Research Report 208

**ARTIFICIAL
RADIOACTIVITY REFERENCE HORIZONS
IN GREENLAND FIRN**

by

**G. Crozaz
C.C. Langway, Jr.
E. Picciotto**

AUGUST 1966

**U.S. ARMY MATERIEL COMMAND
COLD REGIONS RESEARCH & ENGINEERING LABORATORY
HANOVER, NEW HAMPSHIRE**

DA Task IV014501B52A02

Distribution of this document is unlimited

PREFACE

This investigation was conducted by Drs. G. Crozaz and E. Picciotto, Service de Géologie et Géochimie Nucléaires, Université Libre de Bruxelles, and Dr. C. C. Langway, Jr., Snow and Ice Branch, Research Division, U. S. Army Cold Regions Research and Engineering Laboratory.

Financial support of the Institut Interuniversitaire des Sciences Nucléaires de Belgique and the United States National Science Foundation is gratefully acknowledged. We also wish to thank Mr. J. Saufney for laboratory technical assistance.

USA CRREL is an Army Materiel Command laboratory.

CONTENTS

	Page
Preface-----	ii
Summary-----	iv
Introduction-----	1
Artificial atmospheric radioactivity in arctic regions-----	1
Core sample and firn stratigraphy-----	2
Experimental procedures-----	5
Results-----	5
Natural radioactive nuclides-----	5
Gross beta activity-----	5
Strontium-90 deposition-----	7
Literature cited-----	8

ILLUSTRATIONS

Figure

1. Gross beta activity in the air at Kodiak and Thule in relation to the main nuclear bomb test series in the atmosphere. Variation of Sr-90 activity with depth in a snow core from Site II----- 3
2. Density, stratigraphic profile and yearly accumulation from the Camp Century firn core----- 4
3. Gross beta activity of snow with respect to depth of summer surfaces, from Figure 2----- 6

SUMMARY

Total beta measurements have been made on melt water samples from a stratigraphically dated firn core profile from the inland Greenland ice sheet ($77^{\circ} 10' \text{N}$, $61^{\circ} 08' \text{W}$). A marked increase in radioactivity is found in the 1953 firn layer which corresponds to the first important fallout from nuclear test bombs. The pre-1953 natural beta activity is 5 dpm/kg. The influx of artificial debris from the Ivy tests in 1953 is noted by a sharp rise in beta activity to 10 dpm/kg. Total ^{90}Sr deposit to June 1964 is 9.3 ± 1.5 millicuries/ km^2 ($24 \pm 4 \text{ mCi}/\text{mi}^2$). Average ^{210}Pb activity at time of deposit is 3.9 ± 0.4 dpm/kg.

ARTIFICIAL RADIOACTIVITY REFERENCE HORIZONS IN GREENLAND FIRN

by

G. Crozaz, C.C. Langway, Jr. and E. Picciotto

INTRODUCTION

The determination of snow accumulation rates is a vital factor in estimating the net budget of polar ice sheets. These measurements are generally based on the stratigraphic interpretation of profiles from exposed pits or augered cores, and less frequently on direct measurements (stakes) or the interpretation of stable isotope ratio variations (usually oxygen). These classical measuring methods incorporate a variable amount of personal interpretation. Several investigators have discussed or demonstrated that it is possible to measure the average snow accumulation rate over the last decade by using reference levels formed by the fallout of radioactive debris from high atmospheric yield nuclear bomb tests (Martell, 1959; Picciotto *et al.*, 1962; Picciotto and Wilgain, 1963; Vickers, 1963; Wilgain *et al.*, 1965).

The best measure of radioactive bomb debris concentration is given by the emission activity of a well defined nuclide, such as Sr-90. Measurement of Sr-90 activity, however, requires rather elaborate chemical separation and samples of the order of several kilograms. On the other hand, measurement of the gross beta activity is a much simpler procedure requiring samples of only 10 to 100 g. Using gross beta activity measurements has two main disadvantages: (1) the contributions of the most important natural radioactive nuclides (Pb-210 and K-40) must be taken into account, and (2) the beta activity of mixtures of nuclear bomb debris change and decay with time in a complicated and often unpredictable way.

Picciotto and Wilgain (1963) have shown that in Antarctica the contributions from natural radioactive nuclides are negligible compared to artificial radioactivity concentrations. A reference horizon was found in the 1954-55 austral summer layer (Wilgain *et al.*, 1965) which appears to extend over all of inland Antarctica and corresponds to the fallout from the Castle bomb test series. This horizon is marked by a sudden increase in both the gross beta and Sr-90 activities.

The objective of this study is to investigate the gross beta activity in the Greenland firn layers and to determine whether artificial nuclide variations exist which could be used as accumulation reference horizons. The Pb-210 activity is also measured in order to assess the contribution of the natural radioactive nuclides and to appraise the application of the Pb-210 dating method (Goldberg, 1963; Crozaz *et al.*, 1964) for this region of the Greenland ice sheet.

ARTIFICIAL ATMOSPHERIC RADIOACTIVITY IN ARCTIC REGIONS

The general evolution of artificial atmospheric radioactivity in the Arctic since 1950 is known. Gross beta activity measurements of arctic air have been made from 1950 to 1960 by the U.S. Naval Research Laboratory (NRL) at Kodiak, Alaska (58°N, 152°W), and from 1958 to 1962 at Thule, Greenland (76°33'N,

2 ARTIFICIAL RADIOACTIVITY HORIZONS IN GREENLAND FIRN

68°49'W) (Lockhart, 1961; Lockhart et al., 1964). The results of these measurements are summarized in Figure 1 (fine line curve). Activities below 0.1 dpm/m³* are not recorded. The yearly averages (wide line curve) are computed from summer to summer to allow direct comparison with our firn core measurements. The Kodiak data are plotted for 1951-1959; the Thule data between 1958-1964. The dotted curve, showing the air activity at Thule, Greenland, after January 1963, is extrapolated from air measurements made at Thule after the 1958 bomb tests and from the known ratios of stratospheric injections in 1958 and 1962. Figure 1 also contains the data of the main atmospheric nuclear bomb test series. From these data the following interpretations are made:

- (1) The small or large yield Soviet tests that took place in Siberia or in Novaya-Zemlya had a strong and immediate influence at the measuring sites, indicating tropospheric fallout.
- (2) The low yield (less than 1 megaton) U.S. tests carried out at the Nevada site had a weaker influence. Detection of this fallout at the measuring stations in less than 1 month also indicates a tropospheric process.
- (3) The high yield U.S. and U.K. tests conducted at the Pacific Ocean sites did not immediately contaminate the air at the measuring sites, indicating no tropospheric fallout.
- (4) The spring following, high yield Soviet nuclear tests are clearly indicated in the northern latitudes air measurements.

In addition to these ground level atmospheric radioactivity measurements, important information has been published by Martell (1959) on the Sr-90 concentrations in a well dated firn profile from Site II, Greenland (76°59'N, 56°04'W). These results are also presented in Figure 1. The Sr-90 activity is shown to be negligible before the summer of 1953. Thereafter the activity rises to 1 dpm./kg in the 1953 and 1954 snow deposits and increases rapidly to 3.8 dpm./kg after the 1955 summer.

CORE SAMPLE AND FIRN STRATIGRAPHY

The firn core used in this investigation was obtained in the vicinity of Camp Century, Greenland (77°10'N, 61°08'W), in July 1964. Camp Century is located 210 km (130 miles) east of Thule at an elevation of 1885 m (6180 ft) and, from deep bore hole measurements, has a mean annual surface air temperature of -24.6C (B. L. Hansen, personal communication).

The 12-m deep hand-augered core (7.6 cm diam) was bored about 5 km southwest of the main camp. After recovery the core was placed on a table where light was transmitted through the core to show subtle stratigraphic features. Density measurements were subsequently performed on the stratigraphically defined core pieces. These data are shown in Figure 2 along with the interpretations of the yearly boundaries (late summer to late summer). The heavy melt shown in 1953 is percolation from the warm July 1954 period. Average net snow accumulation for the 1947-64 period varies from 16.4 to 49.4 g/cm² -yr, with a mean value of 31.2 g/cm² -yr over the 17-year profile. Independent snow stake measurements from the general Camp Century area (Mock, 1965) give accumulation values of 32 to 36 g/cm² -yr for the 1961-63 period.

*Disintegrations per minute per cubic meter.

Figure 1. Gross beta activity in the air at Kodiak and Thule (Lockhart, 1961; Lockhart et al., 1964) in relation to the main nuclear bomb test series in the atmosphere. Variation of Sr-90 activity with depth in snow core from Site II (Martell, 1959).

Figure 1 (Legend)

Test series	Total yield (megatons)	Test series	Total yield (megatons)	Test series	Total yield (megatons)
Nevada		Pacific Proving Ground (PPG)		USSR	
R-Ranger	0.040	I-Ivy	~ 3	1	0.04
B-J-Buster Jangle	0.072	C-Castle	~ 53	2	1
T-S-Tumbler Snapper	0.104	Re-Redwing	~ 20	3	0.1
U-K-Upshot Knothole	0.252	G1-Grapple	~ 6	4	3
T-W-Teapot Wigwam	0.197	G2-Grapple		5	5
P-Plumbbob	0.343	G3-Grapple		6	
HII-Hardtack II	0.019	HI-Hardtack I	~ 37	7	12
		D-Dominic	37	8	
				9	
				10	30
				11	120
				12	180

Figure 2. Density, stratigraphic profile and yearly accumulation from the Camp Century firn core.

After the detailed stratigraphic observations and density measurements, the external surfaces of the individual core pieces were carefully scraped and placed in precleaned wide-mouthed polyethylene containers. The containers were capped, allowed to melt at room temperature, and sealed with a special Teflon tape. A water level line was placed on the outside of each container.

EXPERIMENTAL PROCEDURES

Before transfer of melt water from the containers the samples were acidified with nitric acid. The gross beta activity was measured on about 50-ml aliquot portions using a procedure similar to that described by Picciotto and Wilgain (1963). Strontium and lead carriers were also added to each original melt water container. After removing the 50-ml aliquot for beta activity measurements, all samples were combined for measurement of total Sr-90 and average Pb-210 concentrations. The experimental procedures for Sr-90 are described in Wilgain *et al.* (1965) and for Pb-210 in Crozaz *et al.* (1964).

RESULTS

Natural radioactive nuclides

Measurements of the ionic concentrations of dissolved solids in the 1963-64 snow accumulation layer at Camp Century show that the average potassium concentration is 0.05 mg/l (Langway, in press). This value corresponds to a K-40 beta activity of the order of 0.1 dpm/kg, which is negligible in comparison to the Pb-210 and other artificial beta activities.

The average Pb-210 activity in the 1947 to 1964 snow accumulation is 3.9 ± 0.4 dpm/kg with corrections applied for radioactive decay since time of deposit. The estimated error takes into account the statistical counting fluctuations, the error of the chemical yield, and the uncertainty in the age of the samples. This value is appreciably higher than the 1.2 dpm/kg given by Goldberg (1963) for the initial Pb-210 activity in snow samples from the south Greenland Station Crete (71°07'N, 37°19'W). It is not possible at this time to determine if this difference is due only to a geographical effect.

Gross beta activity

The results of the distribution of gross beta activity with depth, expressed in dpm/kg of snow, are shown in Figure 3. The counter was arbitrarily calibrated with a potassium standard in thin layer. Taking the yearly accumulation from the stratigraphic analysis as the time standard (Fig. 2), certain features are evident in the distribution of the gross beta activity over the depth profile. One observes an almost constant activity of 5.4 ± 0.5 dpm/kg from 1947 to 1952. A sharp rise to 10 dpm/kg is noted in the beta activity between the summers 1952 and 1953, followed by an irregular but steady increase to 1958. A steep decrease is noted between the summers 1959 and 1961, and, finally, a second important increase is shown after the summer of 1961.

The behavior of the gross beta activity in the firn profile is in good agreement with the atmospheric data from the Naval Research Laboratory (Fig. 1) and is easy to interpret on this basis. A closer agreement is not to be expected since most of the maximum peaks seen in the NRL results are due to tropospheric fallout of young products with short half-lives, most of which have completely decayed before our measurements (see for instance the peak in air activity in October-December 1951).

Figure 3. Gross beta activity of snow (measured Jan 1965) with respect to depth of summer surfaces, from Figure 2.

From 1947 to 1952, the gross beta activity in the firn layers is attributed entirely to natural radioactive nuclides, primarily Pb-210 and Bi-210. The first increase in artificial activity shows up at 7.80 m, the spring of 1953. We are confident of the stratigraphic position of this layer because of an exceptionally good meteorological index horizon (heavy melting) at the 1954 level which is clearly marked over the north Greenland ice sheet (Langway, 1961). This first noticeable increase in beta activity, therefore, must be due to the fallout of the stratospheric debris from the Ivy tests. It occurs too early to be attributed to the first Soviet thermonuclear bomb tests (August 1953).

From 1953 to 1958, the progressive increase of the activity corresponds to the almost continuous succession of high yield bomb tests. The core samples were cut too thick to show the expected seasonal fluctuations. The drop in activity during the years 1959, 1960 and 1961 corresponds to the moratorium on atmospheric nuclear bomb testing. The resumption of the tests during the fall of 1961 is clearly shown by an increase in the beta activity marking a second reference horizon. It is of interest to point out that in Antarctica the Ivy test fallout is not as prominent as in Greenland, and that the main radioactive reference horizon in Antarctica is the February 1955 firn layer resulting from the Castle tests (Picciotto and Wilgain, 1963). On the Greenland ice sheet, the fallout from the Castle tests did not form a clearly marked horizon, because of atmospheric mixing of the debris with the products of the Soviet tests, which had a predominant influence due to their geographical location.

To conclude the discussion of the gross beta activity measurements on the Greenland firn core we show the existence of a distinct radioactive layer, which corresponds to the spring-summer 1953 snow deposit and a second important index horizon at the 1961 level. These horizons could be very useful to glaciologists for estimating the average snow accumulation rate since 1953 over all of the Greenland ice sheet. Confirmation of these results requires additional measurements on more stratigraphically dated firn profiles from various inland locations.

Strontium-90 deposition

Since no direct Sr-90 measurements exist for the latitudes between 70°N and 90°N it was of interest to measure the total cumulative Sr-90 deposit between 1947 and July 1964 as reflected in the firn core samples. Results indicate that the cumulative Sr-90 deposit is 9.3 ± 1.5 mC/km² (24 ± 4 mC/mile²). This value is the lowest found in the Northern Hemisphere and is compatible with the 17 mC/mile² value reported at Thule during the October 1959-June 1964 time interval (U.S. Atomic Energy Commission, 1964).

SUMMARY

Melt water samples from a 12 m deep vertical firn core from Camp Century, Greenland, were measured for gross beta and total Sr-90 content. Annual firn layers representing the time interval 1947-64 were dated by classical stratigraphic techniques. Average net snow accumulation is 31.2 g/cm² - yr over the profile. An easily identifiable index horizon is found to have formed by the first important fallout of radioactive debris from nuclear test bombs. The spring 1953 layer is clearly marked by a sudden increase in gross beta activity which corresponds to the stratospheric fallout from the Ivy test series. The autumn 1961 firn layer will be a useful future index horizon. The gross beta activity before 1952 (measured in January 1965) is about 5 dpm/kg and rises above 10 dpm/kg in 1953 and to over 100 dpm/kg after 1962. The total deposition of Sr-90 to June 1964 is 9.3 ± 1.5 mC/km². The average Pb-210 activity at the moment of deposition is 3.9 ± 0.4 dpm/kg.

8. ARTIFICIAL RADIOACTIVITY HORIZONS IN GREENLAND FIRN

LITERATURE CITED

- Crozaz, G., Picciotto, E. and DeBreuck, W. (1964) Antarctic snow chronology with Pb-210, Journal of Geophysical Research, vol. 69, p. 2597-2604.
- Goldberg, E.D. (1963) Geochronology with Pb-210, Symposium on Radioactive Dating, International Atomic Energy Agency, Athens, Nov. 1962, p. 121-130.
- Langway, C.C., Jr. (1961) Accumulation and temperature on the inland ice of north Greenland, 1959, Journal of Glaciology, vol. 3, p. 1017-1044.
- _____ (in press) "Chemical composition studies" in Stratigraphic analysis of a deep ice core from Greenland, U.S. Army Cold Regions Research and Engineering Laboratory (CRREL) Research Report 77 (in press).
- Lockhart, L.B., Jr. (1961) Atmospheric radioactivity at Kodiak and Wales, Alaska, U.S. Naval Research Laboratory, Report 5658, 16 p.
- Lockhart, L.B., Jr.; Patterson, R.L., Jr.; Saunders, A.W., Jr.; and Black, R.W. (1964) Summary report on fission product radioactivity in the air along the 80th meridian (West) 1957-1962, U.S. Naval Research Laboratory, Report 6104, 29 p.
- Martell, E.A. (1959) Atmospheric aspects of strontium-90 fallout, Science, vol. 129, p. 1197-1206.
- Mock, S.J. (1965) Glaciological studies in the vicinity of Camp Century, Greenland, USA CRREL Research Report 157.
- Picciotto, E. and Wilgain, S. (1963) Fission products in Antarctic snow, a reference level for measuring accumulation, Journal of Geophysical Research, vol. 68, p. 5965-5972.
- Picciotto, E.; Wilgain, S.; Kipter, P.; and Boulenger, R. (1963) "Radio-activité de l'air dans l'Antarctique en 1958 et profil radioactif entre 60°N et 70°S" in Radioisotopes in the physical sciences and industry, International Atomic Energy Agency, Vienna, p. 45-56.
- U.S. Atomic Energy Commission (1965) Health and safety report, HASL-164, p. 111.
- Vickers, W.W. (1963) Geochemical dating techniques applied to Antarctic snow samples, International Association of Scientific Hydrology, Publication no. 61, p. 199-215.
- Wilgain S., Picciotto, E. and Debreuck, W. (1965) Strontium-90 fallout in Antarctica, Journal of Geophysical Research, vol. 70, p. 24.

DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)		2a. REPORT SECURITY CLASSIFICATION	
U.S. Army Cold Regions Research and Engineering Laboratory, Hanover, N.H.		Unclassified	
3. REPORT TITLE		2b. GROUP	
ARTIFICIAL RADIOACTIVITY REFERENCE HORIZONS IN GREENLAND FIRN			
4. DESCRIPTIVE NOTES (Type of report and inclusive dates)			
Research Report			
5. AUTHOR(S) (Last name, first name, initial)			
Croaz, G.; Langway, C.C., Jr., and Picciotto, E.			
6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS	
Aug 1966	12	13	
8a. CONTRACT OR GRANT NO.	9a. ORIGINATOR'S REPORT NUMBER(S)		
b. PROJECT NO.	Research Report 208		
c. DA Task IV014501B52A02	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)		
d.			
10. AVAILABILITY/LIMITATION NOTICES			
Distribution of this document is unlimited			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY	
		U.S. Army Cold Regions Research and Engineering Laboratory	
13. ABSTRACT			
<p>Total beta measurements have been made on melt water samples from a stratigraphically dated firn core profile from the inland Greenland ice sheet (77°10'N, 61°08'W). A marked increase in radioactivity is found in the 1953 firn layer which corresponds to the first important fallout from nuclear test bombs. The pre-1953 natural beta activity is 5 dpm/kg. The influx of artificial debris from the Ivy tests in 1953 is noted by a sharp rise in beta activity to 10 dpm/kg. Total ⁹⁰Sr deposit to June 1964 is 9.3 ± 1.5 millicuries/km² (24 ± 4 mC/mi²). Average ²¹⁰Pb activity at time of deposit is 3.9 ± 0.4 dpm/kg.</p>			

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Ice--Radioactivity--Greenland Snow--Radioactivity--Greenland Radioactive dating--Greenland						

INSTRUCTIONS

1. **ORIGINATING ACTIVITY:** Enter the name and address of the contractor, subcontractor, grantee, Department of Defense activity or other organization (*corporate author*) issuing the report.

2a. **REPORT SECURITY CLASSIFICATION:** Enter the overall security classification of the report. Indicate whether "Restricted Data" is included. Marking is to be in accordance with appropriate security regulations.

2b. **GROUP:** Automatic downgrading is specified in DoD Directive 5200.10 and Armed Forces Industrial Manual. Enter the group number. Also, when applicable, show that optional markings have been used for Group 3 and Group 4 as authorized.

3. **REPORT TITLE:** Enter the complete report title in all capital letters. Titles in all cases should be unclassified. If a meaningful title cannot be selected without classification, show title classification in all capitals in parenthesis immediately following the title.

4. **DESCRIPTIVE NOTES:** If appropriate, enter the type of report, e.g., interim, progress, summary, annual, or final. Give the inclusive dates when a specific reporting period is covered.

5. **AUTHOR(S):** Enter the name(s) of author(s) as shown on or in the report. Enter last name, first name, middle initial. If military, show rank and branch of service. The name of the principal author is an absolute minimum requirement.

6. **REPORT DATE:** Enter the date of the report as day, month, year; or month, year. If more than one date appears on the report, use date of publication.

7a. **TOTAL NUMBER OF PAGES:** The total page count should follow normal pagination procedures, i.e., enter the number of pages containing information.

7b. **NUMBER OF REFERENCES:** Enter the total number of references cited in the report.

8a. **CONTRACT OR GRANT NUMBER:** If appropriate, enter the applicable number of the contract or grant under which the report was written.

8b, 8c, & 8d. **PROJECT NUMBER:** Enter the appropriate military department identification, such as project number, subproject number, system numbers, task number, etc.

9a. **ORIGINATOR'S REPORT NUMBER(S):** Enter the official report number by which the document will be identified and controlled by the originating activity. This number must be unique to this report.

9b. **OTHER REPORT NUMBER(S):** If the report has been assigned any other report numbers (*either by the originator or by the sponsor*), also enter this number(s).

10. **AVAILABILITY/LIMITATION NOTICES:** Enter any limitations on further dissemination of the report, other than those imposed by security classification, using standard statements such as:

- (1) "Qualified requesters may obtain copies of this report from DDC."
- (2) "Foreign announcement and dissemination of this report by DDC is not authorized."
- (3) "U. S. Government agencies may obtain copies of this report directly from DDC. Other qualified DDC users shall request through _____."
- (4) "U. S. military agencies may obtain copies of this report directly from DDC. Other qualified users shall request through _____."
- (5) "All distribution of this report is controlled. Qualified DDC users shall request through _____."

If the report has been furnished to the Office of Technical Services, Department of Commerce, for sale to the public, indicate this fact and enter the price, if known.

11. **SUPPLEMENTARY NOTES:** Use for additional explanatory notes.

12. **SPONSORING MILITARY ACTIVITY:** Enter the name of the departmental project office or laboratory sponsoring (*paying for*) the research and development. Include address.

13. **ABSTRACT:** Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as (TS), (S), (C), or (U).

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

14. **KEY WORDS:** Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.