INVESTIGATIONS ON FRESHWATER LIMESTONES FROM THE CARPATHIAN-BASIN: PALAEOCLIMATOLOGICAL AND SEDIMENTOLOGICAL STUDIES

Theses of the PhD dissertation

SÁNDOR KELE

Eötvös Loránd University, Faculty of Science
Doctorate School in Earth Sciences
Doctorate Program in Earth Sciences

Head of Doctorate School in Earth Sciences: Dr. Miklós Monostori D.Sc., professor
Head of Doctorate Program in Earth Sciences: Dr. Miklós Monostori D.Sc., professor

Supervisor: Dr. Csaba Szabó, PhD, associate professor
Consultant: Dr. Attila Demény, D.Sc., director

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Introduction and aim of the work

Freshwater limestones (travertines) deposited from warm and hot springs, and calcareous tufa formed from ambient temperature karstic water are frequent deposits in the Carpathian Basin. During the Pliocene and Pleistocene the area of the Gerecse and Buda Mts. were characterised by strong hydrological activity, depositing more than hundred individual travertine occurrences. After the first field observations (Kormos and Schréter 1916, Schréter 1953) sedimentological and morphological investigations on travertines became popular research subjects after the 1970's (Scheuer and Schweitzer 1988). The travertines of the Gerecse and Buda Mts. were related to the terrace levels of River Danube and its tributaries by Pécsi (1959) and Kretzoi and Pécsi (1982), based mainly on geomorphological, sedimentological and biostratigraphical considerations. Although further detailed studies have been conducted (e.g. Bakacsi 1993, Korpás *et al.* 2004, Kele *et al.* 2003, etc.), stable isotope analyses of travertines were almost completely missing in Hungary until the early 2000's (except Szöőr *et al.* 1992, Korpás 2003, Kele *et al.* 2003). Radiometric age data from these deposits have rarely been reported (e.g. Pécsi 1973, Kretzoi and Pécsi 1982, Hennig *et al.* 1983), although they are essential for exact palaeohidrological, palaeoclimatological and tectonic conclusions.

During my PhD work – based on mainly sedimentological studies, stable isotope geochemical analyses, and radiometric (U/Th series) age data – I' ve put the travertines of the Gerecse and Buda Mts. in a new chronological frame; I clarified their depositional environment and the connection between travertines and the Danube terrace system; I determined the incision rate of Danube (uplift rate of the Mts.); I defined certain periods of travertine formation in Hungary and I fit these stages to the global paleoclimatological model. With a complex sedimentological-geochemical study of the recent Egerszalók travertine I described many general observations related to the stable isotope fractionation processes during travertine deposition, which can be applied during the research of fossil travertines, too.

Applied methods

The Egerszalók travertine and associated thermal water were sampled along sections with increasing distance from the spring's orifice with the most important physico-chemical parameters (i.e. T, pH, Ec) measured on the field. I performed petrographic studies on the travertines and determined their mineralogical, stable carbon- and oxygen isotope- and trace element composition, completed with electron microprobe observations. With GPS measurements I determined the geographic position and elevation of more than hundred travertine occurrences from the Gerecse and Buda Mts. I performed petrographic observations on the main lithofacies types of each occurrences (on 360 samples) using optical microscopy and I completed more than 625 stable carbon and oxygen isotope measurements using Finnigan MAT delta S and Finnigan delta Plus XP mass spectrometers in the stable isotope laboratory of the Hungarian Academy of Sciences, Institute for Geochemical Research. I completed 87 U/Th dating measurements in the laboratory of the National Taiwan University (NTU) on 58 travertine samples collected from 53 sites from the Gerecse and Buda Mts. using Thermo Electron Neptune mass spectrometer and Multi Collector – Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS) technics.

Theses

1. With the sedimentological, mineralogical, stable isotopic and trace element analyses of the recent Egerszalók travertine and thermal water I proved that during the deposition of travertines the morphology-related CO_2 outgassing, evaporation and microbiological activity played the major roles. My investigations confirmed that the stable carbon and oxygen isotope values of the precipitating travertines increases downstream due to the three factors mentioned above. I proved that the local depositional environments of travertines (i.e. vent, channel, pond, proximal slope with microterraces and small cascades, distal slope and marsh-pool environments) can be distinguished not only by means of field and petrographic observations, but also by trace element and stable isotope analyses. This could be of high importance, if we are interested in the origin of the carbonate depositing fluids, since in this case it is necessary to distinguish the secondary processes, that can affect significantly the isotopic composition of travertine deposited. The effect of these processes are getting stronger with increasing distance from the thermal well. My observations can also be used to distinguish depositional environments and facies in case of fossil travertines as well.
2. I pointed out that the $\delta^{18}O$ values of the Egerszalók travertine show isotope shifts away from the equilibrium fractionation curve of Friedman and O'Neil (1977), due to the rapid calcite precipitation and transportation along the flow path. The data closely follow the temperature–$\Delta^{18}O_{calcite–water}$ relationship observed for other travertine localities, which is slightly displaced from the experimental curve and determines an empirical 'travertine curve'. Based on my studies this shift (and the use of the new 'travertine curve') may result in an approximately 8 °C difference in paleotemperature calculations (Kele *et al.* 2008). This correction is valid for

the travertines precipitated at the vent, where the secondary processes (carbonate transportation and mineralogical changes) are neglected.

3. In my dissertation I clarified the genesis and depositional environments of numerous travertine occurrences from the Gerecse and Buda Mts. by field studies, sedimentological and geochemical analyses. Based on my studies I found that the travertine occurrences of the Gerecse formed in characteristically lower temperature lakes (e.g. Les-hegy, Süttő, Dunaszentmiklós), whereas the travertines of Buda Mts. precipitated from higher temperature, warm or hot thermal water, reflecting hydrothermal effects. Although the remnants of spring cones (e.g. Gellért-hegy Ifjúsági-park, Törökvész lejtő) are common in the Buda Mts., the majority of the travertine occurrences formed also in shallow lakes (e.g. Buda Castle-Hill, Budakalász, Üröm-Hill, etc.) and travertine cones occur also in the Gerecse (e.g. Alsóvadács).

4. With my U/Th measurements I demonstrated that the travertines of the Gerecse and Buda Mts. are significantly younger than considered before, based on the "traditional terrace-system", geomorphological considerations, using their elevation above the sea level and paleomagnetic age data (e.g. Pécsi 1959, Scheuer and Schweitzer 1988). The U/Th analyses showed that the Upper-Pannonian freshwater limestone formation intensive spring activity characterised both mountains during the Middle-Pleistocene. After the Middle-Pleistocene, in both the Gerecse and Buda Mts. 3-3 periods can be characterised by intensive travertine formation and I named them B-I., B-II., B-III. and G-I., G-II., G-III. periods. My measurements confirmed, that there is no age data that proves travertine formation in the Buda Mts. from 180 ky until the Holocene. Differently, in the Western-Gerecse travertines have been formed also in the past 100 ky (e.g. Tata, Porhanyó-bánya, Öreg-tó, Angol-kert).

5. Based on my U/Th age data and using the altitude of travertine localities above sea level defined by GPS measurements, I estimated accurately and separately the uplift rate of the different areas covered with travertine (i.e the incision rate of River Danube and its tributaries). I proved that the minimal average incision rate was around 0.3-0.4 mm/year in the Gerecse and Buda Mts., whereas the incision rate of the individual sub-areas in both mountains may have varied in a certain degree (e.g. Gellért-Hill: 0.47-0.52 mm/year; Buda Castle-Hill: 0.11-0.14 mm/year). The difference in the incision rates of the individual sub-areas points to the fact that the elevation of the travertines does not definitely show their relative age, which means that it is possible to make only rough estimations regarding the changes of paleokarstwater-level using the elevation of travertines.

6. The most obvious method to define the age of the different fluvial terraces is the age determination of terrace-cover deposits (e.g. travertines). Based on my U/Th measurements I concluded, that in the Buda Mts. the travertines at higher elevation (400-500 m asf) are the oldest, and the travertines covering the tI-tIII terrace levels are the youngest. The previously published U/Th data (<360 ky) served as a good base for the dating of the tI-tIV terrace levels situated on lower elevations above the sea level, but the age of the tV-tVII levels was previously determined older than 780 ky (Upper-Pliocene - Lower-Pleistocene), based on mainly geomorphological considerations, sporadic paleontological findings and paleomagnetic measurements. With my U/Th measurements I proved that the travertines covering the tV-tVII terraces are much younger than it was believed before and were formed mostly during the Middle-Pleistocene. I concluded that both the Gerecse and Buda Mts. were characterised by an intensive paleospring activity during the Middle-Upper period of the Middle-Pleistocene, and as a consequence numerous travertine occurrences were formed mainly on the former fluvial terrace deposits. I pointed out that in the Buda Mts. the age of the travertines located on the same elevation above sea level can differ significantly, whereas in the Gerecse there is a closer relationship between altitude and travertine age.

7. With the radiometric age data of travertines I could reconstruct the migration and development of paleokarst springs. In the Upper-Pannonian the travertine formation in the Buda Mts. was characteristic of the Széchenyi-hegy-János-hegy area (>400 m asf). Later on the spring orifices moved eastwards and in the Middle-Pleistocene a significant spring-migration and thermal spring activity started in the Buda Mts., while the northern part of Budapest, and the territory between the Solymár-Valley and Ős-Dera Creek became the center of water discharge. The travertine formation in the Budakalász area (210-240 m asf) started about 550-600 ky ago, while in the Ördögárok-Valley the Máriaremete travertine occurrence (280 m asf) was formed only 400 ky ago. During the Middle-Pleistocene travertine was formed on the Buda Castle-Hill, and 350 ky ago the Rózsadomb area (160-190 m asf) was also characterised with intensive thermal spring activity. About 250 ky ago there was a decrease in the discharge level of the springs and the springs moved eastwards, towards River Danube, while the individual areas (e.g. Rózsadomb, Gellért-Hill, Buda Castle-Hill) uplifting differently. Travertines of the same age located at different elevations are the evidences of the different uplift rate (excluding downward sliding). The springs, from wich the Máriaremete travertine occurrence deposited discharged to the surface in E-SE direction at lower elevation in the Ördögárok-Valley about 200-250 ky ago. Due to the further relative decrease in the water level, about 240 ky ago travertine formation started at the Farkastorki str. (150-160 m asf) and 180 ky ago on the Kiscelli plateau (140 m asf) and the springs moved in EW direction from the Rózsadomb area. Based on my measurements the presence of travertines from the Upper-Pleistocene until the Holocene is not evidenced in the Buda Mts. The present discharge level of the thermal springs might have been evolved around the end of Pleistocene adn early Holocene.

8. With my measurements I proved that in the Gerecse between 250-350 m (asf) generally those travertines deposited, which are older than the upper limit (600 ky) of the U/Th method. The oldest travertine occurrences are located in the Western, Central and Eastern Gerecse, towards the centre of the Gerecse. During the Middle-Pleistocene, the discharge points of thermal springs in the Gerecse moved to the marginal areas and the spring activity started at lower (150-250 m asf) elevations (e.g. Vértesszőlős). After the deposition of the Middle-Pleistocene travertines of the central parts of Gerecse (e.g. Alsóvadács: 329 m asf; Vékonycser: 238 m asf) the travertines of Réz-Hill (466 ky) and Öreg-Hills (391 ky) at 225-235 m (asf) were formed in NE direction from the Middle-Pleistocene occurrences. The Süttő travertine occurrence, which is located on the nothern part of Central-Gerecse (220-250 m asf), was considered previously as Upper-Pleistocene-Lower-Pleistocene, but according to the new U/Th data it was formed during the Middle-Pleistocene as well. During the Middle Pleistocene in the Eastern-Gerecse the spring orifices also moved eastwards. During the Upper-Pleistocene the thermal spring activity and travertine formation moved to the surroundings of Tata (Porhanyó-quarry), while at the same time travertine formation in the Central- and Eastern-Gerecse cannot be proved with age data. The travertine, which is located at the eastern side of the Öreg-Lake (Tata) was formed 50 ky ago, and further Holocene and recent occurrences can be found in the Angol-kert at Cseke-Lake, at the Fényes-springs and at the Csokonai-spring (Dunaalmás, 110-140 m asf).

9. Based on my analyses, the $\delta^{13}C$ és $\delta^{18}O$ values of the travertines of the Gerecse and Buda Mts. deviate characteristically from the recent calcareous tufa precipitations of the Bükk and Mecsek Mts. (e.g. Sebesvíz, Tettye), which have been deposited from cold karstic water. The low $\delta^{13}C$ and high $\delta^{18}O$ values of the recent calcareous tufa deposits are due to the low temperature of karstic water, elevated contribution of the organic matter and the close relationship with climate. I proved that there is a significant difference between the $\delta^{13}C$ values of travertines of the Gerecse and Buda Mts. I found that the $\delta^{13}C$ values vary primarily in spatial distribution, however they reflect different microfacies types and climatic signals as well. There is a significant difference in the $\delta^{13}C$ values between the travertines of the Gerecse and Buda Mts., possibly due to the difference in the nature of spring activity. Besides, longer residence time of water in the bedrock and the length of the underground flowpath can also increase the $\delta^{13}C$ values of travertines, due to the longer contact with marine limestones.

In the Gerecse Mts. I found spatial variations in the $\delta^{13}C$ values of travertines in SW-NE direction. The low $\delta^{13}C$ values of the travertines at Tata might be explained with the contribution of the organic CO_2, but the role of magmatic CO_2 can not be excluded. In the Buda Mts. the $\delta^{13}C$ and $\delta^{18}O$ values are mainly connected to the age and genetics of the occurrences. The Upper-Pannonian travertine occurrences near the Szabadság-Hill have a more positive $\delta^{18}O$ and negative $\delta^{13}C$ values compared to the Pleistocene and Holocene travertines of the Buda Mts. situated at lower altitudes. The $\delta^{13}C$ values of the Pleistocene and Holocene travertines are less negative than the values of the Upper-Pannonian travertines, which can be related to the dominance of carbon deriving from dissolution of ^{13}C rich marine limestones. Since the individual mountains and their sub-areas can be characterized by distinct stable isotope values, the stable isotope measurements are useful to define the location of travertine formation.

10. Using the new U/Th age data series obtained on the travertines of the Gerecse and Buda Mts. I could complement the age histogram curve (created from numerous U/Th data measured on travertines younger than 350 ky) of Pentecost (2005) up to 500 ky, showing a frequency peak during the Mindel/Riss interglacial, too. Based on my studies the formation of travertines was characteristic not only during the interglacials (Mindel/Riss, Riss/Würm), but also during glacials (e.g. Riss), and travertines precipitated in every periods, when the precipitation was enough to provide water supply for the springs. With my studies I demonstrated that in Hungary during glacials humid periods could be also occurred, which favoured travertine deposition. With the detailed stable oxygen isotope analyses of travertine occurrences I showed that the Hungarian travertines are suitable to record the global change in climate. The correlation of the stable oxygen isotope data measured from Hungarian travertines with the curve created from stable oxygen isotope composition of planktonic foraminifera, and correlation with the paleotemperature curve created from the Vostok ice core (Petit *et al.* 1999) highlights the role of Hungarian travertines in the paleoclimate research. In case of freshwater limestones similar correlation with global climate signals has not been mentioned yet.

Conclusions

During my PhD research work I proved that the sedimentological and geochemical analyses of the Hungarian freshwater limestones, together with radiometric dating methods are suitable to provide important palaeoenvironmental, palaeoclimatic and tectonic information. The study of the recent Egerszalók travertine resulted in many general observations, that can be used during the investigations of fossil travertines, too. With the help of the new U/Th data of freshwater limestones of the Buda and the Gerecse Mountains it was possible to reconstruct the migration and development of paleokarst springs, to clarify the relation between freshwater limestones and former Danube-terraces, and to calculate the uplift rate of the Gerecse and Buda Mts. (incision rate of River Danube). Petrographical analyses of the freshwater limestones collected from almost 100 occurrences helped to determine their depositional environment. Based on the new U/Th age data the periods of freshwater limestone formation were determined, in comparison with glacial-interglacial periods. The results of my thesis may serve as a base of further detailed studies on individual travertine occurrences, providing new data on the paleoclimatology of the Carpathian Basin and on the local paleoenvironmental and tectonic conditions.

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