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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 Focşani 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, Tărăpoancă *et al*., 2003) in the Focşani 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; Tărăpoancă *et al*., 2003).

Focşani 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 (Tărăpoancă *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 (Pană, 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). Palaeomagnetic 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 Focşani 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

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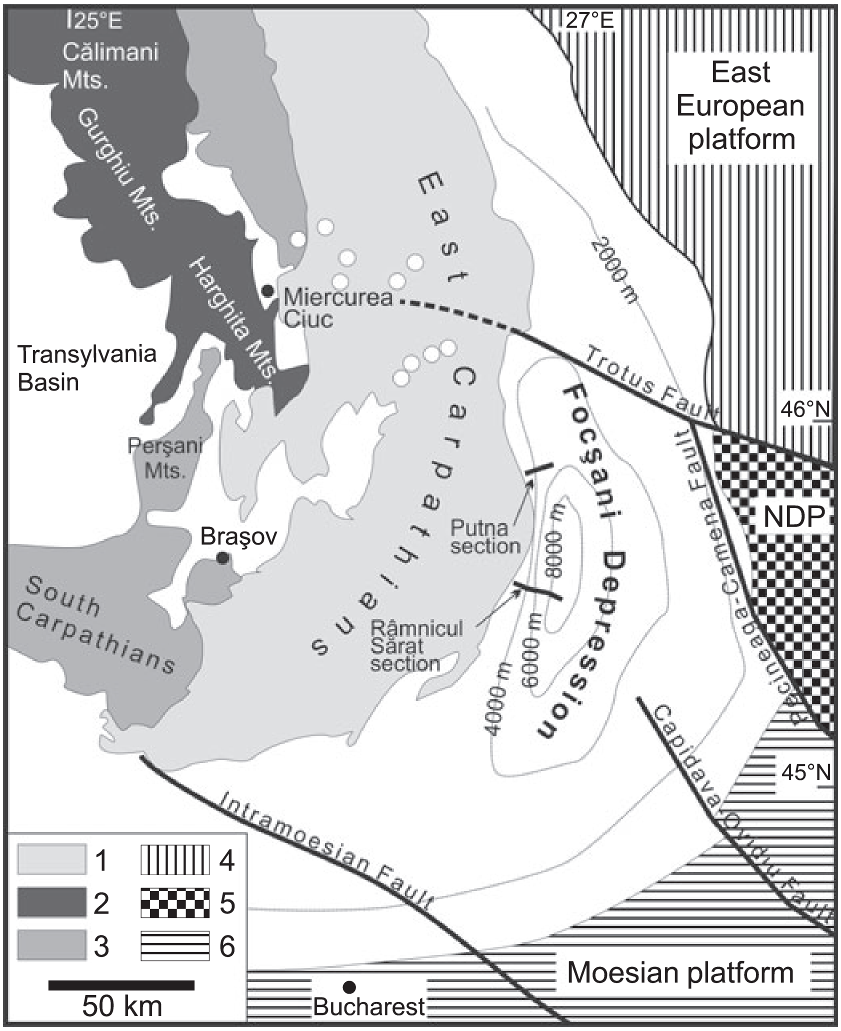

Fig. 1 Geological sketch map showing the location of the two studied sections and the position of the Focşani foredeep basin (Focşani Depression on the map) with respect to potential source areas. 1 = East Carpathians Cretaceous–Tertiary nappe system; 2 = Neogene-Quaternary volcanic arc; 3 = Crystalline basement and its Mesozoic cover; 4 = East European Platform; 5 = North Dobrogean Promontory; 6 = Moesian Platform, white dots = sampling areas from potential sources (40 samples), labelled isolines = isobaths in metres at the base of Neogene (after Dumitrescu and Săndulescu, 1970).

further modal analyses. Samples showing dissolution of feldspars, large and inhomogeneous matrix content, and high degree of clast's fractures have been discarded. The studied sandstones are compositionally immature because of high content of feldspar and lithics, and also texturally immature because of highly angular grain shapes and low degree of grain-size sorting.

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 (Pecskay *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

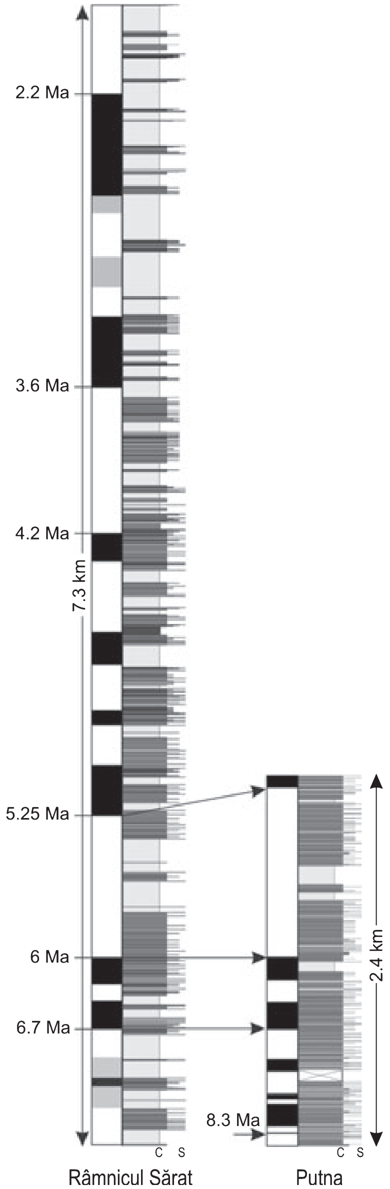

Fig. 2 Lithologic logs of Putna and Râmnicul Sărat sections and their correlation based on magnetostratigraphy (Vasiliev *et al.*, 2004). C = clay, S = sand, black intervals = normal polarity, white intervals = reversed polarity, grey intervals = undefined polarity.

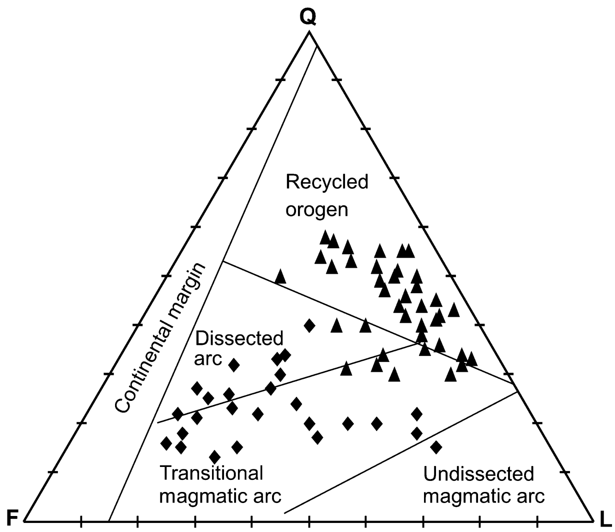

Fig. 3 Sandstones petrography and their potential source area based on Dickinson (1985) diagram and point counting method (300 grains counted per thin section, e.g. Ingersoll *et al.*, 1984). Diamonds = Sarmatian to Meotian (8.5–6 Ma) sandstones; triangles = Pontian to Romanian (6–2 Ma) sandstones, Q = quartz grains (both monocrystalline and polycrystalline), F = feldspar grains, L = lithic fragments.

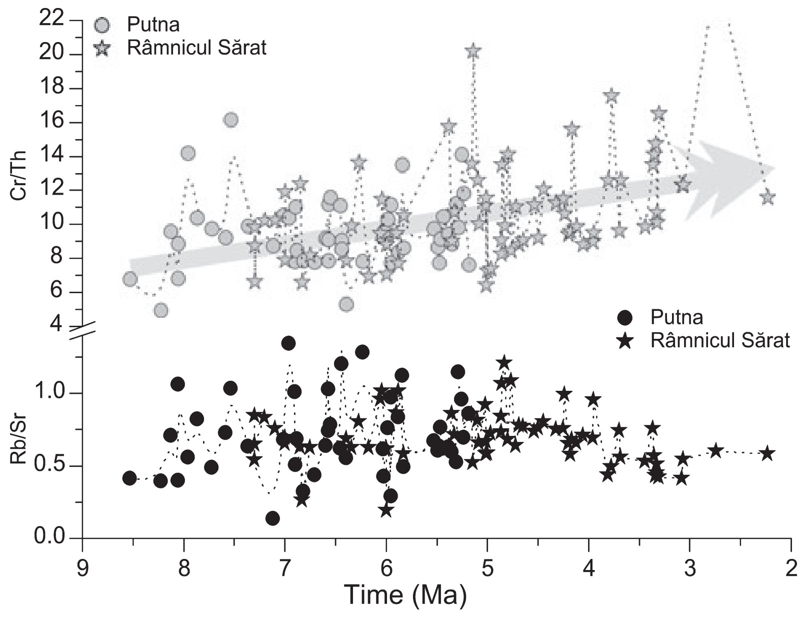

Fig. 4 Rb/Sr and Cr/Th geochemical ratios from Focşani foredeep basin discussed in text, illustrating the general trend of increasing Cr content through time.

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 Al_2O_3 and Cr ($r = 0.63$), Zr ($r = -0.61$), TiO_2 ($r = 0.55$). 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 Trotuş fault's prolongation. The upper part fits well with Ti/Al average values from rocks located south of Trotuş 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 Dabard, 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–Gurghiu volcanic rocks (Mason *et al.*, 1998). In the upper part (5–2 Ma), Ti/Nb ratio oscillates

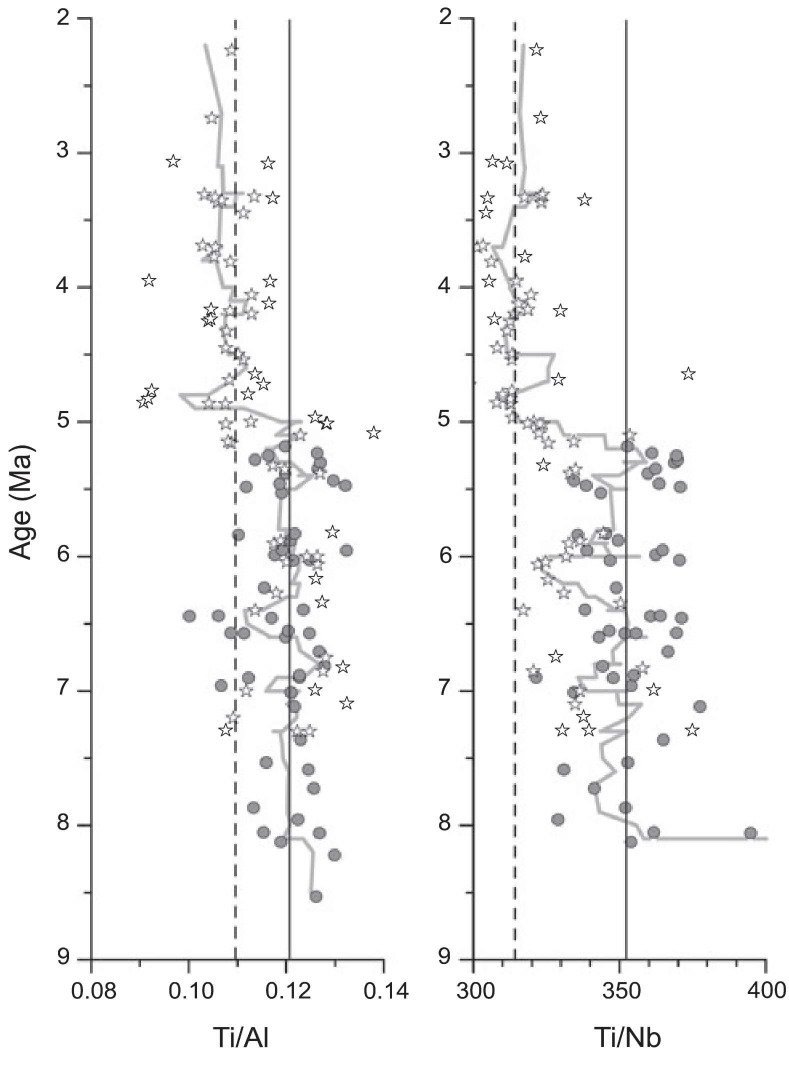

Fig. 5 Ti/Al and Ti/Nb geochemical ratios of mudrocks (circles = Putna section; stars = Râmnicul Sărat section) showing significant changes in the source area around 5 Ma. Thick grey line = 5 points moving average, vertical lines = average values from Cretaceous–Palaeogene flysch (solid line = north of Trotuş fault, dashed line = south of Trotuş fault).

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 Trotuş 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 Râmnicul Sărat 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 Râmnicul Sărat section is explained by partial dissolution of primary magnetite grains and by authigenesis of iron sulphide crystals in more reducing and distal sedimentary environments.

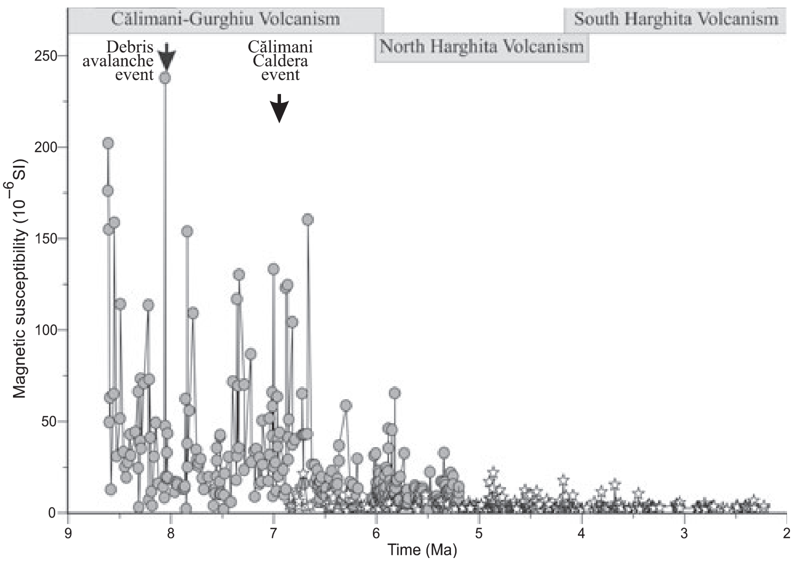

Fig. 6 Mudstone's magnetic susceptibility and the possible sources of exceptional high values. Circles = Putna section; Stars = Râmnicul Sărat section.

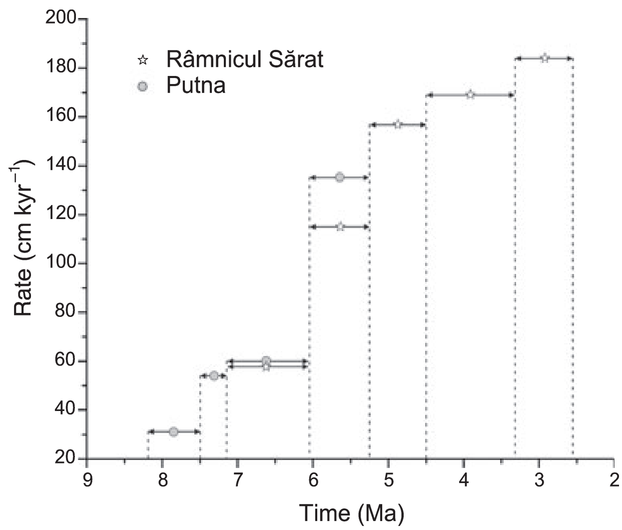

Fig. 7 Sedimentation rates calculated between the calibrated points of the magnetostratigraphy with APTS (after Vasiliev *et al.*, 2004). Note the doubling of sedimentation rate after 6 Ma.

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 Focşani 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 Trotuş fault was not yet exhumed before 5 Ma. This interpretation (except timing) is similar to that presented by Tărăpoancă *et al.* (2003), but their model was based on correlation of the subsidence and uplift rates from adjacent basins. They also used a timescale (Andreescu, 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 thrusting phase (Hippolyte *et al.*, 1999).

Sedimentation rates have been recalculated from compacted sediment thickness and magnetostratigraphic 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 until the limit between Dacian and Romanian deposits (4 Ma). As shown in Fig. 7, in the time interval between 8.5 and 6 Ma, sedimentation rate was relatively low, just reaching 60 cm kyr^{-1} and being similar to the time integrate erosion rate of 50 cm kyr^{-1} for the central part of the East Carpathians (Sanders *et al.*, 1999). Between 6 and 5 Ma, there is a jump in sedimentation rate from 60 to 150 cm kyr^{-1}, which cannot be attributed to the differences in compaction degree. The increase of sedimentation rate after 6 Ma, as well as the dominance of sedimentary and metamorphic sources between 6 and 2 Ma, all converge to the appearance of a new source area, which can only be the Cretaceous–Tertiary nappe system. This system became exposed between 5 and 6 Ma in the southern part of East Carpathians as shown by geochemical data. Doubling in the sedimentation rate after 6 Ma is in good agreement with doubling the erosion rate for the bending zone (100 cm kyr^{-1}, Sanders *et al.*, 1999) and the increase of subsidence (Mat enco *et al.*, 2003; Tărăpoancă *et al.*, 2003).

Our results based on data from basin's sediments and from potential source area are compatible with previous tectonic models and they offer additional spatial and temporal constraints on the connection between exhumation history of the southern part of East Carpathians and the evolution of Focşani foredeep basin.

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