

Report for Camp Century Project

# **Radioactivity in an Ice Core from Camp Century, Greenland**

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## Summary

An ice core was extracted in July 2017 from Camp Century in Northwest Greenland and subsequently examined for indications of radioactive contamination from the operation of a nuclear reactor during 1960-1964. It is concluded that there is no indication that significant radioactive contamination was released into the air as a result of the Camp Century reactor operation. It might be possible that the measured activities for  $^{90}\text{Sr}$ ,  $^{129}\text{I}$  and  $^{137}\text{Cs}$  are underestimated as we find activity concentrations for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  lower than historical concentrations corrected for decay to the time of sampling of the ice core. Further investigation will be carried out to evaluate the absorption of  $^{137}\text{Cs}$  activity onto particles by careful experimental design and low-level gamma measurement for the upper layer (above 2000 cm) ice core samples.

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## Introduction

During 1960-1964 a portable nuclear reactor of moderate size, type PM-2A, operated below the surface of the ice sheet at Camp Century in Northwest Greenland. The reactor had a thermal power of 10 MW that was used for electricity (1.5 MW) and heating. Conventional and radioactive wastes arising from the operation of the camp were discharged into the Greenland Ice Sheet. The Danish-American agreement permitted discharge of liquid radioactive waste into the ice cap up to 2 GBq/y (50 mCi/y) while all solid radioactive waste must be removed from Greenland (Nielsen and Nielsen, 2017).

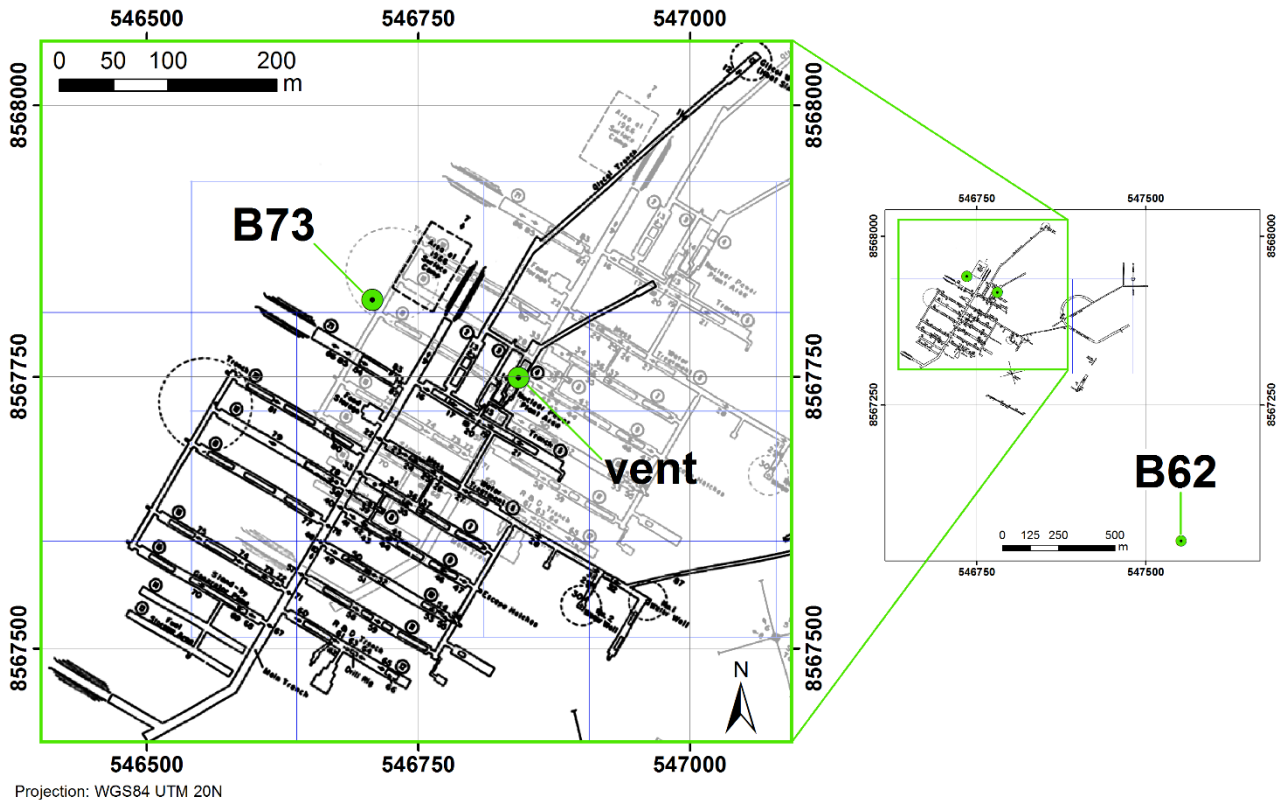
An environmental monitoring programme was carried out by Camp Century staff during the operation of the reactor covering alpha/beta/gamma measurements of background radiation and measurements of radioactivity in discharged coolant, surface air and snow, and the subsurface drinking water supply. In 1961 and 1962 alpha and beta radioactivity in air and snow showed increased levels due to fallout from atmospheric nuclear weapons testing by the Soviet Union. The measurement data was shared with Danish authorities who concluded in 1964 that the reactor operation has not resulted in any significant increase in levels of radiation or radioactivity in Camp Century or surroundings.

Today, concern about potential remobilization of radioactive pollution at Camp Century under climate change, and its impact on the environment, has prompted further investigations. Between December 2017 and June 2018 the Center for Nuclear Technology, Technical University of Denmark (DTU Nutech) carried out an initial screening programme for anthropogenic radioactivity in an ice core recovered about 150 m downwind from the estimated present-day location of the reactor ventilation stack. The radionuclides considered included tritium ( $^3\text{H}$ ; radioactive hydrogen), strontium-90 ( $^{90}\text{Sr}$ ), cesium-137 ( $^{137}\text{Cs}$ ) and iodine-129 ( $^{129}\text{I}$ ). These radionuclides are fission products, which are formed during operation of nuclear reactors and detonation of nuclear bombs. Furthermore, the half-lives of these radionuclides are sufficiently long (12 y, 30 y, 29 y and 16 My, respectively) that they may still be detectable after 60 years decay. The overall results are presented and discussed in this report.

## Ice Core Data

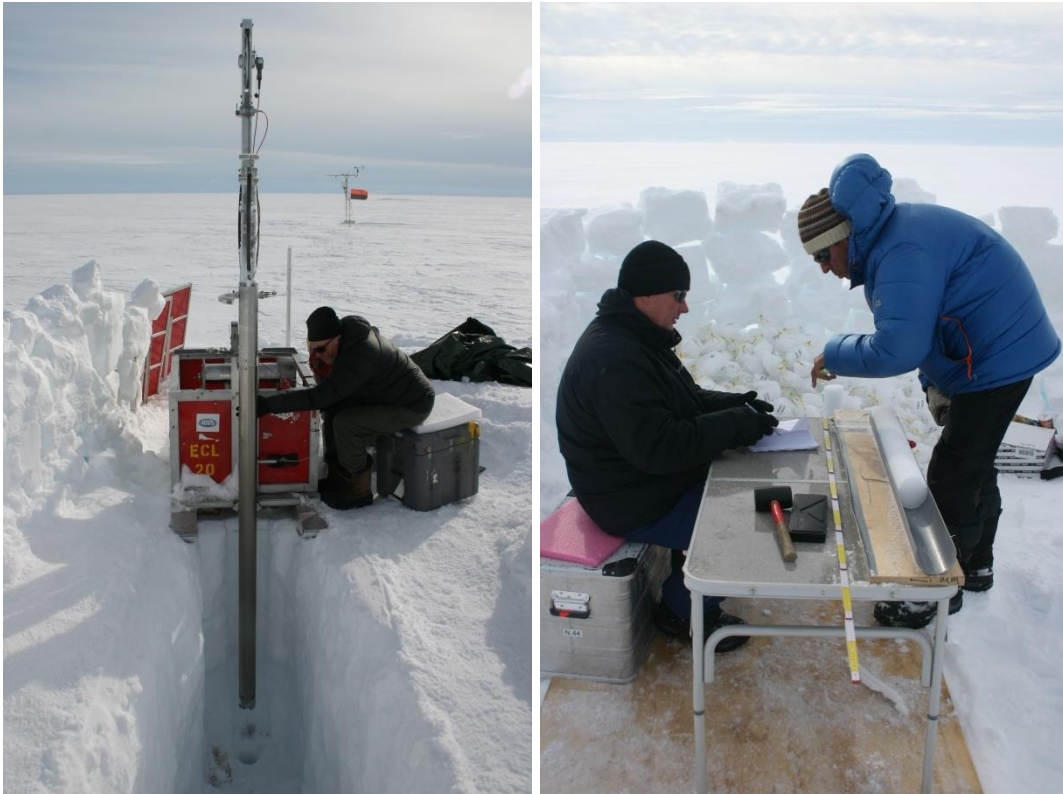
The present-day location of Camp Century's subsurface infrastructure was estimated prior to fieldwork by georeferencing a historical site map (Kovacs, 1970). This georeferencing used a single tie-point, the location of the original drill tower, corrected for motion since its last precise survey in 1986 (Gundestrup et al., 1987). The nuclear reactor vent is assumed to be located directly above the nuclear power plant trench. The general ice core location was pre-selected to be downwind of this estimated reactor vent position. The final ice core location (B73 at 77.1826 °N, 61.1125 °W and 1887 m above

sea level) was selected using ice-penetrating radar to find a drill site with no subsurface debris. We estimate that the ice core was recovered 150 m downwind relative to the reactor vent (Figure 1).



**Figure 1. Ice core location (B73) relative to the estimated reactor vent position. The dark map depicts Camp Century infrastructure georeferenced to present-day coordinates (Kovacs, 1970). The lightly shaded map depicts estimated infrastructure in 1960, prior to movement due to ice flow. Inset depicts the location of ice core B62.**

The ice core was recovered using the University of Wisconsin Eclipse2 ice drilling system. This relatively lightweight (800 kg) tip-drill system operates without drilling fluid to depths of approximately 300 m (Blake et al., 1998). Ice core was recovered from between 1.80 m and 40.00 m depth over two days (25 and 26 July 2017). Core sections were cut into samples of 10.0 cm in length, or 9.9 cm accounting for saw loss, and weighed for density on an open-air core logging table (Figure 2). Given an internal drill diameter of 8.0 cm, each sample was equivalent to between 145 mL (at 300 kg/m<sup>3</sup> firn density) and 440 mL (at 915 kg/m<sup>3</sup> pure ice density) of water. Samples were sealed into individual Whirlpak bags within approximately 15 minutes of extraction. The samples were allowed to melt in transit back to Copenhagen.

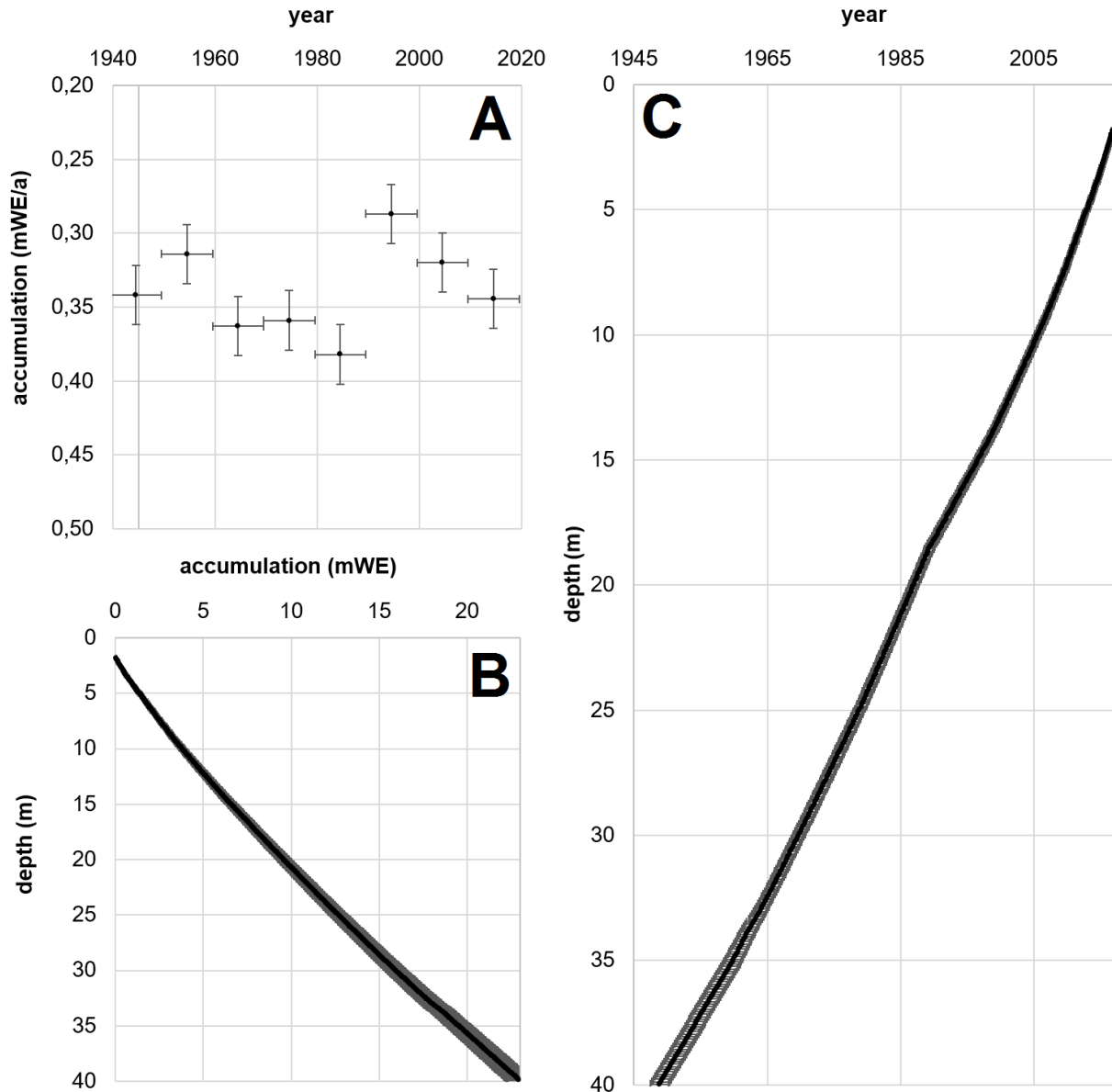


**Figure 2. Left: The University of Wisconsin Eclipse2 ice-drilling system at B73 operated by Mike Jayred. Right: Allan Pedersen (L) and Horst Machguth (R) processing ice core sections for density and samples. (Photos: W. Colgan)**

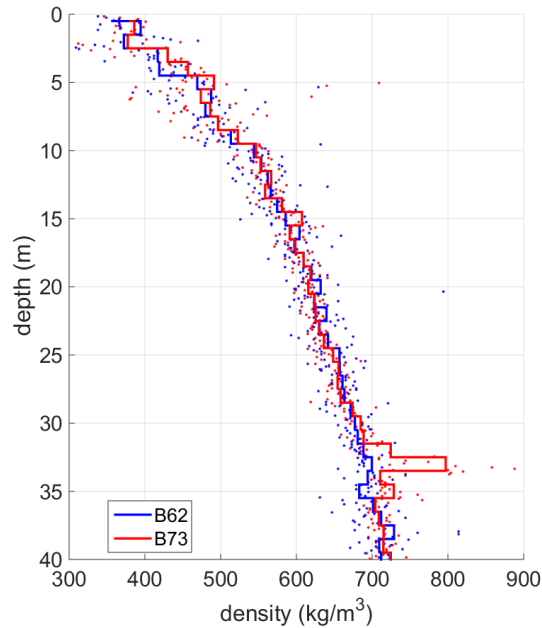
The age-depth relation of the ice core and its samples was estimated by combining the measured accumulation-depth profile with a previously derived decadal accumulation rate history (Buchardt et al., 2012). The measured accumulation-depth profile, in metres of water equivalent (mWE), was derived from depth-integration of individual sample densities (Figure 3). Uncertainties in the accumulation-depth profile and accumulation rate history, which are assumed to be independent, were summed in quadrature. Date uncertainty increases from  $2016.8 \pm 0.3$  at 1.80 m depth to  $1948.9 \pm 1.4$  at 40.00 m depth. The accumulation rate history, derived from a remote undisturbed core site, likely underestimates the local accumulation rate at Camp Century during its active period. Thus, ice core dates may be biased older than calculated below c. 1967.

This age-depth relation for B73 suggests that ice deposited during the 1960.0 to 1964.9 nuclear power plant operational period is now located between 32.55 and 34.95 m depth. Comparison of the B73 density-depth profile with that of ice core B62, which was contemporaneously recovered 1.5 km away at an undisturbed upwind site, highlights that densities inside the debris field are significantly higher than outside the debris field between 32 and 35 m depth (Colgan et al., 2018; Figure 4). This high-

density layer, which is discoloured slightly yellow in appearance and contains visible macroscopic particulates, likely reflects enhanced local compaction and pollution during the Camp Century active period. The recovered ice samples therefore span the entire Camp Century active period.



**Figure 3. A: Previously derived decadal accumulation rate history in the vicinity of Camp Century (Buchardt et al., 2012). B: Measured accumulation with depth profile, in metres of water equivalent (mWE). C: Age-depth relation for ice core B73. In all subplots, whiskers denote associated uncertainty.**



**Figure 4. Near-surface density profiles measured both inside (B73) and outside (B62) the Camp Century debris field. Points denote individual 10-cm samples. Lines denote 1-m averages. The high-density layer between 32 and 35 m depth within the debris field likely reflects enhanced local compaction during the Camp Century active period.**

### Sample Preparation

The ice core samples were received at DTU Nutech in late September 2017 where analyses focused on the depth range 25 to 40 m, which represents the  $1978.7 \pm 0.8$  to  $1948.9 \pm 1.4$  period. On arrival, a significant fraction (about 18%) of the total sample mass was found to be lost in transit even though each 10-cm ice sample was placed in double Whirlpak plastic bags after extraction. About one third of the samples showed signs of leakage and 4% of the bags were completely empty.

Analyses of tritium and  $^{129}\text{I}$  were individually carried out on melted 10 cm ice-core samples. From each sample, 10 mL water was used for tritium analysis and the remainder used for iodine analysis involving filtration and subsequent chemical analysis of the filtrate.

Analyses of  $^{137}\text{Cs}$  were performed on 50 cm samples obtained by pooling five consecutive 10 cm samples comprising water and chemicals remaining after iodine analyses and plastic bags. The samples were evaporated and the plastic bags compacted prior to gamma analysis. Pooling was carried out in order to limit the number of samples for analysis of  $^{137}\text{Cs}$  and to increase the amount of radioactivity in the samples analysed.

Analyses of  $^{90}\text{Sr}$  were performed on a subset of the samples from the  $^{137}\text{Cs}$  analyses covering the evaporated samples and ashed plastic bags.

## Radionuclide Analyses

### Tritium ( $^3\text{H}$ )

Aliquots of 10 mL from each water sample (Figure 5) were analysed using scintillation counting (liquid scintillation counter, LSC: Tri-Carb 3170 TR/SL and Quantulus, PerkinElmer) by direct measurements. Blank samples were prepared using tritium dead water. The standards were prepared with 10 mL of NIST-SRM-4361 Hydrogen-3 standard. The samples, standards and blanks were transferred to a 20 mL plastic vial, and 10 mL Ultima Gold LLT scintillation cocktail (PerkinElmer) was added to each sample. The samples were measured using LSC for 50 min in three cycles. The detection limit for tritium as described above is about 4 Bq/L. Standard uncertainties range from 11% (highest measured activities) to 34% (close to detection limit).



**Figure 5. Incoming inspection and sorting of samples in Whirlpak bags.**

### Iodine-129 ( $^{129}\text{I}$ )

Two  $^{129}\text{I}$  standard solutions with a total iodine ( $^{127}\text{I}$ ) concentration of 0.10 mg/mL and  $^{129}\text{I}/^{127}\text{I}$  atomic ratios of  $1.07 \times 10^{-10}$  and  $9.97 \times 10^{-12}$  respectively were prepared by diluting a  $^{129}\text{I}$  standard solution (NIST-SRM-4949c) with  $^{127}\text{I}$  carrier solution.  $^{129}\text{I}$  standards were prepared as AgI precipitate by



addition of  $\text{AgNO}_3$  and centrifugation for calibration of the AMS (accelerator mass spectrometer) in measurements of  $^{129}\text{I}/^{127}\text{I}$  ratios. Procedure blanks were prepared using the same procedure as for samples (one blank for every seven samples).

The melted ice core samples were first filtered through  $0.45\ \mu\text{m}$  membrane filters, and 300-400 mL of filtrate of each sample was weighed to a glass beaker, 1.0 mg  $^{127}\text{I}$  carrier and  $\text{K}_2\text{S}_2\text{O}_8$  (to a final concentration of 5 g/L) were added and stirred until the  $\text{K}_2\text{S}_2\text{O}_8$  was dissolved, and 3 M  $\text{HNO}_3$  was added to adjust  $\text{pH} < 2$ . The prepared samples were heated on a hot plate at  $60^\circ\text{C}$  for 20 h with a glass watch cover to decompose organic iodine to inorganic iodate. The cooled sample was transferred to a separation funnel,  $\text{CHCl}_3$  and 1 mL of 0.5 M  $\text{NH}_2\text{OH}\cdot\text{HCl}$  solution were added to convert iodate to  $\text{I}_2$  and extract it to organic phase. After the organic phase was separated, a new  $\text{CHCl}_3$  was added to the aqueous phase and the extraction procedure was repeated. 10 mL  $\text{CHCl}_3$  and 0.1 mL of 1 M  $\text{NaNO}_2$  were added to the aqueous phase to convert any iodide to  $\text{I}_2$  and extract it to the organic phase. The aqueous phase was used for determination of the other radionuclides (i.e.  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ). All  $\text{CHCl}_3$  phases were combined and transferred to a new separation funnel. 0.2 mL of 0.05 M  $\text{NaHSO}_3$  solution and 15 mL of deionized water were added to reduce iodine to iodide and back extract it to the aqueous phase. The organic phase was discarded, and 10 mL new  $\text{CHCl}_3$  was added to the aqueous phase, 0.5 mL of 1 M  $\text{HNO}_3$  and 0.1 mL of 1 M  $\text{NaNO}_2$  was added to oxidize iodide to  $\text{I}_2$  and extract it to  $\text{CHCl}_3$  phase, repeat extraction with another 10 mL  $\text{CHCl}_3$ . The  $\text{CHCl}_3$  phase was combined and transferred to a separation funnel, 0.2 mL of 0.05 M  $\text{NaHSO}_3$  and 3 mL water were added to back extract iodine to the aqueous phase. Finally, the water phase containing iodide was transferred to a centrifuge tube, 1 mL of 3 M  $\text{HNO}_3$  was added to adjust the  $\text{pH} < 2$ , and 0.5 mL of 1 M  $\text{AgNO}_3$  was added to precipitate iodide as  $\text{AgI}$ . The precipitate was washed with deionized water until neutral.

The prepared  $\text{AgI}$  precipitates (including samples, procedure blanks and standards) were dried at  $60^\circ\text{C}$  for 2-3 hours, and then ground to fine powder, and mixed with niobium powder (325 mesh) in a mass ratio of 1:5. The mixture was pressed into a copper holder for measurement of  $^{129}\text{I}/^{127}\text{I}$  atomic ratios using a 3 MV Tandem AMS system (HVEE) at Xi'an AMS Center, China.  $^{127}\text{I}^{5+}$  ions were measured as charges (current) using a Faraday cup, and  $^{129}\text{I}^{5+}$  was measured using a gas ionization detector. All samples were measured for six cycles and 5 min per sample in each cycle. The details of AMS system and measurement method for  $^{129}\text{I}$  have been reported elsewhere. The measured  $^{129}\text{I}/^{127}\text{I}$  ratios in procedure blanks were below  $2 \times 10^{-13}$ , which is at least 1 order of magnitude lower than that in the samples. The  $^{129}\text{I}/^{127}\text{I}$  ratios in the samples were corrected by subtracting the blank values.

### Caesium-137 ( $^{137}\text{Cs}$ )

Processed water samples were prepared for  $^{137}\text{Cs}$  measurement. The water phases after iodine extraction of five samples were combined and evaporated to and transferred to beakers (Figure 6) (with average volumes of around 20 mL) for gamma measurement. The samples consisted of  $\text{K}_2\text{SO}_4$  solution with some precipitated crystals covering the bottom of the beaker. The plastic bags were compacted using a 5 tonne press into 200 mL plastic beakers. Only the bags corresponding to the five most active processed water samples were measured. Due to low activities samples were measured for 3-5 days each.

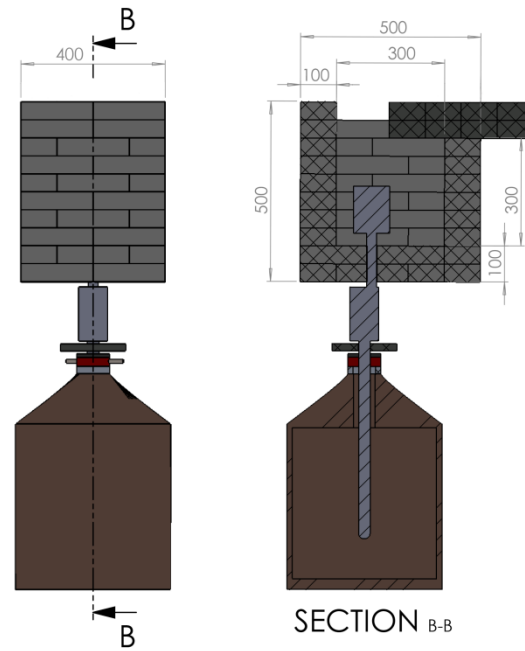


**Figure 6. Processed water sample in a 150 mL beaker.**

The Radioecology Section of DTU Nutech operates eighteen high-purity germanium (HPGe) detectors with relative efficiencies ranging from 15% to 50%. Three Canberra BEGe 5030 type detectors procured in 2015, named detector 8 (DET08), 9 (DET09) and 10 (DET10), were used in this study. Detectors are ~50% relative efficiency (germanium crystal diameter 81 mm, thickness 31 mm) in an ultra-low background dipstick cryostat (Model 7500SL-RDC-6-ULB) with remote preamplifier. To reduce the ambient gamma background the detectors are placed in 10 cm thick lead shields (Figure 7), manufactured from lead with low  $^{210}\text{Pb}$  content.

Preamplifier pulses are processed through standard analogue chain (NIM modules: amplifier, ADC and AIM unit) and spectra are acquired using Genie 2000 software (Canberra Industries, 2013). Spectra were saved with 8k ADC resolution, covering a 10-2700 keV energy range. For peak location and area calculation, Canberra Genie 2000 software was used via the interactive peak fit package (Canberra, 2009). Factory calibration with the LabSOCS software (Canberra/Mirion Technologies, 2013) was used for the activity calculation. Quality assurance of the analytical results is achieved by monthly checking of the important parameters in the spectrometric systems and through regular participation in international intercalibration and intercomparison programs, which demonstrate that

our analytical accuracy is better than 5%, e.g. (National Physics Laboratory, 2018). For quality control, sources of evaporated  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  embedded in epoxy glue in glass LSC vials are measured monthly to check efficiency and energy calibration of the detectors. Short background checks are performed regularly, while long background measurements are taken once or twice per year (Nikola Marković et al., 2018). The detection limit for analysis of  $^{137}\text{Cs}$  in a sample by gamma spectrometry is about 0.05 Bq.

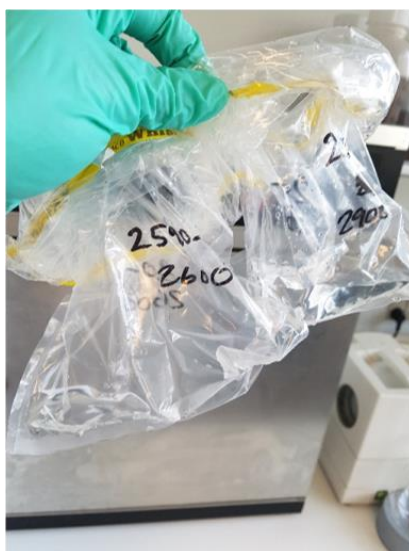


**Figure 7. BEGe detector inside a lead shield (dimensions are in mm). Shield opens by sliding the lid; section B-B shows the shield in the open position.**

### Strontium-90 ( $^{90}\text{Sr}$ )

Selected ice core samples which were analysed for  $^{137}\text{Cs}$  (from depths 3160 to 3400 cm) were used for  $^{90}\text{Sr}$  analysis. The plastic bags (Figure 8) used for sample collection were ashed at  $500^\circ\text{C}$  for 48 hours. The ash was dissolved with 14 M nitric acid and combined with the water samples.  $^{85}\text{Sr}$  was added to the samples in order to determine the chemical recovery for  $^{90}\text{Sr}$ . Moreover, a strontium carrier and calcium chloride were added to the water samples. In the following, strontium was pre-concentrated as insoluble carbonate. The precipitation was dissolved in 4 M nitric acid and a co-precipitation with ferric hydroxide was applied to remove yttrium (Y) and calcium. Afterwards, the dissolved strontium in the supernatant was precipitated as strontium carbonate (separation from potassium), dissolved with concentrated nitric acid and adjusted to a concentration of 8 M  $\text{HNO}_3$  in order to prepare for Sr-column

separation. Yttrium carrier was added to the sample solution.  $^{90}\text{Sr}$  was separated from other interfering radionuclides using an Eichrom pre-packed 2 mL cartridge, 50-100  $\mu\text{m}$  resin. After column separation, the Sr chemical yield was determined using gamma measurement of the  $^{89}\text{Sr}$  tracer. A defined amount of yttrium carrier was added to the sample solution and the samples remained untouched for three weeks to wait for the ingrowth of Y-90. Afterwards, yttrium was separated from strontium using oxalate precipitation (Chen et al., 2002). The oxalate precipitation was filtrated and prepared for the measurement on a gas flow proportional counter (Beta Multicounter System Risø GM-25-5). The detection limit for  $^{90}\text{Sr}$  in a sample is about 0.01 Bq. Additional background measurements were carried out after Y-90 has been completely decayed. The filter samples were ashed and then dissolved with nitric acid. Recovery of the yttrium was determined in the nitric acid solution using ICP-OES.

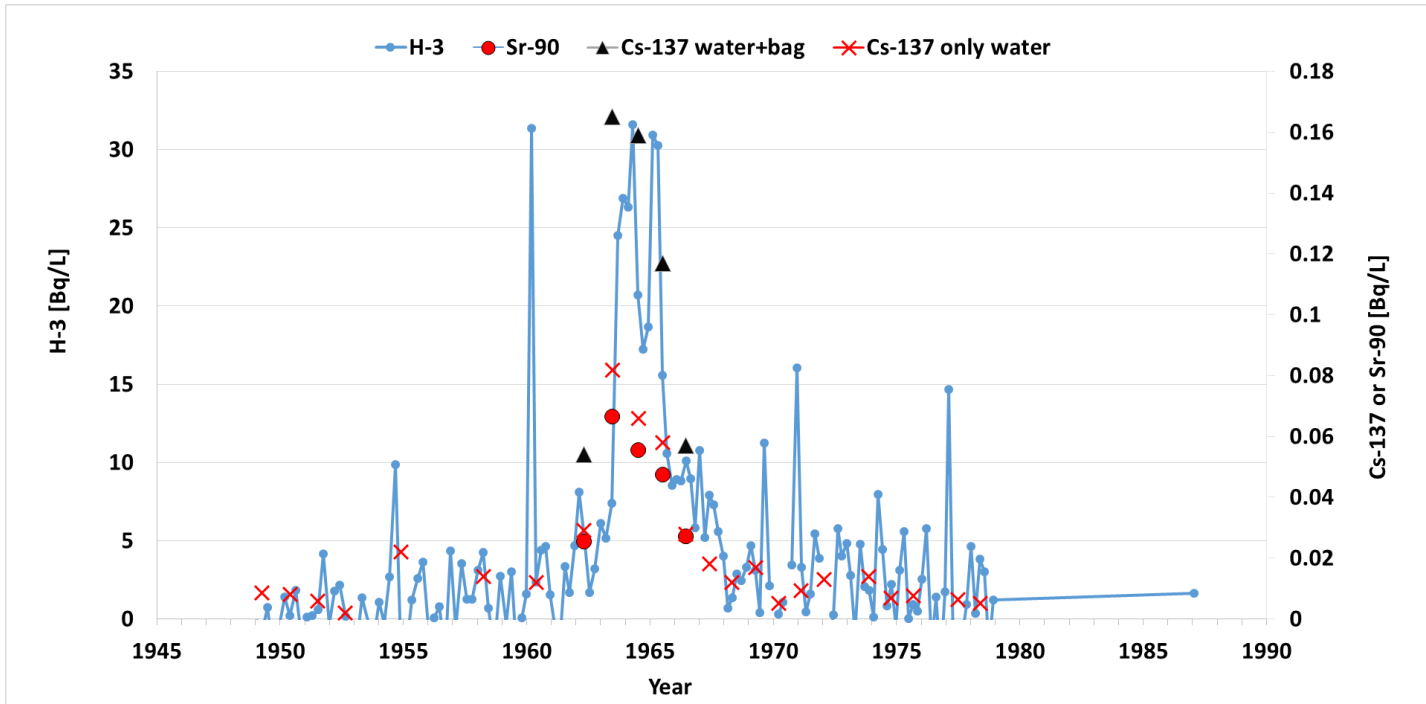


**Figure 8. Plastic bag with water sample.**

## Results and Discussion

The results for  $^3\text{H}$  are summarized in Table 1. Graphic illustrations of the depth profile for  $^3\text{H}$ , as well as for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , are shown in Figure 9. In order to compare their inter-correlation of their depth profiles, decay corrections were performed for all radionuclides to the time of sampling of the ice core. Notably high  $^3\text{H}$  concentrations (17-32 Bq/L) are observed at depths of 3240-3320 cm, which correspond to the time period of  $1963.7 \pm 1.0$  to  $1965.3 \pm 0.9$ . This corresponds to the most intense peak in atmospheric nuclear weapons testing by the USA and the Soviet Union (c. 1962-1964). During this time, tritium concentrations in Greenland precipitation reached levels of up to 600-1200 Bq/L (IAEA, 1969). This corresponds to present-day levels of 30-60 Bq/L when corrected for decay between

atmospheric deposition and ice-core sampling. The tritium concentrations from the ice core correspond reasonably well with the concentrations in precipitation observed in Greenland in the 1960's from the nuclear weapons fallout, and show no indications of contributions from the operation of the nuclear reactor.



**Figure 9. Graphic illustration of  $^3\text{H}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  depth profiles in the ice core.**

**Table 1. Analytical results for  $^3\text{H}$  activity concentrations in the ice core.**

Ice core depth [cm]	H-3 activity [Bq/L]	Ice core depth [cm]	H-3 activity [Bq/L]	Ice core depth [cm]	H-3 activity [Bq/L]
2000	< DL	3030	< DL	3540	< DL
2500	< DL	3040	4.71	3550	< DL
2510	< DL	3050	< DL	3560	< DL
2520	< DL	3060	< DL	3570	< DL
2530	< DL	3070	< DL	3580	4.24
2540	< DL	3080	< DL	3590	< DL
2550	4.62	3090	< DL	3600	< DL
2560	< DL	3100	4.04	3610	< DL
2570	< DL	3110	5.60	3620	< DL
2580	< DL	3120	7.31	3630	< DL
2590	< DL	3130	7.94	3640	4.35
2600	14.68	3140	5.22	3650	< DL
2610	< DL	3150	10.79	3660	< DL
2620	< DL	3160	5.83	3670	< DL
2630	< DL	3170	8.98	3680	< DL
2640	< DL	3180	10.11	3690	< DL
2650	5.79	3190	8.82	3700	< DL
2660	< DL	3200	8.91	3710	< DL
2670	< DL	3210	8.53	3720	< DL
2680	< DL	3220	10.59	3730	< DL
2690	< DL	3230	15.59	3740	9.88
2700	5.61	3240	30.27	3750	< DL
2710	< DL	3250	30.95	3760	< DL
2720	< DL	3260	18.65	3770	< DL
2730	< DL	3270	17.24	3780	< DL
2740	< DL	3280	20.73	3790	< DL
2750	4.45	3290	31.58	3800	< DL
2760	7.99	3300	26.33	3810	< DL
2770	< DL	3310	26.89	3820	< DL
2780	< DL	3320	24.50	3830	< DL
2790	< DL	3330	7.38	3840	< DL
2800	4.80	3340	5.18	3850	< DL
2810	< DL	3350	6.12	3860	< DL
2820	< DL	3360	< DL	3870	4.18
2830	4.82	3370	< DL	3880	< DL
2840	4.00	3380	5.18	3890	< DL
2850	5.79	3390	8.12	3900	< DL
2860	< DL	3400	4.68	3910	< DL
2870	< DL	3410	< DL	3920	< DL
2890	< DL	3420	< DL	3930	< DL
2900	5.44	3430	< DL	3940	< DL
2910	< DL	3440	< DL	3950	< DL
2920	< DL	3450	< DL	3960	< DL
2930	< DL	3460	4.65	3970	< DL
2940	16.06	3470	4.41	3980	< DL
2950	< DL	3480	< DL	3990	< DL
2970	< DL	3490	31.35	4000	< DL
2980	< DL	3500	< DL	4020	< DL
3000	< DL	3510	< DL	4050	< DL
3010	11.24	3520	< DL		
3020	< DL	3530	< DL		

Tables 2 and 3 present  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  activity concentrations in selected sections of the ice core.  $^{137}\text{Cs}$  activity concentrations are calculated by dividing the measured activity with the volume of water present inside the bag, for both processed water and plastic bag samples. As there was some leakage from the plastic bags during transport the real activity concentration would be lower, assuming some radioactivity (e.g.  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ) was deposited in the bag prior to subsequent leaking. The results are reported as the best estimates (ISO, 2010), which introduces a small positive bias for low activity samples where the measurement uncertainty is comparable with the calculated activity, as there are no negative activity values (IAEA, 2017).

**Table 2. Analytical results for  $^{137}\text{Cs}$  activity concentrations in selected sections of the ice core.**

Depth (cm)		Water (Bq/L)		Plastic bags (Bq/L)		Total (Bq/L)	
start	end	Act.	Unc.	Act.	Unc.	Act.	Unc.
2510	2550	5.10E-03	2.3E-03			5.10E-03	2.3E-03
2560	2600	6.40E-03	2.2E-03			6.40E-03	2.2E-03
2660	2700	7.60E-03	2.4E-03			7.60E-03	2.4E-03
2710	2750	6.90E-03	2.1E-03			6.90E-03	2.1E-03
2760	2800	1.40E-02	2.3E-03			1.40E-02	2.3E-03
2860	2900	1.30E-02	2.9E-03			1.30E-02	2.9E-03
2910	2950	9.40E-03	2.6E-03			9.40E-03	2.6E-03
2960	3000	5.20E-03	2.6E-03			5.20E-03	2.6E-03
3010	3050	1.70E-02	3.1E-03			1.70E-02	3.1E-03
3060	3100	1.20E-02	2.5E-03			1.20E-02	2.5E-03
3110	3150	1.80E-02	2.5E-03			1.80E-02	2.5E-03
3160	3200	2.80E-02	3.2E-03	2.90E-02	3.4E-03	5.70E-02	4.7E-03
3210	3250	5.80E-02	4.0E-03	5.95E-02	5.6E-03	1.17E-01	6.9E-03
3260	3300	6.60E-02	4.5E-03	9.26E-02	7.8E-03	1.59E-01	9.0E-03
3310	3350	8.20E-02	5.1E-03	8.29E-02	6.2E-03	1.65E-01	8.0E-03
3360	3400	2.90E-02	2.7E-03	2.51E-02	3.2E-03	5.41E-02	4.2E-03
3460	3500	1.20E-02	3.3E-03			1.20E-02	3.3E-03
3560	3600	1.40E-02	2.4E-03			1.40E-02	2.4E-03
3710	3750	2.20E-02	2.2E-03			2.20E-02	2.2E-03
3810	3850	2.00E-03	1.4E-03			2.00E-03	1.4E-03
3860	3900	6.00E-03	1.7E-03			6.00E-03	1.7E-03
3900	3950	8.10E-03	2.4E-03			8.10E-03	2.4E-03
3960	4000	8.50E-03	2.7E-03			8.50E-03	2.7E-03

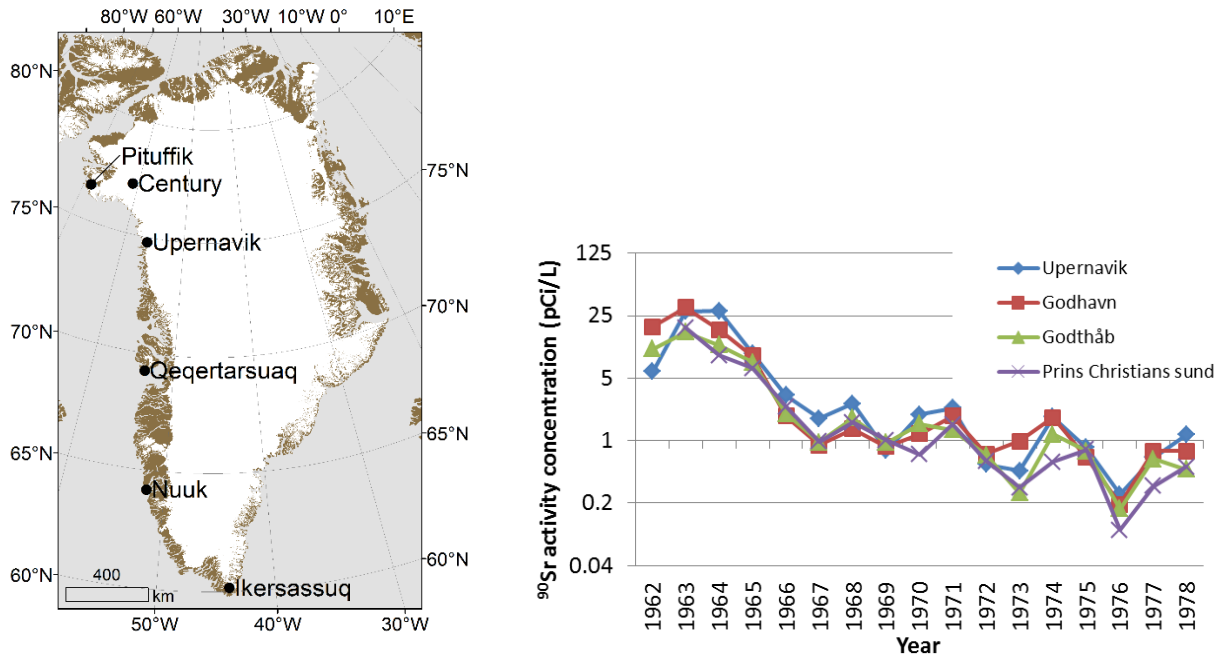
**Table 3. Analytical results for  $^{90}\text{Sr}$  activity concentrations in selected sections of the ice core.**

Depth (cm)		Water (Bq/L)	
start	end	Concentration	Standard uncertainty
3160	3200	2.70E-02	2.9E-03
3210	3250	4.74E-02	4.9E-03
3260	3300	5.56E-02	5.7E-03
3310	3350	6.65E-02	6.9E-03
3360	3400	2.55E-02	2.7E-03

During 1961-1964, the levels of  $^{137}\text{Cs}$  in precipitation in Denmark were in the range 0.1-1 Bq/L, while the  $^{137}\text{Cs}$  in Greenland during that period is not available. The levels of  $^{90}\text{Sr}$  in precipitation in Denmark showed levels similar to those for  $^{137}\text{Cs}$ . Risø reports on environmental Radioactivity in Greenland 1962-1978 contain  $^{90}\text{Sr}$  activity concentrations in precipitation from different sampling locations (Aarkrog et al., 1963-1979). Data from Upernavik, Qeqertarsuaq (Godhavn), Nuuk (Godthåb) and Ikersassuq (Prins Christians Sund) (see Figure 10) are selected as there is a good coverage with yearly mean activity concentrations for almost all the years in the selected time span. The HASL-281 report data was also used for the comparison (Health and Safety Laboratory, 1974). Health and Safety Laboratory, fallout program quarterly summary report from 1974 has Pituffik (Thule)  $^{90}\text{Sr}$  activity concentrations in precipitation data for years 1959-1973. Some data missing, not all years fully covered but the data is in agreement with data from the Risø reports. Data from Risø reports have been converted to Bq/L units and decay-corrected to the day of Camp Century sampling, 25 June 2017. This approach was chosen as uncertainty in the ice core depth-age relation makes it complicated to decay correct the observed concentrations of radionuclides in the ice core samples to the 1960's.

The activity ratios  $^{137}\text{Cs}/^{90}\text{Sr}$  in fallout from atmospheric nuclear weapons testing in the 1960's had values about 1.5 (UNSCEAR, 2000), a value of 1.6 reported for precipitation at Pituffik (HASL station at Thule, Aarkrog, 1979) and a value of 1.4 measured in sea water around Greenland (Aarkrog et al., 1980). This activity ratio changes only slowly with time – due to the similar half-lives of the two radionuclides – and concentrations of  $^{137}\text{Cs}$  in fallout may thus be estimated from concentrations of  $^{90}\text{Sr}$  by applying this ratio. Figure 11 shows a comparison of  $^{137}\text{Cs}$  concentrations estimated from historic  $^{90}\text{Sr}$  data and  $^{137}\text{Cs}$  concentrations measured in the ice core samples, all data decay corrected to time of sampling of the ice core.



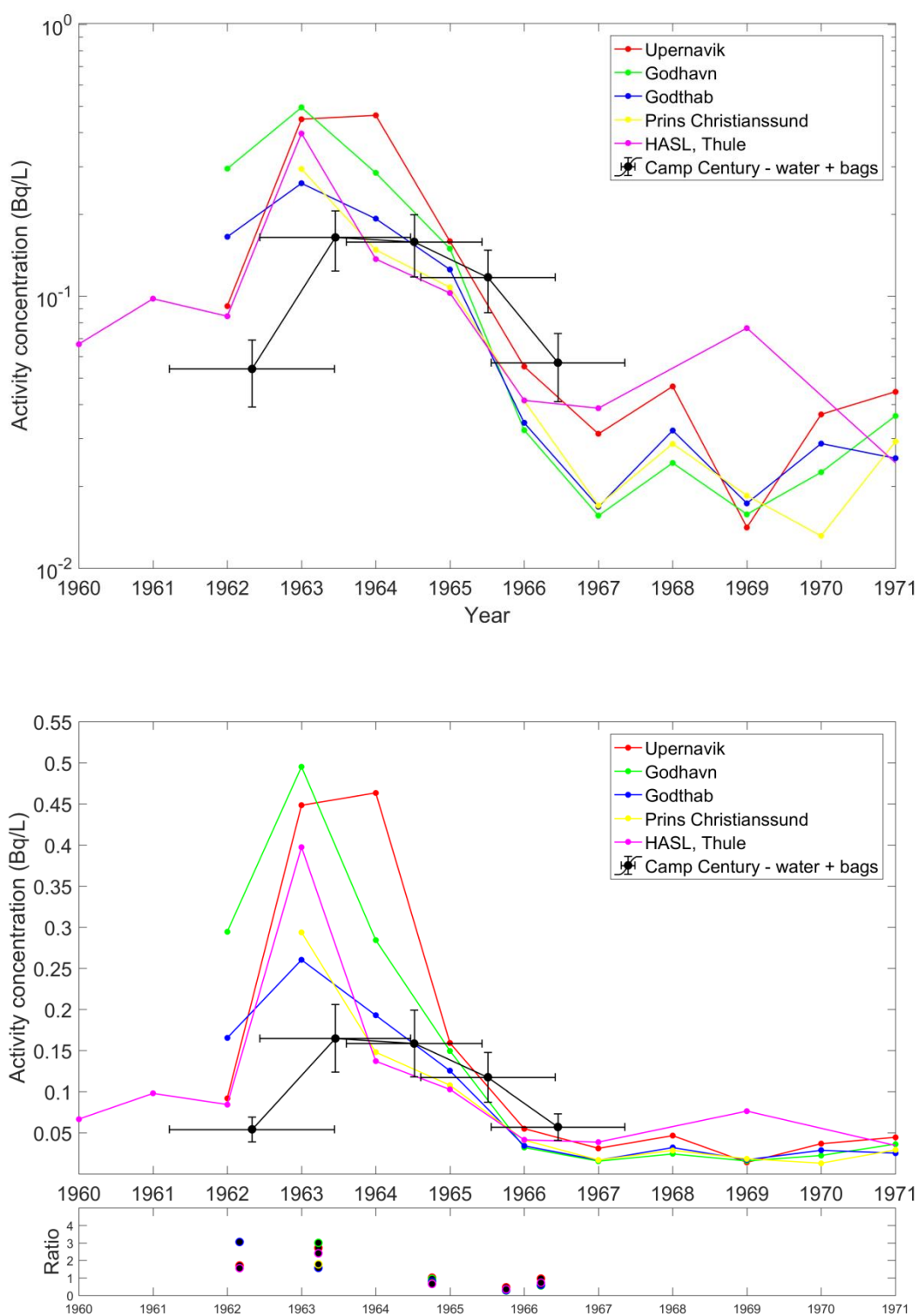


**Figure 10. Sampling locations and annual specific mean activities of <sup>90</sup>Sr in precipitation (data taken from Risø reports) on Greenland used for comparison in this work.**

At first, only activity in the processed water samples was considered. Activity concentrations were lower by at least factor of four when compared with the historical <sup>90</sup>Sr data under assumption of <sup>137</sup>Cs/<sup>90</sup>Sr ratio of 1.5. The activities in the plastic bags were subsequently measured. The activities deposited in the bags were found to be of the same order as the activity in the processed water samples (Table 2). Combined values are presented in Figure 11. Activity concentrations in the peak years (1962-1965) are still lower than expected from historical fallout levels.

From Figure 11, we see no excess of <sup>137</sup>Cs activity during the period of Camp Century reactor operation. However, it is still possible that the measured activities are underestimated as we still have activity concentrations at least two times lower than expected. A possible reason for underestimation of <sup>137</sup>Cs levels include the absorption of <sup>137</sup>Cs onto particles which were filtered out and discarded during the <sup>129</sup>I analysis.

For activity concentration calculations, the activities on the bags were divided by the water volumes in the bags after the leakage (not original volumes) so we even overestimated the activity concentration (probably it was deposited on the surface of the bags even from the part of volume that leaked). Because the water samples were filtered through 0.45 μm membrane filters in the iodine analysis step, all <sup>137</sup>Cs associated with particles bigger than 0.45 μm was lost during this step. Papers by (Steffensen, 1985; Unnerstad and Hansson, 2001) report that a significant fraction of non-soluble particles in ice core water is >0.5 μm. Steffensen (Steffensen, 1997) presents results on particle size distribution through the history.



**Figure 11. Comparison of  $^{90}\text{Sr}$  measurements from Risø reports and HASL-281 report (Greenland, Thule, Air Force Cambridge Research Laboratory, Geopole Station) multiplied with the factor of 1.5 and combined  $^{137}\text{Cs}$  activity concentrations measured in Camp Century water and plastic bag samples (linear and log scale).**

Activity levels of  $^{137}\text{Cs}$  reported in the Wendel et al. (Wendel et al., 2013) are in accordance with our results. Wendel's data are taken from Pinglot et al. (Pinglot et al., 2003). The other reference with  $^{137}\text{Cs}$  activities is Fritzsche et al. (Fritzsche et al., 2002). Levels are  $<100$  mBq/kg at the time of deposition (except for the Devon 98 ice core, Figure 3 in Pinglot et al., where it is around 750 mBq/kg). But this is not in agreement with  $^{90}\text{Sr}$  measurements in precipitation over Greenland (Risø reports) where the values  $>200$  mBq/kg are reported (the same is with HASL and other maximum yearly average values in precipitation) or general UNSECAR fallout reports. In Pinglot and Fritzsche there is not enough info on sample preparation so it is possible that they also lost the particle bound fraction. The concentrations of  $^{90}\text{Sr}$  in the ice core samples shown in Table 3 may be compared with historical concentrations in precipitation in Greenland (Figure 10). Here concentrations peaked in 1963 at values in the range 0.6-1.1 Bq/L which correspond to 0.16-0.31 Bq/L when corrected for decay to the time of sampling of the ice core. This, however, is higher than the concentrations from the ice core by about a factor three. The average activity ratio for  $^{137}\text{Cs}/^{90}\text{Sr}$  from the ice core (Tables 2 and 3) is 2.4, which is significant larger than the reference value of 1.5. The reason for this may be due to fractionation during preparation of the samples. The data thus shows no sign of excess  $^{90}\text{Sr}$  from the operation of the reactor at Camp Century.

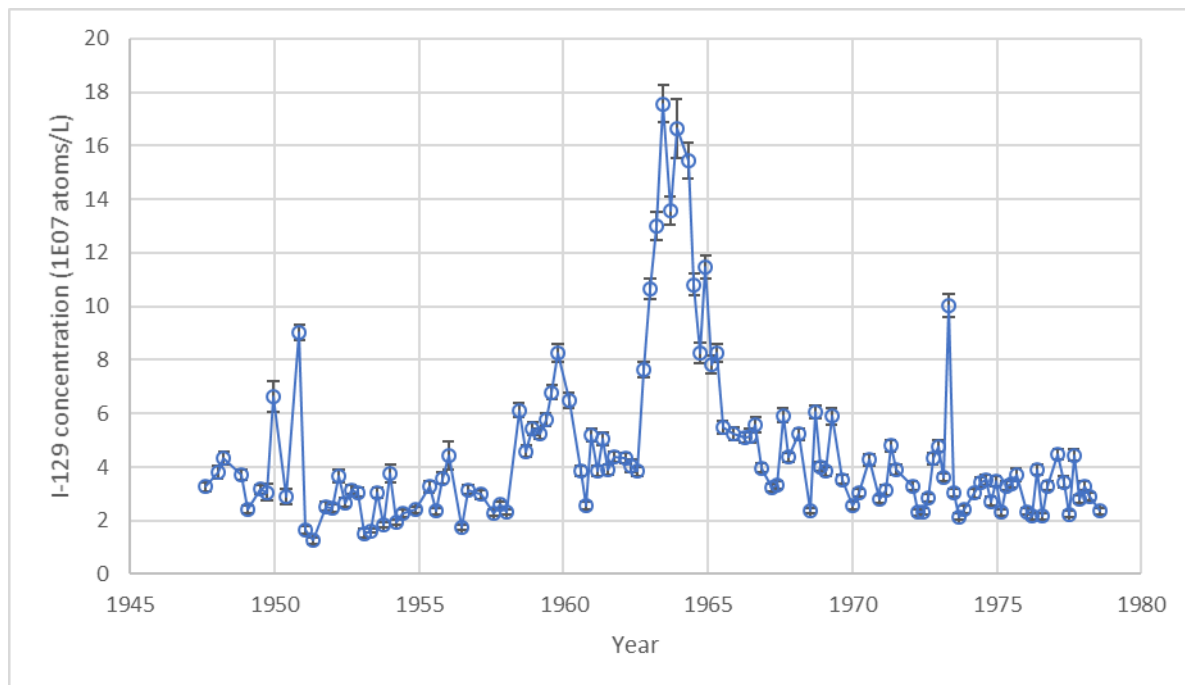
The analytical results of  $^{129}\text{I}$  in the ice core samples are presented in Table 4 and Figure 12 below. Uncertainties are standard uncertainties.

**Table 4. Analytical results of  $^{129}\text{I}$  concentrations in the ice core from Camp Century.**

Depth, cm	$^{129}\text{I}$ concentration, $10^7$ atoms/L		Depth, cm	$^{129}\text{I}$ concentration, $10^7$ atoms/L	
	Value	Uncertainty		Value	Uncertainty
2510	2.35	0.12	3280	15.43	0.67
2530	2.88	0.14	3300	16.63	1.11
2540	3.30	0.19	3310	13.57	0.53
2550	2.80	0.15	3320	17.56	0.69
2560	4.45	0.21	3330	13.00	0.51
2570	2.24	0.13	3340	10.67	0.38
2580	3.44	0.23	3350	7.64	0.29
2590	4.46	0.19	3360	3.85	0.17
2610	3.29	0.16	3370	4.04	0.22
2620	2.16	0.12	3380	4.34	0.19
2630	3.90	0.18	3400	4.38	0.19
2640	2.16	0.14	3410	3.90	0.19
2650	2.33	0.12	3420	5.04	0.24
2670	3.70	0.24	3430	3.85	0.18

2680	3.39	0.15	3440	5.22	0.24
2690	3.30	0.18	3450	2.55	0.15
2700	2.30	0.12	3460	3.85	0.20
2710	3.45	0.19	3480	6.47	0.29
2720	2.72	0.17	3500	8.26	0.34
2730	3.51	0.18	3510	6.79	0.28
2740	3.42	0.19	3520	5.78	0.24
2750	3.03	0.17	3530	5.24	0.21
2770	2.42	0.14	3540	5.44	0.24
2780	2.14	0.12	3550	4.59	0.21
2790	3.02	0.14	3560	6.12	0.25
2800	10.02	0.42	3580	2.33	0.11
2810	3.63	0.15	3590	2.60	0.12
2820	4.78	0.20	3600	2.29	0.13
2830	4.32	0.21	3620	2.98	0.16
2840	2.87	0.15	3640	3.15	0.16
2850	2.34	0.13	3650	1.75	0.10
2860	2.31	0.12	3670	4.44	0.53
2870	3.28	0.17	3680	3.58	0.22
2900	3.90	0.18	3690	2.35	0.13
2910	4.80	0.20	3700	3.27	0.21
2920	3.13	0.18	3720	2.39	0.12
2930	2.80	0.16	3740	2.28	0.12
2950	4.26	0.20	3750	1.93	0.10
2970	3.05	0.14	3760	3.77	0.34
2980	2.56	0.14	3770	1.83	0.10
3000	3.54	0.17	3780	3.03	0.19
3020	5.90	0.31	3790	1.63	0.09
3030	3.84	0.16	3800	1.51	0.17
3040	4.01	0.16	3810	3.06	0.15
3050	6.06	0.24	3820	3.16	0.15
3060	2.37	0.11	3830	2.68	0.15
3080	5.22	0.20	3840	3.65	0.24
3100	4.37	0.17	3850	2.44	0.14
3110	5.93	0.25	3860	2.53	0.15
3120	3.34	0.14	3880	1.25	0.11
3130	3.21	0.15	3890	1.65	0.13
3150	3.97	0.16	3900	9.01	0.91
3160	5.57	0.27	3920	2.89	0.28
3170	5.16	0.25	3940	6.64	0.58
3180	5.10	0.21	3950	3.05	0.31
3200	5.25	0.22	3960	3.16	0.18
3220	5.46	0.24	3980	2.42	0.15
3230	8.24	0.33	3990	3.70	0.19
3240	7.85	0.33	4020	4.34	0.23
3250	11.47	0.45	4030	3.81	0.25

3260	8.25	0.39	4050	3.26	0.16
3270	10.82	0.40			



**Figure 12. Concentrations of  $^{129}\text{I}$  in the ice core ( $10^7$  atoms/L) shown as function of time.**

No historical data on  $^{129}\text{I}$  in precipitation in Greenland is available for comparison with the observations from the ice core. It is noted, however, that the ice core data show peak concentrations at depths corresponding to the years 1962-1964 when fallout from the atmospheric nuclear weapons were most intense and that this is in agreement with the findings for tritium,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ .

## Conclusions and perspectives

It is concluded that there is no indication that significant radioactive contamination was released into the air as a result of the Camp Century reactor operation. It might be possible that the measured activities for  $^{90}\text{Sr}$ ,  $^{129}\text{I}$  and  $^{137}\text{Cs}$  are underestimated as we find activity concentrations for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  lower than historical concentrations corrected for decay to the time of sampling of the ice core. Further investigation will be carried out to evaluate the absorption of  $^{137}\text{Cs}$  activity onto particles by careful experimental design and low-level gamma measurement for the upper layer (above 2000 cm) ice core samples.

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