Provenance analysis as a key to orogenic exhumation: a case study from the East Carpathians (Romania)

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ABSTRACT

Provenance analysis of the sediments from foredeep basins is crucial in understanding the contemporaneous orogenic exhumation processes. We report in this paper complex sediment provenance analysis using sandstone petrography and mudstone geochemistry, combined with magnetic susceptibility of the Upper Miocene to Pliocene deposits from Focșani foredeep basin (Romania). Data show a change of source area between 5 and 6 Ma, from an active volcanic arc towards a recycled orogenic belt, concurrent with an important increase of accumulation rate. This change was triggered by exhumation and erosion of the outer nappes from East Carpathians.

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Introduction and Geological overview

The East Carpathians (Fig. 1) represent an uplifted fold-and-thrust belt attached to a Neogene volcanic arc which provided sediments both to the Transylvania and foredeep basins. According to fission-track data, erosion of the northern East Carpathians began around 12 Ma, while in the southern part erosion started only at 4–5 Ma (Sanders et al., 1999). Contemporaneous volcanic activity took place in the Călimani, Gurghiu and Harghita Mountains (Fig. 1). Thus large quantities of detrital material derived from uplifted orogen and from active volcanic sources became available. The foredeep basin formed during Middle Miocene to Pliocene times in front of the East Carpathians, reached anomalous thickness (13 km, Tărăpoană et al., 2003) in the Focșani Depression (Fig. 1). During Pliocene times deformation of this foredeep basin started, resulting in near vertical tilting and erosion on the western flank (Dumitrescu et al., 1970; Matenco and Bertotti, 2000; Tărăpoană et al., 2003).

Focșani foredeep basin received recently much attention because its relationship with subduction plane was not yet deeply understood (Cloetingh et al., 2004); also our knowledge about its temporal and spatial relationship with other basins in the Paratethys region had some weak points (Jipa, 1997; Vasiliev et al., 2004), and virtually nothing was known about the source area of the huge amount of sediments accumulated in this basin (Tărăpoană et al., 2003).

The basin was filled up with Mio-Pliocene (Upper Sarmatian to Romanian in Eastern Paratethys chronostratigraphic nomenclature) shallow marine to shallow lacustrine sedimentary deposits. Very good exposures are on the western flank of the basin along the almost continuously outcropping Putna and Râmnicul Sărat river sections (Fig. 1). These sections consist in the lower part (Upper Sarmatian–Meotian) of alternating shallow marine sandstones and shales (Saulea, 1956) tilted to near vertical positions and in the upper part (Pontian–Dacian–Romanian) of brackish to lacustrine deltaic shales, siltstones, sandstones and coals (Pâă, 1966; Grasu et al., 1999), progressively less tilted to about 20–30°E. Most of the palaeocurrent features (measured on both river sections) show a dominant NNW–SSE trend, which agrees with the general N–S facies distribution: proximal facies in the north (Putna) and slightly distal facies towards the south (Râmnicul Sărat). However, during the entire basin evolution, no important change in the water depth occurred, as all sediments have sedimentary features typical for shallow water facies, despite the salinity changes.

Magnetostratigraphy provided the high-resolution age control on these sediments, and showed that their ages range from ~8.6 Ma (or the 9.5 Ma second option) at the base to ~2.5 Ma at the top (Vasiliev et al., 2004). However, after detailed analysis of magnetic susceptibility and timing of main volcanic eruptions, the optimal correlation of the magnetostratigraphy with astronomical polarity timescale (APTS) is Vasiliev’s first option; thus Putna section ranges from 8.6 to 5 Ma and Râmnicul Sărat section ranges between 7.3 and 2.5 Ma (Fig. 2). Paleomagnetic data from these two sections show that no significant rotations affected these deposits after their deposition (Dupont-Nivet et al., 2005).

Our case study shows that detailed provenance analysis gives important additional spatial and temporal constraints on the history of exhumation events and on the palaeogeographic evolution of the Focșani foredeep basin (Romania).

Provenance analysis

Sandstone petrography

We used optical microscopy to assess the clast’s origin from sandstones of the Putna and Râmnicul Sărat sections. After careful examination of the degree of diagenetic alteration, several cemented sandstones were selected for
Sandstones from the lower part of the sections (8.6–6 Ma) commonly contain zoned plagioclase (andesine 32–45% An) grains, without any sign of weathering, many unaltered volcanic lithoclasts (andesite, basaltic andesite) and large biotite crystals (Fig. 3), suggesting their provenience from a direct volcanoclastic source without sedimentary recycling. Therefore, the source area is limited to the Călimani and Gurghiu Mountains which were active during the same period (9.4–6.5 Ma according to Seghedi et al., 2004, 2005). No other volcanic province qualifies for source area: the nearby Harghita Mountains are too young (<6 Ma) and the older (12–9 Ma) are mainly intrusive sills and dikes located far north of Călimani Mountains (Peckskay et al., 1995; Mason et al., 1998). However, besides volcanic clasts, these sandstones also contain variable amount of metamorphic and recycled sedimentary clasts (20–45%) and quartz (15–35%).

Sandstones from the upper part of the sections (6–2.5 Ma) are dominated by metamorphic and sedimentary lithoclasts, being free of volcanic lithoclasts (Fig. 3). Frequent unaltered microcline and albite grains have been observed, as well as lot of quartz with undulatory extinction. All these features suggest a metamorphic source area, but no metamorphic complex is presently exposed in the region; the nearest ones are located 150 km to the west or north-west. These areas cannot be considered as potential
source areas, because they are located just near contemporaneously active volcanic chains (Harghita Mountains) and our studied sandstones contain no volcanic material from these rocks. Consequently, the only possible source area for the upper sandstones is the Cretaceous and Palaeogene flysch deposits. These are mainly sandy and muddy turbidites having a metamorphic source area themselves (Vinogradov et al., 1983; Grasu et al., 1998).

Geochemistry
We complemented the petrographic provenance analysis of sandstones with geochemistry of mudrocks as the two studied sections contained large proportion (more than 70%) of mudrocks: siltstones and shales. For comparisons, we also analysed 40 rock samples collected from the presumed source areas (metamorphic and sedimentary rocks, see Fig. 1 for locations). Whole-rock major and trace-element concentrations were acquired...
by X-ray fluorescence using Bruker-AXS spectrometer from University of Utrecht. Detection limits are in the 1–5 ppm range. Trace elements, such as Nb, Th, Zr, Ti, Sc, Cr, Ni, Rb are of main interest, because of their relatively low mobility during weathering, transport, and diagenesis (Condie, 1993; Girty et al., 1994; Fralick and Kronberg, 1997). In addition, we used several geochemical ratios between major and trace elements to discriminate the potential sources of the rocks. To test the influence of grain size on trace elements (mainly on Cr, Zr, Ti which tend to concentrate in the coarser fraction) we computed correlation factors between Al₂O₃ and Cr ($r = 0.63$), Zr ($r = -0.61$), TiO₂ ($r = 0.55$). At least for Cr and Ti, these correlation factors show the affinity to clay minerals and it seems that Zr is mostly fractionated in the coarser part; therefore, Cr and Ti from our mudrocks have been used for further source area discrimination.

To distinguish between mafic and felsic source rocks, we used Cr/Th ratio as a proxy. Pulses of high Cr/Th ratio can be observed, but also a general trend of increasing Cr content towards the top of the section (Fig. 4). The Cr (and also the Ni) content is slightly too high (100–150 ppm) for sediments originating from upper crustal sources (McLennan, 2001). Cr/Ni has quite low values (1.3–2.5 range) reflecting the presence of mafic or ultramafic rocks in the source area (Garver et al., 1996; von Eynatten, 2003). More studies are needed to clearly distinguish the source of Cr and Ni.

On the other hand, Rb/Sr is a rough monitor of changes in siliciclastic-carbonate ratios; it shows that siliciclastic input has high amplitude pulses in the first half of the section and less fluctuation in the second half (Fig. 4). These high pulses are most likely related to the alternation of marine and brackish environments with possibly some diagenetic cementation. As can be seen from the parallel diagram of the two ratios (Fig. 4), no correlation exists between them; thus Cr and Th (same is valid also for Ti and Nb, not shown) are independent of changes in sedimentary environments and of diagenetic overprints, reflecting mainly the source area composition.

Ti/Al ratio is considered a marker for the flux of siliciclastic material from the newly uplifted orogen with high values occurring with high erosion rate without much in situ weathering (Sageman et al., 2003). As can be seen in Fig. 5, the Ti/Al ratio is generally very high (> 0.1) which indicate high sedimentation rate, also individual values are highly variable, but when computing the 5-point moving average, a significant change can be observed around 5 Ma. In the lower part, Ti/Al has a good fit with average values of Ti/Al from rocks collected in the East Carpathians from potential source areas located north of Trotuș fault’s prolongation. The upper part fits well with Ti/Al average values from rocks located south of Trotuș fault. Almost a similar pattern is reflected also by Ti/Nb ratio (Fig. 5). Ti/Nb ratios decrease from basic to acidic compositions in both orogenic and anorogenic settings (Hofmann, 1988; Bonjour and Daubard, 1991). In our case, the highly oscillating values from the lower part (8–5 Ma) reflect the pulses of volcanic material with very low Nb content; which is a characteristic feature for Călimani–Gurghiului volcanic rocks (Mason et al., 1998). In the upper part (5–2 Ma), Ti/Nb ratio oscillates...
less and is significantly lowered showing the dominance of more acidic source composition compatible with geochemical pattern of the rocks from source area south of Trotuș fault.

Magnetic susceptibility

Magnetic susceptibility measurements have been done on a Kappabridge KLY-3 at University of Utrecht, on the same samples as the magnetostratigraphy (Vasiliev et al., 2004) which means all kind of rocks: shales, siltstones and fine-grained sandstones. Plotting the results against age reveals some specific features (Fig. 6): (1) the Putna section comprises rocks with relatively strong, but largely varying magnetic susceptibilities (highest values are from sandstones) carried mainly by detrital magnetite; (2) the Râmniciul Sârat section contains rocks with relatively weak magnetic susceptibility and less variation (however, less sandstones in this section) carried mainly by iron sulphides (Vasiliev et al., 2004).

We conclude that the episodic input of magnetite grains and proximal sedimentary environments (oxygenated waters) are responsible for the susceptibility pattern of Putna section. Large amount of magnetite grains could have been derived from contemporaneously active volcanoes like those from Călimani and Gurghiu Mountains. The higher values of magnetic susceptibility strongly reduce towards the upper part of the Putna section (6.5–5.5 Ma). The weaker susceptibility in the rocks from the Râmniciul Sârat section is explained by partial dissolution of primary magnetite grains and by authigenesis of iron sulphide crystals in more reducing and distal sedimentary environments.

East Carpathian’s exhumation

Previous geochronological studies demonstrated that exhumation of the East Carpathians began 12 Ma in the northern part, and that the southern part started to be eroded later (c. 4–5 Ma) (Sanders et al., 1999). This study combined sandstone petrography with magnetic susceptibility and trace-element analyses of mudrock to better constrain the sedimentary provenance of Upper Miocene to Pliocene infill of the Focșani foredeep basin.

Sandstone petrography showed a change in source area from a volcanic arc province to a recycled orogen province. The geochemical signature of the mudrocks illustrates pre-6 Ma, a contamination with volcanogenic material derived from contemporaneously active volcanoes and a significant change of source area around 5 Ma. Direct-input volcanogenic material into the shallow-water marine to deltaic deposits of Upper Sarmatian to Meotian age implies that there was a major connection between the Călimani and Gurghiu Mountains and the basin. This proves that the sedimentary basin has extended westward and that the present-day Cretaceous–Tertiary nappe system located south of the Trotuș fault was not yet exhumed before 5 Ma. This interpretation (except timing) is similar to that
presented by Tărăpoanca et al. (2003), but their model was based on correlation of the subsidence and uplift rates from adjacent basins. They also used a timescale (Andrescu, 1979) which later was proved to be inaccurate (Vasiliev et al., 2004).

Magnetic susceptibility data are also consistent with this interpretation. The high proportion of magnetite can be related to the main volcanic events in the East Carpathians. Such events could be the huge debris avalanche recently dated at 8 ± 0.5 Ma (Seghedi et al., 2005) and the caldera collapse dated at 6.9 Ma (Seghedi et al., 2004) (Fig. 6).

No other major pyroclastic eruption is known previous to 8.5 Ma (Seghedi et al., 2005). The decay of susceptibility after 6 Ma, when the volcanoes were still active and even closer (eruption phases related to Harghita Mountains), demonstrates that the previous connection with the internal structures of the East Carpathians was closed by a watershed which evolved, may be due to a late continental or oceanic crust. (Hipolyte et al., 1999).

Sedimentation rates have been recalculated from compacted sediment thickness and magnetostatigraphic dating (Vasiliev et al., 2004), using the calibrating points from APTS. We used compacted thickness, because from field observation, the degree of sediment compaction was not significantly different throughout the sections of the Dacic basin and correlation with neighboring areas. Ann. Geol. Pays Hellen., 4, 129–138.


McLennan, S.M., 2001, Relationships between the trace element composition


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