Radiological environmental monitoring at the ESS facility – Annual report 2020

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

2021

Document Version:
Publisher's PDF, also known as Version of record

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 2020

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
Sören Mattsson	soren.mattsson@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 2021:01
Malmö 2020

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

Report BAR-2021/01
Lund 2020

SUMMARY

Results from the radiological environmental monitoring of the European Spallation Source (ESS) are presented for year 2020.

Among the 63 environmental samples analysed for gamma emitting radionuclides, no elevated levels of anthropogenic radioactivity were observed. All samples (except two samples of lichen) had activity concentrations below the MDA for the natural radionuclides as well as ^{137}Cs, and normal levels of ^{40}K. In sewage sludge, detectable levels of ^{177}Lu and ^{131}I were observed in samples that were measured within one week after sampling.

Mobile gamma spectrometry surveys were carried out around ESS and MAX IV (when operating at high frequency during a test run). No deviating radiation levels were observed.

The samples analysed for their ^{3}H activity concentration (120 samples) were generally below the detection limit using the current procedure, instrumentation and analysis time (MDA typically 1.6 Bq L^{-1}). The results show no evidence of any local contamination of ^{3}H in Lund during 2020.

The ^{14}C data in the 26 samples of grass, fruits, berries, crops, milk, honey and meat of in the Lund area and in southern Sweden is consistent with the declining ^{14}C specific activity in atmospheric CO_2. Seasonal variations were observed in the ^{14}C data. No evidence of anthropogenic ^{14}C contamination in the Lund area was noted during 2020.

The report presents the first base-line measurements of ^{129}I (10 samples) in the ESS environmental monitoring programme. The ^{129}I activity concentrations ranged between $(3.18 \pm 0.20)\cdot10^{11}$ atoms/kg d.w. for grass and $(1.31 \pm 0.03)\cdot10^{13}$ atoms/kg d.w. for moss. These values are in the expected range.

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

The radiological environmental monitoring of the pre-operation phase of the European Spallation Source (ESS) started in 2017 with an extensive programme [1], which was followed up with additional yearly investigations in 2018-2019 [2, 3]. Results of the radiological environmental monitoring of ESS during year 2020 are presented in the present report. ESS was during 2020 still in its pre-operational phase.

1.1. Aim

The aim of the current report is to provide continued preoperational radiological environmental data for ESS for year 2020.

2. METHODOLOGY

2.1. Description of pre-operational sampling programme

The design of sampling programmes for 2017-2019 is presented in [1-3]. The programme covered assessment of natural and anthropogenic radionuclides, including gamma-emitters and the pure beta-emitters ^{3}H (tritium) and ^{14}C. For year 2020, analysis of ^{129}I has been included for some samples of soil, grass, lichen and moss.

The majority of the sampling sites for the environmental monitoring programme are located within a few km of ESS. The sites have been chosen to cover different wind directions from the ESS site and additional sampling sites have been added gradually.

Table 1 summarizes the measurements performed 2017-2019 [1-3] and also includes measurements that were performed during 2018 in a project financed by the Swedish Radiation Safety Authority (SSM) [4]. Table 2 lists monitored constituents and frequencies for sampling for year 2020. The dairy farm where e.g. milk samples were collected for years 2017-2018 [1-3] was closed in 2020. Thus, a new dairy farm was chosen for the pre-operational sampling programme. Meat from this dairy farm was also added to the programme. Another sample type that has not been included in previous years includes the bioindicator honey. Surface waters from Höje river, Kävlinge river and Vombsjön were included for the first time (analysed for ^{3}H). In a project financed by the Swedish Radiation Safety Authority (SSM), seaweed has been collected along the Swedish coast and analysed e.g. for gamma and ^{14}C. Some of these data are of relevance to the ESS environmental monitoring programme and will be published in international journals and a report to SSM during 2021. The results from the seaweed measurements for 2020 will be summarized in the upcoming annual report for 2021.

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 [4]	Annual report year 2019 [3]
Airborne	*External radiation*				
	In situ gamma spectrometry	21 sites			
	Mobile	Ambient dose equivalent rate at 29 sites. One car assessment			
	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		13 gamma and 6 ^{3}H 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		4 gamma samples, monthly ^{3}H in tap water
	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 at ESS site for ^{3}H analysis. Monthly basis, start April 2018. Results until April 2019 in [4].	
	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 [4].	
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

Table 2 Monitored parameters (methods) and frequencies of sampling and measurement for zero point assessments 2020.

Discharge	Monitored parameters	Number of sites/frequency
Airborne	*External radiation*	
	In situ gamma spectrometry	At 4 sites
	Mobile	Once around the ESS and once around MaxIV
	Air, deposition	
	Soil	4 samples for ^{129}I analysis
	Foodstuff and/or ingestion	
	Fruits, berries	9 samples for analysis of gamma, 10 for ^{3}H and 8 for ^{14}C
	Crops	1 sample for ^{14}C analysis 16 samples for gamma analysis 1 sample for ^{3}H analysis 1 sugar beet for gamma and ^{3}H analysis
	Milk and forage	1 milk and 1 forage grass sample for gamma, and 2 samples for tritium and ^{14}C analysis
	Meat	1 sample for gamma, tritium and ^{14}C analysis
	Terrestrial indicators	
	Grass	9+1 samples for gamma analysis 13 samples for ^{14}C analysis 4 samples for ^{129}I analysis
	Honey	1 sample for gamma and ^{14}C analysis
	Lichen, moss and other bioindicators	3 lichen samples for gamma (2 samples also for ^{129}I) analysis 1 sample of moss for gamma and ^{129}I analysis 10 other bioindicators for gamma
	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*	
	Ground water	1 at reference site Grevie
	Surface water	Monthly grab sampling of pond water at 3 sites (ESS, MaxIV and 1 urban reference site) for ^{3}H analysis. 11 samples for ^{3}H in water from streams and rivers
	Tap water	8 samples of Lund tap water
	Sewage sludge	13 samples for gamma and 11 for ^{3}H analysis
	Seaweed	Measured within SSM project SSM2019-5225, to be published

2.2. Sampling locations

Sampling sites and sample/measurement types for year 2020 are listed in Table 3 (the previous sample site notations used in Ref [1] for ^{3}H and ^{14}C are included in Table 3). The dairy farm Södervidinge 302-36 (site 64), replacing the phased-out farm in Ref [1], is located 14 km NW (318°) from ESS. Other new sites in Table 3 are sites 74-84, used for extended measurements for surface waters of relevance to ESS, and site 86.

Table 3 Sampling locations and type of measurements performed during year 2020.

Site	Location	Latitude	Longitude	Old site nr ^{14}C	Old site nr ^{3}H	Gamma	^{3}H	^{14}C	^{129}I
1	Västra Odarslöv 341	N55.7431	E13.2477	C5		apple	apple	apple	
2	Östra Odarslöv 651	N55.7380	E13.2736	C25		honey, apple	apple	honey, apple	
4	Ladugårdmarken 461	N55.7384	E13.2314	C6		grass			
5	Switchgear (SW of ESS)	N55.7347	E13.2418			rowan berries, fern, HPGe	rowan berries		
6	Möllegården	N55.7304	E13.2441	C3		grass, pine needles, moss, lichen, sun flower, apple, HPGe	apple	grass	soil, grass, moss
7	Odarslövs kyrka	N55.7509	E13.2482			yellow lichen, gray lichen			
13	Östra Odarslöv 264	N55.7362	E13.2760			grass			
14.2	Kävlingeån, Gårdstånga kyrka	N55.7589	E13.3267				surface water		
27.3	ESS SE corner (at stones)	N55.7346	E13.2597	C12		grass, herb, HPGe		grass	soil, grass
30	Kopparstaden windmill	N55.7385	E13.2543	C30		grass, HPGe		grass	soil, grass
31.3	ESS area (3)	N55.7298	E13.2436						
31.6	ESS area (6) (ESS offical pond 4)	N55.7358	E13.2442		T2b		surface water		
31.20	ESS area (20), weather station	N55.7366	E13.2455		T2a		precipi-tation, air humidity		
32.1	Dammstorpsvägen 16	N55.7284	E13.2556	C10					soil
32.2	Dammstorpsvägen 16 (field 1)	N55.7279	E13.2574			sugar beet	sugar beet		
32.4	Dammstorpsvägen 16 (field 3)	N55.7288	E13.2520			wheat		wheat	
32.5	Dammstorpsvägen 16 (ground water 304/305)	N55.7278	E13.2545						
32.6	Dammstorpsvägen 16 (field 4)	N55.7304	E13.2515			barley			
34.4	MaxIV area (4), apple tree	N55.7270	E13.2363	C29		pear	pear	pear	
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 sludge	sewage sludge		
36.2	Svenstorp's gods, farmland (2) field "6-0"	N55.7583	E13.2508			winter barley			
36.3	Svenstorp's gods, farmland (3) field "8-0"	N55.7449	E13.2442			winter barley			
36.4	Svenstorp's gods, farmland (4) field "5-0"	N55.7515	E13.2397			winter barley			
36.5	Svenstorp's gods, farmland (5) field "21-0"	N55.7400	E13.2671			barley			
36.6	Svenstorp's gods, farmland (6) field "25-0"	N55.7446	E13.2808			barley			

36.7	Svenstorp's gods, farmland (7) field "10-0"	N55.7509	E13.2597			wheat			
36.8	Svenstorp's gods, farmland (8) field "24-0"	N55.7465	E13.2707			barley			
36.9	Svenstorp's gods, farmland (9) field "8-1"	N55.7378	E13.2416			wheat			
36.10	Svenstorp's gods, farmland (10) field "22-0"	N55.7422	E13.2594			barley			
36.11	Svenstorp's gods, farmland (11) field "9-0"	N55.7445	E13.2529			barley			
36.12	Svenstorp's gods, farmland (12) field "1-0"	N55.7616	E13.2423			rape seed	rape seed		
36.13	Svenstorp's gods, farmland (13) field "4-0"	N55.7572	E13.2349			winter barley			
36.15	Svenstorp's gods, farmland (15) field "28-0"	N55.7516	E13.2855			barley			
36.16	Svenstorp's gods, farmland (16) field "29-0"	N55.7568	E13.2936			barley			
42	Ladugårdsmarken (cell tower)	N55.7347	E13.2283			pears	pears	apples	
47	Borrby	N55.4256	E14.2236	C1 (rural ref)				grass, rowan berries	
48	Timjanvägen 5, Lund	N55.7186	E13.1828	C2 (urban ref)	T1a	apple	apple, tap water, air humidity	apple	
54	Active Biotech	N55.7169	E13.2206					rowan berries	
60	Skillinge	N55.47	E14.28	C1c				grass,	
62	Grevie PV5 well	N55.6131	E13.1970		T0		ground water		
63	Monument park	N55.7182	E13.1851		T1b		surface water		
64	Södervidinge 302-36	N55.827	E13.098	C32		grass, meat, milk	grass, meat, milk	grass, meat, milk	grass
74.1	Glomsjön, inlet, at bridge, cow's drinking spot	N55.7206	E13.2661				surface water		
75	Farmland SW of ESS	N55.7318	E13.2429			herbs			
76	Höje å, Trolleberg	N55.7022	E13.1439				surface water		
77	Höje å, Lomma kyrka	N55.6878	E13.0781				surface water		
78	Höje å, Lomma hamn	N55.6756	E13.0578				surface water		
79	Lödde å, Strömnäsvägen	N55.7357	E13.0050				surface water		
80	Lödde å, Högs mölla	N55.7798	E13.0755				surface water		
81	Kävlingeån, Håstad	N55.7769	E13.2350				surface water		
82	Kävlingeån, Flyinge kungsgård	N55.7500	E13.3550				surface water		
83	Kävlingeån, Revingeby	N55.7299	E13.4670				surface water		
84	Kävlingån, Vombbron	N55.6986	E13.5543				surface water		
86	Glorias apple farm	N55.7432	E13.2268			apple	apple	apple	

The sites for sampling, for measurement with gamma spectrometry, and *in situ* gamma spectrometry are shown in Figure 1. The sites were selected based on the assessments during previous years and the majority of the samples were analysed for ^{3}H as well. At the sites 5, 6, 27.3 and 30, *in situ* gamma spectrometry was performed in parallel to the sampling of grass (2020-08-05).

Figure 1 Sampling sites for gamma spectrometry measurements (and *in situ* gamma spectrometry at sites 5, 6, 30, 27.3) for year 2020. See Table 3 for more information.

The sites for ^{3}H sampling in 2020 are shown in Figure 2. Reference/background sites for ^{3}H were the same as in previous reports [2, 3]:

- **Sites 48 and 63, located ~4.3 km from ESS (~246°).** Rural reference sites in northern Lund for sampling of air humidity and surface water, respectively. Some of the tap water samples (tap water from VA Syd) were also collected at site 48.
- **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} [5]).

Figure 2 Sampling sites for ^{3}H samples for year 2020. See Table 3 for more information.

The sites for collecting samples for ^{14}C analysis for year 2020 are shown in Figure 3. Two terrestrial reference sites were included: the rural reference site Borrby (site 47), located 70 km ESE from the ESS site, and the urban reference site in northern Lund (site 48), located 4.6 km (WSW) from the ESS site. Skillinge was chosen as marine reference site. This site is located close to the rural reference site Borrby (site 47).

Figure 3 Sampling sites for ^{14}C samples for year 2020. See Table 3 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, 4]. Weather data was obtained from a Davis Vantage Pro2 weather station located at the ESS site 31.20 (see Ref [4] for details).

In 2020, gamma spectrometry was carried out around ESS by car (with two NaI(Tl)) and by stationary measurements, so called *in situ* spectrometry, using an HPGe detector. Two mobile (car) measurements were performed in august 2020 (during dry weather), at a speed of about 30-40 km/h (except on the E22 motorway where the speed was 100 km/h). During the same month, *in situ* high resolution gamma spectrometry was carried out at four selected locations just outside the fenced area of ESS. Detailed gamma spectrometry and ambient dose rate studies were also carried out around MAX IV when testing the cyclotron at a high operational frequency (10 Hz into the rings, which is the highest intended injection frequency for the facility, normally operating at 2 Hz) for detecting any potential increase in the background radiation level by *e.g.* skyshine.

During 2020, various types of samples were collected for analysis of gamma emitting radionuclides. This include samples of grass (n=10) at selected locations in spring/autumn, fruits and berries (n=9) growing around ESS, various types of crops from the nearby fields (n=17), various types of bioindicators that can be found in the nearby area (n=14, including ground beef, milk and honey). Samples of sewage sludge from Källby sewage treatment plant (VA SYD) in Lund (n=13) were also included. In 2020 special efforts were made to identify contributions to the concentration of gamma emitting radionuclides in sewage sludge from radionuclides originating from diagnostics and therapy with radiopharmaceuticals at Skåne University Hospital in Lund. For this purpose, samples of sewage sludge were collected each week during the summer, each sample representing the average amount of radionuclides in the sludge from the week before (*i.e.* each analysed sample was an average of several sub-samples of sewage sludge, each sub-sample was taken daily over a period of one week). A standard monthly averaged sample (as previously) was analysed as well, for comparison.

All samples analysed with laboratory high resolution gamma spectrometry were measured for about 24 hours.

2.4. Methods for sample collection and analysis of ^{129}I

Samples of grass (sites 27.3, 30, 64), soil (sites 6, 27.3, 30, 32), moss and lichen (site 6) were analysed for their activity concentration of ^{129}I. Since this radionuclide was not analysed before within this zero-point program, a short description of the method is provided, previously published by Schmidt et al [6]. In summary, a small amount of sample (typically between 0.1 and 0.2 g) was mixed with six grams of sodium hydroxide and 4 to 7 mg of iodide carrier in 1 ml of water in a nickel crucible. The crucible was covered and alkaline leaching was carried out for 1 h at 150°C, then for 2 h at 200°C followed by 3 h at 275°C. The melt was then extracted by water and centrifuged. To reduce all iodine species to iodide, N_2H_4 was added to the supernatant. The resulting solution was centrifuged again. Then, $NaNO_2$ was added to the supernatant, acidified with HNO_3 and iodine was extracted into chloroform in a separatory funnel. After reduction by N_2H_4, iodide was back-extracted into water as previously described. The purification by extraction and back-extraction was carried out two more times. Finally, AgI was precipitated by adding an $AgNO_3$ solution, pressed on a copper cathode and measured by Accelerator Mass Spectrometry (AMS) at Centro Nacional de Aceleradores (CAN), University of Seville, Spain.

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 [1-3].

3. RESULTS

3.1. Activity concentration of gamma emitting radionuclides in various types of samples year 2020

In 2020, a total of 63 samples have been analysed for the activity concentration of gamma emitting radionuclides. The sites were selected to overlap with those monitored during the previous years, for samples of grass, crops, bioindicators, fruits and berries, sewage sludge. New sites have been identified for samples of fruits and berries as well as milk and ground beef that was collected from a new dairy farm. Bioindicators were collected where available, at or close to the existing sites. Tables A1.1-1.5, in Appendix 1, present the results from the measurements of the activity concentration of gamma emitting radionuclides in the samples collected in 2020.

The detected activity concentrations of gamma emitting radionuclides in **grass** were below the minimum detectable activity concentration (MDA) for the measurement time: ^{137}Cs (2 Bq kg^{-1} < MDA < 13 Bq kg^{-1}); ^{226}Ra (27 Bq kg^{-1} < MDA < 221 Bq kg^{-1}); ^{228}Ac (5 Bq kg^{-1} < MDA < 38 Bq kg^{-1}), except for ^{40}K that ranged between 284 Bq kg^{-1} to 971 Bq kg^{-1} (average: 620±114 Bq kg^{-1}). This is similar to the previous years [1-3].

For the samples of **crops**, the detected activity concentrations of gamma emitting radionuclides were below the MDA for the measurement time: ^{137}Cs (0.5 Bq kg^{-1} < MDA < 2 Bq kg^{-1}); ^{226}Ra (9 Bq kg^{-1} < MDA < 44 Bq kg^{-1}); ^{228}Ac (2 Bq kg^{-1} < MDA < 7 Bq kg^{-1}), except for ^{40}K that ranged between 99 Bq kg^{-1} to 306 Bq kg^{-1} (average: 158±36 Bq kg^{-1}), also similar to previous years [1-3].

As for the 9 samples of **fruits and berries** analysed, the detected levels of gamma emitting radionuclides were below the MDA for the measurement time: ^{137}Cs (1 Bq kg^{-1} < MDA < 5 Bq kg^{-1}); ^{226}Ra (15 Bq kg^{-1} < MDA < 85 Bq kg^{-1}); ^{228}Ac (3 Bq kg^{-1} < MDA < 13 Bq kg^{-1}), except for ^{40}K that ranged between 202 Bq kg^{-1} to 490 Bq kg^{-1} (average: 317±69 Bq kg^{-1}).

Among the several **bioindicators** found and measured in 2020, ^{226}Ra was not detected in any of the samples using a measurement time of about 24 h: (7 Bq kg^{-1} < MDA 45 Bq kg^{-1}). However, activity concentrations (slightly above the MDA) of ^{137}Cs were observed in two samples of lichen: 5.4±1.5 Bq kg^{-1} and 30±10 Bq kg^{-1}, respectively. As for ^{228}Ac, only one sample (of lichen) were above MDA with the activity concentration 36±5.2 Bq kg^{-1}. This could, however, be attributed to the small amount of dirt remaining on the sample that was not successfully separated Two samples had activity concentrations of ^{40}K below the MDA, “pine needles” (MDA(^{40}K) at 29 Bq kg^{-1}) and “honey” (MDA(^{40}K) at 8 Bq kg^{-1}), whereas the others ranged between 106 Bq kg^{-1} to 1385 Bq kg^{-1}, depending on the type of bioindicator for the radionuclide.

In total, 13 samples of **sewage sludge** were collected in 2020. The majority of the samples were collected on a one-week basis, but also “monthly averaged” samples were included as in previous reports. It is clearly indicated by the results that it is necessary to measure these samples directly after collection, not to miss any short-lived radionuclides. Notable levels of ^{177}Lu and ^{131}I are observed in the sewage sludge when measured close to the sampling date. The radionuclides ^{177}Lu and ^{131}I are used for therapy of cancer patients (prostate cancer and treatment of the thyroid, respectively). The levels observed in the sewage sludge is dependent on the amount of radionuclides distributed to patients at SUS Lund, the residence of the patient and the transportation time of sewage water to the treatment plant, and other factors. As many patients return to their homes after treatment or diagnosis and there they release their radioactive excreta. It is difficult to correlate the amount of activity administered

to patients to the activity concentration of the same radionuclide that is observed in the sewage sludge later on.

However, during the summer months of the sampling, the concentration of ^{177}Lu and ^{131}I varied between 991-8669 Bq kg^{-1} and 1.6-517 Bq kg^{-1}, respectively[1]. At the same time (week 23-32), the average administered activity of these radionuclides at SUS Lund were 11 125 MBq (up to 16 GBq per week) and 5 156 MBq (up to 9 GBq per week) for ^{177}Lu and ^{131}I, respectively.

3.2. *In situ* gamma spectrometry around ESS

The map in Figure 4 shows the mobile gamma spectrometry (NaI(Tl)) measurements carried out around ESS in August 2020. The average Spectrum Dose Index (SDI)[2] over the whole area was 0.090 µSv h^{-1} (C_v = 19%, min = 0.0 56 µSv h^{-1}, max = 0.132 µSv h^{-1}) and 0.093 µSv h^{-1} (C_v = 26%, min = 0.058 µSv h^{-1}, max = 0.198 µSv h^{-1}), during the two occasions, respectively. The slightly elevated SDI levels seen in the map are due to natural occurring radionuclides in constructions and surrounding materials. Single high and low values may also be attributed to electrical noise within the detector system.

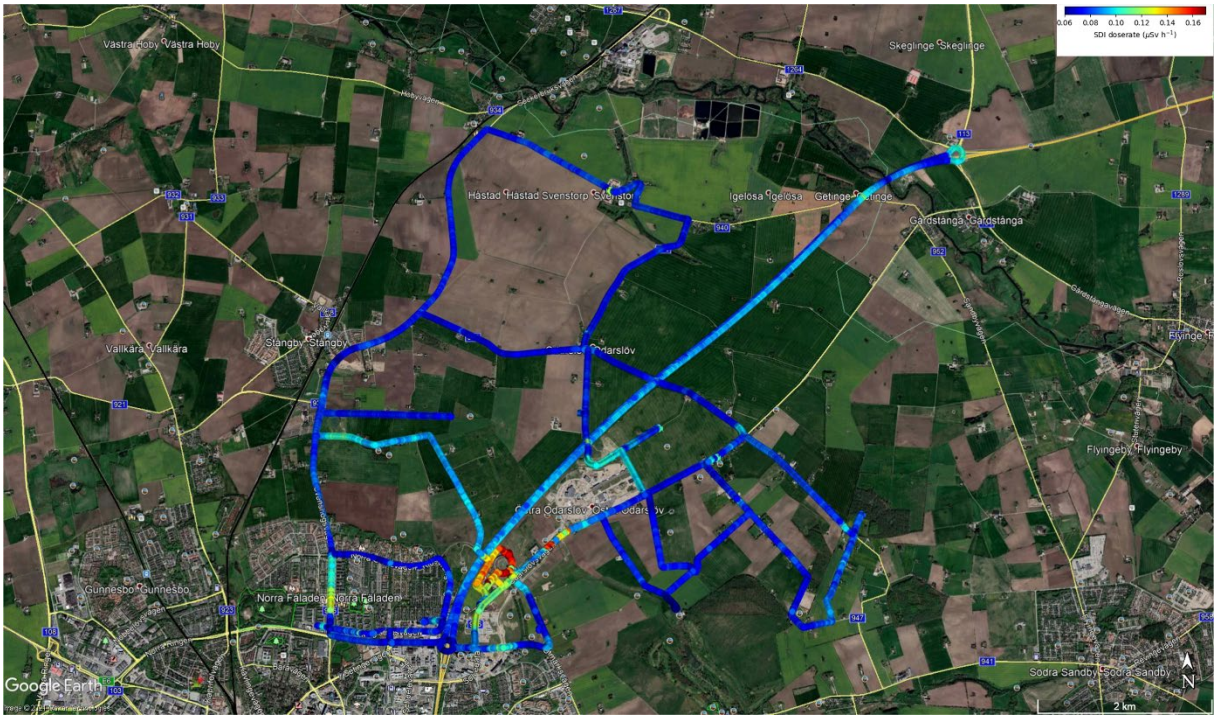

Figure 4 Mobile (car) gamma spectroscopy (NaI(Tl)) around ESS in August 2020. The colour scale shows the average dose rate (µSv h^{-1}) as detected by the NaI(Tl) detectors, averaged over one second, from blue (0.06 µSv h^{-1}) to red (0.17 µSv h^{-1}). Although within normal background levels for the area, the highest values observed are due to naturally occurring radionuclides in nearby soil piles and buildings.

[1] Note that the measured activity concentration is dependent on the time between sampling and measurement.

[2] Spectrum Dose Index (SDI) is a dose-rate (µSv h^{-1}) calculated by multiplying the number of detected pulses in a pulse height distribution by an energy dependent factor to present the total photon dose rate.

In situ high resolution gamma spectra from the four locations around ESS are provided as separate files. The detected radioactivity in air at these sites (35, 36, 37, 38) was evaluated. The activity concentrations of ^{226}Ra, ^{228}Ac and ^{40}K were evaluated as homogenously distributed in the soil (Bq kg^{-1}), whereas for ^{137}Cs the activity was evaluated as surface equivalent activity (Bq m^{-2}). For all sites, ^{137}Cs and ^{226}Ra were below the MDA. ^{228}Ac was below the MDA for two sites (36 and 37) and slightly above the MDA for the two other sites. The activity concentration ^{40}K was on average 500 Bq kg^{-1}, although below MDA at site 37.

A mobile (car) gamma spectrometry (NaI(Tl)) survey around the MAX IV facility (November 2020), when operating at maximum injection frequency, is shown in Figure 5. The figure illustrates the results from several such measurements, during various testing of the facility. Simultaneous dose rate measurements at various locations close to the wall of the cyclotron ring as well as at various distances away from the ring did not show any notable increase in the dose rate. However, it should be noted that potential skyshine gradients and maxima might have been unobserved, due to the terrain and time limit of the tests.

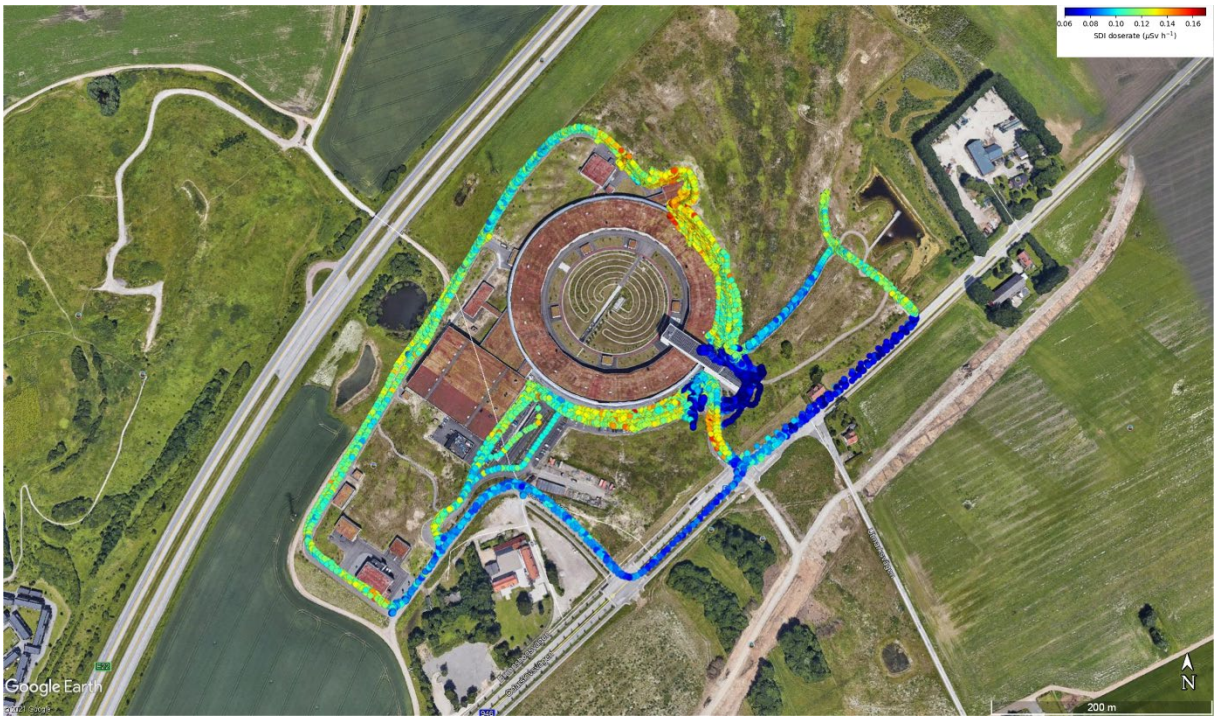

Figure 5 Mobile NaI(Tl) measurements around MAX IV during tests of the facility up to 10 Hz, autumn 2020. The SDI dose rate is indicated as µSv h^{-1} and the scale goes from blue (0.06 µSv h^{-1}) to red (0.17 µSv h^{-1}).

3.3. ^{3}H analysis year 2020

The activity concentration of tritium in the water content of several types of sample was measured by liquid scintillation counting (LSC). The results of the tritium measurements of samples of precipitation, air humidity and surface water collected in 2020 are shown in Tables A1.6-A1.10 in Appendix 1. Repeated measurements on Lund tap water can be found in Table A1.11, and the results from the analyses of sewage sludge, fruits and crops are presented in Table A1.12. Two measurements on milk samples and one beef sample from the dairy farm at site 64 are reported in Table A1.13. The observed levels are generally below the detection limit using the current procedure, instrumentation and analysis time (MDA typically 1.6 Bq L^{-1}). The handful of samples with values above the MDA are still in the

range of expected environmental values with a maximum of 3.2 ± 0.5 Bq L^{-1} for one of the air humidity samples. The results show no evidence of any local contamination of ^{3}H in Lund during 2020.

3.4. ^{14}C analysis year 2020

The results of the ^{14}C analysis of grass, fruits, crops, milk and meat are presented in Table A1.14 in Appendix 1. The results are expressed as $F^{14}C$ [7, 8], see Ref [1] (p. 92-94) for definition and conversion to other activity concentration units[3]. Figure 6 shows $F^{14}C$ at the various sites. The average $F^{14}C$ value of all ^{14}C samples analysed for year 2020 samples was 1.009 (STD: 0.004; SUM: 0.001). The corresponding specific activity is 226 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.

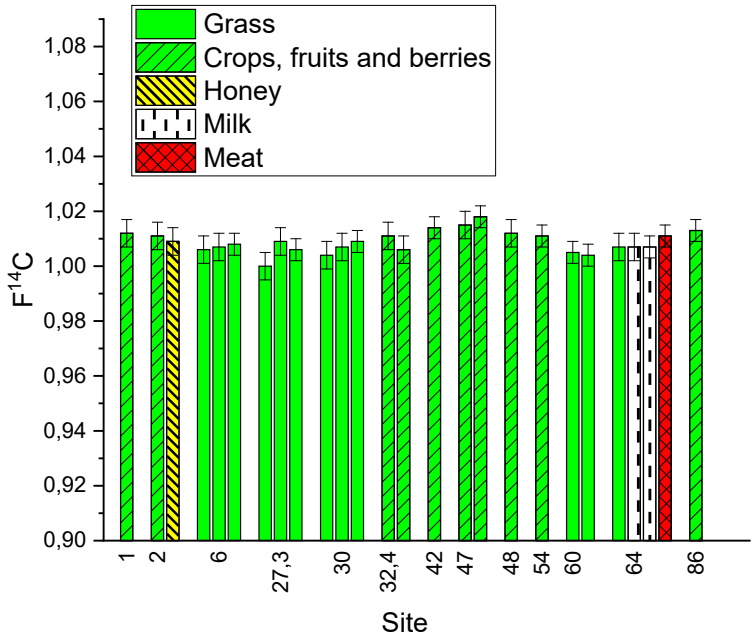

Figure 6 Results of the ^{14}C measurements from 2020. Uncertainties represent 1 standard deviation.

Figure 7 shows the average $F^{14}C$ values obtained in for all ESS zero point assessments so far. For comparison, Figure 7 also includes ^{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

[3] $F^{14}C$ 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)[4] [9-13]. As can be seen in Figure 7, the trend of declining $F^{14}C$ values in atmospheric CO_2 continue (see section 3.4 in Ref [3]).

Seasonal variations, with higher ^{14}C levels in the summer than in the winter, seen in the European and Hyltemossa data in Figure 7, are also visible in the new data for terrestrial organic material from 2020 (see Figure 8 which displays $F^{14}C$ as a function of sampling date). The mean values of the $F^{14}C$ for the 3 sampling seasons (late spring, late summer and late autumn) are significantly different (at the 0.05 level according to an ANOVA test), with higher $F^{14}C$ in the summer than at the other two seasons.

.

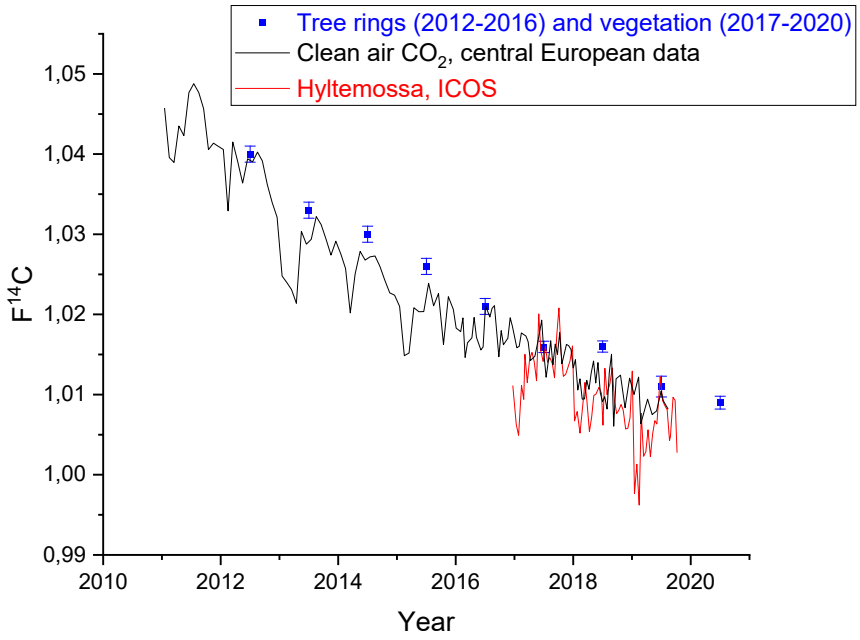

Figure 7 Average $F^{14}C$ values (uncertainty represented by the SUM) obtained for all ESS zero point assessments so far, along with ^{14}C data in atmospheric CO_2 collected at rural background stations in central Europe and at the Swedish ICOS station Hyltemossa [9-13].

[4] 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.

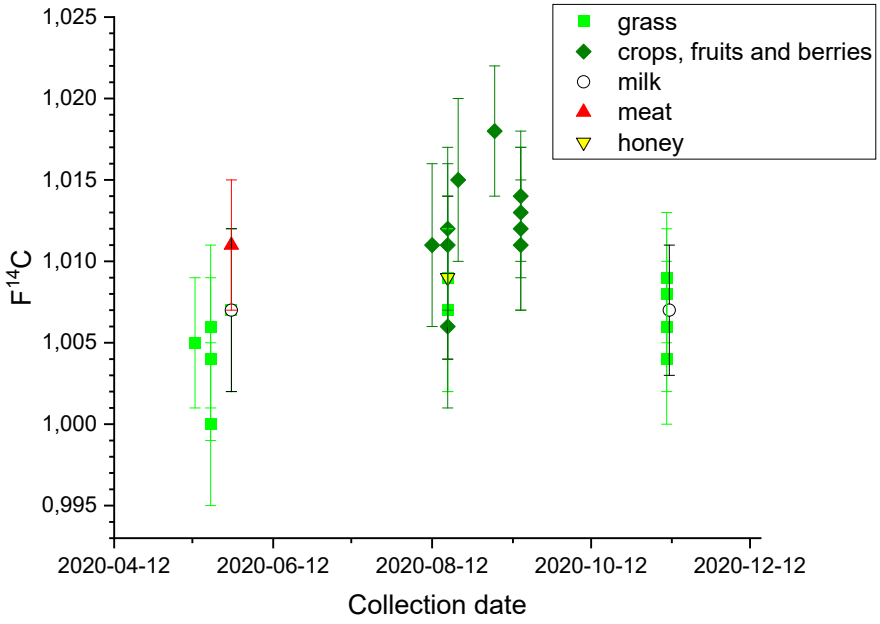

Figure 8 $F^{14}C$ as a function of sampling date. Uncertainties represent 1 standard deviation.

3.5. ^{129}I analysis year 2020

The ^{129}I levels observed, in the limited number of samples so far collected, indicate normal environmental levels for these samples. The results are provided in Table A1.15 in Appendix 1. The ^{129}I activity concentrations ranged between $(3.18 \pm 0.20)\cdot10^{11}$ atoms/kg d.w. for grass and $(1.31 \pm 0.03)\cdot10^{13}$ atoms/kg d.w. for moss. These values are similar to others reported in the literature [14, 15].

3.6. 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 [16].

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].

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

Low levels of gamma emitting radionuclides were observed in the samples collected in 2020, with no significant contribution of anthropogenic radionuclides except for the samples of sewage sludge that were measured within a week after sample collection. The source of the observed levels of ^{177}Lu and ^{131}I in sewage sludge is the hospital SUS Lund. The variability over time and patients places of residence in the region *i.e.* living outside the catchment area of Källby and the potential use of other radionuclides in the future, are important parameters to consider. However, this shows that it is of importance to identify the current sources of anthropogenic radionuclides, although short-lived, within the Lund area.

Among the other samples collected for analysis of gamma emitting radionuclides, the investigated radionuclides were below the MDA, except for ^{40}K that varied slightly depending of type of sample. However, some of the bioindicators (lichen) had detectable levels of other gamma emitting radionuclides, at environmental levels. Hence, this indicates the importance of bioindicators for identifying the presence of various types of radionuclides.

The tritium measurements were extended with new sample types and sites compared to previous years, *e.g.* including the water streams Höje river, Kävlinge river and Lödde river, and meat from cow. The vast majority of the measured tritium levels were below the MDA of 1.6 Bq L^{-1}, with a few exceptions (the maximum tritium value of 3.2±0.5 Bq L^{-1} was registered for an air humidity, sample collected at the urban reference site in the north of Lund in the beginning of March 2020). The data are similar to the tritium data reported in previous annual reports and are close to expected environmental levels.

The ^{14}C data in terrestrial samples in the Lund area and in southern Sweden is consistent with the declining ^{14}C specific activity in atmospheric CO_2. Seasonal variations were observed in the ^{14}C data. No evidence of anthropogenic ^{14}C contamination in the Lund area was noted during 2020.

The ^{129}I activity concentrations ranged between $(3.18 \pm 0.20)\cdot10^{11}$ atoms/kg d.w. for grass and $(1.31 \pm 0.03)\cdot10^{13}$ atoms/kg d.w. for moss, which are considered as expected values.

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. Thanks to Mattias Olsson for assistance in sample preparation for the ^{14}C samples and to professor Rafael Garcia-Tenorio at Universidad de Sevilla for the ^{129}I-analysis.

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.* 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. Lund University. 2020.

[4] 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.

[5] 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.

[6] Schmidt, A., Schnabel, C., Handl, J., Jakob, D., Michel, R., Synal, H.-A., Lopez, J., et al. *On the analysis of iodine-129 and iodine-127 in environmental materials by accelerator mass spectrometry and ion chromatography.* Science of The Total Environment, 223(2-3): 131-156, 1998.

[7] 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.

[8] 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.

[9] Levin, I., Kromer, B. *The tropospheric $^{14}CO_2$ level in mid latitudes of the Northern Hemisphere (1959-2003).* Radiocarbon, 46: 1261-1271, 2004.

[10] 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.

[11] 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.

[12] 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-lXPKZlO4DWX7wncsLQ7akY; http://calib.org/CALIBomb/. 2019.

[13] 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.

[14] Muramatsu, Y., Takada, Y., Matsuzaki, H., Yoshida, S. *AMS analysis of 129I in Japanese soil samples collected from background areas far from nuclear facilities.* Quaternary Geochronology, 3(3): 291-297, 2008.

[15] Krausová, I., Kučera, J., Světlík, I. *Determination of 129 I in biomonitors collected in the vicinity of a nuclear power plant by neutron activation analysis.* Journal of Radioanalytical and Nuclear Chemistry, 295(3): 2043-2048, 2013.
[16] 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 2020

Available upon request. Please contact the authors.