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Impact of Geomorphological Variables in Weighing the Lithological Influence on Geochemical Composition of Stream and Overbank Sediments: A Regression Model for the Žumberak Area (NW Croatia)

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Key words: Drainage basin, Channel–reach morphology, Sediment supply, Lithology, Geochemical composition, Stream sediment, Overbank sediment, Multiple regression analysis, Žumberak, Croatia.

Abstract

The two multiple regression models were compared on the assumption that the geochemical composition of the alluvial material is basically originally derived from the bedrock lithology of a drainage basin of the Žumberak Area (NW Croatia). The latter was integrated with both stream sediment and overbank sediment geochemical data via the two essentially different approaches as regards the catchment basin geomorphological data: (1) as the relative area of influence representing a portion of the catchment basin occupied by a certain rock type; (2) as the relative "line" of influence representing a narrow tract of a certain rock type traversed by the majority of perennial streams which form the active stream network. The model comparison was accomplished on the basis of the goodness-of-fit test for both experimental designs and for the same set of data. The most important result of these experiments highlights the linear approach as the more appropriate model for ascertaining the lithological influence on the analyzed sample media. This can be explained by the stronger erosive and transportational power of the water which flows in the well defined channel network in contrast to the unconcentrated surface runoff in the interfluvial area, coupled with the buffering effects of rich slope and riparian vegetation, which is characteristic of the Žumberak heartland.

1. INTRODUCTION

Much research has been undertaken to elucidate the problem of the relationship between drainage basin lithology and geochemistry of the drainage system deposits, whether the latter be confined strictly to the stream channel, or spread over the extended area beyond its banks. In both cases attention was largely concentrated on evaluating the geochemical background and detecting the elevated values which reflect possible mineralization in a survey area (e.g. ROSE et al., 1970; BONHAM-CARTER et al., 1987), as well as on modelling the geochemical variability and dilution effects (e.g. JENSON & TRAUTWEIN, 1984; HAWKES, 1976; BONHAM-CARTER & GOODFELLOW, 1984; BONHAM-CARTER et al., 1987; CARRANZA & HALE, 1997). Regression analysis has been utilized as a particularly useful mathematical method in relating the desired chemical element to the mapped lithological units. This aptitude is most assuredly rooted in the broad agreement that compositional (geochemical, mineral) and other properties of drainage basin fills are inseparably connected with drainage basin evolution through the delicate process–response relationship and feedback mechanisms in alluvial systems (e.g. JOHNSSON, 1993; WELTJE et al., 1998). In all the above cases however, regression models invariably made use of the areal proportions of a particular lithology within a catchment area putting these in the role of independent variables. This concept simply disregards the fact that not all parts of the drainage basin supply its valley and channel floors with products of weathering and erosion uniformly in space and time. In scaling the eroding and resisting forces in drainage basin systems many parameters interact, causing the sediment yield to depend not only on the physiographic properties of the rocks but also on the character of the forces of denudation. These, in turn, operate on different temporal and spatial scales with regard to slopes and channel–reach areas. Denudation, as a total sum of erosion processes, assumes that sediment is derived in equal portions from all subareas of the watershed, but it tells little about the redistribution of weathered and eroded material within a drainage basin (RITTER, 1978). It is also well known that only the erosive power of water flowing in clearly defined channels represents a truly effective transportation and erodive agent, while, on the other hand, unconcentrated overland flow on slopes acts more slowly and less efficiently, particularly in the presence of rich vegetation cover (KIRKBY, 1969). Thus, the two basin subsystems seem to operate independently and out-of-phase, which is the reason why the understanding of their process–form relationship becomes fraught with difficulties. Their co-dependence during historical times (<200 yrs; RITTER, 1988) can be observed almost solely in a narrow belt of slope adjacent to the stream channel, particularly in its upstream reaches. This is where incising streams erode both vertically and laterally, affecting not only the channel floor but also the lower

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portions of the valley sides. Often they undercut channel slopes, particularly during flood events, inducing rapid mass-movements as a result (usually bedrock and regolith landslides). In contrast, slopes that lie higher up the hillside, in the vicinity of the watershed, rarely experience such energetic geomorphologic events which, together with the mitigating effects of dense slope and riparian vegetation, may effectively reduce the amount of regolith reaching the channel (OWENS et al., 1999). Therefore eroded material, which is transported in various forms of stream-channel load and deposited downstream as stream or overbank (floodplain) sediment, seldom evenly reflects the lithological characteristics of both drainage basin subsystems either in geochemical or mineral composition. Moreover, wider valleys with alluvium infill in the downstream reaches of a channel usually serve as a buffer zone, considerably decreasing their susceptibility to disturbance from slope processes (MONTGOMERY & BUFFINGTON, 1997).

The question arises how to best fit geomorphological variables into the scheme of assessing the real lithological imprint on the geochemical composition of stream and overbank sediments, especially when considering that interfluvial areas occupy a vastly greater proportion of the catchment area. The areal approach that utilizes the relative proportions of distinctive lithology within a drainage basin takes into account inappropriately large portions of the whole catchment characterized by very slow denudation processes. These, in turn, provide the valley floors with relatively small quantities of waste material over a short time interval (geomorphological steady time). In this case, accumulation of the sediment load in channels appears to be quite out of phase with erosion and transportation processes on far reaches of the upper slopes in the vicinity of the watershed where almost no erosion occurs. We are prone to use the linear approach instead, which takes into account only the section of distinctive lithology along the active stream reaches, expecting a closer relationship between the deposits with pertinent channel-floor or bank lithology.

Thus, the aim and scope of this study is to compare the two approaches by the use of the multiple regression model for quantifying the impact of various types of rocks on the selected set of elements. This was accomplished on the basis of the goodness-of-fit test for two different experimental designs and for the same set of data including geochemical content of stream and overbank sediment samples.

2. GEOLOGICAL FRAMEWORK OF THE STUDY AREA

The study area encompasses a large proportion of the Žumberak region, situated in western Croatia (on the Slovenian border), near the capital, Zagreb. Broadly, it is a mountainous system occupying the interfluvial belt between the Sava, Kupa and Krka rivers, with the Žumberak Mt. to the west, and Samobor Mt. to the east as its main constituent bodies (Fig. 1). The landscape combines many characteristics from the two distinct topographic and geologic provinces: Dinaric carbonate terrains with a relatively poor and undeveloped drainage network, in which a variety of karstic phenomena abound, and Pannonian, mostly non-carbonate, highly dissected terrains.

Geologically, it represents a complex terrain with different tectonic and lithological settings (Fig. 2). Interaction and feedback between tectonics and surface processes produced a heterogeneous massif with a number of major tectonic blocks which have suffered horizontal or vertical movements of various intensity. The greatest proportion of the study area is composed of the autochtonous Žumberak Tectonic Unit which consists of various rocks deposited from the Middle Permian to Palaeogene (ŠIKIĆ et al., 1977). Its western part is an upthrown area which is lithologically relatively monotonous due to the predominance of carbonate rocks – Triassic dolomites and Jurassic limestones – over pelagic siliciclastic rocks of Upper Cretaceous age (PLENIČAR et al., 1975; PREMRU et al., 1977; BUKOVAC et al., 1983; HERAK & BUKOVAC, 1988). In contrast the Samobor Mt. is much more tectonically dissected and lithologically variable, with Permian rocks, predominantly sandstones, in the core of the autochthonous block. Its border with the allochthonous Žumberak–Medvednica Knappe Unit is often fragmented by vertical neotectonic faults. The latter consists of the sedimentary rocks of Triassic, Jurassic and Cretaceous age which were thrust over the older substructure during the Alpine orogenesis, but the present appearance of the horizontally dislocated mass resulted mostly from the subsequent partitioning of individual minor blocks which reduced its original proportions. In effect, both of these units suffered intense fragmentation during the neotectonic phase, particularly since the end of the Upper Pliocene, which, together with surface processes, were an important factor in shaping the modern uplifted territory. In contrast, the southern borders of the study area went through the different tectonic and morphological changes which resulted in a steady tectonic subsidence and final formation of the Karlovac Depression (VELIĆ, 1983).

Lithology

The composite (Dinaric–Pannonian) character of the Žumberak territory is well reflected in the intricate lithological patterns. There is a variety of carbonate and siliciclastic facies combined in a fractured mosaic of blocks of different ages which, for the purpose of this work, is limited to a very simplified and sparse sketch-map (Fig. 2).

Throughout the area the Mesozoic carbonate rocks predominate, with the emphasis on the Middle and Upper Triassic dolomite series. These rocks are mostly massive, with almost no fossil content, which prevents further chronostratigraphic determination (ŠIKIĆ et al.,

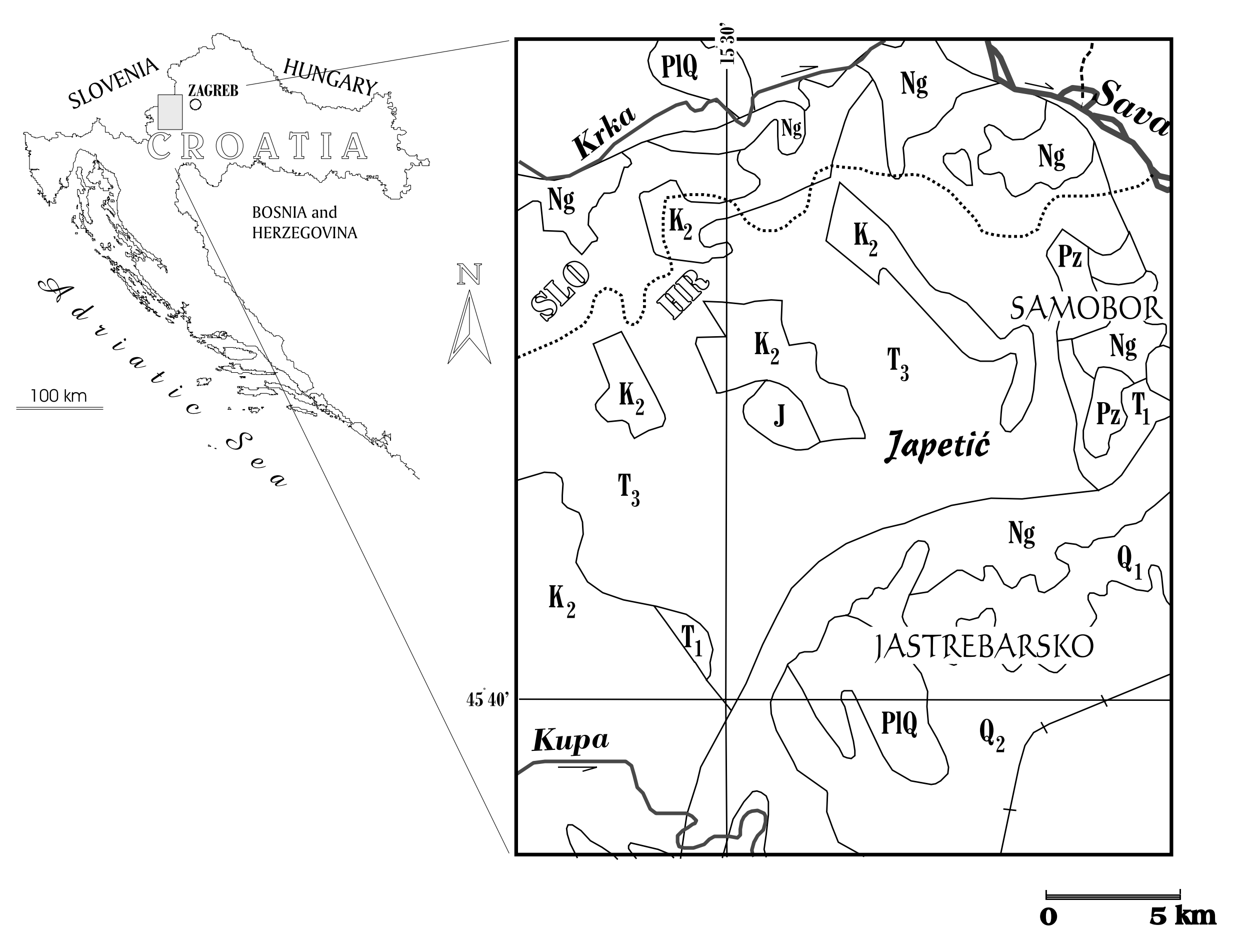

Fig. 1 Generalized geological map showing location of the study area (after ŠIKIĆ et al., 1977; PLENIČAR et al., 1975, and BUKOVAC et al., 1983): Q_2 – Holocene in general; Q_1 – Pleistocene in general; PlQ – Plio–Quaternary (unconsolidated sediments); Ng – Neogene in general (clastic rocks); K_2 – Upper Cretaceous (limestone, dolomites and flysch-like series); J – Jurassic in general (predominantly limestones); T_3 – Upper Triassic (predominantly dolomites); T_1 – Lower Triassic (predominantly clastic rocks); Pz – Middle and Upper Permian (predominantly clastic rocks).

1977). The bulk of the dolomite material, particularly of Upper Triassic age, originated from dolomitization of the calcareous sediments, most probably in the early phase of diagenesis. Later, due to the long lateral transport, the allochthonous mass has become thickly jointed, prone to crushing and easily erodable. Dolomites (Upper Triassic in particular) provide the major supply for both the active stream sediment and alluvial fill in many intermontane valleys. The other member of carbonate lithology is represented by the various types of limestones. In contradistinction to the Triassic monotonous dolomite sequence where stromatolite-type facies is dominant, limestones are distinguished by ample facies diversity throughout the whole stratigraphic column ranging from the Middle Permian to the Miocene. Their vertical and horizontal distribution, though, is thin and locally very confined. Often they are interbedded within the more frequent siliciclastic sedimentary rocks. Only Jurassic limestones can be postulated as representing almost entirely uninterrupted sedimentation, and specifically under pelagic conditions, during the entire period. Their sequence is represented by pelagic (bio)micrites (mostly Liassic–Doger) and (bio)detrital limestones with interbedded cherts (mostly Malmian). Cherts originated from the silicification of calcareous rocks which can be seen in progressive transition from the micritic limestones.

The siliciclastic rocks occupy a considerably lesser portion of the study area. Most generally, they consist of two different sequences of stratified rocks which are temporally widely separated. The older group of sedimentary rocks is bound to an earlier, Permo–Triassic sedimentation cycle. Another sequence comprises younger, mostly Neogene, siliciclastic material which is often unconsolidated or loosely consolidated and thus easily erodable. The oldest rocks are of Middle and Upper Permian age, developed in a molasse-type facies. These are very diverse, but represent mostly sandstones and, more rarely, (quartz)conglomerates. Shales and siltites occur only as intercalations between the former. Associated with them are rare occurrences of copper and iron ore, and also, towards the Triassic boundary,

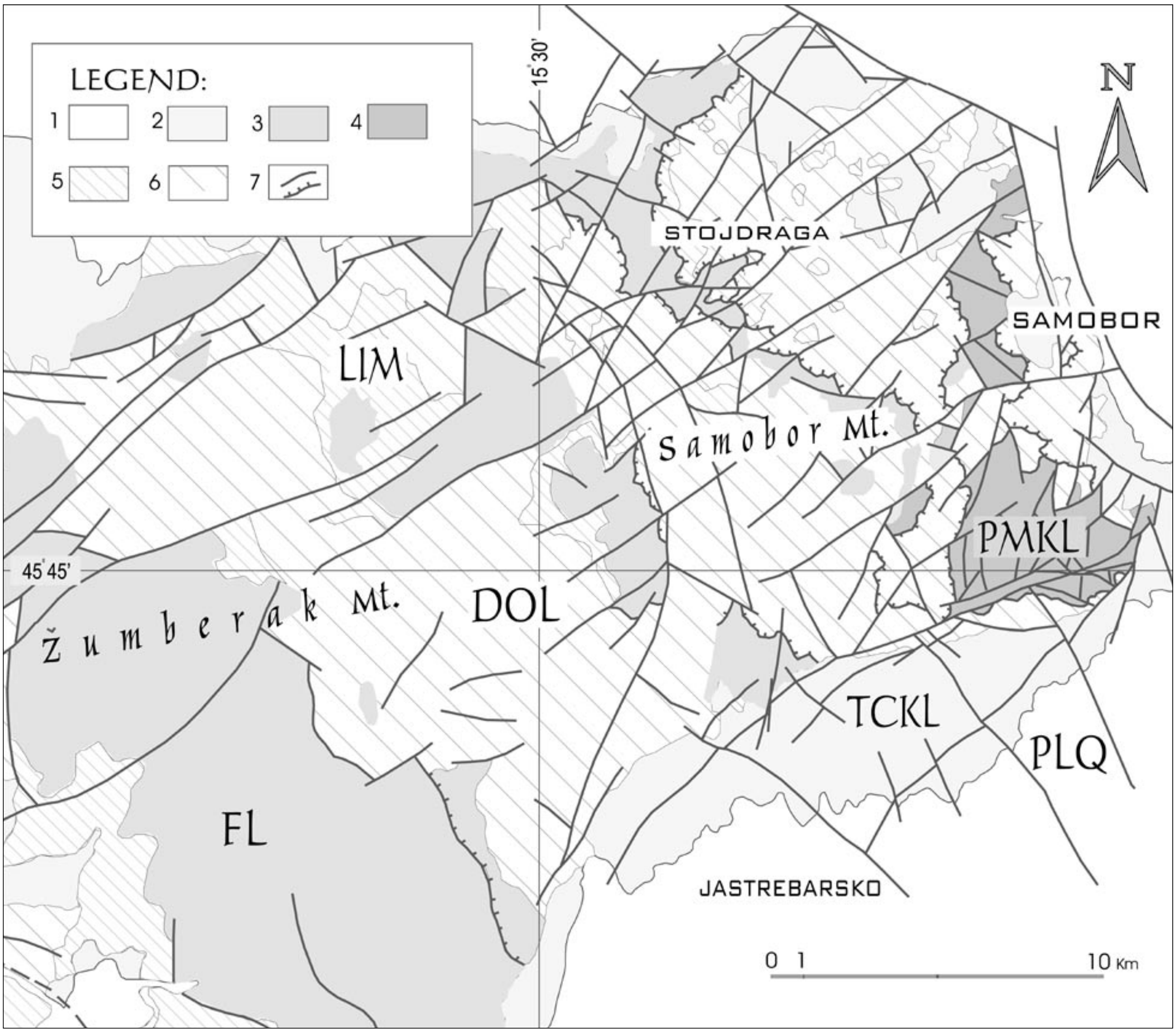

Fig. 2 Lithological sketch-map of the study area (compiled from the geological maps of ŠIKIĆ et al., 1977; PLENIČAR et al., 1975 and BUKOVAC et al., 1983): 1 – PlQ (Plio–Quaternary and Quaternary unindurated siliciclastic sediments; 2 – TCKL (Tertiary clastic sedimentary rocks); 3 – FL (Cretaceous flysch-like series); 4 – PMKL (Upper Palaeozoic–Triassic clastic sedimentary rocks); 5 – LIM (limestones, predominantly Jurassic); 6 – DOL (dolomites, predominantly Middle and Upper Triassic); 7 – faults.

gypsum and anhydrite beds as a result of shallowing of the sedimentary basin (ŠINKOVEC, 1971; ŠIFTAR, 1989). The Lower Triassic also developed a typical shallow-water sandstone facies. In the lower part of the series the terrigenous clastics are dominant, but the carbonate component increases toward the younger strata. This Permo–Triassic complex is exposed in the eastern part of the study area (Samobor Mt.) with patches of Lower Triassic on its extreme southwest fringes.

A characteristic clastic series is associated with the Upper Cretaceous transgression and subsequent deepening of the sedimentary environment. It consists of various flysch-type rocks, spreading over most part of the Žumberak. Genetically, these contain pelagites, hemipelagites, turbidites and contourites (BABIĆ, 1974; MRINJEK, 1992) with the most frequent lithologic members such as calcareous and clayey marls, shales, calcarenites and breccias (basal), occasionally interbedded with cherts and platy limestones.

The peripheral area of Žumberak is composed of diverse clastic facies deposited during the Tertiary period, mainly in the Neogene. These rocks overlie the southern part of the terrain in an almost uninterrupted stratigraphic sequence ranging from the Miocene to Pliocene–Quaternary. To the east and north (Slovenia) they appear only as limited erosion caps transgressively overlying the older Mesozoic, mostly carbonate rocks. In the central parts of Žumberak they are virtually nonexistent. Depending on the unstable sedimentary conditions during the observed time span, the lithology varied from a typical transgressive series beginning with conglomerates, to deep-water clastic deposits marked by calcareous and clayey marls. Due to progressive shallowing of the sedimentary environments towards the end of Neogene the bulk of sedimented material is of a sandy and marly type. Marls, marly clays, sands, sandy marls and clays predominate through the entire Pliocene and extend into the Quaternary (Plio–Pleistocene). This material is weakly consolidated or, most often, unconsolidated and thus easily erodable, which is why peripheral valleys in the southern parts of Žumberak are filled almost solely with its weathering products.

3. DRAINAGE NETWORK

Dendritic and centrifugal patterns, with minor modifications, characterize the drainage network of the Žumberak mountainous landscape. The pattern is primarily tectonic, particularly owing to the vigorous uplift and vertical faulting during the Pliocene–Pleistocene periods (PRELOGOVIĆ, 1969; DUJMOVIĆ & BOGNAR, 1995). The majority of streams with any alluvium fill, which traverse this block-faulted structural morphology are of the fifth-, less frequently of the sixth-order. Draining mostly the peripheral mountain faces they are directed to the main regional base-levels – the valleys of the Sava, Kupa and Krka rivers. Only two of the sixth-order streams transverse its central part to which a great many fifth- or lower-order streams gravitate (the centripetal, mountain drainage network). Of them, the Bregana is a tributary of the Sava river to the east, while Kupčina empties into the Kupa river to the south. Both of these, their tributaries, and the majority of the fifth-order streams in general, contain depositional flood plains. In contrast, due to their higher-gradient channels, the fourth-order valleys only rarely experience aggradational processes, sometimes even cutting their channels into bedrock.

The channel–reach processes

The majority of sampled streams in the study area have developed a certain type of alluvial-channel morphology. Bedrock channel substrate is only found in exceptional cases at least as drainage basins of fourth- or higher order are concerned. This is of great importance as regards the interaction of channel and slope processes which control the delicate relationship between erosion, transport and deposition of the weathered material. In streams cut in bedrock (mostly third-order, or lower), with steep channel gradients, there is a strong coupling between processes in channels and on their adjacent slopes. Since interfluvial distances are often considerably reduced by a progressively branching valley network (often with colluvial substrates where streams are present) in the upper reaches of a drainage basin, vertical incision and upslope debris flow operate more in unison. The mixing of waste material from both environments may be so effective as to smooth the impact of linear and areal aspects of lithology on the compositional variables of channel sediment. This does not occur in the downstream reaches of the channel with progressively lower gradients where, due to the valley widening, the sediment supply from the valley sides is virtually cut off, leaving the bedload composition to depend almost solely on the processes along the stream channel. Depending on the channel gradient and discharge, these may vary from scour (mostly lateral) to deposition of the eroded material from upstream distances which would leave the linear basin aspects as the only geomorphological variable relevant for determining the influence of bedrock lithology on geochemistry or mineralogy of the load. In the investigated area these channel reaches are essentially of three morphological types (according to MONTGOMERY & BUFFINGTON, 1997): the Žumberak heartland abounds with drainage basins of the plane-bed, more rarely step–pool and intermediate channel–reach morphologies, while on its borders, particularly within the Kupa catchment area, mostly pool–riffle morphology occurs (Figs. 3a–d). All of these are distinguished by relatively low gradients varying from <0.015 to 0.03 m/m (measured from the sampling sites upstream) which represents typical values for the majority of mountain drainage basins (TUCKFIELD, 1980; MONTGOMERY & BUFFINGTON, 1997), but the intricate relationship between the bank erosion and character of bedload must be treated with caution (PAOLA, 2000). Thus, no bank (lateral) erosion will be found representative of mountain valleys because of the prevalence of coarse material transported along the channel. The peripheral, mostly sandy-bedded streams will experience occasional bank failures, especially under high discharge events, as may be the case in alluvial segments with silt–sand overbanks on the southern slopes of Samobor Mt. (PEH & MIKO, 2001).

Dry valleys

Another, even greater, problem in evaluating the geomorphological variables may be the presence of dry valleys in the upper reaches of valley segments, which are irrespective of modern channel type and processes. The majority of third- or lower order valleys which occupy the upper areas of the larger drainage basins in the Žumberak region are streamless (Fig. 3e). These valleys must have been cut by streams some time in the past when climatic conditions were different. Climatic changes in the post-glacial period, which commenced at the termination of the Pleistocene ice age, were characterised with increasing amounts of atmospheric moisture. Streams swollen by meltwater and higher rainfalls must have had an immediate impact on raising the level of the water table and, coupled with other factors (such as, local and regional uplift), might have ultimately resulted in prompt expansion of the drainage network (e.g. GREGORY & WALLING, 1973; OLLIER, 1981). Later, with a fall in the average precipitation owing to retrograde climatic changes up to the present time, the latter was contracted to its present extent. Thus, dry valleys portray a major or minor

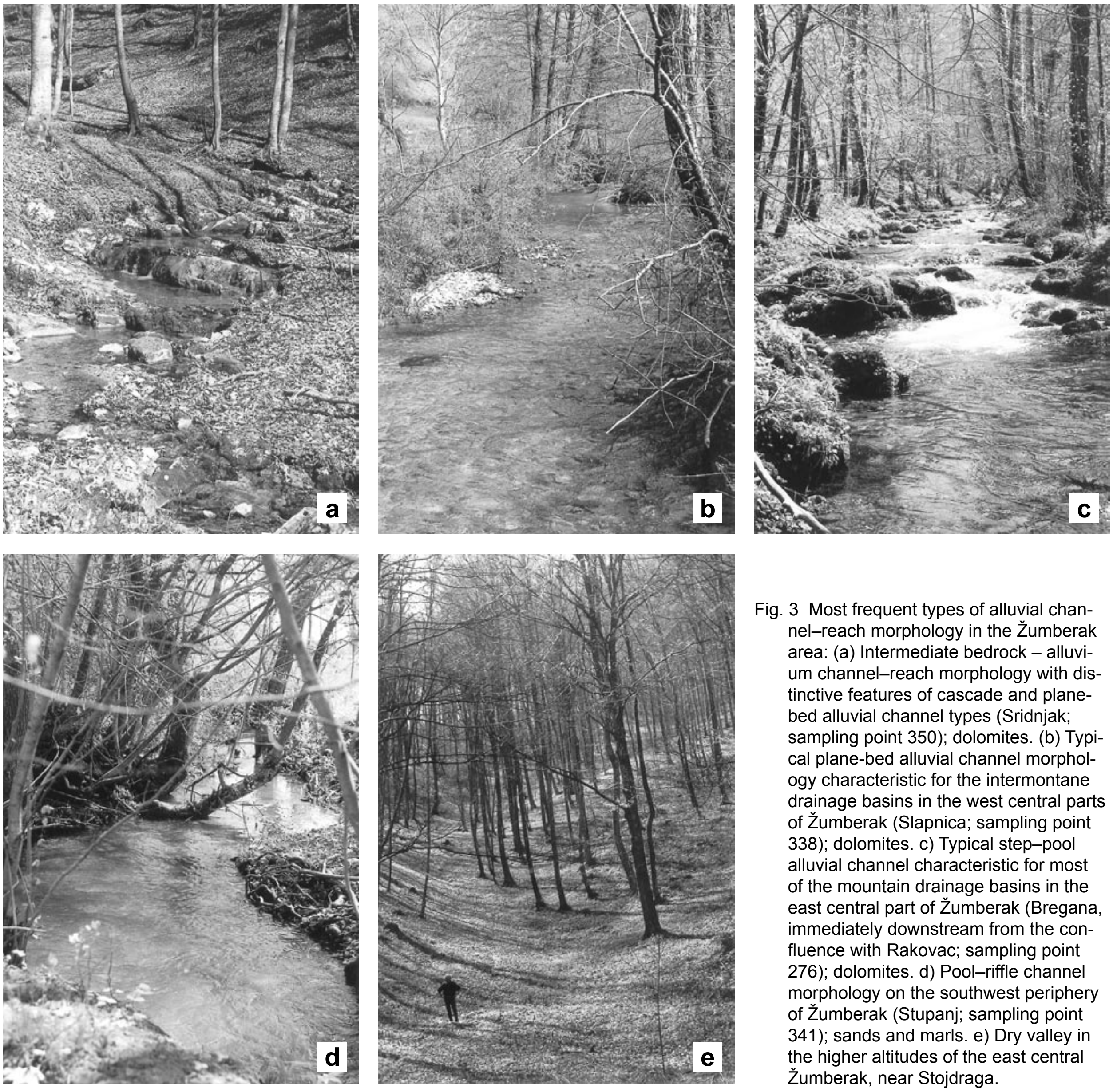

Fig. 3 Most frequent types of alluvial channel–reach morphology in the Žumberak area: (a) Intermediate bedrock – alluvium channel–reach morphology with distinctive features of cascade and plane-bed alluvial channel types (Sridnjak; sampling point 350); dolomites. (b) Typical plane-bed alluvial channel morphology characteristic for the intermontane drainage basins in the west central parts of Žumberak (Slapnica; sampling point 338); dolomites. c) Typical step–pool alluvial channel characteristic for most of the mountain drainage basins in the east central part of Žumberak (Bregana, immediately downstream from the confluence with Rakovac; sampling point 276); dolomites. d) Pool–riffle channel morphology on the southwest periphery of Žumberak (Stupanj; sampling point 341); sands and marls. e) Dry valley in the higher altitudes of the east central Žumberak, near Stojdraga.

discrepancy between the more restricted size of the present stream pattern with regard to the valley pattern which it partly occupies (GREGORY & WALLING, 1973). This discrepancy has a major impact on the effective cutoff of the higher parts of the mountain drainage basins from the rest which contain the active stream network, as regards modern geomorphological processes. It was shown by the morphometric factor model of the bordering region (Hrvatsko Zagorje) that coupling between the slope and channel processes is effective only when the active stream network (stream density) is considered, while the total valley network (drainage density) relates to an altogether different subsystem of drainage basin processes (PEH, 1994, 1997). In such a case one would relate erosion, transport and deposition only to the valleys with perennial streams while intermittent streams, and dry valleys in particular, contribute little to the recent basin change. Consequently the compositional scheme of the waste, transported and deposited load, sampled at the basin outlets, would be in concordance with more recent conditions along the stream channels and their active tributaries than with those in interfluvial areas occupied by relic fluvial landforms.

4. SAMPLING

Field investigations encompassed an area of about 600 km^2 with 46 fourth- and fifth-order mountain drainage basins ranging in size from 0.65 to 26.00 km^2 (Table 1, Fig. 4). In the majority of basins both stream and overbank material were obtainable for sampling which was carried out in a “pair” design on each site wherever possible. Therefore both media were regularly collected from the same spot within a few metres

Case	Sample	Drainage basin	STR	OV	Order	Catchment	Slope (m/100 m)	Area (km^2)
1	4	Kamenjak	+	–	4	Sava	16.67	2.28
2	7	Potok 7 (Pod vrh)	+	–	4	Sava	3.33	1.09
3	41	Ludvić	+	+	4	Sava	4.21	4.41
4	194	Orejovec	+	+	4	Kupa	0.88	5.25
5	196	Piroški potok (SL)	+	+	4	Krka	2.67	6.03
6	197	Skradnja (SL)	+	+	4	Krka	2.35	5.07
7	214	Fučanski jarak	+	+	4	Sava	6.67	2.81
8	220	Jarak	+	–	4	Sava	4.00	3.64
9	221	Velika draga	+	+	4	Sava	6.67	2.99
10	349	Jaševnica	+	+	4	Kupa	1.33	12.02
11	350	Sridnjak	+	–	4	Kupa	10.67	1.38
12	351	Ponornica 351	+	+	4	Krka?	1.82	1.01
13	354	Ponornica 354	+	+	4	Kupa?	0.62	2.86
14	365	Dobri potok	+	–	4	Sava	6.67	3.36
15	366	Vorbaščica	+	+	4	Kupa	2.86	4.56
16	367	Potok 367	+	–	4	Kupa	3.81	2.23
17	371	Potok 371	+	+	4	Kupa	2.85	0.65
18	19	Škrobotnik	+	+	5	Sava	2.20	8.79
19	21	Breganica	+	+	5	Sava	1.60	11.21
20	42	Lipovačka g.	+	+	5	Sava	0.72	26.00
21	43	Rudarska g.	+	+	5	Sava	0.72	15.48
22	178	Reka	+	+	5	Kupa	1.60	9.19
23	187	Okićnica	+	+	5	Kupa	0.50	19.11
24	190	Potok 190	+	+	5	Kupa	0.75	3.15
25	191	Stošinec	–	+	5	Kupa	0.77	3.54
26	198	Sušica (SLO)	+	+	5	Krka	1.40	9.79
27	275	Bregana	+	+	5	Sava	2.32	14.52
28	276	Rakovac	+	+	5	Sava	2.11	8.39
29	328	Žumberačka reka	+	+	5	Kupa	1.33	15.96
30	331	Sušica	+	+	5	Krka	2.86	8.51
31	333	Suvaja	+	+	5	Kupa	0.98	22.91
32	334	Potok	+	+	5	Kupa	2.22	5.44
33	335	Svilnica	+	+	5	Kupa	3.08	3.63
34	337	Ponikva	+	+	5	Kupa	0.19	6.46
35	338	Slapnica	+	+	5	Kupa	0.57	16.25
36	339	Puškarov jarak	+	+	5	Kupa	1.74	9.77
37	340	Brebrovac	+	+	5	Kupa	0.97	5.69
38	341	Stupanj	+	+	5	Kupa	0.77	4.48
39	342	Stiska	+	+	5	Kupa	0.91	11.43
40	343	Malunja	+	+	5	Kupa	0.98	6.87
41	344	Gonjeva	+	+	5	Kupa	1.13	8.11
42	345	Kamenica	+	+	5	Kupa	1.00	17.23
43	346	Bukovica	+	+	5	Kupa	0.77	12.25
44	347	Slatinek	+	+	5	Kupa	0.63	4.33
45	368	Stiper	+	+	5	Kupa	0.8	5.72
46	370	Selna	+	+	5	Kupa	0.37	5.32

Table 1 General data describing the sample sites. Legend: STR – stream sediments; OV – overbank sediments.

distance except in those cases where valley or channel fills were absent. For the six fourth-order drainage basins the overbank sample was unavailable. In one drainage basin of fifth order the stream sediment was not collected. The former case was due to the absence of alluvium in high-gradient valleys, while in the latter the sediment load was present but not sampled because of the obvious household contamination in the vicinity of the sampling site. Sometimes the fine-grained, silt–clay content of the bed similar to that of the adjacent banks arose suspicion as to its origin and the possibility for mistaking the sample media, but earlier investigations have shown that this is rarely the case (PEH & MIKO, 2001).

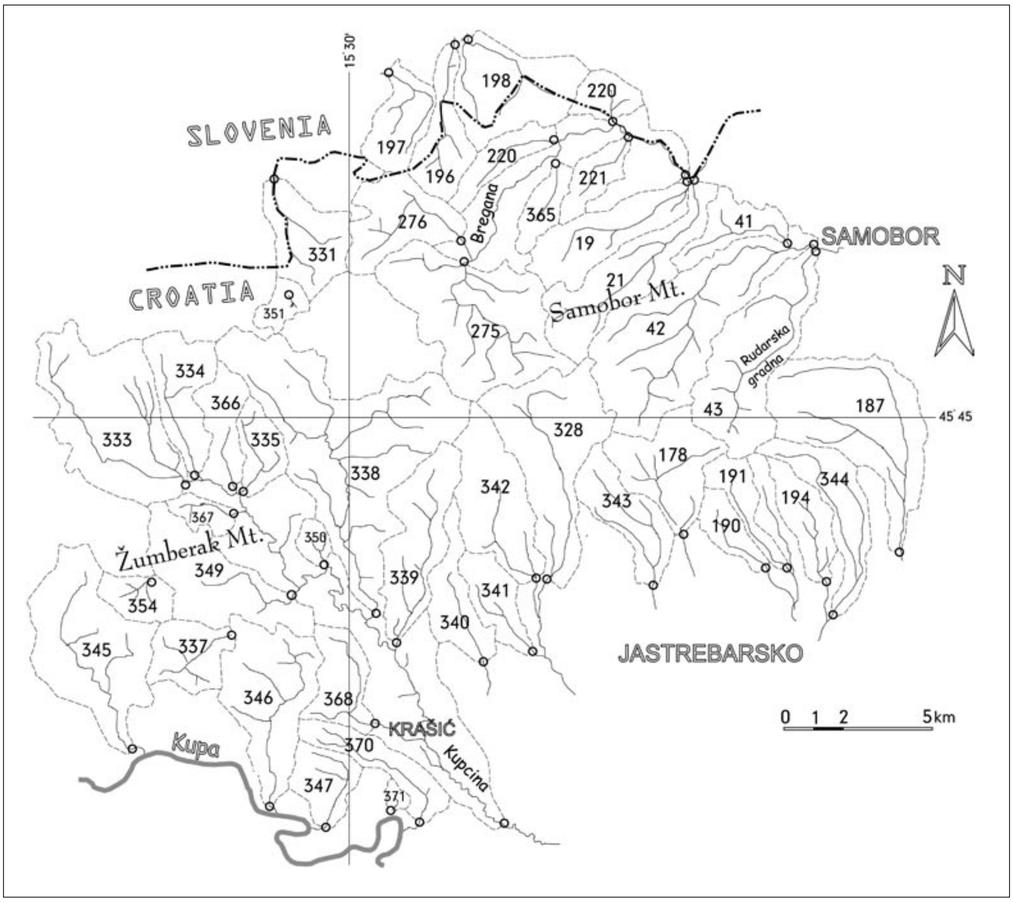

Fig. 4 Sample map showing distribution of fourth- and fifth order drainage basins in the study area.

The sample sites were selected at the basin outlets, sufficiently upstream from the confluence with higher (or the same order) streams in order to avoid sampling the sediment that may result from mixing of material from the two channels during the flood flow. Active stream sediment, which represents the composite of the recently deposited bed load material, was collected from several spots (ordinarily 5–10 as recommended by SALMINEN et al., 1998) over a short channel stretch upstream of the selected site. Simultaneously, a single overbank sediment sample was taken from approximately the same point at the exposed area of either bank of a channel. The latter is composite material collected from the bank section ranging in height from 25 cm beneath the surface down to the water level (usually 0.5 to 2 m thick), while the first 25 cm of the upper, near surface horizon was avoided because of possible anthropogenic disturbance and pedogenesis. In both cases a quantity of about 3 kg of sediment was collected to yield enough representative material for sieving and analysis.

Sample preparation

Collected samples were air-dried (at <40ºC) for approximately three months. After drying, the samples were disaggregated in a porcelain mortar, homogenized, and finally dry-sieved through stainless-steel screens to the <125 μm size fraction. This fraction was preferred because the highest concentration of most of the elements, especially trace elements, occur in the fine-grained, usually 63–125 μm size fraction (e.g. RHOADS & CAHILL, 1999). Also, some studies show that the <125 μm size fraction can contribute to more than 95% of the particles in most samples (SWENNEN et al., 1998).

Analytical methods

Analytical work was performed at the ACME Analytical Laboratories in Vancouver, Canada, where samples were subjected to multi-acid digestion ICP analysis and geochemical Hg analysis by flameless AA. A total of 36 elements were thus analyzed with Au, Be, Bi, Mo, U and W having concentrations invariably, and Ag, Sb, Sn and Cd mostly under the detection limit.

5. REGRESSION ANALYSIS

Regression analysis is one of the most often utilized techniques in multivariate as well as bivariate analysis for assessing the relationship between variables in a data set. In multivariate solutions a multiple linear regression is used to define the simultaneous relationship between one dependent (or response) variable

Y against a number (n) of independent (or predictor) variables X_{1-n}. This is done by finding the "best-fit" straight line via the method of least squares estimation in which deviations of the dependent variable from that line are minimized. The problem on which the analyis is most frequently centred, as in this case, is to assess the contribution of predictor variables to the overall regression model. In other words, it is useful to find that portion of the total variance in a response variable which can be explained by a set of influencing variables. This value is usually termed the "goodness-of-fit", or a multiple coefficient of determination (R^2), which is, in fact, the multiple correlation coefficient of a particular variable squared. In the actual case, the content of each analysed element represents a particular dependent variable in a regression model, while the lithological influences, expressed either in the form of areal or linear proportions of a specific rock unit within a drainage basin, are represented by a set of independent variables.

Variables

An assemblage of 26 elements consisting of eight major and 18 minor and trace elements were selected as independent variables in the regression analysis. In order to obtain the best possible fit of the model a number of these variables should have been submitted to the transformation procedure. Positively skewed frequency distributions were observed among most of the minor and trace elements in both stream sediment and overbank sediment data sets. Thus, the conventional log-transformation was carried out for most of the minor and trace elements such as Cu, Pb, Zn, Ni, Co, Mn, As, Th, Sr, Ba and Hg, but also for some major elements such as Ti, Mg and P.

Six rock types that dominate the terrain lithology were specified as independent variables in the regression analysis (Fig. 2). These are: (1) Clastic sedimentary rocks of the Tertiary period (mostly Neogene) – TCKL; (2) Clastic sedimentary rocks of Upper Palaeozoic–Triassic – PMKL; (3) Cretaceous flysch-like series – FL; (4) Limestones (dominantly Jurassic) – LIM; (5) Dolomites (Middle and Upper Triassic types are prevalent) – DOL; (6) Plio–Quaternary and Quaternary sediments – PlQ. The above scheme may seem grossly oversimplified, but it serves its purpose to the extent to which the great variety of clastic and carbonate facies is expected to contribute to the overall geochemical composition of the sample media. The simultaneous use of variables belonging to the same realm of sedimentary rock types, namely dolomites and limestones, is necessary when considering their strongly contrasted weathering patterns. Crushed and mylonitized dolomite rocks represent a classical "clastic factory" (HOVIUS & LEEDER, 1998) supplying the valley floors with abundant sediment flux, while limestones are much more resistant to physical weathering. These six variables are given the form of the relative proportions of each rock unit within a drainage basin, yet, because calculated on different grounds, they formulate the two experimental designs employed here. The first approach is well-known and operates on the basis of areal proportions of the specified drainage basin lithology, while the second is more specific and takes into account its linear proportions along the perennial streams. In the latter case all blue streamlines have been scanned from the topographic maps (scale 1:25,000).

Defined as above, the lithological variables can be regarded as a compositional data set the statistical treatment of which may be met with difficulties because of the so-called constant sum constraint. The nature of data (zero values in a number of cases due to the absence of some rock types) ruled out the most frequent way of solving the problem which uses the log-ratios (e.g. AITCHISON, 1986) so that initial matrix ill-conditioning was amended optionally, by dropping the surplus or "unnecessary" variable from the closed data set. The lithologic variable subtracted from the original six-variable data set is represented by the youngest rock unit – the Plio–Pleistocene and Quaternary unconsolidated deposits of various types (mostly of freshwater, fluvial–lake origin) – which are the reworked older material of very heterogeneous composition, occupying large portions of the marginal south-eastern drainage basins. Therefore, the supposedly small variance (effected by mixing) contained in the sixth variable was easily sacrificed for the necessities of untangling the compositional data problem. The remaining five lithological variables were deemed quite sufficient to faithfully reflect the main lithologic features of the study area liable for geochemical behaviour in the observed media.

6. RESULTS AND DISCUSSION

The results of regression analysis are described separately for each of the two experimental designs and displayed in Tables 2 and 3. The columns in the tables represent the values of the coefficient of determination R^2, and beta coefficients (β) for independent variables. Tabulated data show a great variance between the values of R^2 which range from 11% for strontium (areal) to 73% for magnesium (areal), both in stream and overbank sediments. Better insight into the variations in each individual element, which reflect the average differences among specified rock units, can be gained by categorizing the R^2 values into simple classes similar to those used for simple correlation coefficients: (1) <0.3 for no correlation; (2) 0.3–0.5 for weak correlation; (3) 0.5–0.7 for good correlation; and (4) >0.7 for strong correlation. Thus one can more easily trace the degree to which the elements are explained by predictor variables if the two approaches, namely linear and areal, are compared. Of the 26 elements reported in this work only P, Sr and Y have R^2 values invariably less than 0.3, which means

	Linear						Areal					
El.	R^2	TCKL	PMKL	FL	LIM	DOL	R^2	TCKL	PMKL	FL	LIM	DOL
Fe	0.50					–	0.44					–
Ca	0.54					+	0.51					+
Mg	0.67					+	0.73				+	+
Ti	0.24						0.26				–	–
Al	0.49	–				–	0.43					–
Na	0.52	–				–	0.42					–
K	0.45					–	0.41					
P	0.14						0.20	+				
Cu	0.37		+				0.30		+			
Pb	0.63		+				0.63		+			
Zn	0.45		+				0.47		+			
Ni	0.64					–	0.41					
Co	0.55					–	0.48					–
Mn	0.47						0.45					
As	0.24		+				0.23		+			
Th	0.36	–	–	–		–	0.40	–		–		–
Sr	0.13						0.11					
V	0.50	–	–			–	0.41	–				–
La	0.35		–			–	0.36					–
Cr	0.53						0.46		+			
Ba	0.52		+				0.61		+			
Zr	0.59	–				–	0.58				–	–
Y	0.26	–	–			–	0.26					–
Nb	0.42	–				–	0.41					–
Sc	0.53	–	–			–	0.50	–				–
Hg	0.31		+				0.43		+			

Table 2 Summarized results of two experimental designs for stream sediment data. Legend: El. – element, R^2 – goodness-of-fit, TCKL – Clastic sedimentary rocks of the Tertiary period; PMKL – Clastic sedimentary rocks of Upper Palaeozoic–Triassic; FL – Cretaceous flysch-like series; LIM – limestones; DOL – dolomites.

that more than 70% of their variation is not explained by the average differences between the lithologic units. To this group can also be added elements such as As, Ti and Nb whose residual variability may also exceed this value. Their lithological origins are better mirrored in overbank material combined with linear proportions of map units, particularly in the case of titanium. The relationship between the stream and overbank sample media will not be discussed here at length because it has been thoroughly considered in an earlier work (PEH & MIKO, 2001). Here, it will suffice to say that the principal geochemical difference between the two sample media was perceived primarily in the presence of a higher content of the bulk of analyzed elements in the overbank sediment as well as in its greater homogeneity. This property certainly extends usefulness of the latter in explanatory purposes due to its greater inherent variability. Here, we are basically concerned with the differences in variation explained by linearly and areally weighted bedrock lithology in a drainage basin. It must be noted, also, that regression designs were narrowed to the variations in geochemical content due to average differences in geological background. Other contributing factors, such as scavenging by Fe and Mn oxides and hydroxides, influence of pH, or anthropogenic impact, were not considered.

The effect of linear vs. areal proportions is most evident from the regression results summarized as the tabulated values of R^2 (Tables 4 and 5; Figs. 5 and 6). The greater part of the analyzed elements fall within the two classes whether R^2 was calculated from linear or form areal proportions, namely of weak (0.3–0.5), or good (0.5–0.7) explanatory potential, respectively. But, it is obvious that the variation of geochemical content is better explained by linear than by areal proportions of rock units, as the number of elements change their R^2-class when the tables are cross-compared, with the higher values in the first case. This is particularly characteristic of Ca and Ti among major, and Cr, Zr, Sc and Ni among minor elements. Explanatory potentials of Nb and Co are also considerably augmented while Mg, interestingly, remains steadily associated to the areally defined bedrock lithology which presupposes the constant supply of dolomite material, spatially unconstrained because of its characteristic weathering pattern. The preponderance of a linearly over areally defined lithological imprint in the sample media is even more noticeable if the R^2 values for the two

El.	Linear						Areal					
	R^2	TCKL	PMKL	FL	LIM	DOL	R^2	TCKL	PMKL	FL	LIM	DOL
Fe	0.67		+				0.62		+			
Ca	0.56					+	0.45					+
Mg	0.69						0.72					
Ti	0.56					–	0.45					–
Al	0.47					–	0.37					
Na	0.47					–	0.37					–
K	0.48						0.43		+			
P	0.22				+		0.16				+	
Cu	0.55		+	+	+		0.53		+			
Pb	0.54		+				0.59		+			
Zn	0.48		+		+		0.42		+			
Ni	0.59				+		0.42					
Co	0.70		+		+		0.64		+	+	+	
Mn	0.46		+		+		0.38		+	+		
As	0.46		+				0.38		+			
Th	0.42					–	0.39					–
Sr	0.21						0.17					
V	0.45					–	0.34					
La	0.40					–	0.31					–
Cr	0.62				+		0.47					
Ba	0.55		+				0.53		+			
Zr	0.57					–	0.48					
Y	0.21						0.16					
Nb	0.39					–	0.24					
Sc	0.51					–	0.40					
Hg	0.52		+				0.62		+			

Table 3 Summarized results of two experimental designs for overbank sediment data. Legend: El. – element, R^2 – goodness-of-fit, TCKL – Clastic sedimentary rocks of the Tertiary period; PMKL – Clastic sedimentary rocks of Upper Palaeozoic–Triassic; FL – Cretaceous flysch-like series; LIM – limestones; DOL – dolomites.

experimental designs are represented as the difference which may be either positive or negative (R^2 linear - R^2 areal). The diagrams (Figs. 5 and 6) show the better fit of the linear aspects which in the case of overbank sediments is quite remarkable because only Mg among major, and Pb and Hg among trace elements, are at variance with the general scheme. As a contrast, with stream sediments not only a smaller number of elements are better explained by the linear approach but also the positive R^2 difference is generally much less (with the exception of Ni and Cu). Thus, some extra minor and trace elements such as Ba, Zn and Th tend to be areally better associated to lithology, while almost all others also reduce to some extent their affinity toward linearly defined bedrock units (cf. Figs. 5 and 6). The overbank sediment is, as expected, a more contrasting background for discriminating between the two regression models. This is the result of the different fluvial dynamics which, even on a small scale, include local fluctuations either due to a low or seasonal variability in flow along the channel or to floods (PAOLA, 2000). Also the geochemical composition of the sediment flux cannot be separated from other aspects of process-based fluvial scenarios, particularly those related to morphodynamics. Depositional history of the drainage basin sampling media reflects either a slow and steady aggradation processes, or catastrophic (flood-induced) redistribution of the previously (temporarily) deposited bedload. Active sediment on the channel floor past any given spot along the channel is more or less balanced with stream transport capacity, having a limited range of supply, transport and deposition. Overbank sediment, on the other side, originates from hazard events with increased water discharge, high average velocities, and high sediment discharges (GREGORY & WALLING, 1973) which affect the whole stream network upstream of the sampling site. Furthermore, besides inundating the wash load over its banks this event may also result in episodic erosion of the channel floor and banks of the feeder streams throughout their length.

Relationship to source lithology

The parent lithology as a source for various elements in alluvial media can be deduced from regression coefficients (β) given in Tables 2 and 3. Only the sign of significant β values is highlighted in corresponding lithological columns marking the increase or decrease

R^2	LINEAR	AREAL
<0.3	**Ti**, **P** As, Sr, Y	**Ti**, **P** As, Sr, Y
0.3–0.5	**Al**, **K** Cu, Zn, Mn, Th, La, Nb, Hg	**Fe**, **Al**, **Na**, **K** Cu, Zn, Ni, Co, Mn, Th, V, La, Cr, Nb, Hg
0.5–0.7	**Fe**, **Ca**, **Mg**, **Na** Pb, Ni, Co, V, Cr, Ba, Zr, Sc	**Ca** Pb, Ba, Zr, Sc
>0.7	– –	Mg –

Table 4 Tabulated values of goodness-of-fit R^2 for both regression models (stream sediments).

R^2	LINEAR	AREAL
<0.3	**P** Sr, Y	**P** Sr, Y
0.3–0.5	**Al**, **Na**, **K** Zn, Mn, As, Th, V, La, Nb	**Ca**, **Ti**, **Al**, **Na**, **K** Zn, Ni, Mn, As, Th, V, La, Cr, Zr, Sc
0.5–0.7	**Fe, Ca, Mg, Ti** Cu, Pb, Ni, Cr, Ba, Zr, Sc, Hg	**Fe** Cu, Pb, Co, Ba, Hg
>0.7	– Co	Mg –

Table 5 Tabulated values of goodness-of-fit R^2 for both regression models (overbank sediments).

in the share of the explanatory variables (rock units) to the regression model (elements).

A number of minor and trace elements derive their origin from the clastic sedimentary rocks of the Upper Palaeozoic to Lower and Middle Triassic age (PMKL). The β coefficients for this rock unit are always positive, signaling that (Fe), Cu, Pb, Zn, Co, Mn, As, Hg and Ba are hosted in these rocks mostly in the sulphide-rich layers or veins in the fine-grained sandstones. This mineralization is notable for the central parts of the Samobor Mt. (Rudarska Gradna stream) sometimes even in the form of small-scale siderite–haematite–sulphide ore bodies with chalcopyrite, cinnabarite and galena–barite paragenesis (ŠINKOVEC, 1971; ŠIFTAR, 1989). All these elements can be traced equally in both alluvial media, and following both approaches.

The β coefficients are not necessarily positive when other rock types are considered. This is especially evident with dolomite bedrock to which most of elements, except Ca and Mg, are negatively associated. Negative values denote the elements which are generally deficient within the relevant lithology, such as Fe, Ti, Al, Na, V, La, Zr, Nb and Sc in dolomite (DOL), Th in the flysch-like series (FL), if statistically significant, as well as Al, Na, Th, V, Zr, Nb and Sc in the Tertiary clastics (TCKL). Thorium is, interestingly, negatively related to all bedrock types where the β coefficients are significant, though its lithological origin

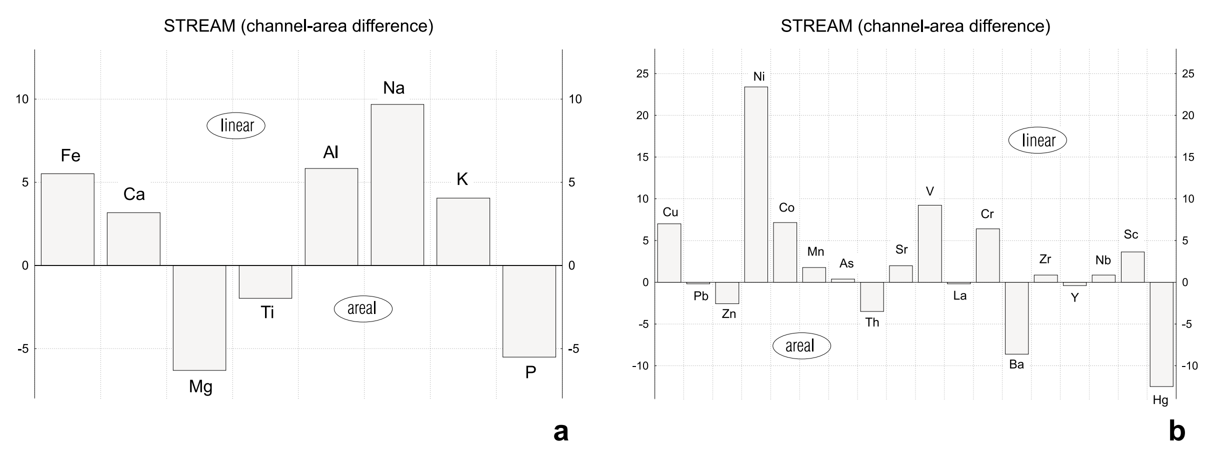

Fig. 5 Linear vs. areal proportions of lithological influence in stream sediments: (a) major elements; (b) minor and trace elements.

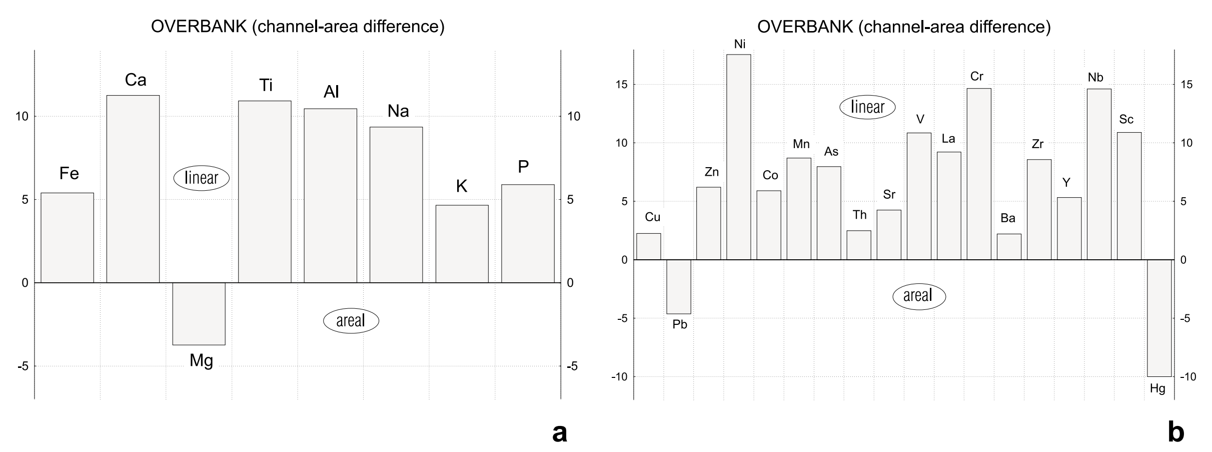

Fig. 6 Linear vs. arial proportions of lithological influence in overbank sediments: (a) major elements; (b) minor and trace elements.

is accounted for by 42% at most. Other elements which are also poorly explained by the variations in the rock types but show statistically significant betas must be pondered carefully. Phosphorus, for example, is a major element falling within this group. Not more than 22% of the variation in this element is due to differences between lithological units but, nevertheless, siginificant β values may appear which relate it positively to limestones (in overbank), or Tertiary clastics (in streams). Magnesium however, of all the elements is best explained by lithological effects (over 70%), albeit the regression coefficient for overbank are proved non-significant whether by the linear or areal approach. Such examples must be regarded only as the case wherein the element has no preferential association with respect to parental rock and the sediment influx with Mg-containing minerals comes in equal proportions from all rock types, not only dolomite (see Tables 2 and 3). Otherwise, Mg in stream sediments is positively related to dolomites, even with limestones (where these occur interbedded with dolomites as carbonate rocks of secondary importance) which can be ascribed to the selective power of the streams precipitating the suspended load not far from the eroded reaches of the channel and valley sides.

A characteristic relationship of a group of trace metals, such as Cu, Zn, Ni, Co, Cr and Mn to the limestone bedrock (Table 3) gives rise to a long-disputed problem of the sediment supply from the limited weathering of “hard” rocks, particularly in carbonate terrains (DURN, 1996; DURN et al., 1999; MIKO et al., 1999). In contrast to the “soft” rocks (including dolomites) which are readily decomposed so that soil formation is not necessary for erosion (LEEDER et al., 1998), limestones predispose some kind of influx of weathered material from different geochemical surroundings. In the Žumberak steep intermontane catchments the regolith which mantles the limestone bedrock is most probably of allochtonous origin, and the unrelated group of elements must have derived their origin from the different bedrock. As expected, all these considerations do not interfere with the stream sediments (Table 2) as the finer silt–clay topsoil material with its trace element contents is easily dispersed in suspension and deposited as the overbank sediment downstream. Besides, the linear approach is proved again as the more faithful regression model which gives emphasis to a limited channel reach as the main receptacle for drainage basin forms and processes.

The Cretaceous flysch-like series and Tertiary siliciclastic rocks do not appear as a predisposed source repository for most of the analyzed elements. This is especially characteristic of the flysch-like series where almost all betas are rendered non-significant, while the latter repeats the scenario of correspondence between the low element content and stronger presence of pertinent lithology. The majority of elements, including Al and Na, are negatively regressed. The fact that in both linear (mostly) and areal (more rarely) approaches these elements readily show lower concentrations in stream sediment can, perhaps, be explained the other way round: by the scenario in which the streams running through the unconsolidated and easily erodible Tertiary siliciclastic rocks easily wash away the fine-grained particles containing clay minerals. None of the analyzed elements in overbank sediment is significantly related to these rocks, which may be indicative of their depositional history – the same way as the Tertiary clastics represent older reworked material, so the recent alluvium represents the Tertiary clastics (or both) recycled and mixed within the area where the latter occupy greater expanses of the Žumberak region (on the border of the Karlovac Depression in the southeast). Furthermore, a great number of elements in overbank sediment which show non-significant β values for each rock unit in both regression models may once again signal indifference of its geochemical composition to climatic or physiographic control on sediment production in late Holocene times.

7. CONCLUSIONS

The two regression models were compared on the assumption that the geochemical composition of alluvial material is essentially derived from the bedrock lithology of a drainage basin. Integration of drainage basin lithology with both stream sediment and overbank sediment geochemical data suites was carried out using the low- to medium-order mountainous catchment basins and their drainage networks in two ways: (1) as the area of influence occupying the whole basin upstream and upslope from the sampling site; and (2) the "line" of influence representing a narrow tract along all of the perennial streams which form the active stream network upstream and upslope from the sampling site. The most straightforward general result of these experiments is the one which underpins the linear approach as the more fitted model of the lithological influence on the analyzed sample media (88% vs. 62% of analyzed elements). This is explained by the stronger erosive and transporting power of the water which flows in the well defined channel network with respect to the unconcentrated surface runoff in the interfluvial area, coupled with the buffer effects of rich slope and riparian vegetation, which is characteristic of the Žumberak heartland. A number of dry valleys in upper basin reaches also contribute to this, as their geomorphologically inactive bottoms considerably reduce the available area required for continuous sediment supply and transport of weathered material down to the lower parts occupied by perennial and intermittent streams.

The other important regression results, which relate geochemical composition of alluvium to the source terrain lithology, can be briefly summarized as follows:

- the low goodness-of-fit values for elements like P and Sr indicate that their lithological origins are effectively screened by other factors not accounted for by the regression models;
- Mg is, as the best fitted element, steadily associated to the areally defined bedrock lithology (tectonically predisposed weathering pattern);
- trace metals (Cu, Pb, Zn, As, Hg, Ba) derive their origin from the Palaeozoic and Lower Triassic siliciclastic rocks (PMKL) as indicated by significant positive beta values;
- majority of elements do not display significant associations with certain lithological variables such as the Cretaceous flysch-like series (FL) and limestones (LIM);
- negative element associations are typical for dolomites (DOL) and Tertiary siliciclastic rocks (TCKL);
- affinity of a group of trace elements – Cu, Zn, Ni, Co, Mn and Cr – for limestones indicates their allochthonous relation to bedrock.

The regression results acquired in this admittedly restricted study indicate that the geochemical signature of drainage basin sediments grossly reflects their sourcing from the basin parent lithology, but the patterns of compositional variation are not easy to assess as these depend on a variety of processes. Although the better fit of the "linear" model highlights the channel processes as a particularly responsible factor in compositional modifications, one needs a greater number of basin predictor variables for unravelling the specific geochemical behaviour of the analyzed elements. The results indicate the overbank sediments as the better recipient of multivariate geochemical supply signals from the study area. This will narrow future studies to the small-scale provenance analysis of alluvia with more detailed bedrock lithology and other variables included which will encapsulate all relevant local–regional geomorphological and other earth-surface processes responsible for its geochemical composition.

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