

ACCELERATOR MASS SPECTROMETRY WITH FULLY STRIPPED ³⁶Cl IONS

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ABSTRACT. A description of accelerator mass spectrometry (AMS) measurements with the long-lived radioisotope ³⁶Cl is given. All measurements were made at the Munich tandem accelerator laboratory. Results are presented for ³⁶Cl measurements in ground waters, in the meteorite Bjurböle, in ice-core samples of the Vernagtferner, Austria, and in granite samples from Hiroshima, Japan, irradiated by the atomic bomb explosion in 1945.

INTRODUCTION

In recent years we have undertaken a program of developing the AMS technique with ³⁶Cl ($T_{1/2} = 3.01 \times 10^5$ y) by using completely stripped ions. The accelerator facility consisting of a MP tandem, a heavy ion post-accelerator (Nolte *et al.*, 1979) and an achromator system can be used for AMS measurements with completely stripped ions of masses up to 40 (Kubik, Korschinek & Nolte, 1984). This method results in a strong suppression of background events originating from isobares with atomic numbers smaller than the radioisotope. Applying this technique, we performed a series of AMS measurements. Results are presented for ³⁶Cl in ground waters, in the meteorite Bjurböle, in ice-core samples of the Vernagtferner, Austria, and in granite samples from Hiroshima, Japan, irradiated by the atomic bomb explosion in 1945. Details about the accelerator and experimental set-up are reported elsewhere (Kubik, Korschinek & Nolte, 1984). A brief description of this system is given below.

THE DETECTION SYSTEM

The detection system is shown in Figure 1. For use in the cesium sputter source, the samples are transformed into AgCl (typical weight: 4 to 10 mg). A small amount of ³⁶S, typically 0.15% relative to chlorine, is added and thoroughly mixed with the AgCl samples. This allows continuous monitoring of the system and the use of the tandem slit-current to stabilize the terminal voltage. The beam passes through three stripper foils: one in the tandem terminal, a second in front of the booster, and a third in the symmetry plane of the achromatic beam analyzing system after the booster. Ca 7% of the ³⁶Cl ions that enter the third stripping foil are completely stripped. The ³⁶Cl¹⁷⁺ ions are separated by magnetic deflection from the ³⁶Sⁿ⁺ beam compounds ($n \leq 16$). The suppression of ³⁶S ions is ca 10^8 . The ³⁶Cl particles are detected by a Bragg-curve spectroscopy detector. The overall transmission from the entrance of the tandem to the detector is $\approx 3 \times 10^{-3}$. A ³⁶S monitor signal (scattered ³⁶S ions) is extracted via a rate-meter from the energy signal of the ionization chamber. The improvement

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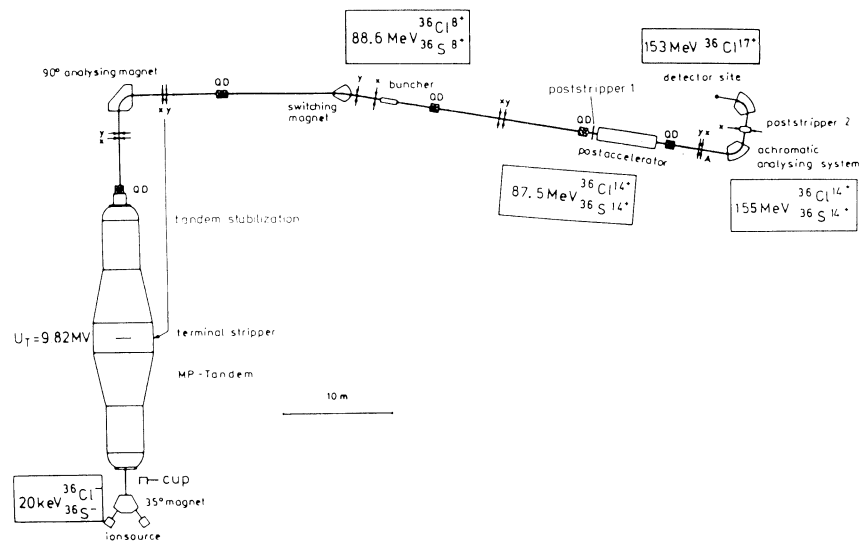


Fig 1. The beam-line system of the Munich heavy ion post-accelerator for AMS measurements

in the transmission, as compared to our previous measurements (Kubik, Korschinek & Nolte, 1984) is due to the new position of stripper 2 in front of the booster. This improves acceptance by a factor of ca 2.

SOME EXPERIMENTAL RESULTS

Measurement of ^{36}Cl in Ground Water

Measurement and interpretation of ^{36}Cl contents in ground waters were previously done by Bentley and Davis (1981). We started a program for determining ^{36}Cl concentrations in ground waters from south central Europe. Other radioactive and stable isotopes (Egger *et al*, 1983; Andrews *et al*, 1985) were already studied in these ground waters. Groundwater samples were collected from Munich (Germany), Rainbach, Braunau and Pattigham (Upper Austria, Fig 2). The measured $^{36}\text{Cl}/\text{Cl}$ ratios in some ground waters, range from 65 to 910×10^{-15} (Table 1), and can be preliminarily interpreted as follows: the sample from Munich with no detectable ^3H content and with a ^{14}C model age of ca 1.5×10^4 y (Egger *et al*, 1983) is assumed to give an initial $^{36}\text{Cl}/\text{Cl}$ ratio for ^{36}Cl dating in this area ($^{36}\text{Cl}/\text{Cl} = 690 \pm 160 \times 10^{-15}$; 2σ error). This value is in good agreement with a value measured previously by Kubik *et al* (1984) ($^{36}\text{Cl}/\text{Cl} = 630 \pm 160 \times 10^{-15}$). The Rainbach 2 sample clearly shows the influence of bomb-produced ^{36}Cl as evident from the ^3H concentration. The Braunau 1 sample has the lowest $^{36}\text{Cl}/\text{Cl}$ ratio and the highest chloride content; this could be caused by the admixture of ascending formation waters, as suggested by Goldbrunner (1984; Fig 3). The Pattigham 2 sample shows a $^{36}\text{Cl}/\text{Cl}$ ratio comparable with the sample from Munich, but due to the low chloride content, the absolute ^{36}Cl concentration is relatively low. Assuming that the sole source

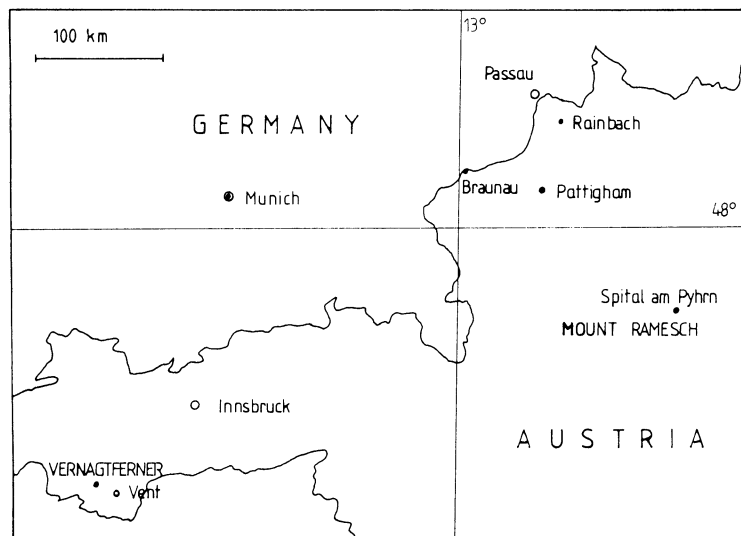


Fig 2. Map of the area of sample locations: Vernagtferner, Munich, Braunau, Pattigham, and Rainbach

of chloride to ground water, with Cl^- concentration of $<1\text{mg/L}$, is by atmospheric contribution, and that underground production of ^{36}Cl is negligible, the ^{36}Cl model age of the Pattigham 2 sample is lower than ca 10^5y .

The ^{36}Cl Concentration in the Bjurböle Meteorite

^{36}Cl was measured in meteorites by counter methods (Honda & Arnold, 1967), aided by the high ^{36}Cl concentration present in most meteorites. AMS measurements of ^{36}Cl in meteorites were already done by the Rochester group (Nishiizumi *et al*, 1983). The advantages of AMS are smaller sample size and higher sensitivity. For our first measurement, we chose the Bjurböle meteorite, where the ^{36}Cl concentration had not been measured yet. The Bjurböle meteorite fell on March 12, 1899 in Nyland (Finland). It belongs to type L4 chondrite meteorite and contains 21.8% iron, 1.2% nickel, and 0.6% cobalt. These elements are almost exclusively concentrated in small grains within the bulk meteorite and were separated magnetically. The $^{36}\text{Cl}/(\text{Fe, Co, Ni})$ ratio in the metallic phase was $3.6 \times$

TABLE 1
 ^{36}Cl , Cl^- and ^3H contents (using 2σ error) of groundwater samples

Sample	Date of sampling	$^{36}\text{Cl}/\text{Cl}$ ($\times 10^{15}$)	Cl^- [mg/L]	^{36}Cl atoms/L ($\times 10^{-6}$)	^3H [TU]
Munich, Löwenbräu 1	17.09.81	690 ± 160	0.44 ± 0.06	5.2 ± 1.4	<0.2
Rainbach 2	06.11.84	910 ± 260	1.04 ± 0.06	16.0 ± 4.7	2.1 ± 0.6
Braunau 1	06.11.84	65 ± 60	6.36 ± 0.32	7.0 ± 6.5	<0.7
Pattigham 2	06.11.84	790 ± 220	0.26 ± 0.04	3.5 ± 1.1	<0.7

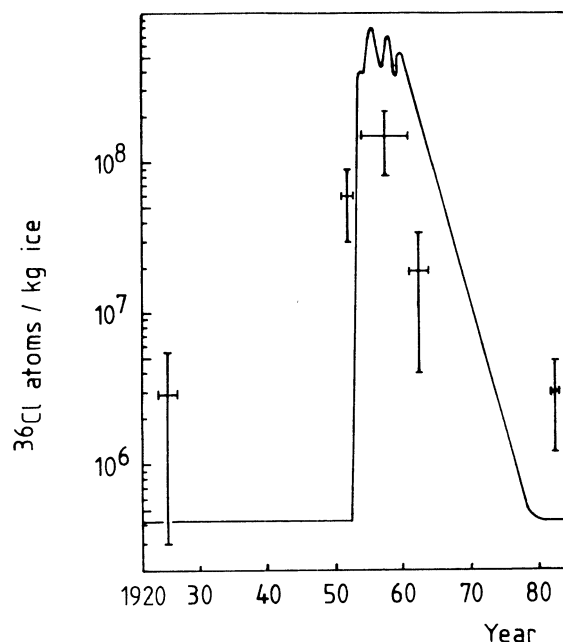


Fig 3. ^{36}Cl atoms per kg ice measured and estimated (after Bentley, Phillips & Davis, in press), on samples from the Vernagtferner (Oetztal Alps, Austria) at different deposition years (determined from ^2H , ^{18}O , and ^3H measurements by Stichler *et al*, 1982; Oerter & Rauert, 1982; Oerter, pers commun, 1984).

10^{-13} . Since ^{36}Cl is in saturation (Heusser, pers commun, 1984), the production rate can be calculated as $2.8 \pm 0.8 \times 10^{-4} \text{ s}^{-1} \text{ g}^{-1}$.

The Distribution of ^{36}Cl at Various Depths in the Vernagtferner (Oetztal Alps, Austria)

One of our previous measurements (Kubik, Korschinek & Nolte, 1984) showed a rather high ratio of $^{36}\text{Cl}/\text{Cl}$ (5×10^{-12}) in the drip water from a cave in Mount Ramesch (Styria, Austria). We attributed this to ^{36}Cl produced by the hydrogen bomb tests in the 1950s and 1960s. Measurements of Greenland ice (Elmor *et al*, 1982) showed that bomb-induced ^{36}Cl production has been some orders of magnitude larger than cosmic-ray-induced production. In order to detect bomb-produced ^{36}Cl , samples from the Vernagtferner ($46^\circ 52' \text{ N}$, Oetztal Alps, Austria) were measured (Table 2). The ice core date was determined from ^2H , ^{18}O , and ^3H measurements by Stichler *et al* (1982), Oerter and Rauert (1982), and Oerter (pers commun, 1984). The ^{36}Cl atoms/kg indicated for the sample (1954–1961) is clearly due to hydrogen bombs. This can also be seen in Figure 3, where the ^{36}Cl concentrations measured together with the predicted natural fallout concentrations are shown. Predicted values were estimated by taking calculated values from Bentley, Phillips, and Davis (in press) at the same latitude and a mean annual precipitation rate of ca 2000 kg m^{-2} . The measured value in the peak is ca 30% of the predicted value. In comparison to the

TABLE 2
 ^{36}Cl , Cl^- and ^3H contents (using 2σ error) of ice-core samples from the
 Vernagtferner (Oetztal Alps, Austria)

Core	Date of sampling	Depth [m]	Year*	$^{36}\text{Cl}/\text{Cl}$ ($\times 10^{15}$)	Cl^- [mg/kg]	^{36}Cl atoms/kg ($\times 10^{-6}$)	^3H [TU]
I/25-29	15.03.79	17-19	1961-1964	2720 ± 2000	0.41 ± 0.06	19 ± 15	353 ± 23
I/30-39**	15.03.79	19-25	1954-1961	8500 ± 2200	1.04 ± 0.20	150 ± 70	—
I/40-43	15.03.79	25-28	1951-1953	16600 ± 5700	0.21 ± 0.04	59 ± 30	16.8 ± 1.3
I/79-83	15.03.79	49-52	1924-1927	1400 ± 800	0.12 ± 0.04	2.9 ± 2.6	3.3 ± 0.7
IV/1-5	05.03.83	1-4	1982-1983	430 ± 260	0.40 ± 0.04	3.0 ± 1.8	29.5 ± 2.2

* Determined from ^2H , ^{18}O and ^3H measurements (Stichler *et al.*, 1982; Oerter & Rauert, 1982; Oerter, pers commun, 1984)

** Not all core segments were available for measurement

estimated values (Fig 3) the ^{36}Cl concentrations of the two samples from 1951-1953 and 1961-1964 seem to give younger ages. This can be attributed to infiltration of melt water, which transports younger ^{36}Cl fallout to older layers in the glacier ice, thus altering the ^{36}Cl concentrations in the ice samples.

Measurements of ^{36}Cl from Granite Samples Irradiated in 1945 by the Atomic Bomb Explosion in Hiroshima

The aim of these measurements is the determination of the RBE factor for fast neutrons. In spite of the vast medical information yet known, there

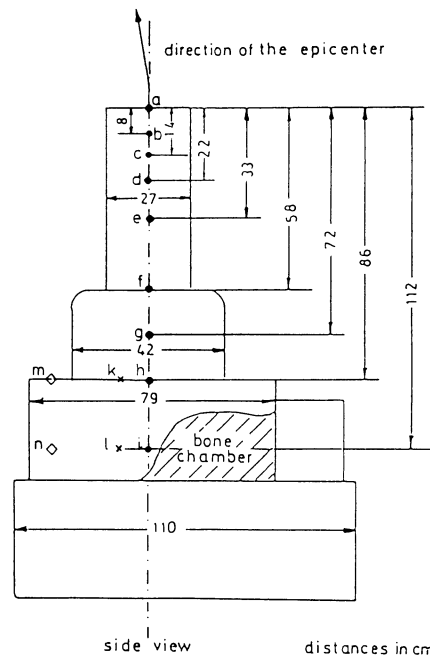


Fig 4. Position of the samples in a granite gravestone

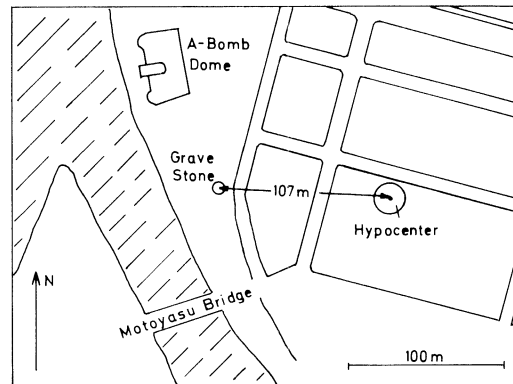


Fig 5. Location of the gravestone in Hiroshima

are still uncertainties concerning the neutron dose at the surface of the earth due to the atomic bomb explosion in Hiroshima. Until now, only one measurement of the dose of fast neutrons in Hiroshima is available (Yamasaki & Sugimoto, 1945). They measured ^{32}P activity ($T_{1/2} = 14.3$ d) produced by the reaction $^{32}\text{S} (n_{\text{fast}}, p) ^{32}\text{P}$. Measurements of ^{36}Cl by AMS is a

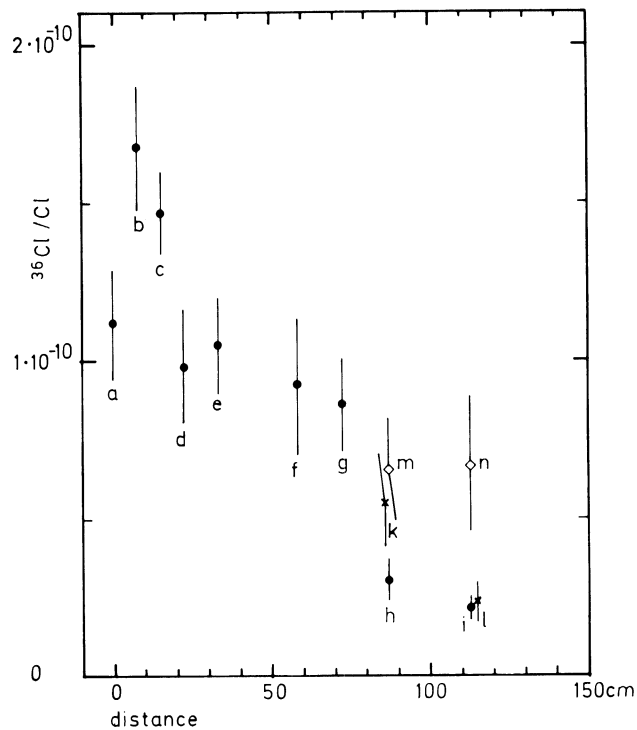


Fig 6. $^{36}\text{Cl}/\text{Cl}$ ratio of samples from various distances from the top of the gravestone

useful way for getting information about both, the thermic and the fast neutron flux. The ^{36}Cl concentration in samples from various depths in a granite gravestone were measured (see Fig 4). The irradiation of this gravestone took place at 107m from the hypocenter (epicenter 580m above ground), as shown in Figure 5. The $^{36}\text{Cl}/\text{Cl}$ ratio measured at various distances from the top of the gravestone is shown in Figure 6. According to the chemical composition of the granite, the reaction $^{35}\text{Cl}(n, \gamma) ^{36}\text{Cl}$ is the main source of ^{36}Cl production. Assuming the ^{36}Cl concentration at the surface is due only to thermic neutrons, a thermic dose of $3.5 \pm 1.0 \times 10^{12}\text{cm}^{-2}$ can be calculated. Calculations of the neutron spectrum at the gravestone are in progress.

ACKNOWLEDGMENTS

The groundwater and glacier-ice samples were kindly provided by J Goldbrunner, H Oerter, and W Rauert. We would like to thank S Hengst and E Sturm for the chloride and tritium analyses.

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