discreet slabs or ‘fingers’ of lithosphere along the strike of the arc. Down-dip breaks in the subducted slab could be caused by a steepening of the overall subduction zone during rollback. As rollback occurred, the change in the dip of the slab was not accompanied by a change in the curvature of the trench, leading to fracturing of the subducted lithosphere (Sperner, 1996). The slab beneath the Vrancea area was thus explained as being only the most SE-detached piece of the subducted oceanic lithosphere. Here we use the model...
of Sperner (1996) to postulate that magmatism in the CGH arc was related to a westward-dipping subducted slab, further north from the Vrancea slab, where compression was more E-W directed. Subduction of this lithosphere may have occurred in the Late Miocene with probable slab breakoff during the Pliocene.

Migration of magmatism along the CGH arc must be directly related to the migration of the subducted slab through the magma-generating zone in the mantle. The mechanism for this migration is unclear but it was most probably related to the geometry of the subduction. If oceanic lithosphere or thinned continental lithosphere were initially subducted obliquely along the East Carpathians, it would have reached magma-generating depths in the most northerly part of the CGH arc whilst remaining at shallower depths further south (Fig. 9a). As subduction continued the subducted lithosphere would reach progressively greater depths along the arc and on reaching a depth of ~80 km would have initiated magmatism in the overriding plate. The time at which the magma-generating window was reached would be later in the south than in the north of the arc. A southwards progression of the initiation of volcanic activity in the overriding plate would be accompanied by closure of the narrow subducted basin at the surface and, for a basin of uniform width, closure would initially occur in the north. As thicker continental lithosphere started to enter the subduction zone in the north, stress would be transmitted into the overriding plate. The subduction system would become locked in the north whilst basin consumption continued further south (Fig. 9b).

After 2–3 Ma in the collisional part of the arc, the down-going slab, still under the influence of slab pull at depth, would start to shear and rupture. Meanwhile, further south, crust continued to be subducted at the surface. As a result, a tear may have progressed along the arc from NW to SE along a zone of weakness in the down-going plate, most probably at the transition zone between different types of lithosphere (Fig. 9c). As continental lithosphere entered the subduction zone at progressively more southerly parts of the subduction system, slab breakoff would have migrated southward along the arc together with the volcanism. Volcanic activity in the north of the arc would therefore have ceased as the slab sank into the deeper mantle whilst in the south the final stages of subduction, mantle metasomatism and volcanism
continued. The convergence rate would have slowed as more buoyant continental crust entered the trench and consequently the downward motion of the slab would have slowed during the final stages of subduction in the south of the CGH arc. A consequence of the slow-down would be that the depth of slab breakoff would become shallower, migrating from depths >120 km to depths <80 km, thus shutting off the magma generation progressively along the arc. Eventually slab breakoff would be complete and the lithosphere would sink into the deeper mantle leaving behind the Vrancea slab further SE along the arc (not shown).

5.4. Asthenospheric upwelling following slab breakoff

Minor extension occurred during the Plio-Pleistocene at the southern end of the CGH arc, facilitating the rise of the ETB alkaline magmas through the Tisza–Dacia microplate and calc-alkaline magmas through part of the Cretaceous flysch. The major lithospheric attenuation normally associated with the generation of alkaline magmas is not seen in this area. The Transylvanian Basin is a thermal collapse or piggyback thrust basin exhibiting very low present-day heat flow (Veliciu and Visarion, 1982; Demetrescu and Andreescu, 1994) and did not undergo significant extension in the Plio-Pleistocene. The close proximity of the ETB alkaline magmatism to the CGH arc axis (Fig. 1) is incompatible with models of back-arc extension which suggest that extension occurs over a broad area some distance behind the arc (e.g. Karig, 1971). Thus rifting and decompressive melting driven by slab rollback are unlikely to have been responsible for the alkaline magmatism. Extension was much more localised along the E–W trending south Transylvanian fault and occurred during a period of transtensional stress during the Pliocene (Szakács and Seghedi, 1996; Balintoni et al., 1998).

Slab windows are areas along arcs that are not underlain by a subducted oceanic slab (Dickinson and Snyder, 1979). Asthenospheric mantle may rise into the void that is produced by slab breakoff and, if this occurs at sufficiently shallow depths (<50 km), decompression melting of the asthenosphere may occur. Major lithospheric extension in the overriding plate is not required by this model. Alkaline magmas produced in this way have been recorded from the Antarctic Peninsula, Baja California and British Columbia (Hole et al., 1991). All of these examples occurred just as subduction-related calc-alkaline arc magmatism was ceasing. The seismic gap in the presently active Vrancea tectonic zone to the south east of the CGH arc (Onescu et al., 1984) may be a slab window, similar to that postulated for a more northerly subducted slab in the Plio-Pleistocene.

According to Davies and von Blankenburg (1995), slab breakoff will occur at shallow depths if the subduction rate is slow. This may have happened during the final stages of subduction beneath the South Harghita area of the CGH arc as convergence waned and continental crust entered the trench. Subsequent asthenospheric upwelling could have caused direct melting to produce the alkaline magmas, these melts having the chemical and isotopic signature of the asthenosphere slightly metasomatised by slab-fluids.

Present-day heat flow measurements around the southern part of the CGH arc are the highest in the East Carpathian area, with temperatures estimated to be in excess of 800°C at 20 km depth (Demetrescu and Andreescu, 1994). Calc-alkaline magma generation occurred during the probable upwelling of hot asthenosphere to shallow depths, into the gap behind the detached slab. The increased fluid contribution in the final stages of arc calc-alkaline magmatism (Fig. 6) may reflect more efficient dehydration of the subducted lithosphere in a hotter subduction regime. Some of the unusual geochemical features at the south end of the CGH arc (high-K magmas, strong LREE/HREE fractionation (Fig. 8), high Sr/Y) may be related to increased temperatures in the subduction zone due to mantle upwelling. This may have also given rise to melting of the subducted oceanic crust in the eclogite field where garnet is stable.

6. Conclusions

We propose that magmatism in the East Carpathian arc was directly related to subduction of an oceanic lithospheric slab during the Miocene. Subduction of the slab through the magma-generating window caused slab dehydration, fluid meta-
somatism and melting of the mantle wedge. These processes continued at depth during the Pliocene, long after basin closure had ceased at the surface. The time gap between the main period of convergence and the timing of magmatism in the East Carpathian arc is consistent with the time required for subducted lithosphere to reach magma-generating depths, to cause mantle wedge metasomatism and melting, and to establish magmatic pathways for melts to reach the surface. Along-arc migration of magmatism is linked to a corresponding migration of the melt-generating zone at depth, together with progressive tearing of the slab from north to south, and was controlled by the geometry of rotation and/or width of the subducted basin.

The subducted lithosphere at the northern end of the CGH arc probably underwent slab breakoff at depths within or below the magma-generation zone (>100 km) during the Pliocene. A decrease in convergence rate during the final stages of subduction, in the extreme south of the CGH arc, caused breakoff to occur at shallower depths (~50 km) beneath the Southern Harghita area. Subsequent upwelling of asthenospheric mantle through a 'slab window', combined with more efficient dehydration of the descending slab under hotter conditions, had an important effect on the chemistry of the final stages of calc-alkaline magmatism.

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