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**Paleoecological reconstructions of S-Transdanubian and E-Croatian
loess-paleosol sequences using malacological and sedimentological
investigations**

Theses of Ph.D. dissertation

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1. Preliminaries, aims

One of the main targets of Quaternary paleoecological researches is to give an overall view of the environmental and climatic conditions of a determined area. According to this purpose commonly several supplementary, so-called multi-proxy examinations were made from a certain site. These examinations could be suitable to draw paleoecological conclusions, even carrying significant information, this is why became one of the favourite methods of the Quaternary researches.

Since loessy sediments cover 15% of the area of Hungary the examinations already started on them during the 19th century. The statistical and comparability potential of fossil mollusc shells in loessy sediments were soon discovered, thus the first fauna distribution and paleoclimatic reconstructions were born from the Carpathian Basin in no time. But the importance of malacological analyses accomplished itself only in the 20th century. Using these analyses the reconstruction of Pleistocene climate changes become possible, than after the 1950s, because of consummating sampling method, additional important results were born by using malacology. It is discovered that the malacofauna is capable in itself for making paleoecological reconstructions and on the basis of this the various fauna associations could be classified into special paleoecological zones, so-called biozones (Krolopp, 1973, 1983). Further on, malacological examinations were amplified with several other loess-sediment analyses giving more detailed paleoecological reconstructions (Sümegei, 1989, 1995b, 1996b, 2001, 2005, 2007). The extended results related to mosaic-like environmental and climatic patterns in the Carpathian Basin during the Pleistocene, which had effect the composition of the malacofauna at a certain site.

Multi-proxy paleoecological investigations had not elaborated at the following S-Transdanubian (Hungary) and E-Croatian loess-paleosol sequences. In the examined areas 5 sequences were investigated using multi-proxy methods (Villánykövesd, Máza and Szálka from Hungary, Šaregrad and Zmajevac from Croatia), to create complex paleoecological reconstructions on them.

By the investigations of the 5 loess-paleosol sequences some essential questions could be answered, which might contribute to make the loessy sediments' paleoecological and sedimentological reconstructions more accomplished in the Carpathian Basin. Some of these questions discuss common paleoecological problems, others deal with the necessity and applicability of the multi-proxy investigations.

1. The first question is about the mosaic-like environmental patterns, proved from various parts of the Carpathian Basin (Sümegei, 1995b, 1996b; Sümegei & Krolopp, 1995, 2002; Sümegei & Hertelendi, 1998). I wondered *whether the examined five loess-paleosol sequences show any similarity to each other, and if yes, what these are; or every sequence contains individual paleoecological and sedimentological information.*
2. The second question can be related to the location of the sequences. Every investigated sequence is situated at the right bank of the Danube River, some are close, others are farther. Comparing the paleoecological results of the sequences to others at the left bank of the Danube River (Bácska, Vojvodina), could be any similarities observed, or not. Accordingly *is it possible that different mollusc faunas developed at the right and left banks at the same time proving the geographic/ecological barrier role of the Danube River?*
3. During the examinations of snail populations a couple of questions were raised. The first and most important *whether can be a particular mollusc taxa's refuge area (where can survive the uncongenial climatic and environmental conditions) determined only by its dominance values in an investigated loess-paleosol sequence.*
4. Essential attributes of the mollusc faunas are the dominance and abundance. Adequate abundance values are necessary for correct statistical analyses (Krolopp, 1983), thus the high number of snail specimens in a sample is the better (Krolopp, 1962, 1965, 1973, 1983), besides standard sampling is also necessary. On the other hand it is possible that the abundance is fairly low or even zero in a

sample. Meeting with a problem like this a question is raised whether *which factors can control the abundance values in an examined loess-paleosol sequence.*

5. To make paleoecological reconstruction on a particular loess-paleosol sequence, it is necessary to adjust it in the geological time. For this, geochronological and dating methods can be applied. But it can happen that *a loess-paleosol sequence does not have any absolute dating data, thus could it be correlated with other loess-paleosol sequences only by its fauna composition or sedimentological attributes? Is correlation possible by using stratigraphic methods only?*
6. The last question is about the multi-proxy investigations. *As investigating a loess-paleosol sequence using this method, could the paleoecological reconstruction be more complete and flawless?*

2. Material and methods

The most important link of the multi-proxy examinations series was the malacological examination. The sampling distance which was used by the malacological examinations was also adopted for sedimentological examinations. This sampling distance was 12 cm at the Hungarian segments, 25 cm at the Croatian segments and 10 cm at a certain segment. Every sample contained approximately 5 kg sediment which was wet-screened by a 0.8 mm mesh sieve. After the wet-screening the shells were selected and identified (Krolopp, 1983). The number of pieces (abundance) and the proportion (dominance) of the shells was represented in every sample according to the depth. These results were supplemented with the environmental and climatic demands of the snail species thus the environmental and climatic conditions could be reconstructed on the examined area.

The examinations of the organic matter and carbonate content were executed by the loss on ignition (LOI) method (Dean, 1974). The significance of this method is that the dry and powdered samples are ignited at several temperatures in a furnace then the weight of the samples after every ignition is measured thus by the weight loss the organic matter and carbonate content can be determined. The ignition of the organic matter was at 550°C and the carbonate content was at 900°C and the results were represented according to the depth on a diagram.

The examination of the grain-size distribution was executed by the laser method (Bokhorst et al., 2011). Dried samples were treated with hydrogen peroxide (30% H₂O₂) and hydrochloric acid (10% HCl) to remove the organic matters and carbonates and the samples were put into an ultrasound cleanser for 10 minutes to dissolve the coagulation among the grains before the examination. The measurement was executed with an Easysizer 20 laser sedimentograph by Omeq. This appliance can measure the samples in 42 grain ranges and count frequency and cumulative values by the given results. The given values were settled into grain-size ranges according to the Wentworth-scale (Wentworth, 1922) and the results were represented on a 100% accumulated diagram. The proportion of the several grain classes can be derived to numerous events. Most important of them could be the

weathering. During the weathering the greater sediment grains get broken up into small pieces thus the proportion of fine fractions increase in weathered sediments. Moreover the proportion of sand fraction is the other fraction which can worth to follow into the examined segment. Because the loess contains mostly medium and coarse silts (Smalley & O'Hara-Dhand, 2012) so the relatively high sand content (and also the clay content) could prove the change of the accumulation conditions (Pye, 1995).

The MS examination defines the magnetisable elements content in a certain unit capacity. The most frequent elements are the magnetite (Fe_3O_4), maghemite ($\gamma\text{-Fe}_2\text{O}_3$) and hematite (Fe_2O_3). The examinations were executed with the MS2K sensor of a Bartington MS2 appliance at the Department of Geology and Paleontology. After the measurement of a certain segment the result is a curve (so-called MS-curve) which can help correlating MS-curves of other sequences and also can be helpful in stratigraphical introduction. The importance of correlation ability and the low and high values were recognised in the 1980's (Kukla et al., 1980; Kukla, 1987) and since that time the Chinese loess-paleosol segments' MS-curves had become correlation references (Zhou et al., 1990; Maher, 1998, Ding et al., 2005; Gibbard & Cohen, 2008).

The geochemical examination consists of numerous steps which were regulated by the MSZ 21470-50:2006 standard (Hungarian Standards Institution, 2006). The examinations begin with the extraction of the correct dried samples by crystal water. The aggressive acid dissolves the dissolvable elements from the sediment which will be found in the liquid. The supernatant pipette after a certain time (24 or 48 hours) than dilute and pour in a sample holder and put them into a refrigerator till the examination. The examination was executed with a PerkinElmer AAnalyst 100 atomic absorption spectrophotometer which can measure the dissolved elements contain in dry matter content (mg/kg). These examinations were prepared to main elements (calcium, magnesium, sodium, potassium, iron, manganese). The geochemical examination provide several information about the postgenetic proceeds or the origin of the sediments grains (Liu et al, 1993; Gallet et al., 1996, 1998; Ding et al., 2001; Jahn et al., 2000; Jeong et al, 2011). Considerable fact that reaction of the elements in a geochemical impact, especially in the case of the pedogenetic process could be

different. During the pedogenesis several main elements can dissolve and disappeared from the examined level but some elements can accumulate at the place of the soil formation (Bohn et al., 1993; Gallet et al., Jahn et al., 2000; Kabata-Pendias, 2001).

The most important factor is the time in case of the examined segments. The base condition of the correlation is to define our results in the time (Sümeği, 2005). There are several absolute chronology method are available, in our case radiocarbon dating (DirectAMS Labor, Seattle, USA) and luminescence dating (IRSL) (Galović et al., 2009; Wacha et al., 2013) were used. The graphic representation of the results was executed by Psimpoll software package (Bennett, 1992). The rough radiocarbon data was calibrated with the Calib 6.1.0 software (Stuiver & Reimer, 1993). The colour of the stratigraphical units was defined on the site was used by Musell colour scale (Munsell, 1905).

3. Summary of results

3.1. Mosaic-like environmental patterns

The first question dealt with the possible paleoecological and sedimentological similarity or dissimilarity of the investigated sequences. The investigated sequences are fundamentally different to each other, of paleoecological view. Various environmental conditions developed at the same time in the investigated areas, thus it can be said, similarly to the other parts of Carpathian Basin (Sümegei, 1995, 1996b; Sümegei & Krolopp, 1995, 2002; Sümegei & Hertelendi, 1998), mosaic-like environmental patterns were developed in S-Transdanubia and E-Croatia during the Middle and Late Pleistocene.

Even some paleoecological horizons can be found similar to each other (cool/cold and mild/warm climatic periods during the MIS 2 and MIS 3 stages), but their vegetation cover was still different.

3.2. Role of Danube River in the diffusion of snail populations

The second question is about the geographic/ecologic barrier role of the Danube River. Was it possible that same or similar snail populations could develop in both banks of the Danube River? Malacological results of the investigated 5 sequences were compared with some sequences from the left bank of the river: Katymár, Hungary, (Sümegei, 2007), Madaras, Hungary (Hupuczki, 2012) and Črvenka, Serbia (Sümegei et al., 2012b).

Increasing dominance of the thermophilous *Pupilla triplicata* can be observed in all three sequences, but only in horizons older than 25,000 cal BP years. This high dominance was supplemented with the meaningful dominance of the mesophilous *Vallonia costata* at Madaras and Črvenka. This dominance pair is traceable in the mollusc fauna of Villánykövesd, Máza, Szálka and Zmajevac-upper sequences in the horizons older than 25,000 cal BP years. However this high dominance of the 2 species at Villánykövesd can be traceable until the Holocene.

At the horizons younger than 22,000 cal BP years at Madaras, a mollusc association can be observed with the significant dominance of *Succinella oblonga*, *Thocholus hispidus*, *Vallonia tenuilabris* and *Columella columella* species. Quite similar dominance relations can be discovered at the same aged horizons of Máza brickyard sequence, but the mollusc association is supplemented with the *Pupilla muscorum* species there. Similar dominances of Katymár sequence can be noticed with the absence of *S. oblonga* species. The oldest parts of Madaras and Katymár sequences are about 35,000 cal BP years old, so for the comparison of older populations the mollusc fauna of Črvenka can be used.

At the older horizons a *P. triplicata*, *V. costata* and *Helicopsis striata* species constituted mollusc association developed in Villánykövesd, Máza, and partly in Szálka sequences. The mollusc fauna of Zmajevac-middle horizon partially correspond to the same aged fauna of Črvenka. The *Cochlicopa lubricella*, *P. triplicata*, *Chondrula tridens* and *V. costata* snail association can be found in the cca. 60,000 cal BP years aged horizons at both sequences.

On the basis of these relations it is certain the mollusc fauna of the right and left bank of the river show similarities. Excluding the local snail species, concrete associations are traceable in both banks. This could be traced back to 2 reasons. The first reason could be the failure of the Danube River as a geographic/ecological barrier, in fact it could functioned as a corridor encouraging the diffusion of the snail species. The second reason could be the ancestors of the Pleistocene snail fauna have dispersed before the modern channel of Danube River had formed. After that, the snail associations developed isolated.

3.3. Determining one species' refuge area

The third question discusses marking off the refuge area of one snail species on the basis its dominance values in a loess-paleosol sequence.

Checking out the dominances of the investigated loess-paleosol sequences and recalling the definition of refuge area (where a species can survive the uncongenial climatic and environmental conditions) it is clearly obvious the

unravelling is in the Villánykövesd sequence. Aside from a few samples *Pupilla triplicata* species was found everywhere in the sequence with average dominance values around 30% and 40%.

Comparing the environmental demands of *P. triplicata* (thermophilous, dry and open vegetation lover) with the reconstructed paleoecological horizons in Villánykövesd sequence it is clear that the basically mild climatic conditions established the opportunity for Villánykövesd sequence to become a refuge area of *P. triplicata*.

3.4. Abundance changes in mollusc fauna

The fourth question is dealt with abundance changes in snail populations and the possible occasioning processes. It is known that the number of snail shells in loessy sediments is high and in paleosol layers is low or what is still worse, zero.

The process which could be the low abundance is linked with, the decarbonisation during the soil formation in the paleosol layer. Under the influence of decarbonisation the carbonates and also the constitutive elements dissolve and leave the paleosol layer (Dokuchaev, 1879; Ding et al., 2001). Most of the snail shells are contained of carbonates (calcite or aragonite) (Soós, 1943), thus with the other carbonates the material of them could also dissolve and migrate from the pedogenic layer. Under the paleosol layer, in the carbonate accumulation horizon the carbonate precipitates contain only the constitutive elements, thus the shells themselves practically become nothingness. Consequently the main abundance influencing factor is the decarbonisation in paleosol layers, but some postgenetic processes (weathering) even could alter the original abundance values in a whole sequence.

So the non-pedgenetic weathering processes could have effect on the whole mollusc fauna in one sequence. The intensity of weathering could be linked to local climatic factors, in the majority of cases to precipitation. Consequently higher amounts of precipitation could conclude by examining a moderately weathered loess-paleosol sequence. Since calcite dissolves in neutral and slightly acidic pH, the more precipitation input could start the

dissolution of snail shells. Of course precipitation-caused dissolution of snail shells affects the abundance values, for example at Szálka loess-paleosol sequence, where a moderately weathered sequence was reconstructed with sparse fossil content.

3.5. Correlating of a sequence without absolute chronological data

The fifth question concerned that is it possible to correlate a segment without absolute chronological data with other segments which have? For example at Szálka loess-paleosol sequence which contained too few fossils for radiocarbon dating. If we could correlate our segment with the help of attributes which take into consideration, such lithological qualities, and results of sedimentological analyses or with the mollusc fauna of the segment.

In our case the lithological structure of the segment from Szálka showed considerable difference from the structure of others rather shows similarity with the segments from Máza but in this case there was great difference among the situation of the paleosol layers. If we take the sedimentological results into account it was noticeable that the segment of Szálka was the most weathered among the examined 5 segments. Thus the correlation on the base of this index was not possible without an only exception which was the MS-curve.

With the help of the MS-curve was compared the loess-paleosol segments (Kukla et al., 1987; Zhou et al., 1990). The MS-curve of Szálka showed similarity with the curve of Máza and Villánykövesd loess-paleosol sequences, thus it is supposable these segments evolved in the same time. Notwithstanding the malacological matter and the conditions of dominance could be helpful during the correlation. The conditions of the dominance of a certain species could show similarity in several segments during a period. Noticeable the increasing dominance of the *P. triplicata* and maybe the *V. costata* species in those layers older than 25.000 cal BP years (mentioned above) in the case of the segments from Szálka, Máza, Villánykövesd and Zmajevac.

It can be told that if a segment does not have absolute chronological data but undergone on malacological and MS measures there will be a good chance to correlate with other segments which have absolute chronological data.

3.6. Importance of the multi-proxy examinations

The sixth and last question was about the multi-proxy examination series which was executed on a segment, what kind of supplementary information could provide in addition the malacological examinations. The aim of the malacological examination is to reconstruct the changes of the narrow environment of a segment, for example temperature conditions, vegetation covering and humidity. With the help of these three data, a complex paleoecological reconstruction could make. But we have to take several important factors into consideration.

However the malacological examination does not provide information about those proceeds which affect the sediment or the evolving conditions of the fossiliferous sediments. The organic matter and carbonate content analyses show the weathering intensity of a sequence and also give information about the location of decarbonisation levels.

The examination of the grain-size distribution gives information about the intense of weathering thus could draw consequence about the quantity of the precipitation on a certain area. The proportion of the change of coarse fraction (fine sand 63-125 μm) shows the evolving of the conditions of the air current and water flow during the sedimentary process.

The MS examination helps to show the weathered soil levels and also applicable for correlation of several segments. The separation of weathered levels and the intense of the weathering could be determined with the geochemical main element examination.

After these if we supplement the malacological examination with the multi-proxy examinations get a multiple paleoecological reconstruction thus produce an overall view from those environmental and climatic conditions which was existed during the sediment evolving.

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