Radiological environmental monitoring at the ESS facility

annual report 2019

Bernhardsson, Christian; Eriksson Stenström, Kristina; Pédehontaa-Hiaa, Guillaume; Jönsson, Mattias

2020

Faculty of Medicine
Department of Translational Medicine
Medical Radiation Physics
Environmental Radiology and Emergency Preparedness Group

Faculty of Science
Department of Physics
Division of Nuclear Physics
Biospheric and Anthropogenic Radioactivity (BAR) Group

Radiological environmental monitoring at the ESS facility – Annual report 2019

List of authors

Christian Bernhardsson	christian.bernhardsson@med.lu.se
Kristina Eriksson Stenström	kristina.stenstrom@nuclear.lu.se
Guillaume Pedehontaa-Hiaa	guillaume.pedehontaa-hiaa@med.lu.se
Mattias Jönsson	mattias.jonsson@med.lu.se

Approved by: Lars E. Olsson, LU, lars_e.olsson@med.lu.se

Department of Translational Medicine
Medical Radiation Physics
Carl-Bertil Laurells gata 9
SE-205 02 Malmö

Report MA RADFYS 2020:03
Malmö 2020

Department of Physics
Division of Nuclear Physics
Professorsgatan 1
SE-223 63 Lund

Report BAR-2020/03
Lund 2020

SUMMARY

Radiological environmental monitoring of the European Spallation Source (ESS) started in 2017. This report presents results from year 2019, including gamma spectrometry of grass, crops, sewage sludge and ground water; tritium measurements of precipitation, air humidity, surface water, crops and sewage sludge; ^{14}C measurements of grass and fruits/berries. Very low levels of gamma emitting radionuclides were observed in the samples, with no significant contribution of anthropogenic radionuclides. All of the tritium values measured were low and fit the expected environmental levels. The ^{14}C data in vegetation is consistent with the declining ^{14}C specific activity in atmospheric CO_2 and shows no evidence of anthropogenic ^{14}C contamination in the Lund area during 2019.

TABLE OF CONTENT PAGE

LIST OF TABLES

LIST OF FIGURES

List of abbreviations

d.w.	Dry weight
ESS	European Spallation Source
$F^{14}C$	Fraction Modern Carbon
LSC	Liquid Scintillation Counting
LU	Lund University
MDA	Minimum Detectable Activity concentration
SSM	Swedish Radiation Safety Authority
STD	Standard Deviation
SUM	Standard Uncertainty of the Mean

1. BACKGROUND

This report presents the results of the radiological environmental monitoring of the European Spallation Source (ESS) during year 2019, during the pre-operational phase of the facility. The radiological environmental monitoring of the pre-operation phase of the ESS started in 2017 with an extensive programme [1], which was followed up with some additional measurements in 2018 [2].

1.1. Aim

The aim of the current report is to continue to provide preoperational radiological environmental data for the ESS facility for year 2019.

2. METHODOLOGY

2.1. Description of pre-operational sampling programme

The sampling programme for the base-line measurements in 2017 and 2018 is described in detail in [1, 2]. Natural as well as man-made radionuclides were assessed, including gamma-emitters, the pure beta-emitters ^{3}H (tritium) and ^{14}C. Most sampling sites were located within a few km of the ESS site, chosen to cover different wind directions from the ESS site. Monitored constituents / frequencies for the 2017-2018 programmes [1, 2] and the additional measurements performed during year 2019 are summarized in Table 1. The table also lists measurements that were performed during 2018 in a project financed by the Swedish Radiation Safety Authority (SSM) [3].

Table 1 Monitored constituents and frequencies of sampling and measurement for zero point assessments.

Discharge	Monitored constituents	Number of sites/frequency for the 2017-2018 report [1]	Annual report year 2018 [2]	SSM report, for year 2018-2019, ref [3]	This report, year 2019
Airborne	*External radiation*				
	In situ gamma spectrometry	21 sites			
	Mobile	Ambient dose equivalent rate at 29 sites. One assessment by car covering the ESS nearby areas (about 2 km in radius).			
	Air, deposition				
	Soil, gamma-emitting radionuclides	Down to a depth of 20 cm at 22 sites. Down to a depth of 7 cm at 29 sites.			
	Foodstuff and/or ingestion				
	Fruits, berries	^{14}C at 12 sites	^{14}C at 10 sites		^{14}C at 6 sites
	Crops	Gamma-emitters at 12 sites, ^{14}C at 6 sites, ^{3}H at one site.	^{14}C at 2 sites		Gamma-emitters in 13 samples, ^{3}H in 6 samples
	Milk and forage	Gamma emitters, ^{3}H and ^{14}C at one site on one occasion.			
	^{14}C in annual tree rings	Years 2012-2016 at 4 sites (2 around ESS, 1 urban background and 1 rural background site).			
	^{14}C in fullerene soot monitors	Same sites as tree rings, four 4-week periods.			
	Drinking water and/or well water	^{3}H at 4 sites	^{3}H at one site		Gamma-emitters in 4 samples, Lund tap water appr monthly for ^{3}H
	Terrestrial indicators				
	Grass	Gamma-emitters at 20 sites, ^{14}C at 12 sites	^{14}C at 8 sites		Gamma-emitters at 6 sites, ^{14}C at 2 sites
	Lichen, moss	Gamma-emitters at 13 sites, ^{14}C at 12 sites			
	Precipitation and air				
	Precipitation		Continuous sampling for ^{3}H analysis. Urban reference site 2018-03-19 to 2018-04-13; ESS site 2018-04-13 to 2018-05-03.	Continuous sampling of precipitation for ^{3}H analysis at ESS site. Monthly basis, start April 2018. Results until April 2019 in [3].	
	Air humidity		Grab sampling for ^{3}H analysis. One sample at urban reference site, two samples at ESS site.	Grab sampling for ^{3}H analysis. Monthly basis, start May 2018. At ESS site and urban reference site. Results until April 2019 in [3].	
Liquid	*Water bodies*				
	Ground water	^{3}H at 12 sites			Gamma in 4 samples
	Surface water	^{3}H at 8 sites			Appr monthly at 3 ponds, ^{3}H. Källby pond.
	Sewage sludge	Gamma-emitters and ^{3}H at Källby: Monthly samples from April 2017 – April 2018.	Gamma-emitters and ^{3}H at Källby: two occasions		Gamma-emitters and ^{3}H in 2 samples

2.2. Sampling locations

Sampling site for year 2019 are listed in Table 2 (the previous sample site notations used in Ref [1] for ^{3}H and ^{14}C are included in Table 2).

Table 2 Sampling locations and type of measurements performed during year 2019.

Site	Location	Latitude	Longitude	Corre-sponding site nr ^{14}C	Corre-sponding site nr ^{3}H	Gamma	^{3}H	^{14}C
4	Ladugårdmarken 461	N55.7384	E13.2314	C6		X[2]		
5	Switchgear (SW of ESS)	N55.7347	E13.2418			X[1]		
6	Möllegården	N55.7304	E13.2441	C3		X[1]		X[1,10]
27.3	ESS SE corner (at stones)	N55.7346	E13.2597	C12		X[1]		
30	Kopparstaden windmill	N55.7385	E13.2543	C30		X[1]		
31.2	ESS area (2)	N55.7327	E13.2427			X[2]		
31.3	ESS area (3)	N55.7298	E13.2436			X[2]		
31.6	ESS area (6)	N55.7358	E13.2442		T2b		X[5]	
31.20	ESS area (20)	N55.7366	E13.2455		T2a		X[6,7]	
32.3	Dammstorpsvägen 16 (field 2)	N55.7266	E13.2554			X[4]		
32.4	Dammstorpsvägen 16 (field 3)	N55.7287	E13.2519			X[4]		
34.4	MaxIV area (4)	N55.7270	E13.2363	C29				X[10]
34.5	MaxIV area (5)	N55.7283	E13.2376				X[5]	
35	Källby (sewage treatment plant) VA SYD	N55.6952	E13.1637			X[12]	X[12]	
36.1	Svenstorp's gods (1), at castle	N55.7669	E13.2532			X[4]		
36.2	Svenstorp's gods, farmland (2) field "6-0"	N55.7583	E13.2508			X[3]	X[3]	
36.3	Svenstorp's gods, farmland (3) field "8-0"	N55.7449	E13.2442			X[3]	X[3]	
36.4	Svenstorp's gods, farmland (4) field "5-0"	N55.7515	E13.2397			X[3]	X[3]	
36.7	Svenstorp's gods, farmland (7) field "10-0"	N55.7509	E13.2597			X[4]		
36.9	Svenstorp's gods, farmland (9) field "8-1"	N55.7378	E13.2416			X[4]		
36.10	Svenstorp's gods, farmland (10) field "22-0"	N55.7421	E13.2594			X[4]		
36.11	Svenstorp's gods, farmland (11) field "9-0"	N55.7445	E13.2529			X[4]		
36,12	Svenstorp's gods, farmland (12) field "1-0"	N55.7616	E13.2423			X[3]	X[3]	
36,13	Svenstorp's gods, farmland (13) field "4-0"	N55.7572	E13.2349			X[3]	X[3]	
36.14	Svenstorp's gods, farmland (14) field "11-0"	N55.7571	E13.2742			X[4]		
47	Borrby	N55.4256	E14.2236	C1 (rural ref)				X[1,11]
48	Timjanvägen 5, Lund	N55.7186	E13.1828	C2 (urban ref)	T1a		X[7,8]	X[10]
52	Professorsgatan 1	N55.7097	E13.2047	C18				X[11]
54	Active Biotech	N55.7169	E13.2206	C21				X[11]
62	Grevie PV5 well	N55.6131	E13.1970		T0		X[9]	
63	Monument park	N55.7182	E13.1851				X[5]	

1 – grass; 2 – ground water; 3 – spring barley; 4 – winter wheat; 5 – surface water; 6 – precipitation; 7 – air humidity; 8 – tap water; 9 – ground water (background); 10 – apple; 11 – rowan berries; 12 – sewage sludge;

In 2019, a rather limited sampling and measurement campaign was carried out for studying the concentration of gamma emitting radionuclides in the area. The main focus was on crops from the nearby fields, whereas a limited number of water, grass and sewage sludge samples was collected and analysed. The sites for gamma spectrometry measurements are shown in Figure 1.

Figure 1 Sampling sites for samples for gamma spectrometry measurements for year 2019, except Site 35 "Källby VA SYD" that is located in the southern part of Lund. See Table 2 for more information about the sampling sites.

The sites used for the ^{3}H sampling in 2019 are shown in Figure 2. Reference (background) sites for ^{3}H were:

- **Sites 48 and 63, located ~4.3 km from ESS (~246°).** This site in the north of Lund acted as a rural background site for air humidity and surface water, respectively. Tap water (from VA Syd) was also collected at this site.
- **Site 62, located ~13.8 km from ESS (~193°).** Water from the deep well Grevie PV5 (depth 71-72 m) at site 62, operated by VA Syd, was used as background water (supported by a previous study: ^{3}H concentration of about 0.02 TU, corresponding to 0.002 Bq L^{-1} [4]).

Figure 2 Sampling sites for ^{3}H samples for year 2019. See Table 2 for more information about the sampling sites.

The sampling sites for ^{14}C for year 2019 are shown in Figure 3. Two sites close to the ESS were assessed: Möllegården (site 6) and MaxIV (site 34.4). Two background sites were also included: the rural background site Borrby (site 47), located 70 km ESE from the ESS site, and the urban sampling site in northern Lund (site 48), located 4.6 km (WSW) from the ESS site. As for the 2017 and 2018 assessments, two samples were collected in the north-eastern part of Lund, where potential sources of anthropogenic ^{14}C exist: sites 52 and 54.

Figure 3 Sampling sites for ^{14}C samples for year 2019. See Table 2 for more information about the sampling sites.

2.3. Methods for sample collection and analysis

All collection and sample preparation procedures are described in Refs [1, 3]. An updated procedure for sampling, sample preparation and measurements of ^{3}H activity concentration is provided in Appendix 1. Weather data was obtained from a Davis Vantage Pro2 weather station located at the ESS site 31.20 (see Ref [3] for details). Average wind speed and directions were logged every hour.

3. RESULTS

3.1. Weather data 2019

Main wind directions in year 2019 (see Figure 4) were WSW (12.3% of the time), ESE (12.2%), W (11.6%) and SW (11.3%). Average annual wind speeds for these sectors were 4.2 m/s, 3.8 m/s, 3.8 m/s and 5.2 m/s, respectively. Wind from W, WSW, SW and SSW accounted for 44.2% over the year. Average wind speed for all wind directions over the whole year was 3.7 m/s.

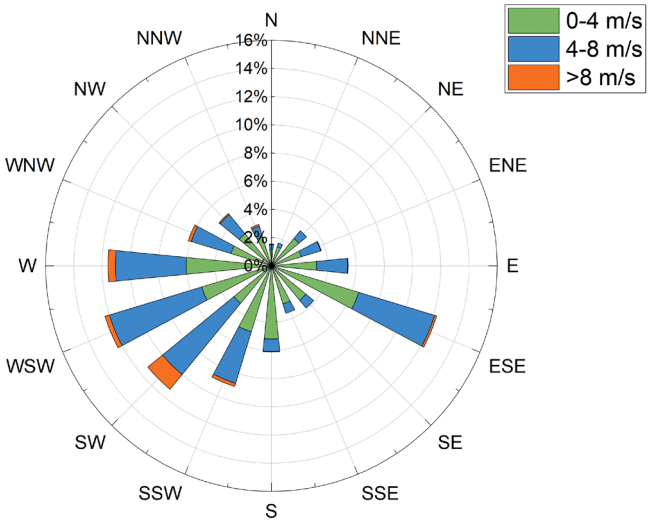

Figure 4 Wind rose for ESS for year 2019, obtained from hourly measurements with a Davies Vantage Pro weather station located at ESS site 34.20.

3.2. Activity concentration of gamma emitting radionuclides in various types of samples year 2019

Tables A2.1-2.4, in Appendix 2, present the results from the measurements of the activity concentration of gamma emitting radionuclides in the 25 samples collected in 2019.

The detected levels of gamma emitting radionuclides in the four water samples were below the detection limit for the measurement time: ^{137}Cs (0.2 Bq kg^{-1} < MDA < 1.6 Bq kg^{-1}); ^{226}Ra (9 Bq kg^{-1} < MDA < 30 Bq kg^{-1}); ^{228}Ac (2.3 Bq kg^{-1} < MDA < 4.7 Bq kg^{-1}), except for one sample that had a measurable level of ^{40}K: 23±12 Bq kg^{-1}. Also, the six grass samples had

activity concentrations that were below the detection levels for the measurement time: ^{137}Cs (3 Bq kg^{-1} < MDA < 7 Bq kg^{-1}); ^{226}Ra (37 Bq kg^{-1} < MDA < 92 Bq kg^{-1}); ^{228}Ac (9 Bq kg^{-1} < MDA < 23 Bq kg^{-1}), but measurable levels of ^{40}K, ranging between 426 Bq kg^{-1} to 658 Bq kg^{-1} (average: 552±67 Bq kg^{-1}).

Thirteen samples of crops (spring and autumn barley) collected in 2019 had activity concentrations that were below the detection limit for the measurement time: ^{137}Cs (0.6 Bq kg^{-1} < MDA < 2.6 Bq kg^{-1}); ^{226}Ra (9 Bq kg^{-1} < MDA < 49 Bq kg^{-1}); ^{228}Ac (2.1 Bq kg^{-1} < MDA < 7.6 Bq kg^{-1}), but measurable levels of ^{40}K, ranging between 122 Bq kg^{-1} to 213 Bq kg^{-1} (average: 162±36 Bq kg^{-1}).

The two samples of sewage sludge were analysed too long after collection to identify any radionuclides used at the hospital, SUS Lund, e.g. ^{131}I and ^{177}Lu. However, radionuclide concentrations of ^{137}Cs, ^{226}Ra, ^{228}Ac and ^{40}K were measurable in one of the samples: 3±0.5 Bq kg^{-1}, 27±13 Bq kg^{-1}, 33±2 Bq kg^{-1} and 182±33 Bq kg^{-1}, respectively. The activity concentration in the other sample was below the detection limit for ^{226}Ra (MDA = 72 Bq kg^{-1}) and ^{137}Cs (MDA = 4 Bq kg^{-1}), whereas the concentrations of ^{228}Ac and ^{40}K were determined as 31±6 Bq kg^{-1} and 137±134 Bq kg^{-1}, respectively.

3.3. Analysis of ^{3}H year 2019

The results of the tritium measurements of samples of precipitation, air humidity and surface water collected since end of April 2019 are shown Tables A2.5-A2.8 in Appendix 2. Values for the beginning of the year can be found in Ref [3] (report to SSM). Repeated measurements on Lund tap water can be found in Table A2.9, and the results from the analyses of sewage sludge and crops are presented in Table A2.10. The observed levels are generally below the detection limit using the current procedure, instrumentation and analysis time (MDA typically 1.2 – 1.6 Bq L^{-1}). The results show no evidence of any local contamination of ^{3}H in Lund during 2019 (compare Figure 10 in Ref [3]).

3.4. ^{14}C analysis year 2019

The results of the ^{14}C analysis of grass and crops are presented in Table A2. Table A2.11 in Appendix 2 and Figure 5. The results are expressed as F^{14}C [5, 6], see Ref [1] (p. 92-94) for definition and conversion to other activity concentration units[1].

[1] F^{14}C values corresponding to naturally produced ^{14}C are close to 1. Maximum F^{14}C values observed in 1963 due to testing of atmospheric nuclear weapons in the late 1950s and early 1960s was around 2. F^{14}C in atmospheric CO_2 is currently approaching the pre-bomb levels. Typical F^{14}C values found in environmental samples in the vicinity of light water reactors may be elevated by up to several % compared to F^{14}C values at sites remote from such facilities

[7] Stenström, K., Skog, G., Nilsson, C.M., Hellborg, R., Leide-Svegborn, S., Georgiadou, E., Mattsson, S. *Local variations in ^{14}C – How is bomb-pulse dating of human tissues and cells affected?* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 268(7–8): 1299-1302, 2010, [8] Stenström, K., Erlandsson, B., Hellborg, R., Wiebert, A., Skog, G. *Environmental levels of carbon-14 around a Swedish nuclear power plant measured with accelerator mass spectrometry.* Ibid.113(1–4): 474-476, 1996.

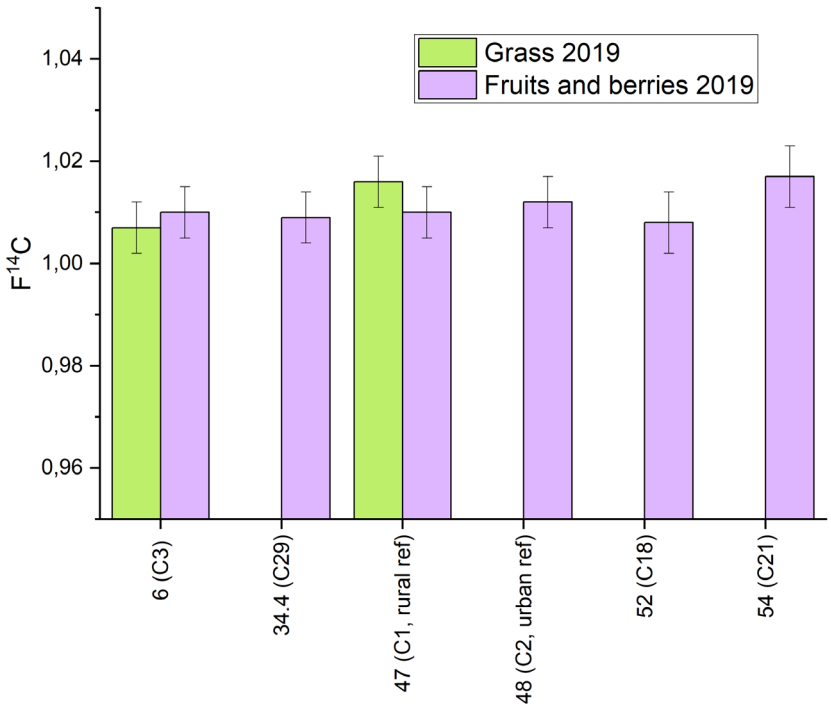

Figure 5 Results of the ^{14}C measurements of grass and fruits/berries from 2019. Uncertainties represent 1 standard deviation.

The average $F^{14}C$ value of the 2 grass samples and 6 fruit/berries samples was 1.011 (STD: 0.004; SUM: 0.001). This corresponds to a specific activity of 227 Bq kg^{-1} C using $\delta^{13}C$= - 25 ‰ (see Annex B4 in Ref [1]). The data is normally distributed, with no outliers according to Grubb's test.

The average $F^{14}C$ values obtained in for all ESS zero point assessments so far are shown in Figure 6, along with ^{14}C data in atmospheric CO_2 collected at rural background stations in central Europe (high altitude) and at the Swedish ICOS station Hyltemossa (56.0976 N, 13.4189 E, 115 m above sea level, sampling height 150 m above ground)[2] [10-14]. The declining $F^{14}C$ values in atmospheric CO_2 result from two effects: 1) uptake of bomb-^{14}C into the oceans (and biosphere), and 2) dilution of ^{14}C specific activity due to input of ^{14}C-free CO_2 from fossil fuels into the atmosphere. The ^{14}C concentration in vegetation is expected to differ somewhat from the average ^{14}C concentration in atmospheric CO_2. Firstly, plants only absorb CO_2 at daytime during the growing season. Secondly, CO_2 respired from microbial processes in soil is taken up by the plants through photosynthesis [15-17]. Near-ground air may thus contain more bomb-pulse ^{14}C (from decomposing of litter from previous years) than the atmosphere on average. These effects may be reflected in some of the data in Figure 6. Local differences may also occur between sites with different influence from fossil fuel CO_2.

[2] ICOS ^{14}C data ($\Delta^{14}C$) has been recalulated into $F^{14}C$ according to $F^{14}C = \left(\frac{\Delta^{14}C}{1000} + 1\right) e^{(y-1950)/8267}$, where *y* is the year.

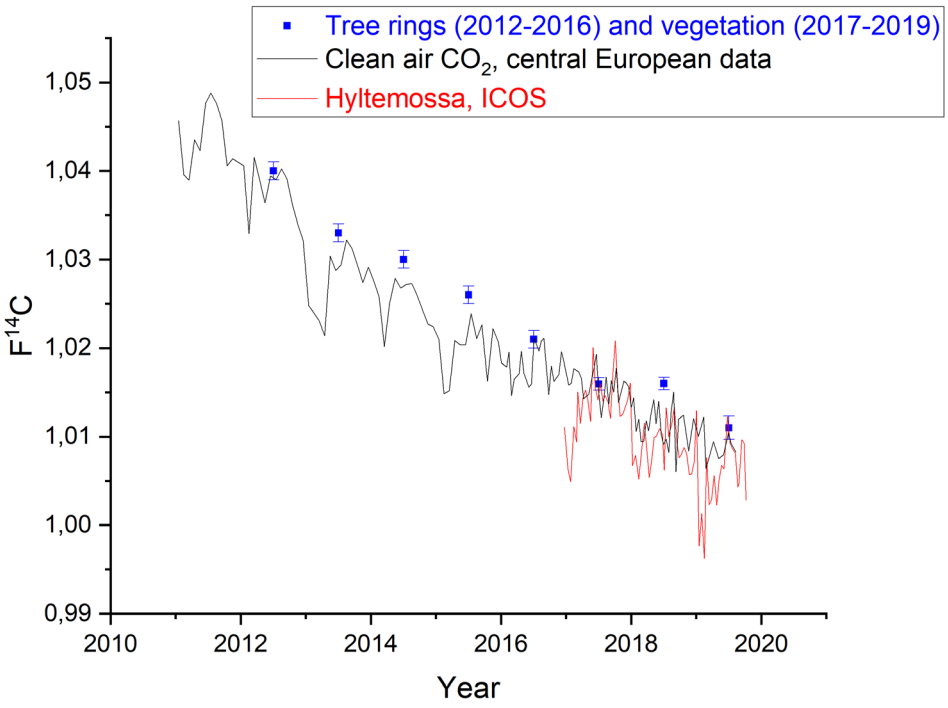

Figure 6 Average $F^{14}C$ values (uncertainty represented by the SUM) obtained in for all ESS zero point assessments so far, along with ^{14}C data in atmospheric CO_2 collected at rural background stations in central Europe and at the Swedish ICOS station Hyltemossa [10-14].

3.5. Quality assurance

Samples of deep well water (Grevie-Bulltofta verket, VA Syd) with a well-documented low tritium concentration were used as background and dilutions of tritiated water samples with certified values (from the inter-comparison exercise Procorad, 2019) were used as control in the tritium measurements. A quenching curve was also obtained using the method described by the cocktail provider Perkin Elmer [18].

The high purity germanium spectrometry systems (HPGe) are regularly checked for efficiency, gain stability and energy resolution using a standard solution containing ^{60}Co, ^{137}Cs and ^{241}Am. To ensure that the detector or lead cage has not been contaminated, a background spectrum is obtained and studied at each quality assurance occasion. The laboratory participates in the annual IAEA proficiency test (IAEA-TEL). In 2019, we also participated in the PROCORAD intercomparison test. Our results have during the last years been satisfying and we are confident in our methods for sample preparation, measurements and evaluation.

For the gamma spectrometry measurements, the activity concentrations of several naturally occurring radionuclides are low, especially in biological and water samples. The minimum detectable activity (MDA) will decrease with increasing counting time. To match the rate of incoming samples and still have a reasonable detection limit, we have chosen a counting time of at least two days. Samples with very small mass are usually measured over the weekend. Since the counting time varies between different samples, the MDA will not be

constant. The detection limit is also specific for each radionuclide and detector. The MDA value is therefore presented together with the activity estimate and in the result section.

The quality of the ^{14}C data was assured by measurement and analysis of secondary standards as described in Ref [1].

4. SUMMARY AND CONCLUSIONS

Very low levels of gamma emitting radionuclides were observed in the samples collected in 2019, with no significant contribution of anthropogenic radionuclides. It is however suggested to monitor e.g. sewage sludge more carefully (for example by weekly samplings followed by direct analysis and to continue to sample sludge on a monthly basis) as this is an excellent indicator of the anthropogenic radionuclide inventory that is managed in the area. Furthermore, it would be useful to correlate the magnitude and variability of the radionuclide inventory in sewage sludge, after such sampling, and to correlate it with the administered radioactivity at SUS Lund during the same period. Finally, it should be noted that seasonal variations of the concentration of gamma emitting radionuclides should be followed in e.g. samples of grass.

All of the tritium values measured were low and fit the expected environmental levels. A slight increase of tritium activity concentration is observed in the surface water samples during the spring: this phenomenon called “spring leak” is also expected. The slightly higher values measured in the sewage sludge were also observed in 2018.

The ^{14}C data in vegetation is consistent with the declining ^{14}C specific activity in atmospheric CO_2 and shows no evidence of anthropogenic ^{14}C contamination in the Lund area during 2019.

5. ACKNOWLEDGMENT

This work was financed by ESS under collaboration agreement ESS-0093103. The authors (Medical Radiation Physics in Malmö and Division of Nuclear Physics, Lund University) would like to express gratitude to the individuals, families and companies that have helped with providing access to the sites investigated and samples collected during this programme.

6. REFERENCES

[1] Bernhardsson, C., Eriksson Stenström, K., Jönsson, M., Mattsson, S., Pedehontaa-Hiaa, G., Rääf, C., Sundin, K., et al. *Assessment of "Zero Point" radiation around the ESS facility.* Report MA RADFYS 2018:01, Report BAR-2018/04. Lund University, Sweden. https://portal.research.lu.se/portal/sv/publications/assessment-of-zero-point-radiation-around-the-ess-facility(2153e07c-b465-4191-abc3-dbfcaf28b85b).html. 2018.

[2] Bernhardsson, C., Eriksson Stenström, K., Pedehontaa-Hiaa, G. *Radiological environmental monitoring at the ESS facility – Annual report 2018.* Lund University. 2020.

[3] Eriksson Stenström, K., Barkauskas, V., Pedehontaa-Hiaa, G., Nilsson, C., Rääf, C., Holstein, H., Mattsson, S., et al. *Identifying radiologically important ESS-specific radionuclides and relevant detection methods.* SSM report of project SSM 2018-1636.

https://www.stralsakerhetsmyndigheten.se/publikationer/rapporter/stralskydd/2020/202008/. 2020.
[4] Åkesson, M., Suckow, A., Visser, A., Sültenfuβ, J., Laier, T., Purtschert, R., Sparrenbom, C.J. *Constraining age distributions of groundwater from public supply wells in diverse hydrogeological settings in Scania, Sweden.* Journal of Hydrology, 528: 217-229, 2015.
[5] Reimer, P.J., Brown, T.A., Reimer, R.W. *Discussion: Reporting and calibration of post-bomb ^{14}C data.* Radiocarbon, 46(3): 1299-1304, 2004.
[6] Eriksson Stenström, K., Skog, G., Georgiadou, E., Genberg, J., Johansson, A. *A guide to radiocarbon units and calculations.* Internal Report LUNFD6(NFFR-3111)/1-17/(2011). Lund University. D.o.N.P. Dep of Physics. http://lup.lub.lu.se/search/ws/files/5555659/2173661.pdf. 2011.
[7] Stenström, K., Skog, G., Nilsson, C.M., Hellborg, R., Leide-Svegborn, S., Georgiadou, E., Mattsson, S. *Local variations in ^{14}C – How is bomb-pulse dating of human tissues and cells affected?* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 268(7–8): 1299-1302, 2010.
[8] Stenström, K., Erlandsson, B., Hellborg, R., Wiebert, A., Skog, G. *Environmental levels of carbon-14 around a Swedish nuclear power plant measured with accelerator mass spectrometry.* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 113(1–4): 474-476, 1996.
[9] Bernhardsson, C., Eriksson Stenström, K. *Point zero measurements for the ESS facility.* Report in preparation. Lund University. 2018.
[10] Levin, I., Kromer, B. *The tropospheric $^{14}CO_2$ level in mid latitudes of the Northern Hemisphere (1959-2003).* Radiocarbon, 46: 1261-1271, 2004.
[11] Levin, I., Kromer, B., Hammer, S. *Atmospheric Δ14CO2 trend in Western European background air from 2000 to 2012.* Tellus B: Chemical and Physical Meteorology, 65(1): 20092, 2013.
[12] Hammer, S., Levin, I. *Monthly mean atmospheric D14CO2 at Jungfraujoch and Schauinsland from 1986 to 2016.* heiDATA: Heidelberg Research Data Repository [Distributor] V2 [Version]. http://dx.doi.org/10.11588/data/10100; www.calibomb.org. 2017.
[13] Conen, F., Emmenegger, L., Leuenberger, M., Steger, D., Steinbacher, M. *"ICOS RI, 2020. ICOS ATC 14C Release, Jungfraujoch (10.0 m), 2016-01-04_2019-08-12".* https://hdl.handle.net/11676/X-IXPKZlO4DWX7wncsLQ7akY; http://calib.org/CALIBomb/. 2019.
[14] ICOS (Integrated Carbon Observation System). *ICOS ATC 14C Release from Hyltemossa 2016-12-22 – 2019-10-08.* https://data.icos-cp.eu/portal/. PID: 11676/WE07MZ3UlYcJau76jaTIW_Sg. ICOS DATA is licensed under CC4BY (http://creativecommons.org/licenses/by/4.0/). 2020.
[15] IRSN. *Carbon-14 and the environment*. Available at http://www.irsn.fr/EN/Research/publications-documentation/radionuclides-sheets/environment/Pages/carbon14-environment.aspx. 2012. Accessed: 2018-02-01.
[16] Trumbore, S. *Radiocarbon and Soil Carbon Dynamics.* Annual Review of Earth and Planetary Sciences, 37(1): 47-66, 2009.
[17] Mobbs, S., Smith, K., Thorne, M., Smith, G. *Modelling approaches to C-14 in soil-plant systems and aquati environments.* SSM2014:30. 2014.
[18] Thomson, J. *Use and Preparation of Quench Curves in Liquid Scintillation Counting, Application Note.* Perkin Elmer Inc., Waltham Massachusetts United States. https://www.perkinelmer.com/se/liquidscintillation/images/APP_Use-and-Preparation-of-Quench-Curves-in-LSC_tcm151-171749.pdf. 2014.

APPENDIX 1. PROCEDURE SAMPLING, SAMPLE PREPARATION AND MEASUREMENT OF ^{3}H ACTIVITY CONCENTRATION USING LIQUID SCINTILLATION COUNTING

SAMPLING, SAMPLE PREPARATION AND MEASUREMENTS OF ^{3}H ACTIVITY CONCENTRATION USING LIQUID SCINTILLATOR COUNTING

Purpose

The purpose of this appendix is to describe the sampling, sample preparation and measurement of the activity concentration of ^{3}H using liquid scintillation counting (LSC) analysis for the framework agreement entitled "For the provision of projects, services and reporting linked to the Radiological and Environmental Monitoring System" (contract ESS-2702700).

Applicability

The description (procedure) is applicable to various environmental samples measured at the Lund University laboratory at the Medical Radiation Physics group in Malmö using LSC. The procedure is applicable to future measurements of tritiated water (HTO) in various samples collected from the Lund area to assess the radiological impact of ESS. The current method does not allow measurement of the organically bound tritium.

Methodological outline

For the background mapping of ^{3}H, samples of water, sewage sludge, milk, meat and crops have been sampled, measured and analysed.

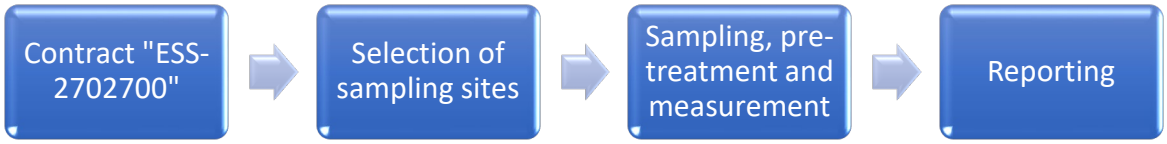

Figure 3. 1. Flow of actions for achieving the zero point measurements of the ^{3}H activity concentration in various types of samples by laboratory LSC analysis.

Each sample measured for the ^{3}H concentration in the LSC is prepared according to a procedure that was initially developed during the ESS-0093103 contract period. The sewage sludge, milk, meat and crops were freeze dried in order to extract water from the samples, prior to the chemical pre-treatment and LSC analysis. After evaluation of raw data, the results are presented in a final report.

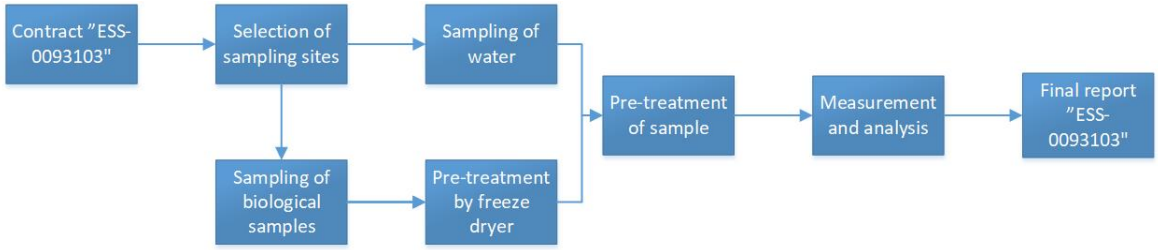

Figure B3. 1. Details of the workflow of the ^{3}H procedure developed during the ESS-0093103 contract.

Methodological details

Selection of sampling sites

Several sampling sites were selected within the fenced area of the ESS, in the close vicinity of the ESS site and around Lund municipality.

Ground water was sampled in the existing drill holes on the ESS site as well as in the close vicinity of the ESS site. In addition, water samples were collected in some private wells (although not all of them were in operation), in households within the approximately 1500 m radius from ESS, and from the five water outlet ponds at Källby sewage treatment facility in Lund.

Surface water was collected from the existing ponds and streams within the site perimeter and in Lund municipality.

Precipitation was collected as an average monthly sample on the ESS site.

Air humidity was collected on the ESS site and on an urban background site in Lund.

Sewage sludge is an indicator of the collective radionuclide concentration in human population in the catchment area of the sewage treatment facility. To a varying degree the sludge also contains radionuclides from runoff and from household and processed water.

Milk and meat were sampled from one farmer. Although there are currently few dairy or meat farms close to ESS, those products are an important pathway of transfer to humans from ground deposited radionuclides.

Crop including sugar beets, apples, berries, buckwheat and barley were collected as bioindicators in the farmlands surrounding the ESS site.

Tap water received in Lund households was also measured for comparison purposes.

Output	Representative sampling locations for determination and follow up of ^{3}H activity concentration in water, sewage sludge and foodstuff, for ^{3}H background dose estimates to representative persons.

General aspects on sampling

It is recommended to wear gloves during sampling, especially when in direct contact with the sample itself. At the site, each sample taken must be carefully documented and marked in temporary containers that are carefully sealed. To avoid cross contamination, special precaution should be taken when several samples are taken and when a ^{3}H contamination is suspected.

Special sampling tools are required for sampling of surface water (a long enough rod to reach out in the water without disturbing it) and ground water (a long enough rope to reach the water level).

The site itself should be documented by its GPS coordinates, photographs from different directions and preferably also at various distances to include ambient information that might be of importance for the future.

Between samplings, it is important to carefully clean the sampling probe, *e.g.* by washing of the sampler with clean water.

The information collected during the sampling must be digitalised as soon as possible and after that stored and secured.

Output	Reducing the risk of cross-contamination between samples. Guarantees sampling locations for the future. Minimizes the risk of loss of data.

Sampling and sample preparation

Pre-treatment of the samples

The water was sampled in 500 ml plastic bottles which were kept in a refrigerator until preparation. To reduce quenching and remove ions and the organic matter from the sample, all ground and surface waters were distilled.

Water from the sludge and foodstuff samples was extracted using freeze-drying. The sludge and the foodstuff were frozen at -18 °C directly after being collected. Using a hammer, the sludge was crushed to smaller pieces and placed in an aluminium tray. The freeze dryer process started in the morning and was turned off at the end of the day. In the following morning, the extracted water from the sample was retrieved from the condenser of the freeze dryer.

The mass of the samples was measured before and after the freeze-drying process. To extract all water from the sludge, the samples were placed in a drying cabinet until they were completely dry. Thereafter, the mass and water content were determined.

The method was tested using a tritium tracer to establish the freeze-drying yield. It shall be noted that only the water fraction is measured in this method and not the organically bound tritium in the solid residue.

^{3}H analysis using liquid scintillator counting

Samples were split in two to duplicate the measurements, 10 ml of water were mixed with 10 ml of Ultima Gold LLT scintillation cocktail (Perkin Elmer) in a 20 ml polyethylene vial. Samples were then shaken for 10 min and stored in the dark for 48 hours to decrease chemical quenching before measurement. Each duplicate was then measured in a Beckman LS 6500 multi-purpose LSC during a total of 10 hours (5*2h) and evaluated using a dedicated user programme.

Samples of deep well water (Grevie-Bulltofta verket, VA Syd) with a well-documented low tritium concentration were used as background and samples with certified values (Procorad 2019) were used as control. The Horrocks's method was used for quenching correction.

Uncertainty estimate

The uncertainty in the results is dominated by the statistical uncertainty in the count rates of the sample and background. Uncertainties introduced in volume and efficiency measurements are negligible compared to the statistical uncertainty in the number of counts.

The confidence interval of the activity concentration, $\bar{A}$, was calculated using the following equation:

$$\bar{A} = \frac{t_{n-1;0.95} \times \sigma}{\sqrt{n-1}} \qquad \text{(Eq. B3.1)}$$

where *n* is the number of measurements, σ is the standard deviation of the repeated measurements and *t* is the Student t-value for a two-tailed distribution with a degree of freedom of *n*-1 and a confidence interval of 95%.

Minimum detectable activity

The minimum detectable activity, MDA, is defined according to [1]:

$$MDA = \frac{3.29 \cdot \sqrt{\left(\frac{cpm_b}{t_s}\right) + \left(\frac{cpm_b}{t_b}\right) + \left(\frac{2.71}{t_s}\right)}}{60 \cdot E \cdot V} \qquad \text{(Eq. B3.2)}$$

where cpm_b is the count rate of the background (counts per minute), t_s is the measurement time of the sample (min), t_b is the measurement time of the background (min), *E* is the efficiency and *V* is the sample volume (l). The background count rate was established using deep well water with a well-documented low tritium level. The counting time was set to 1200 minutes. This long counting time and the cumulative background measurements gave MDA values going from 1.5 to 3 Bq l^{-1}. This method provides results with MDA levels close to the natural levels of tritium in the environment and well below the limit of 100 Bq l^{-1} in drinking water set by the National Food Agency [2].

Output	Activity concentration of tritiated water in various types of samples.

Storage of samples

An overview of storage procedures for ^{3}H samples is provided in [3].

Sampling, pre-treatment and measurement

Before analysis, the water samples are stored in air tight and closed containers in a fridge (-5 °C). After analysis, the water containers are transferred to a storage room and kept a room temperature.

Sewage sludge and foodstuff are stored in closed containers in a freezer (-18 °C).

Output	The samples collected are possible to re-analyse in the future. However, when the sample comes in contact with any other material such as the container or the atmosphere, the tritium content may change. A re-analyse of a background sample is thus of limited value.

Output

Representative sampling locations for determination and follow up of ^{3}H activity concentration in water, sewage sludge and foodstuff, for background ^{3}H dose estimates to representative persons.

^{3}H activity concentration for the sampled matrices.

Reducing the risk of cross-contamination between samples. Guarantees sampling locations for the future. Minimizes the risk of loss of data.

References

[1] Currie, L.A. *Limits for qualitative detection and quantitative determination. Application to radiochemistry.* Anal Chem, 40(3): 586-593, 1968.

[2] Livsmedelsverket. *Vägledning till livsmedelsverkets föreskrifter (SLVFS 2001:30) om dricksvatten*. 2016.

[3] Kim, D., Croudace, I.W., Warwick, P.E. *The requirement for proper storage of nuclear and related decommissioning samples to safeguard accuracy of tritium data.* Journal of hazardous materials, 213: 292-298, 2012.

APPENDIX 2. DATA FROM MEASURENTS 2019

Available upon request. Please contact the authors.