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¹³⁷Cs gamma-ray detection at Summit, Greenland

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ABSTRACT. Global fall-out from atmospheric testing of thermonuclear weapons produced horizon markers corresponding to the initiation of testing in 1953 and the maximum fall-out in 1963. The radioactive isotope ¹³⁷Cs associated with these events has a half-life of 30.2 years. Therefore, with the appropriate radiation detectors, this fall-out can be used as a long-term temporal indicator in glaciers and snowpack. A prototype γ -ray detector system was successfully tested and was used to make in-situ measurements of the ¹³⁷Cs marker in a borehole at Summit, Greenland. The system consisted of a 7.6 cm by 7.6 cm NaI(Tl) scintillation crystal/photomultiplier detector, commercial pre-amplifier, amplifier and power supplies, and a microcomputer-based pulse-height analyzer. The measurements were made in boreholes of 25.4 cm and 12.7 cm diameter to depths of 22 m. Based on the results reported here, the γ -ray detection technique promises to be a powerful way to locate quickly horizon markers in the field.

INTRODUCTION

Determining snow-accumulation rates is an obvious, but critical, component of glaciologic mass-balance studies. Glaciochemical investigations also demand knowledge of snow-accumulation rate; particularly temporal variations therein. A number of physical and chemical properties of snow (e.g. stable isotopes, soluble ionic species, dust) display annual variations that allow identification of annual increments of snow in polar and temperate snowpacks (Lorius and others, 1970). In practice, interpretation of any one of these "annual indicators" can be quite subjective; hence, several are often applied in any given study. In addition, "marker horizons" of known absolute age are invaluable to corroborate and validate annual-layer counting.

Global fall-out from atmospheric testing of thermonuclear weapons has long been recognized as providing several nearly ideal event horizons corresponding to the initiation of testing in 1953-54 and the maximum in fallout in 1963 (e.g. Picciotto and Wilgain, 1963; Picciotto and others, 1964, 1967; Lorius and others, 1970; Ambach and Dansgaard, 1970). A significant fraction of the fallout radioactivity consisted of 90 Sr (half-life = 28.8 years) and ^{137}Cs (half-life = 30.2 years), so the labeled layers of snow are still readily detectable (e.g. Gunten and others, 1983; Clausen and Hammer, 1988; Dibb, 1992) and should remain so for about 100 years. The accident at Chernobyl created a new ¹³⁷Cs-labeled layer corresponding to spring 1986 on many glaciers in the Northern Hemisphere (e.g. Pourchet and others, 1986; Davidson and others, 1987, 1989; Ambach and others, 1988; Haeberli and others, 1988; Dibb, 1989; Pinglot and Pourchet, 1989).

With the possible exception of visible stratigraphy, all techniques for annual-layer counting require post-field-

season processing and analysis before results are available. Radiochemical analyses to locate fall-out horizons also generally have significant turn-around times. However, the radioactivity in the labeled layers of snow provide the prospect of making measurements *in the field* that would immediately yield the depth to two or three time lines (i.e. 1953, 1963 and 1986).

¹³⁷Cs, present in bomb and Chernobyl fall-out, emits 662 keV γ -rays that, if present in sufficient quantity, are readily observable by a variety of scintillation detectors. A down-hole γ -spectrometry "package" of the kind described below including scintillator, photomultiplier tube, pre-amplifier and heaters can be assembled and lowered into 4 or 5 in [10.1 or 12.7 cm] diameter boreholes (see also Pinglot and Pourchet, 1989). With such an instrument and a very light drill (or hand auger), the depth to the marker horizons (hence, accumulation rates) can be determined relatively rapidly at a large number of sites. Such markers could be used to determine whether the desired depth for sampling had been reached. The core recovered when creating the boreholes would be available for a variety of other analyses. Sampling of these cores could be focused on known time intervals of highest interest, reducing the amount of material that would have to be returned to the laboratory for analysis.

We have assembled and tested a prototype down-hole γ -spectrometer to measure the bomb fall-out history at Summit, Greenland. Our results indicate that such a system will allow rapid determinations of fall-out history at many sites in the Northern Hemisphere and perhaps in Antarctica.

GAMMA-RAY DETECTION SYSTEM

The γ -ray detector used for the measurements was a cylindrical thallium-activated sodium iodode (NaI(Tl))

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scintillator, 7.6 cm in diameter and 7.6 cm thick. The scintillator crystal was optically coupled to a photomultiplier tube and both were enclosed in a stainless steel case. The scintillator/photomultiplier unit (model 3M3/3PSS) was obtained from the Bicron Corporation. The energy resolution of the detector, $\Delta E/E$, was measured during calibration and testing and was found to be 6.8% at 662 keV. Here, ΔE is the full width at half maximum (FWHM) of the photopeak.

For mechanical and thermal protection, the detector unit was enclosed in ~3 cm of ethafoam insulation and encased in a cylinder of plastic (12.7 cm outer diameter, 0.3 cm wall thickness) made from Lexan pipe. Since NaI(Tl) is sensitive to thermal shock and its light output depends on temperature (~0.6% °C⁻¹), two resistive heaters were attached to the metal case of the detector. The heaters could dissipate a maximum power of 40 W and were controlled with a thermostat set to operate at a temperature of 28°C. The heater voltage was provided by a variable voltage supply. The cabling between the detector and the control/data electronics consisted of a coaxial cable for high voltage, a coaxial cable for the preamplified signal and cables for the pre-amplifier and heater power.

Figure 1 shows a functional block diagram of the detector system. The photomultiplier high voltage was provided by a Tennelec TC952 power supply, operating at a nominal voltage of 835 V. A pulse pre-amplifier (Tennelec TC145) was mounted in the detector container close to the detector to drive the long (~30 m) signal cable and to minimize problems from noise pick-up in the cable. The signal pulses were shaped and amplified with a Tennelec TC246 amplifier/single-channel analyzer. The output was fed to a Nucleus PCA card, a 1024 channel pulse-height analyzer unit that plugs into the back plane of an IBM personal computer. Operation of the card and data handling were controlled with software that was packaged with the card. γ -ray spectra were accumulated



Fig. 1. Block diagram of the γ -ray detector system.

for pre-set lifetimes (generally 1000 s), displayed on the computer monitor and stored on floppy disk.

To analyze the data, we used several features of the PCA software: identification of regions of interest, calculation of peak centroids, calculation of FWHMs, etc. For more extensive analysis and graphing, spreadsheet software was used.

In order to monitor the stability of the detector continuously, a ¹⁰⁹Cd source was mounted at the scintillator end of the unit. This source produces a primary γ -ray line at 88 keV (Heath, 1964). The 88 keV line was used to monitor the gain of the system, which could be adjusted with the amplifier as necessary to keep a standard energy to pulse-height relationship.

DATA ACQUISITION

Two depth profiles of in-situ γ radioactivity were obtained at the GISP2 (Greenland Ice Sheet Project 2) deep-drilling camp at 72°34'N, 38°28'W in the summer of 1992. The first test was conducted in a hole that had been drilled in 1989. Originally, the hole had a 15.0 cm diameter but had been reamed to 25.4 cm for various logging instruments. In addition, the top 4 m of this hole had been lined with CIBA pipe to facilitate relocation and re-occupation of the hole. The second test was conducted in the hole created during recovery of a new 10.2 cm diameter firn core during the 1992 season. The hole diameter was approximately 12.7 cm.

For the first test, 1000s counts were collected in the 1989 hole at 1.0 m intervals from 5 to 25 m on 14 May. On 15 May, the depth resolution was improved by counting at the 0.5 m intervals between 10 and 19 m. The second test was conducted while drilling was still ongoing. During the evening of 3 June, 1000 s counts were collected every meter to a depth of 10 m, then every 0.5 m to 16.5 m, which was the current depth of the hole. The following evening, the profile was continued to 22 m depth, with 0.5 m resolution to 20 m and at 1.0 m steps below. On-site examination of the spectra revealed that activity in the ¹³⁷Cs window was essentially constant below 20 m and had a local maximum at 14.5 m. A longer count (4000 s) was then conducted at 14.5 m depth to confirm that the detector was in fact responding to ¹³⁷Cs.

During both tests, gain stability was ensured by monitoring the position of the 88 keV line from the ¹⁰⁹Cd source included in the detector package. In addition, before and after each series of down-hole measurements, a ¹³⁷Cs source was placed next to the detector and spectra were recorded to verify the location of the primary energy region of interest.

GAMMA-RAY SPECTRA AND DEPTH PROFILES

To show the major features of the γ -ray spectra measured in the boreholes, we use only the data from the 12.7 cm diameter borehole. The spectra from the 25.4 cm diameter borehole are similar but the signal produced by the ¹³⁷Cs deposition is somewhat smaller because of the increased distance between source and detector. Figure 2 shows γ -ray spectra accumulated at three different depths



Fig. 2. Gamma-ray count spectra (2.7 keV channel width) accumulated for 1000s at three different depths in the Summit borehole; the upper panel shows spectrum at 1 m, the center panel shows spectrum at 14.5 m and the lower panel shows spectrum at 22 m. Logarithmic scales on righthand side refer to the spectra plotted as a solid line; linear scales on lefthand side refer to spectra plotted as a dashed line. Sources of important features in the spectra are identified.

in the 12.7 cm diameter borehole: 1.0, 14.5 and 22.0 m. The spectrum at 1.0 m has three low-energy peaks due to the ¹⁰⁹Cd source at 23 ke V (¹⁰⁹Ag X-ray), 60 ke V (X-ray escape peak) and 88 keV (primary peak) (Heath, 1964). A relatively weak peak at 266 ± 8 keV from the ¹⁰⁹Cd source may be due to a 257 keV de-excitation γ -ray from an isomeric state in the ¹⁰⁹Ag daughter (Brandi and others, 1964). There is also a strong continuum, which dominates the spectrum at energies above 100 keV, and a peak at 519 ± 13 keV. We associate the continuum spectrum and the peak with cosmic-ray background (see discussion below). No significant contribution from ¹³⁷Cs at 662 keV is seen at this depth.

The spectrum at 14.5 m was accumulated at a depth at which we expect the greatest contribution from ¹³⁷Cs, based on a β -particle counting analysis of an ice core from a nearby borehole (Dibb, 1992). In fact, a peak at $675 \pm 20 \text{ keV}$ with a counting rate of $0.090 \pm 0.008 \text{ s}^{-1}$ is present in a 4000 s spectrum taken at this depth. The continuum is lower than that in the 1.0 m spectrum by a factor of ~4 (in the 132–211 keV range), and the peak near 519 keV is no longer present. Finally, the spectrum at 22.0 m shows only a weak continuum and the peaks from ¹⁰⁹Cd.

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In order to evaluate possible sources of the spectral features, we plot the depth profiles of counting rates for interesting energy ranges (Fig. 3). The depth profiles have been fitted with a four-parameter model consisting of an exponential, a constant and a normalized profile based on β -activity measurements by Dibb (1992) of an ice core from Summit, Greenland. The exponential component is most likely caused by cosmic-ray-produced γ -rays. The constant component is due to the ¹⁰⁹Cd calibration source and, possibly, a much weaker contribution from intrinsic radioactivity in the detector materials. The β -activity profile (see Fig. 4), since it is primarily due to ¹³⁷Cs and ⁹⁰Sr deposition, is expected to have the same depthdependence as γ -rays from ¹³⁷Cs. The β -profile was smoothed with a linear-chapeau filter to match the coarser position resolution of the γ -ray detector. (The position resolution is broadened by the transmission of the γ -rays through the firn, which has an attenuation length of about 20 cm.) The β -profile was shifted down in depth by 1.6 m to fit the data better. This shift is accounted for by snowfall in the 3 years between the core sampling and the γ -ray measurements. So the model of the γ -ray counting rate as a function of depth, R(d), can be written as

$$R(d) = a_1 e^{-d/a_2} + a_3 + a_4 f_\beta(d)$$

where d is the depth in meters, $f_{\beta}(d)$ is the smoothed and



Fig. 3. Counting rate-depth profiles in the Summit borehole for four energy ranges. A significant component from the "bomb-layer" is present in the range that contains the ¹³⁷Cs 662 keV photopeak (c) and a range where ¹³⁷Cs γ -rays from Compton scattering are expected (b).



Fig. 4. Upper panel shows β -activity profile measured by Dibb (1992) in an ice core from Summit, Greenland (dashed line) and smoothed profile using a seven-point linear-chapeau filter (solid line). Years of some significant changes in fall-out from nuclear weapons testing are indicated. Lower-panel data points show residual counting rate in the γ -ray energy range (576– 742 keV) containing the ¹³⁷Cs photopeak after the exponential and constant components are subtracted from the total rate. The solid line is the smoothed β -activity profile from the upper panel.

shifted β -activity as a function of depth and $a_1 - a_4$ are the four free parameters.

Figure 3a shows the depth-dependence of the counting rate in the energy range (63-130 keV) that contains the 88 keV ¹⁰⁹Cd peak. The error bars were calculated from Poisson counting statistics (i.e. \sqrt{n}/t , where *n* is the number of counts and *t* is the accumulation time). Most of the counting rate in the ¹⁰⁹Cd peak region is due to the ¹⁰⁹Cd source itself (~166 s⁻¹), but there is a component with approximately exponential depth-dependence that is caused by the underlying continuum. The scatter in the data points, which is greater than what is expected from counting statistics, is caused by detector-gain changes. In this and the following plots, a small number of outliers due to substantial gain changes have been removed.

Figure 3c shows the depth profile in the 576–742 keV energy range, which includes the 662 keV ¹³⁷Cs peak. In addition to an exponential decrease with depth and a small constant rate, a significant component with the same profile as the β -activity is present. Therefore, we identify this component with ¹³⁷Cs deposited during the era of above-ground nuclear bomb testing. The significance of the contribution of the bomb layer to the profile can be quantified with the "F test" statistic (Bevington, 1969). Applying this test, we find that this contribution is significant with a probability exceeding 99.9%.

In Figure 3d, we plot the depth profile of an energy range (793–1032 keV) above the ¹³⁷Cs peak. Here, the fall-out component is not significant. This strengthens the conclusion that the "bomb-test profile" is caused by ¹³⁷Cs deposition, since it is absent in an energy range where the ¹³⁷Cs emission will have no effect. In contrast, an energy range below 662 keV (415–576 keV) plotted in Figure 3b does show a significant bomb-test component. This is due to γ -rays from the 662 keV line that are Compton scattered to lower energies, both in the firn and in the detector.

We summarize the data on the depth profiles for various energies in Table 1. The table lists the model parameters from "least-squares" fits to the depth profiles. Also shown is the reduced χ -squared (χ^2) per degree of freedom for each fit. The exponential depth-dependence has an e-folding depth of \sim 3 m at all energy ranges. This continuum spectrum is probably caused by electromagnetic "showers" from cosmic rays interacting in the firn. The e-folding depth of 3 m corresponds to $150 \,\mathrm{g \, cm^{-2}}$ of water for a firn density of 500 kg m⁻³. This is in reasonable agreement with the e-folding depth of $\sim 180 \,\mathrm{g \, cm^{-2}}$ for cosmic-ray-produced γ -rays in the atmosphere (Peterson, 1963; Ryan and others, 1979). This comparison should be valid, since the nuclear and γ -ray interaction properties of air and water are similar when expressed in units of mass thickness, such as g cm⁻² (Zombeck, 1990; Particle Data Group, 1992). The cosmic-ray origin of this background is further supported by the presence of a peak at 519 ± 13 keV with an e-folding depth of 2.9 m in the borehole spectra. This line is probably the 511 keV

Energy range keV	a_1 counts s ⁻¹	<i>a</i> 2 m	a ₃ counts s ⁻¹	a_4 counts s ⁻¹ /counts h ⁻¹ kg ⁻¹	χ^2/dof^*
63-130	15.6 ± 3.6	3.85 ± 0.9	166.4 ± 1.1	0.01 ± 0.015	17.7
415-576	3.29 ± 0.5	2.89 ± 0.3	0.184 ± 0.010	0.00053 ± 0.00026	1.02
576-742	1.52 ± 0.21	2.96 ± 0.4	0.138 ± 0.005	0.00057 ± 0.00018	1.11
793-1032	1.30 ± 0.28	2.73 ± 0.5	0.10 ± 0.02	0.000073 ± 0.00013	1.79

Table 1. Parameters for fits to depth profiles

* dof, degrees of freedom = number of data points – number of parameters.

positron-annihilation line, which is also a prominent feature of the atmospheric γ -ray spectrum, where it has an *e*-folding depth of $185 \,\mathrm{g\,cm^{-2}}$ (Peterson, 1963). The constant term in the model could be due to intrinsic activity in the detector itself, in addition to the ¹⁰⁹Cd calibration source. The "hard" component of the cosmic rays, which has a much larger *e*-folding depth than the total component (e.g. Hayakawa, 1969), could also contribute to this term.

DISCUSSION AND CONCLUSIONS

The foregoing dissection of individual γ -ray spectra and examination of the depth-dependence of the response of the down-hole γ -spectrometry package provides a high level of confidence that the observed response in the ¹³⁷Cs window is in fact due to bomb fall-out. The underlying continuum is reasonably well understood and well represented by an exponential decay function and a constant (Table 1). Subtraction of these components from the measured activity in the ¹³⁷Cs window (576–742 keV) at each depth yields residuals which reflect the bomb fallout history at the Summit site (Fig. 4).

Figure 4 reproduces the detailed β -activity profile measured on a core collected in 1989 (Dibb, 1992) and the results of smoothing this record with a seven-point linear-chapeau filter. (This smoothed record is also $f_{\beta}(d)$, discussed above.) When the residual ¹³⁷Cs activity is superimposed on the smoothed beta record (with a 1.6 m depth offset to account for snow accumulation between 1989 and 1992), the agreement is striking. In particular, the in-situ γ -activity measurements capture the sudden onset of artificial radioactivity in the early 1950s, the decrease during the bilateral test moratorium (1959-61) and the sharp maximum in fall-out in 1963. There is also an indication of increased ¹³⁷Cs activity near 10 m depth, apparently reflecting French and Chinese atmospheric tests in the late 1960s and early 1970s, although this signal appears to be slightly offset (no more than 1 m) from the correlative peaks in the smoothed β -activity profile.

It must be pointed out that the detailed β -activity profile in Figure 4 includes 146 points, with a sampling interval of 10 cm in the depth range where the fall-out peaks are encountered. Analysis of these samples required nearly 3 months of counting time alone. In contrast, the entire in-situ γ -activity profile was collected in 35 000 s (just under 10 h). Despite some loss of depth resolution, the in-situ γ -ray measurements clearly provide nearly real-time access to several absolute time horizons in firm on the Greenland ice sheet.

Since we do not expect Greenland to be especially favored as a site of fall-out depositions, the in-situ technique should be useful, in general, on glaciers in the Northern Hemisphere. However, its feasibility in the Southern Hemisphere, and in Antarctica in particular, is not as clear. Based on the report by Dibb and others (1990) of radioactivity in the snowpack at the South Pole, we calculate that about 100 counts from ¹³⁷Cs would be measured by a 10 cm by 10 cm NaI(Tl) γ -ray detector in 1 h at the depth of the bomb fall-out maximum. The significance of such a measurement would depend, of course, on the background radiation at the site but this application certainly seems to be worth testing.

The Chernobyl marker has already been measured in situ on a glacier in the French Alps by Pinglot and Pourchet (1989). The signal from 137 Cs at this location varied markedly among boreholes separated by 500 m or less. In some boreholes, the signal was clearly observed within a counting time of 300 s; in others, it was not significant. Since the Chernobyl deposition is also known to be highly variable over larger areas (e.g. Dibb, 1989), its reliability as a marker depends strongly on location.

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