International
Progress Report

IPR-02-68

Äspö Hard Rock Laboratory

TRUE-1 Continuation project

Use of radon concentrations for estimation of fracture apertures - Part 1: Some method developments, preliminary measurements and laboratory experiments

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October 2002

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Report no.	No.
IPR-02-68	F83K
Author	Date
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Checked by	Date
Anders Winberg	Jan 2003
Approved	Date
Christer Svemar	03-04-17

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Keywords: Aperture, fracture, pulse shape analysis, radon

Abstract

Some experimental activities have been performed in order to investigate the possibility of assessing fracture apertures from radon concentrations in groundwater. A new method for sampling groundwater and maintaining overpressure during the γ-spectrometry measurement has shown that reproducible results can be obtained. By sampling at the TRUE-1 site at the Äspö Hard Rock Laboratory and performing subsequent radon measurements, indications have been obtained that radon concentrations are very similar for waters sampled from the same identified geological structures.

Laboratory experiments have been done in order to see if a simple diffusion cell approach combined with liquid scintillation measurements with pulse shape analyses (PSA) can be used to measure radon fluxes from intact rock surfaces. The results are very promising and indicate that determination of radon fluxes from most rock materials should be possible to perform.

Radon fluxes have also been estimated from some different fault gouge materials; all sampled at the Äspö Hard Rock Laboratory. The results provides no evidence that the flux from the unconsolidated gouge material in the fracture should be the dominant source for the radon in the groundwater.

Sammanfattning

En experimentell studie har genomförts i syfte att undersöka möjligheterna att bestämma sprickaperturer i berg från radonkoncentrationer i grundvattnet. En ny metod har utvecklats för att provta grundvatten och bibehålla övertrycket under den γ-spektrometriska mätningen. Provtagning vid TRUE-1 siten i Äspölaboratoriet och efterföljande radonmätningar har indikerat att radonkoncentrationerna är sinsemellan väldigt lika för vatten som provtagits från samma geologiska struktur.

Laboratorieexperiment har utförts i syfte att undersöka om en enkel diffusionscell i kombination med vätskescintillationsmätningar med Pulse Shape Analyses (PSA) kan användas för att mäta radonflöden från intakta bergytor. Resultaten är mycket lovande och indikerar att känsligheten är tillräcklig för att kunna möjliggöra bestämningar av radonflöden från de flesta bergmaterial.

Uppskattningar har även utförts av radonflöden från vissa finfördelade sprickfyllnadsmaterial (fault gouge material); alla dessa material provtagna i Äspölaboratoriet. De preliminära resultaten ger ingen tydlig antydan av att radonflödet till grundvattnet i en naturlig bergsspricka skulle fullständigt domineras av bidraget från det finfördelade gougematerialet utan att bidraget från det intakta berget kan spela en betydande roll.

Content

1 Introduction

Within the SKB site investigation programme, an investigation has been performed concerning the possibilities of using the concentration of ^{222}Rn in groundwater in order to assess the fracture apertures in crystalline rock in an average sense. The conceptual idea is that the progenitor of ^{222}Rn, i.e., ^{226}Ra, is almost completely associated to the rock phase in groundwater system. The decay of radium produces the noble gas radon which can be transported through the pores and microfractures of the crystalline rock and therefore reach successively larger fractures. However, the lifetime of ^{222}Rn is comparatively short ($t_{½}$=3.82 d) which gives the radon a very short time to diffuse out to the rock fractures. Based on the general experiences of diffusion properties of crystalline rock, one can therefore assume that only the radon produced in the rock at mm-cm distance from the fracture surface can reach the conductive fractures. The concentration of radon in the groundwater can thus be regarded as a measure of the amount of rock that is interacting with the groundwater. If the radon concentrations in the groundwater can be combined with the radon flux rate of the rock (determined in laboratory experiment, e.g., Andrews et al. 1986) an estimation of the fracture aperture can be made. The simplest approach to this problem is of course using an assumption of a single parallel fracture (Figure 1-1) having a radon concentration, C_{Rn} (Bq/m^3), with a radon flux, Q (s^{-1}m^{-2}), from the fractue surfaces. At a steady state, the number of radon atoms decaying in the fracture is equal to the number of atoms diffusing in to the fracture. Assuming that the radon flux can be considered as independent of the radon concentration in the fracture (i.e., the diffusion from the fracture in to the matrix can be neglected) the fracture aperture, b (m), can be calculated from:

$$b = \frac{Q}{C_{\mathrm{Rn}}} \tag{1.1}$$

Restrictions to this simple approach have been made by Neretnieks (2001) who has presented a model that accounts for the impact of the diffusion from the fracture in to the rock matrix. This approach is much more complicated and thus involves some additional parameters, e.g., the pore diffusivity of radon, D_{p} (m^2/s), the porosity of the matrix, ε (-) and the fraction of the radon atoms that is not trapped within the mineral crystals, η (-). Calculations made by Neretnieks (2001) indicate that the smaller the fracture aperture gets, the larger deviation is obtained between a simple surface production model and the diffusion model.

The fracture aperture has a close relationship to the flow wetted surface per volume of rock (Elert 1997) and can therefore be an important parameter in the performance assessment. A schematic illustration of the concept is given in Figure 1-1.

There are several aspects of experimental limitations and sources of uncertainty which complicates this approach of estimating fracture apertures, e.g.;

- The flow of radon across a fracture surface is estimated to be rather low. It is therefore likely that improvements of low activity measurements of radon has to be done in order to be able to apply these measurements in rock types that do not have particularly high radium concentrations
- It is possible that the distribution of the uranium series radionuclides and radium in particular is very heterogeneous in the rock. Especially any fracture filling material (e.g., fine fractioned gouge material, described in e.g., Andersson et al. 2002) may be a significant and heterogeneous source of radon to the fracture groundwater.
- Sampling of deep groundwater causes a release of the pressure, a process that is suspected to cause losses of dissolved gasses, e.g., radon.

Since the TRUE-1 site at Äspö Hard Rock Laboratory has been used for several types of investigations (e.g., tracer experiments, geo-hydrological investigations, chemical analyses of the groundwater, Winberg et al. 2000) it was proposed to use the TRUE-1 site as a target area for a test of the method. The purpose of the project is to extract information of the radon concentrations in groundwater and radon fluxes from geologic material as possible from the target area and to use this information to assess the applicability of the radon method in the SKB site investigation programme. Furthermore, the project could potentially provide the TRUE-1 and the TRUE Block Scale project (e.g., Andersson et al. (2002) with improved assessment flow wetted surface data and could also be coupled to dilution measurements at ambient conditions. These tasks will be included as a part of the TRUE-1 Continuation programme.

The first part of the project (which is dealt with in this report) consists of method development and preliminary conclusions of the applicability of the method.

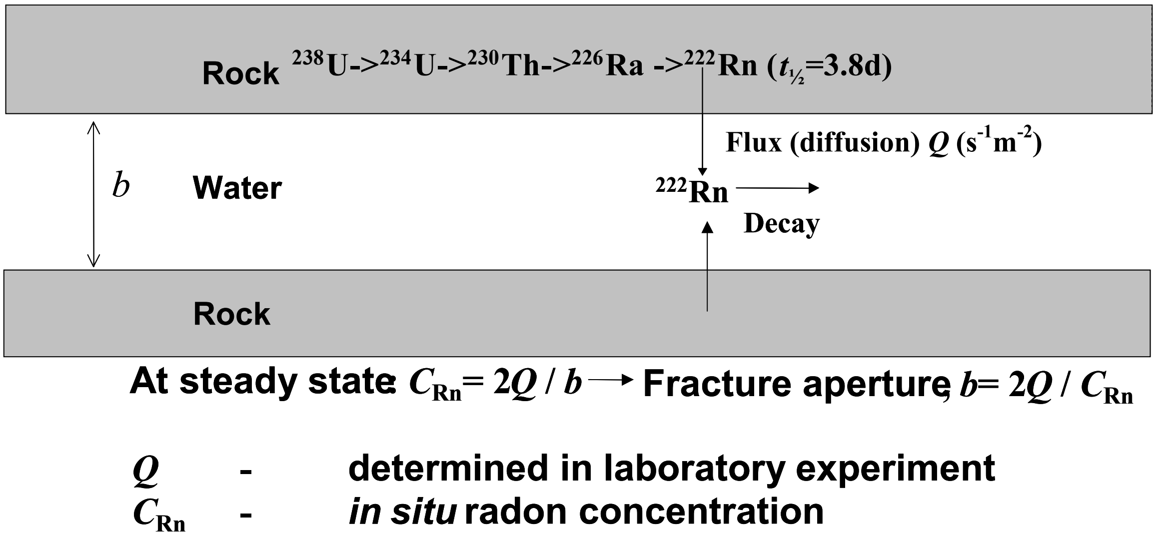


Figure 1-1 *Simplified conceptual description for the use of radon concentrations in determination of fracture apertures, see text for details.*

2 Measurements of radon concentrations in sampled groundwater

2.1 Background and identification of problems

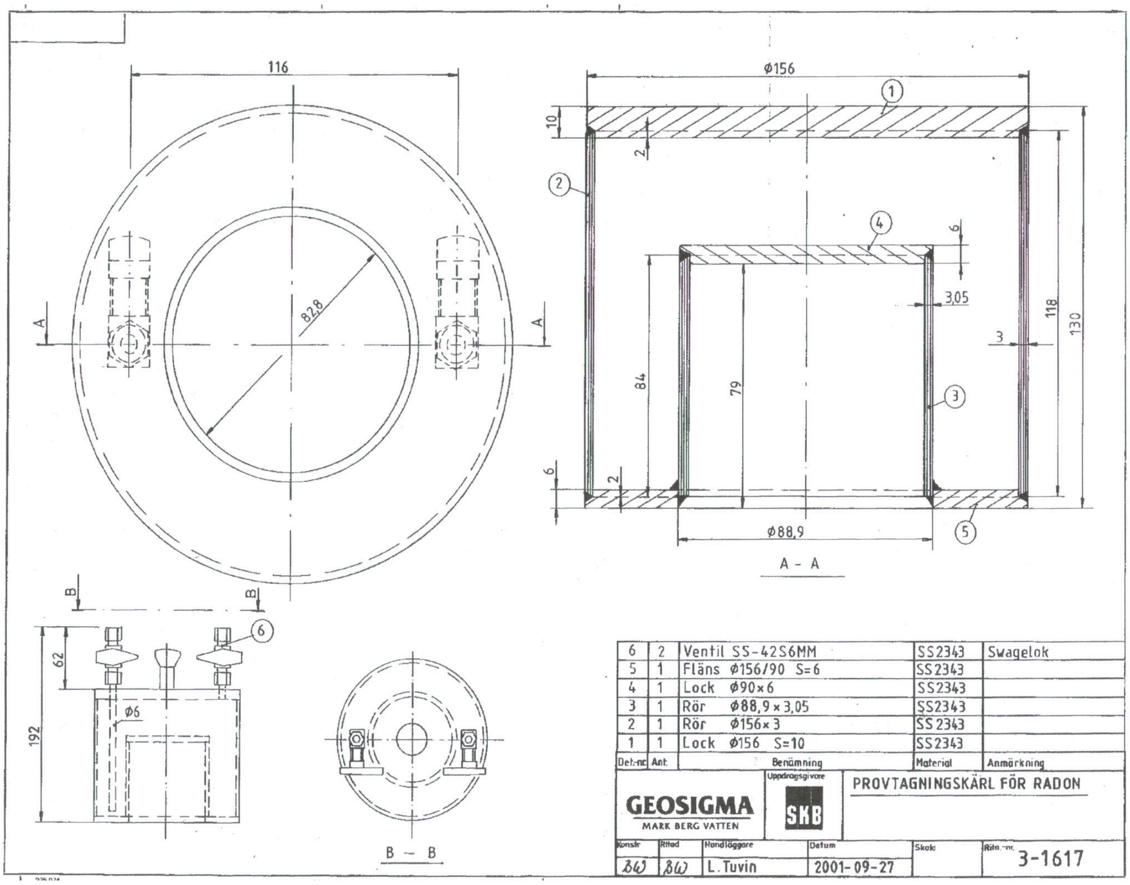
The normal underground sampling procedure involves extraction of groundwater from a borehole section where two packers isolate the test section. The packers are normally located such that a water conducting fracture transecting the borehole will have one packer on both sides of the fracture. If there is only a small exchange between the groundwater in the isolated section and the groundwater in the fracture (i.e., low flow rate in the fracture), the groundwater in the borehole section will have a radon concentration reflecting the ratio of the rock surfaces and the groundwater volume of the borehole section. The aim of the present investigation was to measure the surface volume ratio in the rock fractures and it was therefore important that the influence of the borehole conditions was minimised. It was thus important that the sampled groundwater was rather quickly transferred from the fracture to the sampling device, for this reason the sampling flow rate had to be rather high. Furthermore, in order to avoid influences from stagnant water in the borehole section, it was advantageous if a large volume of the pumped water is discarded before the actual water sample is taken. However, pumping out large volumes of water may cause in-flow of water from surrounding features. Considering these demands, it was decided that the sampling should be performed with maximum flow rate and that a water volume corresponding to one borehole section should be flushed before sampling occurred. For some very large borehole sections involved, this demand could not be fulfilled for practical reasons. For these sections, the sampling volume was determined from the assumption that only the borehole volume between the fracture and the inlet in the section was influenced by the sampling.

2.2 Construction of high-pressure sampling vessels

A problem with sampling of groundwater for radon measurement is that, due to release of the pressure, degassing is likely to occur which will cause losses of radon. Furthermore, sampling into plastic flasks may cause losses of radon, due to diffusion, between the time of sampling and the time of measurement. It was therefore considered advantageous to develop a sampling system consisting of a metal container that could keep a slight overpressure (~5bar) in the sampling situation. This sampling container should be possible to put directly on the γ-spectrometry detector at BASLAB, i.e., without any release of the overpressure or any exposure of the water to the surrounding air, which a transfer to a separate measurement flask would involve.

A sample vessel was constructed to enable sampling and avoiding a full pressure release. Furthermore, the sampling container was constructed to enable position on a γ-spectrometry detector without having to transfer the solution to a separate measurement beaker. In order to optimise the γ-spectrometry measurement efficiency, the sampling container was therefore constructed as a Marinelli beaker. A principal drawing of the sampling device is given in Figure 2-1.

In order to prevent losses of radon due to post-sampling diffusion, the sampling vessels was manufactured in a non-diffusive material, i.e., stainless steel. The vessel was dimensioned for a water pressure up to ~5 bar. Two valves were placed on the vessel which were designed to open at a certain pressure; 6 bar for the incoming water and 5 bar for the outgoing water.



Det.-nr	Ant.	Benämning	Material	Anmärkning
6	2	Ventil SS-42S6MM	SS2343	Swagelok
5	1	Fläns Ø156/90 S=6	SS2343	
4	1	Lock Ø90×6	SS2343	
3	1	Rör Ø88,9×3,05	SS2343	
2	1	Rör Ø156×3	SS2343	
1	1	Lock Ø156 S=10	SS2343	

Figure 2-1 *Construction drawing of the high pressure metal sampling vessel to be used for radon sampling and γ-spectrometry measurement*

2.3 Measurement of radon concentrations in the TRUE-1 site

Two campaigns of sampling of groundwater combined with radon measurements in the TRUE-1 site were performed, the first was performed 2001-11-22 and the second campaign was performed during the period 2001-12-10 - 2001-12-12. In the first campaign, only water from 7 borehole sections were sampled; in the second campaign 17 borehole sections were sampled for radon analyses.

Directly before the sampling took place, water corresponding to one section volume was extracted from the borehole. Subsequently, approximately 8 litres of water was sampled as a part of the program for the chemical analyses of Äspö groundwater. When the sampling of these waters was terminated, the radon sampling vessel was connected to the outlet from the borehole. The sample vessel was first filled up with water (1.5 litres) and thereafter, an additional 4.5 litres of water was flowed through; this was done in order to decrease the amount of first arrival water that was exposed to air contact during its sampling. After this volume of water had been flowed through, the valves were closed so that an overpressure of 5 bar was maintained.

The samples vessels were thereafter transported to BASLAB/CLAB where the samples vessels were put directly on the HPGe-detector (18% relative efficiency, EG&G Ortec) and were thus measured directly using γ-spectrometry without any depressurisation of the water. A separate sample vessel had been manufactured that had the same geometry but was instead, for calibration purposes, filled with a mixed γ-spectrometry solution (QCY B41, AEA Technology). The evaluation of the γ-spectrum was performed using the software GammaVision 5.31 (EG&G Ortec). In the quantification of the ^{222}Rn activity, the daughter radionuclide ^{214}Pb ($t_{½}$=26.8min) was used under the assumption of secular activity equilibrium between these isotopes. Using 3h in-growth time after the sampling, the activity of ^{214}Pb is >99% of the ^{222}Rn activity.

2.3.1 Results and discussion

The results of the measurements are illustrated in Figures 2-2. As can be seen the concentrations seem to be very dependent on the fracture being sampled. For example, the concentrations from the sections that has been interpreted to contain the Feature B fracture system all have radon concentration around 600 Bq per litre while samples originating from the Feature A fracture system (including NW-2′) have concentrations around 400 Bq per litre. From the results of the borehole sections measured in both sampling campaigns, it is indicated that the results are reproducible.

One could suspect that the volumes of the borehole section that the water is sampled from, influences the measured radon concentrations. The reason for this is that the rock surface to water volume ratio is much lower for the borehole section volume compared to the fracture. A long duration in the borehole section (and also in the plastic tubings) for the sampled groundwater can therefore be suspected to give erroneous radon concentrations. To address these potential sources of error, studies of the radon concentrations as a function of sampling flow rate and borehole section volume were performed. However, the results (Figure 2-3) showed no indication of a correlation of the measured parameters.

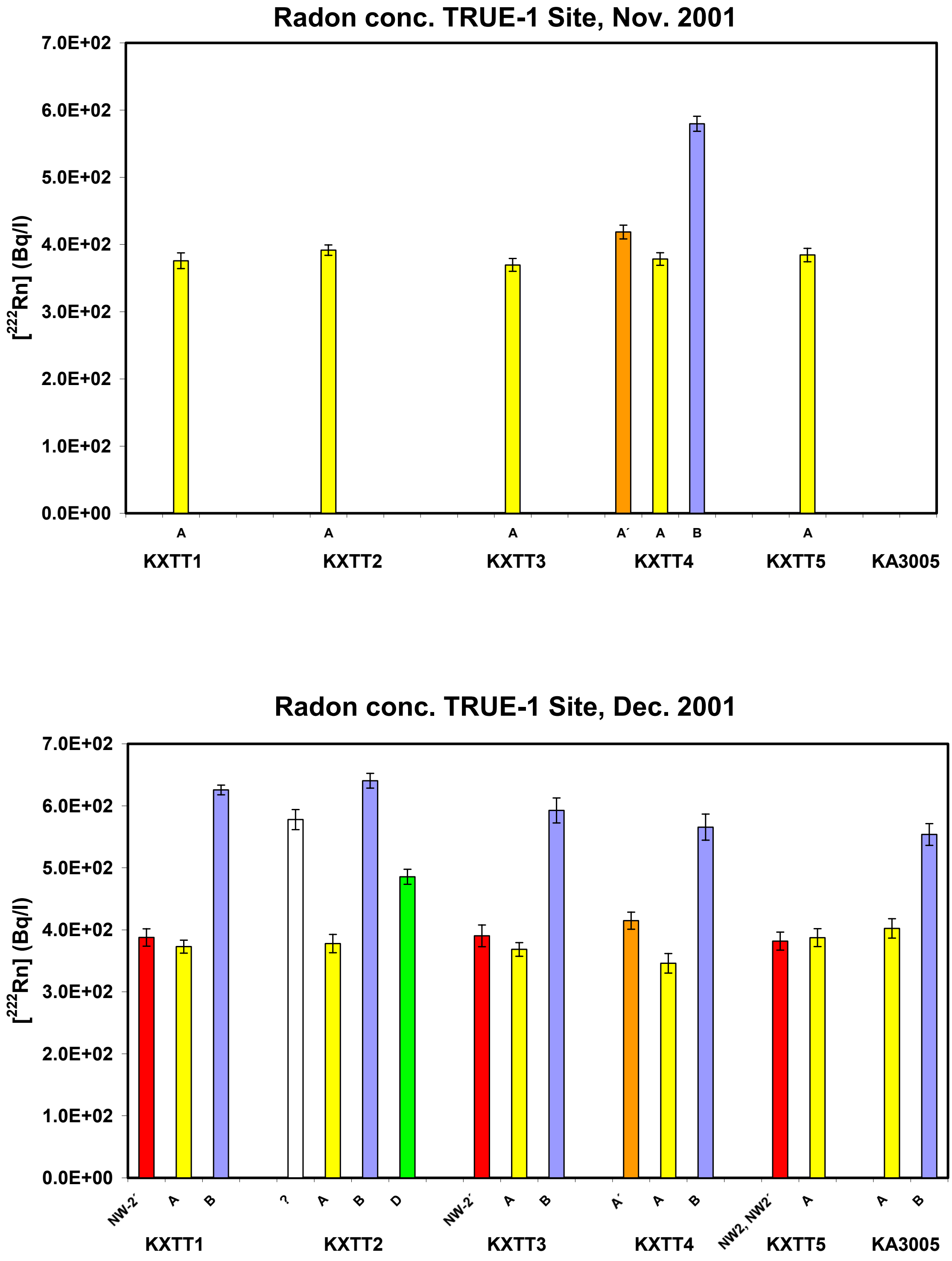
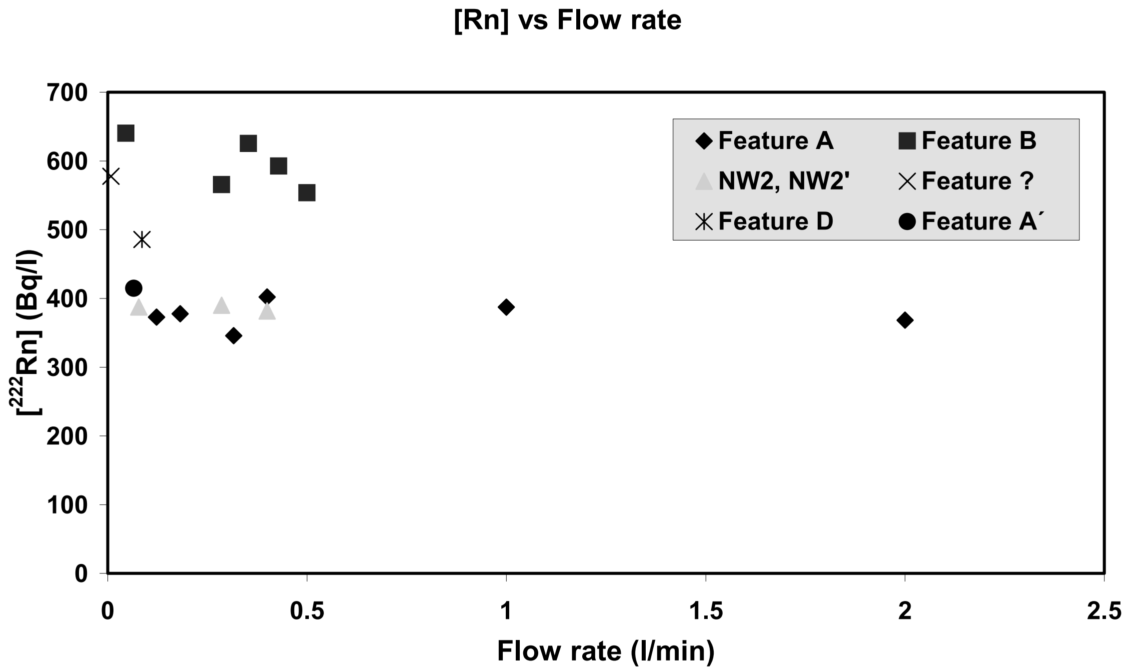


Figure 2-2 *Radon concentration in sampled groundwater in the TRUE-1 site. Results are given for the two campaigns, November 2001 (top) and December 2001(bottom) . The results refer to the different boreholes (KXTT1-KXTT5 and KA3005) and the isolated sections in these boreholes which correspond to different features (see Winberg et al. 2001 for further details)*



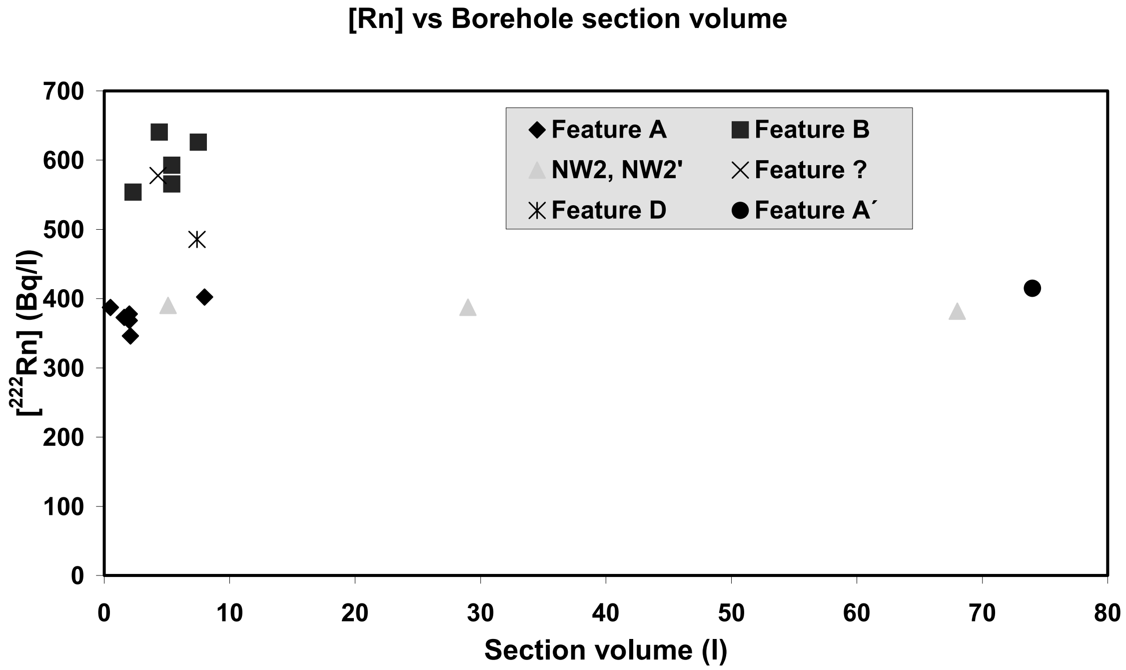


Figure 2-3 *Radon concentrations in groundwater sampled at the TRUE-1 site, presented as a function of sampling flow rate (top) and of borehole section volume (bottom).*

2.3.2 Comparison with supporting measurements

In addition to the sampling for radon described in Section 2.3, samplings and chemical analysesaccording to the Äspö HRL Class 5 were performed. The results of these measurements will be published elsewhere. However, the measurements of isotopes included in the uranium decay series, distributed as Excel spreadsheets, will be presented in this report.

Complementary measurements of radon was performed by sampling in 5 litres plastic beakers and sending the samples to Studsvik Nuclear AB for analyses using γ-spectrometry measurements. This means that, contrary to the samples taken with the metal vessel sampler, a full depressurisation of these samples was obtained. Furthermore, the samples were stored a longer time (i.e., days-weeks) before measurements were performed. A comparison of the results obtained from the seven different borehole sections where the two different sampling techniques were applied (Figure 2-4) shows lower radon concentrations when a sampling procedure is applied which includes depressurisation and storing the samples in plastic beakers. Furthermore, the trend of very similar radon concentrations for the different Features can not be seen in the results for this simplified sampling procedure. It is thus indicated that degassing of radon in depressurised samples is a problem which is solved with the newly constructed metal sampling vessel and associated analysis methodology.

Besides the concentration of ^{222}Rn, the activities of the uranium decay series isotopes ^{238}U, ^{234}U, ^{230}Th and ^{226}Ra were measured by Studsvik Nuclear AB in seven different groundwater samples. The results are presented in Table 2-1 amd further details will be given by Smellie and Tullborg, in prep. The results show that there is an enrichment in the groundwater of some of the daughter radionuclides in the uranium series, e.g., ^{234}U and ^{226}Ra. However, as expected these activities are far much lower than the measured activities of ^{222}Rn. This verifies that the major part of the radon found in the groundwater in the fracture systems originates from ^{226}Ra decay associated to the geologic material phase and only to a very limited part to the groundwater.

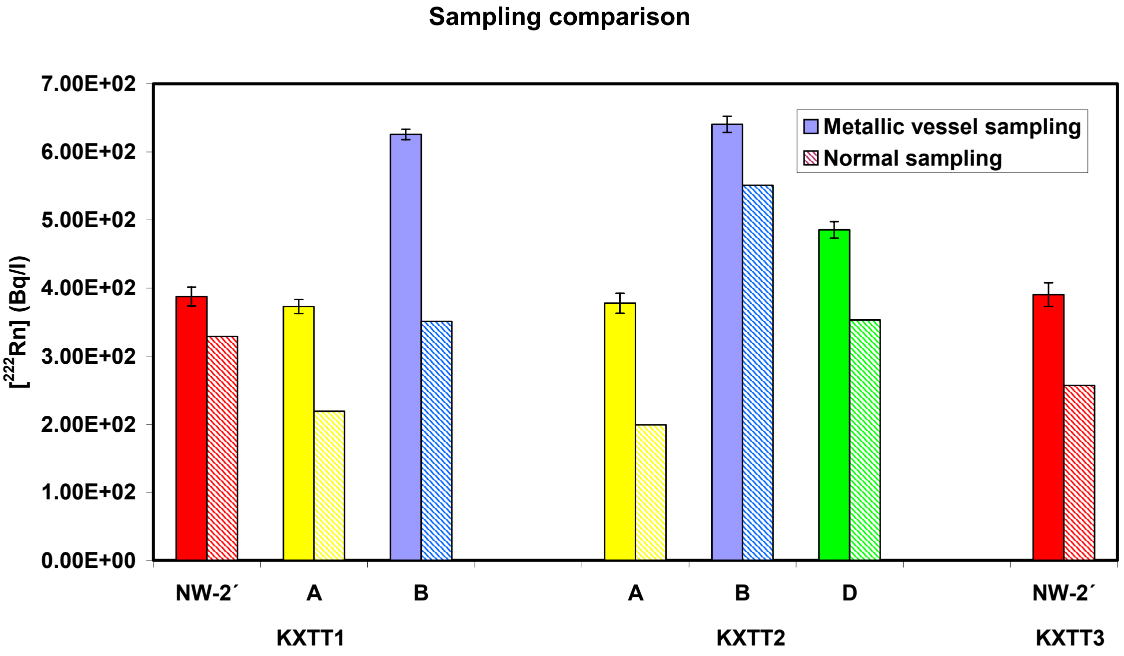


Figure 2-4 *Comparison of radon concentrations obtained with the metal vessel sampling method and with the normal sampling method, see text for details.*

Table 2-1 Activities of the uranium series radionuclides measured in the groundwater sampled at the TRUE-1 site. The measurements were performed by Studsvik Nuclear AB by using α-spectrometry (^{238}U, ^{234}U and ^{230}Th) and by using BaSO$_4$(s) co-precipitation followed by γ-spectrometry (^{226}Ra)

Borehole section	Feature	^{238}U (mBq/kg)	^{234}U (mBq/kg)	^{234}U/^{238}U	^{230}Th (mBq/kg)	^{226}Ra (Bq/l)	^{226}Ra/^{238}U
KXTT1:R1	NW-2´	8.1±2.4	34±10	4.2±2.0	<0.27	0.97	95±28
KXTT1:R2	A	6.4±1.9	28±8	4.4±2.1	<0.31	0.78	116±35
KXTT1:R3	B	8.3±2.5	30±9	3.6±1.7	<0.55	0.97	102±31
KXTT2:R2	?	5.0±1.5	23±7	4.5±2.1	<0.45	0.63	137±40
KXTT2:R3	A	4.3±1.3	22±7	5.0±2.4	<2.1	0.89	174±52
KXTT2:R5	B	3.7±1.1	15±4	4.0±1.9	<0.49	1.2	152±46
KXTT3:R1	NW-2´	5.2±1.6	25±7	4.7±2.2	<5.7	0.88	147±44

3 Measurements of ^{222}Rn at low concentration using PSA analysis

3.1 Background

To enable laboratory radon flux measurements from fracture surfaces in drill cores in contact with water solutions, very low concentrations of radon needs to be measured very accurately. A promising method which is likely to produce a high signal to noise ratio is liquid scintillation with α/β pulse shape analysis (PSA). In this work, measurements using this technique were performed using the equipment at BASLAB/CLAB and at the Department of Nuclear Chemistry, Chalmers University of Technology.

3.2 Studies of the influence of the pulse shape analyses (PSA) level

Detection of α-decay using liquid scintillation measurement produces electronic pulses with significantly different shape compared to pulses originating from e.g., β- and γ-activity (cf. Figure 3-1). With electronic discrimination of pulses with respect to their shape (i.e., Pulse Shape Analyses, PSA)favourable conditions are obtained for measurements of very low α-activity in presence of a significant background level of β- and γ-activity activity in the sample. However, the level used for the pulse discrimination, i.e., the PSA-level has to be optimised, since:

- The PSA-level can be set so that a high acceptance of pulses is obtained. The advantage of this method is that a high efficiency is obtained but the disadvantage is that many of the pulses originating from β- and γ-activity are falsely accepted as α-decay. Since the most background activity originates from β- and γ-activity, a high acceptance of pulses gives a high background count rate and, consequently, a higher detection limit.
- On the other hand the PSA-level can be set so that a low acceptance of pulses is obtained. The advantage is that practically all pulses from β- and γ-activity are discarded so that a low background count rate is obtained. However, by this method the efficiency of the α-counting is somewhat lowered which also can have negative impact on the detection limit.

Measurements with variations of PSA-level were performed on a Wallac Rackbeta 1219 on Department of Nuclear Chemistry, Chalmers University of Technology, and on a Wallac 1414 Guardian, BASLAB/CLAB. For all measurements, glass scintillation vial together with liquid scintillation cocktail with high flashpoint (Ultima Gold AB)were used.

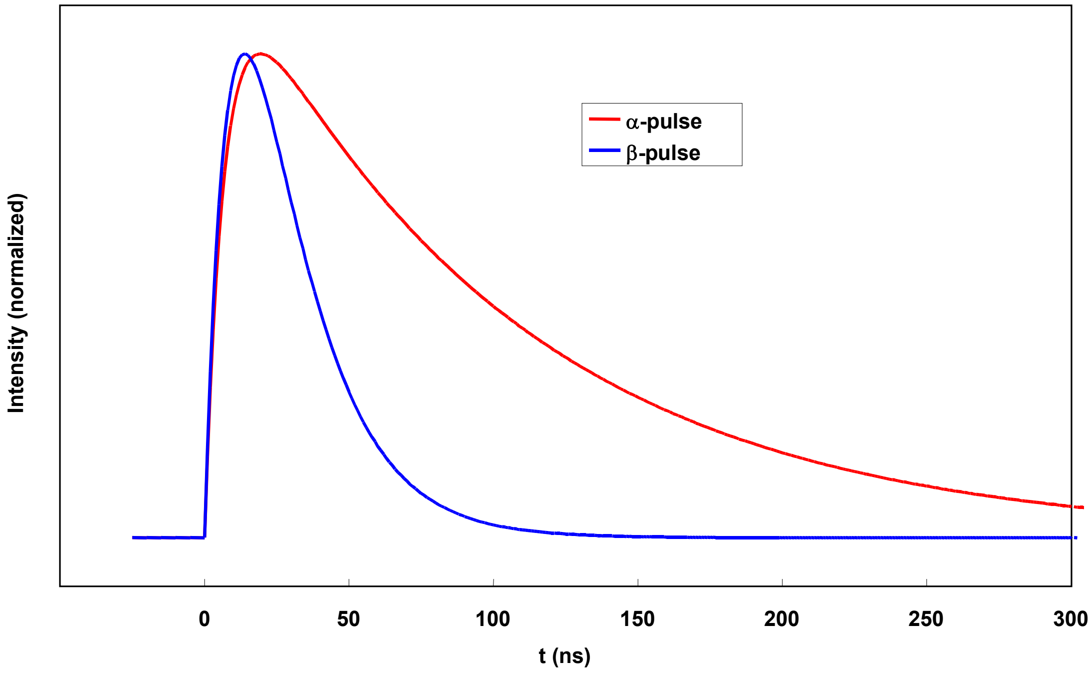


Figure 3-1 *Principal illustration of the shape of α- and β-pulses obtained in liquid scintillation measurements. The α-pulses have a longer duration in time and by varying the PSA-level, β-pulses can be electronically discriminated.*

3.2.1 Measurement on Wallac Rackbeta 1219 (CTH)

The efficiency and background level as a function of the PSA-level was checked using a standard solution of ^{241}Am (α-emitter). In these measurements, energy discrimination was performed so the only pulses obtained in the upper half of the energy spectrum (channels number 500-1024) were registered. The efficiency was studied as a function of the PSA level used. Besides that, a sample containing ^{137}Cs (β- and γ-emitter) was also measured. The "β-spillover" i.e., the amount of β-pulses from ^{137}Cs falsely registered as α-pulses was thus registered. The results are given in Figure 3-2 and indicate a level of approximately 100% measuring efficiency up to a PSA-level of 150. The β-spillover drops drastically with PSA levels of >125, and it is thus indicated that good measuring properties (high and stable efficiencies combined with low spillover) are obtained in the PSA-region of 125-150.

A check was also done in order to estimate the background count rate as a function of the PSA-level. The results of this test are given in Figure 3-3 and indicates that a drastic decrease in background count rate is obtained in the PSA region 140-150. This shift occurs somewhat later compared to the decrease in spillover from ^{137}Cs but still much earlier compared to the decrease in α-counting efficiency. From these observations one can conclude that:

- Radon measurements where the concentration is >1Bq per litre is best done using PSA-levels ~135. The total count rate is then high enough that the uncertainty from the background count rate will have a minor impact on the total uncertainty. One can then benefit from being on a ~100% efficiency plateau, i.e., a low uncertainty in the efficiency value can be obtained.

- Low activity measurements of radon (<1Bq) is probably best done using PSA-levels ~160. One is then on the slope of the efficiency curve but the uncertainty added by this fact is probably compensated by the decrease in uncertainty obtained by the significant decrease of the background count rate.

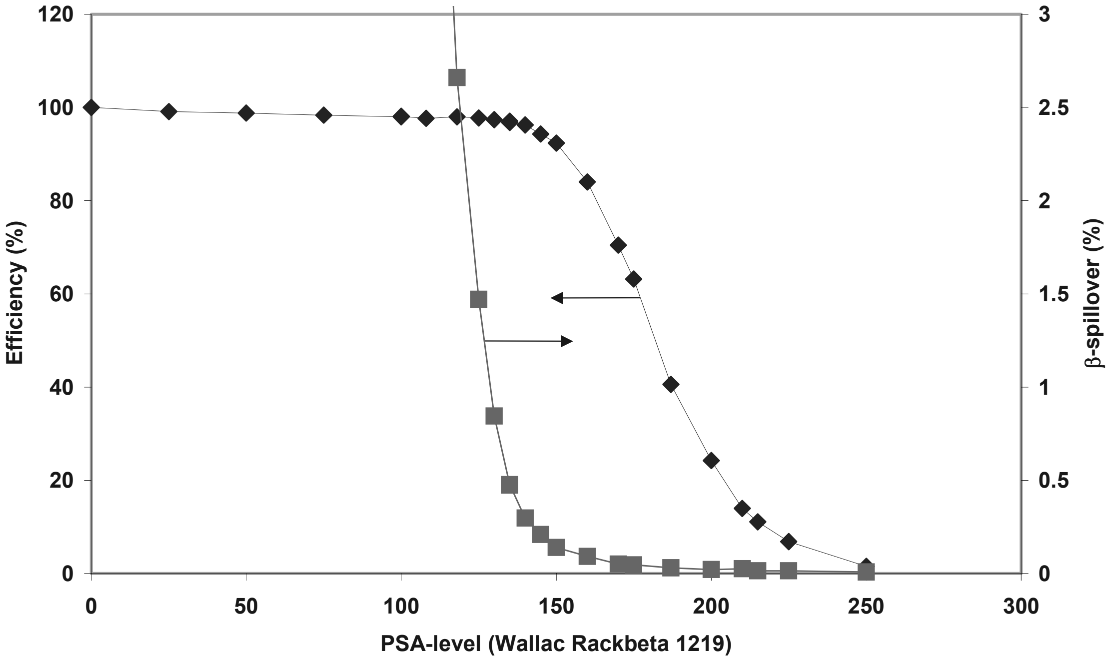


Figure 3-2 *Efficiency of α-counting of* ^{241}*Am (diamonds, projected on the left y-axis) and the corresponding β-spillover from* ^{137}*Cs (squares, projected on the right y-axis), given as a function of the PSA-level. Measurement was performed using the Wallac Rackbeta 1219 at the Department of Nuclear Chemistry, CTH.*

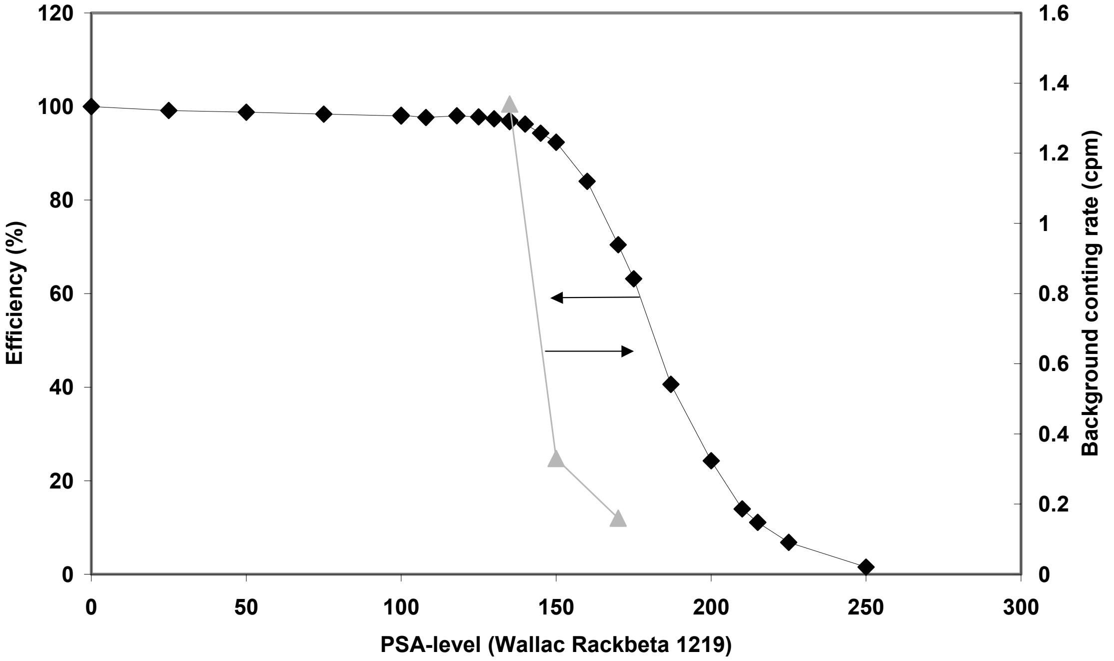


Figure 3-3 *Efficiency of α-counting of* ^{241}*Am (diamonds, projected on the left y-axis) and the corresponding background count rate (triangles, projected on the right y-axis), given as a function of the PSA-level. Measurement was performed using the Wallac Rackbeta 1219 at Department of Nuclear Chemistry, CTH.*

Measurements were also done in the same way by studying the α-efficiency and β-spillover as a function of the PSA-level with varying water composition. Besides the distilled water used in the previously described experiment, Äspö groundwater (saline) and groundwater from Delsjön, Göteborg (low-saline) were exposed to the same experiment procedure as described above. The results (Figure 3-4) indicate that variations in chemical compositions seem to have very small influence on the efficiencies and on the β-spillover.

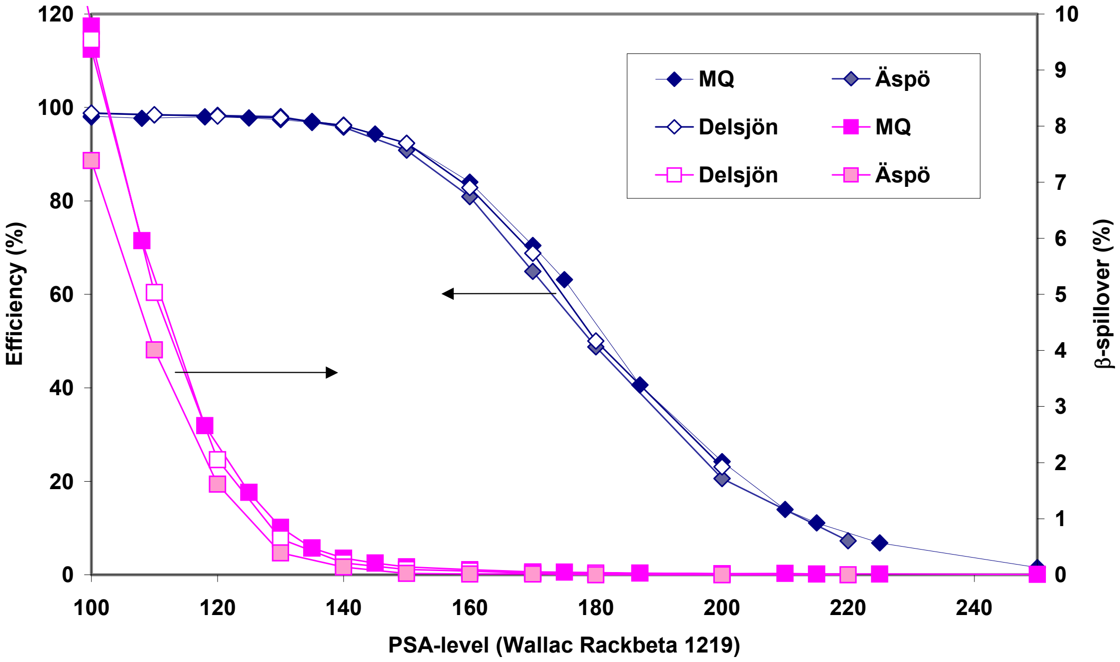


Figure 3-4 *Efficiency of α-counting of ^{241}Am (diamonds, projected on the left Y-axis) and the corresponding β-spillover from ^{137}Cs(squares, projected on the right Y-axis), given as a function of the PSA-level. Measurement was performed using the Wallac Rackbeta 1219 at the Department of Nuclear Chemistry, CTH.*

3.2.2 Measurement on the Wallac 1414 Guardian, BASLAB/CLAB

In these measurements, sampled groundwater from the Äspö Hard Rock Laboratory was used. Fromthe results of the γ-spectrometry measurements (cf. Chapter 2) this water is presumed contain ^{222}Rn in several hundreds of Bq per liter) 5 ml of the water was mixed with scintillation cocktail and measurement was performed. The count rate of this sample was measured and registered as a function of the PSA-level. A blank sample (corresponding amount of distilled water with scintillation cocktail) and was measured the same way in order to obtain the characteristics of the background count rate. The results are presented in Figure 3-5. Unfortunately, a somewhat confusing fact is that the numerical setting of the PSA-levels works on the Wallac 1414 Guardian (Baslab/CLAB) is defined in a different way than the corresponding settings on the Wallac Rackbeta 1219 (Chalmers). Increasing the numerical value on the PSA level on the Wallac 1414 Guardian instead gives a lower discrimination level and the characteristics of the plots from the two machines are therefore directly opposite.

The results indicate a sharp increase of the sample count rate between the PSA-level of 20 and 30. After that, a plateau is reached and increasing the PSA level above 60 gives increasing count rate. The characteristics are thus quite different from the characteristics observed for the measurement of ^{241}Am where a straight plateau was reached. A possible explanation to this is that the plateau between the PSA level of 30 and 50 corresponds to the characteristics of 100% efficiency for the α-decay of the radon series radon nuclide (i.e., the decay of ^{222}Rn, ^{218}Po and ^{214}Po, respectively). Further increase of the PSA level gives a gradually increased acceptance of the β-particles produced in the decay series of the radon daughters ^{214}Pb and ^{214}Bi. This explanation is supported by the characteristics of the background count rate (most probably originating from β-decay) which also increases significantly at PSA level >50. This explanation is thus also compatible with the β-spillover studies presented in section 3.2.1. An evaluation of the of the α-efficiency and the β-spillover characteristics according to the proposed explanation is presented in Figure 3-5 which thus indicate that with high PSA-levels, <25% of the detected pulses could originate from β-decay from the radon daughters. The radionuclide suspected to have the strongest interference in the α-counting is ^{214}Bi which have a rather high-energetic β-decay (> 3.3 MeV).

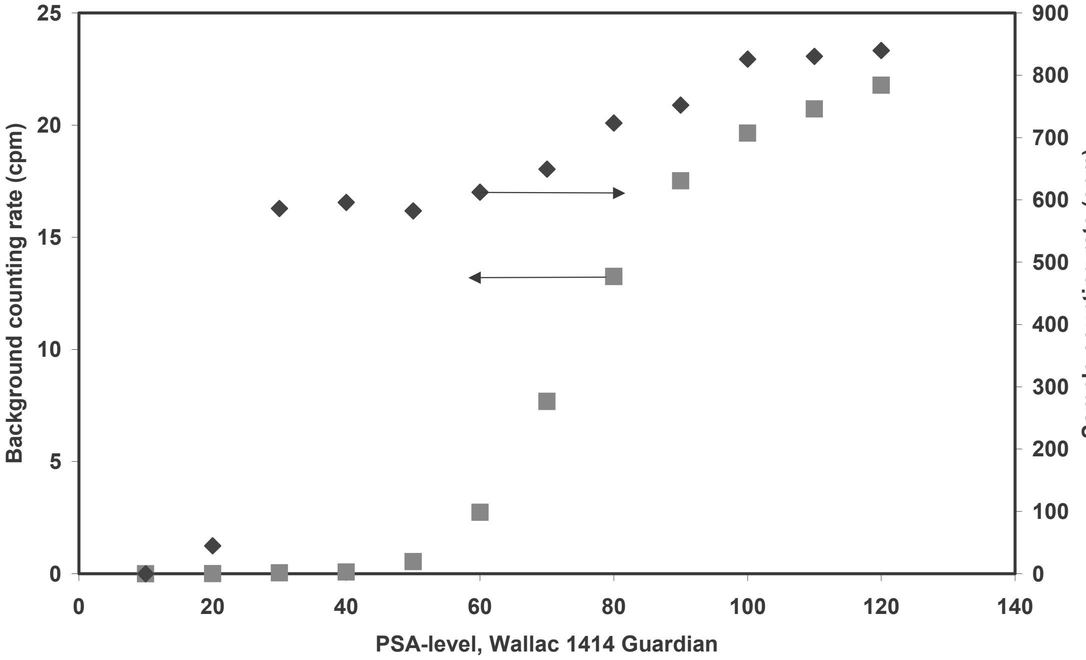


Figure 3-5 *Count rate of a groundwater sample containing radon (diamonds, projected to the right Y-axis) presented as a function of the PSA-level used. As a comparison, the background count rate (squares, projected to the left Y-axis) at different PSA-levels is also given in the figure.*

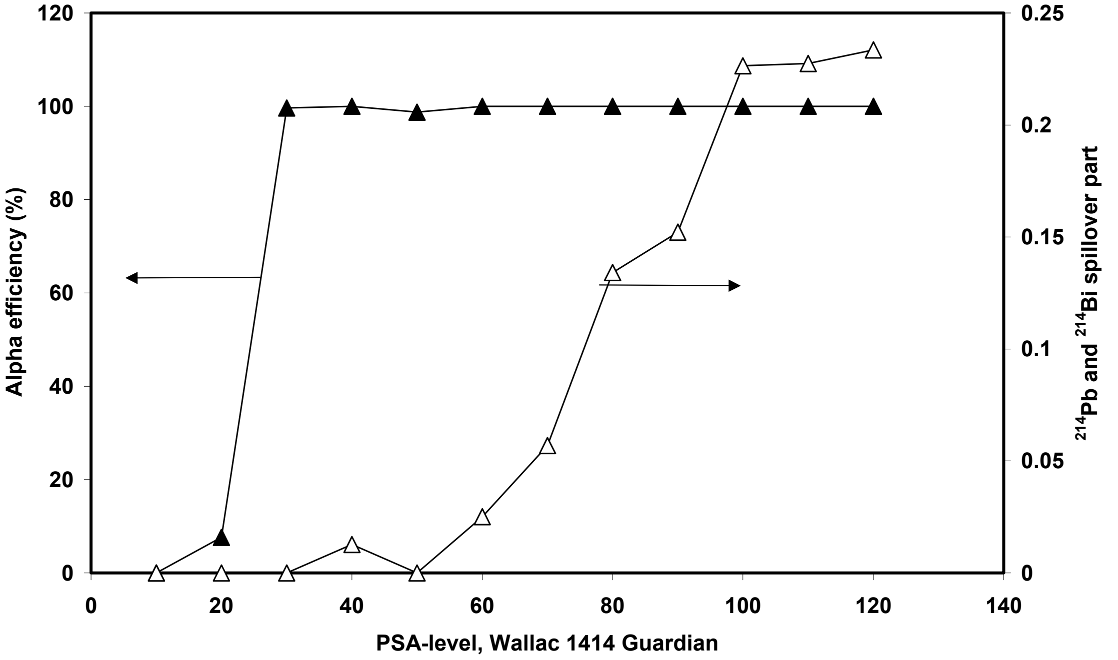


Figure 3-6 *Interpretation of the PSA-level characteristics obtained in the measurements of groundwater sample, original data presented in Figure 3-5. It is proposed that the α-efficiency (filled triangles, projected on the left Y-axis) is 100% already at a PSA-level of 30. The increase in the count rate with PSA-levels above 60 is caused by gradually increased β-spillover (opened triangles, projected on the right Y-axis) from the β-emitting radionuclides in the radon decay series, i.e., ^{214}Pb and ^{214}Bi.*

The α-spectrum of the radon sample, measured with a PSA level of 50, is shown in Figure 3-7. The non-symmetric double peaks can be explained by the decay of ^{222}Rn (5.5MeV) and ^{218}Po (6.0 MeV) which causes the broad low energy peak while the somewhat sharper high energy peak is caused the decay of ^{214}Po (7.7MeV). As a comparison, the spectrum of the pulses rejected in the pulse shape analyses, i.e., the β-decay pulses, is also given in the figure.

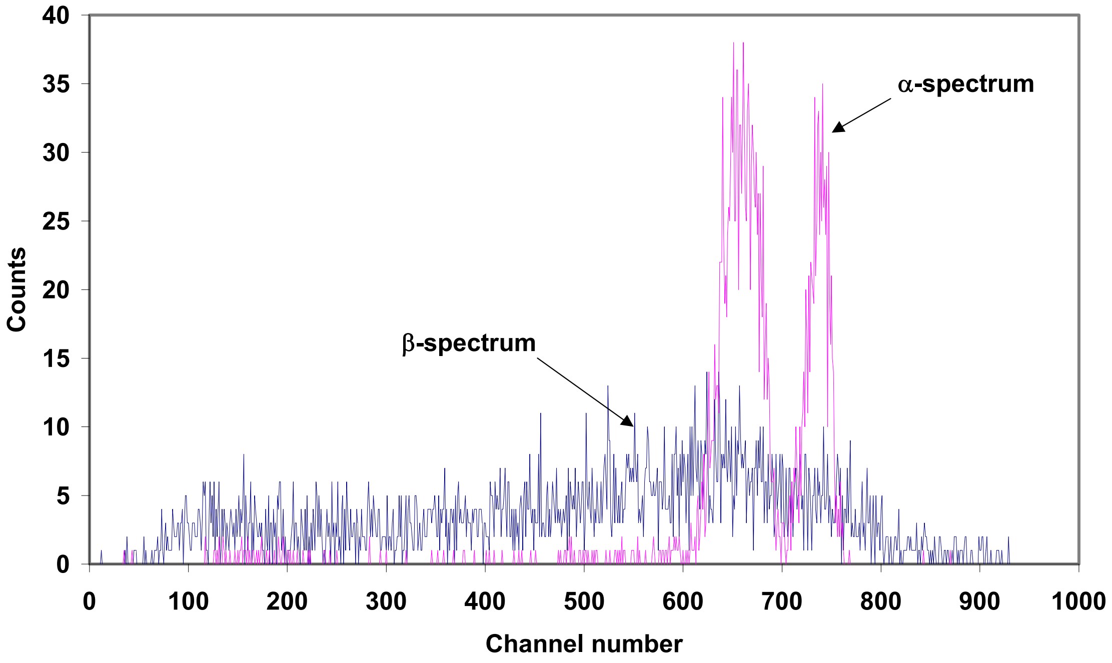


Figure 3-7 *α-spectrum (i.e., the counts registered in a measuring time of 11000s given as a function of the channel numbers) measured with PSA level 50 on the Wallac Guardian 1414. Besides the α-spectrum, a corresponding spectrum is given for the pulses rejected as α-pulses, i.e., the β-spectrum.*

Measurements were also performed using the same groundwater sample and repeating the liquid scintillation counting with time intervals of ~3 days. The results show (Figure 3-8) that the decrease of the measured count rate follow the tabulated half-life of ^{222}Rn, i.e., 3.824d. (Evaluation of the half-life from the experimental data gives 3.77±0.08 d). This observation shows that leakage of radon from the glass vials during the measurements is a minor problem. Furthermore, the results show that the concentration in the groundwater of the progenitor to ^{222}Rn, i.e., ^{226}Ra, is at least a factor of 100 lower than the radon concentration.

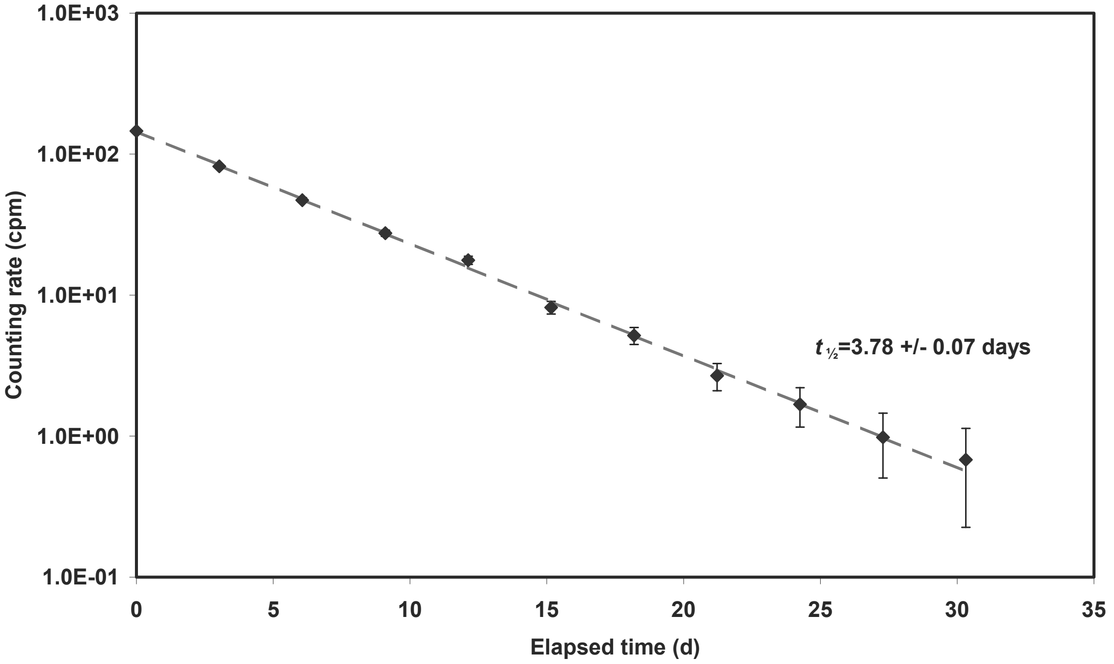


Figure 3-8 *Repeated measurements of a groundwater sample containing radon. The Results are given as the count rate reduced with the background counting. The results show that the activity decreases according to the tabulated half-life of* ^{222}*Rn, i.e., 3.824 d.*

Measurements were also performed by diluting the sampled groundwater 10 and 100 times. With this procedure, studies of the linearity of the count rate versus the radon concentration were obtained. The measurements were repeated 3 times with a 12 h interval. 4 different measurement series were obtained. The results (Figure 3-9) indicate a near linear relationship between the count rate and the radon concentration.

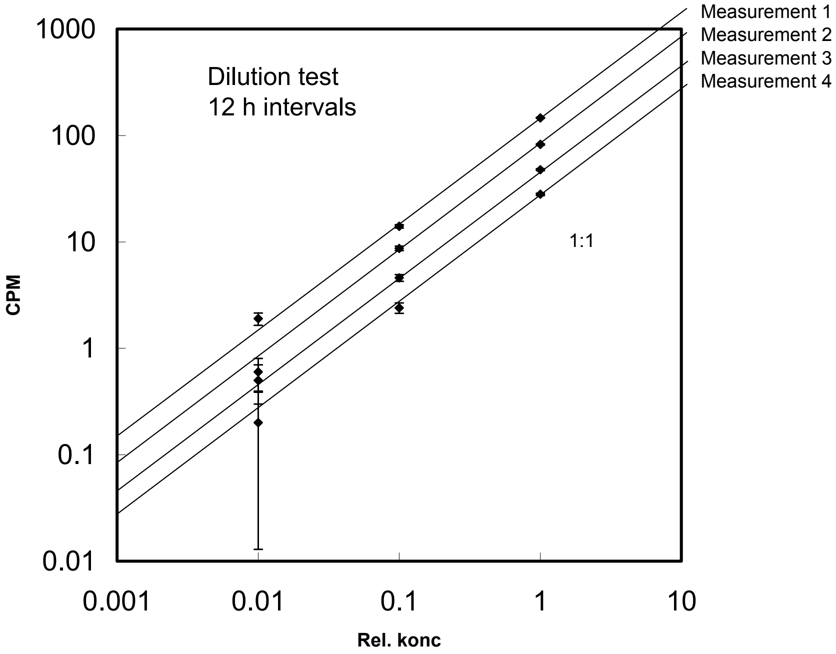


Figure 3-9 *Count rate of a sampled groundwater with dilutions of 10 and 100 times, see text for details*

An attempt to make an efficiency calibration was performed by sampling 5 ml volumes of the sampled groundwaters that previously had been measured with γ-spectrometry (Section 2.3). These small samples were measured using liquid scintillation counting (PSA-level of 40) and the count rate was registered as a function of the activities measured using γ-spectrometry. The results are shown in Figure 3-10. Using the assumption that only the α-decay in the radon decay series is registered (i.e., 3 pulses should registered per decay of ^{222}Rn) the apparent detection efficiency of the liquid scintillation was determined to 83.3±2.8 percent. However, this is in contradiction of the interpretation of the detector characteristics, previously presented in Figure 3-6, where a 100% α-efficiency was interpreted for a PSA-level of 40. A possible explanation of this deviation is that sampling of the pressurised groundwater may cause radon losses due to degassing of the sample. The obtained deviation from 100% efficiency in the liquid scintillation measurement may therefore be a result of losses during sampling rather than actual non-100% detection efficiency. Further investigations of this problem are probably necessary to sort this problem out.

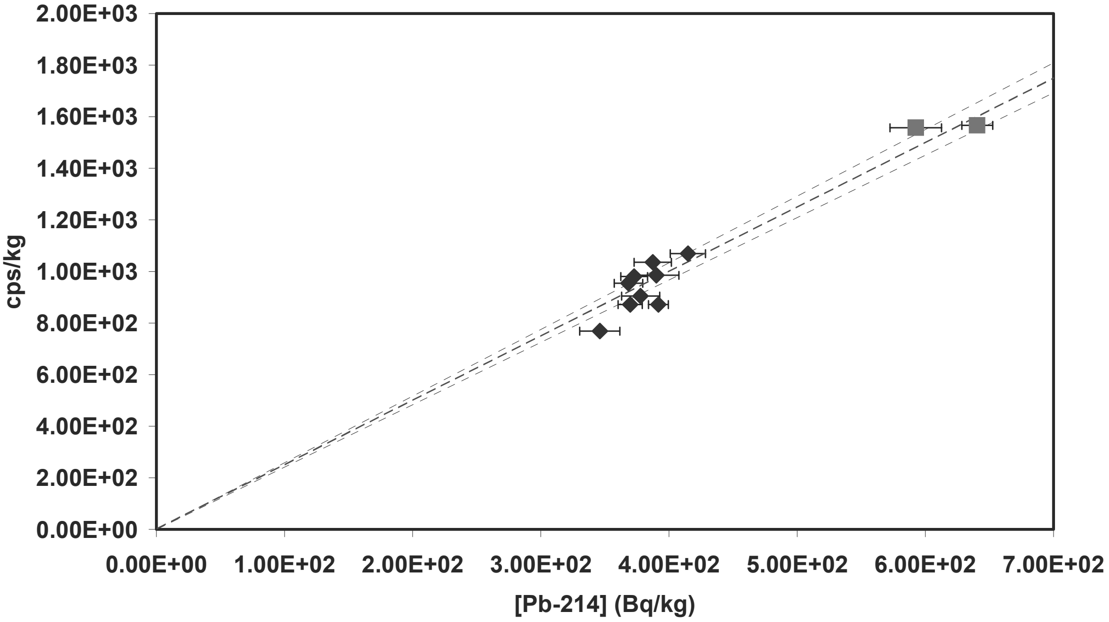


Figure 3-10 *Count rate in the liquid scintillation counting (PSA-level 40) as a function of the concentration ^{214}Pb (radon daughter product) in the same samples.*

4 Laboratory measurements of radon fluxes from rock material

4.1 Experimental

4.1.1 Gouge material

In this work, three different gouge materials originating from different places in the ÄHRL were selected for the measurements. The materials are noted KI0025F 194m, 1303-NE-1 and KAS 04:65m EW-1, cf. Smellie and Tullborg in prep. for the additional descriptions. The material was sieved and only the fraction <0.125mm was used in the experiment. Results of the elemental analysis of the fault gouge materials are given in Appendix A.

Duplicate samples were prepared of each gouge material and 1-1.5 g of the material was contacted with 5.2-5.8 g (exact amount registered by weighing) distilled water in a centrifugation vial. The amount of water was chosen in order to completely fill up the vial, i.e., to avoid as much air contact with the water phase as possible.

The samples were stored in a refrigerator for 42 days of in-growth of the radon. Refrigerator was used in order to mimic the *in situ* temperature in rock (diffusivity is temperature dependent) and the contact time was selected to ensure a full in-growth of the ^{222}Rn isotope and thus obtaining full steady-state conditions. After that, the vial was centrifuged (10000 rpm) and thereafter sampling of the water phase was performed. As much of the water phase as possible (4.3-5.0g, exact amount registered by weighing) was sampled using a syringe. The water sample was thereafter passed through a syringe filter (0.45μm) in to a scintillation vial filled with 15 ml of scintillation cocktail. The vial was subsequently completely filled up with scintillation cocktail; thereby avoiding radon degassing to any air phase in the scintillation vial.

4.1.2 Diffusion cells

In this experiment, 20 ml-diffusion cells (earlier used for the determination of diffusivity in intact unaltered Äspö rocks, i.e., Äspö diorite and Fine-grained granite, Byegård et al. 1998) were used for the determination of radon fluxes from generic rock types. In these cells (Figure 4-1) 20 ml of water was contacted to a surface of 10cm^2 of the rock specimen. Both sides of the diffusion cell were utilised for the radon flux measurement. Three different cells for each rock type were used, i.e., a total number of 6 radon flux measurements were obtained.

Besides the diffusion cells with intact rock types, investigation of the radon fluxes was also performed using material from the Feature A rim zone, the target fracture system for the TRUE-1 experiments (Winberg et al. 2000). In this experiment 50 ml-diffusion cells were used and a 4cm^2 area was exposed to this volume of distilled water. Three different diffusion cells were used; two with mylonite and one with altered diorite. Further details and descriptions of earlier experiments performed with these particular rock samples are found in Byegård et al. (2001).

Before the start of the experiment, a number of water exchanges (3 for the 20 ml cells, 2 for the 50 ml cells) were performed. All the water in the cells was extracted and was replaced by the same volume of fresh distilled water. After the last exchange, the cells were completely filled up with distilled water and were put in a refrigerator for radon in-growth (42 days). Subsequently, sampling was performed according to the procedures above (except that centrifugation was not performed and that a 5.0-5.3g water was sampled, exact amount registered by weighing). Since the first measurements showed that no radon flux could be detected from the Äspö diorite and the Feature A rim zone material, the experiment procedure was repeated for these samples in a second measurement campaign, performed similar as the one described above

Figure 4-2 *Photograph of a diffusion cell device used for the estimation of radon emanation from intact rock surfaces.*

4.1.3 Radon measurements

During the first measurement campaign, liquid scintillation measurements were performed on a Wallac Rackbeta 1219 at the Department of Nuclear Chemistry, Chalmers University of Technology. The PSA level was set to 135. Each sample was measured for 1.5 hours and 4-5 measurements of each sample was performed during a 7 day period. The energy discrimination was set such that only the α-pulses in the channels #500-900 (total number 1024) were registered.

Since the results of the first measurement campaign indicated that the background count rate was too high to statistically verify any radon flux from the intact Äspö diorite samples and the Feature A rim zone material, it was decided to repeat the experiment for these material. To possibly improve the measurement conditions, the following changes were applied in the second measurement campaign:

- The liquid scintillation counting device at BASLAB/CLAB (Wallac 1414 Guardian) was used for the measurement, applying a PSA-level of 40. Complementary to these measurements, the same samples were measured at Chalmers; however, with a PSA-level set to 160. Based on the earlier measurements (e.g., cf. Figure 3-2) it could be anticipated that these PSA levels would give a significantly decreased background count rate. However, a decrease in the efficiencies to the range of 80-90% should also be anticipated.

- It was decided that for the second measurement campaign, only to include the low activity side for the Äspö diorite samples that previously had been used for through diffusion experiment using $^{137}Cs^{+}$ and $^{133}Ba^{2+}$ tracers, respectively. As a consequence only 10 samples were measured and the measurement time could therefore be increased to 4.2 hours.

4.1.4 Evaluation procedure

Data was evaluated by means of a least squares sum fit of the count rate R_0. The error square sum, q, of the measured data points and the model is

$$q = \sum_{i=1}^{n} \left(y_i - f(x_i) \right)^2 \tag{4.1}$$

where y_i is the observed data and $f(x_i)$ is the model with the parameters to be evaluated. In the case herein (a radiometric measurement following the decay of the measured radionuclide) the error square sum can be written

$$q = \sum_{i=1}^{n} \left(R_i - R_0 \cdot e^{-\lambda \cdot t_i} \right)^2 \tag{4.2}$$

where R_i is the observed count rate at time t_i, R_0 the count rate at time $t_0=0$ (which is the parameter to be evaluated) and λ the decay constant of ^{222}Rn. The necessary condition for q to be minimum, with respect to R_0, is

$$\frac{\partial q}{\partial R_0} = 0 \tag{4.3}$$

Some algebra and solving for R_0 gives

$$R_0 = \frac{\sum_{i=0}^{n} R_i}{\sum_{i=0}^{n} e^{-\lambda \cdot t_i}} \tag{4.4}$$

This evaluation method has some basic metrological advantages:

- It is fully transparent, meaning R_0 is expressed in a mathematical formula which easily can be implemented in a combined uncertainty calculation
- A validation of the measured radionuclide is obtained, since any measurement not following the decay of ^{222}Rn will show data points outside the decay interval

Figure 4-2 illustrates the evaluation procedure from measurements of three experiments (numerical results are given in Section 4.2). The solid exponential functions in each plot show the decay interval at the 95% confidence level. Each data point is shown with a combined uncertainty having a coverage factor $k=2$, i.e., given with ~95% confidence. Combined uncertainty calculations were done according to the ISO/GUM approach (International Organisation of Standard, 1995). It can be observed in the different figures that all experiments follow the decay of ^{222}Rn.

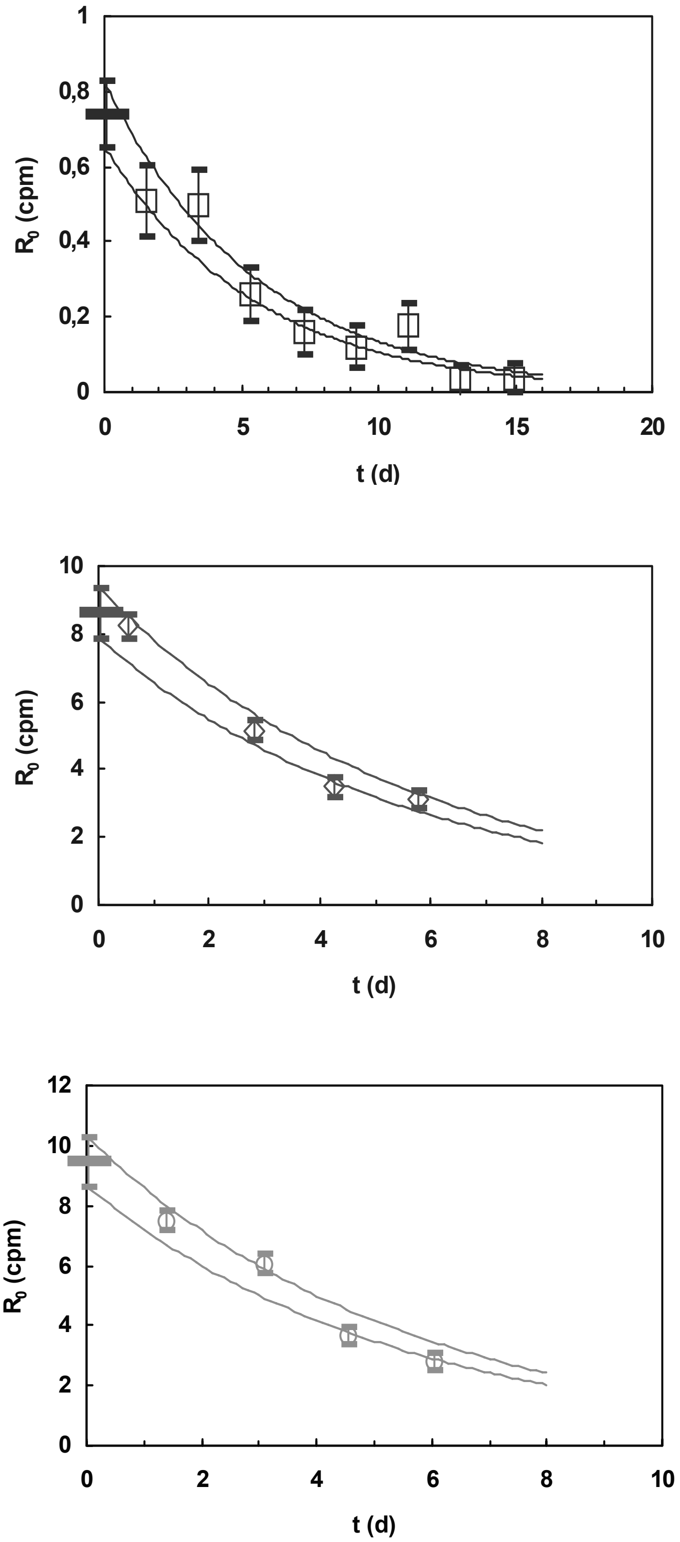


Figure 4-2 *Count rate (α-mesurements) of water in contact with different geologic material (top: intact unaltered Äspö diorite, middle: Fault gouge material, bottom: Fine grained granite. Repeated measurements were done using ~1.5 days interval. The evaluated count rate at t=0(sampling time) is illustrated together with the uncertainty obtained for that value.*

4.2 Results

4.2.1 Background measurements and detection limits

The lowest background levels were obtained for the second measurement series. In these measurements, a background count rate of 0.054±0.006 counts per minute (cpm) was obtained. Applying a detection limit (paired observation) according to Currie (1968) would correspond to a detection limit of 0.028 cpm, corresponding to 0.03 Bq/l. This can be transferred to radon flux detection limits, according to: :

- 0.6 $s^{-1}m^{-2}$ for the experimental device of 20 ml of water in contact with 9.7 cm^2 rock surface area (i.e., the diffusion cells for the unaltered rock types)
- 3.2 $s^{-1}m^{-2}$ for the experimental device of 50 ml of water in contact with 4 cm^2 rock surface area (i.e., the diffusion cells used for the Feature A rim zone rock material)
- 0.16 $s^{-1}m^{-2}$ for the experimental device of 10 ml of water in contact with 20 cm^2 rock surface area (i.e., an experimental device optimised for radon flux measurements)

The detection limits are based on a single measurement and are given under the assumption that all α-activity measured originates from ^{222}Rn. However, to obtain a better verification of the existence of ^{222}Rn in the water, a decrease in the count rate compatible with the half-life of ^{222}Rn should be addressed in the evaluation. With an application of the evaluation procedure described in Section 4.1.4, a detection limit of 11 $s^{-1}m^{-2}$ was estimated for one of the cells containing Feature A rim zone rock material. One can thus estimate that a demand of a half-life verification of ^{222}Rn in the measurements will increase the given detection limits above by approximately a factor 3.

4.2.2 Spectrum measurements

Examples of α-spectrum obtained in these low-concentration measurements of radon and background measurements are shown in Figure 4-3. Although the activity is much lower, the shape of the spectrum is similar to the ones obtained for the groundwater measurements (cf. Figure 3-6). The characteristic double peak is clearly observed and can be regarded as an experimental proof for the existence of radon in the sample.

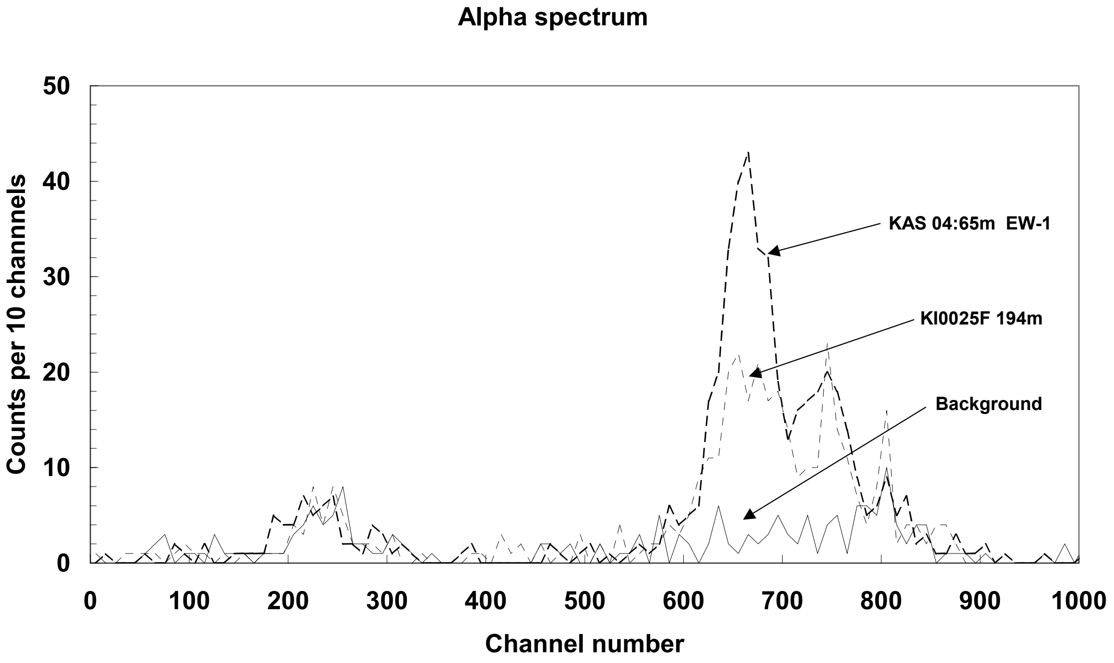


Figure 4-3 *α-spectra obtained for the low-concentration radon measurements. Examples are given for two samples (fault gouge materials) and these spectra are compared to the spectrum obtained in a background measurement. In this presentation, the counts registered in 10 channels have been summed up in each individual data point.*

4.2.3 Half-life measurements

Besides the spectrum information, qualitative identification of radon may be obtained by measuring the decrease of the count rate with the elapsed measuring time, i.e. half-life verification. As can be seen in Figures 4-4, 4-5 and 4-6, the decrease in activity follows very well the rate given by the tabulated half-life of ^{222}Rn (3.824 d)

4.2.4 Radon concentrations

The obtained radon concentrations in the different laboratory experiment are presented in Table 4-1 and Table 4-2; together with different supporting data. A general trend is that the radon emission from Fine Grained granite samples and the fault gouge materials can be rather easily quantified. On the contrary, the radon emission from intact unaltered Äspö diorite and from the rim zone material of Feature A is more difficult to quantify. An interesting observation is that the uranium concentration in Fine grained granite according to the available data is not very much higher than what is found for the intact Äspö diorite. The reason for the very different radon fluxes for these materials can not be explained by the uranium concentration data. It is thus indicated that the distribution of radium and the textural composition of the rock material could have an impact in the radon flux of the rock material. Further research is probably necessary in order to answer these questions.

Several of the data-sets obtained for the intact rock samples were not possible to use for evaluation of the radon flux. The samples #22A and #23A had earlier been used as the high activity side for ^{137}Cs^{+} in diffusion experiment. In the radon measurements of water sample from these cells, a high α-count rate was observed which showed no indication to decrease according to the half-life of ^{222}Rn. The interpretation to this is that desorption of $^{137}Cs^+$ had occurred from the rock surfaces. In the radon measurements the desorbed $^{137}Cs^+$ gave "β-spillover" in the measurements (cf. Section 3.2.1) and the results could therefore not be used. The same interferences occurred for sample #19A which had been used as the high activity side for $^{133}Ba^{2+}$ in diffusion experiment in intact Äspö diorite. However, the data obtained for #19A (high activity side for $^{133}Ba^{2+}$ in diffusion experiment in intact Fine grained granite) showed no pronounced interferences in the α-counting. The reason for this is that Fine grained granite is less sorptive compared to Äspö diorite and also that Ba is not such a strong sorbing tracer as Cs (Byegård et al. 1998). Furthermore, the decay characteristics of ^{133}Ba (ε,γ) is not suspected to give as much "β-spillover" as ^{137}Cs.

The radon concentration in the diffusion cells containing Feature A rim zone material gave α-count rates that could not be distinguished from the background count rate. The only data set that was used was the one from #T2A, the B-side, which was used for an estimation of a radon flux detection limit for this experimental device. No evaluation of the other data-sets from diffusion cells containing Feature A rim zone material was peformed

When assessing no measurable radon flux from the Feature A rim zone material, one should have in mind that the experimental device used consists of comparatively large water volumes (two 50 ml cells) in contact with very small amounts of rock material (2x2 cm surfaces with a depth of 3, 5 and 8 mm respectively). Laboratory experiments with similar material (Byegård et al. 2001) have indicated a very heterogeneously distributed porosity with several microfractures present. A possibility is that radon is produced in microfracture can diffuse rather long distance. The limited depth used, especially in these experiments, may give erroneous results.

Based on the assumption that ^{226}Ra is in a secular equilibrium with the uranium, one can from the measured uranium concentration of the gouge material estimate that 20-36% of the Rn produced in the fault gouge material reaches the water phase. This means that even for the very fine-powdered fault gouge material (<0.125mm) the vast majority of the radon atom produced in the decay of the radium will be trapped in the crystalline rock structure. It seems reasonable to assume that for the intact unaltered rock where the grains are larger, the numbers of radon atoms that after the decay will be trapped within the crystalline structure rock will be even higher. For evaluation of the non-released part in the experiments using intact crystalline rock, diffusion calculations (e.g., according to the concept presented by Neretnieks 2001) are necessary in which the porosity and the diffusivity have to be known.

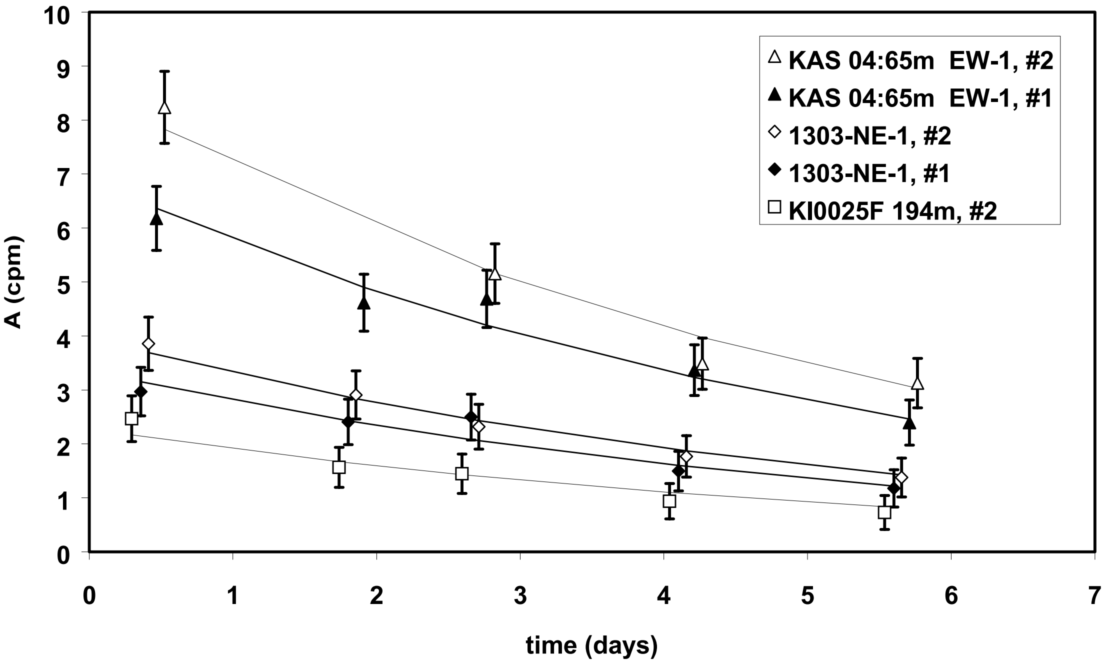


Figure 4-4 *Count rate from the water samples in contact with the fault gouge material (size<0.125mm). The dots corresponds the measured count rate, the uncertainty given is based on counting statistics given with a 95% confidence interval. The lines represent the best fit of a decay curve with a half-life of 3.824 d (the tabulated half-life of ^{222}Rn)*

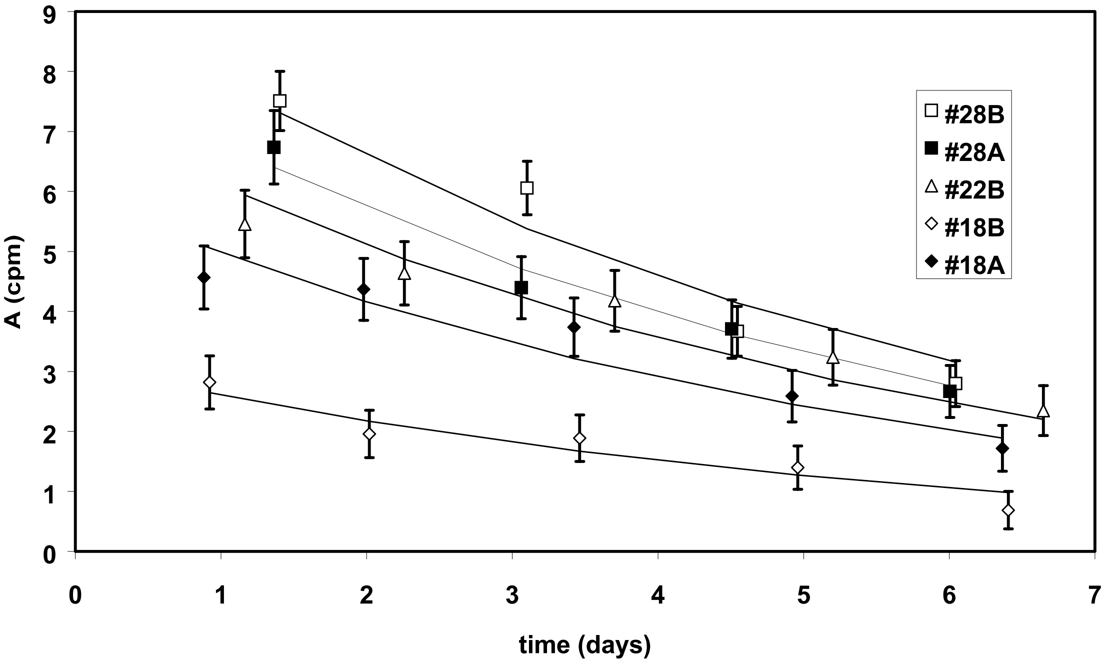


Figure 4-5 *Count rate from the water samples in contact with the Fine grained granite (intact un-altered rock). The dots corresponds the measured count rate, the uncertainty given is based on counting statistics given with a 95% confidence interval. The lines represent the best fit of a decay curve with a half-life of 3.824 d (the tabulated half-life of ^{222}Rn)*

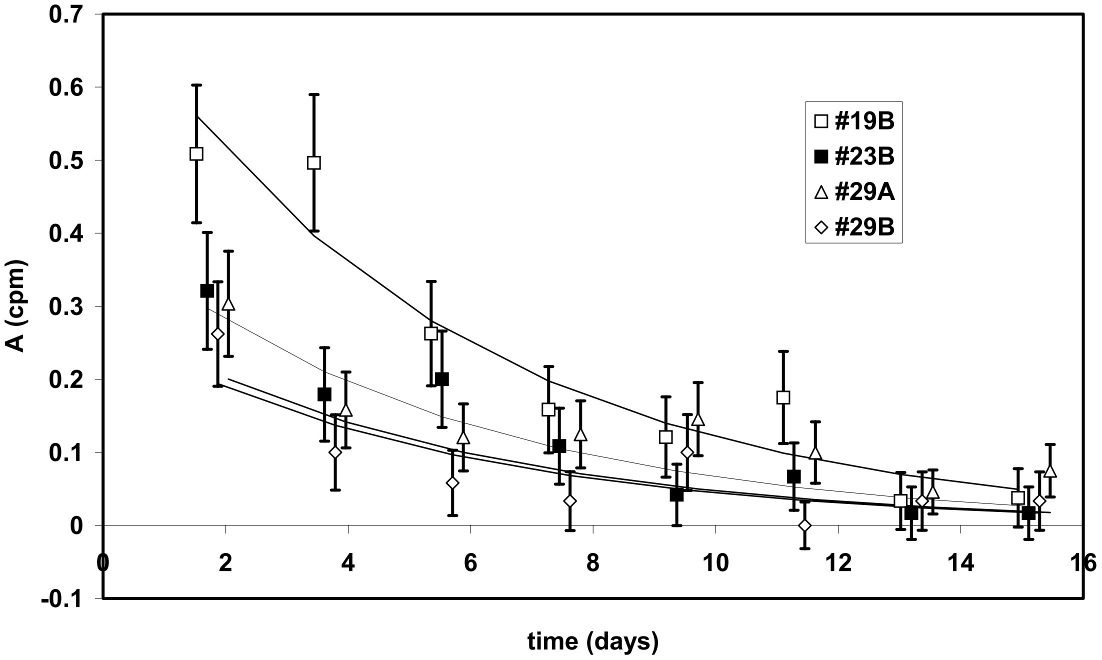


Figure 4-6 *Count rate from the water samples in contact with the Äspö diorite samples (intact unaltered rock). The dots corresponds the measured count rate, the uncertainty given is based on counting statistics given with a 95% confidence interval. The lines represent the best fit of a decay curve with a half-life of 3.824 d (the tabulated half-life of ^{222}Rn)*

Table 4-1 Results of the radon flux measurements on intact rock material, given combined with some geological properties of the different samples that have been used in the investigation.

#	side	Rock type	Sample length (cm)	Contact area (cm^2)	ε A) (vol%)	D_e B) (m^2/s)	[U] C) (ppm)	Cell. Vol. (ml)	[^{222}Rn] Bq/l	Rn–flow (m^{-2}s^{-1})
18	A	FGG	1	9.7	0.12	6.8e-14	3.8	20	6.6±0.8	137±17
	B							20	3.5±0.8	73±17
19	A	ÄD	1	9.7	0.35	9.6e-14	2.6	20	Excluded	
	B							20	0.74±0.08	16±2
22	A	FGG	1	9.7	0.41	1.8e-13	3.8	20	Excluded	
	B							20	8.0±0.8	165±20
23	A	ÄD	1	9.7	0.21	1.4e-13	2.6	20	Excluded	
	B							20	0.46±0.08	9.6±1.7
28	A	FGG	2	9.7	Not	Not	3.8	20	8.9±0.9	186±24
	B				Meas.	Meas.		20	11±1	224±20
29	A	ÄD	2	9.7	Not	Not	2.6	20	0.31±0.08	6.5±1.6
	B				Meas.	Meas.		20	0.31±0.08	6.4±1.6
T2A	A	Myl.	0.5	4	0.54	1.6e-13	5.0	50	Not evaluated	
	B							50	<0.098	<11
T2B	A	Myl.	0.3	4	0.41	4.4e-14	5.0	50	Not evaluated	
	B							50	Not evaluated	
T2C	A	Alt. ÄD	0.8	4	0.33	1.6e-13	3.7	50	Not evaluated	
	B							50	Not evaluated	

A) Sample 18-29, Johansson et al. 1997, T2A-T2C, Byegård et al, 2001.

B) Sample 18-29, Johansson et al. 1997, T2A-T2C, Byegård et al, 2001, diffusivity determined for tritiated water in the direction A→B

C) Byegård et al. 1998

Table 4-2 Results of the radon flux measurements of gouge material (sieved, <0.125mm size fraction used in the experiment).

Sample	#	Rock material (g)	[U] A) (ppm)	^{234}U/^{238}U A)	Water (g)	[^{222}Rn] Bq/l	Rn–flow (kg^{-1}s^{-1})	η
KI0025F 194m	1		6.2	1.08			Sample failure	
	2	1.52			5.21	2.5±0.7	10±3	0.20±0.05
1303-NE-1	1	1.44	8.9	1.2	5.43	3.6±0.7	17±3	0.23±0.04
	2	1.48			5.53	4.0±0.7	17±3	0.23±0.04
KAS 04:65m EW-1	1	1.09	21	1.34	5.76	7.0±0.7	42±5	0.36±0.04
	2	1.42			5.66	8.7±0.8	40±4	0.34±0.03

A) Data a-spectrometry measurements, Smellie and Tullborg, in prep

5 Conclusions

- The *in situ* sampling and radon measurements at the TRUE-1 site has shown that very similar radon concentration is measured for different groundwater samples within a singular Feature.
- Scintillation with pulse shape analysis (PSA) has proved to be a highly sensitive method for low-level measurements of radon concentrations.
- Using diffusion-cell device in laboratory experiments has shown that radon fluxes can be measured using a rather simple experimental set-up. An optimised experimental set-up, i.e., 10 ml of water with $20 cm^2$ of rock surface area should enable measurement of radon fluxes of $0.15\ s^{-1}m^{-2}$. However, with demand of a half-life verification of the radon, the detection limit is increased to $\sim 0.5\ s^{-1}m^{-2}$.
- The results obtained for the radon fluxes from the fault gouge materials involved in this study has indicated that no evidence of that the gouge material should be the dominant source of radon to the groundwater to the fracture.

6 Recommendations for future research

Some complementary tasks that remain and are associated to the activities described in this report are:

- Obtaining a better understanding of the liquid scintillation counting efficiency in the radon measurements and the impact of the "β-spillover" as a function of the PSA-level
- Obtaining a quantification estimate of the magnitude of the degassing of radon in sampling involving depressurization.

Based on the progress made in the project, the following tasks are foreseen for obtaining increased understanding of the interaction of the radon and the groundwater.

- Investigate the possibilities of non-destructive radium measurements of intact rock samples using γ-spectrometry.
- *In situ* experiments in order to verify the mechanisms of radon flux from rock surfaces to groundwaters. This experiment should be performed in a dipole flow geometry experiment in which water without dissolved radon is injected. The decrease in radon concentration obtained during the "breakthrough" of this water should provide confirmation of the basic principles assumed for the mechanisms of the radon flow. Furthermore, information could be obtained on the magnitude of the radon diffusion parameters, the release factor (η), the pore diffusivity (D_p) and the rock porosity (ε).
- Sensitivity analyses of the diffusion parameters. If very low fracture apertures are estimated from the experimental results (i.e., low radon flux from the laboratory samples combined with high radon concentration in the sampled groundwater) it has been shown (Neretnieks 2001) that a simple surface production model is not valid for the calculation of the fracture aperture. Instead a diffusion model has to be applied in which the release factor (η), the pore diffusivity (D_p) and the rock porosity (ε) will be included. Theses are likely to be assigned with a considerable uncertainty and the combined uncertainty calculation should be performed to estimate the sensitivity of the different parameters.
- The potential impact of heterogeneously distributed porosity should be investigated
- A test should be made to see if the indication of unique radon concentrations in individual fractures in the TRUE-1 site can be verified by radon measurements in the TRUE Block Scale site (cf. Andersson et al. 2002)

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Appendix A

Chemical analyses of the fault gouge material used in the investigation, analyses performed using ICP-MS at SGAB, Luleå, except for the isotope activity ratios which have been determined by α-spectrophotometry at Studsvik Nuclear AB. Further details are given by Smellie and Tullborg, in prep

	KI0025F 194m	**1303-NE-1**	**KAS 04:65m EW-1**
U (mg/kg)	6.24	8.9	21
$^{234}U/^{238}U$	1.08	1.2	1.34
$^{230}Th/^{234}U$	0.93	0.89	0.94
Th (mg/kg)	37.4	8.55	20
$^{228}Th/^{232}Th$	1.14	1.4	1.36
Al_2O_3 (%)	12.8	11.3	22.2
CaO	1.23	1.16	0.609
K2O	6.33	5.81	10.7
MgO	0.932	1.74	2.04
MnO	0.0392	0.0635	0.0353
Na_2O	2.41	0.142	0.14
P_2O_5	0.139	0.108	0.114
SiO_2	70.2	72.1	55
TiO_2	0.389	0.243	0.528
LOI	1.4	3.1	4.7
Sum	97.7	97.2	94.9
Ba (mg/kg)	1040	1380	1190
Be	1.69	7.35	13.2
Co	<6	7.89	8.91
Cr	18.4	139	15.3
Cu	201	133	396
Ga	45.9	48.2	51.9
Hf	6.69	2	11.8
Mo	66.4	11.1	<2
Nb	16.6	5.99	38.5
Ni	<10	29.9	<10
Rb	166	226	595
Sc	2.09	5.35	6.11
Sn	2.68	7.68	20
Sr	184	294	110
Ta	0.557	0.484	3.33
V	41.8	92.1	23.7
W	158	1.84	3.04
Y	24.6	17.1	46
Zn	71.8	140	153
Zr	396	99.7	566
La (mg/kg)	39.2	30.3	38.2
Ce	89.7	63.4	60.7
Pr	10.1	7.15	8.68
Nd	36.7	23.2	30
Sm	6.67	4.18	4.79
Eu	0.979	0.428	0.644
Gd	5.41	2.68	4.46
Tb	0.843	0.395	0.884
Dy	4.18	1.56	6.08
Ho	0.876	0.351	1.42
Er	1.91	0.879	6
Tm	0.266	0.147	0.877
Yb	1.93	0.937	5.76
Lu	0.308	0.191	1.01