



Radioactivity in the Risø District July-December 2019

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Radioactivity in the Risø District July-December 2019



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Report DTU-ENV-RAS-0001

June 2020

By

Jixin Qiao, Kasper G. Andersson and Arne Miller

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Preface

A specific monitoring programme in the vicinity of the nuclear installations at the Risø site is carried out by DTU Environment on behalf of and as a contractor to Danish Decommissioning (DD). This report presents the analytical results of the monitoring and sampling carried out in the period July-December 2019. The materials and methods used in connection with the monitoring programme are described in pages 27-28.

Risø, May 2020

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Summary

The environmental surveillance of the Risø environment was continued in July-December 2019. The mean concentrations in air were: $0.17 \pm 0.10 \mu\text{Bq m}^{-3}$ of ^{137}Cs , $2.45 \pm 0.94 \text{ mBq m}^{-3}$ of ^7Be and $0.26 \pm 0.16 \text{ mBq m}^{-3}$ of ^{210}Pb (± 1 standard uncertainty). The depositions by precipitation at Risø in the second half of 2019 were: $0.032 \pm 0.005 \text{ Bq m}^{-2}$ of ^{137}Cs , $596 \pm 60 \text{ Bq m}^{-2}$ of ^7Be , $42.0 \pm 4.2 \text{ Bq m}^{-2}$ of ^{210}Pb and $<1.3 \text{ kBq m}^{-2}$ of ^3H . The average background dose rate (TLD) at Risø (Zone I) was measured as $58 \pm 9 \text{ nSv h}^{-1}$ compared with $56 \pm 5 \text{ nSv h}^{-1}$ (± 1 standard uncertainty) in the four zones around Risø.

Table 1. Radionuclides in ground level air collected at Risø (cf. Figs. 1, 1.1 and 1.2), July-December 2019 (Unit: $\mu\text{Bq m}^{-3}$)

Date	^7Be	^{137}Cs	^{210}Pb
02-Jul-19 – 08-Jul-19	2286(10%)	0.044(18%)	79(10%)
08-Jul-19 – 15-Jul-19	2823(10%)	0.043(14%)	191(10%)
15-Jul-19 – 22-Jul-19	3569(10%)	0.103(12%)	299(10%)
22-Jul-19 – 29-Jul-19	2911(10%)	0.114(11%)	329(10%)
29-Jul-19 – 05-Aug-19	4011(10%)	0.186(12%)	326(10%)
05-Aug-19 – 12-Aug-19	3098(10%)	0.055(12%)	140(10%)
12-Aug-19 – 19-Aug-19	3066(10%)	0.053(12%)	127(10%)
19-Aug-19 – 26-Aug-19	2743(10%)	0.092(12%)	156(10%)
26-Aug-19 – 02-Sep-19	4921(10%)	0.249(12%)	598(10%)
02-Sep-19 – 09-Sep-19	2100(10%)	0.081(12%)	109(10%)
09-Sep-19 – 16-Sep-19	1830(10%)	0.074(12%)	112(10%)
16-Sep-19 – 23-Sep-19	2381(10%)	0.136(12%)	126(10%)
23-Sep-19 – 30-Sep-19	3018(10%)	0.331(12%)	380(10%)
30-Sep-19 – 07-Oct-19	2772(10%)	0.171(12%)	209(10%)
07-Oct-19 – 14-Oct-19	3543(10%)	0.102(12%)	256(10%)
14-Oct-19 – 21-Oct-19	2053(10%)	0.128(12%)	266(10%)
21-Oct-19 – 28-Oct-19	2451(10%)	0.124(12%)	357(10%)
28-Oct-19 – 04-Nov-19	1734(10%)	0.179(13%)	249(10%)
04-Nov-19 – 11-Nov-19	2855(10%)	0.303(10%)	268(10%)
11-Nov-19 – 18-Nov-19	2924(10%)	0.326(12%)	658(10%)
18-Nov-19 – 25-Nov-19	1233(10%)	0.346(12%)	512(10%)
25-Nov-19 – 02-Dec-19	1105(10%)	0.285(12%)	449(10%)
02-Dec-19 – 10-Dec-19	1161(10%)	0.085(11%)	43(10%)
10-Dec-19 – 17-Dec-19	2245(10%)	0.179(12%)	132(10%)
17-Dec-19 – 23-Dec-19	2434(10%)	0.352(11%)	319(10%)
23-Dec-19 – 30-Jan-19	767(10%)	0.144(12%)	113(10%)
Mean	2540	0.165	262
SD	935	0.102	160

*Figures in brackets are relative standard uncertainties

Table 2.1. Radionuclides in precipitation in the 10 m² rain collector at Risø (cf. Fig. 8.1), July - December 2019. (Unit: Bq m⁻³)

Month	⁷ Be	¹³⁷ Cs	²¹⁰ Pb
July	2231(10%)*	0.258(15%)	134(10%)
August	2111(10%)	0.122(14%)	93(10%)
September	1344(10%)	0.072(14%)	100(10%)
October	1005(10%)	0.032(22%)	67(10%)
November	3576(10%)	0.159(16%)	373(10%)
December	2009(10%)	0.112(22%)	158(10%)

*Figures in brackets are relative standard uncertainties

Table 2.2. Radionuclides in precipitation in the 10 m² rain collector at Risø (cf. Fig. 8.1), July - December 2019. (Unit: Bq m⁻²)

Month	Precipitation (m)	⁷ Be	¹³⁷ Cs	²¹⁰ Pb
July	0.025(10%)*	56(14%)	0.0065(18%)	3.4(14%)
August	0.072(10%)	153(14%)	0.0088(17%)	6.7(14%)
September	0.077(10%)	104(14%)	0.0056(17%)	7.7(14%)
October	0.112(10%)	113(14%)	0.0035(24%)	7.5(14%)
November	0.034(10%)	120(14%)	0.0053(19%)	12.6(14%)
December	0.025(10%)	50(14%)	0.0028(25%)	4.0(14%)
Sum	0.346(5%)	596(10%)	0.0325(15%)	41.9(10%)

*Figures in brackets are relative standard uncertainties

Table 2.3. Tritium in precipitation collected at Risø (cf. Figs. 1, 2.3.1 and 2.3.2). July - December 2019. (Unit: kBq m⁻³)

Month	10 m ² rain collector*
July	<3.4
August	<3.4
September	4.6(26%) ^a
October	<3.4
November	<3.4
December	<2.7
Double determinations*.	

^a Figures in brackets are relative standard uncertainties

Table 2.4. Tritium in precipitation collected at Risø (cf. Fig. 1). July - December 2019 (Unit: kBq m⁻²)

Month	Precipitation (m)	10 m ² rain collector
July	0.025(10%) ^a	<0.085
August	0.072(10%)	<0.250
September	0.077(10%)	0.345(28%) ^a
October	0.112(10%)	<0.381
November	0.034(10%)	<0.116
December	0.025(10%)	< 0.068
Sum	0.346(5%)	< 1.245

^a Figures in brackets are relative standard uncertainties

Table 3.1. Radionuclides in sediment samples collected at Bolund in Roskilde Fjord.(cf. Fig. 3.1) July - December 2019. (Unit: Bq kg⁻¹ dry)

Date	¹³⁷ Cs	K*
23 September	0.66(16%) ^a	14.2(10%)

*Unit: g kg⁻¹ dry

^a Figures in brackets are relative standard uncertainties

Table 4.1. Radionuclides in seawater collected in Roskilde Fjord (cf. Fig. 4.1) July - December 2019. (Unit: Bq m⁻³)

Date	¹³⁷ Cs
4 September	8.8(12%) ^a

^a Figures in brackets are relative standard uncertainties

Table 4.2. Tritium in seawater collected in Roskilde Fjord (Risø pier) (cf. Fig. 4.2) July - December 2019*^a.

Month	kBq m ⁻³
September	<3.4
December	<3.4

* Double determinations

^a Figures in brackets are relative standard uncertainties

Table 5.1. Radionuclides in grass (* snow) collected at Risø near the Waste Treatment Station, location I P3, Fig. 1, July - December 2019. (**Measured on bulked ash samples)

Week no. or month	Date	K (g kg ⁻¹ fresh)	¹³⁷ Cs (Bq kg ⁻¹ fresh)	¹³⁷ Cs (Bq m ⁻²)
27	1 July	3.3(10%) ^a	<0.5	
29	15 July	6.1(10%)	<0.6	
31	29 July	0.2(10%)	<0.5	
33	12 August	7.1(10%)	<0.5	
35	26 August	6.8(10%)	<0.3	
37	9 September	5.1(10%)	<0.5	
39	23 September	5.7(10%)	<0.6	
41	07 October	4.5(10%)	<0.4	
43	21 October	2.9(10%)	<0.2	
45	4 November	2.6(10%)	<0.3	
47	18 November	5.8(10%)	<0.6	
49	2 December	4.6(10%)	<0.6	
51	17 December	4.9(10%)	<0.6	
53	30 December	7.4(10%)	<0.7	
** July		6.1(10%)	0.038(58%)	0.018(60%)
** August		7.3(10%)	<0.036	<0.008
** September		5.0(10%)	0.033(56%)	0.021(58%)
** October		3.8(10%)	<0.043	<0.066
** November		3.4(10%)	0.062(33%)	0.021(35%)
** December		5.7(10%)	0.059(17%)	0.025(20%)

^a Figures in brackets are relative standard uncertainties

Table 5.2. Radionuclides in *Fucus vesiculosus* collected at Bolund in Roskilde Fjord. July - December 2019. (Unit: Bq kg⁻¹ dry)

Date	¹³⁷ Cs	K*	% dry matter
18 July	2.0(10%) ^a	27(10%)	20(10%)

*Unit: g kg⁻¹ dry

^a Figures in brackets are relative standard uncertainties

Table 7.1. Waste water collected at Risø (cf. Fig. 1), July - December 2019.

Week Number	Total beta (eqv. mg KCl l ⁻¹)	¹³⁷ Cs (Bq m ⁻³)	¹³¹ I (Bq m ⁻³)	²²⁶ Ra (Bq m ⁻³)
27	103(10%) ^a	<114	<146	<291
28	86(11%)	<132	<131	<296
29	111(10%)	<114	<131	<275
30	158(10%)	<119	<153	<291
31	152(10%)	<130	<133	<298
32	129(10%)	<46	<150	<97
33	143(10%)	<28	<33	<57
34	109(10%)	<53	<41	<110
35	97(11%)	<59	<48	<115
36	99(11%)	<107	<77	<1470 ^b
37	109(12%)	<101	<85	<1370 ^b
38	78(10%)	<115	<115	<262
39	108(10%)	<122	<121	<278
40	89(10%)	<101	<91	<220
41	77(10%)	<83	<76	<180
42	87(10%)	<118	<123	<279
43	48(11%)	<121	<134	<291
44	53(11%)	<129	<130	<308
45	67(10%)	<126	<152	<305
46	54(10%)	<138	<140	<319
47	85(11%)	<146	<145	<341
48	61(11%)	<121	<138	<290
49	63(13%)	<52	<41	<107
50	57(11%)	<119	<132	<294
51	55(12%)	<54	<41	<105
52	60(12%)	<52	<101	<114
53	45(15%)	<29	<33	<55
Mean	88.3	<102	<105	<367
SD	32.0			

^a Figures in brackets are relative standard uncertainties

^b Enhanced level of detection limit due to technical conditions. Total beta figures are not enhanced for this period.

Table 8.1. Background dose rates around the border of Risø (cf. Fig. 8.1) measured with thermoluminescence dosimeters (TLD) in the period November 2018 – November 2019. (Results are normalized to nSv h^{-1})^b

Location	nSv h^{-1} ^a
1	44(10%) ^a
2	50(10%)
3	60(10%)
4	54(10%)
5	53(10%)
6	74(10%)
Mean	55(5%)

^a Figures in brackets in Table 8.1 and 8.2 are relative standard uncertainties

^b Note: due to an internal misunderstanding at DTU, background dose rate figures in Tables 8.1 and 8.2 represent an entire year, and not as normally reported 6 months. The results in Table 8.3 are recorded at the same time as TLD collection. All these results were thus also presented in the previous report in this series.

^c This result in Table 8.2 is unusually high. The measurement is believed to be correct. The reason for the deviation from the expected value is unknown.

Table 8.2. Background dose rates around Risø (cf. Fig. 8.2 and Fig. 1) measured with thermoluminescence dosimeters (TLD) in the period November 2018– November 2019. (Results are normalized to $nSv h^{-1}$)^b,

Risø zone	Location	$nSv h^{-1}$ ^a
I	1	47(10%)
I	2	57(10%)
I	3	73(10%)
I	4	55(10%)
I	5	58(10%)
Mean		58(5%)
II	P1	55(10%)
II	P2	60(10%)
II	P3	49(10%)
II	P4	49(10%)
Mean		53 (10%)
III	P1	56(10%)
III	P2	Dosimeter lost
III	P3	53(10%)
Mean		54(8%)
IV	P1	51(10%)
IV	P2	Dosimeter lost
IV	P3	57(10%)
IV	P4	53(10%)
IV	P5	55(10%)
IV	P6	53(10%)
IV	P7	63(10%)
Mean		55(4%)
V	P1	61(10%)
V	P2	55(10%)
V	P3	121(10%) ^c
V	P4	51(10%)
V	P5	55(10%)
V	P6	68(10%)
V	P7	49(10%)
V	P8	62(10%)
V	P9	51(10%)
V	P10	62(10%)
Mean		63(4%)

Table 8.3. Terrestrial dose rates at the Risø zones (cf. Fig. 8.2 and Fig. 1) November 2018–November 2019. Measured with a NaI(Tl) detector. (Unit: nSv h⁻¹)

Risø zone	Location	October
I	P1	42(10%) ^a
I	P2	52(10%)
I	P3	318(10%)
I	P4	40(10%)
I	P5	49(10%)
Mean		100(5%)
II	P1	45(10%)
II	P2	48(10%)
II	P3	41(10%)
II	P4	42(10%)
Mean		44(4%)
III	P1	47(10%)
III	P2	48(10%)
III	P3	42(10%)
Mean		46(6%)
IV	P1	41(10%)
IV	P2	45(10%)
IV	P3	43(10%)
IV	P4	44(10%)
IV	P5	36(10%)
IV	P6	48(10%)
IV	P7	52(10%)
Mean		44(4%)
V	P1	58(10%)
V	P2	55(10%)
V	P3	56(10%)
V	P4	50(10%)
V	P5	49(10%)
V	P6	43(10%)
V	P7	45(10%)
V	P7a	46(10%)
V	P8	45(10%)
V	P9	56(10%)
V	P10	45(10%)
Mean		50(4%)

^a Figures in brackets are relative standard uncertainties

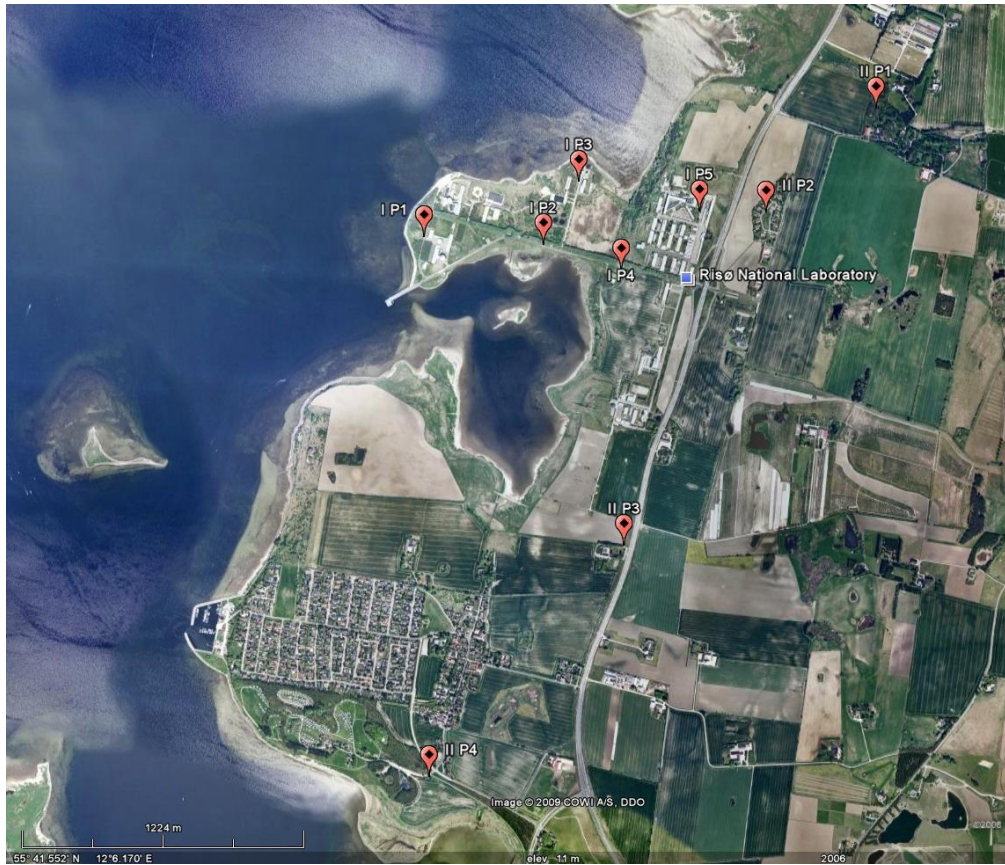


Fig. 1. Locations for measurements of gamma-background radiation Zone I and II (cf. Tables 8.2 and 8.3)

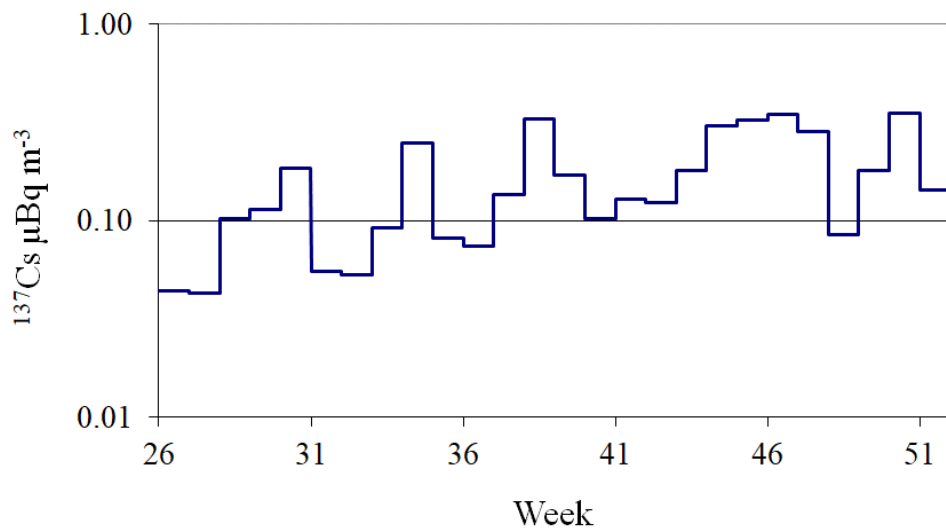


Fig. 1.1. Caesium-137 in ground level air collected at Risø in July-December 2019. (Unit: $\mu\text{Bq m}^{-3}$)

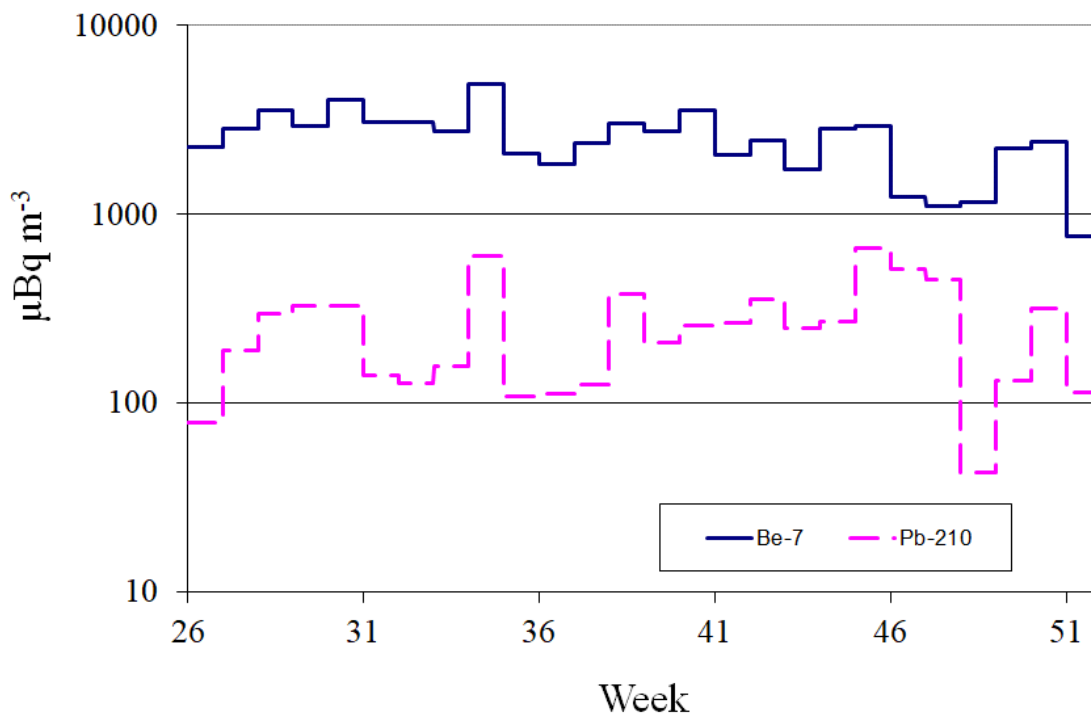


Fig. 1.2. Beryllium-7 and Lead-210 in ground level air collected at Risø in July-December 2019. (Unit: $\mu\text{Bq m}^{-3}$)

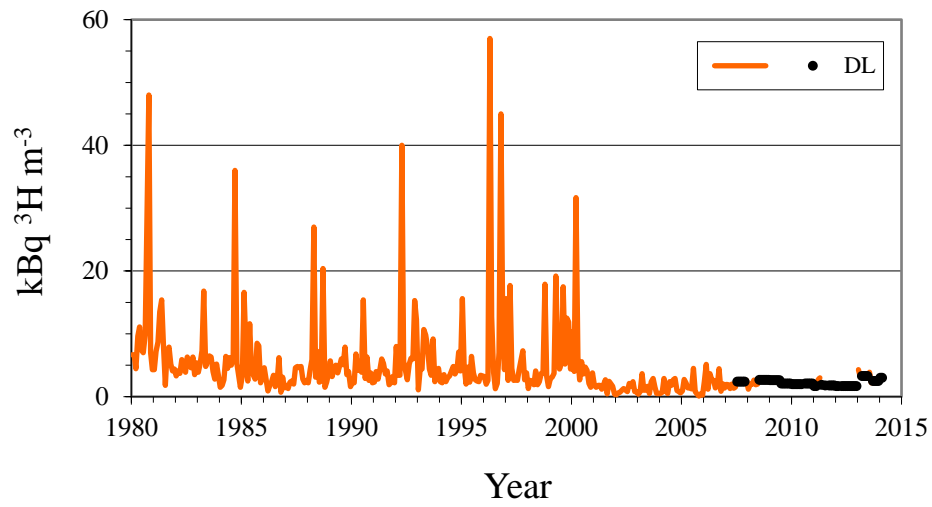


Fig. 2.3.1. Tritium in precipitation collected at Risø (1 m² rain collector) 1980 - 2013. (Unit: kBq m⁻³; DL = detection limit). This rain collector was taken out of operation in 2013.

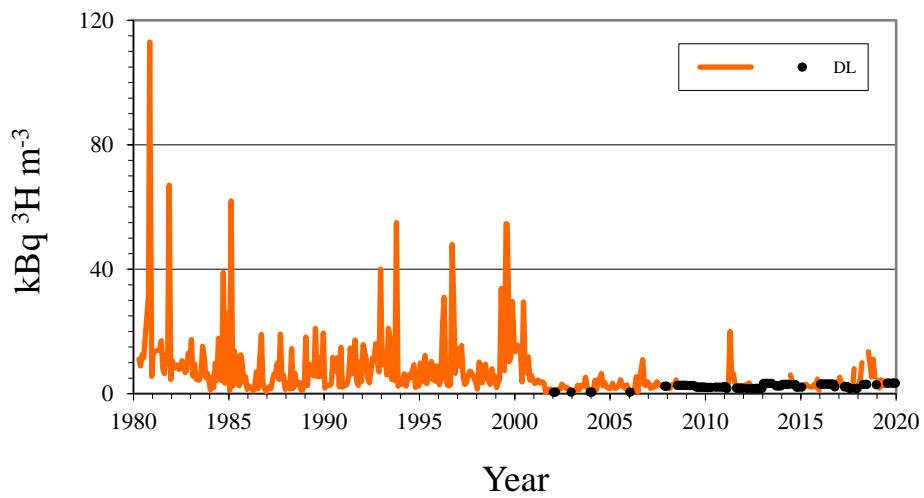


Fig. 2.3.2. Tritium in precipitation collected at Risø (10 m² rain collector) 1980 - 2019. (Unit: kBq m⁻³; DL = detection limit)

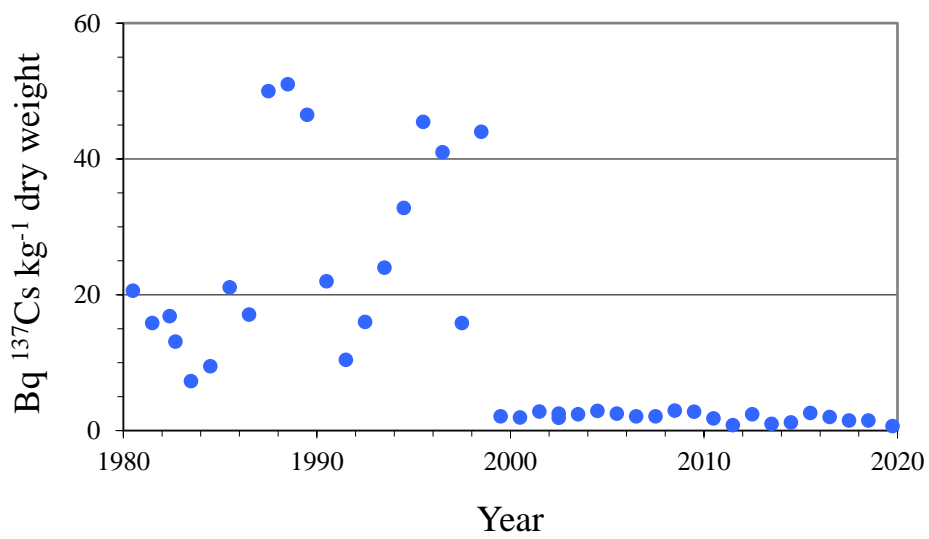


Fig. 3.1. Caesium-137 in sediment samples collected at Bolund in Roskilde Fjord. 1980 – 2019. (Unit: Bq kg⁻¹ dry matter)

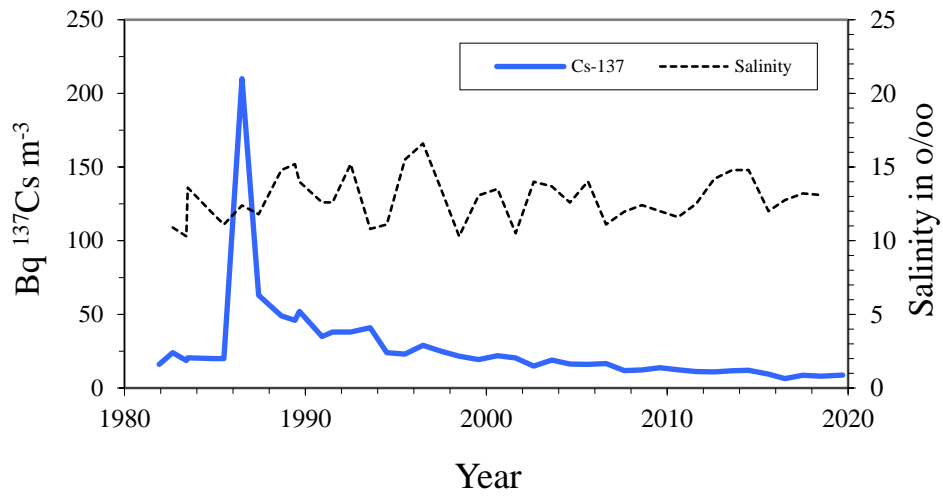


Fig. 4.1. Caesium-137 in seawater collected in Roskilde Fjord 1980 – 2019. (Unit: Bq m^{-3})

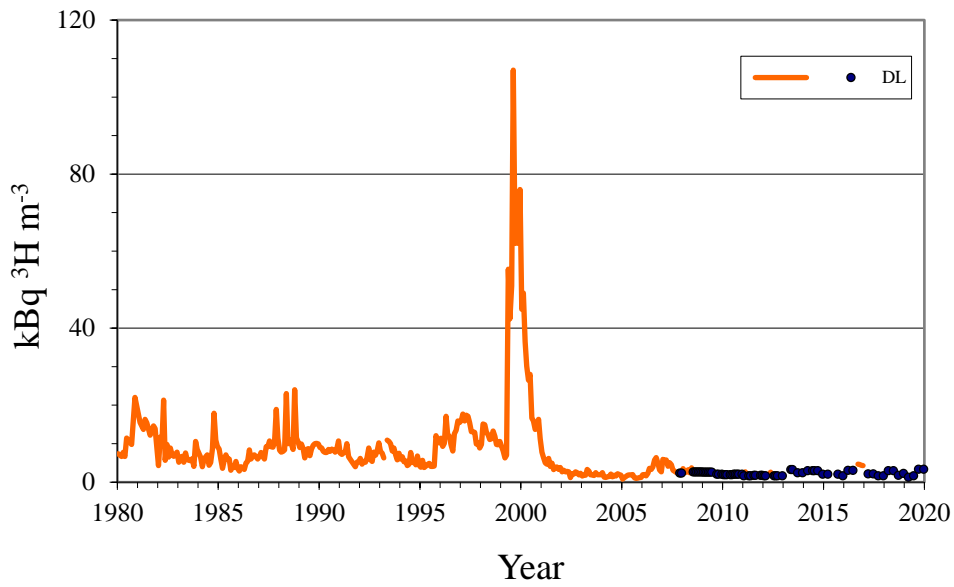


Fig. 4.2. Tritium in seawater collected in Roskilde Fjord 1980 - 2019. (Unit: kBq m^{-3} ; DL = detection limit)

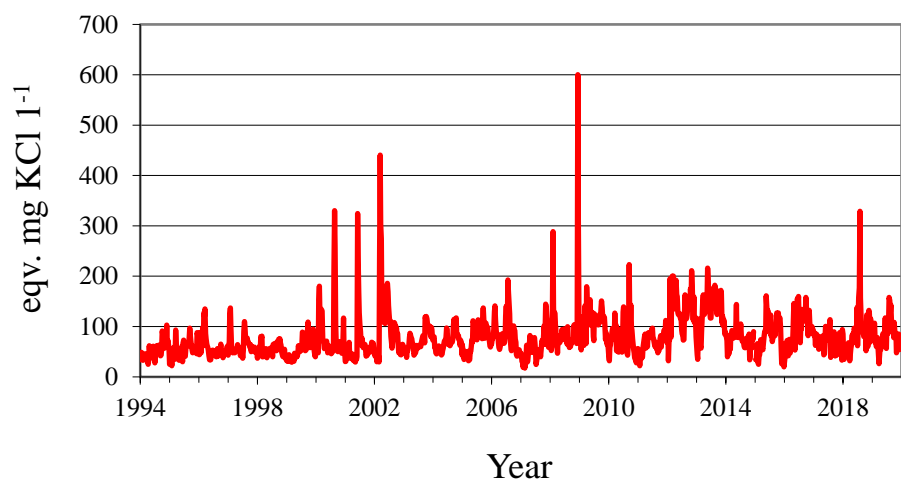


Fig. 7.1. Total-beta radioactivity in waste water collected at Risø 1994 - 2019.
(Unit: eqv. mg KCl l⁻¹)

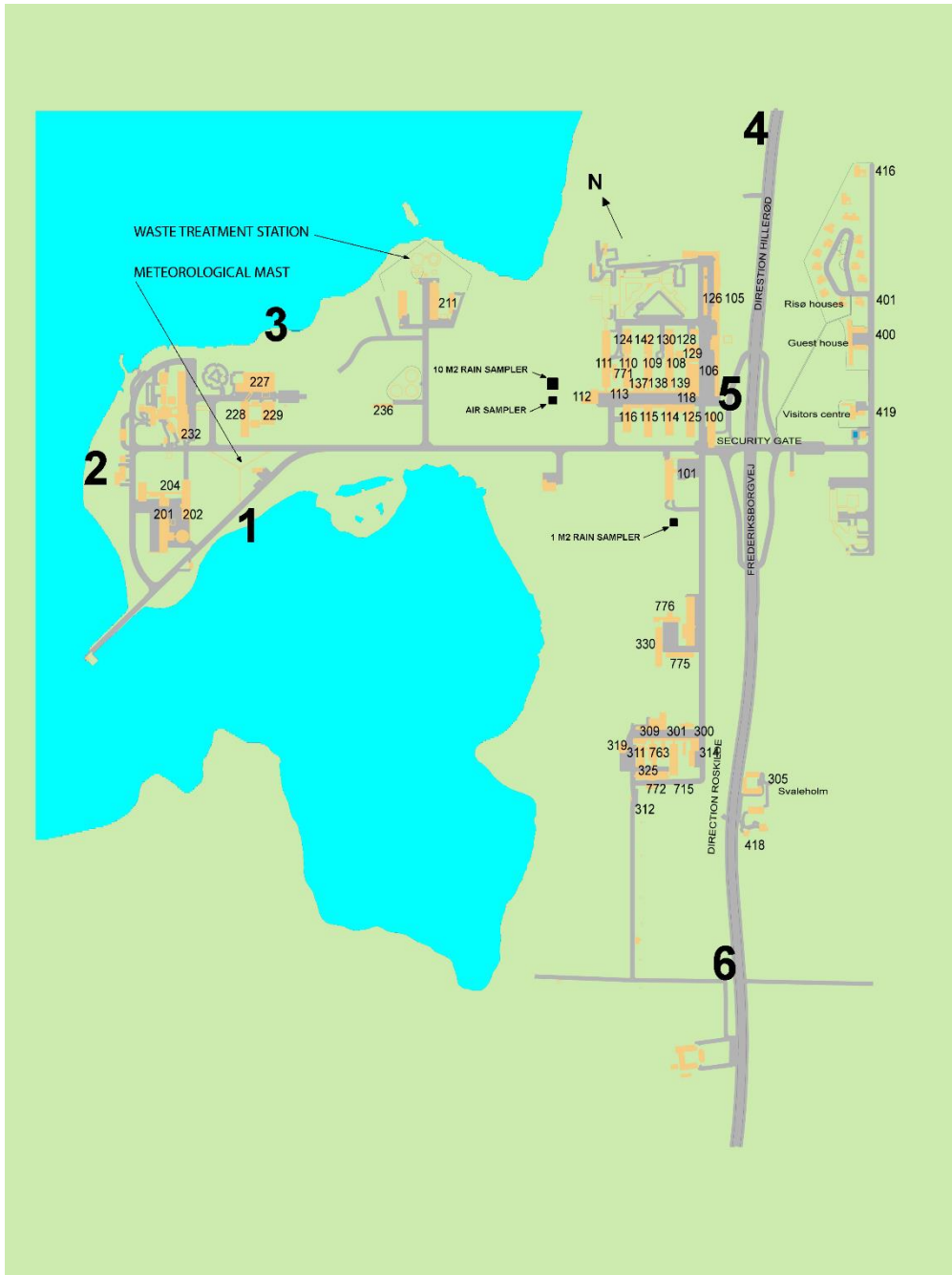


Fig. 8.1. Locations (1-6) for TLD measurements around the border of Risø (cf. Table 8.1).



Fig. 8.2. Locations for measurements of background radiation around Risø in Zones III, IV and V.

Materials and methods

External gamma dose rate monitoring

Monitoring of external gamma dose rate is carried out with the following devices

- Thermoluminescence dosimeters TLD: LiF, measurement frequency annually from May to April. TLD equipment manufacturer: ALNOR/RADOS
- NaI detector: 3x3 inch, SAM 935 Surveillance and Measurement System, Berkeley Nucleonics Cooperation, USA, visual read-out

Calibration of TLD is carried out by irradiation of dosimeters at a calibration irradiator. Traceability of delivered doses is ensured through calibration of the dose rate of the calibration irradiator by the National Institute of Radiation Protection (NIS). Calibration has been verified by measurement with ionisation chamber from NPL, UK. The NaI detector is calibrated periodically vs. a Reuter Stokes high-pressure ionisation chamber.

Air sampler

The sampler at Risø is manufactured by DTU. Air is drawn through a polypropylene filter at a rate of about 2000 m³/h. The filter is normally changed weekly. The flow rate is monitored by a gas meter connected to a shunt. The gas meter reading is compared to that of a reference gas meter intermittently.

DTU analyse the filters by gamma spectrometry shortly after filter change to check for the presence of short-lived man-made radionuclides. The air filters are subsequently stored for a minimum of one week to allow for decay of short-lived naturally occurring radionuclides before repeated gamma analysis. Filters are analysed for ¹³⁷Cs, ⁷Be and ²¹⁰Pb and other gamma emitters.

Deposition collector

The Risø site operates a large rain collector of 10 m². The collector is heated and water is passed through an ion exchange column to a large tank. The 10 m² collector provides monthly samples of rain water analysed for tritium and ion exchange resin which is analysed by gamma spectrometry for ⁷Be, ¹³⁷Cs and ²¹⁰Pb and other gamma emitters.

Water and sediment

A waste water sample from the Waste Treatment Station is collected weekly and analysed for total beta radioactivity and the radionuclides ¹³¹I, ¹³⁷Cs and ²²⁶Ra. Water samples from Roskilde Fjord are collected each quarter and analysed for tritium, annually for ¹³⁷Cs. A sediment sample is collected annually from Roskilde Fjord and analysed for ¹³⁷Cs.

Terrestrial and aquatic biota and flora

Grass samples are collected weekly at the Risø site and analysed by gamma spectrometry. Samples are bulked to monthly samples which are analysed for ^{137}Cs .

Seaweed samples are collected annually from Roskilde Fjord at Risø and analysed for ^{137}Cs .

Sample reception and preparation

Sample identification numbers are entered in log books. Sample preparation methods include drying, freeze drying, ashing, sorting and sieving. Selected samples are archived.

Sample measurements

Radioactivity in samples is measured by total beta counting and gamma spectrometry.

Measurement devices

- Ge detectors for gamma spectrometry. Calibration of detectors is based on mixed-nuclide standards used occasionally. Monthly checks are made of detector efficiency and energy resolution. Background measurements of gamma systems are made a few times per year.
- Low-level Geiger-Müller counters for total beta counting, manufactured by DTU. Calibration based on standards of KCl. Counting efficiency and background are checked monthly.
- Liquid scintillation spectrometer for analysis of tritium in water. Samples are analysed with a calibration standard.

Analytical results, data handling and reporting tools

Analytical results are printed on paper, recorded in log books and stored in a data base on intranet. Results below detection limits recorded as such. Spreadsheets are used for calculating results from raw data.

Quality assurance, laboratory accreditation and intercomparison exercises

Analytical results are checked by experienced staff and discussed with senior scientists if questions arise.

DTU is accredited to testing for radioactivity by DANAK according to the international standard ISO 17025. The accreditation covers testing for certain non-gamma emitting radionuclides but not for radionuclides occurring in the environment and food in general.

DTU participate regularly in international intercomparisons on laboratory analyses of radionuclides.

Conclusions

This report shows the results of the environmental surveillance monitoring programme carried out at and around the Risø site in July-December 2019. The mean concentrations in air were: $0.17 \pm 0.10 \mu\text{Bq m}^{-3}$ of ^{137}Cs , $2.45 \pm 0.94 \text{ mBq m}^{-3}$ of ^7Be and $0.26 \pm 0.16 \text{ mBq m}^{-3}$ of ^{210}Pb (± 1 standard uncertainty). The depositions by precipitation at Risø in the second half of 2019 were: $0.032 \pm 0.005 \text{ Bq m}^{-2}$ of ^{137}Cs , $596 \pm 60 \text{ Bq m}^{-2}$ of ^7Be , $42.0 \pm 4.2 \text{ Bq m}^{-2}$ of ^{210}Pb and 1.3 kBq m^{-2} of ^3H . The average background dose rate (TLD) at Risø (Zone I) was measured as $58 \pm 9 \text{ nSv h}^{-1}$ compared with $56 \pm 5 \text{ nSv h}^{-1}$ (± 1 standard uncertainty) in the four zones around Risø. None of the recorded levels of radioactivity and radiation have given rise to concern.

DTU Environment is working to develop new environmentally friendly and sustainable technologies and disseminate this knowledge to society and new generations of engineers. Research in Radioecology & Tracer Studies (RTS) aims at developing methods and instruments for analysing manmade and naturally recurring radionuclides in the environment and samples from nuclear facilities. The RTS Section is responsible for carrying out the environmental radioactivity monitoring program in Denmark.

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