Radiological environmental monitoring at the ESS facility – Annual report 2021

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

2022

Document Version:
Other version

Link to publication

Citation for published version (APA):
Eriksson Stenström, K., Bernhardsson, C., Jönsson, M., & Pédehontaa-Hiaa, G. (2022). *Radiological environmental monitoring at the ESS facility – Annual report 2021*. Lund University.

Total number of authors:
4

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 2021

List of authors

Christian Bernhardsson	christian.bernhardsson@med.lu.se
Kristina Eriksson Stenström	kristina.stenstrom@nuclear.lu.se
Mattias Jönsson	mattias.jonsson@med.lu.se
Guillaume Pedehontaa-Hiaa	guillaume.pedehontaa-hiaa@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 2022:01
Malmö 2022

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

Report BAR-2022/01
Lund 2022

SUMMARY

Results from the radiological environmental monitoring of the European Spallation Source (ESS) are presented for 2021. Previous zero-point assessments (2017-2020) have mainly focussed on terrestrial samples. New sample types for 2021 include a sediment sample from a pond at the ESS and brown seaweed (*Fucus*) from Lomma bay and from the east coast of Scania (Skillinge). For gamma-emitting radionuclides, increased levels of anthropogenic radioactivity (^{177}Lu and ^{131}I) originating from hospital use, were only observed in sewage sludge samples. For tritium, the majority of the samples had activity concentrations that were below the minimum detectable activity (MDA) of 1.62 Bq l^{-1}. Expected environmental levels, without any evidence of local contamination, were also seen in the ^{14}C data.

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
MDA	Minimum Detectable Activity concentration
SDI	Spectrum Dose Index
SSM	Swedish Radiation Safety Authority
STD	Standard Deviation
SUM	Standard Uncertainty of the Mean

1. BACKGROUND

Results of the radiological environmental monitoring of the pre-operation phase of the European Spallation Source (ESS) have been reported since 2017 [1-4]. In this report we present results for 2021.

1.1. Aim

The aim of the current report is to provide continued preoperational radiological environmental data for ESS for year 2021.

1.2. Current status of the ESS activities involving ionising radiation

In 2021, there were no operational activities at ESS that could lead to emissions of radioactivity to the environment. In the report [5] the progress work is described in more detail: "During the year 2021 ESS had continued operation of klystrons and began testing of cryomodules in Test Stand 2 as well as testing of klystrons in G02, the so called klystron gallery on level 100 which is next to the accelerator tunnel one floor up. Commissioning of the Normal-Conducting Linac (NCL) started with beam up to Medium-Energy-Beam-Transport (MEBT) Faraday cup, proton beam energy up to 3.6 MeV, in October 2021."

2. METHODOLOGY

2.1. Description of pre-operational sampling programme

The sampling programmes for 2017-2020 are presented in detail in [1-4], covering measurements and analysis of natural and anthropogenic radionuclides, including gamma-emitters, ^{3}H (tritium), ^{14}C, and for 2020 also ^{129}I. Sampling was mainly performed within a few km of the ESS site. The sampling sites have been chosen to cover different wind directions from the ESS site and additional sampling sites and sample types have been added gradually.

Table 1 summarizes the measurements performed 2017-2020 [1-4], and also includes measurements that were performed during 2018 in a project financed by the Swedish Radiation Safety Authority (SSM) [6]. In another project financed by SSM, seaweed was collected along the Swedish coast in 2020 and analysed *e.g.* for gamma and ^{14}C. Some of these data are of relevance to the ESS environmental monitoring programme and have recently been published [7, 8], but were not included in Lund University annual report for radiological environmental monitoring for 2020 [4].

Table 2 lists monitored constituents and frequencies for sampling in 2021. The programme now also includes some sediments samples analysed with gamma spectrometry and seaweed.

Table 1 Monitored parameters (methods) and frequencies of sampling and measurement for zero point assessments.

Discharge	Monitored parameters	Number of sites/frequency for the 2017-2018 report [1]	Annual report year 2018 [2]	SSM report, for year 2018-2019, ref [6]	Annual report year 2019 [3]	Annual report year 2020 [4]
Airborne	*External radiation*					
	In situ γ spectrometry	21 sites				4 sites
	Mobile	Ambient dose equivalent rate at 29 sites. One car assessment				ESS and MaxIV
	Air, deposition					
	Soil, γ-emitting radionuclides	Down to a depth of 20 cm at 22 sites. Down to a depth of 7 cm at 29 sites.				
	Soil, ^{129}I					4 sites
	Foodstuff and/or ingestion					
	Fruits, berries	^{14}C at 12 sites	^{14}C at 10 sites		^{14}C at 6 sites	γ-emitters at 9 sites, ^{3}H at 10 sites and 8 ^{14}C
	Crops	γ-emitters at 12 sites, ^{14}C at 6 sites, ^{3}H at one site.	^{14}C at 2 sites		13 γ and 6 ^{3}H samples	γ-emitters at 16 sites, ^{3}H at 1 site, ^{14}C at 1 sites, 1 sugar beet for γ and ^{3}H
	Milk and forage	γ-emitters, ^{3}H and ^{14}C at one site on one occasion.				1 milk, 1 forage grass for γ, 2 ^{3}H and ^{14}C
	Meat					1 γ, t^{3}H and ^{14}C
	^{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		4 γ, monthly ^{3}H in tap water	
	Terrestrial indicators					
	Grass	γ-emitters at 20 sites, ^{14}C at 12 sites	^{14}C at 8 sites		γ at 6 sites, ^{14}C at 2 sites	9+1 samples for γ, 13 ^{14}C
	Lichen, moss and other bioindicators	γ-emitters at 13 sites, ^{14}C at 12 sites				3 lichen samples for γ (2 also for ^{129}I) analysis, 1 moss for γ and ^{129}I, 10 other bioindicators for γ
	Honey					1 γ and ^{14}C
	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 at ESS site for ^{3}H analysis. Monthly basis, start April 2018. Results until April 2019 in [6].		Precipitation at ESS site for ^{3}H analysis. Monthly basis.
	Air humidity		Grab sampling for ^{3}H analysis. 1 at urban reference site, 2 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 [6].		Grab sampling for ^{3}H at ESS site and urban reference site. Monthly basis.
Liquid	*Water bodies*					
	Groundwater	^{3}H at 12 sites			γ in 4 samples	1 at reference site Grevie (^{3}H)
	Surface water	^{3}H at 8 sites			Appr monthly at 3 ponds, ^{3}H. Källby pond.	Monthly grab sampling of pond water at 3 sites for ^{3}H analysis. 11 ^{3}H in water from streams and rivers
	Tap water					8 samples of Lund tap water (^{3}H)
	Sewage sludge	γ-emitters and ^{3}H at Källby: Monthly samples from April 2017 – April 2018.	γ-emitters and ^{3}H at Källby: two occasions		γ-emitters and ^{3}H in 2 samples	13 samples for γ and 11 for ^{3}H
	Seaweed					SSM project SSM2019-5225 [7]

Table 2 Monitored parameters (methods) and frequencies of sampling and measurement for zero point assessments 2021.

Discharge	Monitored parameters	Number of sites/frequency
Airborne	*External radiation*	
	In situ gamma spectrometry	3 sites (summer and winter)
	Mobile	1 assessment (summer) 1.5-2 km from ESS
	Air, deposition	
	Soil	N/A (to be continued during ESS operation)
	Fruits, berries	7 samples for gamma and ^{14}C analysis, 5 samples for ^{3}H analysis
	Crops	15 samples for gamma analysis, 2 samples for ^{3}H analysis
	Milk and forage	2 milk samples for gamma, ^{3}H and ^{14}C analysis
	Meat	N/A (to be continued during ESS operation)
	Grass	9 samples for gamma, 3 samples for ^{14}C analysis
	Honey	2 samples for gamma and 1 sample for ^{3}H analysis
	Lichen, moss and other bioindicators	2 samples of lichen, 2 samples of moss, 1 sugar beet for gamma analysis
	Precipitation and air	
	Precipitation	Continuous sampling of precipitation at ESS site for ^{3}H analysis. Monthly basis.
	Air humidity	Grab sampling for ^{3}H analysis at ESS site and urban reference site. Monthly basis.
Liquid	*Water bodies*	
	Groundwater	3 samples for gamma analysis, 1 sample from Grevie (background)
	Surface water	Monthly grab sampling of pond water at 3 sites (ESS, MaxIV and 1 urban reference site) for ^{3}H analysis. 3 samples for ^{3}H in water from streams and rivers
	Tap water	12 samples of Lund tap water (^{3}H)
	Sewage sludge	8 samples for gamma and ^{3}H analysis
	Seaweed	2 samples for gamma and ^{14}C analysis
	Sediment	1 test sample for gamma, site 31.6 (ESS pond 4), ~5 months sampling with bottom trap

2.2. Sampling locations

Sampling sites and sample/measurement types for 2021 are listed in Table 3.

Table 3 Sampling locations and type of measurements performed during year 2021.

Site	Location	Latitude	Longitude	Gamma	^{3}H	^{14}C
1	Västra Odarslöv 341	N55.7431	E13.2477	apple	apple	apple
2	Östra Odarslöv 651	N55.7380	E13.2736	pear, honey	pear	pear
4	Ladugårdmarken 461	N55.7384	E13.2314	groundwater		
6	Möllegården	N55.7304	E13.2441	grass, apple, lichen, moss, *in situ*	apple	grass, apple
14.2	Kävlingeån, Gårdstånga kyrka	N55.7589	E13.3267		surface water	
27.3	ESS SE corner (at stones)	N55.7346	E13.2597	grass, *in situ*		grass
30	Kopparstaden windmill	N55.7385	E13.2543	grass, moss, *in situ*		grass
31.3	ESS area (3)	N55.7298	E13.2436	groundwater		
31.6	ESS area (6) (ESS offical pond 4)	N55.7358	E13.2442	sediment	surface water	
31.20	ESS area (20), weather station	N55.7366	E13.2455		precipitation, air humidity	
32.2	Dammstorpsvägen 16 (field 1)	N55.7279	E13.2574	spring barley		
32.4	Dammstorpsvägen 16 (field 3)	N55.7288	E13.2520	winter wheat		
32.5	Dammstorpsvägen 16 (groundwater 304/305)	N55.7278	E13.2545	groundwater		
34.4	MaxIV area (4), apple tree	N55.7270	E13.2363	apple		apple
34.5	MaxIV area (5), pond	N55.7283	E13.2376		surface water	
35.1	Källby (sewage treatment plant) VA SYD	N55.6952	E13.1638	sewage slude	sewage slude	
36.2	Svenstorp's gods, farmland (2) field "6-0"	N55.7583	E13.2508	rapeseed		
36.3	Svenstorp's gods, farmland (3) field "8-0"	N55.7449	E13.2442	rapeseed		
36.4	Svenstorp's gods, farmland (4) field "5-0"	N55.7515	E13.2397	rapeseed		
36.5	Svenstorp's gods, farmland (5) field "21-0"	N55.7400	E13.2671	spring barley		
36.6	Svenstorp's gods, farmland (6) field "25-0"	N55.7446	E13.2808	spring barley		
36.7	Svenstorp's gods, farmland (7) field "10-0"	N55.7509	E13.2597	winter wheat		
36.8	Svenstorp's gods, farmland (8) field "24-0"	N55.7465	E13.2707	spring barley		
36.9	Svenstorp's gods, farmland (9) field "8-1"	N55.7378	E13.2416	winter wheat		
36.10	Svenstorp's gods, farmland (10) field "22-0"	N55.7422	E13.2594	spring barley		
36.11	Svenstorp's gods, farmland (11) field "9-0"	N55.7445	E13.2529	rapeseed		
36.13	Svenstorp's gods, farmland (13) field "4-0"	N55.7572	E13.2349	rapeseed		
42	Ladugårdsmarken (cell tower)	N55.7347	E13.2283	pear, sugar beet	sugar beet	pear
48	Timjanvägen 5, Lund	N55.7186	E13.1828	apple	air humidity, apple	
52	Professorsgatan 1, Lund	N55.7097	E13.2047			rowan berries
54	Active Biotech	N55.7169	E13.2206			rowan berries
59	Gamla Bjärred, Pilevägen	N55.7071	E13.0334	seaweed		seaweed
60	Skillinge	N55.47	E14.28	seaweed		seaweed
62	Grevie PV5 well	N55.6131	E13.1970		ground water	
63	Monument park	N55.7182	E13.1851		surface water	
64	Södervidinge 302-36	N55.827	E13.098	milk	milk	milk
74.1	Glomsjön, inlet, at bridge, cow's drinking spot	N55.7206	E13.2661		surface water	
76	Höje å, Trolleberg	N55.7022	E13.1439		surface water	
77	Höje å, Lomma kyrka	N55.6878	E13.0781		surface water	
86	Glorias apple farm	N55.7432	E13.2268	apple	apple	
87	Östra Odarslöv 541 (bee keeper)	N55.737793	E13.287011	honey	honey	
89:6	Lundaslättens drift, field F	N55.723129	E13.246233	barley		
89:7	Lundaslättens drift, field G	N55.718712	E13.249172	winter wheat		

The sites for sampling, for measurement with laboratory gamma spectrometry, and sites for *in situ* gamma spectrometry are shown in Figure 1. Site 31.6 (ESS pond 4) was also used for collection of a sediment sample.

Figure 1 Sampling sites for gamma spectrometry measurements for year 2021. See Table 3 for more information. The lower figure is zoomed around the Lund area.

Figure 2 shows the sites for ^{3}H sampling in 2021. As in previous annual reports, sites 48 and 63 are urban reference sites, used for sampling of air humidity and surface water. Water from the deep well “Grevie PV5” (depth 71-72 m) at site 62, operated by VA Syd, was used as background water (a previous study has reported a ^{3}H concentration of about 0.02 TU, corresponding to 0.002 Bq L^{-1} [9]).

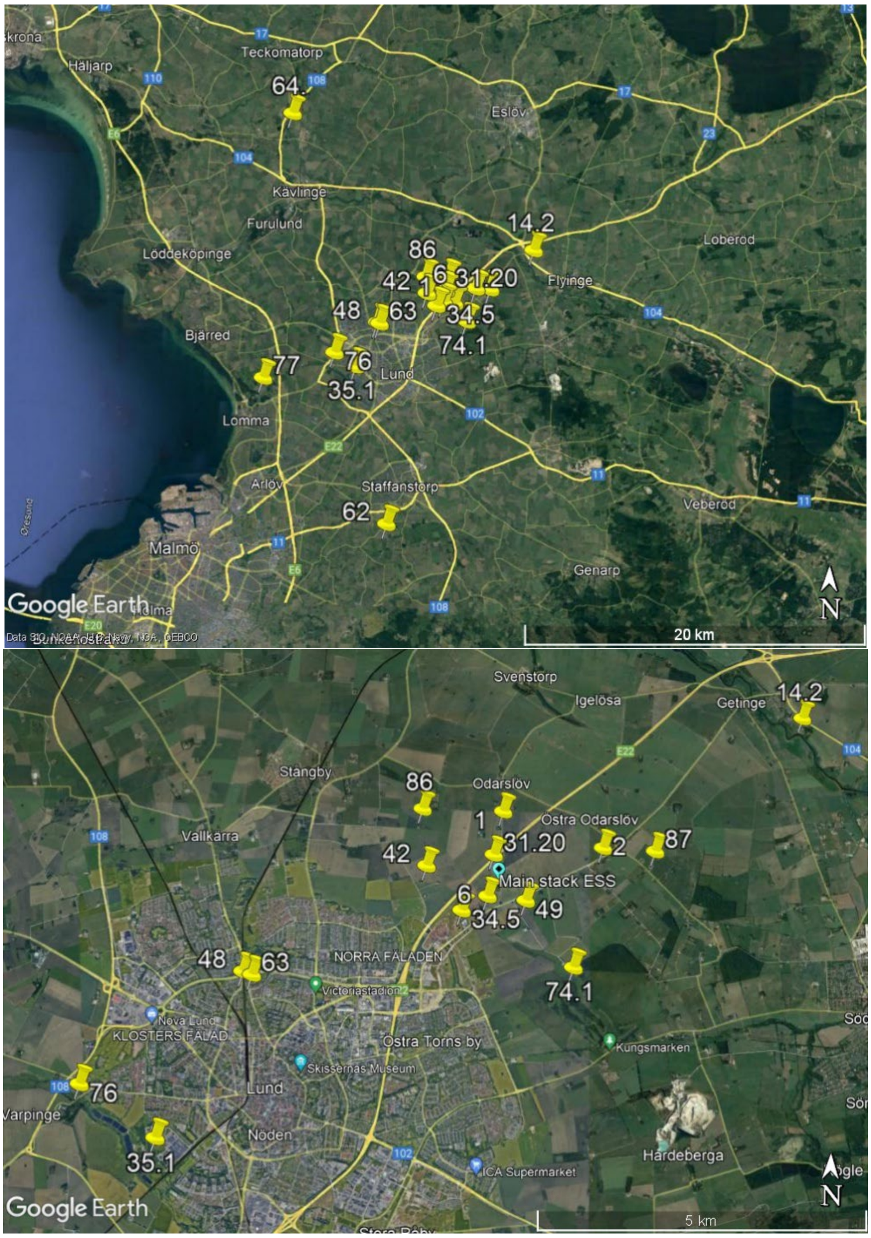

Figure 2 Sampling sites for ^{3}H samples for year 2021. See Table 3 for more information. The lower figure is zoomed around the Lund area.

Samples collected for ^{14}C analysis are shown in Table 4, which also indicates which of the samples have been analysed so far. Samples not analysed are available for future analysis.

Table 4 Sampling locations for ^{14}C samples in 2021.

Analysed	Id	Site nr	Site name	Date	Species
	E14C_271:6	6	Möllegården	2021-06-01	*Poaceae sp.* (grass)
	E14C_272:27.3	27.3	Kärrpengavägen bus stop, tram garage	2021-06-01	*Poaceae sp.* (grass)
	E14C_273:30	30	Kopparstaden wind mill, ESS 1st wind power tower	2021-06-01	*Poaceae sp.* (grass)
X	E14C_274:64	64	Södervidinge 302-36	2021-06-14	Milk
X	E14C_275:60	60	Skillinge	2021-08-12	*Fucus vesiculosus* (brown algae)
	E14C_276:60	60	Skillinge	2021-08-12	*Poaceae sp.* (grass)
	E14C_277:88	88	Åhus Revhaken	2021-08-13	*Fucus vesiculosus*
	E14C_278:88	88	Åhus Revhaken	2021-08-13	*Poaceae sp.* (grass)
	E14C_279:88	88	Åhus Revhaken	2021-08-13	*Pyrus communis* (pear)
	E14C_280:48	48	Timjanv 5, Lund	2021-08-27	*Malus domestica* (apple)
X	E14C_281:27.3	27.3	Kärrpengavägen bus stop, tram garage	2021-08-27	*Poaceae sp.* (grass)
X	E14C_282:6	6	Möllegården	2021-08-27	*Poaceae sp.* (grass)
X	E14C_283:6	6	Möllegården	2021-08-27	*Malus domestica* (apple)
X	E14C_284:30	30	Kopparstaden wind mill, ESS 1st wind power tower	2021-08-27	*Poaceae sp.* (grass)
X	E14C_285:34.4	34.4	MaxIV	2021-08-27	*Malus domestica* (apple)
X	E14C_286:42	42	Ladugårdmarken cell tower	2021-08-27	*Pyrus communis* (pear)
X	E14C_287:2	2	Östra Odarslöv 651	2021-08-27	*Pyrus communis* (pear)
X	E14C_288:54	54	Active Biotech	2021-08-31	*Sorbus aucuparia* (rowan berries)
X	E14C_289:52	52	Professorsgatan 1	2021-08-31	*Sorbus aucuparia* (rowan berries)
X	E14C_290:1	1	Västra Odarslöv 341	2021-08-31	*Malus domestica* (apple)
X	E14C_291:59	59	Gamla Bjärred	2021-09-02	*Fucus vesiculosus* (brown algae)
X	E14C-292:64	64	Södervidinge 302-36	2021-10-29	Milk

The sites presented in Figure 3 are those sites, from which samples have been analysed for ^{14}C in 2021 (marked with x in Table 4).

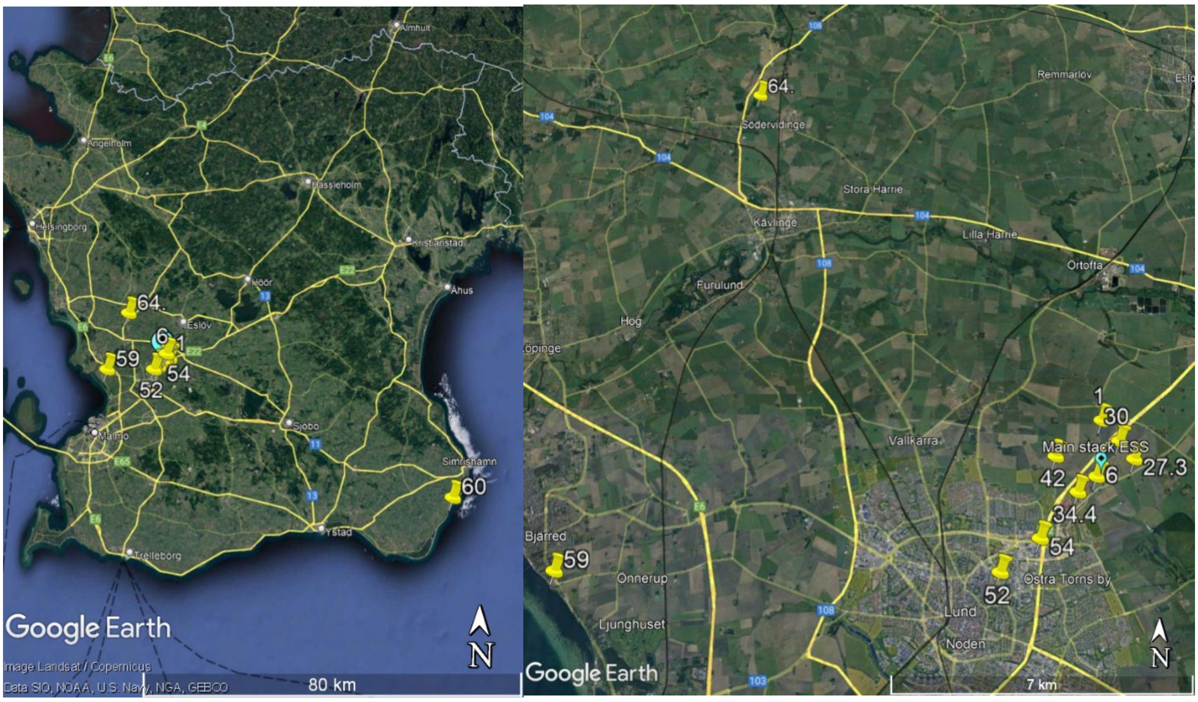

Figure 3 Sampling sites for ^{14}C samples for year 2021. See Table 4 for more information.

2.3. Methods for measurements, sample collection and analysis of gamma emitting radionuclides

The collection and sample preparation procedures are described in Refs [1, 6]. Two sediment samples were collected using a trap (5 L plastic bucket) located at the pond bottom (Site 31.6) between 22 June and 10 November 2020, and between 12 November 2020 and 16 September 2021. The two samples were pooled into one before gamma spectrometric analysis. This sediment sample is the first of this sample type within the ESS zero-point assessments, and part of development work for establishing sampling procedures for sediment sampling. A separate report will describe the method and sampling procedure when fully developed.

In 2021, two samples of seaweed (*Fucus vesiculosus*) were collected, on the west coast outside Lomma (Site 59) and on the east coast outside Skillinge (Site 60), for analysis of gamma emitting radionuclides. The sample preparation of the seaweed was done the same as for other similar samples [8, 10]. First, the seaweed was dried in a heating cabinet (a couple of hours at 70 °C), after which the samples were grinded and put into 200 ml beakers that were analysed in lead shielded HPGe detectors (acquisition time 24-72 h).

2.4. Method for *in situ* and mobile gamma spectrometry around ESS

The methods for *in situ* and mobile gamma spectrometry are described in Ref [1, 6]. *In situ* gamma spectrometry was repeated at three specific sites (same as used for grass sampling) close to the ESS, in different directions: south, east, and north, corresponding to Sites 6, 27.3 and 30, respectively. The acquisition times were at least 40 minutes. As during previous years of the zero point program, a car-borne mapping of the gamma dose rate (including spectrometry) in terms of spectrum dose index (SDI, see [11] for definition) was carried out during the summer 2021 on the available roads within 1.5-2 km around the ESS.

2.5. Methods for tritium and ^{14}C

Sample collection, preparation, measurement and analysis of tritium was identical to the one described in the annual report for 2019 [3]. Sample collection, preparation, measurement, and analysis of ^{14}C was also performed according to previous reports [1-4].

3. RESULTS

3.1. Activity concentration of gamma emitting radionuclides in various types of samples year 2021

For various reasons it was not possible to sample sewage sludge at Källby treatment facility (Site 35) each month during the year. However, the samples collected cover the different seasons of the year and for the natural radionuclides there are no obvious trends over the seasons. For ^{177}Lu (patients from SUS Lund), however, there are slightly higher concentration levels in May-June, about twice as high, as compared to the rest of the year (on average 3.3 kBq kg^{-1} vs 1.4 kBq kg^{-1}). This may be due to a higher number of patients for these months in 2021, but that needs to be verified by the administered activity at SUS Lund. No similar trend is observed for ^{131}I (patients from SUS Lund). It should be noted that there are many factors influencing the measured radioactivity concentration of hospital radionuclides. Therefore, these values should be viewed as a confirmation of the presence rather than the exact concentration in the samples each month. The results of the gamma analysis of sewage sludge are presented in Table A1.1.

Among the 15 samples of crops collected from the ESS nearby farmlands, the activity concentration of the studied radionuclides were below the minimum detectable activity concentration (MDA) for the measurement time used, except for ^{40}K. The ^{40}K activity concentration levels are rather low and slightly higher for rapeseeds than for barley and wheat, on average 311 Bq kg^{-1} and 174 Bq kg^{-1}, respectively. The results of the gamma analysis of crops are presented in Table A1.2.

Nine samples of grass were analysed, three samples from three different sites (two samples during the summer and one during winter, per site). Only ^{40}K was above the MDA. The detected activity concentration in August was about half the value detected in June and January (for sites 6 and 30). This was not observed at Site 27.3, with another type of grass, where the ^{40}K levels were relatively constant. The results of the gamma analysis of grass are presented in Table A1.3.

Among the fruits, bioindicators and the groundwaters analysed, unexpected levels of radioactivity were not observed. The results of the gamma analysis of these samples are presented in Tables A1.4, A1.5 and A1.6.

As for the two types of samples collected for the first time in this zero point program, seaweed and sediment, it was confirmed that seaweed is a good indicator for radioactivity in water (including ^{137}Cs). The sediment sample was a first test of this sample type and showed detectable levels for ^{226}Ra, ^{228}Ac and ^{40}K, but not for ^{137}Cs for the measurement time used. The results of the gamma analysis of seaweed are presented in Table A1.5 and the sediment sample in Table A1.7.

3.2. *In situ* gamma spectrometry around ESS

Due to problems with the germanium detectors in 2021, two *in situ* gamma spectra were acquired during the summer instead of three. Additionally, three spectra were acquired during the winter period, but in 2022. Small levels of ^{137}Cs were observed at the sites (when using the larger HPGe detector and at Site 27.3 when using the smaller HPGe) but no unexpected levels of radioactivity were noted. The results of the *in situ* gamma spectrometry are presented in Table A1.8.

The average SDI dose rate from the 5600 measurement points during the mobile mapping (Figure 4) was 0.06 µSv h^{-1} with a standard deviation of the mean of 19%. It should be noted that the absolute value of the SDI dose rate is based on an internal calibration at one measurement point, and should hence not be observed as exact for each position. The measurement data should rather be viewed in terms of the variation over the mapped area, which for the data in Figure 4, corresponds to a span between 0.036-0.132 μSv h^{-1} (with a median value of 0.053 μSv h^{-1}).

In Figure 4 it can be observed that the radiation background along the small gravel and asphalt roads surrounded by fields with crops is low and homogenous. Areas with slightly elevated levels were close to construction sites or under bridges.

Figure 4 Dose rate map (Google Earth, April 2021) in terms of SDI-dose rate (μSv h^{-1}) around ESS as measured by a car borne 2×3 litres NaI(Tl) detector system on 2021-06-14.

3.3. ^{3}H analysis year 2021

The results of the tritium measurements of samples of precipitation, air humidity, surface water, Lund tap water and streams collected in 2021 are shown in Tables A1.9-A1.14 in Appendix 1. Results from measurements of sewage sludge, fruits and crops are presented in Table A1.15. Two measurements on milk samples from the dairy farm at site 64 are reported in Table A1.16.

For the majority of the samples, the measured activity concentration was below the MDA of 1.62 Bq l^{-1}. Slightly elevated levels of tritium were measured in all the monitored ponds in August and September and in some of them in October and December. It corresponds to the warmest and coldest periods of the year. Tritiated water has a higher melting and boiling point than water. It induces a fractionation of tritium when evaporation is high or when the temperatures are below 0°C. Thus, values superior to the 1.62 Bq L^{-1} detection were measured but they are still in the expected range of environmental levels. Similar observations can be done for air humidity and precipitation samples.

Like the previous years, several food products and sewage sludge samples show slightly elevated levels of tritium. In some of them like honey, grain or sewage sludge, it could be attributed to the partial drying of the product that induces tritium fractionation. All the measured activity concentrations of tritium in the samples are at expected environmental levels.

3.4. ^{14}C analysis year 2021

The results of the ^{14}C analysis of 12 terrestrial samples (grass, fruits, berries and milk) are presented in Table A1.17 in Appendix 1. Marine sample results are presented in Table A1.18 in Appendix 1. The results are expressed as $F^{14}C$ [12, 13], see Ref [1] (p. 92-94) for definition and conversion to other activity concentration units[1].

Figure 5 shows $F^{14}C$ at the various sites. The average $F^{14}C$ value of the terrestrial ^{14}C samples analysed for year 2021 samples was 1.008 (STD: 0.002; SUM:< 0.001). The corresponding specific activity is 226 Bq kg^{-1} C using $\delta^{13}C$= - 25 ‰ (see Annex B4 in Ref [1]). According to Grubb's test, the milk sample from June (ESS_14C_274:64, $F^{14}C$ = 1.012 ± 0.005) is a significant outlier (at the 0.05 level). However, all terrestrial data overlap within 1 σ of the analytical uncertainty, and hence this data point is not to be considered as an outlier.

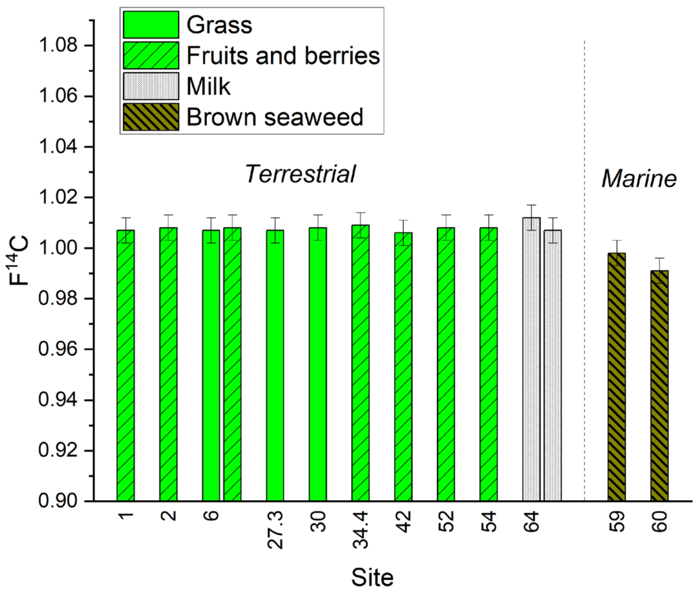

Figure 5 Results of the ^{14}C measurements from 2021. Uncertainties represent 1 standard deviation.

Figure 6 shows the average $F^{14}C$ values obtained in for all ESS zero point assessments so far (terrestrial samples), 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 (N56.0976, E13.4189, 115 m above sea level, sampling height 150 m above

[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.

ground)[2] [14-18]. $F^{14}C$ values in atmospheric CO_2 (remains of bomb-^{14}C) are still decreasing (see section 3.4 in Ref [3]).

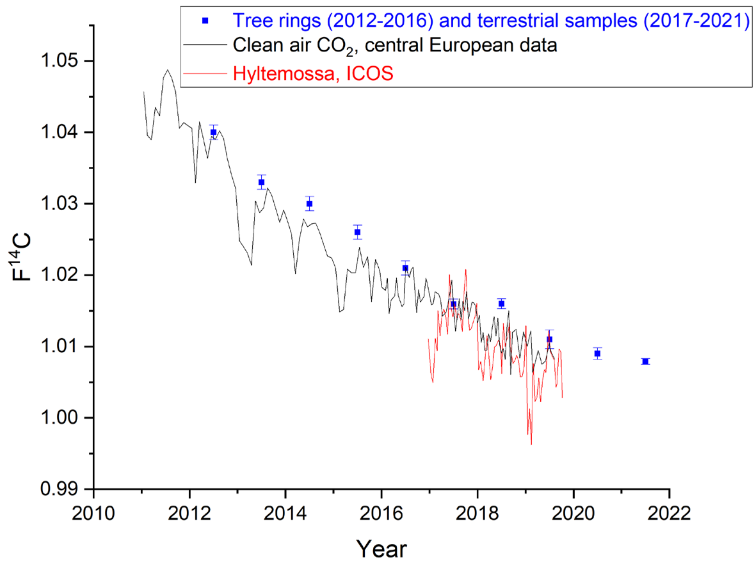

Figure 6 Average $F^{14}C$ values (uncertainty represented by the SUM) obtained for all ESS zero point assessments so far (terrestrial samples), along with ^{14}C data in atmospheric CO_2 collected at rural background stations in central Europe and at the Swedish ICOS station Hyltemossa [14-18].

As shown in Figure 5 (and Table A1.18 in Appendix 1) two samples of brown seaweed (*Fucus vesiculosus*) had $F^{14}C = 0.998 \pm 0.005$ at the west coast (site 59. Gamla Bjärred) and $F^{14}C = 0.991 \pm 0.005$ at the east coast (site 60. Skillinge, marine reference site). In a recent study [7], we analysed ^{14}C in seaweed samples collected along the Swedish coast in 2020. Samples from the west coast as well as from the east coast were analysed, including samples from site 59 and site 60. In general, $F^{14}C$ varies significantly more in the marine environment than in the terrestrial environment, partly due to a slower mixing in the oceans than in the atmosphere (see also [7]). Figure 7 presents $F^{14}C$ for sites 59 and 60 for 2020 [7] and for 2021. The variations in $F^{14}C$ between the different years is not unexpected (see [7]) due inhomogeneity of $F^{14}C$ in the marine environment.

[2] ICOS ^{14}C data ($\Delta^{14}C$) has been recalculated 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.

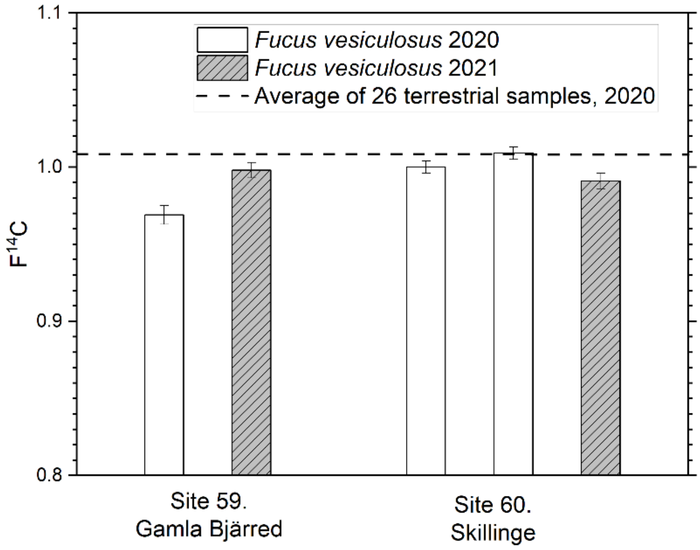

Figure 7 $F^{14}C$ in *Fucus vesiculosus* collected at site 59 (Gamla Bjärred) and site 60 (Skillinge) in 2020 [7] and 2021.

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 scintillation cocktail provider Perkin Elmer [19].

The laboratory participates in the annual IAEA intercomparison tests for gamma spectrometry and in 2019 we also participated in the PROCORAD intercomparison test. Our results have, in general, been satisfying during the last years and we are confident in our secure methods for sample preparation, measurements and evaluation. Detailed information about the QA for gamma spectrometry can be seen in previous reports [1-3].

As previously reported, 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

Among the regular samples collected and analysed for activity concentration of gamma emitting radionuclides, increased levels of anthropogenic radioactivity were only observed in samples of sewage sludge. The increased levels of ^{177}Lu and ^{131}I in sewage sludge is due to radiation therapies of patients at SUS Lund. Furthermore, some of the samples collected were in too small amounts, for the standard measurement time (2-3 days), to achieve acceptable MDA levels. When such small amounts are measured in the future, a longer acquisition time will be used. In 2021, samples of seaweed and one sample of (pond) sediment were included and analysed for gamma emitting radionuclides. These both sample types are believed to be important indicators in the future, during operation of ESS, and showed normal levels of radionuclides in 2021. In seaweed, Chernobyl ^{137}Cs was also observed.

During the summer 2021 a new bicycle path was constructed along Odarslövsvägen. During that construction the groundwater well at site number 31.3 (ESS area (3)) was removed.

For the majority of the samples, the measured activity concentrations of tritium were below the minimum detectable activity (MDA) of 1.62 Bq l^{-1} or at expected environmental levels.

The $F^{14}C$ data in terrestrial samples collected in the Lund area in 2021 are not believed to reflect any local anthropogenic contamination. The preoperational data for ^{14}C has now been extended into the marine environment through measurements from sites relevant to discharges from the ESS (Site 59. Gamla Bjärred), and from a reference site on the east coast of Scania (Site 60. Skillinge).

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) express their gratitude to the individuals, families and companies that have helped with providing access to the sites investigated and samples collected during this programme. Thanks to Mattias Olsson for assistance in sample preparation for the ^{14}C samples.

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. 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.* Report MA RADFYS 2020:02, Report BAR 2020/02. Lund Univeristy. 2020.
3. Bernhardsson, C., Eriksson Stenström, K., Pedehontaa-Hiaa, G., Jönsson, M. *Radiological environmental monitoring at the ESS facility – Annual report 2019.* Report MA RADFYS 2020:03, Report BAR-2020/03. 2020.
4. Bernhardsson, C., Eriksson Stenström, K., Jönsson, M., Pedehontaa-Hiaa, G., Mattsson, S. *Radiological environmental monitoring at the ESS facility – Annual report 2020.* Report MA RADFYS 2021:01, Report BAR 2021/01. 2021.
5. Johansson, L. *ESS annual report to SSM.* ESS internal report (ESS-3246455), 2022, 10 p. 2022.
6. 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.
7. Eriksson Stenström, K., Mattsson, S. *Spatial and temporal variations of ^{14}C in Fucus spp. in Swedish coastal waters.* Journal of Environmental Radioactivity Volume 242, February 2022, 106794, 2022.
8. Eriksson Stenström, K., Mattsson, S. *Project SSM2019-5225: "Marine 14C levels around the Swedish coast" – Additional gamma spectrometry measurements and ICP-MS analysis of brown algae (Fucus spp.).* BAR 2021/02. MA RADFYS 2021/02. https://portal.research.lu.se/en/publications/project-ssm2019-5225-marine-14c-levels-around-the-swedish-coast-a. 2021.
9. 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.
10. Mattsson, S., Eriksson Stenström, K., Pédehontaa-Hiaa, G. *Long-time variations of radionuclides and metals in the marine environment of the Swedish west-coast studied using brown algae (Fucus serratus and Fucus vesiculosus).* Report to the Swedish Radiation Safety Authority (SSM). 2022.
11. Karlberg, O. *Manual och teknisk beskrivning av CEMIK-systemet och NuggetW version 3.2.* 2007.
12. 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.
13. Eriksson Stenström, K., Skog, G., Georgiadou, E., Genberg, J., Johansson, A. *A guide to radiocarbon units and calculations.* LUNFD6(NFFR-3111)/1-17/(2011). Lund University. Dep of Physics. Div of Nuclear Physics. http://lup.lub.lu.se/search/ws/files/5555659/2173661.pdf. 2011.
14. Levin, I., Kromer, B. *The tropospheric $^{14}CO_2$ level in mid latitudes of the Northern Hemisphere (1959-2003).* Radiocarbon, 46: 1261-1271, 2004.
15. Levin, I., Kromer, B., Hammer, S. *Atmospheric $\Delta^{14}CO_2$ trend in Western European background air from 2000 to 2012.* Tellus B: Chemical and Physical Meteorology, 65(1): 20092, 2013.
16. Hammer, S., Levin, I. *Monthly mean atmospheric $D^{14}CO_2$ 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.
17. 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-IXPKZIO4DWX7wncsLQ7akY; http://calib.org/CALIBomb/. 2019.
18. ICOS. *ICOS ATC ^{14}C release from Hyltemossa 2016-12-22 – 2019-10-08.* https://data.icos-cp.eu/portal/. PID: 11676/WE07MZ3UIYcJau76jaTIW_Sg. ICOS DATA is licensed under CC4BY (http://creativecommons.org/licenses/by/4.0/). 2020.
19. 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. DATA FROM MEASURENTS 2021

Available upon request from the authors