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Geochemistry of Eocene flysch sandstones in the NW External Dinarides

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We present the main petrographic and geochemical features of the Lower to Middle Eocene turbiditic sandstones from the northwestern portion of the External Dinaride flysch basin. Sampled areas cover SW Slovenia (Vipava and Brkini Basins) and the Istrian Peninsula (Trieste-Koper and Pazin Basins). Framework constituents of the lithic arenites reveal low-grade metamorphic, acidic plutonic, and to a lesser extent, mafic volcanic and ultrabasic sediment sources, with evidence for a small degree of sediment recycling as well. Among the processes that commonly influence sediment compositions, weathering in the source and sorting were probably negligible, but carbonate contribution of detrital or intrabasinal origin diluted the siliciclastic portions to various degrees. Main and trace element compositional data agree well with petrography and clearly indicate the predominance of felsic, crustal source lithologies. Exposed mafic-ultramafic source units were volumetrically less important.

From the Early Paleogene, extensive sediment mixing occurred in front of the Dinaride orogenic thrust wedge, with the components derived from different Dinaride units of felsic crystalline basement, platform carbonates and ophiolite. In the Eocene, a likely source of the mafic-ultramafic detritus was the Jurassic ophiolitic mélange in the NE Dinarides.

Key words: Tertiary, Dinarides, flysch, sandstone, foreland basin, petrography, geochemistry, provenance

Introduction

A nearly continuous belt of Upper Cretaceous to Miocene flysch successions extends from the Southern Alps along the entire outer margin of the External Dinaride thrust belt. These synorogenic deposits become progressively younger

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toward the SE (Marjanac and Ćosović 2000). Flysch formation is probably related to the large-scale stress field that has affected the whole Adriatic realm since the Late Cretaceous, resulting in foreland basin evolution and deep marine sedimentation (Channell et al. 1979; Marinčić 1981; Aljinović et al. 1990; Tari 2002).

In the Southern Alps-Dinarides junction, the tectonically least disturbed and best exposed sections of the Paleogene External Dinaride flysch are found at the Istrian Peninsula and in SW Slovenia (Fig. 1). Based on the heavy mineral associations of turbiditic sandstone combined with paleocurrent directions toward the SE, Magdalenić (1972) concluded that a large part of the detrital material was derived from the Alps, with minor contributions from the NE, i.e. from the Dinarides. However, Marinčić (1981) and Marinčić et al. (1996) proposed that the entire clastic material was derived from the Dinarides and explained axial flow directions by flow deflection. A third type of paleocurrent data, indicating sediment transport toward the NW, was also reported (Orehek 1991). Debrites and turbidites of pure carbonate composition are intercalated in the succession, which consistently indicate ESE-directed transport (Babić and Zupanič 1996).

The flysch deposits are surrounded by thick carbonate platform sediments, which poses a further problem in source area assignment. Results of heavy mineral analyses by Magdalenić (1972) readily suggest provenance mixing in the foreland basin from various sources which, however, needs to be demonstrated in more detail.

In this paper we present the results of a pilot provenance study, focusing on petrographic and whole-rock geochemical analyses of sandstone from the flysch successions of Istria and SW Slovenia.

Geologic setting and stratigraphy of the flysch

The outermost structural element of the Dinarides, the Adriatic Carbonate Platform s.str. (Vlahović et al. 2005) became emerged in the Late Cretaceous, followed by karstification and bauxite formation. The regional erosional surface is overlain by a Paleogene overstep sequence recording foreland basin sedimentation on a carbonate ramp with progressive deepening and transition into flysch deposition (Košir 1997; Marjanac and Ćosović 2000; Vlahović et al. 2005). During Tertiary nappe stacking the most external part of the dismembered platform was a rigid block acting as a foreland to both the Dinarides and the Apennines. It underwent Late Tertiary CCW rotation of 30° with respect to the External Dinaride nappes (Márton et al. 1990; E. Márton, pers. comm., 2005). The flysch underlies these nappes and vitrinite reflectance data indicate burial up to 3500 m (Rainer 2003).

The flysch successions are made up of siliciclastic turbidite beds, generally of 5–40 cm in thickness, intercalated with hemipelagic marl. The turbidites represent incomplete, Tb-e, and more often, Tc-e and Td-e Bouma sequences. Parallel lamination and current ripples are rarely visible. In the stratigraphic column of

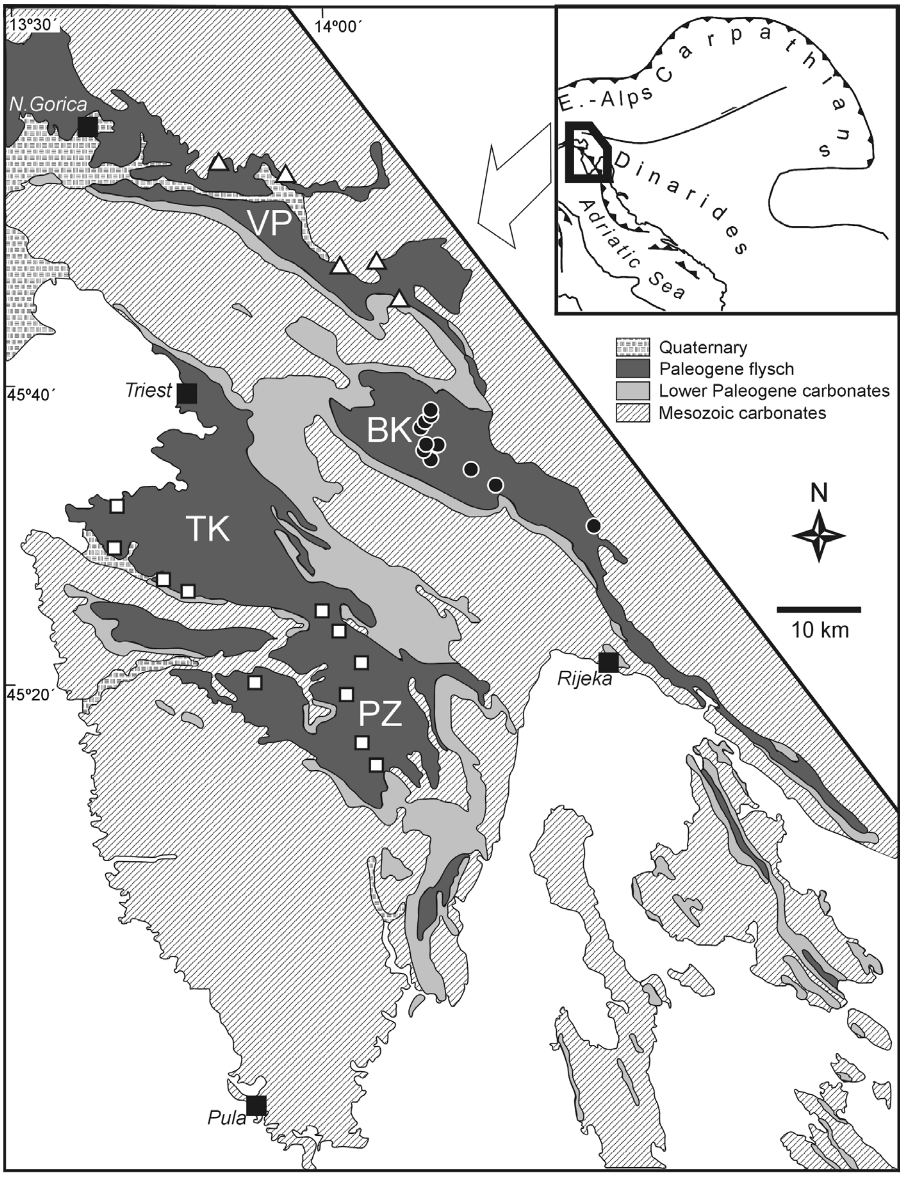

Fig. 1
Geologic sketch map showing the distribution of Paleogene flysch deposits in the NW External Dinarides (after Bigi et al. 1991, simplified). Segments or "sub-basins" of the flysch belt: VP=Vipava [Paleocene to Ypresian] BK=Brkini [Upper Ypresian to Middle Lutetian], TK=Triest-Koper, PZ=Pazin. TK and PZ are Upper Lutetian to Lower Priabonian in age and referred to as Istria in the text. Symbols show sampling locations of turbiditic sandstone; triangles: Vipava Basin, circles: Brkini Basin, squares: Istrian Basin

Istria, a coarsening-upward sequence was established; the turbidite thickness clearly increases whereas those of hemipelagics are constant or decrease (Magdalenić 1972), indicating an increasingly proximal position. Estimates of the thickness of the Istrian flysch vary from 300 m (Marinčić et al. 1996) to 500 m (Pavšić and Peckmann 1996).

Planktonic foraminifera biostratigraphy and available calcareous nannoplankton data prove that in the NW Dinarides flysch sedimentation commenced earlier in the SW Slovenian part, in the Early Eocene (Piccoli and Proto Decima 1969; Drobne 1979). In Istria the flysch is predominantly of Middle Eocene age, covering a time span from Middle/Late Lutetian (Tari-Kovačić 1997) to Early Priabonian (Benić 1991).

Sampling and analytical techniques

Samples were taken from the Lower Eocene Vipava Basin, the Lower to Middle Eocene Brkini Basin, and from the Middle Eocene Istrian Basin. Sampling locations are shown in Fig. 1. Bias due to compositional effects of grain-size variations and of recent weathering was minimized by always sampling very fine to medium-grained material, and whenever possible, from the fresh part of the turbidite beds. Sandstone samples were then investigated by common petrographic techniques. Thin sections were stained for K-feldspar following Houghton (1980). For whole-rock geochemistry, 27 samples were selected and carefully crushed to chips of <4 mm using an iron press and a plastic sieve to minimize contamination. Care was taken to select only material that is unweathered, well-cemented and largely free of calcite veinlets. After pulverization in an agate disc mill, loss on ignition (LOI) was determined gravimetrically following an overnight heating at 1050 °C. Fused borate glass discs were made with Merck™ Spectromelt™ A12. Major and trace (V, Cr, Co, Ni, Zn, Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Nd, Sm, Eu, Yb, Pb, Th and U) element concentrations were determined by X-ray fluorescence analysis at the Department of Geochemistry, University of Tübingen, using a Bruker AXS S4 Pioneer spectrometer. Scandium concentrations of selected samples were determined by INAA in the nuclear reactor at the Technical University of Budapest. Accuracy is within ±1% for major and ±10% for most trace elements. Analytical precision is better than 3% for major elements and better than 10% for most trace elements. Sc analyses are accurate to within ±1% with a precision better than 2%.

Due to the significant carbonate content of the samples (see below) compositional data treated herein were carbonate-corrected prior to further data analysis, by recalculating the analyses on a Ca-free basis. Carbonate dilution is a serious problem; it masks the Ca-contents linked to apatite and silicate components such as plagioclase feldspar or mafic lithic fragments. However, with respect to element ratios dealt with in this paper the dilution effect is largely negligible.

Results and discussion

Petrography

Thin sections were examined to gain basic information on the nature of available lithologies in the source area. Due to the difficulties of correlating between outcrops of the rather monotonous turbidite succession, vertical petrographic trends could not be established. On the other hand, a clear decrease in average grain size is observed from the Brkini Basin toward Istria. In many of the Vipava and Istria samples a reliable framework grain characterization was not possible due to the small grain size and high matrix content, indicating considerable diagenetic overprint.

All sandstone samples are characterized by a low degree of textural maturity and a comparatively high mineralogical maturity. They are always quartz-rich and, compared to the other basins, the greywacke of Istria is richer in quartz at the expense of lithic fragments and feldspar but are texturally less mature, containing matrix up to 30%.

Estimation of framework component abundances shows that all samples are dominated by quartz (60–80%). Lithic fragments (10–25%) predominate over feldspar (5–15%). The amount of dark matrix varies between 5 and 20% in the Brkini Basin and can exceed 30% in Istria. The presence of small amounts of micritic cement is ambiguous; it cannot be readily distinguished from alteration products of unstable framework components.

The clear overall fining trend observed from the Brkini Basin distally, i.e. toward Istria, is accompanied by an increase of quartz among the framework grains and of matrix. In these rocks rare planktonic foraminifera can be recognized. However, they contain up to 40 wt% CaO (see below), suggesting that the carbonate material is fine-grained and thus largely represented by the matrix. Sand-sized detrital carbonate grains were not found in any sample in the Brkini and Vipava basins, but angular, micritic carbonate fragments of extrabasinal origin are common in Istria.

Quartz is angular to subangular and many (~60%) are monocrystalline with undulatory extinction or polycrystalline, consisting of 2–3 subgrains. Non-undulatory quartz and quartz with subgrains are subordinate. This conforms to a low-grade metamorphic source (Basu et al. 1975), but both the breakage of grains upon transportation and the small grain size limit the reliability of this interpretation. Rounded quartz grains were not observed. Among the feldspars, sodic plagioclase prevails. K-feldspar is mainly orthoclase with some microcline, whereas sanidine was not observed, suggesting the predominance of plutonic sources over acid volcanics. Flakes of detrital biotite and muscovite are subordinate and always bent, indicating considerable compaction. Patches of chlorite are common. Zircon, tourmaline, rutile, garnet and Cr-spinel were readily identified as accessory framework constituents.

The lithic fragments include the following (identifiable mainly in the Brkini Basin): Quartz-mica aggregates: here, the mica is either randomly oriented in fragments of hypidiomorphic granular texture, or aligned parallel to sheared quartz grains, suggesting acid plutonic and low-grade, felsic metamorphic origin, respectively. Slate: rounded, dark, often weathered grains with or without recognizable lamination. Orthoquartzite: subrounded grains showing strongly foliated texture, the laminae separated by limonitic films. Felsic volcanics: subangular to subrounded grains of vitrophyric texture with small (~10 μm) feldspar and quartz microphenocrysts embedded in a glassy, usually devitrified, groundmass. Mafic volcanics include subrounded grains mainly of ophitic to intersertal texture. Their groundmass is highly chloritized. Serpentinitic rock fragments are made up of small needles or laths in a complex, mesh-like intergrowth texture. They often incorporate minute grains of opaque phases. About 10% of the lithic fragments is represented by chert, but "ghosts" of radiolarians are rarely preserved.

It follows that most of the examined sandstone samples are classified as lithic arenite according to Pettijohn et al. (1973). Based on their elevated matrix content, some samples in the Istrian Basin are intermediate with the lithic wackes. The framework constituents are derived chiefly from low-grade metamorphic, acidic plutonic, acidic subvolcanic (microcline), mafic volcanic, ultrabasic and older sedimentary sources. Typical textural relationships and frequent framework grains are shown in Fig. 2.

Whole-rock geochemistry

Sandstone compositions are primarily controlled by the bulk lithologic composition of the source. They can be influenced by the interplay of several further factors including the intensity and duration of chemical weathering, hydraulic sorting, diagenesis and sediment recycling (e.g. Johnsson 1993). However, if fresh detritus is rapidly transported from the source to a nearby depositional site (e.g. via short-term fluvial transport followed by marine turbidity currents in the adjacent foreland basin), the element budget of sediments can be used to decipher the overall composition of the source area (Taylor and McLennan 1985). Some trace elements such as Zr, Hf, Y, Ti, Nb, Ta, Sc, V, Th, U and the REE are considered to be nearly immobile in aqueous systems and thus of particular importance: their fractionation during weathering, transport and deposition is comparatively low and they can sensitively indicate minor but important source components (McLennan et al. 1980; Bhatia and Taylor 1981; Bhatia 1985; McLennan et al. 1993).

Results of whole-rock geochemical analyses of the flysch sandstones are shown in Table 1.

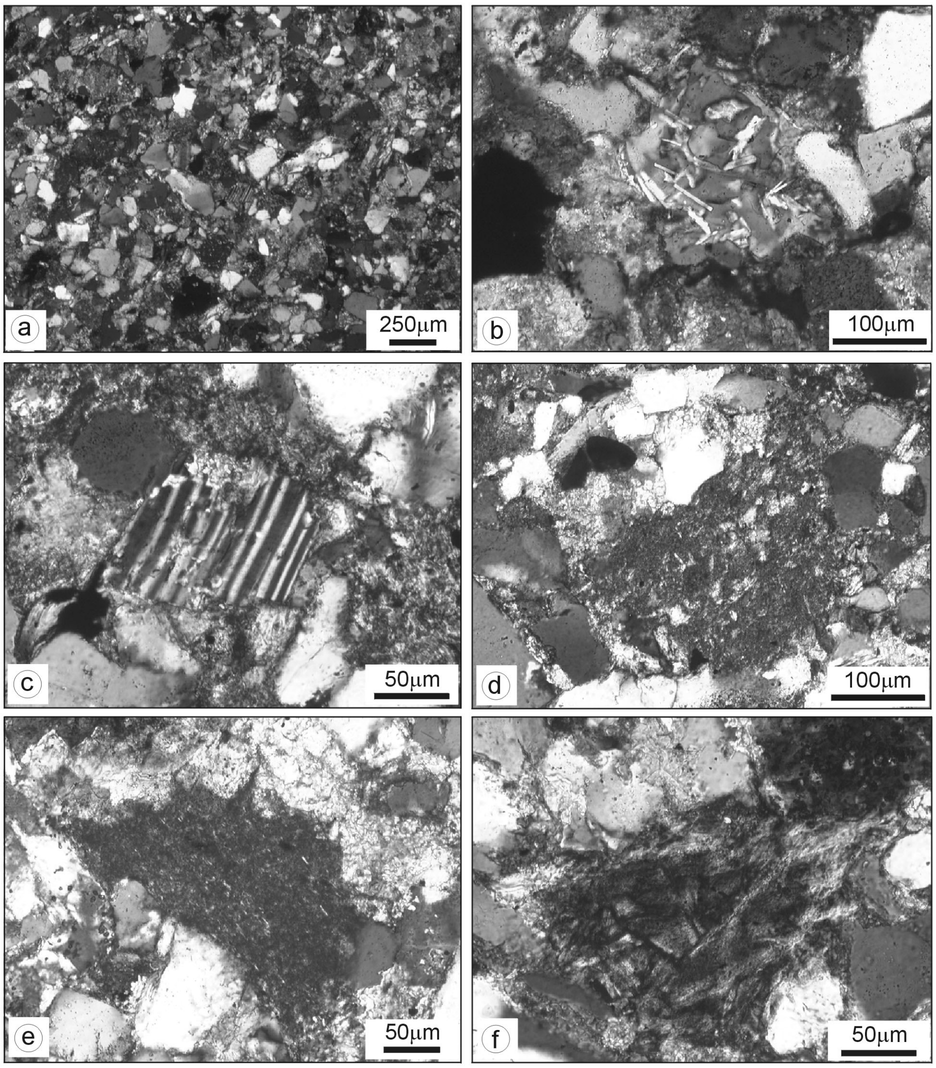

Fig. 2
Typical textural features of the Brkini sandstones. a) Lithic greywacke with abundant subangular quartz fragments; b) quartz-muscovite aggregate; c) sodic plagioclase grain (extinction angle in the symmetrical zone: 16°); d) mafic, microphaneritic volcanic rock fragment with minute patches and laths of feldspar. e) mafic, aphanitic volcanic rock fragment; f) serpentinitized ultramafic rock fragment. Serpentine exhibits fibrous to mesh texture

Table 1
XRF analyses of siliciclastic turbidite sandstone from the NW External Dinaride flysch. Refer to Fig. 1 for basin names. Sc concentrations were determined by instrumental neutron activation analysis (INAA)

Basin	TK	TK	TK	TK	PZ	PZ	PZ	PZ	PZ	PZ	PZ	BK	BK
Sample	IZO-1	MAR-1	SCA-1	CRN-1	LUK-3	SVZ-1	ZAJ-1	KRJ-1	SLK-1	BAS-2	BNE-1	HAR-1	KOZ-4
Grain size (ϕ)	3.5-4	2.5-4	3-4.5	3.5-3	2.5-4	2-3.5	2.5-4	3.5-4	3.5-4	3-4.5	2.5-4	2-3.5	2.5-3.5
Rock type	lw	la	lw	lw	la	lw	lw	lw	lw	lw	lw	la	la
Remark		mc	mc	mc	mc		mc	mc	mc	mc	mc		
SiO_2 (wt%)	49.14	28.26	39.84	29.48	32.68	37.67	34.74	22.50	30.64	41.43	34.78	78.97	66.55
TiO_2	0.33	0.20	0.37	0.26	0.25	0.31	0.29	0.17	0.21	0.40	0.28	0.61	0.50
Al_2O_3	5.18	2.91	5.71	3.08	4.03	4.77	4.26	2.55	3.43	6.62	4.36	9.23	6.82
$Fe_2O_{3\,tot}$	1.86	1.61	2.47	1.64	1.94	1.76	2.38	1.39	1.95	3.24	1.93	3.97	3.08
MnO	0.09	0.08	0.07	0.09	0.08	0.06	0.09	0.06	0.10	0.09	0.10	0.09	0.08
MgO	0.78	0.72	1.20	1.31	1.31	1.27	1.03	1.00	0.98	1.18	0.99	1.40	0.96
CaO	21.53	35.89	26.40	34.90	32.27	27.06	31.00	39.59	33.95	24.13	30.95	0.21	9.60
Na_2O	1.03	0.38	0.70	0.47	0.52	0.90	0.54	0.37	0.48	0.78	0.49	1.22	1.10
K_2O	0.91	0.52	1.00	0.57	0.69	0.64	0.76	0.49	0.64	1.10	0.76	1.24	1.01
P_2O_5	0.07	0.06	0.07	0.06	0.07	0.07	0.07	0.08	0.06	0.09	0.07	0.07	0.08
LOI	18.38	29.31	22.08	28.03	25.96	24.07	24.76	31.72	27.49	20.85	25.21	2.98	9.35
Sc (ppm)	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.
V	29.8	23.5	48.6	23.2	40.3	38.1	36.9	20.0	26.0	57.8	30.4	64.3	53.6
Cr	137.5	124.0	143.1	189.7	103.7	118.4	155.6	82.2	96.4	122.8	112.0	283.5	230.8
Ni	26.0	26.4	37.9	26.6	37.1	10.7	46.3	17.6	32.1	58.9	29.0	144.2	47.1
Co	5.2	3.8	7.9	6.6	6.0	3.8	10.5	2.2	5.5	12.7	3.4	15.9	10.2
Zn	27.5	32.2	49.9	33.3	36.9	25.3	42.8	28.9	37.6	53.8	41.3	45.2	45.4
Rb	33.8	22.2	42.0	22.8	30.9	22.7	35.6	19.5	26.6	47.5	32.7	51.3	43.9
Sr	480.9	309.2	323.1	350.2	387.2	431.0	311.2	416.1	291.0	287.9	340.8	38.0	181.3
Y	14.0	11.5	16.3	13.1	12.0	14.4	12.3	n.d.	11.2	17.7	14.9	24.0	28.2
Zr	117.4	52.1	111.6	110.5	66.2	121.2	74.8	44.7	48.2	89.0	70.6	188.2	212.6
Nb	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Ba	177.3	66.1	133.6	83.7	1268.4	3653.5	106.5	50.1	88.0	118.6	98.0	179.7	98.0
La	55.5	32.8	46.9	48.7	27.1	n.d.	52.9	54.4	63.4	51.4	52.5	26.8	30.7
Ce	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	39.6	40.4
Nd	18.5	16.6	17.8	21.2	15.0	10.4	13.9	15.1	14.4	24.2	22.8	26.2	23.4
Sm	2.4	n.d.	3.5	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	5.2	4.2	4.4	3.1
Eu	1.2	0.7	1.0	0.8	0.9	1.0	0.7	0.9	0.7	1.1	1.1	0.5	0.7
Yb	0.9	0.8	1.2	0.7	0.8	0.9	0.9	0.4	0.7	1.4	1.1	2.0	2.2
Pb	n.d.	n.d.	n.d.	10.0	n.d.	n.d.	n.d.	7.4	4.9	8.7	n.d.	5.6	12.4
Th	n.d.	n.d.	n.d.	5.2	n.d.	n.d.	6.7	5.1	5.2	5.4	n.d.	5.0	7.9
U	1.5	0.6	n.d.	1.5	0.6	2.0	n.d.	1.6	0.6	n.d.	n.d.	n.d.	n.d.
Total (wt%)	99.41	100.00	100.00	100.00	100.00	99.01	100.00	100.00	100.00	100.00	100.00	100.11	99.23
CIA	64.4	67.9	70.3	66.1	69.2	67.0	69.2	65.9	67.4	71.4	70.7	72.3	68.9
molar Na/K	1.71	1.11	1.06	1.26	1.15	2.12	1.08	1.16	1.13	1.08	0.97	1.49	1.65

la: lithic arenite; lw: lithic wacke; mr: mudrock

mc, ms: on raw XRF data of samples with unusually high CaO or SiO_2, out of the range of instrument calibration, a secondary matrix correction was carried out

n.d.: not detected; n.m.: not measured

Table 1 (cont.)
Calculated molar Na/K ratios and the Chemical Indices of Alteration (CIA - Nesbitt and Young 1984) are also tabulated. CIA=$Al_2O_3 \times 100/(Al_2O_3+CaO^*+Na_2O+K_2O)$, where CaO* is silicate-bound calcium, all values are molar

Basin	BK	BK	BK	BK	BK	BK	BK	BK	BK	VP	VP	VP	VP	VP
Sample	TAT-4	ZBC-1	KOZ-2	RJA-1	TAT-3	TAT-5	KOZ-1	KOZ-3	ZJL-1	BAN-2	UST-2	PDN-1	HRU-1	NNS-1
Grain size (ϕ)	1.5-3.5	1.5-3.5	3-3.5	2.5-3.5	2.5-3.5	2-3.5	3-4	3-4	3.5-4	1.5-3.5	2-4	2.5-3.5	2.5-4	4.5-5
Rock type	la	la	la	la	la	la	la	la	la	la	lw	la	lw	mr
Remark		ms		ms		ms								mc
SiO_2 (wt%)	81.66	68.33	66.29	71.54	67.70	81.50	57.13	57.18	53.80	69.41	65.13	70.63	64.12	35.23
TiO_2	0.67	0.50	0.48	0.37	0.45	0.67	0.45	0.50	0.51	0.47	0.49	0.73	0.49	0.40
Al_2O_3	8.59	6.92	8.80	6.29	6.91	8.21	6.85	7.67	6.87	8.09	6.54	12.49	7.20	8.17
$Fe_2O_{3\ tot}$	3.08	2.67	3.23	2.09	2.57	3.27	3.59	3.79	3.75	3.31	3.27	6.77	3.09	7.02
MnO	0.06	0.12	0.11	0.08	0.11	0.08	0.22	0.10	0.23	0.36	0.12	0.08	0.20	0.46
MgO	1.49	1.04	1.15	0.87	1.10	1.17	1.88	1.12	2.65	1.26	0.92	2.19	1.95	2.57
CaO	0.19	9.05	8.60	8.42	9.32	0.09	13.82	13.65	14.76	7.25	11.20	0.45	10.32	22.76
Na_2O	1.75	1.53	1.85	1.49	1.55	1.18	1.17	1.02	1.50	1.84	0.77	2.07	1.53	0.45
K_2O	1.21	1.19	1.21	0.91	1.17	1.28	1.04	1.20	0.77	1.04	1.01	1.37	1.10	1.18
P_2O_5	0.09	0.08	0.09	0.06	0.07	0.06	0.08	0.09	0.09	0.08	0.10	0.13	0.08	0.26
LOI	2.18	8.46	8.46	7.80	8.73	2.36	13.76	12.90	14.87	7.84	10.54	3.11	10.17	21.37
Sc (ppm)	7.40	n.m.	7.20	5.30	n.m	n.m	n.m	n.m	6.70	n.m.	n.m.	n.m.	n.m.	n.m.
V	56.9	37.3	53.3	39.1	37.8	56.5	47.6	67.2	51.1	48.7	59.5	92.3	44.4	86.2
Cr	531.9	288.3	141.2	193.5	240.5	538.9	173.9	225.1	437.3	187.6	130.5	222.4	245.8	87.9
Ni	120.0	55.0	52.7	38.4	48.5	135.7	67.5	75.3	109.9	62.6	30.8	191.6	72.4	76.3
Co	15.5	8.2	12.5	7.8	7.6	14.3	15.0	16.8	14.4	14.5	14.5	27.8	13.6	14.3
Zn	29.3	30.9	36.8	19.7	25.0	31.6	41.2	57.3	37.6	30.5	60.7	77.7	31.3	80.6
Rb	46.1	42.5	50.1	34.4	43.0	47.2	46.1	55.1	34.2	43.5	46.2	59.5	43.5	58.6
Sr	42.6	138.8	150.4	144.0	189.4	32.9	226.3	229.1	209.1	161.6	174.5	74.9	191.7	585.6
Y	28.5	23.2	19.7	18.0	21.2	21.7	19.7	23.6	21.0	20.8	20.3	30.1	18.7	19.1
Zr	256.8	154.1	124.4	129.1	161.0	229.2	123.8	165.0	139.1	144.8	111.9	134.8	143.3	73.5
Nb	9.3	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	12.9	n.d.	n.d.
Ba	176.4	179.2	135.4	111.9	184.3	192.5	135.7	116.9	104.3	184.7	143.4	222.8	147.6	118.3
La	29.1	27.1	27.2	27.5	26.4	23.1	29.3	32.3	22.7	54.2	45.5	51.3	46.2	86.5
Ce	41.0	n.d.	33.0	n.d.	28.5	27.6	26.4	40.6	n.d.	15.5	n.d.	29.6	19.6	24.8
Nd	24.0	32.4	18.0	19.2	22.5	23.6	19.4	21.4	17.8	20.9	18.8	27.3	21.2	22.8
Sm	3.5	3.2	2.4	n.d.	2.5	5.1	3.8	2.6	2.8	4.0	2.9	4.9	2.7	4.3
Eu	0.4	0.6	0.6	0.4	0.7	0.5	0.9	0.8	0.8	0.9	0.7	0.7	0.8	1.9
Yb	2.1	1.9	1.6	1.4	1.6	1.6	1.6	1.8	1.7	1.6	1.7	2.8	1.5	1.9
Pb	12.6	9.0	9.4	8.7	n.d.	15.1	14.0	11.7	12.9	13.8	16.6	11.1	11.4	27.1
Th	8.1	3.4	6.4	2.2	5.1	7.7	6.1	6.4	5.8	7.1	3.3	5.9	n.d.	8.0
U	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Total (wt%)	101.12	100.00	100.35	100.00	99.79	100.00	100.08	99.31	99.92	101.06	100.17	100.13	100.34	100.00
CIA	66.3	63.5	66.0	63.5	63.3	70.1	67.9	70.9	66.4	65.1	72.0	71.1	64.9	78.8
molar Na/K	2.20	1.95	2.32	2.48	2.01	1.40	1.71	1.29	2.96	2.69	1.16	2.30	2.11	0.58

la: lithic arenite; lw: lithic wacke; mr: mudrock
mc, ms: on raw XRF data of samples with unusually high CaO or SiO_2 contents, out of the range of instrument calibration, a secondary matrix correction was carried out
n.d.: not detected; n.m.: not measured

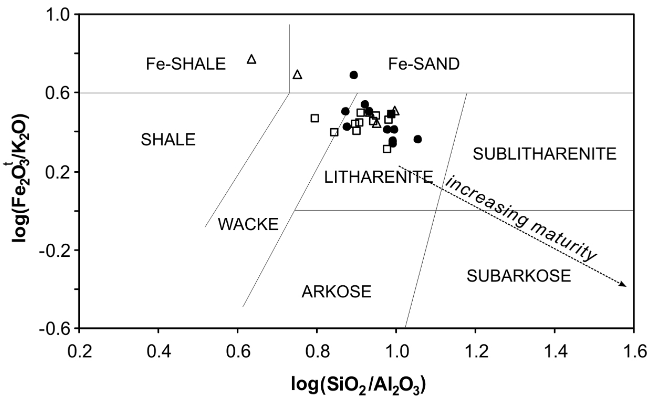

Fig. 3
Classification of the flysch sandstone based on major element compositions, using the scheme of Herron (1988), showing litharenite composition for most samples. In agreement with the petrographic data there is no indication for mineralogical maturation processes (arrow) during sand development, which can imply relatively rapid erosion and short sediment transport. Symbols as in Fig. 1

Sandstone classification

According to the major element compositions, most of the flysch sandstone is classified as litharenite (Fig. 3). The SiO_2/Al_2O_3 values reflect a moderate mineralogical maturity. Most molar Na/K ratios fall between 0.5 and 1.5 (Table 1), with the coarser-grained Brkini samples having higher average values, in agreement with the slight dominance of sodic plagioclase among the feldspars. The lower and largely uniform Na/K ratios in Istria of ca. 1.1 are likely to correspond to a higher amount of illite in the finer-grained sandstone. The elevated Fe_2O_3t/K_2O values point to contribution of mafic minerals in addition to feldspars, and corroborate the low textural maturity of the sandstone. On the other hand, the inferred K_2O mobilization during sandstone development (see below) is also an important factor, which could lead to the observed ratios.

These results do not confirm the petrographic observations insofar that optically quartz is the dominant framework component. This is because a significant part of unstable framework components is transformed into "pseudomatrix" upon diagenesis. Consequently, the difference in mobility of e.g. Na^+ and K^+ during diagenesis may have influenced the bulk compositions.

Major elements

The composition of sediments deposited in a specific tectonic environment dominated by igneous, metamorphic or sedimentary rock suites tends to reflect the particular major element composition of source rocks in that setting. We have adopted the method of Bhatia (1983) who outlined four general, distinct tectonic provenance groups. The derived sediments in the adjacent basins are well separated both chemically and petrographically: (1) oceanic island arc: tholeiitic to calc-alkaline mafics, yielding highly immature volcanogenic sandstone; (2) continental island arc: felsic/intermediate volcanics, represented by volcanogenic lithic greywacke; (3) active continental margin: gneiss and felsic igneous rocks (both volcanic and plutonic) of an uplifted crystalline basement made up of slices of an older orogen, and represented by greywacke, as well as (4) passive continental margin: highly mature quartzose sediments derived from metamorphics and by recycling of older sedimentary units. Sediments of this provenance group are mainly sublitharenites and match the recycled orogen provenance of Dickinson (1985).

Discrimination is performed by means of a so-called territorial plot where for each sample two different linear combinations of the major element data are plotted in a diagram having predefined fields (Bhatia 1983). The calculated discriminant scores for the flysch sandstone are shown in Fig. 4. All samples fall in the passive margin field indicative of the predominance of metamorphic and older sedimentary rock suites in the hinterland. However, the point cluster is elongate in shape and points toward the active continental margin field. Taking

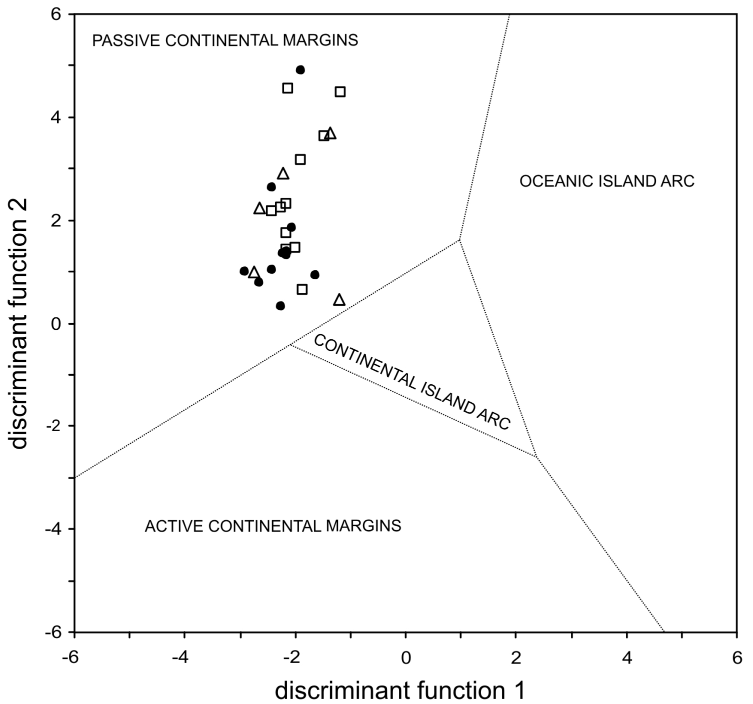

Fig. 4
Plot of discriminant scores for the flysch sandstone, calculated from their major-element compositions. Symbols as in Fig. 1. The plot is subdivided into fundamental tectonic settings. All samples plot in the compositional field typical of passive margin settings. See text for discussion. Discriminant function 1 = $-0.045[SiO_2]-0.972[TiO_2]+0.008[Al_2O_3]-0.267[Fe_2O_3]+0.208[FeO]-3.082[MnO]+0.140[MgO]+0.195[CaO]+0.719[Na_2O]-0.032[K_2O]+7.510[P_2O_5]+0.303$. Discriminant function 2 = $-0.421[SiO_2]+1.988[TiO_2]-0.526[Al_2O_3]-0.551[Fe_2O_3]-1.610[FeO]+2.720[MnO]+0.881[MgO]-0.907[CaO]-0.177[Na_2O]-1.840[K_2O]+7.244[P_2O_5]+43.570$. Discrimination procedure after Bhatia (1983)

into account that the discriminant scores would only plot astride the boundary of two areas if a nearly 1:1 mixing of two contrasting sediment types occurred, it is very probable that the predominant felsic metamorphic and recycled sedimentary detritus was mixed with minor amounts of volcanic, more mafic material. No clear-cut trend exists in the major element compositions in the Istrian, Brkini and Vipava Basins (except for CaO and LOI – see Fig. 8). However, the center of the Brkini data lies closer to the active continental margin field. If this pattern is meaningful, it may reflect that sedimentation in the Brkini Basin was more influenced by igneous rocks, which normally prevail in active continental margin settings.

Trace elements

Using ratios of immobile trace elements has many advantages; they appropriately reflect the ratios within the source rock (e.g. Bhatia and Crook 1986; Floyd and Leveridge 1987) and they circumvent the problem of varying element abundances due to dilution effects, f.i. by extrabasinal carbonate (see Rollinson 1992). Two-source mixing can be modeled using mixing curves (Dinelli et al. 1999), but characterizing the interplay of several sources is difficult.

As for the Istrian, Brkini and Vipava Basins, the ferrous trace element ratios Cr/V vs. Y/Ni provide evidence for small components of ultrabasic units in the ultimate source (Fig. 5). Such ratios are useful in identifying an ophiolite source and in tracing its proximity (McLennan et al. 1993). The ratio Cr/V is a measure of Cr enrichment over the general level of ferrous elements. Cr is concentrated in Cr-spinel, a key mineral in ophiolite, whereas Y is a proxy for heavy REE, typically hosted by zircon and garnet. Ferromagnesian element abundance is expressed by the V and Ni contents and tends to be high in mafic-ultramafic

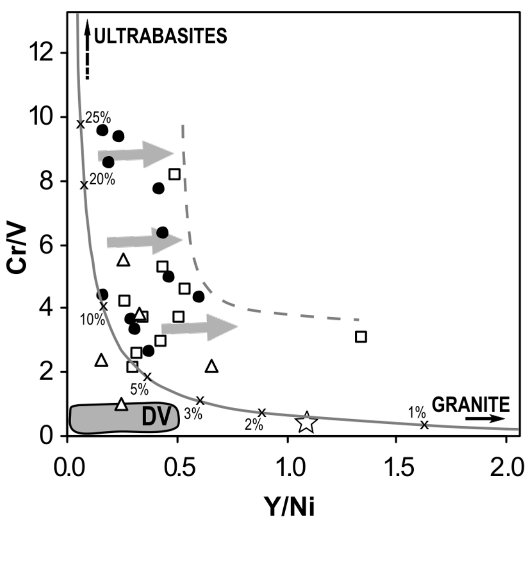

Fig. 5
Cr/V versus Y/Ni plot (McLennan et al. 1993) for the flysch sandstone. Also shown is a mixing line of ultrabasic (Cr/V = 45; Y/Ni = 0.001) and granitic (Cr/V = 0.093; Y/Ni = 8.889) rocks (Turekian and Wedepohl 1961; Dinelli et al. 1999). Percentages show the extent of ultrabasic addition to the mixture. Asterisk: average upper continental crust composition (Taylor and McLennan 1985). DV: typical compositions of mafics and related amphibolite of the Dinaride Ophiolite Belt and the Vardar Zone (Pamić et al. 2002). Bold arrows: shift of the plots from the granitic-ultrabasic mixing curve, possibly due to the incorporation of an Y-rich component. Dashed line: inferred position of the upper limit of this addition

sources. The Brkini Basin probably received slightly more ophiolitic detritus than the Istrian and Vipava Basins as deduced from its generally higher Cr/V values (Fig. 5). Overall the amount of the ultrabasic component within the sediment is estimated to be in the range of 5–25%. It can be also seen that there is an apparent shift of the ratio-ratio plots from the theoretical granitic-ultrabasic mixing curve of Dinelli et al. (1999). This may suggest that felsic igneous detritus was intermingled not only with ultrabasic material but also with a substantial fraction of clastics from a source of different composition. Although an input of basic rocks is possible, as supported by the presence of basic lithic fragments, incorporation of Y-rich rocks into erosion such as garnet-bearing micaschist or felsic crystalline rocks richer in zircon may result in elevated Y/Ni ratios, which is in agreement with the high garnet and zircon content of the heavy mineral spectra (Mikes 2003). Zircon enrichment due to sediment reworking is improbable, as Zr/Sc ratios suggest primary crystalline, rather than reworked, sources for zircon (to be discussed later).

As shown above, major element data suggest a passive continental margin tectonic environment for the flysch sandstone, but trace elements indicate more mafic contribution as well. Another adequate way to achieve tectonic discrimination is using a full range of elemental composition, which sensitively indicates various mafic and heavy mineral inputs within a sedimentary suite (Floyd et al. 1991). Figure 6 shows the upper continental crust-normalized (UCC) element distributions of the flysch sandstone, with elements arranged from left to right in the order of decreasing abundance in the UCC (Condie 1993). Average UCC-normalized values from the Istrian, Brkini and Vipava Basins show a very similar pattern, except for differences in the abundance of some REE (La, Yb), Co and Pb. The high Ba concentration in Istria probably reflects fine crystalline, sedimentary barite that does not occur in the Brkini and Vipava Basins. The REE contents exhibit poorly fractionated normalized LREE/HREE ratios (La_N/Yb_N=1.1 for Brkini, 2.3 for Vipava and 4.2 for Istria) showing that REE distribution is largely controlled by the rocks comprised by an average upper continental crust. Normalizing the REE to the primitive mantle; the according ratios are 10.9; 22.3; 40.6, respectively. This means that contribution from continental crust clearly predominate over mantle components.

In keeping with the low Y/Ni and elevated Cr/V ratios discussed above, strong positive Cr-Ni anomalies indicate high mafic input. Only the passive margin settings would reveal negative anomalies, i.e. normalized Cr and Ni values <1 (Floyd et al. 1991). A strong Ti-Zr-Y positive and strong V negative anomaly, as would be expected for a passive margin, is not signaled; these elements either correspond to, or are slightly depleted relative to the average upper continental crust.

In samples where Nb determination was possible, strong negative Nb anomalies were observed. The extent of this anomaly can be measured by the Nb/Nb* ratio $\{=Nb_N/[(Ni_N+Ti_N) \times 0.5]\}$ (Floyd et al. 1991). These low values

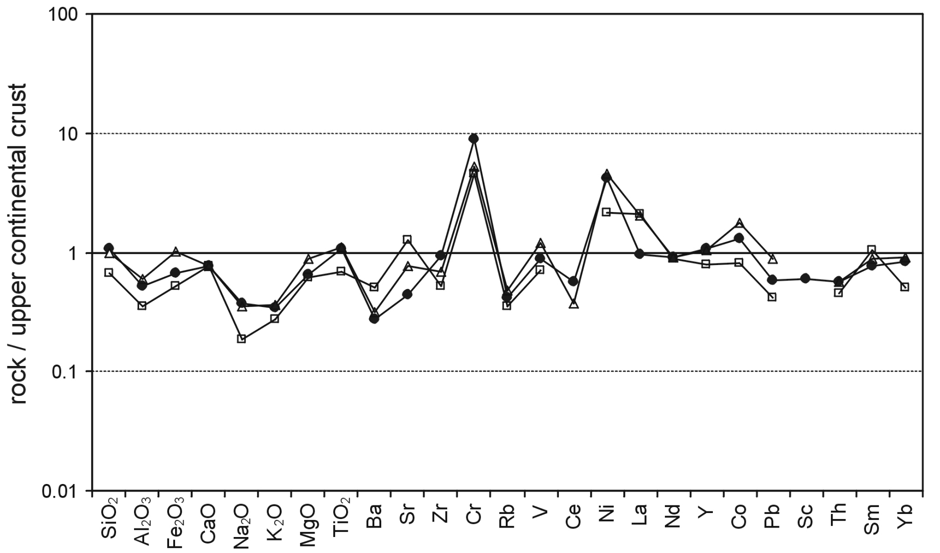

Fig. 6
Upper continental crust-normalized element distributions in the flysch sandstone. Symbols as in Fig. 1. Average analyses from the three basins are plotted. The positive anomalies of Cr and Ni indicate mafic-ultramafic input. Positive Ti-Zr anomalies, characteristic for sediment sources originating largely from passive margins, are not detected. Note: inspection of individual plots has shown that the scatter of those data is small and that they do not decline much from the general pattern of the average plots. Normalization values are from Taylor and McLennan (1985). Elements are arranged from left to right in the approximate order of decreasing abundance in the upper continental crust (see Condie 1993). Ce from Istria samples is not shown as none was detected; Sc analyses are only available from four Brkini samples

(0.13 for Brkini and 0.15 for Vipava) are a typical phenomenon with sediment sources involving subduction-related magmatic rocks. The degree of the anomaly is usually much less (ca. 0.5) for passive margins consisting of old, partly reworked continental crust (Floyd et al. 1991).

The spider diagram of Fig. 6 also reveals that soluble, mobile elements (Na, K, Rb) are slightly depleted in the flysch sandstone.

Finally, data plots in the Ti/Zr vs. La/Sc field confirm that the overall source composition closely resembles the average (granodioritic) UCC, most typical of an active continental margin (Fig. 7).

Effects of weathering and post-erosional processes

Given the many processes acting within the sedimentary cycle and the resulting element mobility that could have exerted control on sandstone composition, it is necessary to examine them so as to check the validity of the above provenance interpretations.

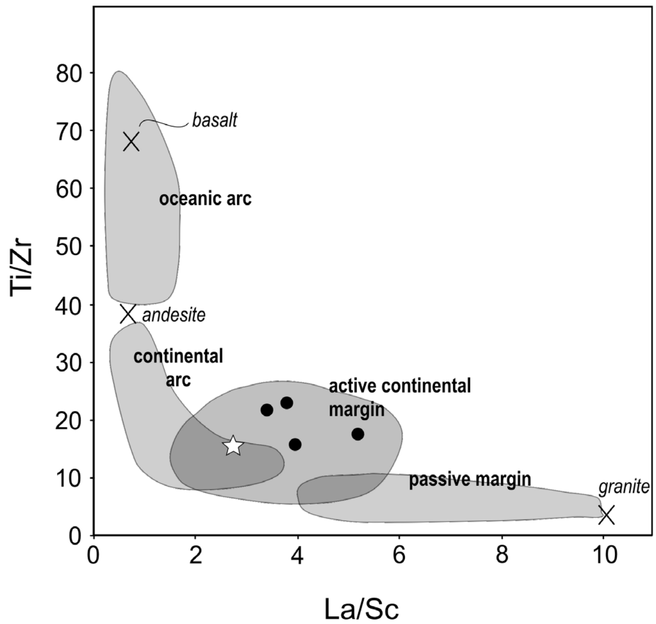

Fig. 7
Ti/Zr vs. La/Sc plot for selected flysch sandstone samples of the Brkini Basin. The values fall in the field of active continental margin compositions, strongly suggesting a granodioritic bulk composition of the various sediment sources of the flysch. Discrimination scheme from Bhatia and Crook (1986), asterisk shows average UCC composition (Taylor and McLennan 1985)

Effect of carbonate addition

A striking feature of all samples is that they contain high and variable amounts of CaO (Fig. 8a, Table 1). These are generally higher in the more distal and finer-grained Istrian Basin (24–40 wt%) and lower in the proximal, coarser-grained Brkini Basin (0–15 wt%) samples. Overall, the CaO wt% reveals an excellent positive correlation with LOI values, suggesting that calcite dilutes the siliciclastic material to various degrees (Fig. 8a). There is no petrographic indication for

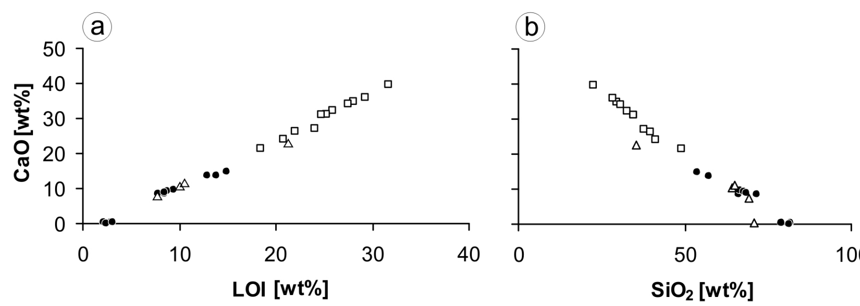

Fig. 8a, b
Bivariate plots of CaO, SiO_2 and loss on ignition (LOI) values. Symbols as in Fig. 1. The linear correlations correspond to the admixture of calcium carbonate to the siliciclastic detritus to extremely varying degrees. Note the consistently higher carbonate contents in Istria

diagenetic growth of calcite in the pore space. Calcite veins may be present in the sandstone, but such sample volumes were readily excluded from analysis. The calcite dilution is reflected by the SiO_2 content as well, which is negatively correlated with CaO (Fig. 8b). Micas, clays and chlorite can be accounted for small deviations from the linear trend of LOI vs. CaO.

CaO addition could be explained by admixture of fine-grained, detrital carbonate although the Brkini samples lack sand-sized carbonate fragments. A diagenetic origin of the calcite is also possible: upon burial, the underlying thick Adriatic Carbonate Platform sediments, or even the intercalated beds of redeposited carbonate could have released sufficient calcium.

Hydraulic and weathering effects

The Fe_2O_3/K2O vs. SiO_2/Al_2O_3 ratios (Fig. 3), together with the general angular to subangular shape of framework and heavy mineral grains, imply short sediment transport distances prior to funneling into the flysch basin. The low textural maturity is not readily seen in the SiO_2/Al_2O_3 ratios, reflecting post-depositional breakdown of the most labile framework components. The arrow in Fig. 3 indicates the trend of increasing sediment maturity, which can integrate weathering (preferential removal of Fe_2O_3-bearing lithics and relative enrichment of quartz) and subsequent hydraulic effects (sorting, winnowing), leading to decreased Fe_2O_3/K_2O and increased SiO_2/Al_2O_3 ratios. Clearly the flysch sandstone was not affected by significant sediment maturation processes.

The relationship of silicate-bound CaO and alkalic elements (K_2O, Na_2O) to Al_2O_3 can be also used to assess the degree of source weathering. In Fig. 9 the

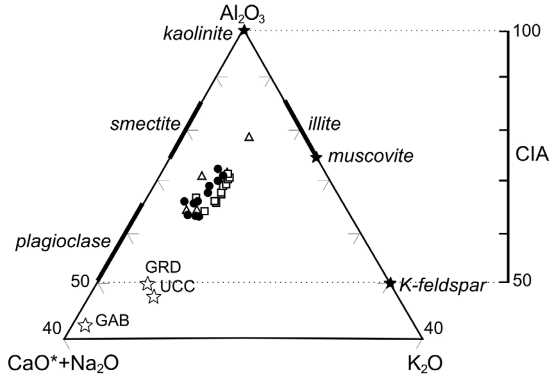

Fig. 9
A-CN-K plot with molar values, indicating moderate Chemical Index of Alteration values (CIA; Nesbitt and Young 1984) for the flysch sandstone, ranging between 0.64 and 0.72. Symbols as in Fig. 1. CaO* is silicate-bound CaO. In spite of the strong masking effect of the carbonate dilution which hampers the precise determination of CaO*, it is necessary to account for it. Thus, based on petrography and whole-rock mass-balance considerations, we have chosen an approach which assumes that CaO* represents 10% of the total Na_2O content. We believe that errors associated with this realistic estimation do not significantly influence the CIA values. Together with the low textural maturity of the sandstone, these indices are most consistent with a negligible weathering in the source or during alluvial storage and thus imply relatively rapid sediment transport. The linear alteration trend toward illite most probably results from slight differences in stages of diagenetic alteration of the sandstone rather than from source area weathering. Asterisks show average UCC composition (Taylor and McLennan 1985) and average granodiorite (GRD) and gabbro (GAB) compositions (Le Maitre 1976)

data exhibit moderate CIA (Chemical Index of Alteration; Nesbitt and Young 1984) values (64–72) but plot away from the average, fresh UCC (granodioritic) composition and suggest a trend towards low-K illite. Furthermore, the Ypresian to Lower Lutetian Vipava and Brkini samples plot closer to the A–CN edge than the mostly Bartonian ones from Istria. The origin of a tentative weathering trend line of the former would lie somewhat closer to more basic compositions, represented on the plot by the average gabbro of Le Maitre (1976). The importance of mafic (possibly also ultramafic) lithologies compared to exposed felsic units could, therefore, be higher in the catchment area of the Vipava and Brkini Basins until Early Lutetian, and have decreased during the later stages of Mid-Eocene, as recorded by the Istrian samples.

Although paleoclimate is an important factor, intense weathering mostly requires effective, long-lasting soil development in a transport-limited system (Johnsson 1993). Therefore, taking into account the low textural maturity, the inferred proximity of the basin to higher-relief source areas, and the fact that most of the mineral reactions also take place during diagenesis, we assume that weathering was not important in influencing any of the sandstone compositions. Rather, diagenetic reactions can probably account for the development of the observed trends, and incorporation of smaller amounts of older sediments having a different weathering history could have also been possible.

Cr/Ni ratios are useful in tackling problems of reworking of ophiolitic components (von Eynatten 2003). Figure 10 indicates the Cr/Ni values which rarely exceed 5. This range is somewhat higher than the compositional range of ultramafic rocks in ophiolite but conforms quite well to that of the associated

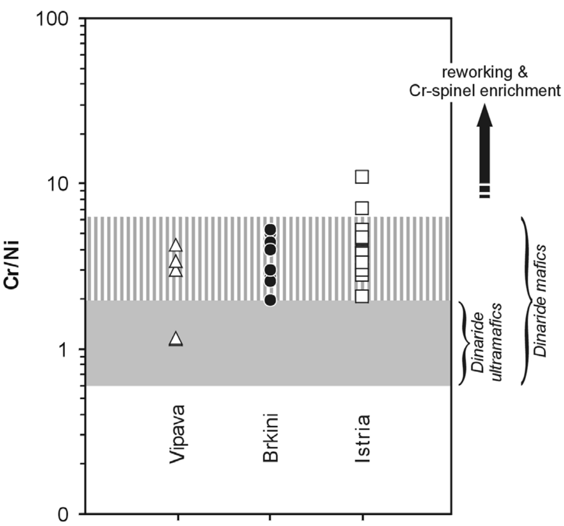

Fig. 10
Synoptic presentation of the Cr/Ni values from the flysch sandstones. This ratio in the parent rocks is retained in the immediately derived sediments, whereas recycling gives rise to Cr-spinel enrichment and depletion of mafic lithics, leading to considerably higher Cr/Ni ratios. The observed sandstone Cr/Ni values outside the ultramafic range but within the mafic field show that neither the contribution of ultramafic lithic fragments alone, nor enrichment of Cr-spinel due to reworking played an important role. A mixed contribution of ultramafic fragments, Cr-spinel, and mafic (basic) magmatic rocks is well in line with the analytical and our other data (von Eynatten 2003). Petrologic data are from Lugović et al (1991), Robertson and Karamata (1994) and Pamić et al. (2002)

mafic rocks. In fact, the amount of mafic magmatic source components is small compared to the ultramafic rocks, as indicated by detrital Cr-spinel chemistry (Mikes 2003). The Cr/Ni ratios, outside the ultramafic range but within the mafic field, could imply that neither ultramafic lithic fragments alone nor enrichment of Cr-spinel due to reworking played an important role. However, a mixed contribution of ultramafic fragments, Cr-spinel, and mafic (basic) lithoclasts is well in line with the analytical and our other data (von Eynatten 2003). The difference in the degree of the UCC-normalized anomalies of Cr and Ni is also in agreement with a mixing of the above source components.

Finally, the Th/Sc vs. Zr/Sc plot was used to assess the degree of sorting and/or reworking of felsic, crustal material (McLennan et al. 1993). Figure 11 illustrates that the samples plot on the igneous differentiation trend, close to the UCC composition, and there are no signs of significant heavy mineral concentration due to zircon enrichment. The lack or very low amount of subrounded/rounded quartz and heavy mineral grains in the sandstones agree well with the Th/Sc and Zr/Sc values. Thus, recycling of older, mature sediments of felsic, crustal origin probably did not play a significant role.

Conclusions

(1) Geochemically, turbiditic sandstone of the Istrian, Brkini and Vipava Basins is classified as lithic arenite. Framework constituents reveal chiefly low-grade metamorphic and acidic plutonic sources. To a lesser degree, basic volcanics and

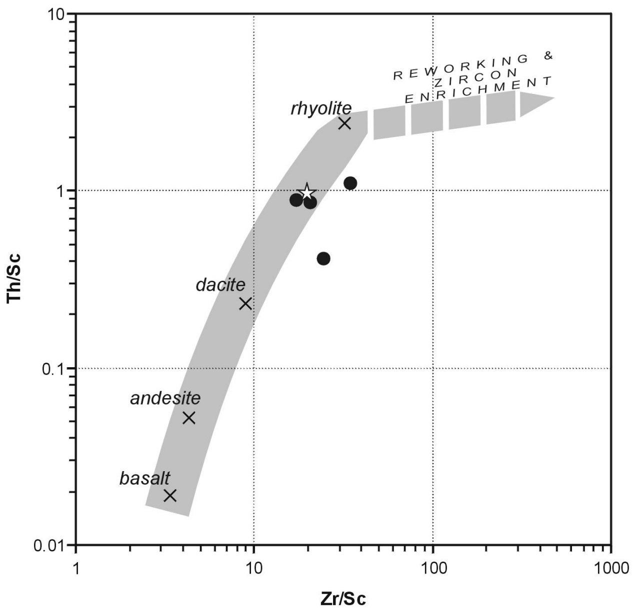

Fig. 11
Th/Sc vs. Zr/Sc plot showing a magmatic arc trend (McLennan et al. 1993; Willan 2003). Sediments derived from (meta)magmatic units plot close to this trend line whereas reworking, that commonly involves zircon enrichment, causes an abrupt shift toward elevated Zr/Sc ratios. Data points of the flysch sandstone from the Brkini Basin fall near the trend, close to the UCC composition (asterisk), and thus preclude significant reworking of felsic, continental source components

ultrabasic sources were also eroded. Small, recycled sedimentary source components can be detected as well.

(2) Calcite of detrital and/or diagenetic origin dilutes the siliciclastic portions to various degrees (0–40 wt% CaO); the Istrian sandstone is more carbonatic than the Brkini sandstone. Effects to modify sandstone composition other than diagenesis, such as weathering and sorting, were probably negligible.

(3) Main element compositional data clearly indicate the predominance of a felsic (meta)igneous, and a subordinate, unspecified, more mafic provenance.

(4) Trace element variations and trace element ratios can be successfully used to refine the nature of mafic components in the source area. The Cr/V vs. Y/Ni plot reveals that in addition to ca. 5–25% ultramafic material, detritus of mafics, and probably small amounts of recycled sedimentary rocks also intermingles with the granitoid-derived sediment. Felsic source lithologies are dominant and the overall contribution by mafic-ultramafic components is comparatively small. Detritus shed in the Brkini Basin during the Early Eocene is slightly richer in mafic-ultramafic components than for the Middle Eocene in Istria.

(5) Tectonic discrimination using major elements alone suggests a passive margin setting. However, this confronts upper continental crust-normalized element distribution, which clearly shows the presence of mafic, probably subduction-related material, not typical for a passive margin. In addition, the Ti/Zr-La/Sc plot indicates deposition at an active continental margin. The contrasting results can be reconciled by assuming that the source types of the sediments are not closely related to the tectonic setting of the studied basins. There is a striking discrepancy between the passive margin setting implied by the main element discrimination techniques used (Bhatia 1983) and the foreland situation argued by Dimitrijević (1974), Marinčić (1981), Pamić et al. (1998) and Tari (2002). In fact, in front of the Dinaride complex orogenic thrust wedge, sediments derived from individual slices of crystalline basement, dismembered ophiolite, and older sediments may have mixed.

(6) The data presented herein show only little variations over larger distances and among different basins and therefore do not support the idea of a combined Alpine-Dinaride provenance, which has arisen from the largely bimodal paleocurrent directions alone (Magdalenić 1972; Orehek 1991). As for the ultrabasic source components, derivation from the Penninic ophiolite of the Eastern Alps is improbable because these formations in the Tauern Window were not yet exhumed in the Eocene (Frisch et al. 2000). A likely source candidate is the Jurassic ophiolitic mélange in the NW Dinarides and research is in progress to further test this hypothesis by means of chemistry of key heavy minerals.

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