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## **Forsmark site investigation**

# **Analysis of radioactive isotopes in near surface groundwater, surface water, biota and soil**

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December 2009

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## Abstract

This report describes complementary analyses of radioactive isotopes in surface waters, groundwaters, biota and soil in the Forsmark investigation area. The study can be divided into two parts. In the first part of the study, radon concentrations were measured in near surface groundwaters. The radon analyses were carried out in the field with a portable radon detector. The purpose was to obtain a spatial distribution of the radon concentrations in near surface groundwaters and to examine the possibility to use radon as a tracer for groundwater flow patterns. In the second part, complementary analyses of radium, uranium and thorium isotopes were conducted in surface waters, soil and vegetation. These isotope analyses were performed by Risø National Laboratory, Roskilde, Denmark. The purpose was to obtain complementary isotope data by using analytical methods with especially low detection limits. The already existing amount of data was considered insufficient, and earlier values were often below the reported detection limit.

The results from the first part of the study show largely varying radon concentrations in the investigation area, but they are still within the normal range for Swedish granitic bedrock areas. The results of the isotope analyses in surface water, vegetation and soil are in the same order of magnitude as the earlier isotope analyses conducted by SKB within the Forsmark area. The  $^{235}\text{U}/^{238}\text{U}$  ratios indicated a natural uranium isotope composition.

# Sammanfattning

Denna rapport beskriver kompletterande analyser av radioaktiva isotoper i ytvatten, grundvatten, biota och jord i Forsmarks undersökningsområde. Studien kan delas in i två delar. I den första delen har radonhalten mätts i ytnära grundvatten. Radonmätningarna gjordes i fält med en portabel radondetektor. Syfte med mätningarna var att få en spatial bild av radonkoncentrationen i det ytnära grundvattnet och att undersöka möjligheten att använda radonhalter för att spåra grundvattenflöden. I den andra delen av studien gjordes kompletterande analyser av radium, uran och thoriumisotoper i ytvatten, jord och vegetation. Dessa analyser utfördes på Risø Forskningscenter, Roskilde, Danmark. Syftet var att erhålla kompletterande isotopdata genom att använda analysmetoder med särskilt låga detektionsgränser. Befintlig datamängd ansågs otillräcklig, och tidigare värden låg ofta under detektionsgränsen.

Resultaten från den första delen av studien visade på kraftigt varierande radonhalter inom det undersökta området. Radonhalten låg dock inom det normala intervallet för granitdominerade områden i Sverige. Resultaten från isotopanalyserna i ytvatten, vegetation och jord visade värden i samma storleksordning som tidigare isotopanalyser gjorda av SKB i Forsmarksområdet. Kvoten  $^{235}\text{U}/^{238}\text{U}$  indikerade en naturlig uranisotopsammansättning.

# Contents

<b>1</b>	<b>Introduction</b>	7
<b>2</b>	<b>Objective and scope</b>	9
<b>3</b>	<b>Samples</b>	11
3.1	Radon measurements in near surface groundwater	11
3.2	Isotope analyses in surface waters, biota and soil	12
<b>4</b>	<b>Equipment</b>	17
4.1	RAD-7	17
4.2	Liquid scintillation counter (LSC)	17
4.3	Alpha spectrometry	19
4.4	Inductively Coupled Plasma mass spectrometry (ICP-MS)	19
<b>5</b>	<b>Execution</b>	21
5.1	Radon field measurements	21
5.2	Radium, uranium and thorium isotope analyses	21
	5.2.1 Collection of samples	21
	5.2.2 Laboratory analyses	21
5.3	Nonconformities	22
<b>6</b>	<b>Results</b>	23
	<b>References</b>	25
	<b>Appendix 1</b> Radioisotope data	27
	<b>Appendix 2</b> Species list for biota samples	31

# 1 Introduction

This document reports the results gained by the activity *Analyses of radioactive isotopes in surface water, groundwater, biota and soil (Analys av radioaktiva isotoper i ytvatten grundvatten, biota och jord)*, which is a complementary activity performed after the site investigation at Forsmark. The work was carried out in accordance with the activity plan AP PF 400-09-002. Controlling documents for performing this activity are listed in Table 1-1. Activity plans are SKB's internal controlling documents.

Sections 4.1– 4.3 and 5.2.2 in this report describe analytical equipments and methods and have been written by Per Roos at Risø National Laboratory, Roskilde, Denmark.

This study performed can be divided into two parts. In the first part of the study radon concentrations were measured in near surface groundwater in order to get a spatial picture of the radon concentration in groundwater. The radon analyses were conducted in the field with a portable radon detector described in Section 4.1. This activity was initiated to obtain an extensive set of radon concentration data in near surface groundwaters. This dataset was used to investigate the possibility to trace groundwater flow patterns using radon as a tracer. During this work it was discovered that many of the reported radon concentration values determined during the early site investigation were not corrected for the decay of radon that takes place between the time of sampling and the time of analyses. This caused significant errors in the radon concentration determinations. The reported radon concentrations that were not corrected for this decay were within the scope of this study identified and marked as unreliable in the database Sicada.

Complementary radon measurements, similar to the ones performed here, were also carried out in near surface groundwaters at Laxemar. This activity was included in the monitoring programme and will be reported in a separate P-report.

In the second part of this study, complementary analysis of radioactive isotopes in surface water, soil and vegetation were made. The isotope analyses were performed by Risø National Laboratory, Roskilde, Denmark. This study was initiated to gain data in addition to the already existing data set. Some radium isotope analyses performed in the early part of the site investigation were proved unreliable, since the reported detection limits were too high to provide reliable results. After a change of laboratory the detection limit was lowered. However, the new detection limit was still too high for radium in surface water. The early radium measurements were marked as unreliable in the Sicada database and will not be used for further analyses.

Original data from the reported activities in this study are stored in the primary database Sicada. Data are traceable in Sicada by the Activity plan numbers AP PF 400-09-002. Only data in databases are accepted for further interpretation and modelling. The data presented in this report are regarded as copies of the original data. Data in the databases may be revised, if needed. Such revisions will not necessarily result in a revision of the P-report, although the normal procedure is that major revisions entail a revision of the P-report. Minor revisions are normally presented as supplements, available at [www.skb.se](http://www.skb.se).

**Table 1-1. Controlling documents for the performance of the activity.**

Activity plan	Number	Version
Analys av radioaktiva isotoper i ytvatten, grundvatten, biota och jord	AP PF 400-09-002	1.09

## 2 Objective and scope

The purpose of the first part of the study was to obtain spatial information on the radon concentration in near surface groundwaters and to examine the possibility to use radon as a tracer for groundwater flow patterns. The objective was also to evaluate the quality of the radon measurements performed with the portable radon detector RAD-7. This report contains the results of the measurements which are further discussed in /Grolander 2010/.

The purpose of the second part of the study was to obtain additional radium, uranium and thorium isotope data from surface waters, biota and soil as a complement to the already available dataset. The results from previous determinations of these isotopes in different material are few and, especially for surface waters, of limited use since many reported values fall below the detection limit. The isotope information will be used to gain a general understanding of the distribution of isotopes in the natural environment at Forsmark. The results will also be used for parameterisation of site specific parameters used in the transport modelling. The isotope analyses are listed in Table 2-1.

**Table 2-1. Isotopes analysed in surface waters, biota and soil.**

<b>Isotopes analysed</b>	226Ra	228Th	230Th	232Th	233U	234U	235U	236U	238U
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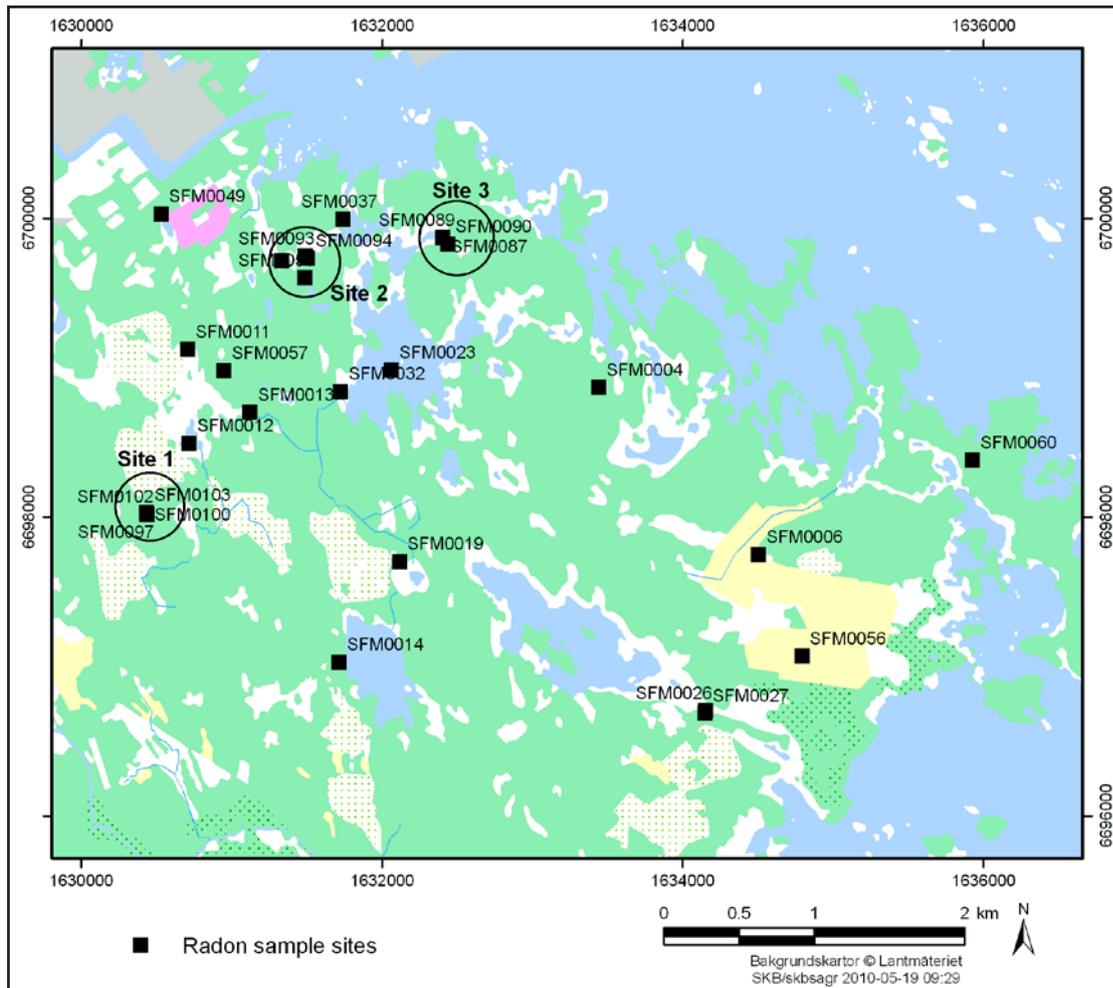
## 3 Samples

### 3.1 Radon measurements in near surface groundwater

The radon samples were collected in groundwater monitoring wells in Quaternary deposits. The sampling was performed during two sampling campaigns: the first in October 2008 and the second in June 2009. Sampling locations and sampling dates are presented in Table 3-1. The locations of the sampled monitoring wells are displayed in Figure 3-1. At three locations, marked with Site 1, Site 2 and Site 3 in Figure 3-1, clusters of monitoring wells were sampled. At these locations several wells were installed with different screening depths. The installed wells were steel pipe pumping wells, HDPE (High Density PolyEthylene) groundwater monitoring wells or wells supplied with BAT-type filter tips. (The special sampling in BAT-type wells is described in Section 5.1.). The well types are listed in Table 3-1. Sample size was 250 ml for all samples, except for some samples collected in BAT-type wells, where only 40 ml of water could be extracted. Also the sample size is listed in Table 3-1.

**Table 3-1. List of sampling locations, sampling dates, sample size and well type for radon samples collected in near surface groundwaters. The wells SFM0013, SFM0014, SFM0019 and SFM0026 were sampled at two occasions.**

IDCODE	Sampling date	Sample size (ml)	Well type
SFM0001	2008-10-09	250	HDPE pipe
SFM0004	2008-10-10	250	HDPE pipe
SFM0006	2008-10-08	250	HDPE pipe
SFM0011	2008-10-09	250	HDPE pipe
SFM0012	2008-10-10	250	HDPE pipe
SFM0013	2008-10-09	250	HDPE pipe
SFM0013	2009-06-11	250	HDPE pipe
SFM0014	2008-10-10	250	HDPE pipe
SFM0014	2009-06-14	250	HDPE pipe
SFM0019	2008-10-10	250	HDPE pipe
SFM0019	2009-06-15	250	HDPE pipe
SFM0023	2008-10-09	250	HDPE pipe
SFM0026	2008-10-09	250	HDPE pipe
SFM0026	2009-06-11	250	HDPE pipe
SFM0027	2008-10-09	250	HDPE pipe
SFM0032	2008-10-07	250	HDPE pipe
SFM0037	2008-10-08	250	HDPE pipe
SFM0049	2008-10-07	250	HDPE pipe
SFM0051	2008-10-08	250	HDPE pipe
SFM0056	2008-10-10	250	HDPE pipe
SFM0057	2008-10-09	250	HDPE pipe
SFM0060	2008-10-09	250	HDPE pipe
SFM0084	2009-06-12	250	HDPE pipe
SFM0086	2009-06-16	40	BAT
SFM0087	2009-06-12	250	HDPE pipe
SFM0089	2009-06-16	40	BAT
SFM0090	2009-06-12	250	Steel pipe
SFM0091	2009-06-09	250	HDPE pipe
SFM0093	2009-06-12	40	BAT
SFM0094	2009-06-09	250	Steel pipe
SFM0095	2009-06-09	250	HDPE pipe
SFM0097	2009-06-10	250	BAT
SFM0100	2009-06-12	40	BAT
SFM0102	2009-06-09	250	BAT
SFM0103	2009-06-09	250	Steel pipe



**Figure 3-1.** The sampling locations for the radon measurements in near surface groundwater. SFM0013, SFM0014, SFM0019 and SFM0026 have been sampled both in June 2009 and October 2008. Site 1, Site 2 and Site 3 represent clusters of monitoring wells, where several wells were samples with different screening depths.

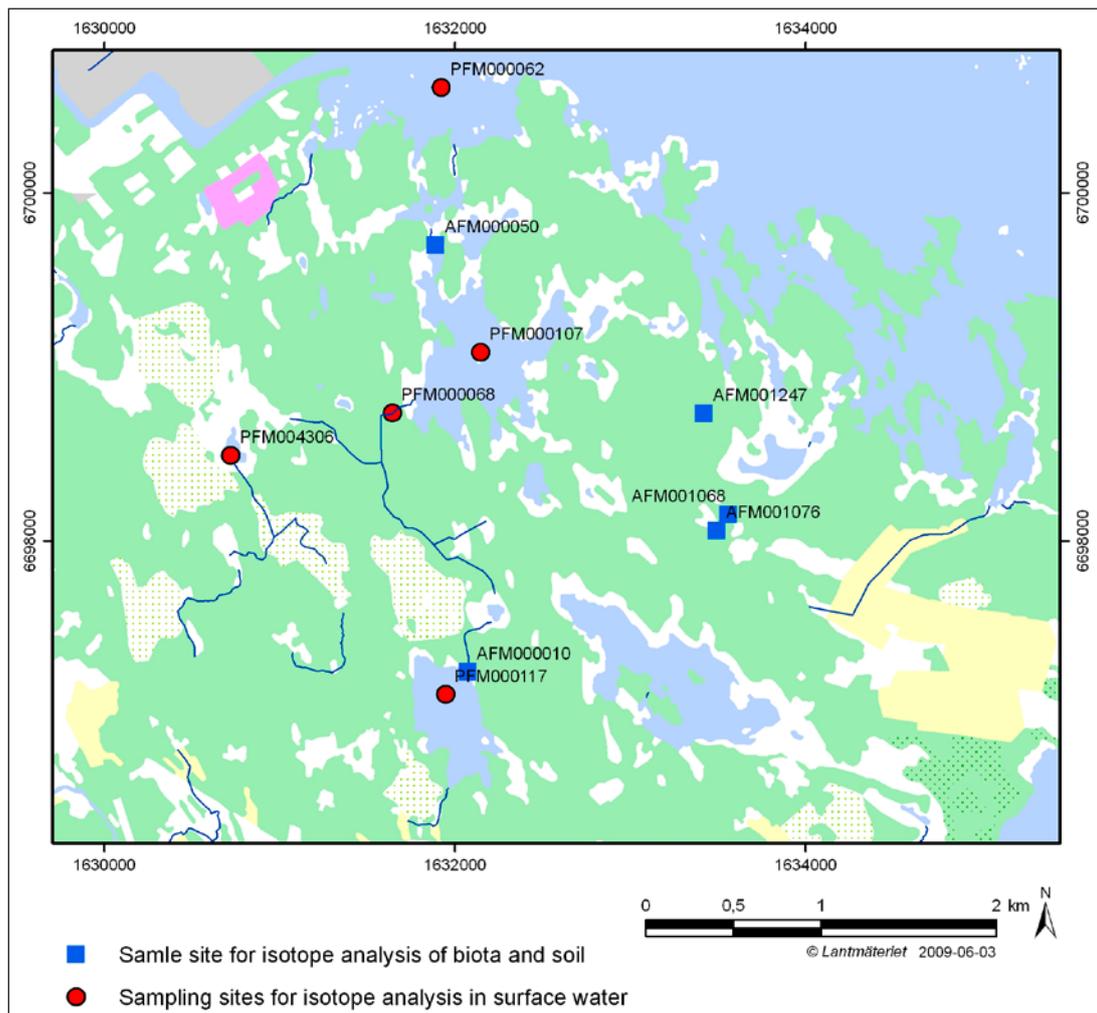
### 3.2 Isotope analyses in surface waters, biota and soil

Samples for radium, uranium and thorium isotope analyses were collected from terrestrial, limnic, and marine environments and consisted of terrestrial and aquatic vegetation, soil and surface waters. The samples are listed in Table 3-2 and information of sampling sites, sample type, sample volume or weight and sampling dates are included. Swedish and English names of the biota samples are presented in Appendix 2.

The sampling locations are presented in Figure 3-2. Soil and terrestrial vegetation samples were collected at three locations (AFM1247, AFM1076 and AFM1068) that have been used for biota and soil sampling in previous investigations. Photos from these sites are presented in Figure 3-3, Figure 3-4 and Figure 3-4, respectively. Limnic biota samples were collected from the two lakes Eckarfjärden (AFM0010) and Bolundsfjärden (AFM0050). Photos from these lakes are displayed in Figure 3-5 and Figure 3-6. Location codes starting with AFM embody area codes covering a defined area. For lakes the area codes represent the whole lake, whereas the markings seen in Figure 3-1 represent the sampling area more precisely, even though the samples were taken from an area and not from a defined point.

**Table 3-2. List of samples for radium, uranium and thorium isotope analyses with short description.**

Sampling Site	Sample type	Sample size Wet weight (g) Volume (L)	Sampling date
<b>Soil</b>			
AFM1247	Pooled soil sample	<2,000 g	2009-06-10
AFM1076	Pooled soil sample	<2,000 g	2009-06-10
AFM1068	Pooled soil sample	<2,000 g	2009-06-10
<b>Biota</b>			
AFM1247	Vaccinium myrtillus	933 g	2009-06-10
AFM1076	Picea abies	698 g	2009-06-10
AFM1076	Carex elata	940 g	2009-06-10
AFM1076	Rubus saxatilis	530 g	2009-06-10
AFM1068	Maianthemum biofolium	297 g	2009-06-10
AFM0050	Chara sp.	1,320 g	2009-06-10
AFM0010	Chara sp.	1,397 g	2009-06-10
<b>Surface water</b>			
PFM0107	Lake water	50 L	2009-06-15
PFM0117	Lake water	50 L	2009-06-15
PFM0062	Sea water	50 L	2009-06-15
PFM0068	Stream water	50 L	2009-06-15
PFM4306	Lake water	50 L	2009-06-16



**Figure 3-2.** The sampling locations for soil, biota and surface water samples collected for determinations of radium, uranium and thorium isotopes.



**Figure 3-3.** Sampling location AFM1247 where a pooled soil sample and a *Vaccinium myrtillus* (blueberry) sample were collected.



**Figure 3-4.** Sampling location AFM1076 where a pooled soil sample and three biota samples of *Picea abies* (Norway spruce), *Rubus saxatilis* (stone bramble) and *Carex elata* (sedge) were collected.



**Figure 3-5.** Sampling location AFM1068 where a pooled soil sample and a *Maianthemum biofolium* (May lily) sample were collected.



**Figure 3-6.** Lake Eckarfjärden from where a water sample (PFM0117) and a biota sample of *Chara* sp. (stonewort) were collected (AFM0010).



**Figure 3-7.** Lake Bolundsfjärden from where a water sample (PFM0107) and a biota sample of *Chara* sp.(stonewort) were collected (AFM0050).

## 4 Equipment

In this study the in-air radon monitor RAD-7 was used to measure radon concentrations extracted from water in the field. The RAD-7 device is explained in Section 4.1 below. Radium, thorium and uranium isotopes were analysed at Risø DTU using a Liquid Scintillation Counter (LSC) for  $^{226}\text{Ra}$  analyses, alpha spectrometry for  $^{228}\text{Th}$  analyses and Inductively Coupled Plasma mass spectrometry (ICP-MS) for  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{230}\text{Th}$  analyses. These analyse equipments are described in Sections 4.2, 4.3 and 4.4 respectively.

### 4.1 RAD-7

RAD-7 is a commercially used device to determine radon concentration in air mostly for indoor-air radon measurements. The RAD-7 is an alpha particle detector that uses a solid state detector to detect alpha particles with different energy levels. A solid state alpha detector can convert the energy from an alpha particle to an electrical signal. This makes it possible to identify the specific element that decays since every element emits alpha particles with different energy levels.

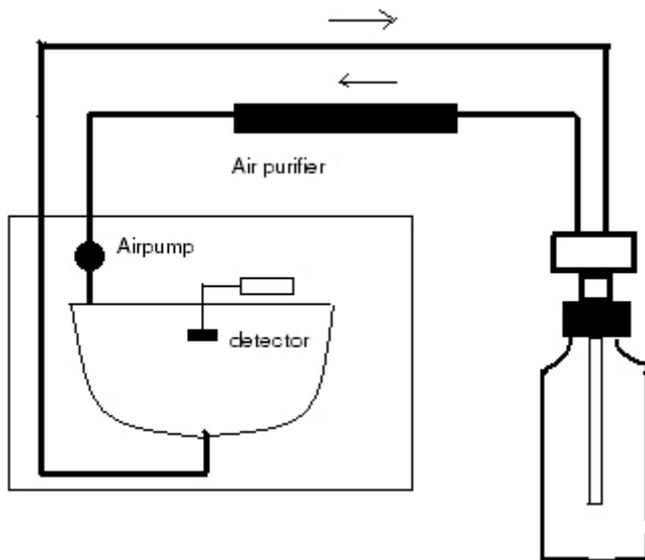
The inside of the RAD-7 consists of a hemisphere that is coated with an electrical conductor. An ion-implanted silicon alpha detector is placed in the centre of the hemisphere as shown in Figure 4-4. A high voltage power circuit creates a high electrostatic field within the measurement chamber. This electrostatic field makes the positively charged radon daughters ( $^{214}\text{Po}$  and  $^{218}\text{Po}$ ) absorbed onto the surface of the detector. This means that when the polonium isotopes decay and emit alpha particles, the alpha particles enter the detector, and an electrical signal with a specific energy level will be produced and recorded. The radon concentration is then calculated from the decay of the radon daughters  $^{214}\text{Po}$  and  $^{218}\text{Po}$  and not from the decay of radon if self. This technique gives the RAD-7 a quick response time, since the half-life of the  $^{218}\text{Po}$  is only 3.05 minutes, and secular equilibrium is reached after only 12 minutes.

There is an accessory to the RAD-7 that makes it possible to measure radon in water. It is called H<sub>2</sub>O accessory and has been used at laboratories for more than a decade. The detection limit is 0.370 Bq/L /DurrIDGE Company 2000/. A picture of the H<sub>2</sub>O accessory is shown in Figure 4-4. The water sample, which is collected in a 250 ml vial, is aerated when an air pump inside the RAD-7 chamber pumps air into the vial through a plastic tube. The air is then lead via a gas purifier into the RAD-7 chamber. This creates a closed loop of air between the vial and the RAD-7 chamber. The RAD-7 chamber is sensitive to moisture, and the relative humidity has to be kept below 10% to obtain good measurement results. Therefore the gas purifier shown in Figure 4-4 is used to absorb all moisture in the ingoing air. The gas purifier is a plastic cylinder filled with CaSO<sub>4</sub> that absorbs water effectively.

The water sample is aerated for five minutes to get all the radon from the water into the air circulation. After the first five minutes of aeration, the RAD-7 waits for five minutes to get the  $^{218}\text{Po}$  in equilibrium with the radon. Then the decay of  $^{218}\text{Po}$  is counted for five minutes and a result is printed. The results include, besides the radon concentration, also a statistical uncertainty for the measurement. After this first reading, three more five-minutes reading are made, and a mean value for the radon concentration in the four measurements cycles is presented, as well as a standard deviation value calculated for the four radon measurements.

### 4.2 Liquid scintillation counter (LSC)

The Quantulus 1220 (Perkin Elmer), shown in Figure 4-2, is an ultra low level liquid scintillation spectrometer, which enables measurements normally only possible in special underground installations. This is made possible by a combination of a massive lead shielding and an active anti-coincidence shielding through a large scintillation tank surrounding the measurement chamber. This instrument was used for the measurement of  $^{226}\text{Ra}$ . The instrument separates events originating from low-density ionisation (electrons, X-ray and gamma-photons) and high-density ionisations (alpha particles) by analysing the light decay from individual scintillation events. The data from the Quantulus LSC are



*Figure 4-1. Schematic picture of the RAD-7 H<sub>2</sub>O accessory (after /DurrIDGE Company 2000/).*



*Figure 4-2. The Quantulus LSC is the cabinet shown in the background behind a set-up of the ordinary anticoincidence shielded GM gas flow counters in a lead cave in the foreground.*

normally corrected individually for each sample based on the so called quenching parameter, which is determined internally by a standard gamma source irradiating the sample with a well-known gamma ray fluence. Apart from the quenching parameter, the cross-talk between the alpha and beta particle windows is adjusted by monitoring alpha and beta events for a standard source with various pulse shape analyser settings (PSA-settings). Counting efficiency for the LSC was determined by a certified <sup>226</sup>Ra standard solution.

A P-type germanium detector (60% efficiency relative a 3×3 inch NaI(Tl)-detector at  $^{60}\text{Co}$  1,332 keV) placed in a 10 cm thick lead-shield was used to determine the  $^{133}\text{Ba}$ -tracer added to samples in the radiochemical separation of  $^{226}\text{Ra}$ . Calibration by counting efficiency was made using a  $^{133}\text{Ba}$  solution in the same geometry and with similar concentration as was used for the samples.

### 4.3 Alpha spectrometry

A Canberra alpha spectrometry system (see Figure 4-3) consisting of 32 PIPS-detectors (Planar Implanted Passivated Silicon) with surface areas of 450 mm<sup>2</sup> connected to low-noise signal processing electronics was used for the analysis of  $^{228}\text{Th}$  and also  $^{230}\text{Th}$  and  $^{232}\text{Th}$  when detectable. A Canberra Genie-2000 system with acquisition interface modules (AIM-modules) was used to acquire control of the detectors. Calibration of the system is normally not needed, since isotope dilution analysis is applied when analysing alpha emitters, and all alpha particle energies are counted with the same efficiency.

### 4.4 Inductively Coupled Plasma mass spectrometry (ICP-MS)

The Thermo X<sup>II</sup> – series ICP-MS (see Figure 4-4) was used for the analysis of  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{230}\text{Th}$ . The instrument is a quadrupol-based ICP-MS with a collision cell. A low-flow concentric nebuliser and a CETAC U5000+ ultrasonic nebuliser were used for the sample introduction into the instrument. Calibration, tuning and optimisation of the instrument were done daily, using standard multi-element solutions as well as certified isotope ratio material for U-isotopes (U-500).



*Figure 4-3. The solid state alpha spectrometry system used for the alpha spectrometry of thorium.*



*Figure 4-4. The ICP-MS system used for the analysis of Th and U isotopes.*

## 5 Execution

### 5.1 Radon field measurements

Groundwater sampling was performed in the monitoring wells in soil presented in Chapter 3 in October 2008 and June 2009. The sampling location can be seen in Figure 3-1. The wells were purged for one to five well volumes, depending on the availability of water and the recovery rate. Some of the sampled wells were supplied with BAT-type filter tips, see Table 3-1. This means that the sampling was done using the BAT-filter equipment. A BAT-type well is supplied with a filter at the screening depth, and the water is sampled through the filter via a septum. To force the water through the filter, a vacuum pressurised glass vial with a needled is used. The needle is pushed through the septum, and the groundwater is sucked up to the sampling vial due to the vacuum pressure. In order to get a representative groundwater sample, the vial was first filled during a period of 10 to 24 hours depending on the recovery rate of the well, and then a second vial was filled from which the sample was taken.

The RAD-7 H<sub>2</sub>O method was used with the 250 ml vials, when possible. For some BAT-type wells 40 ml vials were used, see Table 3-1. The vials were first rinsed with the sampling water before a water sample was taken. The radon concentration was measured with the RAD-7 as soon as possible after the sampling. The time between sampling and measurement was measured, and the loss of radon due to decay was corrected for.

### 5.2 Radium, uranium and thorium isotope analyses

#### 5.2.1 Collection of samples

Surface water samples were collected in two water cans of 25 litres each, in order to get the required sample volume of 50 litres per sampling site. The cans were first rinsed with distilled water and also rinsed once with the surface water before a water sample was collected. The water sampling, which was performed from a boat with exception for the sample taken from the stream, was carried out by lowering the water can below the water surface.

The soil samples were collected from three spade dug holes at each site in order to get a pooled sample representing the sampling area. The samples were collected in plastic bags.

The terrestrial vegetation samples were collected using a scissor to cut off the stems of the plants. The samples were collected at several locations within the sampling area to get a pooled sample representing the vegetation of the sample area. The samples were collected in plastic bags and weighed before they were sent to the laboratory.

The limnic vegetation samples were collected from a boat using a rake. The samples were taken from the bottom of the lake. Before the samples were placed in a plastic bag, they were rinsed in the lake water. The samples were weighed before they were sent to the laboratory.

All vegetation samples were stored in a refrigerator before transport to the laboratory.

#### 5.2.2 Laboratory analyses

Upon delivery, samples were weighed (fresh weight) and then stored frozen until further treatment. Biological and soil samples were freeze-dried during 1–3 days and the dry weight recorded. Following freeze-drying, samples were ashed at 500° C over night and the ash weight recorded. The ash was completely dissolved in the presence of isotopic tracers (<sup>133</sup>Ba, <sup>229</sup>Th and <sup>233</sup>U) using a HNO<sub>3</sub>/HCl/HF mixture. Water samples were acidified with HCl to pH = 1 and a well known amount of <sup>133</sup>Ba added, and samples were stored until further treatment. A comprehensive description of the radiochemical separation methods used can be found in /Chen et al. 2001/.

### **Uranium-isotopes**

All uranium isotopes ( $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$ ) were analysed using ICP-MS following chemical isolation of uranium. As yield determinant  $^{233}\text{U}$  was used. Uranium was isolated from the dissolved biological and soil samples by extracting with Tri-Butyl-Phosphate (TBP) in 8M  $\text{HNO}_3$ . Thorium was eliminated by diluting the TBP with xylene and washing with 1.5M HCl several times. The purified uranium was back-extracted with de-ionised water and directly injected to the ICP-MS where masses 233, 234, 235 and 238 were recorded. Mass-discrimination was applied using standards of natural uranium and NBL U-500 (equal atomic concentration of  $^{235}\text{U}$  and  $^{238}\text{U}$ ). Samples were run in two different dilutions to obtain  $^{234}\text{U}/^{235}\text{U}$  and  $^{235}\text{U}/^{238}\text{U}$  ratios without applying dead-time corrections. The concentration of the  $^{238}\text{U}$  isotope in the dissolved sample was also obtained separately by using a small sub-sample of non-purified solution, where a well known amount of  $^{233}\text{U}$  was added.

For water samples a subsample of about 2 L was used. The isotopic tracer  $^{233}\text{U}$  was added, and uranium was co-precipitated on  $\text{Fe}(\text{OH})_3$  by adding iron and ammonia to the samples. Following centrifugation, the precipitate was dissolved in 8M  $\text{HNO}_3$  and treated in the same way as described above for biological and soil samples. The concentration of the  $^{238}\text{U}$  isotope in the water samples was also obtained separately by using a small sub-sample (typically 10 ml) of raw water, where a well known amount of  $^{233}\text{U}$  was added.

Along with the analysis, 2 blank samples (100 ml deionised water) were analysed.

### **Thorium-isotopes**

Thorium was isolated from the samples by using the discarded material in the uranium-analysis. The 1.5M HCl wash of TBP containing all thorium was evaporated and purified by a ion-exchange procedure. Half of the purified solution was used for ICP-MS measurement to obtain  $^{232}\text{Th}$  and  $^{230}\text{Th}$ . Masses 229, 230 and 232 were monitored during the analysis. A  $^{229}\text{Th}$  standard was used as a yield determinant in the analysis. The isotope  $^{232}\text{Th}$  was also analysed directly in a non-purified sub-sample by injecting directly into the ICP-MS and adding  $^{229}\text{Th}$  as yield tracer.

The part of the purified Th-sample not used for ICP-MS was electroplated onto stainless steel discs and counted by alpha spectrometry to determine  $^{228}\text{Th}$ . In many cases  $^{230}\text{Th}$  and  $^{232}\text{Th}$  could be obtained from these analysis as well.

Thorium in water samples was analysed by precipitating onto  $\text{Fe}(\text{OH})_3$  by adding iron and ammonia to the samples. Following centrifugation, the precipitate was dissolved in 8M  $\text{HNO}_3$  and thorium was purified as described above. Half of the purified sample was taken for ICP-MS and half to alpha spectrometry, primarily to determine  $^{228}\text{Th}$ .

### **Radium-226**

Radium in soil and biological samples was analysed by using discarded solutions from the U and Th extraction and ion-exchange procedure. Radium was neither extracted, nor adsorbed in these procedures, but followed the water phase. Radium from these solutions was co-precipitated onto  $\text{MnO}_2$  formed by adding  $\text{KMnO}_4$ ,  $\text{MnCl}_2$  and ammonia to pH = 9. The precipitate was dissolved and radium further purified by precipitate onto  $\text{PbSO}_4$ , which was dissolved and transferred to a liquid scintillation vial. An hydrophobic (oil-based) scintillator was added, and three weeks were allowed for ingrowth of  $^{222}\text{Rn}$  and daughter products, which was sequently determined by counting on the quantulus LSC. The radiochemical yield was determined by using a well-known amount of  $^{133}\text{Ba}$  added to each sample. The  $^{133}\text{Ba}$  in the separated sample solutions was quantified separately by counting on a HPGe-detector.

## **5.3 Nonconformities**

According to the Activity plan the original idea was to collect two vegetation samples from each terrestrial sampling location. This was later changed, and only one sample was taken from sites AFM1068 and AFM1247, whereas three samples were collected from site AFM1076.

## 6 Results

The radon-222 data from this study are presented in Appendix 1, Table A1-1, Radioisotope data.

The radon concentrations in near surface groundwater show a large variability with both low (0.43 Bq/L) and high (430 Bq/L) radon concentrations. Most of the radon concentrations fall within the normal range (40–400 Bq/L) for granitic bedrock areas in Sweden /Skeppström 2005/. The results from these radon measurements are together with the corresponding radon measurements from the Laxemar investigation area used to evaluate the use of radon as a tracer for groundwater discharge in /Grolander 2010/.

The radium, uranium and thorium isotope determinations in the surface water, vegetation and soil samples, which are presented in Appendix 1, Tables A1-2, A1-3 and A1-4, show results in the same order of magnitude as previous isotope analyses in the Forsmark area. The  $^{235}\text{U}/^{238}\text{U}$  ratios show that the uranium isotope composition is natural and not significantly altered by anthropogenic contamination.

A species list for the collected biota samples is presented in Appendix 2, Table A2-1.

## References

**Chen Q, Aarkrog A, Nielsen S P, Dahlgaard H, Lind B, Kolstad A K, Yu Y, 2001.** Procedures for Determination of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{237}\text{Np}$ ,  $^{234}\text{U}$ ,  $^{238}\text{U}$ ,  $^{228}\text{Ac}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{99}\text{Tc}$  and  $^{210}\text{Pb}$ - $^{210}\text{Po}$  in Environmental Materials. Risø National Laboratory, Roskilde Rapport R-1263(EN). (<http://www.risoe.dk/rispubl/nuk/nukpdf/ris-r-1263.pdf>).

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## Radioisotope data

**Table A1-1. Radon-222 concentrations in near surface groundwater. The radon concentrations are corrected for the decay between sampling and analysis.**

IDCODE	Sampling depth (m)	Rn222 (Bq/L)	Rn222 STD (Bq/L)
SFM0001	4.45	35.67	3.12
SFM0004	6.00	23.23	1.88
SFM0006	4.20	3.86	1.09
SFM0011	4.50	99.16	6.05
SFM0012	6.35	65.10	0.45
SFM0013	4.48	430.52	21.97
SFM0013	4.48	410.00	14.30
SFM0014	2.00	149.00	9.06
SFM0014	2.00	142.19	8.01
SFM0019	4.50	39.85	4.62
SFM0019	4.50	39.60	1.18
SFM0023	5.40	189.32	9.10
SFM0026	16.00	165.24	13.80
SFM0026	16.00	144.35	8.93
SFM0027	8.00	314.37	22.77
SFM0032	4.00	51.35	6.65
SFM0037	3.00	31.87	2.19
SFM0049	5.00	12.52	2.59
SFM0051	5.25	53.98	2.90
SFM0056	6.17	1.06	0.13
SFM0057	4.55	66.39	5.25
SFM0060	7.60	79.60	2.27
SFM0084	3.70	42.65	3.34
SFM0086	2.21 (BAT)	23.33	7.15
SFM0087	2.00	3.64	0.96
SFM0089	0.95 (BAT)	0.43	0.50
SFM0090	3.07	53.08	8.01
SFM0091	1.90	71.28	4.99
SFM0093	1.01 (BAT)	3.75	1.32
SFM0094	2.24	32.16	1.56
SFM0095	5.00	374.00	19.20
SFM0097	2.76 (BAT)	19.80	2.61
SFM0100	1.81 (BAT)	4.25	1.21
SFM0102	1.26 (BAT )	2.68	0.57
SFM0103	4.90	230.00	22.08

**Table A1-2. Uranium isotope determinations in vegetation and soil samples.**

IDCODE	Sample content	Wet weight (g)	Dry weight (g)	<sup>238</sup> U	<sup>238</sup> U uncertainty	<sup>234</sup> U	<sup>234</sup> U uncertainty	Atom Ratio <sup>234</sup> U/ <sup>238</sup> U	<sup>234</sup> U/ <sup>238</sup> U uncertainty
AFM001247	Humus	3.52	1.90	164	5	182	5	0.0072	5
AFM001247	Vaccinium myrtilus	0.91	0.21	0.056	5	0.059	5	0.0073	5
AFM001068	Humus	2.65	1.94	37	5	38	5	0.0072	5
AFM001068	Maianthemum biofolium	0.34	0.05	0.085	5	0.086	5	0.0073	5
AFM001076	Rubus saxatilis	0.73	0.12	0.068	5	0.069	5	0.0073	5
AFM001076	Carex elata	0.89	0.17	0.200	5	0.22	5	0.0071	5
AFM001076	Humus	5.41	2.96	54	5	56	5	0.0072	5
AFM001076	Picea abies	0.67	0.10	0.016	5	0.017	5	0.0073	5
AFM000050	Chara sp.	1.37	0.12	7.7	5	8.4	5	0.0072	5
AFM000010	Chara sp.	1.46	0.23	4.1	5	4.2	5	0.0072	5

**Table A1-3. Thorium and radium isotope determinations in vegetation and soil samples.**

IDCODE	Sample content	Wet weight (g)	Dry weight (g)	<sup>228</sup> Th	<sup>228</sup> Th uncertainty	<sup>230</sup> Th	<sup>230</sup> Th uncertainty	<sup>232</sup> Th	<sup>230</sup> Th uncertainty	<sup>226</sup> Ra	<sup>226</sup> Ra uncertainty
AFM001247	Humus	3.52	1.90	14.1	5.2	0.02310	0.0047	12.2	4	29.1	3.1
AFM001247	Vaccinium myrtilus	0.91	0.21	1.7	4	0.00001	0.0084	0.0093	5.4	7.6	5.7
AFM001068	Humus	2.65	1.94	9.9	4.7	0.01040	0.0045	10.0	4	20.1	3.9
AFM001068	Maianthemum biofolium	0.34	0.05	0.52	5.5	0.00004	0.013	0.039	4.3	1.6	5.5
AFM001076	Rubus saxatilis	0.73	0.12	0.061	7	0.00002	0.013	0.016	4.6	0.3	7.4
AFM001076	Carex elata	0.89	0.17	0.15	5.9	0.00001	0.011	0.0062	4.7	1.3	6.1
AFM001076	Humus	5.41	2.96	7.0	6.2	0.00799	0.0061	6.8	4	13.1	4.2
AFM001076	Picea abies	0.67	0.10	0.015	6.3	0.00000	0.013	0.0026	4.6	0.8	8.1
AFM000050	Chara sp.	1.37	0.12	6.6	4	0.00130	0.0047	0.48	4	23.1	3.6
AFM000010	Chara sp.	1.46	0.23	3.5	5.6	0.00104	0.0061	0.42	4	16.0	3.9

**Table A1-4. Radium, uranium and thorium isotope determinations in surface waters.**

IDCODE	Water type	<sup>226</sup> Ra (Bq/L)	<sup>226</sup> Ra dev (Bq/L)	<sup>238</sup> U (mBq/L)	<sup>234</sup> U (mBq/L)	<sup>232</sup> Th (mBq/L)	<sup>230</sup> Th (mBq/L)	<sup>228</sup> Th (mBq/L)	Atomic ratio <sup>234</sup> U/ <sup>238</sup> U
PFM000062	Sea water	0.0031	0.0042	6.37	7.55	-0.001	0.004	0.387	0.00719
PFM000107	Lake water	0.0058	0.0041	28.53	32.31	0.004	0.010	0.619	0.00719
PFM000117	Lake water	0.0038	0.0040	12.31	13.16	0.024	0.060	0.533	0.00719
PFM000068	Stream water	0.0044	0.0040	21.84	23.90	0.486	1.077	4.130	0.00719
PFM004306	Lake water	0.0075	0.0044	57.03	60.14	0.115	0.492	1.169	0.00719

## Species list for biota samples

Table A2-1. List of sampled biota in Latin, Swedish and English.

Latin	Swedish	English
<i>Vaccinium myrtillus</i>	Blåbär	Blueberry
<i>Rubus saxatilis</i>	Stenbär	Stone Bramble
<i>Picea abies</i>	Gran	Norway Spruce
<i>Carex elata</i>	Bunkestarr	Sedges
<i>Maianthemum biofolium</i>	Ekorrbär	May lily
<i>Chara</i> sp.	Kransalg	Stonewort