

# **COSMOGENIC $^{10}\text{Be}$ DATING OF DANUBE TERRACES: TECTONIC AND CLIMATIC INFLUENCE ON QUATERNARY RIVER INCISION IN HUNGARY**

## **Final research report of the OTKA PD 83610**

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### **INTRODUCTION AND PRELIMINARIES**

Vertical movement of rock masses is a possible expression of neotectonic deformation. If denudation does not counterbalance vertical motion, the surface suffers uplift. In this scenario, the older are the landforms and their related deposits, the higher is their topographic position. Formation of a terrace system is one expression of such a process. On the other hand, discharge and sediment supply of a river are mainly regulated by the climate. In periods of high discharge coupled with low sediment supply the river tends to incise its valley and when discharge is low relative to the sediments to be transported, the river prefers to widen its valley and to lay down its sediment load. Accordingly, formation of a terraced valley is also possible due to climate driven surface processes, but only in an environment dominated by a constant or repeated base level drop. In an intra-continental setting insensitive to the global sea level changes most common trigger of the base level drop is surface uplift (e.g. (Bridgland, 2000; Ruszkiczay-Rüdiger, 2007; Häuselmann et al, 2007; Baotian et al., 2013; Fuchs et al., 2014; Bender et al., 2016).

In such environment, if a river is cutting through an uplifting region – like the Danube river crossing the Transdanubian Range in the Pannonian Basin – climate and tectonics are co-acting in the formation of river terraces. It is difficult to distinguish between these two signals, as both have been preserved by the terrace levels. This is why terrace altitude-age data pairs are suitable for the determination of long-term incision rates, which are a good measure of vertical deformation rates. Alternately, some characteristic climate events (like abrupt warming by the end of a glacial phase) may have led to river incision and abandonment of certain terrace levels, linking terrace carving to climate events (Gábris and Nádor, 2007; Gábris, 2008; Bridgland and Westaway, 2008; Rixhon et al, 2017).

### **Terrace studies along the Hungarian Danube valley**

Recent studies on the terraces of the Hungarian section of the Danube river (Ruszkiczay-Rüdiger et al., 2005a, 2005b; Ruszkiczay-Rüdiger, 2007; Gábris, 2008) evidenced the need of numerical age constraints for a thorough revision of the Hungarian terrace system established by Pécsi (1959). This system basically remained untouched, only slight modifications were suggested by further studies (Kretzoi and Pécsi, 1982; Pécsi et al., 1985; Gábris, 1994; 2008; Gábris and Nádor, 2007). Previous data allowed recognising differential vertical motions along the valley, with the fastest uplift in the axial zone of the Transdanubian Range, the Danube Bend. However, the former chronology, based mostly on relative age data was valid only at certain river sections and provided scarce numerical constraints of the terrace horizons (Ruszkiczay-Rüdiger, 2007).

Main objective of the proposed research was to provide new geochronological data on the abandoned terrace surfaces along the Danube river to enable a better understanding of the differential role of neotectonics and climate changes in river incision. In the framework of this project numerical age determination of the terraces was targeted by the application of surface exposure age dating using terrestrial in-situ produced cosmogenic radionuclides (CRNs).

## **Terrestrial in-situ produced cosmogenic nuclide applications in landscape evolution in Hungary**

Since the pioneering work of Lal (1991), terrestrial in-situ produced cosmogenic nuclides are of increasing importance in geochronology and in quantitative geomorphology worldwide. The method have developed at an amazing speed and converted into one of the most popular tools of Quaternary geochronology (Gosse and Phillips, 2001; Dunai, 2010; Balco, 2011; Granger et al, 2013; Phillips et al, 2016)

Application of terrestrial in-situ produced cosmogenic nuclides for surface exposure age dating in Hungary started by a ground-breaking study using a stable terrestrial cosmogenic nuclide ( $^3\text{He}$ ) on uncovered strath terraces in the Danube Bend region (Ruszkiczay-Rüdiger et al., 2005b). The first study using the CRN  $^{10}\text{Be}$  in Hungary was on wind-polished rock surfaces of the Balaton Highland (Ruszkiczay-Rüdiger et al., 2011). Results of this work suggested that age determination of landforms affected by certain degree of denudation was not possible using surface samples only. Therefore, the application of depth profiles of  $^{10}\text{Be}$  was introduced, enabling the estimation of denudation rate-corrected exposure ages (Siame et al., 2004; Braucher et al., 2009; Hidy et al., 2010).

Based on these experiences the present study put forward the application of  $^{10}\text{Be}$  depth profiles on the terraces covered by alluvial material along the Danube. The choice of  $^{10}\text{Be}$  is reasonable as it is applicable on quartz-containing lithology, and sediments deposited by the Danube are rich in quartzite sand and gravel. The depth-profile technique seemed to be necessary due to the proposed old age (several 100 ka) of the higher terraces affected by surface denudation, evidenced by the incomplete alluvial sequences and reduced thickness of terrace material (Pécsi, 1959; Ruszkiczay-Rüdiger et al., 2005a).

On the other hand, the demand of the depth profile technique for processing significant number of samples delivered the need for launching a sample preparation facility in Hungary, the second objective of the proposed research.

Previously all samples have been chemically processed at CEREGE - CNRS, LN2C (Centre Européen de Recherche et d'Enseignement des Géosciences de l'Environnement - Centre National de la Recherche Scientifique; Laboratoire National des Nucléides Cosmogéniques), Aix en Provence, France. Accelerator Mass Spectrometry (AMS) measurement of the nuclide ratios of the processed samples occurred also at CEREGE by the 5 MV AMS facility ASTER (Accélérateur pour les Sciences de la Terre, Environnement, Risques; Arnold et al., 2010).

The setup of a sample preparation laboratory for cosmogenic nuclides in Hungary appeared to be reasonable because despite the rapidly increasing international popularity of the method, its application was hindered by the lack of expertise and available laboratory background in the region. The sample preparation laboratory to be launched in the framework of the proposed research is the first in the eastern Central European region. The CEREGE has assured to be open to collaborate with the proposed cosmogenic sample preparation laboratory in Hungary in AMS measurement of nuclide ratios and finishing sample processing (if necessary).

### **THE TIMING OF THE RESEARCH**

The first research year of this study started in 01.02.2011, when the project was hosted by the Eötvös University of Budapest. During the first three months of the project I was working on a review paper on the fluvial and aeolian landscape evolution of Hungary (Gábris et al., 2012), connected to which I revised and re-evaluated previous works on Danube terraces. Then, because of the birth of my second child, I successfully asked for the suspension of my research program for 2 years of maternity leave, from 01.05.2011 to 31.04.2013.

After the maternity leave I changed my research institution: I moved from the Eötvös University, to the Institute for Geological and Geochemical Research (FGI) of the Research Centre for

Astronomy and Earth Sciences (CSFK), Hungarian Academy of Sciences (MTA). As a consequence, the host institute of this project was also changed: from 01.05.2013 it is affiliated to the MTA CSFK FGI.

Sampling and data interpretation occurred in accordance with the workplan. However, the first results were not always as expected, some study sites proved to be unsuitable or suboptimal for  $^{10}\text{Be}$  exposure age dating, hence new study areas and new CRN techniques were also introduced in the research. Besides, the laboratory setup occurred faster and to a more advanced stage, compared to the original plans. The new study areas coupled with the need for the introduction of new CRN strategies, which fortunately came together with the extension of laboratory facilities suitable for the entire chemical extraction process of cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$  from quartz containing rocks. These possibilities meant extra tasks as well, giving rise to a need for the extension of the project by 1.5 years, which I successfully applied for. This period of time was used for 1) testing new CRN dating techniques on new sample locations along the Danube river and simultaneously achieving new chronological data; 2) making laboratory tests to set and verify the reliability of the sample preparation protocol used in the newly launched Cosmogenic nuclide sample preparation laboratory of Budapest (Cosmo-lab).

## METHODOLOGICAL APPROACH AND ITS EVOLUTION DURING THE PROJECT

### Exposure age determination

Exposure age dating relies on the production of cosmogenic nuclides in the surface layers of the lithosphere (Lal, 1991). In this study a weighted mean spallation production rate at sea level and high latitude (SLHL) of  $4.02 \pm 0.36$  atoms/g  $\text{SiO}_2/\text{yr}$ . This production rate is the weighted mean of recently calibrated production rates in the Northern Hemisphere (Ruszkiczay-Rüdiger et al., 2016a). All exposure and burial age calculations and modelling, if not mentioned otherwise, were carried out using the equation (1), scaling and constants described in Ruszkiczay-Rüdiger et al., (2016a).

Exposure age of a terrace is the time elapsed since its abandonment by the river. Taking samples only from a terrace surface enables the *simple exposure age determination* in condition that neither surface denudation nor sediment accumulation have occurred since terrace abandonment. Both processes lead to an underestimation of the true terrace age, therefore simple exposure age dating is suitable for the determination of the minimum exposure age of a landform (e.g. Gosse, and Phillips, 2001; Dunai, 2010). The faster is the denudation and/or longer is the time elapsed since terrace abandonment the major will be the difference between the true and calculated terrace age. On the other hand, in case of aggradational terraces the CRN inventory of the sediment from previous exposure (so called inheritance) may lead to age overestimation if not accounted for (e.g. Anderson et al, 1996; Braucher et al., 2009).

Because of the high probability of surface denudation since terrace formation and unknown amount of inherited cosmogenic nuclides, *cosmogenic  $^{10}\text{Be}$  depth profile*-approach was applied on the gravel terraces of the Danube river. This method consists of sample collection at several subsequent subsurface depths ( $n=5-8$  or more) at the same location, down to at least 3-4 m depth. The method requires the terrace material to be undisturbed and with no hiatuses, and a well preserved original terrace surface.

The exposure age of the sampled landform is calculated considering the depth dependence of CRN production. If CRN concentrations are not in equilibrium with landscape processes at depth yet, results will converge to a unique solution of exposure age, erosion rate, and inheritance when accounting for both nucleogenic and muogenic production pathways (Siame et al., 2004; Braucher et al., 2009; Hidy et al., 2010; Rixhon et al., 2014; Stange et al., 2014; Laloy et al., 2017). If the concentration of cosmogenic nuclides is in steady state for the entire profile, a minimum age and

the time averaged denudation rate can be calculated (Siame et al., 2004; Braucher et al., 2009; Hidy et al., 2010; Ruszkiczay-Rüdiger et al., 2016a).

Sampling procedures and age calculation methodology have been described by Ruszkiczay-Rüdiger et al. (2016a).

### Burial age determination

The first results on depth profile dating of the high terraces of the Győr-Tata terraces were puzzling and difficult to interpret, as the profiles were in steady state for CRN production and loss due to decay and surface denudation. Therefore, in search of a more powerful tool for the age determination of the old terraces, some sample sets were taken to be processed for burial age determination using the pair of the CRNs  $^{26}\text{Al}$  and  $^{10}\text{Be}$ . The choice of the  $^{26}\text{Al}/^{10}\text{Be}$  nuclide pair is reasonable as both nuclides are produced and retained within the crystal lattice of quartz and can be separated from the same sample through a standard extraction chemistry procedure (Merchel and Herpers, 1999).

Burial age determination relies on the radioactive decay of cosmogenic nuclides ( $^{26}\text{Al}$   $t_{1/2} = 708 \pm 17$  ka (Nishiizumi, 2004),  $^{10}\text{Be}$   $t_{1/2} = 1387 \pm 12$  ka (Korschinek et al., 2010; Chmeleff et al., 2010) after being buried and thus shielded from further cosmic irradiation. These cosmogenic nuclides accumulate during surface exposure prior to burial in the selected mineral fraction with a well constrained  $^{26}\text{Al}/^{10}\text{Be}$  spallogenic production rate ratio of  $6.61 \pm 0.52$  (updated from the Nishiizumi et al. (1989) by Braucher et al. (2011)).

The *simple burial age determination* is based on the difference between the half-lives of the two nuclides.  $^{26}\text{Al}$  concentrations decrease roughly twice as fast as the  $^{10}\text{Be}$  concentrations.  $^{26}\text{Al}/^{10}\text{Be}$  concentration ratio decreases exponentially with an apparent half-life of  $1.48 \pm 0.01$  Ma, allowing for the estimation of burial durations from  $\sim 100$  ka to  $\sim 5$  Ma (Granger and Muzikar, 2001; Granger, 2006).

If the burial is not thick enough ( $>10$ - $15$  m) to efficiently stop cosmogenic nuclide production (like it frequently occurs in the case of fluvial terrace material of a few meters thickness), CRN concentrations measured in the samples correspond to the sum of the inherited cosmogenic nuclide concentrations (accumulated during previous surface exposure and denudation), that have undergone radioactive decay and of the cosmogenic nuclide concentrations accumulated during the burial period at the sample depth. This simple approach is represented by the exposure-burial diagram, where by plotting the  $^{26}\text{Al}/^{10}\text{Be}$  ratio against the  $^{10}\text{Be}$  concentration of the samples (Granger 2006, see more in Results section). This postburial CRN inventory increases CRN concentrations in the rocks at depth, therefore may lead to an underestimated burial age if not accounted for. Accordingly, this approach will provide a minimum burial age estimate. Assuming stable conditions after burial, accounting for postburial CRN production (steady denudation rate since burial) leads to a maximum age estimate. If no further geological constraints are available, these bracketing ages can be used to assess the length of the burial period (Granger, 2006, Lebatard et al., 2014). This approach has been used in the Gerecse Hills during the second part of the research period.

Another possible way to overcome the possible age underestimation caused by the postburial CRN production is the application of *isochron burial dating* (Balco and Rovey, 2008). Isochron burial dating uses several distinct samples (i.e. 4-6 individual cobbles, preferably of different lithology) from the same subsurface depth, within the same stratigraphic horizon. Accordingly, the sample set shares the same post-depositional history, which therefore can be taken as a constant. If the sampled cobbles have been deposited with a wide enough range of initial nuclide concentrations (different denudation rates in the source area), on a Be-Al diagram their nuclide concentrations will plot on a straight line providing that the inherited  $^{26}\text{Al}/^{10}\text{Be}$  ratio (R) is constant. The slope of this line depends on the burial duration and is independent of the postburial production (Balco and Rovey, 2008). As post-depositional CRN inventory is constant for all samples, the method is not

sensitive to shallow subsurface depths and surface denudation (as long as the inherited nuclide inventory remains significant compared to the post-depositional production) (Bender et al., 2016; Erlanger et al., 2016.; Zhao et al, 2016).

This method requires the samples to arrive with different inherited amounts of CRNs, but with the same  $^{26}\text{Al}/^{10}\text{Be}$  ratio, i.e. no sediment storage occurred within the catchment, that could reduce the inherited ratio of a few samples to an unknown value and therefore would prevent them from plotting on the isochron. This technique has been applied on a Danube terrace in the Vienna basin during the last phase of the project.

## **DEVELOPMENT OF THE COSMOGENIC NUCLIDE SAMPLE PREPARATION LABORATORY**

### **Setup**

The setup of the Cosmogenic nuclide sample preparation laboratory (Cosmo-lab) started in 2013, in the FGI. The installation of a separate room for the Cosmo-lab equipped with a new ventilation hood, suitable for strong acid treatments including evaporation of HF (which processes are part of the extraction chemistry of in-situ cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$ ) has been supported by the CSFK.

During the 2<sup>nd</sup> year of the project (2014) the Cosmo-lab was equipped for the quartz separation and subsequent chemical leaching (the first, so called “dirty-lab” processing phase, delivering the cleaned quartz aliquots (20-40 gr/sample). These aliquots are ready for total dissolution and subsequent extraction chemistry (second, so called “clean-lab” phase). To prepare the Cosmo-lab for the “clean-lab” phase, appeared to be outside the conceivable objectives of laboratory construction proposed by the workplan and budget of this project.

### **First results**

To test the suitability of the facility of the Cosmo-lab, a sample set of moraine boulders from the Retezat Mts (Southern Carpathians) was processed for exposure age determination using in-situ cosmogenic  $^{10}\text{Be}$ . This sample set, together with the depth profile samples from Ács (Győr-Tata terraces) were the first to be partially processed in the Cosmo-lab (physical sample preparation and cleaning of the quartz) in 2014. The AMS measurements (at ASTER) delivered good results: the novel  $^{10}\text{Be}$  data for the moraine samples provided a consistent dataset with a formerly published and recalculated sample batch (Ruszkiczay-Rüdiger et al, 2016b), suggesting that the first, “dirty lab” phase of sample processing could be safely done in the Cosmo-lab of Budapest.

### **Laboratory expansion**

In the CSFK I have found several unforeseen possibilities compared to the expectations at the time of project submission 3 years earlier in a different institution. Most important among these were the new ventilation hood and availability of several essential “clean-lab” equipments, like the ultra-pure water system in the Soil laboratory of the Geographical Institute (FTI) and the laboratory centrifuge, high temperature oven in the FGI. Besides, in-house availability of sieving facilities in the FTI and accessible quality control of the cleaned quartz by optical microscope and also by XRD and XRF in the FGI supported the progress of the Cosmo-lab.

These capacities allowed me to use the funding of the present proposal for the further development of the Cosmo-lab. As a result, in the coming year it was possible to prepare the Cosmo-lab for the entire sample processing for both in-situ cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$ , including extraction chemistry by ion chromatography and production of oxides by ignition.

In the autumn of 2016 the Cosmo-lab has been complemented by a second room, also equipped with a proper ventilation hood. This permitted the quartz cleaning (“dirty-lab”) phase and the

extraction chemistry (“clean-lab”) phases of the sample processing to be physically separated, which was a great improvement towards safer and faster sample treatment.

These achievements mean that the Cosmo-lab of Budapest is now able to produce BeO and Al<sub>2</sub>O<sub>3</sub> from quartz-bearing rock samples (<http://www.geochem.hu/kozموgen/lab.html>), ready to be loaded in the AMS cathodes, which is far beyond the planned level (“dirty lab” phase only) in the framework of this project.

### **New possibilities and tasks**

The above described technological development had 2 main consequences:

(1) The research along the Danube valley had the chance of applying new CRN techniques, which have amplified significantly the possibilities of age determination.

(2) The outcome of the “clean-lab” must have been tested for low and stable background levels and for the accuracy and repeatability of the cosmogenic <sup>10</sup>Be and <sup>26</sup>Al concentrations of the processed aliquots. Special attention had to be paid for samples to be processed also for <sup>26</sup>Al for several reasons: (i) Aluminium is abundant in the lithosphere, therefore thorough cleaning of the quartz aliquots from any trace of other Al containing minerals is essential. (ii) During the dissolution of the quartz and evaporation of HF Al may form insoluble fluoride precipitates, which may lead to loss of Al from the sample. (iii) Independent determination of the <sup>27</sup>Al content of the sample is necessary, as unlike the <sup>9</sup>Be, small amount of <sup>27</sup>Al may be present in the quartz. Additionally, as previously I’ve been working mainly with <sup>10</sup>Be samples, I had little experience with the behaviour of <sup>26</sup>Al samples.

After the first experiments, some methodological improvements were introduced in the Cosmo-lab with ongoing testing (see more in the Results section). Geochronological and methodological achievements of the last sample sets were published on conferences (Ruszkiczay-Rüdiger et al., 2016c, 2017; Neuhuber et al., 2016).

### **Quality control on processing phases in external laboratories, testing independent <sup>27</sup>Al determination possibilities**

External laboratories have been involved in two steps of sample preparation: crushing of rock samples (1) and determination of stable <sup>27</sup>Al content (2).

(1) Until 2017, the first step of the physical treatment of rock samples - i.e. the crushing and grinding - occurred at the Eötvös University (Budapest) using Retsch BB200 Jaw Crusher. Safe handling and elimination of cross contamination was guaranteed as I was allowed to handle the equipment myself.

In 2017, in the framework of the “Proposal for the development of research infrastructure” of the Hungarian Academy of Sciences, I assisted to the successful application of the FGI for the newest jaw crusher model (Fritsch Jaw Crusher Pulverisette 1, Model 2, Premium Line) (IF-004-2017). The crusher has arrived and has been installed in the FGI in July 2017; meaning that only one step remained to be done by an external laboratory.

2) Determination of native stable <sup>27</sup>Al content of samples for paired <sup>26</sup>Al/<sup>10</sup>Be analysis is essential (e.g. Gosse and Phillips, 2001) to decide whether Al carrier is to be added to a sample or not. Its uncertainty will directly affect the age/denudation rate results, as the cosmogenic <sup>26</sup>Al concentration is calculated from the <sup>26</sup>Al/<sup>27</sup>Al AMS ratio using the measured value of the native <sup>27</sup>Al (plus amount of the added carrier, if necessary). Therefore, accurate <sup>27</sup>Al measurements are of primary importance.

Aliquots for <sup>27</sup>Al determination were analysed by an Agilent MP-AES 4100 instrument at the Hertelendi Laboratory of Environmental Studies of the Institute for Nuclear Research of the MTA (HEKAL, Debrecen, Mihály Braun). To test the correctness and precision of HEKAL instrument duplicate samples were sent to be measured at the CEREGE by ICP-OES (n=23). The <sup>27</sup>Al values (converted to ppm

in the quartz dissolved) resulted to be systematically higher by 2-6% if measured by MP-AES compared to values provided by ICP-OES.

In the following, another sample set has been analysed by ICP-MS at the Nuclear Security Department, Centre for Energy Research, MTA (NSD; Budapest, Kornél Fél) and duplicates have been sent again to the CEREGE. The results from CEREGE have not arrived yet.

Besides, two samples were processed for a round robin test of the determination of their  $^{27}\text{Al}$  content. The participating instruments: MP-ES (HEKAL), ICP-MS (HEKAL), ICP-OES- (CEREGE), ICP-MS (NSD). (A sub sample has also been sent to an ICP-MS at the Geological Survey of Vienna, but the measured values were significantly (>70%) off for both samples compared to the values measured by the other laboratories, therefore this lab was excluded from the test). According to the first results of this test, the  $^{27}\text{Al}$  concentrations experienced a ~4% standard deviation around the mean of all values, with a maximum difference of 12%. These differences seem to be small, but converting them into ages they may cause up to 50% difference in burial age for short burial times (<200 ka). For longer burial time this difference reduces to values ~10%, and for burial times over 1 Ma to values <5%. To sum up, determination of native  $^{27}\text{Al}$  is of major importance, and has a considerable effect, especially for lower burial times. The testing is going on to find the best cooperation partner.

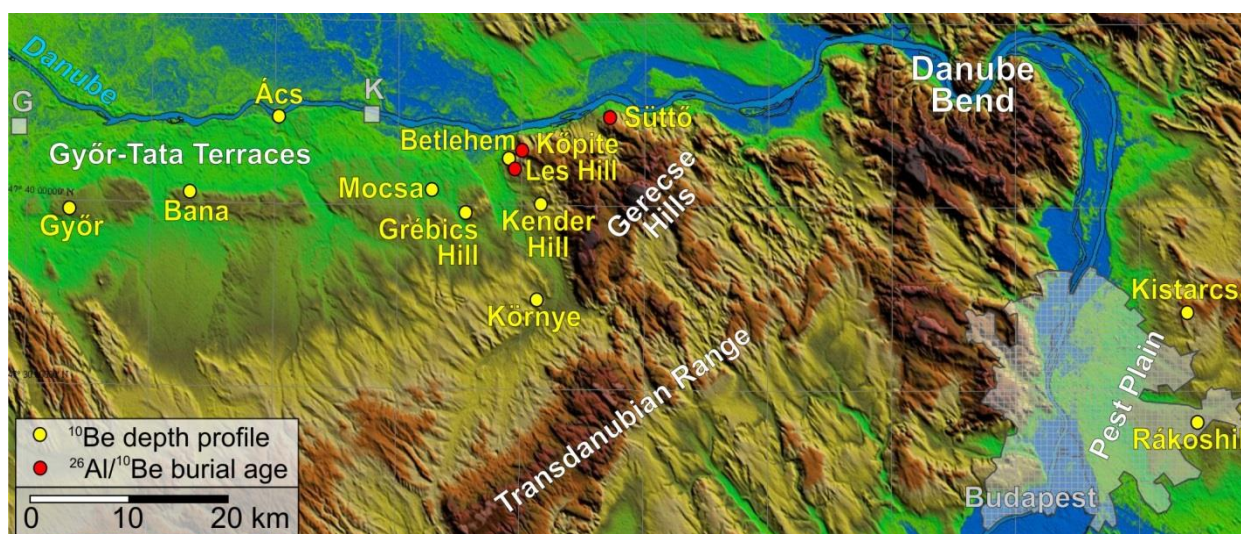
### Accelerator Mass Spectrometry

Both Beryllium and Aluminium nuclide ratios of the samples processed by the Cosmo-lab were measured by the ASTER facility at CEREGE-LN2C, Aix en Provence, France in the framework of scientific cooperation.

During the duration of this project I visited the Cosmogenic Nuclide Laboratory at CEREGE three times for consultation and to finish sample processing (2014-2015). These visits were supported by a bilateral MTA-CNRS project (NKM-31/2014-2015).

### GEOCHRONOLOGICAL RESULTS

In the Hungarian Danube valley two lowland areas the Győr-Tata Terrace region and the Pest Plain; and a hilly region, the Gerecse Hills were targeted areas for terrace age determination by this project.



**Fig.1.** Sample locations (yellow and red dots) of CRN age determination in the Hungarian Danube valley

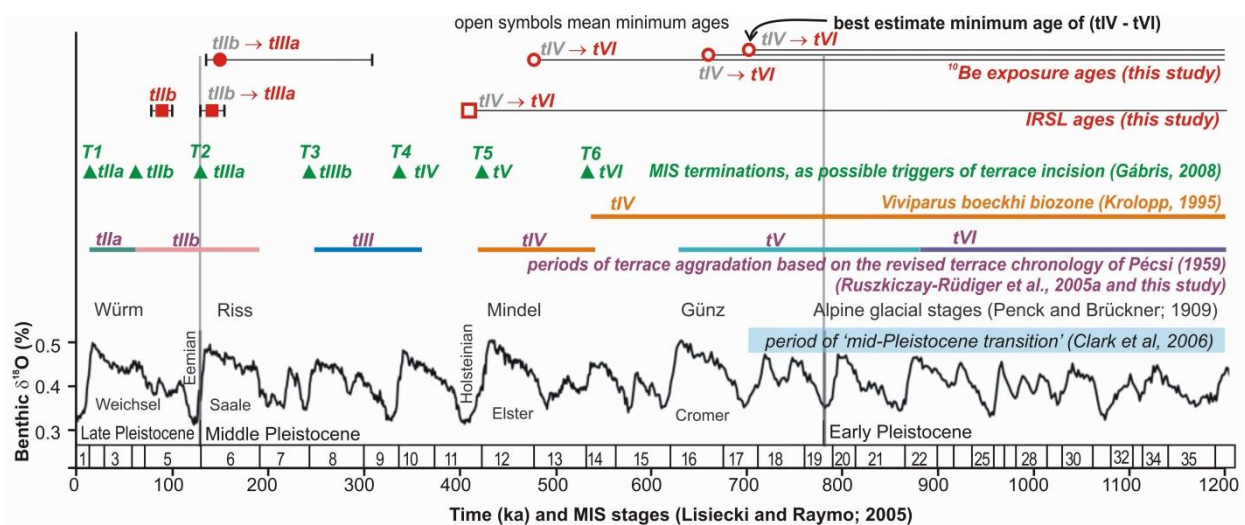
## The Győr-Tata Terraces

A total number of 53 samples was collected and analysed at 6 locations (Győr, Bana, Grébics, Mocsá, Ács, Környe; Fig.1) in the Győr-Tata terrace region in order to establish the chronology of terrace formation by  $^{10}\text{Be}$  exposure age dating of terrace sediments. To achieve a more robust terrace chronology, CRN dating was combined with luminescence dating (post-IR IRSL measurements on K-feldspar). The results were published by Ruszkiczay-Rüdiger et al. (2016a), therefore only a short summary will be provided here.

Because of the high probability of surface denudation since terrace formation and unknown amount of inherited cosmogenic nuclides, the cosmogenic  $^{10}\text{Be}$  depth profile approach was applied. To model the exposure age-denudation rate-inheritance triplets using the concentration-depth relationship of the CRN  $^{10}\text{Be}$  depth profile data  $\chi^2$  minimisations by Monte Carlo simulations were performed (Hidy et al., 2010) to obtain the denudation rate - corrected exposure ages.

Remnants of the highest terrace horizon of the study area provided a best estimate erosion-corrected minimum  $^{10}\text{Be}$  exposure age of >700 ka. We proposed that the abandonment of the highest terrace of the Hungarian Danube valley was triggered by the combined effect of the onset of tectonic uplift and the beginning of major continental glaciations of Quaternary age (mid-Pleistocene climate transition; ~1.2-0.8 Ma; Clark et al., 2006), a process described for several river systems in Western Europe (Häuselmann et al., 2007; Gibbard and Lewin, 2009). The >700 ka age is considerably older than the formerly suggested >420 ka age of this horizon, therefore it rather appears to be coeval with the tVI horizon than with the tIV of the “traditional terrace system”. For these old terraces both Post-IR IRSL and CRN dating was able to provide minimum ages only, nevertheless, minimum ages of the CRN method were considerably older, thus most probably closer to the true age of the terraces.

For the lower terraces the new chronology enabled to differentiate between a higher and a lower level of the former tIIb terrace, maintaining the name tIIb for the lower one (~90 ka; MIS 5b-c) and assigning the higher-older level (~140 ka; MIS 6) to the tIIIa of Gábris, (2008). However, for these young horizons affected by surface processes (e.g. denudation, cryoturbation) the post-IR IRSL method proved to be a more suitable chronometer than the age determination using CRN depth profiles (Fig.2). A correlation of the terrace carving periods to MIS terminations was possible for some of the lower terraces, but for higher terrace formation could not be assigned to certain climate periods, most probably because of the poor resolution of the data (Fig. 2).



**Fig. 2.** Periods of terrace formation in the Hungarian Danube valley according to previous studies and results of the terrace age determinations plotted above the benthic  $\delta^{18}\text{O}$  record and MIS stages (Fig.4. of Ruszkiczay-Rüdiger et al., 2016a).



Surface denudation rates were well constrained by the cosmogenic  $^{10}\text{Be}$  depth profiles between 5.8 m/Ma and 10.0 m/Ma for all terraces. The presented CRN and post-IR-IRSL ages are the first numerical age data on these terraces, and the CRN denudation rates are the first constraints on surface denudation affecting this area.

The minimum terrace ages were used for the calculation of maximum incision/uplift rates of the Danube river, relevant for the above determined >700 ka time span. Incision rates increase from west (<0.06 mm/a) to east (<0.13 mm/a), toward the more elevated Transdanubian Range. The higher Late Pleistocene incision rates derived from the age of the lower terraces (0.13-0.15 mm/a) may suggest a slight acceleration of uplift towards present.

In addition, one further sample set was collected in the hinterland of the Győr-Tata terraces on a supposed pediment postdating the highest terraces (fQ4, >300ka; Csillag et al., in prep) of the Győr-Tata region (Környe, n=5). However, this outcrop was shallow (140 cm), with a lithological change around 1 m depth, therefore could not provide enough information for a proper age-erosion rate modelling. The cosmogenic in-situ  $^{10}\text{Be}$  data could be interpreted in two ways:

(1)  $^{10}\text{Be}$  concentrations are interpreted as the minimum exposure age of this site suggest a very young, ~50 ka exposure age, clearly unconformable with the geomorphological position of the site (Csillag et al., in prep). Therefore, this age would propose a relatively young truncation event.

(2)  $^{10}\text{Be}$  concentrations interpreted as denudation rates put forward a ~20-25 m/Ma surface denudation, in which scenario no information on the deposition age of the sediment could be retained by the  $^{10}\text{Be}$  inventory of the sediment.

Neither version could provide an age estimate of the targeted pediment surface, and the location was only marginally connected to the Győr-Tata terraces, therefore this site was omitted from the published dataset of Ruszkiczay-Rüdiger et al. (2016a).

## **The Gerecse Hills**

Between the Győr-Tata Terraces and the Danube Bend the Danube river has incised into Late Miocene siliciclastic rocks deposited on a basement composed mainly of Mesozoic carbonate rocks and Paleogene to Neogene siliciclastic cover. The carbonate rocks are exposed to the south of the Danube valley, in the Gerecse Hills. River terraces on northern slopes of the Gerecse Hills, whose development was linked to the incision of the Danube were the main targets of this study, as these may provide useful information on the vertical deformation and climate oscillations of the area.

The first results of this work were presented on conferences (Ruszkiczay-Rüdiger et al., 2016c), and have been integrated in the Explanatory book to the geological map of the Gerecse Hills (Csillag et al., submitted; Budai et al., submitted). Preparation of a peer reviewed paper has started in spring 2017, when results of the AMS measurements of the last sample set have arrived (Ruszkiczay-Rüdiger et al., in prep). As these results haven't been published yet, a more detailed description of this research area and results will be provided in the following.

### ***The terrace material and methodology***

Terrace deposits of the Gerecse Hills consist of cross-bedded sandy gravels, gravelly sands, in a very variable thickness from a few centimetres up to ~20 m. The terraces are frequently covered by travertine deposited by karstwater springs upwelling from the underlying carbonate rocks (Scheuer and Schweitzer, 1988; Kele, 2009; Török et al., 2017) and by loess (Novothny et al., 2011). Terraces of considerable thickness have been preserved where the fluvial sediments were protected by the overlying travertine.

In order to establish a new and robust geochronological timeframe of the Quaternary landscape evolution of the Gerecse Hills governed by the incising Danube river, novel CRN data had to be inserted in an up-to-date geological, geomorphological and geochronological timeframe. Results of this work are being published by Csillag et al. (submitted) and Budai et al. (submitted). During

the present study novel CRN data have been compared to published U-series data of travertines (Kele, 2009; Sierralta et al., 2010) and with luminescence ages from the lower terraces (post-IR IRSL on K-feldspar; work of Ágnes Novothny and Edit Thamó-Bozsó). The revision of the paleontological findings was assisted by Attila Virág and Bálint Szappanos. Fieldwork and re-interpretation of geological, geomorphological and field data was carried out together with Gábor Csillag, László Fodor, Zoltán Lantos. A total number of 29 samples were taken from 5 locations for CRN analysis from the Gerecse Hills and were processed for  $^{10}\text{Be}$  only (n=15) and for paired  $^{26}\text{Al}$ - $^{10}\text{Be}$  analysis (n=14).

This comprehensive overview of former and new data allowed us to settle a new geochronological timeframe of the terrace formation and of the incision of the Danube in the study area. Assisted by the new chronological data, the traditional terrace system of Pécsi (1959) has been challenged. On the new geological map of the Gerecse Hills one Pliocene (fQ9) and ten Pleistocene (fQ8-fQ2a) terrace levels have been distinguished (Csillag et al., submitted). These are labelled by Arabic numbers with a prefix “fQ” (fQ2a – fQ9) to clearly differentiate the new terminology from the former system, when roman numbers were used (Table 1). This nomenclature was already used for the terrace chronology of the Gerecse Hills presented by Ruzskiczay-Rüdiger et al. (2016c).

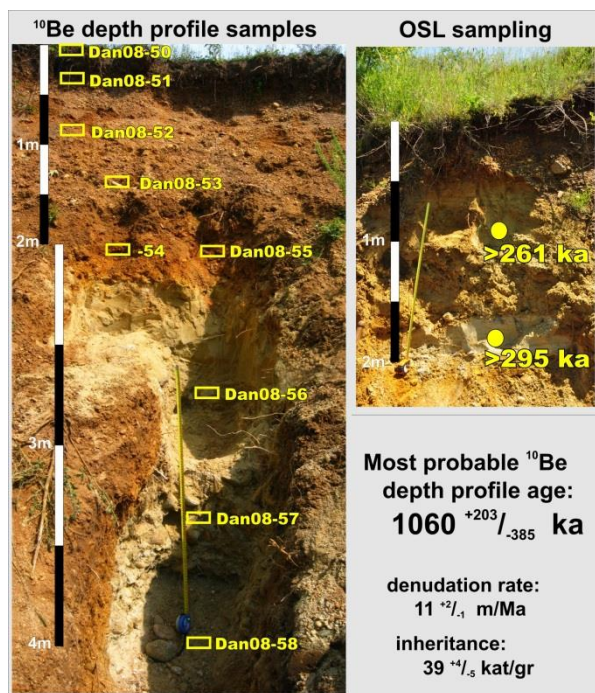
RT	TT	RA (m, asl)	TA (m, asl)	Age range	Method	Reference
fQ9	VII	273-285	270-310	2.6-3.2 Ma	CRN	3
fQ8	VI	265-278	240-270	n.a.	-	-
fQ7	V	215-223	190-220	2.1-3.2 Ma	CRN	2,3
fQ6	V	190-205	190-210	>700 ka; >410ka	CRN OSL	1 1
fQ5	IV	170-185	160-180	680-1260 ka	CRN	2,3
fQ4	IV	~160 m	160-180	n.a.	-	-
fQ3b	III	~150 m	140-150	210-570 ka; >263 ka	CRN OSL	2,3 2,3
fQ3a	III	132-138	140-150	n.a.	-	-
fQ2c	IIb	125-132	117-127	130-156 ka	OSL	1
fQ2b	IIb	118-120	117-127	86-128 ka	OSL	1,2,3
fQ2a	IIa	113-116	111-113	n.a.	-	-

**Table 1.** The “traditional” terrace labels and elevations compared to the revised terrace system.

**RT:** Revised terminology of Danube terraces (Csillag et al., submitted; Ruzskiczay-Rüdiger et al., 2016b); **TT:** “Traditional” terrace terminology (Pécsi, 1959); **RA:** Revised terrace altitude in the Gerecse Hills (Csillag et al., submitted); **TA:** Terrace altitude in the Gerecse Hills after Pécsi (1959); **Age range:** min-max age range of a terrace level based multiple samples and locations (if relevant) including dating uncertainties. References: 1: Ruzskiczay-Rüdiger et al., 2016a; 2: Ruzskiczay-Rüdiger et al., 2016c; 3: Ruzskiczay-Rüdiger et al., in prep.

### *CRN sample locations and results*

Initially, as it was proposed by the research plan, terrace locations suitable for  $^{10}\text{Be}$  depth profile dating were searched for and sampled in the Gerecse Hills. Two locations (Bethlehem quarry, Kender Hill) appeared to be suitable for this technique (n=15). At both locations, similarly to the Győr-Tata Terraces, exposure age-denudation rate-inheritance triplets were modelled using the Monte Carlo approach of Hidy et al. (2010) (as described by Ruzskiczay-Rüdiger et al., 2016a). The first results have been presented by Ruzskiczay-Rüdiger et al. (2016c).



In the abandoned gravel pit of the *Betlehem quarry* near Szomód (Figs. 1, 3) a Mid-Pleistocene terrace of the Danube (175 m asl. tIV according to Pécsi, 1959; fQ5 in the new system) is exposed. Samples were collected from the sandy gravel down to a depth of 4 m (n=9). The most probable exposure age of this terrace is  $1.06^{+0.20/-0.39}$  Ma with a denudation rate of  $11^{+2/-1}$  m/Ma. Minimum burial age provided by the post-IR IRSL measurements (>295 ka) and the revised malacological age of ~0.78-1 Ma (Krolopp, 2014) are in agreement with the CRN exposure age of this horizon.

**Fig. 3.** The outcrop of the Betlehem quarry with CRN and OSL sample locations and results.

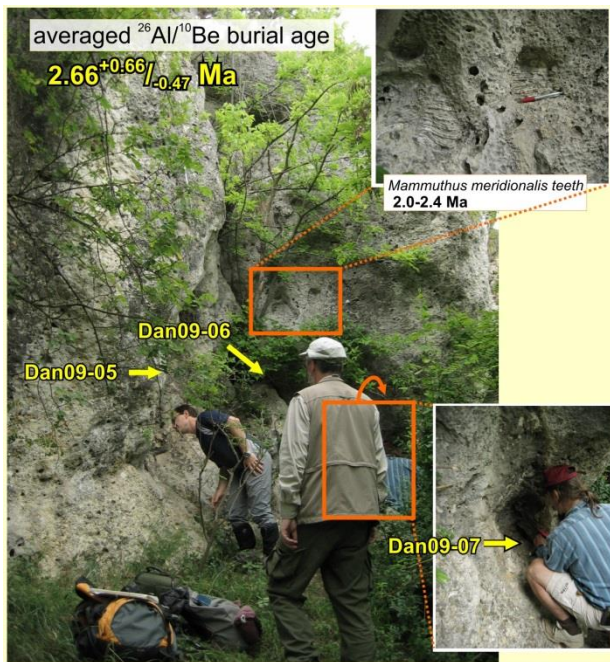
In the abandoned sandpit of the *Kender Hill* (165 m asl) close to Tata (Figs. 1, 4) a pediment surface (pQ4-3b) is exposed, which is most probably connected to the fQ3b terrace on the western side of the Gerecse Hills, in the Által-ér valley. Samples were collected along a 5 m deep depth profile (n=6; the uppermost sample could not be analysed) The most probable exposure age of this surface is  $377^{+211/-165}$  ka with a denudation rate of  $20^{+8/-3}$  m/Ma. The CRN age is in agreement with the minimum burial age assessed by the post-IR IRSL measurements (>456 ka).



**Fig. 4.** The outcrop of the sandpit at Kender Hill with CRN and OSL sample locations and results

A high terrace level (fQ7; former tV), outcropping in a travertine cemented fluvial succession in the abandoned travertine quarry on the *Les Hill* (Figs. 1, 5) was sampled for a combined  $^{26}\text{Al}/^{10}\text{Be}$  depth profile and burial dating (n=5). At the upper part of the profile the overbank fine sands, while at 4-6 m depth sandy gravel with cm size pebbles are exposed and have been sampled. Unfortunately, fine sand content of the samples taken from top surface was consumed during quartz separation and leaching and only the remaining 3 samples from the 4.7-6.0 m depth range could be used for the measurement of their  $^{26}\text{Al}$  and  $^{10}\text{Be}$  concentrations. Assuming stable conditions after burial and considering postburial production (which, given the long exposure time, is significant at this subsurface depth) a maximum burial age of  $3.41 \pm 0.42$  Ma was calculated for the terrace as an average for the three samples. This maximum age is conformable with a surface denudation rate of 27.7 m/Ma that would suggest a total denudation of 94.4 m material; a very unlikely scenario.

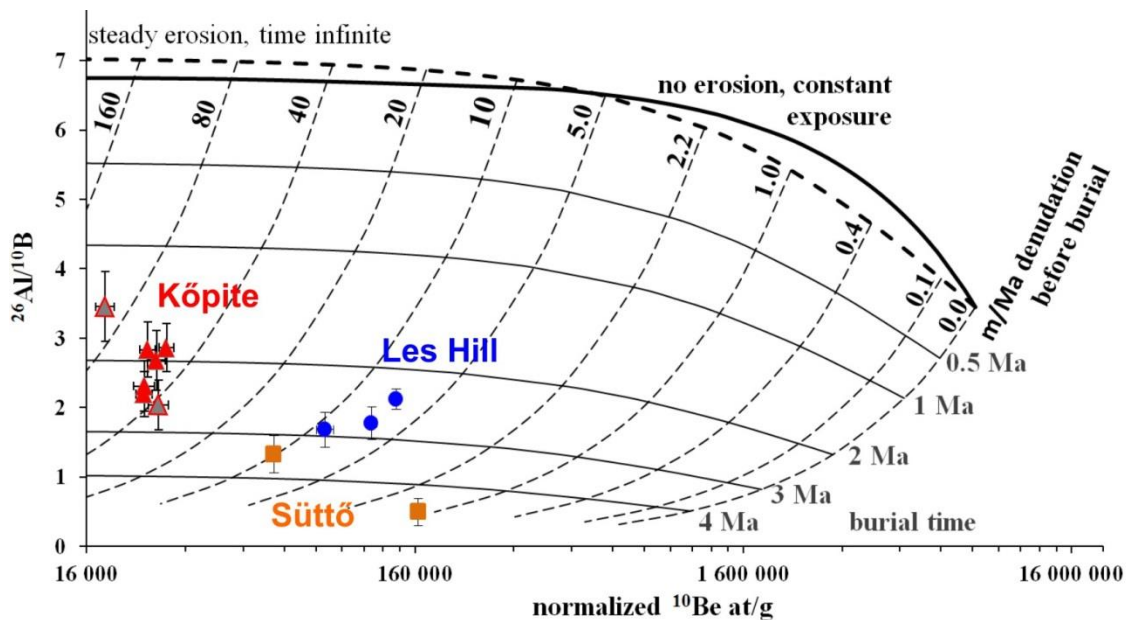
As another limit, the minimum burial time has been assessed proposing complete burial and no postburial production resulting in a *minimum burial* age of  $2.37 \pm 0.17$  Ma for the youngest sample and  $2.66^{+0.66}/_{-0.47}$  Ma for the mean of the three samples (Figs. 5, 6).



The minimum CRN burial ages agree within error with the ~2.0-2.4 Ma most probable paleontological age of the site determined by the morphometric analysis of 3 *Mammuthus meridionalis* teeth (Virág, 2013) found within the sandy gravel body (Fig. 5). This suggests that the sampled rock have been shielded from cosmic irradiation for most part of their burial history and a recent truncation event have removed >10 m of rock from above the sampled outcrop, which is a possible scenario knowing that the area has been used for quarrying freshwater limestone from the age of the Roman empire.

**Fig. 5.** The outcrop of the Les Hill with sample locations and *Mammuthus meridionalis* teeth

On the other hand, Th/U ages between  $273 \pm 73$  ka and  $408 \pm 73$  ka of the freshwater limestone of the Les Hill (n=3) (Kele, 2009) suggest considerably younger age of the travertine than the that of the siliciclastic terrace material and enclosed paleontological findings.

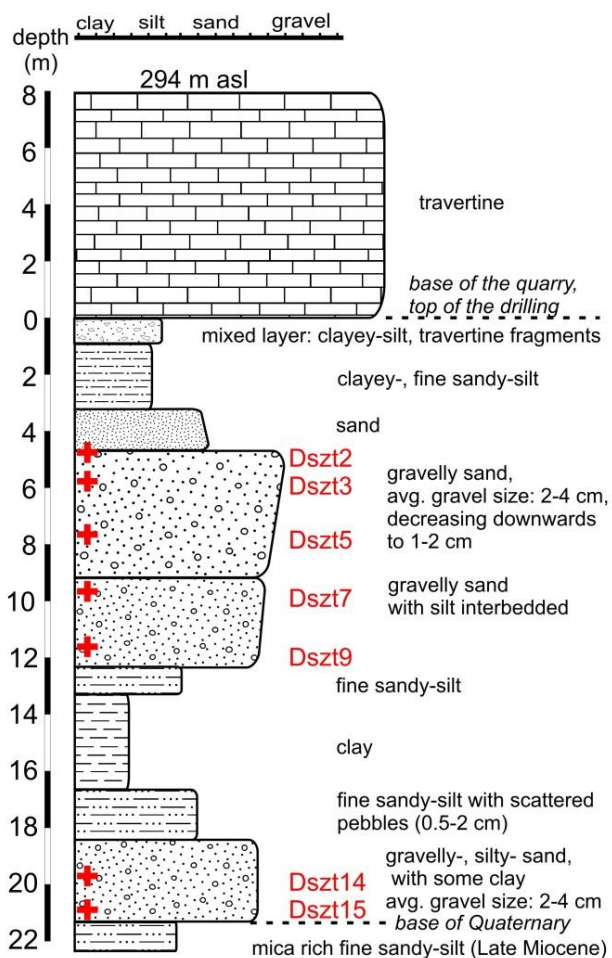


**Fig. 6.** Exposure-burial diagram of the samples taken for burial age determination in the Gerecse Hills. Samples plotted in grey were excluded as outliers from the Köpíte sample set. Error bars represent 1 $\sigma$  analytical uncertainty, uncertainty of the  $^{10}\text{Be}$  and  $^{26}\text{Al}$  half-lives, 1% uncertainty of AMS machine deviation, and 6% uncertainty production rate and scaling and the uncertainty of the sample mass depth.

During the second part of the project two additional locations were sampled for  $^{26}\text{Al}/^{10}\text{Be}$  burial age dating (Süttö, Köpíte).

Two samples were collected from the gravelly sediment underlying the travertine complex of *Süttő*, (215 and 218 m asl). This sediment was described as a Danube terrace (tV, Pécsi, 1984) underlying the thick 20-50 m travertine complex of *Süttő*. Samples were collected from the gravelly clay and from a travertine-cemented conglomerate underlying the thick travertine body at the bottom of a paleo-valley. Low  $^{26}\text{Al}/^{10}\text{Be}$  isotopic ratios suggest  $3.34\pm 0.68$  Ma and  $5.39\pm 2.1$  Ma simple burial age of the sampled material (Fig.6), which is clearly unconformable with the 422-210 ka Th/U age of the overlying travertines (Kele, 2009, Sierralta et al., 2010). A revision of the drillings and outcrops in and around the *Süttő* travertine body disproved the original position of these pebbles as a Danube terrace, instead proluvial origin of this sediment has been postulated with the gravel re-deposited from an older terrace or from the Eocene siliciclastic lithologies (Csillag et al., submitted). In this case, the sampled pebbles must have arrived with a lowered initial  $^{26}\text{Al}/^{10}\text{Be}$  ratio, allowing for the low CRN ratios measured in the samples. Thence, unfortunately this location is not suitable for the age determination of Danube terraces.

The last sample set of the Gerecse Hills (n=7) was collected along a drill-hole in the autumn of 2015 (AMS results arrived in spring 2017) from the *Kőpíte Hill* (286 m tszf.) (Figs. 1, 7). This location is one of the representatives of the highest terrace of the Danube in the area (fQ9, former tVII), with a supposed Early-Pleistocene to Late-Pliocene age (Pécsi, 1959). Th/U age of this travertine is beyond the 600 ka limit of the method. The drilling (supported by the OTKA NK 83400 project) occurred in an abandoned travertine quarry using a spiral head. The estimated amount of the recently removed travertine cover by quarrying is ~8m (based on the map of the Third Military Survey of Hungary; 1869-1887). Samples were taken in a 1 m interval from 5 to 21 m depth and seven samples were selected from these for  $^{26}\text{Al}/^{10}\text{Be}$  burial age determination.



The coherence of the data set was tested using the reduced  $\chi^2$  test (Ward and Wilson, 1978) and burial age and denudation rate estimates appear to be consistent for the sample set except for 2 samples (Dszt-02, -14) (Fig. 6). These samples have been omitted from further modelling and discussion.

The dataset enabled to model the terrace age in several different ways. These are presented here from the simplest to the more complex models:

- (1) simple minimum-maximum burial age determination,
- (2) considering similar pre- and post-depositional histories of the samples
- (3) as (2) and accounting for a hiatus between the terrace abandonment and travertine precipitation.

**Fig. 7.** The sediment sequence reconstructed on the basis of the drillhole on the *Kőpíte Hill* with CRN sample locations.

- (1a) The model without postburial production assuming infinite burial depth yields a **minimum burial age** estimate for each sample between  $1.87\pm 0.23$  Ma and  $2.32\pm 0.37$  Ma with a mean of  **$2.10\pm 0.31$  Ma** (and  $53\pm 8$  m/Ma mean pre-burial denudation rate). These solutions appear on the exposure-burial diagram (Fig. 6).
- (1b) **Maximum burial ages** varied between  $2.35\pm 0.37$  Ma and  $3.85\pm 0.63$  Ma, with a mean of  **$3.08\pm 0.46$  Ma** (with  $68\pm 10$  m/Ma mean pre-burial and  $12\pm 2$  m/Ma mean post-burial denudation rates). The model with postburial production assumes that the surface after terrace formation have undergone steady denudation and samples have accumulated CRNs during burial, allowing for the determination of more realistic burial ages (Lebatard et al., 2014).
- The calculations accounting for postburial production are considered to be representative of the true terrace age. The calculated mean postburial denudation rate enables the estimation of the total thickness of travertine eroded (and dissolved) to be  $37\pm 5$  m during the last 3.08 Ma, a value around the upper limit of the travertine thickness values based on the maximum thickness of other, better preserved travertine bodies of the area (Török et al., 2017).
- (2a) In a next model samples from the upper sand body are forced to share similar burial age and denudation rate values, as they are supposed to share the **similar pre and postburial histories**. According to the sedimentary record (Fig. 7) the deepest sample might belong to a previous sedimentation phase, thus its pre-burial denudation rate has been allowed to be independent. This model provides a burial age of  **$2.61\pm 0.39$  Ma**, coupled with a postburial denudation rate of  $16.4\pm 2.4$  m/Ma. In this scenario the total thickness of travertine eroded/dissolved from above the terrace is  $43\pm 6$  m, suggesting an initial travertine thickness of travertine around 45-57 m, a thickness slightly above the largest described in the area.
- (2b) As a subsequent step, denudation rate was constrained at 10 m/Ma, the maximum value calculated for from  $^{10}\text{Be}$  depth profile data at the Győr-Tata Terraces (Ruszkiczay-Rüdiger et al., 2016a). Applying this limitation the calculated burial duration is  **$2.93\pm 0.43$  Ma** suggesting  $29\pm 4$  m travertine eroded and an thus an original thickness of 25-41 m, a value in accordance with the travertine thickness data of the area.
- (3) The previously described models do not consider the possibility of a **hiatus between terrace abandonment and travertine formation**. Therefore, as a next step a phase of hiatus (terrace exposure before burial by the travertine) was introduced in the model. Interestingly, this approach provided zero exposure time for the terrace before travertine deposition, and thus the estimated ages remained similar to those of model (2).

As a summary, the most probable  $^{26}\text{Al}/^{10}\text{Be}$  burial age of the terrace at Kőpíte Hill is supposed to be between  $2.61\pm 0.39$  Ma and  $2.93\pm 0.43$  Ma, which suggests a Late Pliocene age of the fQ9 level of the Gerecse Hills. This age is in accordance with the Early Pleistocene age of the fQ7 level supported by the burial age data of the Les Hill and also by paleontological records.

Modelling of the burial age of the Kőpíte Hill terrace is still in progress: a model accounting for a period of travertine aggradation is under development. According to the preliminary results, no major change of the terrace age is expected.

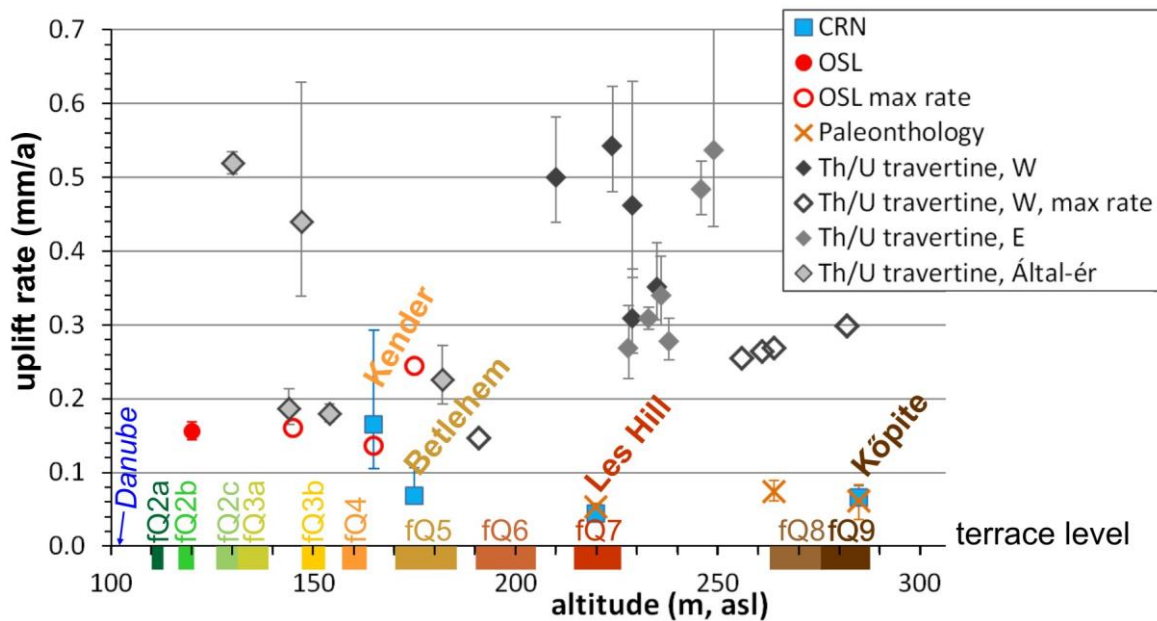
### ***Discussion and conclusions:***

#### ***uplift rate of the Gerecse Hills in the light of the new chronological data***

The presented ages are the first numerical constraints on the incision history of the Danube before Late Pleistocene times delivering new data on the related neotectonic vertical deformation and climate change.

In most cases the compiled dataset of geomorphological position, revised paleontological ages, new luminescence and CRN data are in good agreement. The dataset suggests incision rates (from terrace formation to present) to remain below 0.1 mm/a for older terraces (Early Pleistocene or older). No uplift rate above 0.2 mm/a is suggested by the data, except for those derived from Th/U

ages of travertines (not including the maximum rate calculated from the OSL min age of the Betlehem quarry; Fig. 8).



**Fig. 8.** Uplift rates derived from numerical age data plotted against sample altitude, with the name of the CRN locations. Terrace levels with their altitude range (coloured strips) as in Table 1. For maximum uplift rates no error bars appear on the plot. **W**: data from the Western Gerecse Hills; **E**: data from the Eastern Gerecse Hills; **Által-ér**: data from the Által-ér valley.

On the other hand, incision rates derived from the Th/U ages of the freshwater limestone occurrences perform a considerable scatter and vary between 0.15 mm/a and 0.54 mm/a. Most values are above 0.3 mm/a (Ruszkiczay-Rüdiger et al., 2016c; in prep). Th/U ages apparently show little or no age-elevation relationship, as it would be expected in a scenario when their precipitation occurred at or close to the former base level (Pécsi et al., 1984; Schulte et al., 2008; Zentmyer et al., 2008. Wang et al., 2017).

Unreasonably high uplift rates derived from travertine ages suggest that freshwater limestone precipitation was possible considerably above the base level. This is a possible scenario because locations of the recent karst-water springs in the region are not governed only by the base level fluctuations, but follow the paths of the karstwater flow systems and elevation of the springs represents the hierarchs of the gravity driven regional groundwater flow (Mádl-Szőnyi and Tóth, 2015). Accordingly, the carbonate-rich waters springing up well above base level were flowing down the hillslopes and along pre-existing valleys from their discharge point and travertine precipitation took place in a gently sloping environment in the form of spring-mounds and tetrata systems with ponds of lacustrine environment (Török et al, 2017). These evidences suggest very limited suitability of travertine Th/U age data for the quantification of river incision in the Gerecse Hills.

Uplift rates between ~0.05 mm/a and ~0.16 mm/a are in accordance with other independent geochronological data in the study area (Fig. 8) and are similar to data published for the European Alps (Brocard et al., 2003; Häuselmann et al., 2007; Wagner et al., 2010).

For the terraces younger than ~350 ka it was possible to correlate the periods fast global warming (MIS terminations) with river incision and terrace carving (up to the fQ3b level). For the higher and older horizons, the limited conservation of terrace remnants and the low time resolution of the chronological data do not enable to associate the terraces to certain climate phases.

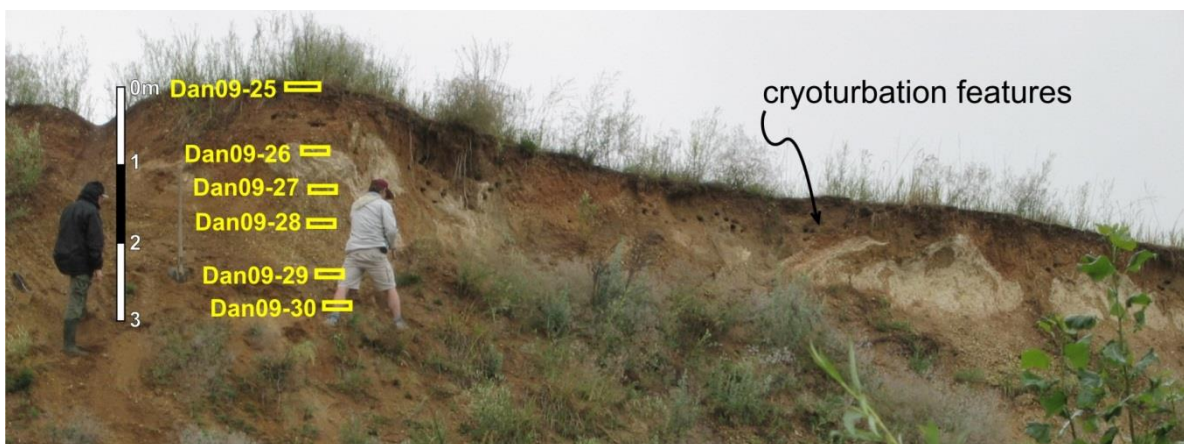
## The Pest Plain

On the Pest Plain previous authors distinguished 5-7 terrace levels, which were considered to be cut into the Early Pleistocene alluvial fan of the Danube (Pécsi, 1959; Kretzoi and Pécsi, 1982). Higher terraces, similarly to the Győr-Tata Terraces are tilted towards the subsiding lowland of the Great Hungarian Plain, and gradually smooth into its alluvial fill-up sequence. Lower terraces are occupied by the town of Budapest thence, unfortunately offered no sample locations (Fig. 1).

Only two abandoned gravel pits were found to be acceptable candidates for sampling for CRN depth profile dating, which were still affected by severe cryoturbation (Fig. 9). Despite the sub-optimal conditions for CRN dating, in the lack of more suitable locations, an attempt was made for the exposure age determination of these terraces. These sample locations were at Kistarcsa (246 m asl., tV, the highest terrace level in the southern-eastern part of the Pest Plain; n=6) and at Rákoshill (153 m asl, tIV or V, in the southern-eastern part of the area close to the Great Hungarian Plain; n=5) (Fig. 1).

The tV terrace of the Pest Plain might share the same age as the highest terrace of the Győr-Tata Terraces (>700 ka, Ruszkiczay-Rüdiger et al, 2016a) and the fQ6 horizon in the Gerecse Hills (1.0-1.9 Ma; Csillag et al., submitted; Ruszkiczay-Rüdiger et al., in prep).

Results of the  $^{10}\text{Be}$  depth profile modelling by Monte Carlo simulations using the Matlab depth profile simulator (Hidy et al., 2010; as described by Ruszkiczay-Rüdiger et al., 2016a) at Kistarcsa provided a most probable exposure age of  $1.57^{+0.35}/_{-0.84}$  Ma coupled with a denudation rate of  $5.3^{+0.3}/_{-0.4}$  m/Ma.



**Fig. 9.** The outcrop of the Kistarcsa gravel pit with sample locations and cryoturbation features.

The profile at Rákoshill was difficult to interpret because it was not possible to take samples from the upper 2.5 m of the outcrop (due to mixing by cryoturbation), which depth range is critical for the exposure age calculations. Without further geological constraints depth profile modelling could not provide a unique age – denudation rate solution for this dataset. The most probable denudation rate at this site is 7-8 m/Ma with a most likely exposure age older than >770 ka, which is in agreement with the previous age estimates.

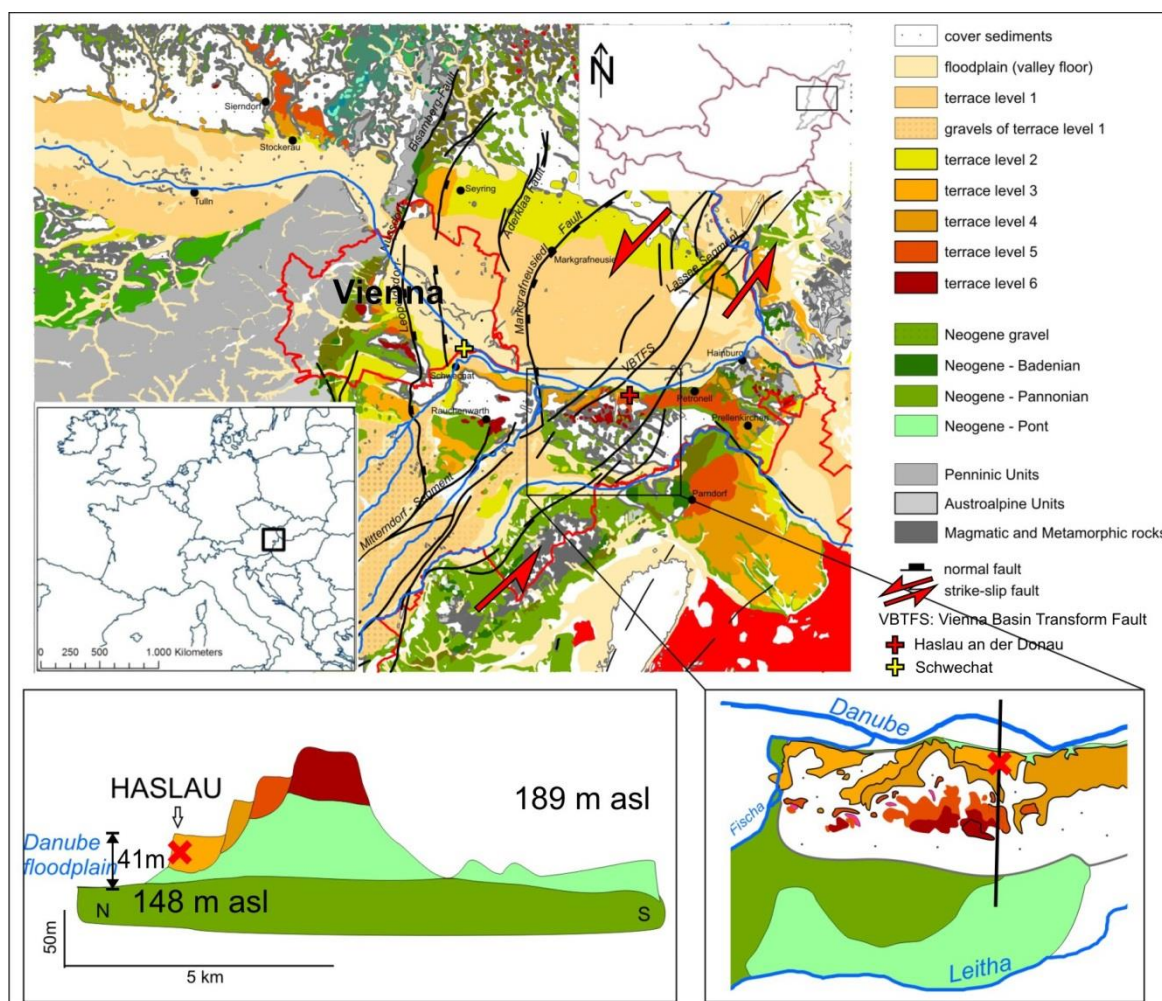
In the lack of additional independent chronological data and more locations suitable for CRN age determinations, the study on the Pest Plain has been suspended and the results of these CRN measurements were not be published so far.



## The Vienna Basin

### *Terrace age determination and inter-laboratory comparison*

Stimulated by the cessation of further studies in the Pest Plain and in search for further research areas at other segments of the Danube valley I established new connections with colleagues from Vienna (Kurt Decker, University of Vienna; Stephanie Neuhuber, BOKU). This resulted in the start-up of a fruitful cooperation and in the extension of my research field to the Danube terraces in the southern part of the Vienna Basin.



**Fig. 10.** The geological map of the Vienna basin (Fuchs and Grill, 1984), with the position of the studied outcrop at Haslau an der Donau.

The inversion-related neotectonic deformation of the Vienna Basin started during Late Miocene times, expressed as NNE-SSW trending transform faulting and development of pull apart basins (Decker et al., 2005; Salcher et al., 2012). Vertical deformation related to the strike-slip faulting led to the development of a terrace sequence of 6 Quaternary levels along the Danube river (Fuchs and Grill, 1984). The sandy-gravelly sediments of the Danube have been uplifted and fragmented leading to the formation of several disconnected terrace levels on different faulted blocks along the river (Fig.10).

The primary research objective of the study in the Vienna Basin has been defined as a better understanding of the Quaternary incision and related uplift history along the Danube river. On the other hand, the first meeting with the Austrian colleagues revealed that a new Cosmogenic sample preparation laboratory was under construction in the BOKU as well, which was run by Stephanie

Neuhuber. This fortunate coincidence made our second objective to be conceived, which was the inter-laboratory comparison of the output of the two recently launched cosmogenic nuclide sample preparation laboratories of Vienna and Budapest, that use different geochemical techniques to separate in-situ produced cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$  from rock samples.

The first sampling in the Vienna Basin and the processing of the first 5 samples were supported by a small project of the OMAA (Austro-Hungarian Action Foundation, 90öu17; April 2015 – March 2016), which work has been continued in the framework of the present project.

### ***Fieldwork and laboratory procedures***

After visiting several gravel pits in the area, a sample set has been collected from a gravel quarry at Haslau an der Donau exposing a middle Pleistocene terrace of the Danube (Figs 10, 11). No numerical age constraints have been available so far for the Danube terraces in the southern part of the Vienna Basin, the estimated age was based on geomorphological and sedimentological observations (Fuchs and Grill, 1984). A set of 14 samples was collected from two depth levels (5.5 m and 11.5 m): six cobbles and a sand sample from each level (Fig. 11) for isochron burial dating of the sediments using in situ produced  $^{26}\text{Al}/^{10}\text{Be}$  ratios. At shallow depth simple burial age dating may considerably underestimate the true terrace age. Isochron burial age determination is insensitive for the postburial CRN production, which may be a significant part of the measured CRN inventory at 5.5 m, the subsurface depth of the higher sampled level (Dan14-10 to-16), and may be present even at 11.5 m depth (see methodology section).



**Fig. 11.** The outcrop at Haslau an der Donau with sample locations and cryoturbation features.

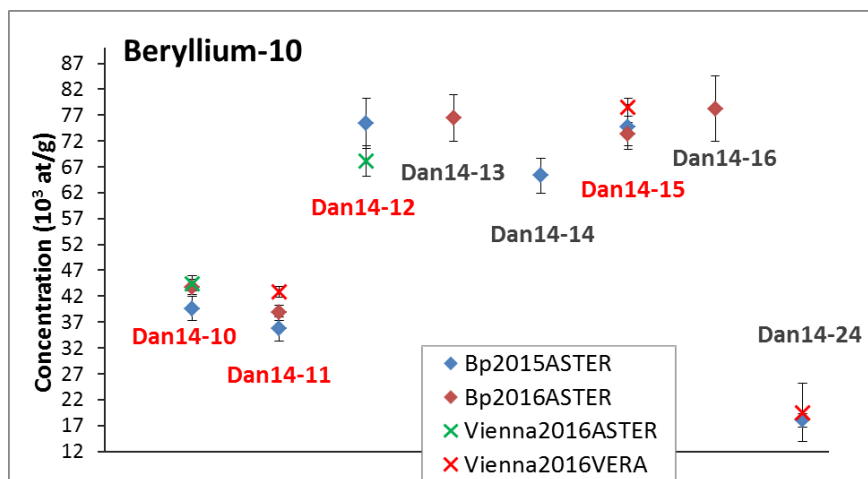
The cobbles were crushed and sieved and the 250-1000  $\mu\text{m}$  grain size fraction was split into two sub-samples. These sub-samples were processed independently in Budapest and in Vienna, using different protocols (Budapest: Merchel and Herpers, 1999; Vienna: Kohl and Nishiizumi, 1992). Sample processing consisted of chemical etching of the samples to obtain pure quartz. The purified samples were dissolved in HF in the presence of a  $^9\text{Be}$  carrier (The two laboratories used different carriers.) The  $^{27}\text{Al}$  content of the samples has been determined independently using ICP-MS (Geological Survey of Austria, Vienna for the Cosmo-lab in Vienna) and MP-AES (Institute of Nuclear Research, Hungarian Academy of Sciences, Debrecen for Cosmo-lab in Budapest) and  $^{27}\text{Al}$  carrier was added if necessary. After substitution of HF by nitric-, then hydrochloric acids, ion exchange columns were used to extract Be and Al. Targets of purified  $\text{BeO}$  and  $\text{Al}_2\text{O}_3$  were prepared for AMS (Accelerator Mass Spectrometry) measurement.

All AMS measurements of  $^{10}\text{Be}/^9\text{Be}$  ratios of the samples from Budapest occurred at ASTER (n=8), and the samples from Vienna were divided between ASTER (n=2) and VERA (Vienna Environmental Research Accelerator, Faculty of Physics, University of Vienna; n=3). All  $^{27}\text{Al}/^{26}\text{Al}$  ratios were measured at ASTER.

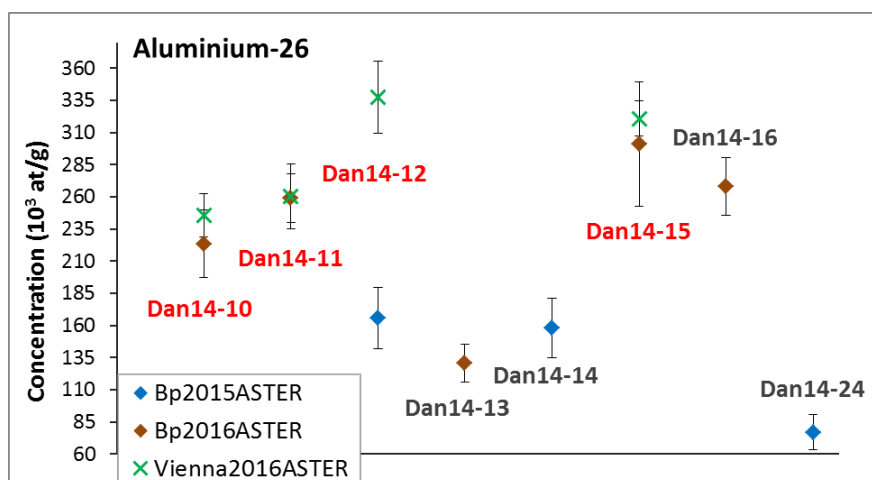
**Results**

In 2015 and 2016, processing started with the upper sampled level at 5.5 m subsurface depth. Eight samples in two sample sets (with 3 duplicate samples) were processed in the Cosmo-lab of Budapest for the determination of cosmogenic  $^{26}\text{Al}$  and  $^{10}\text{Be}$  concentrations, including all samples from the higher (Dan14-10 to-16) and 1 sample from the lower level (Dan14-24) (Fig. 12). In a next sample set, all samples from the lower (11.5 m deep) horizon (Dan14-20 to 26) have been processed and 3 out of the 6 samples are under processing in Vienna, as well. Results on both sample sets of the higher samples level have arrived and first results on the inter-laboratory comparison are available (Neuhuber et al., 2016; Ruszkiczay-Rüdiger et al., 2017), which will be outlined here.

Different protocols regarding total digestion, purification, and chromatography in Vienna and Budapest resulted in overlapping  $^{10}\text{Be}$  isotope concentrations if measured at the same AMS facility (ASTER). Interestingly,  $^{10}\text{Be}$  concentrations of sub-aliquots measured at VERA are slightly higher, but as only 3 samples were measured at both AMS facilities, this might not be significant (Fig. 12).



**Fig. 12.**  $^{10}\text{Be}$  concentrations of the sub-samples processed in Budapest (Bp) and in Vienna. Error bars show  $1\sigma$  analytical uncertainty.



**Fig. 13.**  $^{26}\text{Al}$  concentrations of the sub-samples processed in Budapest (Bp) and in Vienna. Error bars show  $1\sigma$  analytical uncertainty.

Four samples were processed for the determination of their  $^{26}\text{Al}/^{27}\text{Al}$  ratios at both laboratories. For three samples out of four,  $^{26}\text{Al}$  concentrations are similar (Fig. 13). Low  $^{26}\text{Al}$  concentration of at least one sample processed in the first set of Budapest (Bp) may be due to loss of Al as fluoride

precipitates during the volatilization of the fluorides. This initial problem has successfully been avoided for subsequent sample sets by increasing the evaporation temperatures (adding HClO<sub>4</sub>).

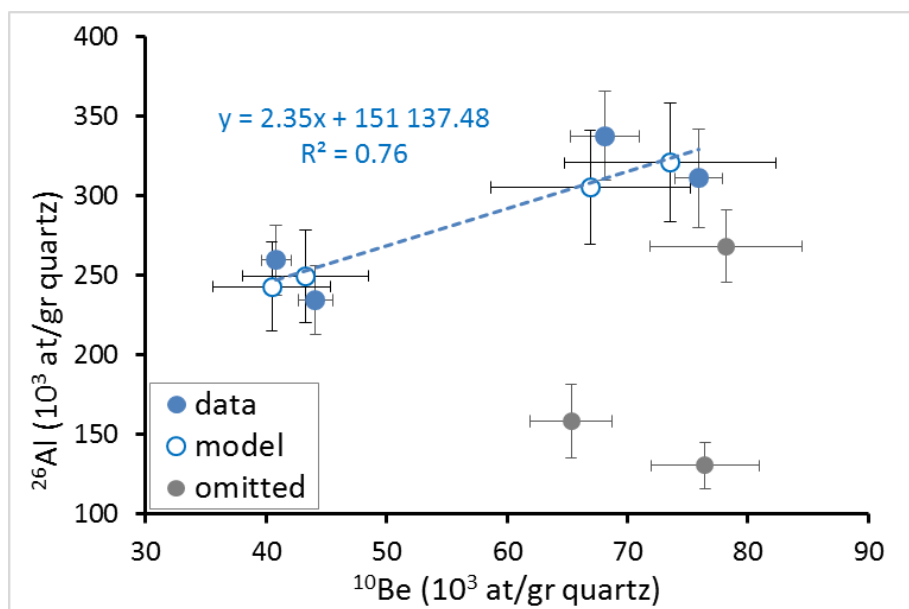
Due to the possible loss of <sup>26</sup>Al in the 1<sup>st</sup> sample set of Budapest, only samples processed in both laboratories with matching nuclide concentrations were used for the burial age calculations with their concentrations averaged (with codes printed in red). For the sample Dan14-12 the <sup>26</sup>Al concentration of the Vienna sample was used.

Most important implications of the inter-laboratory comparison and laboratory setup are as follows:

- 1) The laboratory background is safe for in-situ produced cosmogenic <sup>10</sup>Be and <sup>26</sup>Al extraction at both laboratories (Budapest and Vienna).
- 2) The different geochemical techniques applied by the two laboratories for the extraction of <sup>10</sup>Be and <sup>26</sup>Al lead to similar results.
- 3) There are two major sources of error for any applications involving in-situ produced cosmogenic <sup>26</sup>Al. First is the formation of insoluble fluoride precipitates, which must be broken up by increasing the temperatures during the final stage of HF evaporation. Second is the requirement of an accurate independent determination of stable <sup>27</sup>Al, as this value may modify the calculated nuclide concentrations considerably. These error sources were pinpointed and monitored during the laboratory setup; and the laboratory protocol has been modified when necessary to rule out these error sources.

***<sup>26</sup>Al-<sup>10</sup>Be burial isochron age determination of the Haslau terrace***

Concentrations of <sup>26</sup>Al were plotted against the <sup>10</sup>Be concentrations of the samples in order to determine the slope of the isochron, which depends on the burial age of the sampled sediment (Balco and Rovey, 2008) (Fig. 14).



**Fig. 14.** <sup>26</sup>Al -<sup>10</sup>Be plot for isochron burial age determination. Only samples measured at both laboratories were used for the isochron with averaged concentration values. Error bars show 1σ analytical uncertainty.

The significant dispersion of the data relative to the expected straight line (Fig. 14) may be the result of variable inherited <sup>26</sup>Al/<sup>10</sup>Be ratios associated with material redeposited in the studied terrace after having been remobilized from various depths from older terraces or sediments. This leads to lowered inherited <sup>26</sup>Al/<sup>10</sup>Be ratios and consequently, overestimation of the burial duration.

Accordingly, the calculated  $2.27 \pm 0.22$  Ma isochron burial age is interpreted as the **maximum age** of terrace abandonment.

### ***The incision of the Danube at Haslau***

The maximum terrace age estimated using the first results of isochron burial age determination of the Haslau terrace enabled the calculation of a **minimum incision** rate of  $\sim 18 \pm 2$  m/Ma of the Danube at Haslau. A maximum incision rate of 51-182 m/Ma could be calculated from the previously estimated Middle Pleistocene terrace age. These min-max values bracket a possible range of Quaternary uplift rate for the Haslau block.

In a drill-hole in the neighbouring block to the west (Schwechat Deep, see yellow cross at Schwechat on Fig.10), a burial age of 2.6-2.9 Ma was estimated for a gravel body 7-15 m below the Danube (Lüthgens et al., 2017). The availability of old gravel deposits at low position upstream from the studied terrace supports the possibility of erosion and re-deposition of older terrace material also suggested by the isochron data.

Besides, the Schwechat burial age tentatively enabled to estimate a subsidence rate of  $\sim 2.5$  m/Ma at the Schwechat Deep and to assess the magnitude of Quaternary vertical displacement between the two fault-bounded blocks to be  $>20$  m/Ma. Further age determinations are necessary to decide whether these preliminary ages and calculated vertical deformation rates are close to the true values.

### **Additional results related to the terrace studies**

As an incidental result of the fieldwork on the gravel terraces of the Danube river, large amount of periglacial soil mixing features were observed (Figs 9, 11), as these loose sandy gravel bodies are frequent subjects of cryoturbation. These features are hindering the work with CRN depth profiles due to the elimination of the original bedding of the sediment, as it was the case for the IIb terrace at Ács (see Ruzsiczay-Rüdiger et al., 2016a), or the terrace at Rákoshill. Nevertheless, as these striking features hindered the age determination of the terraces, I intended to use them to achieve more information on periglacial soil deformation processes and on the (paleo)climatic conditions necessary for their development.

A critical revision of the local and of the state of the art international literature on periglacial soil deformation features, together with the new field experiences resulted in a review paper on the importance of cryoturbation in the Pannonian Basin and on the significance and limitations of cryoturbation features as paleoclimate indicators (Ruzsiczay-Rüdiger and Kern, 2016d).

### **FINAL REMARKS**

Result of this project provided essential data for the reconsideration of the terrace chronology in the Hungarian Danube valley. Most important outcomes of the chronological work:

- 1) age determination of geomorphic surfaces which were hitherto datable only by indirect dating methods;
- 2) comparison of the CRN data by other independent numerical ages and also to revised paleontological records, resulting in a more robust chronology on the one hand. On the other hand, this study may serve as a reference work on the applicability fields of the different dating methods (e.g. OSL better for young terraces, CRN better for the older ones; limited suitability of Th/U travertine ages to trace the incision history of the Danube);
- 3) reference data for the revised terrace chronology considering the geochronological, geomorphological and geological results of the past few years from the Gerecse Hills and from the Győr-Tata Terraces, which will be applicable at other sections Hungarian Danube valley as well.

- 4) Despite the better age constraints of the novel terrace chronology, more data with higher resolution would be required to clearly distinguish between the effects of climate and tectonics on terrace formation.
- 5) The new sample preparation laboratory in Budapest, being the first in the East-Central European region, has opened a window of opportunities for the quantification of landscape evolution processes in the Pannonian Basin and its surroundings, which hitherto remained undated due to the lack of suitable techniques.
- 6) The chronological work started in the Vienna Basin was a first step towards the cross-border synchronisation of the Austrian and Hungarian terrace systems, a task that has not been addressed properly yet.

I'm grateful for the OTKA/NKFIH for the support, which allowed me a unique possibility to achieve the above results. I'm also indebted to the excellent colleagues from Hungary and from abroad, who shared their ideas with me and whom I cooperated with throughout this project. They appear as co-authors in the published abstracts, and papers.

Owing to the new acquaintances brought forth by this research I was invited speaker at a seminar in Salzburg (*Quaternary incision history of the Danube River via <sup>10</sup>Be exposure age determination of river terraces, Hungary, Central Europe*. University of Salzburg, 03. 11. 2015) and in Vienna (*Quaternary vertical deformation constrained by fluvial and eolian landforms dated using in situ produced cosmogenic <sup>10</sup>Be, Western Pannonian Basin, Hungary*. Universität für Bodenkultur, Vienna, 19. 10. 2016).

Budapest, 14 August 2017

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