Radiological environmental monitoring at the ESS facility

annual report 2018

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

2020

Citation for published version (APA):
Bernhardsson, C., Eriksson Stenström, K., & Pédehontaa-Hiaa, G. (2020). *Radiological environmental monitoring at the ESS facility: annual report 2018*. (MA RADFYS 2020:02), (BAR-2020/02).

Total number of authors:
3

LUND UNIVERSITY

PO Box 117
221 00 Lund
+46 46-222 00 00

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 2018

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

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 2020:02
Malmö 2020

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

Report BAR-2020/02
Lund 2020

SUMMARY

Radiological environmental monitoring of the pre-operation phase of the ESS started in 2017. The current report presents additional results of ^{3}H in precipitation, air humidity and sewage sludge, gamma radionuclides in sewage sludge and ^{14}C in vegetation.

The sewage sludge sample collected in the summer showed a low level of ^{3}H (2.3±0.4 Bq L^{-1}, MDA = 1.6 Bq L^{-1}) which was similar to the one measured on the winter and spring of the same year on the same location. The results of the tritium measurements in air humidity and precipitation were generally below the detection limit.

The sewage sludge sample collected for analysis of gamma emitting radionuclides indicated presence of ^{131}I (115.7±4.3 Bq kg^{-1}). It is important to note that the measured sample is an average sample of the month (consisting of sub-samples that were collected during each working day of the month) and that the administered e.g. ^{131}I activity at SUS Lund, that end-up in the sewage sludge, is influenced by many different factors (e.g. number of patients, residence of these patients, physical decay). Other natural (^{40}K, ^{226}Ra, ^{228}Ac) and anthropogenic (^{137}Cs) radionuclides were observed at small and expected concentration levels for this type of sample.

The samples of crops had activity concentrations of gamma emitting radionuclides below the MDA for the measurement time (approx. 24 h), except for ^{40}K (131.8±16.6 and 183.4±14.0 Bq kg^{-1}, respectively).

The 20 analysed samples of vegetation showed no signs of anthropogenic contamination of ^{14}C. The average ^{14}C activity concentration from year 2018 was not significantly different from the data obtained in year 2017.

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 2018, with the ESS still being in its pre-operational phase. The radiological environmental monitoring of the pre-operation phase of the ESS started in 2017 in a collaboration project between Lund University (Medical Radiation Physics Malmö and Division of Nuclear Physics) and the ESS [1]. The results of this extensive programme, establishing the “Zero Point” radiation levels during 2017 (and partly during 2018), are presented in the report “Assessment of “Zero Point” radiation around the ESS facility” [2].

The additional 2018 assessment was significantly reduced compared to the 2017 programme and aimed to commence studies of the variability over time of existing radionuclides in the environment of ESS. This meets the criteria for preoperational studies according to the IAEA Safety Guide RSG1.8 [3]. The 2018 measurements focussed on ^{14}C (grass, fruits, berries) and also included a few gamma spectrometry measurements on sewage sludge and crops. Additionally, Lund University measured ^{3}H in precipitation and air humidity during spring 2018.

Lund University continued the ^{3}H measurements during the rest of 2018 in a project financed by the Swedish Radiation Safety Authority (SSM) [4]. The results can be found in the SSM project report in Ref [4]. Apart from these measurements, a base-line study of the tritium levels in urine from 55 individuals, living or working in Lund, was performed in 2018 [4, 5]. This study was also financed by SSM.

1.1. Aim

Environmental monitoring of nuclear or radiation-related facilities may serve different purposes. According to IAEA [3, 6], the specific aims of environmental monitoring of facilities in normal operation are as listed in Table 1.

Table 1 Objectives of environmental monitoring programmes [3, 6].

Criterion	Objective
EMP a)	To demonstrate that authorized discharge limits and public exposure due to direct radiation are not exceeded. If authorized environmental levels exist, to check compliance.
EMP b)	To obtain data and information for dose assessments of populations. To obtain data for assessment of actual or prospective doses to the representative person (see definition of representative person below).
EMP c)	To check operational conditions and to trigger alarms and extended environmental monitoring programmes in case of unusual conditions.
EMP d)	To obtain long-term trends in the environmental radionuclide levels close to the facility. To monitor and evaluate long-term trends in the environmental radionuclide concentrations close to the facility, e.g. due to cumulative impact, and discover any unpredicted changes in the radionuclide levels resulting from unplanned or diffuse releases.
EMP e)	To check predictions of models. To verify the results of the source monitoring and the associated modelling: predictions of environmental levels should be consistent and exposure limits of the public should not be exceeded. To investigate the transport and fate of radionuclides in the environment.
EMP f)	To provide information for the public. To serve as basis for information to the public.

Preoperational data of existing levels of radionuclides, natural as well as man-made, and their variability are thus required to be able to fulfil the objectives of the operational environmental monitoring programme. The aim of the current report is to continue to contribute with such data for the ESS facility.

2. METHODOLOGY

2.1. Description of pre-operational sampling programme

The sampling programme for the extensive base-line measurements in 2017 and beginning of 2018 is described in detail in [2]. The radionuclides selected for the 2017 assessment represent natural as well as man-made radionuclides. Gamma-emitters as well as the pure beta-emitters ^{3}H (tritium) and ^{14}C were included. The majority of the sites in the 2017 programme were located within a few km of the ESS site. The sites were chosen to cover all directions from the ESS site and aimed to be representative and reflect the current population density and ecology in the area. A large number of sites were included in the 2017 programme, with the aim to guarantee that enough number of sites would be accessible for future measurements. The sites selected were located outside the fence of the ESS facility area, except for a limited number of ground- and surface water samples that were taken from drilled holes and ponds inside the ESS area. The 2017 measurements included extensive *in situ* and mobile gamma spectrometry; laboratory-based gamma spectrometry of soil, grass, milk, crops, sewage sludge and bioindicators; ^{3}H analysis of ground water, surface water, some bioindicators, sewage sludge, sugar beets and milk; and ^{14}C analysis of grass, fruits, crops, milk (including fodder of the cows), moss, tree rings and fullerene soot monitors. A summary of the results from the 2017 report is given in section section 4.1.

During year 2018 the continued zero-point measurements included:

- Gamma spectrometry of sewage sludge and crops (to follow long-term trends)
- ^{3}H measurements of precipitation, air humidity, sewage sludge and ground water
- ^{14}C measurements of grass, fruits, berries and crops

Tritium measurements of precipitation and air humidity were not included in the report from year 2017. This is however highly relevant for ESS, since tritium is expected to dominate the source term of continuous radioactive effluents (in $Bq \cdot year^{-1}$) from the ESS [7]. Monitored constituents / frequencies for the 2017-2018 programme [2] and the additional measurements performed during year 2018 are summarized in Table 2.

Table 2 Monitored constituents and frequencies of sampling and measurement for zero point assessments.

Discharge	Monitored constituents	Number of sites/frequency for the 2017-2018 report [2]	This report, for year 2018	SSM report, for year 2018, ref [4]
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	
	Crops	Gamma-emitters at 12 sites, ^{14}C at 6 sites, ^{3}H at one site.	^{14}C at 2 sites, gamma emitters 2 sites	
	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	
	Terrestrial indicators			
	Grass	Gamma-emitters at 20 sites, ^{14}C at 12 sites	^{14}C at 8 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.
	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.
Liquid	*Water bodies*			
	Ground water	^{3}H at 12 sites	At background site	
	Surface water	^{3}H at 8 sites		
	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 after April 2018	

2.2. Sampling locations

Sewage sludge was collected at Källby sewage treatment plant (site 35 in Ref [2]), and the sites for the other ^{3}H and ^{14}C samples collected in 2018 are listed in Table 3. In the present report we refer to a consolidated site number list for all types of radiometric analyses; for clarity the previous sample sites used in Ref [2] for ^{3}H and ^{14}C are included in Table 3. The consolidated site number list will remain in future annual reports.

Table 3 Sampling locations and type of measurements performed during year 2018.

Site	Location	Latitude	Longitude	Corre-sponding site nr ^{14}C	Corre-sponding site nr ^{3}H	Gamma	^{3}H	^{14}C
1	Västra Odarslöv 341	N55.7431	E13.2477	C5				X^{5,6}
2	Östra Odarslöv 651	N55.7380	E13.2736	C25				X^{5,6}
4	Ladugårdmarken 461	N55.7384	E13.2314	C6				X^{6}
6	Möllegården	N55.7305	E13.2441	C3				X^{5,6}
17.1	Klosterängshöjden	N55.7268	E13.2266	C7				X^{5}
27.2	Kärrpengavägen bus stop	N55.7344	E13.2603	C12				X^{5}
31.11	ESS area (11)	N55.7371	E13.2520	C28_1				X^{5}
31.17	ESS area (17)	N55.7346	E13.2429	C28_7				X^{5}
31.20	ESS area (20), at weather station	N55.7366	E13.2455		T2a		X^{2,3}	
35.1	Källby (sewage treatment plant) VA SYD	N55.6952	E13.1637			X^{1}	X^{1}	
36	Svenstorp´s gods	N55.76691	E13.25325			X^{8,9}		
47	Borrby	N55.4256	E14.2236	C1 (rural ref)				X^{5,7}
48	Timjanvägen 5, Lund	N55.7186	E13.1828	C2 (urban ref)	T1a		X^{2,3}	X^{5,6}
49	Dammtorpsvägen	N55.7300	E13.2539	C11				X^{8}
50	ESS 2d wind power tower	N55.7400	E13.2586	C15				X^{8}
52	Professorsgatan 1	N55.7097	E13.2047	C18				X^{7}
54	Active Biotech	N55.7169	E13.2206	C21				X^{7}
55	Stadsodlingen Brunnshög	N55.7208	E13.2411	C22				X^{5}
62	Grevie PV5 well	N55.6131	E13.1970		T0		X^{4}	

1 - sewage sludge; 2 – precipitation; 3 – air humidity; 4 – ground water (background); 5 – grass; 6 – apple; 7 – rowan berries; 8 – wheat; 9 – rape seed

The four sites used for the ^{3}H sampling (one also for gamma) during 2018 were (see also Table 3 and Figure 1):

- **Site 31.20 (referred to as T2 in [4] and Appendix 2), inside the fenced ESS area, located ~ 0.29 km from the ESS target (342°).** Precipitation and air humidity were collected at this site.
- **Site 35.**1: Källby sewage treatment plant in the SW parts of Lund where sewage sludge was collected (also for gamma analysis).

- **Site 48 (referred to as site T1 in [4] and Appendix 2), located ~4.3 km from ESS (~246°).** This site in the north of Lund acts as a rural background site for air humidity. A few precipitation samples were also collected at this site.
- **Site 62 (referred to as site T0 in [4] and Appendix 2), located ~13.8 km from ESS (~193°).** Water from the deep well Grevie PV5 (depth 71-72 m), 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} [8]).

Samples of crops were collected at Site 36, during harvest (August 2018), for analysis of gamma emitting radionuclides.

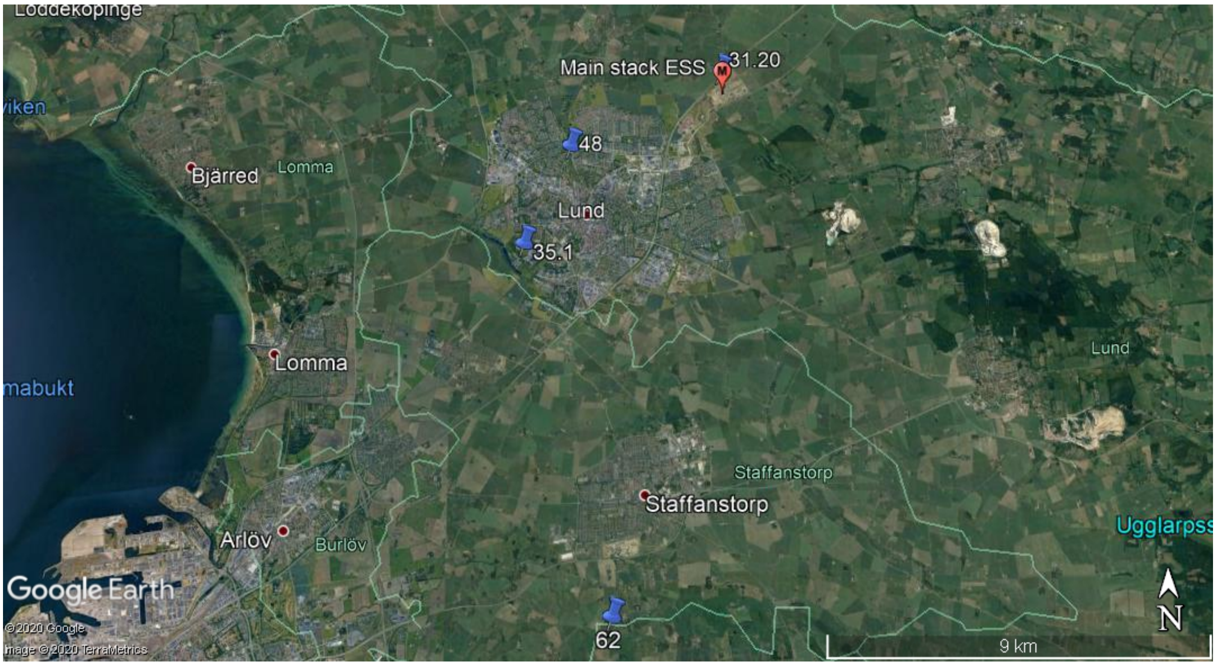


Figure 1 Sampling sites for ^{3}H samples for year 2018. See Table 3 for more information about the sampling sites.

The sampling sites for ^{14}C for year 2018, see Table 3 and Figure 2, represent a selection of the sites in the 2017 report. 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, were included for comparison. As for the 2017 assessment, two samples were collected in the north-eastern part of Lund, where potential sources of anthropogenic ^{14}C exist: sites 52 and 54.

Figure 2. Sampling sites for ^{14}C samples for year 2018. See Table 3 for more information about the sampling sites.

2.3. Methods for sample collection and analysis

The collection and sample preparation procedures for precipitation and air humidity samples are described in Appendix 1 in this report. In summary, precipitation samples were collected on a monthly basis using a rain collector following the recommendations of the IAEA/GNIP precipitation sampling guide [9]. Air humidity was collected using condensation of water vapor (see Appendix 1 further details). A weather station (Davis Vantage Pro2, Davis Instruments) was installed at site 31.20 in the end of September 2018 (see chapter 5.3.2 in Ref [4] for further details). Sewage sludge was collected and pretreated for ^{3}H according to

ANNEX B3 in Ref [2]. Ten ml of the ^{3}H samples were analysed using a Beckman LS 6500 counter according to Appendix 2. The majority of the ^{3}H samples samples were measured for 600 min.

Sewage sludge was collected and pretreated for ^{3}H according to ANNEX B3 in Ref [2]. Ten ml of the ^{3}H samples were analysed using a Beckman LS 6500 counter according to section 3.3 of Appendix 2. The majority of the samples were measured for 1200 min.

Sewage sludge for gamma analysis was collected, pretreated and measured according to ANNEX B2 in Ref [2]. Samples of crops for gamma analysis were collected, pretreated and measured according to ANNEX B2 in Ref [2].

The collection and sample preparation for ^{14}C, as well as measurement and analysis, were the same as ANNEX B4 in Ref [2].

3. RESULTS

3.1. Summary of the 2017 report

The main outcomes of the 2017 assessments were (cited from Ref [2, 10]):

- The concentrations of natural occurring radionuclides in the ground were similar, with only small variations observed between the sites. Expected differences between the sites were observed for ^{137}Cs, in terms of depth distribution and total inventory in the soil.
- No unexpected gamma emitters were observed in the various samples collected:
    - ^{131}I was observed in the majority of the samples of sewage sludge. Other medically used radionuclides might have been found if the time between sampling and measurements had been shortened.
- Only small variations were observed for the radionuclides investigated with *in situ* gamma spectrometry at the various sites, except for ^{137}Cs that was observed to vary between some sites.
- An average value of the ambient dose equivalent rate 1 meter above the ground was determined as well as the variability within and between the sites.
- The tritium levels were below the minimum detectable activity concentration (MDA) in all samples analysed, except two samples of sewage sludge, where the activity concentration was slightly above the MDA.
- The sampling sites in the Lund area showed no signs of local anthropogenic contamination of ^{14}C.

3.2. Analysis of ^{3}H in precipitation and air year 2018

The results of the tritium measurements of the collected precipitation and air humidity samples during the spring 2018 are shown in Appendix 2. As concluded in Appendix 2, the observed levels are generally below the detection limit using the procedure and analysis time described in Appendix 1 and 2. For the results for the rest of year 2018, presented in the SSM report Ref [4], the detection limit was lowered using longer measurement time of samples as well as background.

3.3. Analysis of ^{3}H and gamma-emitters in sewage sludge year 2018

The activity concentration (Bq L^{-1}) of ^{3}H was measured in one sewage sludge collected in July 2018 at Källby wastewater treatment plant (see Appendix 3, Table A3.1). The ^{3}H activity concentration of 2.3±0.4 Bq L^{-1} was above the MDA (1.6 Bq L^{-1}) but still very close to the environmental levels. Similar values measured in sewage sludge in January and April 2018 on the same location were reported previously [2].

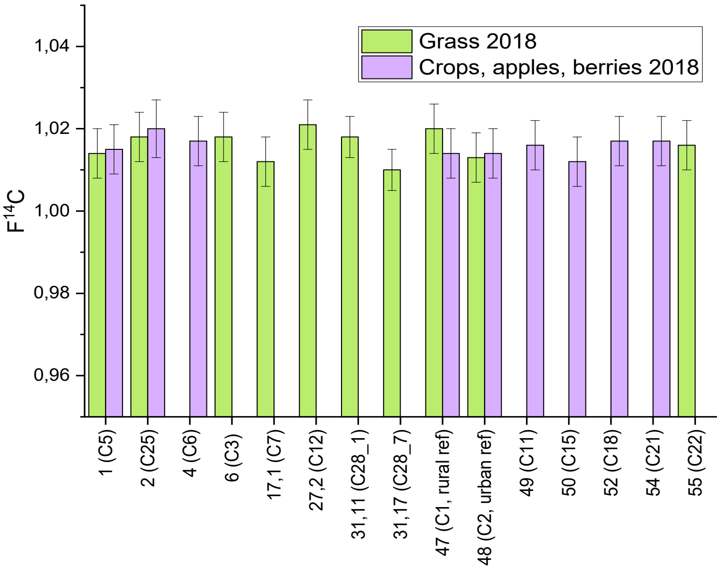
The activity concentration (Bq kg^{-1} d.w.) of gamma emitting radionuclides was measured in one sample of sewage sludge collected in July 2018 at Källby wastewater treatment plant (see Appendix 3, Table A3.2). Apart from naturally occurring radionuclides, the sample also contained 115.7±4.3 Bq kg^{-1} of ^{131}I. Additional monthly samples of sewage sludge were collected in January-April of 2018, for which the concentration of gamma emitting radionuclides has been reported in Ref [2].

3.4. Analysis of gamma-emitters in crops year 2018

The activity concentration (Bq kg^{-1} d.w.) of gamma emitting radionuclides was measured in two samples of crops collected during harvest in 2018 at Svenstorp´s gods (country seat) (see Appendix 3, Table A3.3).

3.5. ^{14}C analysis year 2018

The results of the ^{14}C analysis of grass and crops are presented in Appendix 3 (Table A3.4 and A3.5) and Figure 3. The results are expressed as $F^{14}C$ [11, 12], see ANNEX B4 in Ref for definition[1].



a)

---

[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 [13,, 14].

[13] 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,

[14] 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.

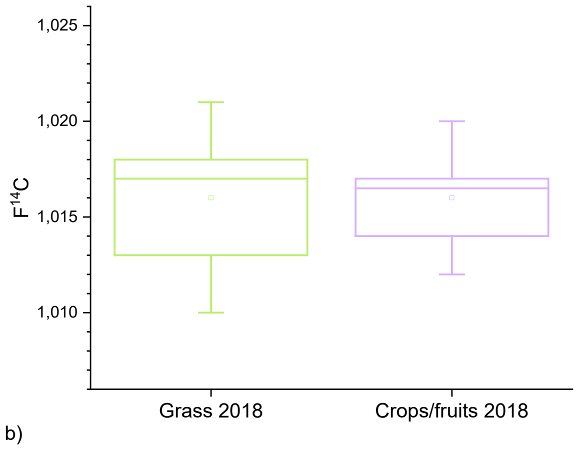


Figure 3. a) Results of the ^{14}C measurements of grass and crops/fruits from 2018. b) Box plot showing that the mean $F^{14}C$ value of grass was not significantly different from the crops/fruit data of 2018 (p>0.05, one-way ANOVA).

For the grass data from 2018 (*N*=10), the average $F^{14}C$ value was 1.016 (STD: 0.004; SUM: 0.001). Crops, fruits and berries from 2018 (*N*=10) also had an average $F^{14}C$ value of 1.016 (STD: 0.002; SUM: 0.001). This corresponds to a specific activity of 228 Bq/kg C using $\delta^{13}C$= - 25 ‰ (see Annex B4 in Ref [2]). The data is normally distributed, and no outliers are identified in the data set using Grubb's test. The average $F^{14}C$ value for all organic samples grown during the previous year, 2017, was 1.017 (*N*=57; SUM: 0.001) as reported in Ref [2]. The average of the $F^{14}C$ data from year 2018 is not significantly different from the data from year 2017. None of the samples analysed show any signs of anthropogenic ^{14}C in the Lund area.

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 samples with certified values (TRY44 number R8/12/123 by Eckert and Ziegler, Germany, 1.5% uncertainty) were used as control. A quenching curve was also obtained using the method described by the cocktail provider Perkin Elmer [15].

The high purity germanium detectors used for determining the activity concentration of gamma emitting radionuclides were calibrated as described in Ref [2]. The detectors are regularly controlled (efficiency, resolution etc.) every 2nd or 3rd month using a calibration source. The MDA levels depend on the measurement time of each sample (normally 24 h) and is specific to each radionuclide. MDA values are provided together with the measurement results.

The quality of the ^{14}C data was assured by measurement and analysis of secondary standards as described in Ref [2].

4. SUMMARY AND CONCLUSIONS

Radiological environmental monitoring of the pre-operation phase of the ESS started in 2017. The current report has presented additional results of ^{3}H in precipitation, air humidity and sewage sludge, gamma radionuclides in sewage sludge and ^{14}C in vegetation.

The sewage sludge sample collected in the summer showed a low but measurable level of ^{3}H (2.3±0.4 Bq L^{-1}, MDA = 1.6 Bq L^{-1}) which was similar to the one measured on the winter and spring of the same year on the same location. The results of the tritium measurements in air humidity and precipitation were generally below the detection limit.

The sewage sludge sample collected for analysis of gamma emitting radionuclides indicated presence of ^{131}I (115.7±4.3 Bq kg^{-1}). It is important to note that the measured sample is an average sample of the month (consisting of sub-samples that were collected during each working day of the month) and that the administered e.g. ^{131}I activity at SUS Lund, that end-up in the sewage sludge, is influenced by many different factors (e.g. number of patients, residence of these patients, physical decay of radionuclides). Other natural (^{40}K, ^{226}Ra, ^{228}Ac) and anthropogenic (^{137}Cs) radionuclides were observed at small and expected concentration levels for this type of sample.

The two samples of crops had activity concentrations of gamma emitting radionuclides below the MDA for the measurement time (approx. 24 h), except for ^{40}K (131.8±16.6 and 183.4±14.0 Bq kg^{-1}, respectively).

The 20 analysed samples of vegetation showed no signs of anthropogenic contamination of ^{14}C. The average ^{14}C activity concentration from year 2018 was not significantly different from the data obtained in year 2017.

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] ESS. *Collaboration Agreement ESS-0093103 Concerning "Monitoring of environmental radioactivity and radiation levels for "Zero Point" Assessment and Contributions to ESS Environmental Monitoring Plan" between European Spallation Source ERIC and Lund University.* ESS-0093103. 2017.

[2] 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.

[3] International Atomic Energy Agency (IAEA). *Environmental and Source Monitoring for Purposes of Radiation Protection. Safety Guide RSG1. 8.* International Atomic Energy Agency. Vienna, Austria. 2005.

[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] Pedehontaa-Hiaa, G., Holstein, H., Mattsson, S., Rääf, C., Eriksson Stenström, K. *Tritium in urine from members of the general public and occupationally exposed workers in Lund, Sweden, prior to operation of the European Spallation Source.* Journal of Environmental Radioactivity, 213: 1-7, 2020.

[6] International Atomic Energy Agency (IAEA). *Programmes and Systems for Source and Environmental Radiation Monitoring.* IAEA Safety Report Series No. 64. International Atomic Energy Agency. Vienna, Austria. 2010.

[7] Ene, D. *Source Term to the Environment from the ESS Target Station*. Nuclear Energy Agency. NEA/NSC/R(2018)2, p. 32-47. 2018.

[8] 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.

[9] IAEA/GNIP. *IAEA/GNIP precipitation sampling guide.* Accessed: Available from: http://www-naweb.iaea.org/napc/ih/documents/other/gnip_manual_v2.02_en_hq.pdf.

[10] Certificate of calibration – multinuclide standard solution (No. 7603; 1991-36). Eckert & Ziegler Nuclitec GmbH, 24937 Avenue Tibbitts, 91355 Valencia CA, USA.

[11] 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.

[12] 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.

[13] 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.

[14] 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.

[15] 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: TRITIUM IN PRECIPITATION AND AIR

Department of Physics
Division of Nuclear Physics
The Biospheric and Anthropogenic Radioactivity (BAR) group

Procedure: Collection of precipitation and air humidity and subsequent measurement of waterborne tritium (HTO)

Authors	Kristina Eriksson Stenström	Professor Division of Nuclear Physics Lund University
	Guillaume Pedehontaa-Hiaa	PhD, Researcher Division of Nuclear Physics Medical Radiation Physics, ITM Lund University
	Vytenis Barkauskas	PhD, Post-doc Division of Nuclear Physics Lund University
Reviewed by	Christopher Rääf	Associate Professor Medical Radiation Physics, ITM
	Daniela Ene	PhD, Senior Scientist Environment, Health and Safety European Spallation Source ESS-ERIC
Approved by	Dirk Rudolph	Professor, Head of Division Division of Nuclear Physics

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

Report BAR-2018/02

Lund 2018

TABLE OF CONTENT PAGE

LIST OF FIGURES

7. PURPOSE

The purpose of the procedure is to describe the necessary steps for measurement of the activity of tritium (^3H) in the atmospheric water vapour and in precipitation, i.e.:

- sampling of atmospheric water vapour and sampling of precipitation;
- sample preparation;
- measurement of tritium using Liquid Scintillation Counting (LSC);
- data analysis to produce the final results as activity concentration of tritium in precipitation and air humidity.

The procedure was applied to the background measurements of tritium reported in Ref [1]. The procedure may be applied to future measurements of tritium-containing water in air and precipitation at the ESS site and surroundings in the frame of the ESS Ambient Control Program to be endorsed by the Swedish Radiation Safety Authority (SSM).

8. PROCEDURE APPLICABILIBTY

The procedure is applicable to environmental sampling of water from precipitation (continuous sampling) and air (occasional sampling). The activity concentration of waterborne tritium (HTO) is measured with LSC. The procedure is applicable to measurements of tritium in the ESS site area both prior to and after start of operation of the ESS facility in order to assess the background and the radiological impact of ESS.

The air humidity sampler must not be used during rainy weather conditions. For continuous monitoring of tritium in air during operation of the ESS facility, a commercially available tritium air sampler, collecting water vapour as well as hydrogen gas, is recommended.

9. SAMPLING OF WATER FROM AIR AND PRECIPITATION

9.1. Procedure map

The procedure map is shown in Figure 1.

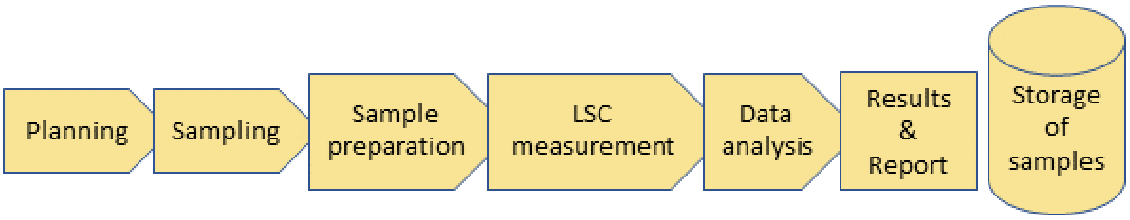


Figure 4 Procedure map.

9.2. Procedure details

Details of the procedure map (planning and sampling) are shown in Figure 2.

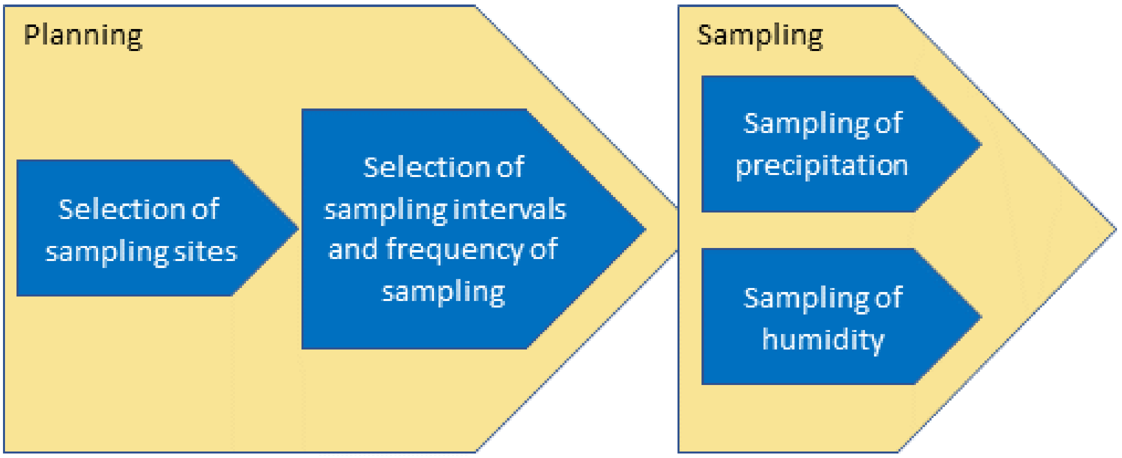


Figure 5 Details of the procedure map (planning and sampling).

9.2.1. Input

The input to the procedure is water-borne tritium in precipitation and air.

9.2.2. Planning

Several criteria for selection of the sampling sites and time intervals are analysed prior to sampling. For sampling of precipitation, the sampling site within the ESS area should be selected considering the recommendations in the IAEA/GNIP precipitation sampling guide [2]. In particular, the influence from structures should be minimized, e.g. by keeping trees and buildings at least as far away as they are high [2]. The sampling location should preferably be within the fenced area to minimize the risk of e.g. theft. For sampling of air humidity, the site should preferably not be in the immediate vicinity of very busy roads (to reduce influence from water vapour produced in combustion engines). The sampling of air humidity requires access to 230 V. Sampling of air humidity for background measurements should strive to cover different weather conditions, wind directions and seasons. During operation of the facility, it is recommended to sample air humidity in the upwind as well as in the downwind direction of the ESS site. Preferably, precipitation samples as well as air humidity samples should also be collected occasionally at an urban background site. Intervals for continuous sampling of precipitation are preferably 1 month as recommended by GNIP [2], unless the purpose of the measurements gives rise to another sampling frequency (e.g. at changes in operating conditions of the facility).	**Responsible:** Radiological expert Planning Selection of sampling sites Selection of sampling intervals and frequency of sampling

For sampling of air humidity, it should not rain, since the sampler is not designed for such weather conditions. Therefore, the weather forecast must be consulted when deciding the sampling date. If possible, sampling of air humidity should be performed at the monthly emptying of the precipitation collector.

Output/product	Selection of sites. Selection of sampling intervals for precipitation. Selection of dates for collection of air humidity.

9.2.3. Sampling

9.2.3.1. Sampling of precipitation

Responsible:

Radiological expert and/or technician

Sampling

Equipment: Precipitation is sampled using a rain collector from Palmex (Croatia) (Standard Rain Sampler RS1, including a siphon inlet) [3]. The sampler can be equipped with a funnel of diameter 13.5 cm or 23 cm for rain sampling. The larger funnel is commonly being used for rain sampling. During winter months a snow tube (diameter 15 cm, height 52 cm) is placed on top of the smaller funnel to assure representative sampling of snow. Water from the precipitation enters a 3 L plastic bottle through the funnel and the plastic siphon tube (reaching the bottom of the 3 L flask). A spare bottle with lid is available and used when emptying the sampler (bottles are switched). This dip-in sampler (and the sampling site) fulfils the recommendations of the IAEA/GNIP precipitation sampling guide [2]. The sampler is mounted on a metal stand (see Figure 3).

Figure 6 The Palmex rain sampler RS1, equipped with the 23 cm funnel.

Sampling of precipi-tation

Method: At the time of retrieving a precipitation sample, the bottle with the precipitation is unscrewed from below, and the flask is closed with the associated lid. The spare bottle is attached to the sampler. The water is taken to the lab where it is transferred to one or several 500 ml water collection bottles (water sampling bottle, sterile, VWR collection, art no 331-0063) using a dedicated funnel. The water is weighed. The sterile water flask is labelled with sampling date interval, amount of collected water, sampling site

(T#), sample number (P#) and signature of the person performing the task. The sterile water flasks with the precipitation is stored in a refrigerator at 4°C at the sample preparation laboratory. The 3 L plastic flask is washed with distilled water and dried. All sampling data should be stored.

9.2.3.2. Sampling of air humidity

Responsible:

Radiological expert and/or technician

Sampling of humidity

Equipment: For sampling of air humidity, a dehumidifier from Wood's (Wood's MRD10, Woods, Canada) [4] is used, see Figure 4. A 24 cm long plastic tubing (inner diameter 8 mm) is connected to the water outlet of the dehumidifier. A sterile 500 ml water collection bottle (VWR collection, art no 331-0063) is used for collection of condensed water vapour. A cable approved for outdoor use shall be used to connect the dehumidifier to a 230 V power supply. Temperature and air humidity are monitored during sampling.

Figure 7 The air humidity sampler Wood's MRD10 located in front of the Palmex rain sampler.

Method: The sampler is transported to the sample collections site by car at the time of sampling. The sampler is placed in a plastic storage box (the red box in Figure 4), which also have space available for the sample collection flask. When sampling at the rain collector, a tray is placed at the base of the metal rack of the rain collector to stabilize the box with the humidity sampler. The dehumidifier is secured onto the metal rack by using straps. If the sampler is used elsewhere, it is suitable to place the sampler and the plastic storage box on a portable table.

The free end of the plastic tubing (the tubing that is attached to the water outlet of the dehumidifier) is put into a 500 ml sample collection bottle (also placed in the red storage box in Figure 4). Care should be taken to protect the socket (connecting the dehumidifier and extension cable) from any water (e.g. by keeping the socket in a water-tight plastic bag).

The dehumidifier is started by pressing the start button (ON/OFF). The mode should be "Continue" (select at HUM ID button), which refers to continuous sampling. Air humidity and temperature are noted at the beginning and at the end of the sampling. The time of sampling depends on the temperature and relative humidity (and possibly on wind speed). In general: sample for 1 hour and then check the amount of water collected, either by visually inspecting the sample or by weighing the sample (at least 20 g of water should be collected).

Switch off the dehumidifier when the sampling is complete. Wait for at least 5 minutes, and then carefully tilt the dehumidifier towards the water sampling bottle to allow water condensed inside the dehumidifier to enter the water sample bottle. The water inside the dehumidifier may also be frozen, which require some additional time for thawing (the dehumidifier is however equipped with a defrosting function, which may be activated during sampling). Remove the water sampling flask, put on the lid, weigh the sample and label the flask with sampling date, sampling interval, amount of collected water, sampling site (T#), sample number (H#) and signature of the person performing the task. The sterile water flasks with the condensed water vapour are stored in a refrigerator at 4°C in the sample preparation laboratory until analysis. All sampling data should be stored, including temperature, air humidity, wind speed and wind direction at the time at sampling (the two latter obtained from a nearby weather station).

Output/product	Water samples from precipitation (integrated sampling). Samples of air moisture (occasional sampling).

9.2.4. Sample preparation

The precipitation samples are filtrated using filter paper to remove small plant or soil pieces that could have entered the collection bottle. The humidity samples are filtrated if collected at very dry and windy conditions.

10 mL of the filtrate is mixed with 10 mL of Ultima Gold LLT LSC cocktail in a 20 mL LSC plastic vial, the vial is shaken for 2 min and stored for 48 h in the dark to reduce chemical quenching before LSC measurement.

Background samples are prepared following the same procedure but with 10 mL of tritium depleted water (also called old water). Such water can be obtained from a deep well at Grevie (N55.6131, E13.1970) - operated by VA Syd - with a well-documented low HTO concentration (well 8 in Ref [5]).

Standard samples are prepared using 10 mL of the same tritium depleted water added with known amounts of tritium from a reference solution (TRY44 number R8/12/123 by Eckert and Ziegler, Germany).

Responsible:

Technician

Sample preparation

Output/product	Filtered water samples mixed with scintillation cocktail, ready for measurement.

9.2.5. LSC analysis

The samples are analyzed in a LSC liquid scintillation counter according to the instructions given by the manufacturer of the instrument. The basic output of the analysis is instrument-dependant. However, in general an LSC instrument provides counts per minute (cpm), the statistical uncertainty of this count (%) and the counting efficiency. Measurements of the standards serve as quality insurance of the LSC results.

Responsible:

Radiological expert

LSC measure-ment

Output/product	Counts per minute (cpm) including statistical uncertainty and counting efficiency of the measurements of unknowns, standards and background.

9.2.6. Calculations

The following should be performed with an external computer if the LSC instrument is not equipped to post-process the results.

Responsible:

Radiological expert

Data analysis

Results & Report

9.2.6.1. *MDA*

The minimum detectable activity, *MDA* (Bq/L), is calculated according to equation (2) [6]:

$$MDA = \frac{3.29 \cdot \sqrt{\left(\frac{cpm_b}{t_s}\right) + \left(\frac{cpm_b}{t_b}\right) + \left(\frac{2.71}{t_s}\right)}}{60 \cdot E \cdot V} \qquad (1)$$

where cpm_b is the count rate of the background (cpm), 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).

9.2.6.2. *Tritium activity concentration in precipitation*

The tritium activity concentration A in precipitation in Bq/L is calculated according to equation (1):

$$A = \frac{cpm_s}{60 \cdot V \cdot E_s} - \frac{cpm_b}{60 \cdot V \cdot E_b} \qquad (2)$$

where cpm_s is the count rate of the sample, E_s is the counting efficiency of the sample, cpm_b is the count rate of the background, E_b is the counting efficiency of the background and V is the volume (L).

9.2.6.3. *Tritium activity concentration in air humidity*

The relative humidity RH is given by:

$$RH = \frac{p}{p_{sat}} \qquad (3)$$

where p is the partial water vapour pressure in air (Pa) and p_{sat} is the saturated vapour pressure over water (Pa). Tetens' equation provides the saturated vapour pressure over water, p_{sat}, given in Pa [7]:

$$p_{sat} = 610.78 \cdot e^{\left(\frac{17.27 \cdot T_{°C}}{T_{°C}+237.3}\right)} \tag{4}$$

where $T_{°C}$ is the temperature in °C.

The ideal gas law states

$$p \cdot V = \frac{m}{M} \cdot R \cdot T \tag{5}$$

where V is the volume, m is the mass, M is molar mass of water vapour ($18.015 \cdot 10^{-3}$ kg/mol), R is the gas constant (8.314 J/(mol·K)) and T is the temperature in K.

The absolute humidity AH is given by:

$$AH = \frac{m}{V} \tag{6}$$

Using equations (3)-(6) above, AH (kg/m^3) can be written as:

$$\begin{aligned} AH &= \frac{m}{V} = \frac{p \cdot M}{R \cdot T} = \frac{RH \cdot p_{sat} \cdot M}{R \cdot T} \\ &= \frac{RH \cdot M}{R \cdot T} \cdot 610.78 \cdot e^{\left(\frac{17.27 \cdot T_{°C}}{T_{°C}+237.3}\right)} \end{aligned} \tag{7}$$

The tritium activity concentration in air humidity, A_{air} (Bq/m^3) is finally given by:

$$A_{air} = \frac{AH \cdot A}{\rho} \tag{8}$$

where A is the measured activity concentration of water (Bq/L) and ρ is the density of water (1.0 kg/L).

9.2.6.4. Uncertainty in LSC measurement

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 total calculated uncertainty (σ_{calc}) in the activity concentration considers the uncertainty of the sample as well as the background:

$$\sigma_A{}^2 = \left(\frac{\partial A}{\partial cpm_s}\right)^2 \cdot \sigma_{cpm_s}{}^2 + \left(\frac{\partial A}{\partial cpm_b}\right)^2 \cdot \sigma_{cpm_b}{}^2 \tag{9}$$

Thus,

$$\sigma_A = \frac{1}{60 \cdot V}\left(\left(\frac{\sigma_{cpm_s}}{E_s}\right)^2 + \left(\frac{\sigma_{cpm_b}}{E_b}\right)^2\right)^{0.5} \qquad (10)$$

The statistical uncertainty in counts *(cpm·t)* of a measurement equals $(cpm\cdot t)^{0.5,}$ thus:

$$\sigma_{cpm} = \frac{(cpm \cdot t)^{0.5}}{t} = \left(\frac{cpm}{t}\right)^{0.5} \qquad (11)$$

and

$$\sigma_A = \frac{1}{60 \cdot V}\left(\frac{cpm_s/t_s}{{E_s}^2} + \frac{cpm_b/t_b}{{E_b}^2}\right)^{0.5} \qquad (12)$$

9.2.6.5. Uncertainty in activity concentration of air humidity

The uncertainty in the calculation of A_{air} is believed to be associated with a) the measurement of the relative humidity *RH*, and b) isotope fractionation during the condensation process.

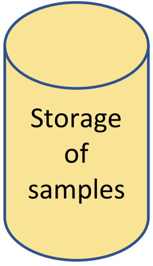
a) The uncertainty in the measurement of the relative humidity RH is conservatively estimated to be 10 % (depending on the instrument used).

b) Isotope fractionation during condensation depends on several factors, such as relative humidity and condensation temperature [8]. In laboratory tests on condensation of water vapour, performed by Deshpande et al [8], the molar concentration of deuterium (^2H) was on average 1.5% higher in the condensate than in the vapour of ambient air (condensation temperature 0 °C). Maximum observed relative molar excess of ^2H was 4% [8]. Assuming that the isotope fractionation of tritium is of the same order of magnitude as for ^2H, the concentration of tritium may be up to 8% higher in the liquid phase than in the vapour (with the experimental conditions of Ref. [8]). Using a conservative approach, the uncertainty due to isotope fractionation during condensation is estimated to be 15%.

The final uncertainty in the HTO activity concentration of air humidity includes the uncertainty of the LSC measurement as well as in *RH* and isotope fractionation.

Output/product	Activity concentrations of tritium in precipitation (in Bq/L) and air (in Bq/m^3). If the sample activity concentration is below the *MDA*, the value is presented as "< *MDA*"). The results shall be presented in a written report, which includes quality assessment of the measurements and interpretation of the results.

9.2.7. Storage of samples

Sample material remaining after LSC analysis is stored according to recommendations by SSM [9].	**Responsible:** Radiological expert 
Output/product	The procedure guarantees that sample material is available until the data has been disseminated.

10. ACKNOWLEDGEMENT

The authors thank Associate Professor Christopher Rääf, Medical Radiation Physics (Malmö) and Dr Daniela Ene, ESS, for constructive reviewing of the report.

11. REFERENCES

[1] Eriksson Stenström, K., Pedehontaa-Hiaa, G., Barkauskas, V. *Tritium in precipitation and air humidity at the European Spallation Source (ESS) site: results from measurements in the spring of 2018.* BAR-2018/03. Lund University, Department of Physics, Division of Nuclear Physics. 2018.

[2] IAEA/GNIP. *IAEA/GNIP precipitation sampling guide*. Accessed: 2018-05-02. Available from: http://www-naweb.iaea.org/napc/ih/documents/other/gnip_manual_v2.02_en_hq.pdf.

[3] Palmex. Accessed: 2018-06-13. Available from: http://www.rainsampler.com/portfolio-page/rain-sampler-rs1/.

[4] Wood's. Accessed: 2018-06-13. Available from: https://woods.se/en/Produkter/dehumidifiers/woods-mrd10/.

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

[7] Tetens, O. *Über einige meteorologische Begriffe.* Zeitschrift für Geophysik, 6: 207-309, 1930.

[8] Deshpande, R.D., Maurya, A.S., Kumar, B., Sarkar, A., Gupta, S.K. *Kinetic fractionation of water isotopes during liquid condensation under super-saturated condition.* Geochimica et Cosmochimica Acta, 100: 60-72, 2013.

[9] *Special conditions for the ESS facility in Lund.* Vers. 2, ESS-0018828. Swedish Radiation Safety Authority. 2015.

APPENDIX 2. REPORT TRITIUM IN AIR AND PRECIPITATION

Department of Physics
Division of Nuclear Physics
The Biospheric and Anthropogenic
Radioactivity (BAR) group

Tritium in precipitation and air humidity at the European Spallation Source (ESS) site: results from measurements in the spring of 2018

Authors	Kristina Eriksson Stenström	Professor Division of Nuclear Physics
	Guillaume Pedehontaa-Hiaa	PhD, Researcher Division of Nuclear Physics / Medical Radiation Physics, ITM
	Vytenis Barkauskas	PhD, Post-doc Division of Nuclear Physics
Reviewed by	Christopher Rääf	Associate Professor Medical Radiation Physics, ITM
Approved by	Dirk Rudolph	Professor, Head of Division Division of Nuclear Physics

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

Report BAR-2018/03

Lund 2018

TABLE OF CONTENT PAGE

LIST OF TABLES

LIST OF FIGURES

1. INTRODUCTION

Tritium is a naturally occurring radionuclide ($T_{1/2}$ = 12.3 years, decay mode β^-), which is also produced anthropogenically e.g. by neutron activation in nuclear reactors or by spallation reactions in accelerator facilities. Tritium will be produced by the ESS facility, and part is expected to be released to the environment during normal operation [1].

This report presents the results of ESS Purchase Order number 24005514: "Measurements of tritium in precipitation and air humidity collected at the ESS site during spring 2018". The tasks include:

- Task #1: Measurement of tritium in 2 air samples (tritium-containing water, HTO)
- Task #2: Measurement of tritium in 2 precipitation samples (HTO)

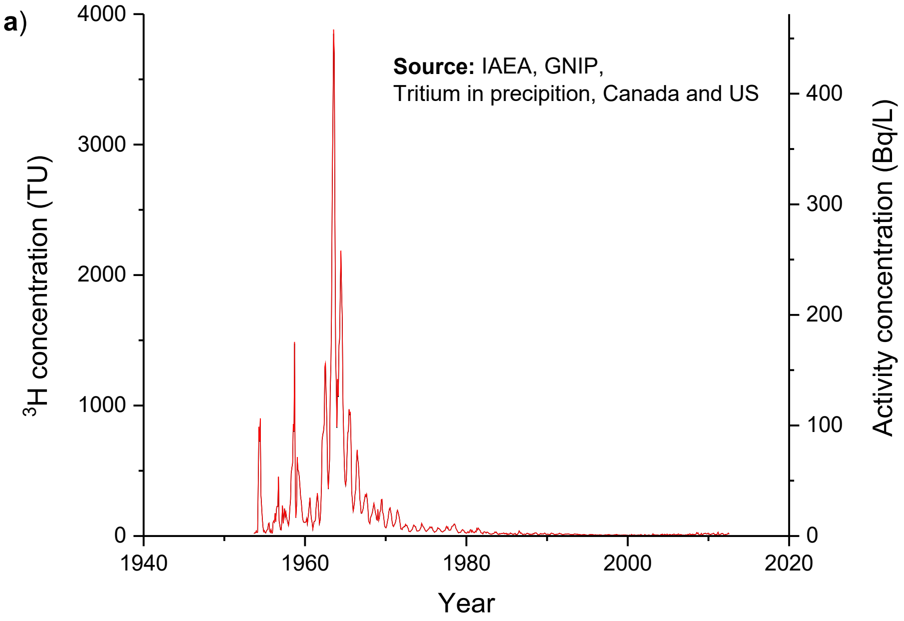
The measurements are complementary to the background assessment of the ESS site performed during 2017 (ESS project *ESS-0093103*) [2].

As a first part of the report, some previous measurements of tritium in different environments are presented.

2. TRITIUM ACTIVITY CONCENTRATIONS IN THE ENVIRONMENT

The prevailing form of tritium in the environment is as water. Thus tritium is transported through the various phases of the hydrological cycle [3, 4]. The natural background level of tritium, resulting from nuclear reactions involving cosmic radiation and atmospheric gases, is between 0.1 and 0.6 Bq/L of water [5].

Man-made tritium mainly originates from testing of hydrogen bombs: Figure 1a shows the tritium activity concentration in precipitation in Canada and the US since middle of the last century. Figure 1b shows the corresponding values from 1990 to 2012. The observed seasonal variations are partly due to the "spring leak", which results from the exchange of tropospheric and stratospheric air masses during winter and spring (see [6] and references therein). Figure 1b indicates that levels from < 1 Bq/L up to a few Bq/L can be expected in precipitation, and also in moisture in air.



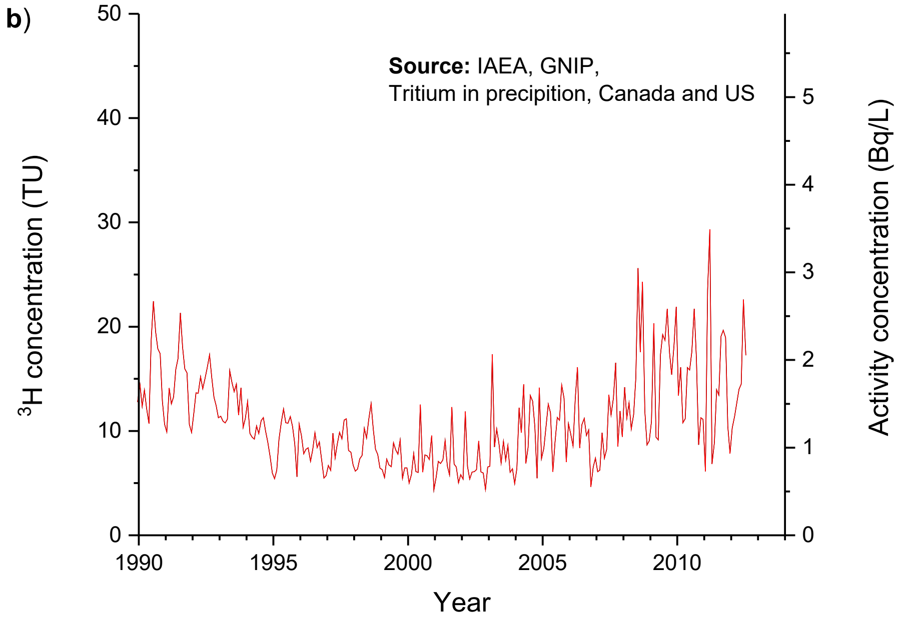


Figure 8 Tritium concentration in precipitation in Canada and the US [7] a) during the bomb-pulse era, b) from 1990 to 2012. 1 TU = 1 Tritium Unit, which equals 1 tritium atom per 10^{18} ^1H atoms, corresponding to 0.118 Bq/L of water.

A few datasets of tritium in Swedish waters are available. Since year 2001, the Swedish Radiation Safety Authority (SSM) collects water samples twice a year at six Swedish waterworks, none however in the southernmost Sweden [8]. The tritium activity concentrations are often below the detection limits of the instruments used (general a few Bq/L, denoted as "< value" in Figure 2, which presents data from two of the waterworks monitored). Typical tritium activity concentrations are from below 1 Bq/L up to a few Bq/L, i.e. similar to the data from Canada and the US in Figure 1.

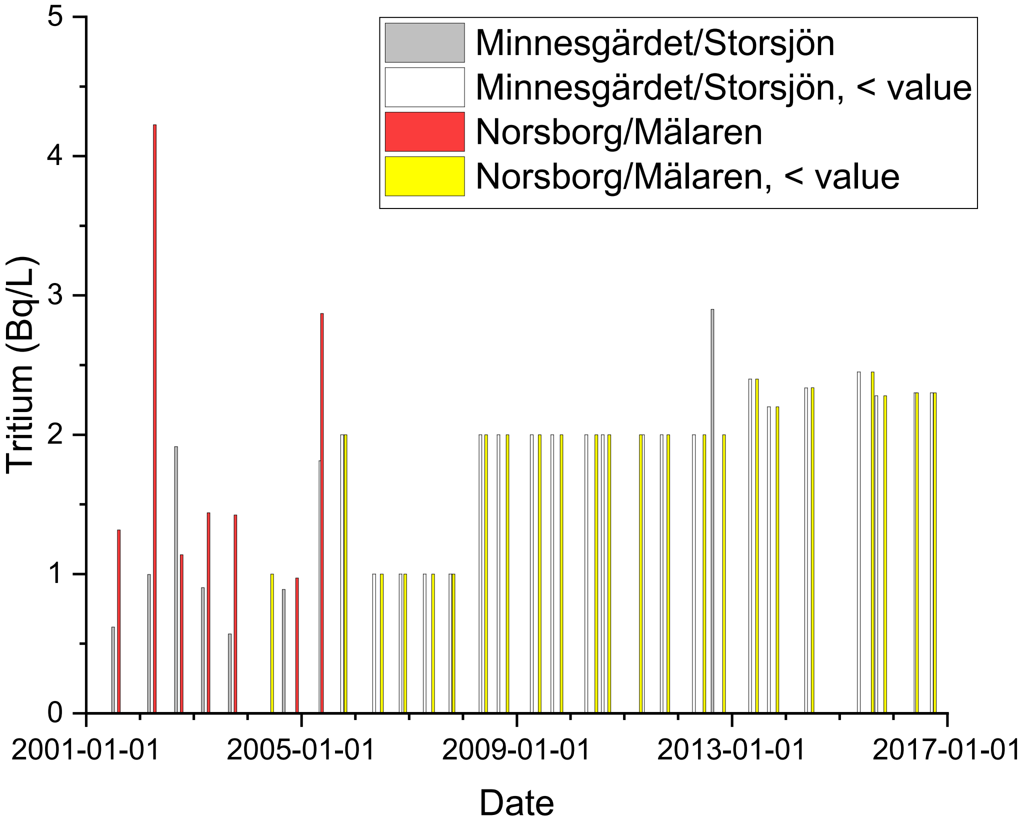


Figure 9 **Tritium activity concentration in drinking water [8].**

Local contamination of tritium can be observed around nuclear power plants, in particular in the environment of heavy water reactors (typically some tens of Bq/L of water) [5]. As part of the national programme for the environmental control at the Swedish nuclear facilities, the Swedish Radiation Safety Authority (SSM) monitors tritium in surface sea waters at six monitoring stations since 2009 [9]. Figure 3 shows the tritium activity concentration in two of the stations, Ringhals in Kattegatt and Fjällbacka in Skagerrak [9]. The Ringhals sampling location (R35) lies 8.2 km from the cooling water release point of the Ringhals nuclear power plant [10]. Fjällbacka is located approximately 160 km in the northerly direction along the coast from Ringhals. The majority of the data in Figure 3 are below the detection limit of a few Bq/L.

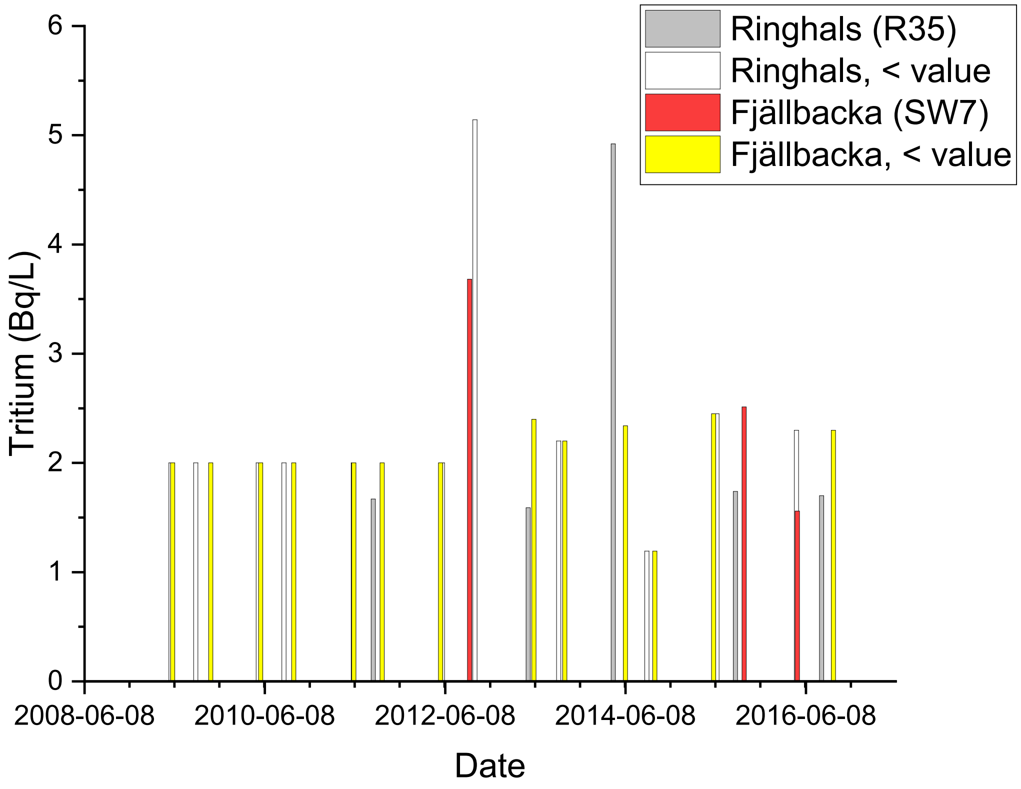


Figure 10 **Tritium activity concentration in sea water [9].**

3. MATERIALS AND METHODS

3.1. Sampling sites

The sampling sites are described in Table 1 and depicted in Figure 4. The Grevie PV5 well (site T0), operated by VA Syd, is used as background water for the liquid scintillation (LSC) analysis. In a previous study by Åkesson et al [11], the tritium activity concentration of this well has been measured as 0.02 TU, corresponding to 0.002 Bq/L. Measurements of ^{39}Ar in the water of this well indicates a mean residence time of 120 years [11]. Site T1 is used as an urban reference site. This site is identical to the carbon-14 sampling site C2 in the final report of the ESS project *ESS-0093103* [2]. The location of site T2 within the ESS site is shown in Figure 5.

Table 4 Description of sampling sites for tritium in water, air and precipitation.

Site	Description	GPS coordinates	Sample type
T0	Grevie PV5 well (Background water)	N55.6131 E13.1970	Water from deep well
T1	Timjanvägen 5, Lund (Urban background site)	N55.7186 E13.1827	Air humidity Precipitation
T2	ESS site, close to pond 4	N55.7366 E13.2454	Air humidity Precipitation

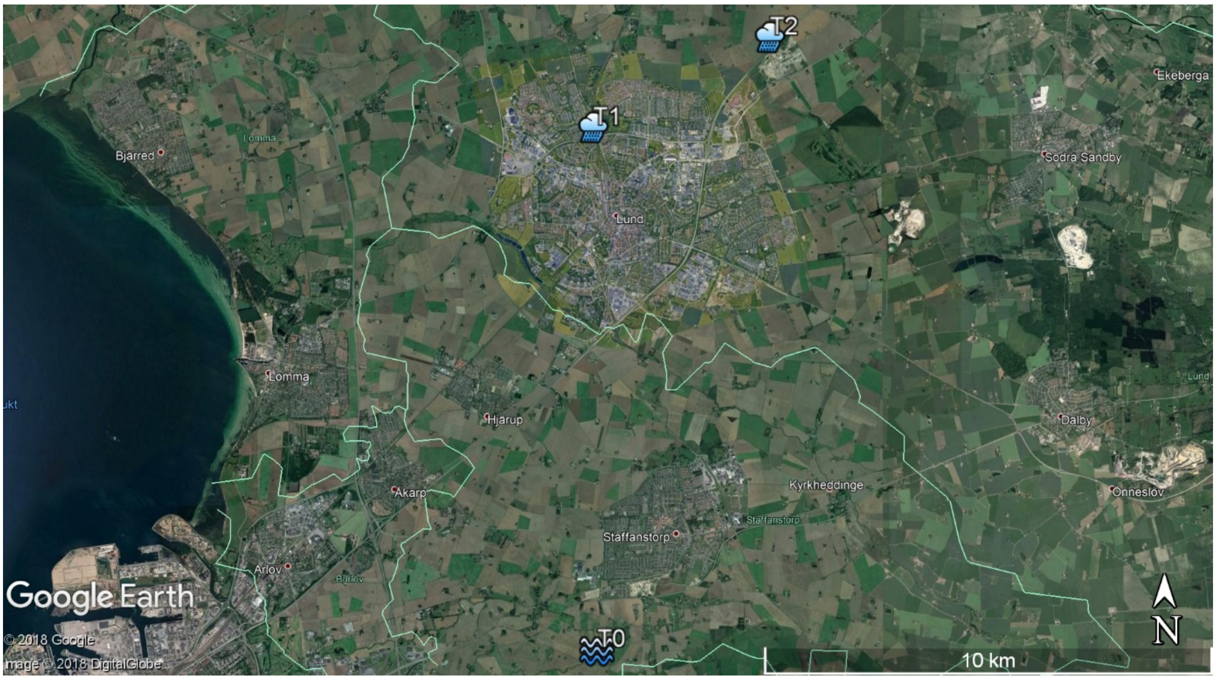


Figure 11 Map of tritium sampling sites.

Figure 12 Position of the rain and precipitation samplers at the ESS site (T2).

3.2. Sampling procedure

The sampling procedure is described in detail in Ref [12]. In brief, the procedure is applicable to:

- Task #1: Grab sampling of air humidity using an air dehumidifier (Wood's MRD10 [13]).
- Task #2: Continuous collection of precipitation using a precipitation sampler (Palmex standard rain sampler RS1 [14]).

3.3. Measurement

Samples were analysed with LSC at Lund University (LU) using a Beckman LS 6500 counter. 10 mL of water and 10 mL of Ultima Gold LLT LSC cocktail were used for each sample. The basic counting time of samples as well as background was 600 minutes. A few samples were measured twice to increase counting statistics, one sample was measured 3 times and another sample was measured 4 times. The output of the analysis was counts per minute (cpm), the uncertainty on this count (%) and the quench level given by Horrock's parameter value (#H) [15]. Efficiency of each measurement was determined using Horrocks's method for quenching correction [16]. Further information of the measurement procedure can be found in Ref [12].

3.4. Data analysis

Details of the data analysis can be found in Ref [12].

4. RESULTS AND DISCUSSION

4.1. Minimum detectable activity

The count rate of the background water sample (V = 10 ml) from site T0 was on average 5.01±0.13 cpm and the efficiency E was 22.43±0.32 %. The minimum detectable activity, MDA, calculated according to Ref. [12], was found to be 3.2 Bq/L of water for a counting time of 600 minutes for sample as well as background. The MDAs for the samples measured twice, three times and four times were 2.8 Bq/L, 2.6 Bq/L and 2.5 Bq/L, respectively.

The MDA from 600 minutes of measuring time fulfils the requirements in Refs [17] and [18], stating that the detection limit must be less than 10^5 Bq/m^3 (100 Bq/L) for instruments measuring releases of tritium to water and less than 10^3 Bq/m^3 for releases to air. Using an air humidity of 8 g/m^3 [19] and a density of 1 kg/L, the latter requirement corresponds to a required detection limit of 125 Bq/L.

4.2. Air humidity (Task #1)

Table 2 shows details during sampling of air humidity as well as results of the LSC measurement. None of the samples shows an HTO activity above the MDA, as can be expected at uncontaminated sites (see Figure 1).

Table 5 Parameters during sampling of air humidity and results of LSC measurements using 10 ml of sampled water (Task #1). Uncertainty corresponds to 1σ. MDA = 3.2 Bq/L.

Site	Sample id	Date	Time	Temp $T_{°C}$ (°C)	Relative humidity *RH* (%)	Absolute humidity *AH* [1] (kg/m^3)	Mass of sampled water *m* (g)	HTO conc. in water A [1] (Bq/L)	HTO conc. in air A_{air} [1] (Bq/m^3)
T1	H4	2018-04-13	10:50-11:50 (60 min)	14	55	0.0066	30	<*MDA*	<0.02
T2	H5 [2]	2018-04-13	13:35-14:30 (55 min)	17.6	48	0.0072	56	<*MDA* [2]	<0.02
T2	H6 [3]	2018-04-23	11:23-12:09 (46 min)	16.9	64	0.0092	81	<*MDA* [3]	<0.02

[1] *Calculated according to Ref [12].*

[2] *Measured four times, MDA = 2.5 Bq/L.*

[3] *Measured three times, MDA = 2.6 Bq/L.*

4.3. Precipitation (Task #2)

Table 3 shows details during sampling of precipitation as well as results of the LSC measurement. Only one sample (sample P7 collected at the ESS site T2) shows an HTO activity above the MDA. The observed value is of the same order of magnitude as can be expected at uncontaminated sites (see Figure 1).

Table 6 Parameters during sampling of precipitation and results of LSC measurements using 10 ml of sampled water (Task #2). Uncertainty corresponds to 1σ. MDA = 3.2 Bq/L.

Site	Sample id	Date interval	Nr of days	Type of precipitation	Mass of sampled water m (g)	HTO conc. in water A [1] (Bq/L)
T1	P2 [2]	2018-03-19 2018-03-20	1	Snow	20 [a]	*<MDA* [2]
T1	P3 [2]	2018-03-20 2018-03-23	2.5	Snow	225 [a]	*<MDA* [2]
T1	P4	2018-03-23 2018-04-06	14	Snow and rain	706 [b]	*<MDA*
T1	P6	2018-04-08 2018-04-13	5	Rain	Not registered	*<MDA*
T2	P7	2018-04-13 2018-04-23	10	Rain	41 [b]	3.8 ± 1.0
T2	P8 [2]	2018-04-23 2018-05-03	10	Rain	732 [b]	*<MDA* [2]

[1] *Calculated according to Ref [12].*

[2] *Measured twice, MDA = 2.8 Bq/L.*

[a] *Using the snow tube (diameter 15 cm).*

[b] *Using the funnel with diameter 23 cm.*

ND No data

5. VALIDATION

Some of the samples were additionally analysed at CARER (Centre for Advanced Research in Environmental Radioactivity at Mangalore University, India) using a LSC Quantulus-1220 counter (counting time 300 min, 2 trials). The background count rate was 0.70 cpm (obtained from measurements of water from the Grevie well/site T0), resulting in an MDA of 1.13 Bq/L at 99% confidence level.

Table 7 Comparison of results obtained from CARER and LU.

Site	Sample id	HTO concentration in water, CARER *A* (Bq/L)	HTO concentration in water, LU *A* (Bq/L)	Agreement between CARER and LU
T2	H4	< 1.13 (CARER MDA)	< 3.2 (LU MDA)	OK
T2	H5	< 1.13 (CARER MDA)	< 2.5 (LU MDA)	OK
T2	H6	1.7 ± 0.6	< 2.6 (LU MDA)	OK
T2	P7	1.3 ± 0.6	3.8 ± 1.0	Overlap within 3σ
T2	P8	1.7 ± 0.6	< 2.6 (LU MDA)	OK

For sample P7 the LU and CARER results overlap within 3σ. The lower detection limit of the Quantulus-1220 counter (CARER) compared to the Beckman 6500 LS instrument (LU) is expected due to the lower instrumental background of the Quantulus-1200 system.

6. CONCLUSION

The observed levels of tritium in air and precipitation show no evidence of any local contamination of tritium in the form of HTO during the monitored period. Reduction of the MDAs requires further investigations.

7. ACKNOWLEDGEMENT

The authors thank VA Syd for giving access to the background water from the Grevie PV5 well. The authors are grateful for the validation measurements performed at Centre for Advanced Research in Environmental Radioactivity (CARER), Mangalore University, India, by Professor Karunakara Naregundi and his team. The authors also thank Associate Professor Christopher Rääf, Medical Radiation Physics (Malmö) for constructive reviewing of the report.

8. GLOSSARY

Term	Definition
CARER	Centre for Advanced Research in Environmental Radioactivity at Mangalore University, India
HTO	Tritium-containing water
LSC	Liquid Scintillation Counting
LU	Lund University, Sweden

9. REFERENCES

[1] Ene, D., Andersson, K., Jensen, M., Nielsen, S., Severin, G. *Management of tritium in European Spallation Source.* Fusion Science and Technology, 67(2): 324-327, 2015.

[2] Bernhardsson, C., Eriksson Stenström, K. *Point zero measurements for the ESS facility.* Report in preparation. Lund University. 2018.

[3] UNSCEAR. United Nations. *Sources and effects of ionizing radiation. UNSCEAR 2000 report to the General Assembly, with scientific annexes. Volume I: Sources*. 2000.

[4] UNSCEAR. United Nations. *Sources, effects and risks of ionizing radiation. UNSCEAR 2016. Report to the General Assembly, with scientific annexes. Annex C. Biological effects of selected internal emitters - tritium*. 2016.

[5] IRSN. *Tritium and the environment*. Available at http://www.irsn.fr/EN/Research/publications-documentation/radionuclides-sheets/environment/Pages/Tritium-environment.aspx. 2012. Accessed: 2018-08-20.

[6] Ducros, L., Eyrolle, F., Vedova, C.D., Charmasson, S., Leblanc, M., Mayer, A., Babic, M., Antonelli, C., Mourier, D., Giner, F. *Tritium in river waters from French Mediterranean catchments: Background levels and variability.* Science of The Total Environment, 612(Supplement C): 672-682, 2018.

[7] IAEA/WMO. *Global Network of Isotopes in Precipitation. The GNIP database*. Accessed: 2017-12-01. Available from: http://www.iaea.org/water.

[8] SSM. *Miljödatabasen/Dricksvatten*. Accessed: 2018-05-29. Available from: https://www.stralsakerhetsmyndigheten.se/omraden/miljoovervakning/sokbara-miljodata/miljodatabasen/dricksvatten/.

[9] SSM. *Miljödatabasen/Havsvatten*. Accessed: 2018-05-29. Available from: https://www.stralsakerhetsmyndigheten.se/omraden/miljoovervakning/sokbara-miljodata/miljodatabasen/havsvatten/.

[10] Lindén, A.-M. The Swedish Radiation Safety Authority. *2004:15 Omgivningskontrollprogram för de kärntekniska anläggningarna, revision*. ISSN 0282-4434, 2004.

[11] 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.

[12] Eriksson Stenström, K., Pedehontaa-Hiaa, G., Barkauskas, V. *Procedure: Collection of precipitation and air humidity and subsequent measurement of tritium.* BAR2018/02. Lund University, Department of Physics, Division of Nuclear Physics. 2018.

[13] Wood's. Accessed: 2018-06-13. Available from: https://woods.se/en/Produkter/dehumidifiers/woods-mrd10/.

[14] Palmex. Accessed: 2018-06-13. Available from: http://www.rainsampler.com/portfolio-page/rain-sampler-rs1/.

[15] *Scintillation system LS 6500 Operating instructions*. Beckman Coulter, Inc, USA. Report,1999.

[16] Horrocks, D., *Applications of liquid scintillation counting*. Elsevier, 1974.

[17] *Special conditions for the ESS facility in Lund.* Vers. 2, ESS-0018828. Swedish Radiation Safety Authority. 2015.

[18] European Union. *COMMISSION RECOMMENDATION of 18 December 2003 on standardised information on radioactive airborne and liquid discharges into the environment from nuclear power reactors and reprocessing plants in normal operation*. 2004/2/Euratom, 2004.

[19] Ene, D. *Environmental impact analysis.* ESS-0001898. 2012.

APPENDIX 3. DATA FROM MEASUREMENTS 2018

Available upon request. Please contact the authors.