Geological Quarterly, Vol. 40, No. 3, 1996, p. 481–486

Jan DZIERŻEK, Jerzy NITYCHORUK, Marek G. ZREDA, Grażyna ZREDA-GOSTYŃSKA

Cosmogenic isotope ^{36}Cl — a new perspective for Quaternary chronostratigraphy of Poland

The new method of absolute dating by (cosmogenic isotope ^{36}Cl) is considered. The method is based on measuring concentrations of isotope ^{36}Cl which is produced in terrestrial rocks exposed at the surface to cosmic radiation. The presence of ^{36}Cl in most of the rocks and its long half-life make it useful in determination of the age of landforms and sediments in different depositional environments within the time range from a few thousand years to about one million years. The initial samples collected in the Polish Tatra Mts. are being analyzed in the U.S.A. The authors believe that cosmogenic ^{36}Cl method becomes a new important tool in Quaternary chronostratigraphy of Poland.

Dating of deposits using thermoluminescence (TL) and radiocarbon (^{14}C) methods, and less often potassium-argon (K–Ar) and uranium-thorium (U–Th) methods, is the basic chronostratigraphic tool for Quaternary stratigraphy in Poland. Each of the isotopic dating methods has certain limitations resulting from the physical fundamentals of the method (which in turn limits the method's applicability to specific types of sediments), analytical precision and accuracy of the measurement of each isotope, and the analytical costs. Because of the applicability of TL to dating of unconsolidated sediments and its relatively low cost, the method has been used widely during the last several years, even though errors associated with poor analytical precision may often be larger than the accepted range of the expected result (M. F. Pazdur, A. Bluszcz, 1987*a*, *b*). Because of analytical problems, the popularity of the TL method for dating of glacial and pluvio-glacial sediments is decreasing, along with the increasing skepticism about the accuracy of the method in general. The radiocarbon method, despite being a more advanced technique, is limited to relatively young organic deposits and carbonate sediments (< 30–50 ky) and provides only indirect (or bracketing) ages of landforms.

In this context, each new method of absolute dating of deposits should be accepted by Quaternary scientists as a chance of dating landforms with a greater confidence. Will cosmogenic ^{36}Cl prove to be such a method?

The principles of the cosmogenic ^{36}Cl dating method were established in the 1950s (R. Davis Jr., O. A. Schaeffer, 1955), but because of the insufficient analytical sensitivity, the potential of the technique could not be realized at that time. Recently, owing to the development of ultra sensitive accelerator mass spectrometry (AMS) for measuring ^{36}Cl (D. Elmore *et al.*, 1979; D. Elmore, F. M. Phillips, 1987), the cosmogenic ^{36}Cl method has been developed to the point at which geological applications are possible. The method has been calibrated (M. G. Zreda *et al.*, 1991; M. G. Zreda, 1994; F. M. Phillips *et al.*, 1996) and used to date young volcanic rocks (M. G. Zreda *et al.*, 1993), glacial sediments and landforms (F. M. Phillips *et al.*, 1990; R. I. Dorn *et al.*, 1991; M. G. Zreda, 1994; M. G. Zreda *et al.*, 1994*b*, 1995; M. G. Zreda, F. M. Phillips, 1995), pluvio-glacial terraces (M. G. Zreda, 1994), shorelines and pluvial beach deposits (M. G. Zreda, 1994), fault scarps formed by prehistoric earthquakes (M. G. Zreda *et al.*, 1995), faults cutting alluvial deposits (P. R. Bierman *et al.*, 1995*a*), meteorite craters (F. M. Phillips *et al.*, 1991), and to study erosion rates (M. G. Zreda, 1994; M. G. Zreda *et al.*, 1994*a*; P. R. Bierman *et al.*, 1995*b*).

The cosmogenic ^{36}Cl method is based on measuring concentrations of isotope ^{36}Cl which is produced in terrestrial rocks exposed at the surface to cosmic radiation. Measured concentrations of ^{36}Cl allow determination of the time that elapsed between the exposure of the rock to cosmic radiation and the time of sample collection. Cosmogenic ^{36}Cl is formed mainly in the narrow surface layer of the rock and its production rates decrease to negligible values at the depth of a few metres. There are several nuclear reactions leading to the formation of ^{36}Cl; the following three are the most important in the top few metres of the Earth's crust (M. G. Zreda, 1994):

1. Spallation of ^{39}K and ^{40}Ca due to bombardment by secondary fast neutrons, which compose the main spectrum of the cosmic ray flux at the Earth surface. These reactions account for 16–80% of produced ^{36}Cl, depending on the concentration of K and Ca in the rock.
2. Activation of ^{35}Cl by slow (thermal) neutrons formed by thermalization of fast neutrons through interactions with different nuclei in the atmosphere and rocks. This process accounts for 11–80% of ^{36}Cl formed, depending on the total Cl content of the rock.
3. Negative muon capture by ^{40}Ca, which accounts for 0.3 to 10% of ^{36}Cl formed.

Under ideal conditions, when the surface of the rock is rapidly exposed to cosmic radiation by a single geologic event and afterwards remains undisturbed, the amount of accumulated ^{36}Cl is a function of the cosmic ray intensity, the chemical composition of the rock and the exposure time. The number of ^{36}Cl atoms in the rock can be described by the following linear differential equation:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = P - \lambda_{36}N$$

where: P — production rate of cosmogenic ^{36}Cl (in atoms/yr); N — number of atoms of ^{36}Cl (atoms); t — exposure time (yr); λ_{36} — decay constant of ^{36}Cl (0.0000023/yr).

The equation describing the exposure time t (surface exposure age), corrected for all pathways leading to the production of ^{36}Cl and for the geographic location of the sample, is written as (M. G. Zreda *et al.*, 1991; M. G. Zreda, 1994):

$$t = \frac{-1}{\lambda_{36}} \ln \left[1 - \frac{R_{36}\lambda_{36}N_{\mathrm{Cl}}}{ELD\left(\Psi_{\mathrm{K}}C_{\mathrm{K}} + \Psi_{\mathrm{Ca}}C_{\mathrm{Ca}} + \Phi_n \frac{\sigma_{35}N_{35}}{\Sigma\sigma_i N_i}\right)} \right]$$

where: R_{36} — atomic ratio of ^{36}Cl to stable Cl, corrected for radiogenic background (i.e., ^{36}Cl produced by activation of Cl by thermal neutrons derived from decay of U and Th present in the rock); λ_{36} — decay constant of ^{36}Cl (0.0000023/yr); N_{Cl} — number of atoms of stable isotopes of Cl (^{35}Cl and ^{37}Cl); Ψ_{K}, Ψ_{Ca} — production rates due to spallation of ^{39}K and ^{40}Ca (in atoms per kilogram of rock per unit concentration of K and Ca, respectively); C_{K}, C_{Ca} — concentration of K and Ca, respectively (expressed as weight % of the oxide); Φ_n — production rate of thermal neutrons (number of neutrons per kilogram of rock per year); σ_{35} — thermal neutron absorption cross section for ^{35}Cl (cm^2); N_{35} — number of atoms of ^{35}Cl; σ_i — thermal neutron absorption cross section of element i in the rock; N_i — number of atoms of element i in the rock; E, L, D — correction factors for the production rates of ^{36}Cl due to elevation above sea level (E), geographic latitude (L) and depth (D).

The production rates of cosmogenic ^{36}Cl from its main target elements (^{35}Cl, ^{39}K and ^{40}Ca) have been determined empirically using independently dated rocks from several locations and of different ages (M. G. Zreda *et al.*, 1991; M. G. Zreda, 1994). These production rates have been refined recently by F. M. Phillips *et al.* (1996).

As can be inferred from the discussion above, the large number of factors affecting the measured concentration of ^{36}Cl isotope may lead to a significant error associated with the result. Analytical errors of the AMS measurements are small, usually smaller than 5% (F. M. Phillips *et al.*, 1990, 1991; M. G. Zreda *et al.*, 1991). Errors associated with the lack of information on the temporal and spatial distribution of cosmic ray intensity may reach 10–20%, but are being systematically reduced through continuing research (M. G. Zreda, 1994). Other geological factors that affect the amount of ^{36}Cl accumulated in exposed surfaces include (M. G. Zreda, 1994; M. G. Zreda, F. M. Phillips, in press):

1. Physical weathering, erosion and spalling of the outermost layer of rock.
2. Chemical weathering leading to the change in chemical composition of the rock.
3. Change in the geometry of the rock (boulder), change of the angle between incoming cosmic rays and the rock surface, or redeposition.
4. Change of elevation above sea level, for example due to tectonic processes.
5. Temporary and/or partial shielding due to covering of the rock surface by vegetation, snow, ice, volcanic ash, etc. The methods of limiting the influence of the above factors on ^{36}Cl dating are described in detail elsewhere (M. G. Zreda *et al.*, 1993; M. G. Zreda, 1994; M. G. Zreda, F. M. Phillips, in press).

Errors in the interpretation of the obtained results may be reduced by:

— careful selection of objects with minimal influence of interfering factors;

— detailed geological, geomorphological and palaeogeomorphological analysis of the study area;

— using of appropriate corrections;

— collection of multiple samples from each dated surface;

— performing additional analyses and measurements, for example other cosmogenic isotopes such as ^{10}Be and ^{26}Al.

Measurements of ^{36}Cl in rocks, together with accompanying analyses, are expensive and at present possible only at a few AMS laboratories in the world, including Purdue University in the United States of America. However, the presence of ^{36}Cl in most rock types and its long half-life ($3.01 \cdot 10^5$ yrs) make it useful in determination of the age of landforms and sediments in different depositional environments in the time range from a few thousand years to about one million years ago. Applications of ^{36}Cl to dating Quaternary landforms in the western U.S.A., the Canadian Arctic, and in Wales demonstrate that this method has been already well established and usually gives geologically acceptable results that correspond with those obtained by other methods (M. G. Zreda, 1994; M. G. Zreda *et al.*, 1991, 1993; F. M. Phillips *et al.*, 1991). Its broad range of applicability gives ^{36}Cl dating a definitive advantage in comparison with other dating methods.

In 1995, we established a cooperation between Warsaw University (Dr. J. Dzierżek and Dr. J. Nitychoruk) and the University of Arizona (Prof. M. G. Zreda). The primary activity of our research group is dating of Late Quaternary glacial landforms and deposits in Poland using the ^{36}Cl dating method. For the initial study, we selected the Tatra Mountains because of the existence of glacial deposits and landforms from the youngest glaciation. Most of these deposits are well preserved and relatively undisturbed by anthropogenic activity. In addition, geology and geomorphology of the area have been extensively studied and detailed stratigraphic records have been produced (J. Dzierżek *et al.*, 1986; 1987; H. Hercman *et al.*, 1987; L. Lindner *et al.*, 1990; 1993; L. Lindner, 1994). This background will allow for a more complete and meaningful interpretation of the ^{36}Cl results and will form a basis for future comparative studies in the Polish Lowland. In 1995, we collected several samples from moraines and glacially polished bedrock in Dolina Pięciu Stawów Polskich (Valley of Five Polish Pools), Dolina Roztoki (Valley Roztoka), Hala Gąsienicowa and in the Morskie Oko area. This year, we will increase the number of samples from the Tatras and from selected areas in the Polish Lowland within the limits of the last glaciation.

Meanwhile, the samples collected during the last field season are being analyzed in the U.S.A. The first results, although only preliminary, are very promising because they date glacial landforms described above to the Late Vistulian, as expected. Our results will be published after more samples are dated using ^{36}Cl, and after thorough examination of all the data. If it is deemed necessary, we will use additional cosmogenic techniques (e.g., other cosmogenic isotopes, using mineral separates from the same sample for ^{36}Cl dating, and distribution of ^{36}Cl below the surface) to verify the obtained results.

Although dating using cosmogenic ^{36}Cl is time consuming and expensive, we believe that our work on cosmogenic ^{36}Cl dating of Quaternary surfaces in the Tatra Mountains and the Polish Lowland will contribute to further improvement of this technique and serve as a basis for verification of our concepts of the Quaternary chronostratigraphy of Poland.

Acknowledgements. Supported in part by the U.S. National Research Council through a COBASE Short-Term Project Development Grant to M. G. Zreda. The authors are indebted to the Head of the Tatra National Park for permission to carry out the field works. Field works were financed by Institute of Geology of Warsaw University — BW/1344/2.

Instytut Geologii Podstawowej
Uniwersytetu Warszawskiego
Warszawa, al. Żwirki i Wigury 93
Department of Hydrology and Water Resources
Department of Geosciences
University of Arizona
Tucson, AZ 85721, U.S.A.
Received: 28.06.1996

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Jan DZIERŻEK, Jerzy NITYCHORUK, Marek G. ZREDA, Grażyna ZREDA-GOSTYŃSKA

KOSMOGENICZNY IZOTOP ^{36}Cl — NOWA PERSPEKTYWA CHRONOSTRATYGRAFII CZWARTORZĘDU POLSKI

Streszczenie

Opisano metodę datowania osadów i form rzeźby, polegającą na pomiarze zawartości izotopu ^{36}Cl skoncentrowanego w przypowierzchniowej warstwie skały. Izotop ten powstaje w wyniku reakcji cząstek promieniowania kosmicznego z atomami potasu i wapnia, występującymi w skale. Zmierzona liczba atomów izotopu ^{36}Cl w próbce pozwala określić (według podanych wzorów) czas przez jaki powierzchnia skały była eksponowana. Ze względu na powszechność występowania chloru i długi okres jego rozpadu metoda pozwala datować obiekty geologiczne i antropogeniczne z zakresu od kilku tysięcy do miliona lat wstecz.

Ze względu na liczne czynniki wpływające na pomierzoną zawartość izotopu ^{36}Cl w skale (wietrzenie fizyczne i chemiczne warstwy przypowierzchniowej, redepozycja obiektu, czasowe ograniczenia od wpływów promieniowania) bardzo ważne jest staranne wytypowanie obiektu badań i dyskusja wyniku w kontekście geologicznym i geomorfologicznym.

Do pierwszych badań tego typu w Polsce wybrano obszary Tatr Wysokich i Niżu Polskiego (zasięg ostatniego zlodowacenia) — próbki są opracowywane w laboratoriach w USA. Otrzymane dotychczas wyniki, jak i dane uzyskane z badań autorów amerykańskich pozwalają mieć nadzieję, że metoda ^{36}Cl może stać się wkrótce ważnym narzędziem chronostratygrafii czwartorzędu Polski, uzupełniając i zastępując dotychczasowe metody.