After the discovery of the cosmic radiation in the 1910’s, Grosse (1934) was the first who suggested that the cosmic radiation could produce radioactive nuclides at the surface of the Earth. The next step towards the utilisation of cosmogenic nuclides in earth sciences was the discovery of Libby (Libby et al. 1949) in the late 1940s: during its interaction with the atmosphere, cosmic radiation produces measurable amounts of $^{14}$C.

In the 1950s Paneth et al. (1952) Davis and Schaffer (1955) suggested that cosmogenic nuclides could be produced within the minerals of the surface rocks. In 1967, Lal and Peters laid the theoretical bases of the method. During the next decade effects of cosmic radiation in meteorites and lunar rocks were studied, e.g. for the determination of the age of lunar craters (e.g. Eugster et al. 1977). However, the very low production rates impeded the application of this method in terrestrial rocks because available analytical instrumentation was unable to measure extremely low concentrations of cosmogenic nuclides produced at the Earth surface.

Until the 1980s the application of cosmogenic nuclides in earth sciences was restricted to the nuclides of atmospheric origin. The most widely used $^{14}$C is atmospheric, but $^{10}$Be, $^{26}$Al, $^{36}$Cl, $^{39}$Ar, $^{32}$Si and $^{81}$Kr isotopes are generated in the air, too. During the hydrologic cycle these can enter the sedimentary systems, which is the basis for their application in earth sciences.

Similarly to the widely accepted terminology (e.g. Wagner 1998, Gosse and Phillips 2001) the term “terrestrial in situ cosmogenic nuclide” (TCN) is used in this study for the cosmogenic nuclides produced within the minerals at, or close to the surface.

In the 1980s, newly developed very sensitive instrumentation, the accelerator mass spectrometry (AMS) and the highly sensitive conventional noble gas spectrometry gave way to the terrestrial application of in situ produced cosmogenic nuclides. Two stable, noble gas isotopes ($^3$He, $^{21}$Ne) and seven radioactive isotopes ($^{10}$Be, $^{26}$Al, $^{36}$Cl, $^{14}$C, $^{39}$Ar, $^{41}$Ca, $^{32}$Si) can be used for exposure dating as these are the ones accumulating in quantitative amount in the lithosphere (e.g. Lal 1991).

In this chapter the basic concepts of the TCN method are provided, aiming at summarising the advantages and problems of this method before turning to the application of $^3$He exposure age dating in the research area, the Danube Bend.

### 4.1. TCN in landscape evolution studies

The study of terrestrial in situ produced cosmogenic nuclides (TCN) provides the possibility to date the exposure age of certain landforms. The concentration of TCN accumulated in the surface layers of the rock is proportional to the time elapsed since it has been exposed to cosmic irradiation (e.g. Kurz 1986a, Lal 1991, Cerling and Craig 1994, Wagner 1998). With the measurement of TCN concentration accumulated in the rock surface, the exposure age can be calculated (Lal 1991). If the sampled rock surface is part of a characteristic geomorphic horizon, the exposure age of the rock equals with the age of formation of this horizon. Such surfaces can develop also with denudation and with accumulation; these genetic features are to be considered during the TCN experiments.

Measuring the TCN concentration in rock-surfaces of known age – e.g. historical, or K/Ar, Ar/Ar dated lava bodies, glacially polished surfaces – with no surface erosion gives the opportunity of the determination of the TCN production rate (e.g. Nishizumi et al. 1989,
Cerling 1990, Kurz et al. 1990, Kubik et al. 1998). The precise knowledge of the production rates is essential for the application of the method.

TCN methods are widely used to solve problems of landscape evolution in a period of the order of magnitude of $10^2$-$10^6$ years. This time range covers the whole Plio-Quaternary, which permits a more extensive usage of the TCN method with respect to the other dating methods applied for the Quaternary. The $^{14}$C, the luminescence and the Th/U methods can only be used for the last $\sim$50, $\sim$130 (occasionally up to 1 Ma), and $\sim$350 ka, respectively. Therefore, after solving the analytical problems with the new instrumentation the application of TCN became widespread. As a consequence of the large number of publications only a few examples are mentioned:

- reconstruction of the glacial events, ice volume variations via exposure age dating of ice polished rock surfaces or moraines (e.g. Briner and Swanson 1998, Schafer et al. 1999, Zreda et al. 1999),
- definition of exposure age of characteristic landforms (e.g. Albrecht et al. 1993, Repka et al. 1997, Dunai et al. 2005, Riihimaki et al. 2006),

### 4.2. Production of TCN

The Earth is exposed to a steady flux of cosmic radiation. When it reaches the Earth’s atmosphere the primary cosmic radiation consists of $\sim$87% highly energetic protons and $\sim$12% $\alpha$ particles. In the atmosphere these highly energetic particles interact with the atoms of the air, mainly with the $\text{N}_2$ and $\text{O}_2$. The secondary particles, mostly protons, neutrons, muons and photons collide with the atmospheric atoms, loose their energy and slow down (e.g. Gosse and Phillips 2001).

The secondary particles approach the Earth surface. Their composition is neutron dominated, due to the higher probability of neutrons to escape during the interaction of primary cosmic rays with the atmosphere (Niedermann 2002). The cosmic ray flux decreases exponentially in the atmosphere, but its energy spectrum in the troposphere remains roughly invariant. The shielding depth of the atmosphere at sea level is 1033 g/cm$^2$ which corresponds to $\sim$3.5 m of rock.

The TCN are produced mostly by the secondary cosmic radiation in the uppermost few meters of the lithosphere, at 3 m depth less than 1% of the surface flux remains (Lal, 1991). Consequently, similar decrease of the production of cosmogenic nuclides is observable. Among the secondary particles, neutrons and muons are energetic enough to create cosmogenic isotopes in the minerals of the surface rocks (e.g. Cerling and Craig 1994, Niedermann 2002).

**Mechanisms of TCN production**

Spallation, thermal neutron capture and muon induced reactions are the most important among the TCN forming processes (Table 4-1, Fig. 4-1).
Spallation is the dominant nuclear reaction in surface rocks and in the atmosphere. Spallation is the nuclear reaction of a fast incoming neutron with a nucleus in which a few protons and neutrons are expelled leaving behind a lighter nucleus. Neutrons are more likely to be expelled; therefore spallation favours the formation of neutron-poor daughter nuclei, i.e. the lighter isotopes of an element (Niederman 2002). The $^3$He, $^{21}$Ne, $^{10}$Be, $^{26}$Al and $^{36}$Cl nuclides formed mostly by spallation are of major importance for geologic, geomorphic TCN application (Table 4-1). Beneath the surface the concentration of fast incoming neutrons decreases exponentially. The path length in which the particle radiation decreases to 1/e (~36.8%) depends on the density of penetrated matter (e.g. Gosse and Phillips 2001).

<table>
<thead>
<tr>
<th>nuclide</th>
<th>half-life (years)</th>
<th>main target elements in the lithosphere</th>
<th>nuclear reaction of production of cosmogenic nuclides in the lithosphere</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^3$He</td>
<td>stable</td>
<td>O, Si, Al, Mg, stb.</td>
<td>spallation, (thermal neutron capture)</td>
</tr>
<tr>
<td>$^{21}$Ne</td>
<td>stable</td>
<td>Mg, Na, Si, Al</td>
<td>spallation, (negative muon capture)</td>
</tr>
<tr>
<td>$^{10}$Be</td>
<td>$1.5 \times 10^6$</td>
<td>O, Si, Al</td>
<td>spallation, (negative muon capture)</td>
</tr>
<tr>
<td>$^{26}$Al</td>
<td>$0.71 \times 10^6$</td>
<td>Si, Al</td>
<td>spallation, (negative muon capture)</td>
</tr>
<tr>
<td>$^{36}$Cl</td>
<td>$0.30 \times 10^6$</td>
<td>Cl, K, Ca</td>
<td>thermal neutron capture, spallation (negative muon capture)</td>
</tr>
<tr>
<td>$^{41}$Ca</td>
<td>$0.10 \times 10^6$</td>
<td>Ca</td>
<td>thermal neutron capture</td>
</tr>
<tr>
<td>$^{14}$C</td>
<td>5730</td>
<td>C, O</td>
<td>thermal neutron capture</td>
</tr>
</tbody>
</table>

Table 4-1. Production and half-life of terrestrial in situ produced cosmogenic nuclides of widest applicability range in geosciences (Cerling and Craig 1994, Wagner 1998, Niedermann 2002).
Chapter 4: Principles of exposure age determination...

The nucleus formed by thermal neutron capture is one atomic mass heavier than the target nucleus. The nuclei are capable of capturing the neutrons slowed down to thermal energy. This process is of minor significance for the light cosmogenic noble gases, but for the radionuclides $^{36}\text{Cl}$, $^{14}\text{C}$, $^{41}\text{Ca}$ the thermal neutron capture is of primary importance (e.g. Niedermann 2002; Table 4-1).

Negative muon capture contributes only a few percent of the total TCN production (Heisinger and Nolte 2000), therefore it is usually neglected in exposure age studies. Negative muons are particles heavier than electrons. Due to their penetrating nature relative significance of negative muons increases with depth, and at a few meters below the surface negative muon capture becomes dominant over spallation (Gosse and Phillips 2001, Niedermann 2002, Heisinger et al. 2002b). Therefore negative muon capture may be important in the case of shielded samples or in areas of fast erosion. Fig. 4-2 shows the connection between the depth dependence of the processes of TCN production with the example of the $^{10}\text{Be}$.

Fast muons induced reactions are an order of magnitude less important than negative muon capture. At a couple of meters depth fast muon reactions can surpass the negative muon reactions (Heisinger and Nolte 2000, Heisinger et al. 2002a Fig. 4-2), but they usually can be neglected in exposure age studies.

4.3. Stable and radioactive isotopes

According to their radioactivity there are two groups of TCN. The first group is formed by non-decaying stable nuclides. The accumulation of these nuclides is time-proportional; their concentration in the lithospheric rocks is constantly increasing during exposure to cosmic irradiation. For stable cosmogenic nuclides, such as $^{3}\text{He}$, and $^{21}\text{Ne}$, the exposure age $t$ is expressed as:

$$t = \frac{N*S}{P}$$

Where $N$ is the measured concentration of the cosmogenic nuclide (atoms/g), $P$ is the production rate at sea level and high latitude (atoms/g/y) and $S$ represents the scaling factors describing changes of the cosmic ray flux as function of altitude and latitude (e.g. Lal 1991, Dunai 2000 & 2001).

In contrast with the stable nuclides, the decay of the radioactive nuclides starts simultaneously with their formation. Therefore the concentration of the radionuclides shows exponential saturation in time (Figs. 4-3, 4-4), which can be expressed as:

$$N = \left(1 - e^{-\lambda t}\right) \times \frac{P}{\lambda}$$

Where $\lambda$ is the decay constant [1/y], and can be

Fig. 4-2. Depth-dependence of production rates of $^{10}\text{Be}$ in the lithosphere (Heisinger and Nolte 2000).

Fig. 4-3. TCN concentration in case of constant production rate and none erosion. The half-lives of the nuclides are shown in brackets (after Cerling and Craig 1994).
defined as:

\[
\lambda = \frac{\ln 2}{t_{1/2}}
\]

Where \(t_{1/2}\) is the half-life [y] of the nuclide \((t_{1/2} = 0.693/\lambda)\). The half-lives of the most important TCN are shown in Table 4-1. The concentration of the radioactive nuclides reaches an equilibrium level around four or five half-lives of each nuclide (e.g. Cerling and Craig 1994). In this case the number of the newly forming and decaying nuclides is equal and the increase of the concentration of the nuclide in the lithosphere comes to an end. Nuclides of shorter half-lives reach their equilibrium level faster then those with longer half lives (Fig. 4-3). This period of time is of great importance, as the method is not applicable for exposure ages exceeding the period of three to four half-lives. The ratio of the production rate and of the decay constant yields the concentration of nuclides at their equilibrium level \((N_e)\):

\[
N_e = \frac{P}{\lambda}
\]

When the rock surface is covered, the concentration of stable nuclides remains constant, which allows determination of the duration of past exposures. The concentration of the radioactive isotopes in covered rock surfaces decreases exponentially, in accordance with their half-lives:

\[
N = e^{-\lambda t} \times \frac{P}{\lambda}
\]

Accordingly, radioactive nuclides are applicable for the calculation of the time elapsed since complete shielding of the surface or sediment (e.g. Wagner 1998, Fig. 4-4).

**4.4. Scaling factors for TCN production**

Secondary cosmic-ray flux changes temporally, with latitude and altitude (e.g. Desilets and Zreda 2001, 2003, Desilets et al. 2006). Therefore the production rate of TCN is not constant in time and space. The application of TCN to solve geoscientific problems requires the TCN production rates to be known at any place on Earth and over geological time ranges. TCN production rates are extremely low, usually in the order of magnitude of 1-100 atoms/g/yr (e.g. Gosse and Phillips 2001).
Spatial changes of TCN production rate

TCN are suitable for age determination at almost any place on Earth. However, as the production rate differs from site to site, scaling factors affecting the production rate are to be taken into account. The most important scaling factors are the elevation above sea level, latitude, geometry of the studied surface, geochemistry of the studied rocks and possible shielding effects (e.g. Lal 1991, Gosse and Phillips 2001).

The geomagnetic field deflects primary cosmic radiation, composed mainly of protons. Only particles exceeding certain cutoff rigidity \( P_c \) \([GV]\) are able to reach the Earth surface in the form of secondary cosmic radiation. Cutoff rigidity is the energy to charge ratio necessary to cross the magnetic field and can be expressed as (Niedermann 2002):

\[
P_c = \frac{M \mu_0 c}{16\pi R^2} \cos^4 \lambda_m
\]

Where \( M \) is the dipole moment of the geomagnetic field \((7.9 \times 10^{22} \text{ Am}^2)\), \( \mu_0 \) is the magnetic permeability of free space \((4\pi \times 10^{-7} \text{ Vs/Am})\), \( R \) is the radius of the Earth \((6.37 \times 10^6 \text{ m})\), and \( \lambda_m \) is the geomagnetic latitude. Accordingly, the cutoff rigidity increases from the Poles to the Equator and as a consequence, the cosmic ray flux decreases towards low latitudes. At latitudes >60° the cutoff rigidity drops below the critical value of cosmic ray particles, thus at these high latitudes cosmic ray flux remains constant.

Geographic pole represents the average geomagnetic pole position for periods longer than 10-20 ka. As majority of the applications cover 10⁴-10⁶ years the calculation of TCN production rates geomagnetic latitude is usually replaceable by geographic latitude (e.g. Cerling and Craig 1994, Fig. 4-5).

When approaching the Earth surface the cosmic radiation is attenuated during the interactions with atmospheric particles. The resulting decrease of cosmic ray flux is approximately exponential (Lal 1991):

\[
N = N_0 e^{-d/\Lambda}
\]

Where \( N_0 \) and \( N \) are the numbers of cosmic ray particles at the top of the atmosphere and at the location of the sampling site respectively, \( d \) is the atmospheric depth (expressed in g/cm²) and \( \Lambda \) is the attenuation length or absorption mean free path length (g/cm²). Cosmic ray flux increases as air pressure decreases; consequently TCN production rates increase with elevation (~1%/10m at low altitude). Correction factors for the effect of the changes in air pressure are usually presented as function of altitude to simplify their use in exposure dating. The long-term average atmospheric pressure \( p \) is used to convert altitude to air pressure (Iribane and Godson 1992 cited by Dunai 2000):

\[
p = p_0 \left(1 - \frac{p_R h}{T_0} \right) \frac{g_e}{\beta_0 R}
\]
where $p_0$ is the sea level pressure (standard pressure 1013.25 mbar; actual value may deviate in some regions); $\beta_0$ is the temperature decrease with elevation (0.0065 K/m); $T_0$ is the sea level temperature in Kelvin (288.15 K=15 °C; appropriate value is required depending on the climate region); $h$ is the altitude in meters; $g_0$ is the acceleration due to gravity at sea level (9.80665 m/s$^2$) and $R_d$ is the gas constant (287.05 J/kg/K).

The attenuation length depends on the penetration depth ($z$) and the density ($\Lambda=\frac{z^*}{\rho}$). The penetration depth expresses the distance in which the radiation flux of the incoming radiation is reduced to its 1/e part upon crossing through material (e.g. air, water, rock). The shielding depth of the atmosphere at sea level is 1033 g/cm$^2$, corresponding to about 3.5 m of rock (Niedermann 2002). The lithospheric value of the attenuation length varies between 120 and 170 g/cm$^2$. This means a penetration depth of about 45-65 cm in a rock of 2.7 g/cm$^3$ (Gosse and Phillips 2001).

The cutoff rigidity of the geomagnetic field is latitude dependent, thus the values of $\Lambda$ are also changing. At high latitudes cosmic ray particles of lower energy are able to cross the atmosphere. For this reason here the attenuation length is shorter; i.e. the incoming particles can penetrate into shallower depth of the rock surface than at low latitudes. For exposure dating calculations TCN production rates at the altitude and latitude of a sample site are normalized to sea level and high latitudes. The attenuation length also depends on the target rock composition and on the nuclide that is produced (Niedermann 2002).

Lal (1991) introduced an easily treatable scaling method for the latitude and altitude dependent variation of the TCN production rate. This was challenged by Dunai (2000), who uses the geomagnetic field inclination instead of the geomagnetic latitude. The objective of both methods is the establishment of a consistent set of scaling parameters, which enables the comparison of results from different locations from various authors (Fig. 4-6). In this study the scaling method of Dunai (2000) will be applied.

![Fig. 4-6. TCN production rates according to Lal (1991) and Dunai (2000) in function of latitude and altitude. Production rates are normalized to sea level and high latitudes ($P_0$). Note that production rates increase with altitude and latitude. Insets show that different scaling methods lead to the largest differences in production rates near sea level at latitudes 20-40°.](image)

**Temporal variations of TCN production rate**

Temporal changes of TCN production depend on the solar activity, on the geomagnetic field strength and on the position of the geomagnetic poles.
The solar activity shows a cyclic variation of 11 years. The cosmic ray flux reaching the Earth is inversely related to the solar activity: the increased sunspot activity results in a decrease of the cosmic ray flux, thus the TCN production rate is also reduced. These variations affect the low-energy branch of the cosmic ray flux. Consequently, at low latitudes the solar cycle have minor effect, as the low-energy particles do not exceed anyway the cutoff rigidity necessary to enter the atmosphere (Cerling and Craig 1994). At high latitudes, during periods of sunspot maximums and minimums the deviation of TCN production rate from the average can reach 25% (Masarik and Beer 1999). Typical time span of exposure age studies comprises several hundreds of solar cycles, for this reason the effects of sunspot activity can usually be neglected.

Secular variations of magnetic field control the cutoff rigidity of the incoming particles, which affects the flux and energy spectrum of the incoming radiation. The inclination, the declination and the intensity are the parameters characterizing the geomagnetic field. The inclination and the declination define the magnetic field vectors. The magnetic field vectors averaged for the Earth determine the position of the geomagnetic poles. For periods of more than a few thousand years the geomagnetic pole can be substituted by the geographic pole, thus geographic latitude can be used for scaling TCN production rates (Niedermann 2002).

The secular variations of the intensity of the Earth magnetic field induce variations of the dipole moment. The weaker magnetic field is more permeable for cosmic radiation, which means that the intensity of the geomagnetic field and TCN production rates are also inversely correlated. The greatest change in TCN production occurs at low latitudes (0-10°), where the cutoff rigidity is higher. At higher latitudes (above 40°) the production rate remains unaffected by the changes of the magnetic field intensity for the period of 10-800 ka (Masarik et al. 2001, Dunai 2001). According to Dunai (2001) the attenuation coefficient changes with the dipole strength, or better cutoff rigidity (eq. 7); at higher elevation these changes are more noticeable.

4.5. Subsurface production of cosmogenic nuclides

Cosmic radiation is attenuated exponentially while crossing the atmosphere according to equation (8). When cosmic ray particles penetrate into the lithosphere, they are further attenuated roughly exponentially:

\[
P(z) = P_0 e^{-\rho z / \Lambda}
\]

Where \( P_0 \) is the production rate at the surface, \( P(z) \) is that at depth \( z \) (cm), \( \rho \) is the rock density (g/cm\(^3\)) and \( \Lambda \) is the attenuation length (g/cm\(^2\)). The exponential decrease described by equation (10) is only valid for the spallation-produced component. The maximum of the thermal neutron capture is at ~30-50 g/cm\(^2\) attenuation length (~10-20 cm depth), but this is important only for some nuclides, e.g. \(^{36}\)Cl (Liu et al. 1994). The muon induced TCN production also shows exponential decrease, however the attenuation length in this case is considerably larger than it is for the spallation (~1300 g/cm\(^2\); ~3 m, Brown et al. 1995).

At the air-rock interface the neutron flux does not change within ~10-20 g/cm\(^2\) of attenuation length above and below their boundary (Masarik and Reedy 1995). This means constant TCN production within the first ~5 cm of penetration into the rock. For this reason for samples of a width of a few cm there is no need for the correction for shelf shielding. The TCN concentration of samples collected from larger depth have to be corrected for shielding according to equation (10).
4.6. TCN production on partially shielded, on inclined and on eroding surfaces

The incident cosmic radiation has a roughly symmetrical distribution around a maximum at vertical. The cosmic irradiation at a large flat surface comes from the entire celestial hemisphere. When nearby object (e.g. large rock, neighbouring mountain) covers part of the horizon, and also at sloping surfaces certain part of the low angle cosmic rays is shaded out, which results in a locally lower TCN production rate. Sampling is ideal at smooth, flat and non-shielded surfaces. The effect of sloping is negligible until ~25°. Production rates at boulder surfaces may be 10-12% lower than values at large flat surfaces (Masarik and Wieler 2003). If samples are taken from major slopes, from boulders or in partially shielded position the TCN production rates have to be corrected to their flat-surface-value (Niedermann 2002; Fig. 4-7).

Erosion removes the exposed, surface layers of the rock. This process gradually brings to the surface previously shielded or partially shielded layers. The exposure of these to cosmic radiation gets started in a certain depth with a very low TCN production rate. As the rocks get closer to the surface, the TCN production rate increases step by step until the surface value.

In the case of constant erosion rate the measured concentration of TCN can be used to calculate the erosion rate (Lal 1991):

\[ \varepsilon = z \left( \frac{P_0}{N} - \lambda \right) \]

In the case of stable nuclides the decay constant \( \lambda = 0 \). The erosion rate is inversely correlated with the TCN concentration, because in the case of slow erosion there is more time for TCN accumulation. The time necessary for establishing a steady state TCN concentration depends on the erosion rate, and on the half-life of the nuclide. The steady-state TCN concentration is lower if the erosion is fast and if the half-life is short (Cerling and Craig 1994; Fig. 4-8).

**Average erosion rates of drainage basins**

The TCN method is applicable for the calculation of average erosion rate of small (1-10 km², e.g. Granger et al. 1996), medium sized (~100 km²; e.g. Schaller et al 2001) drainage basins. The basic concept of these calculations is that each sediment-particle records its exhumation rate. The average TCN concentration of sediments yields the averaged erosion rate of the drainage basin according to equation (11). This method is applicable only if the sample represents the whole area of the drainage basin by the same proportion, thus drainage basins of homogeneous lithology and of moderate topography are required. Slopes of the studied area

![Fig. 4-7. TCN production on slopes. \( P_0 \) is the production rate on horizontal, smooth surfaces (Niedermann 2002).](image)

![Fig. 4-8. Dependence of stable TCN concentration on the erosion rate. The production rate is 1 atom/g/yr (Cerling & Craig 1994).](image)
should erode steadily, thus the hilly regions of slipping slopes are as bad as high mountains where slopes evolve through frequent rockfalls and slope collapses.

### 4.7. Effects of erosion and temporal shielding

Determination of exposure ages or erosion rates is straightforward when the exhumation of the rock surface from a completely shielded position was a short event and it has not been affected by erosion thereafter, or it has been eroded with a constant rate. In these *simple exposure histories* the exposure age or the erosion rate is derived directly from equations (2), (3), (11). In all other cases exposure histories are complex (Niedermann 2002) and consequently derivation of ages and rates is more complicated or even impossible. The recognition of simple or complex exposure histories is essential to avoid misleading conclusions.

**Simple exposure ages**

We talk about simple exposure ages if no erosion, nor temporal burial has affected the surface since its formation. The erosion is negligible if less than a few cm of rock has been removed during the whole time of exposure. In this case the resulting change of the production rate remains less than 10%, which remains within the precision of the TCN method. Both erosion and partial shielding result in a decreased accumulation of TCN in the rock. The measured TCN concentrations are lower for surfaces affected by shielding or erosion than for stable surfaces of similar age. Therefore TCN concentrations yield minimum exposure ages or maximum erosion rates.

The reliability of the exposure age is increased when two cosmogenic nuclides are studied. Using TCN-pairs there has to be significant difference between the half-lives of the nuclides. Fig. 4-9 shows two examples for the application of TCN ratios for determination of simple or complex exposures. The area between the lower solid lines show the case of no erosion, the upper line represents the steady state erosion. The area between the two curves includes all possibilities for simple exposure histories. This area of the plots is called “steady-state erosion island” after Lal (1991). Data to the left of this area indicate complex exposure histories. Data to the right of the “steady state erosion island” is outside the curve of no erosion, thus these would show excess of TCN, which may indicate an experimental error (Niedermann 2002).

**Complex exposure histories**

When several periods of different TCN production rates occur after the formation of a geomorphic surface the exposure history is complex. The surface evolution with partial soil, sediment or snow cover of the past, or with periods characterised by different erosion rates.
impede the exclusive application of the TCN method. In such cases the TCN method has to be combined with other, independent data.

The recognition of complex exposure histories is important for the correct interpretation of the measured data. One way of the recognition of complex exposure histories is the method of two TCN described above (at Fig. 4-9), where the data points outside the “steady state erosion island” indicate complex exposure histories.

Another technique is the compilation of a depth-profile (Gosse and Philips 2001; Fig. 4-10). In sedimentary successions the upper 10-100 cm can be mixed e.g. by bioturbation. In these cases the upper part of the curve of TCN concentration can be completed on the basis of its lower segment. In complex exposure histories the change of TCN concentration with depth does not correspond to the to the decrease described by equation (10). Fig. 4-10 shows several theoretical depth profiles of TCN concentration with different erosion rates and exposure histories. If the depth-profile deviates from an exponential decrease of TCN concentration the exposure history is probably complex. With the application of depth-profiles TCN concentration of sedimentary rocks can be corrected for the inherited component, which makes the calculated ages older then the real age of surface formation. In such cases the curve of exponential decrease does not converge to zero, but the value of the inherited TCN concentration (Fig. 4-10). Accordingly, TCN concentration of the shielded samples yields the inherited component of the nuclide content of the sediment (Gosse and Philips 2001).

**Fig. 4-10.** Theoretical depth profiles of TCN concentration of a sedimentary succession (modified after Gosse & Phillips 2001). Simple exposure histories without (a) and with inherited amount of TCN (b: fast erosion; c: slow erosion; d: inherited amount of cosmogenic nuclides). The upper 40 cm can be mixed e.g. by bioturbation (e). In such case the upper part of the curve of TCN concentration can be completed on the basis of its lower segment (f, g, h). i – a complex exposure history.

### 4.8. Selection of the adequate TCN

Formation of each cosmogenic nuclide requires specific target elements, from which it can develop during the interaction with the cosmic ray particles. On the other hand, minerals of the lithosphere are retentive for different cosmogenic nuclides. Hence, a TCN is only applicable on a certain rock if there are target elements for its formation and minerals retentive for the newly produced cosmogenic nuclides.

One of the major problems of a TCN research is the expected age of the concerned landform. Accumulation of stable nuclides is proportional with time, this means that theoretically they can be applied for any time range. The restriction is usually the stability of the surface, because the probability to overlook an erosion or burial event forging the exposure age increases. The radioactive nuclides however, are applicable only for time ranges 3-4 times larger than their half-lives because the increase of their concentration within the rocks is stopped as they reach their equilibrium level.

In this study cosmogenic $^3$He will be applied for exposure age determination (chapter 5) but a brief overview about the most widely used TCN is also provided. Description of the nuclides...

**Helium-3**

The noble gas helium occurs naturally as $^3$He and $^4$He. Both nuclides are stable. $^4$He amounts to almost 100% of the total helium in the nature. $^3$He is produced by the $\alpha$-decay e.g. within the U-Th series. There are four sources of $^3$He: primordial, radiogenic (decay of $^3$H), nucleogenic and cosmogenic (as a daughter nuclide in neutron induced decay of $^6$Li). Both He isotopes are present on Earth from the beginning of its existence as primordial helium.

The $^3$He/$^4$He ratio in nature depends on the source of the He. The $^3$He/$^4$He ratio in the crust is around $2 \times 10^{-8}$, whereas in the mantle the ratio is considerably higher: $\sim 10^{-4}$–$10^{-5}$. The crust is enriched in uranium and thorium compared to the mantle, thus the radiogenic $^4$He decreases its $^3$He/$^4$He ratio. The atmospheric $^3$He/$^4$He ratio is $1.384 \times 10^{-6}$. Cosmogenic $^3$He/$^4$He ratios are close to 0.1, thus several order of magnitude higher than crustal, mantle and atmospheric ratios.

Several factors are important to consider applying $^3$He. The most important aspects are: (1) Presence of primordial He component in volcanic rocks can be significant. The He components of different origin are distinguished based on where they reside: the primordial isotopes are situated in fluid inclusions of the minerals in contrast with the cosmogenic $^3$He, that is retained within the crystal lattice. Therefore magmatic and cosmogenic He can be separated by mechanical crushing under vacuum allowing the magmatic component to be measured separately. (2) The noble gas $^3$He is very diffusive towards the atmosphere, thus flat surfaces with fresh (not weathered) rocks are required for sampling.

Both helium isotopes are stable, thus the accumulation of cosmogenic $^3$He is proportional to the exposure time of the studied rock surface. The dating range of $^3$He stretches widely from a few 100 y to several My. Cosmogenic $^3$He in rocks is produced mainly by spallation of nuclei, like O, Si, Mg, and Al (Table 4-1). Minerals of low ion-porosity, such as olivine, pyroxenes, hornblende or diamond are able to retain cosmogenic $^3$He quantitatively.

Consequently, cosmogenic $^3$He was selected for the age determination of the strath terraces developed on Miocene volcanites in the Danube Bend because $^3$He covers the expected time range of terrace formation (between 10 ka up to a few Ma) and is suitable for volcanic lithology of the area.

**Beryllium-10**

Beryllium in nature consist of two isotopes: the stable $^9$Be and the radioactive $^{10}$Be. This latter is produced in the atmosphere and in the lithosphere by the spallation of O, N, Si, and Al (Table 4-1). Atmospheric $^{10}$Be can be transported through precipitation into the subsurface where it can be adsorbed to the surface of the minerals. The amount of atmospheric $^{10}$Be in a rock can significantly surpass that of the in situ produced nuclides. Therefore, the removal of the atmospheric component is essential for the application of this nuclide for exposure age studies.

In the lithosphere $^{10}$Be builds up in most of the minerals. For exposure dating silicate minerals, particularly quartz and olivine are utilized. The in situ production rate of the $^{10}$Be is well known, and its concentration in quartz can be measured on the same sample as another cosmogenic isotope, the $^{26}$Al. These advantages beside the fact that quartz is almost ubiquitous in the lithosphere make the $^{10}$Be the most widely used cosmogenic nuclide. The half-life of $^{10}$Be is 1.51 Ma and hence it is applicable for exposure ages up to 4-5 Ma.

**Aluminium-26**

Natural aluminium consists mainly of the stable $^{27}$Al, and only small portion is represented by the cosmogenic $^{26}$Al. This nuclide is radioactive, its half life is 705 ka, therefore it is applicable for periods of time up to 2-3 Ma. The atmospheric component of the cosmogenic $^{26}$Al is negligible because of the rare
occurrence of the target Ar nuclei. The aluminium is the third most abundant element of the crust. The concentration of the cosmogenic in situ 26Al of the lithosphere can be measured mostly on quartz, as the aluminium content of most of the other silicon-minerals is too large. Measuring 26Al together with 10Be improves the confidence of the exposure age and the erosion rate determination.

**Chlorine-36**

The natural chlorine consists mainly of two stable nuclides: the 35Cl and the 37Cl. The cosmogenic 36Cl is produced in the atmosphere and also in the lithosphere. It is radioactive with a half-life of 301 ka. In surface rocks it is produced by thermal neutron capture of 35Cl and spallation of 39Ca and 40K nuclei, therefore it is applicable on calcite and feldspar within an age range of 10^3-10^6 a. It is easily soluble by water, thus the atmospheric component can be removed without difficulty. Nevertheless, during weathering processes the cosmogenic 36Cl can also be leached causing incorrect exposure ages.

**Neon-21**

There are three natural neon isotopes: 20Ne, 21Ne and 22Ne. These nuclides are stable and are noble gases, providing a dating range from 1 ky to several My. In the lithosphere neon of cosmogenic, nucleogenic, atmospheric and primordial origin occurs. The ratio of these nuclides is characteristic for their source, which makes it possible to separate the cosmogenic component during the analysis. The in situ 21Ne is mainly produced by spallation of Si, Mg and Al (Table 4-1). Consequently this nuclide can be applied for exposure age dating or erosion rate determination on lithologies containing quartz, olivine or pyroxene. Quartz is retentive for 21Ne which allows its joint application with 10Be and 26Al.

**4.9. Error considerations**

As with all experimental data there is an uncertainty associated with the data obtained. Also the translation of the analytical data into e.g. ages is associated with uncertainties. The measured data are an approximation of the real values. The error sources can be divided into two groups: systematic and random errors (Wagner 1998). The systematic errors or reliability are caused by e.g. instrumental deficiencies, environmental influences. They modify the results always in the same direction and degree, therefore they cannot be recognised by repeated measurements. The random errors or precision are recognisable by repeated measurements as they are caused by random inaccuracies, like statistical processes, or reading the instrument. The systematic error means the reliability of the method, the random error is the reproducibility of the measurement.

The systematic error sources in the case of the cosmogenic nuclides can be significant. Production rate of the cosmogenic nuclides depend on several factors, like latitude and altitude dependence, neglecting the muogenic component, geomagnetic field properties, solar activity variations or the scaling method. Majority of these factors can only be assessed with limited accuracy; therefore the different scaling models may have systematic differences as large as 1-20 % (Niedermann 2002). All studies using cosmogenic nuclides are affected by these systematic errors, thus they are not included in TCN data tables; however should be kept in mind. The errors quoted for the cosmogenic data (1σ) include the analytical uncertainties only. The precision (±1σ) of cosmogenic 3He data of the Danube Bend samples is ±3%.

Other important error source is the selection of the sample site. Shielding effect of temporary soil or snow cover is usually not traceable after its disappearance, but may reduce the concentration of TCN within the rocks diminishing the exposure age of the landform. During glacials in the central part of the Pannonian Basin, cold and dry periglacial conditions prevailed, with no permanent snow or ice cover. Therefore, the effect of partial snow- or ice-shielding has been neglected, as influence of this factor is within the 1-20% systematic uncertainty of the TCN method.
The effect of erosion decreases TCN concentration in a similar way as temporary burial
does. Although surfaces of the sampling sites did not indicate evidence of erosion since their
abandonment by the river, the possibility of erosion could not be excluded. Erosion removes
layers of irradiated rock from the surface exposing material, which was in shielded position
until then. Therefore, $^3$He exposure ages are defined as minimum ages (Niedermann 2002;
straths may have formed earlier than the exposure age) and the incision rates derived thereof
are maximum incision rates (as older terraces indicate slower incision).

The measured abundances of $^3$He have to be corrected for any contributions of non-
cosmogenic origin. Atmospherically derived $^3$He is usually insignificant but in the case of
magmatic and volcanic rocks the presence of trapped mantle $^3$He is possible. If there is a
significant unrecognised non-cosmogenic component, calculated ages are older (or erosion
rates are slower) than the real values (Niedermann 2002). Trapped noble gas components
originated from the mantle reside in fluid inclusions. This component can be identified and
quantified by measuring the gas fraction released by crushing the aliquot of certain sample
(section 5.4). Therefore in the sample suite, two samples, VS 12 and DB 22, where crushed for
detection of an eventual mantle component. These experiments revealed that for the sample
suite of the present study less than 1% of the noble gases reside in fluid inclusions, thus we
conclude that mantle helium is not a significant source of $^3$He in our samples.