



Comparison of the analytical methods used to determine natural and artificial radionuclides from environmental samples by gamma, alpha and beta spectrometry

Final Report from the NKS-B CAMNAR activity

Pöllänen, Roy; Virtanen, Sinikka ; Kämäräinen, Meerit ; Kaipainen, Maria ; Hou, Xiaolin; Roos, Per; Eriksson, Mats; Ehrs, Sara; Nalbandyan, Anna ; Møller, Bredo; Jonsson, Sofia ; Lagerkvist, Petra; Tovedal, Annika

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samples by gamma, alpha and beta spectrometry

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Abstract

In CAMNAR, an extensive interlaboratory exercise on the analytical methods used to determine several radionuclides present in the environmental samples was organized. Activity concentration of different natural radionuclides, such as Rn-222, Pb-210, Po-210, K-40, Ra-226, Ra-228 and isotopes of uranium, in addition to artificial Cs-137 and Am-241 were analysed from lake sediment samples and drinking water. The measurement techniques were gamma-ray spectrometry, alpha spectrometry, liquid scintillation counting and inductively coupled plasma mass spectrometry. Twenty six laboratories from nine Nordic and European countries participated in the intercomparison. Extraordinary variation between the results reported by different laboratories were revealed for some radionuclides indicating the need of future intercomparisons especially in the case of natural water samples.

Key words

Intercomparison, gamma-ray spectrometry, alpha spectrometry, liquid scintillation counting, inductively coupled plasma mass spectrometry

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(Contract: AFT/B(17)5)

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1. Introduction

Accurate determination of radionuclides from various sources is essential for assessing potential hazards associated with nuclear accidents/incidents as well as in the case of authorised releases to the environment. High-quality analytical capabilities are also necessary, for example in the surveillance of environmental radioactivity and other measurement activities in the field of nuclear safety, security and safeguards.

To ensure reliability of the results, a quality management system need to be established. All the used methods must be validated before they can be applied. This includes assessment of the sensitivity, detection limits and estimation of the uncertainties. In addition, selectivity and specificity, range and linearity, repeatability and reproducibility as well as stability and robustness against external influences are of importance (Ikäheimonen et al., 2006; Quevauviller, 2002; IUPAC, 2002). The accuracy of the method is confirmed by using certified reference materials and spiked samples, and by comparing against results obtained by using other methods or by performing interlaboratory comparisons.

The CAMNAR project aims to improve the quality and the capabilities of the participating laboratories by conducting an extensive interlaboratory exercise on the analytical methods used to determine several radionuclides present in environmental samples. Assessing the usability of the methods and their differences for determining various natural radionuclides (such as Rn-222, Pb-210, Po-210, isotopes of uranium) and some artificial radionuclides (e.g. Cs-137 and Am-241) from the environmental samples are of importance for the project.

The basic ideas on how to carry out the intercomparison and how to select the radionuclides of interest were as follows:

- The intercomparison should not be too easy (i.e. the samples should not contain only Cs-137, for example) but at the same time the case should not be too complicated.
- There should be a possibility of using different measurement techniques (not only gamma-ray spectrometry).
- The samples should contain natural as well as artificial radionuclides.
- The nuclides present in the samples should cover a wide range of gamma-ray energies (46 keV–1460 keV), which is important for validating the efficiency calibration.
- Some difficult-to-determine nuclides should also be present in the samples.

In the intercomparison, the participating laboratories were to analyse the activity concentrations of the selected radionuclides in a lake sediment sample and in a drilled well water sample by employing different analytical techniques routinely used in each laboratory. The measurement techniques include gamma-ray spectrometry, alpha spectrometry, liquid scintillation counting and mass spectrometry. The results as well as descriptions of the methods used in the intercomparison are presented here.

One of the current challenges in the Nordic countries is determination of Rn-222. High activity concentrations of natural radionuclides may occur in groundwater, especially in drilled well water, and to ensure the safety of the public the monitoring of Rn-222 in groundwater is needed. The need for Rn-222 determination has increased after the Commission of the European Communities issued a Council Directive 2013/51/Euratom that lays down requirements for the protection of the health of the general public with regard to radioactive substances in water, intended for human consumption. The directive suggest

remedial actions if the radon concentration exceeds 1000 Bq/L in public or commercial distribution. Water supplies with radon concentrations below 100 Bq/L are exempted from remedial actions.

The importance of Rn-222 measurement in environmental monitoring in its turn has raised a need for accreditation, particularly in laboratories carrying out measurements required by authorities. From the point of view of accreditation, successful participation in intercomparisons is being regarded as the best way of verifying good analytical quality (European Standard EN ISO/IEC 17025, 2005; Ikäheimonen et al., 2006). However, despite this importance, very few interlaboratory comparisons have been performed for Rn-222.

2. Intercomparison samples

Environmental samples – sediment material from lakes and water from a drilled well – chosen in this intercomparison are those that are commonly analysed by the participating laboratories.

The bottom sediment material was taken from several small lakes at 2012–2013 in Kuusamo and Sotkamo (Fig. 1a). The material was put together, dried and mixed to obtain homogeneous sample material (Fig. 1b). Since the amount of the sediment material was limited (around 1 kg, 2.8 L) part of the material was analysed in several laboratories. According to the previous analyses performed for the sediment material, the material contains naturally occurring radionuclides and small amounts of Cs-137 and Am-241.

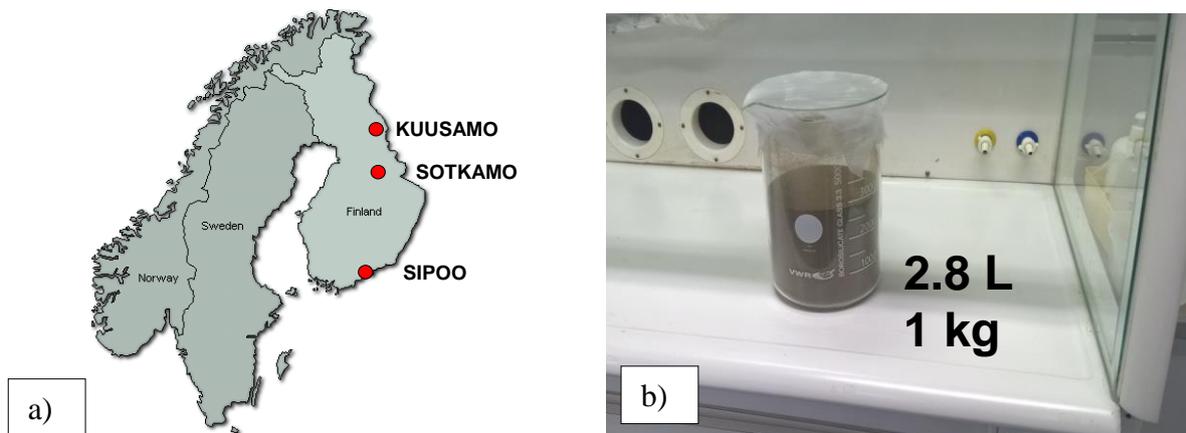


Figure 1. a) Location of the sampling sites. b) Homogenized sediment material.

The water samples were from a drill well taken in Sipoo at 2017. According to the previous analyses the raw water has elevated concentrations of U and Rn. The water was for household consumption and a purification system was used to remove U and Rn from the water (Fig. 2). However, this system was bypassed in order to get the raw water. Water was run for 20 min to empty the pressure vessel and to get the raw water directly from the well.

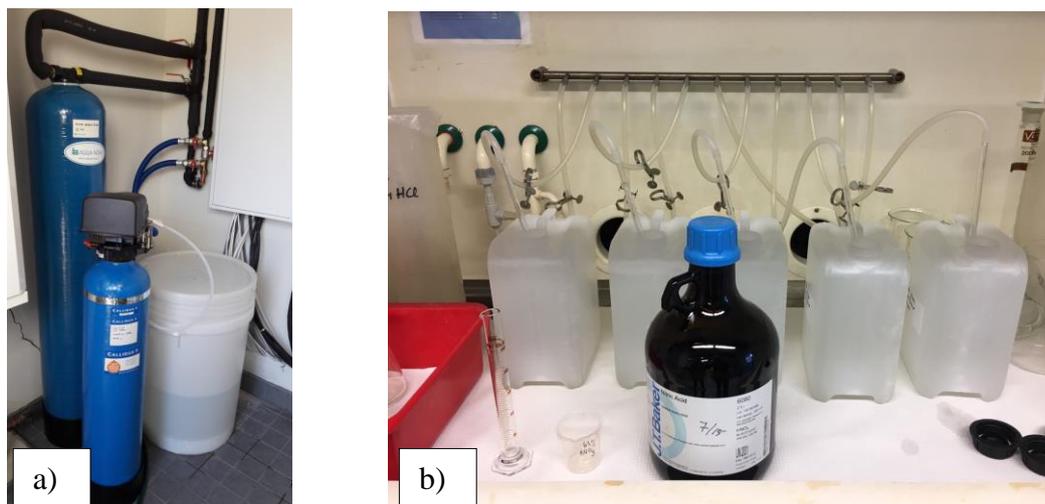


Figure 2. a) Water purification system. b) Bubbling and acidifying the water.

The activity concentration of Rn-222 in the raw water is of the order of 4–6 kBq/L, originating from Ra-226 in the bedrock. However, Ra-226 concentration in water is low. To avoid

possible systematic errors for reporting the results caused by the Ra-226/Rn-222 disequilibrium this excess Rn-222 must be removed from the water. This is why water was bubble aerated and acidified before sending it to participating laboratories. For Rn-222 analyses 1 litre sample in a glass bottle was taken, and this sample was not acidified or bubble aerated. Samples were also taken directly to liquid scintillation bottles that were prefilled with the scintillation cocktails that of each of the participating laboratories uses.

The radionuclides to be determined in the sediment and water samples were specified before the intercomparison. A special format was used for reporting the results (Fig.3).

Nuclide	Activity (Bq/kg)	Uncertainty (k=1)
Cs-137		
Am-241		
K-40		
Pb-210		
Th-234 (U-238)		
Ra-226		
Ra-228		
U-235		
Pa-234m (U-238)		

Nuclide	Activity (Bq/L)	Uncertainty (k=1)
Pb-210		
Th-234 (U-238)		
Ra-226		
U-235		
Pa-234m (U-238)		

Figure 3. Reporting format for the sediment sample (left) and the water sample (right) for gamma-ray spectrometry. Reporting formats for radiochemical measurements and mass spectrometry are not presented here. The nuclides selected for reporting are in the leftmost columns.

Three aliquots from the sediment sample material were measured by gamma-ray spectrometry in order to verify the homogeneity of the material. These tests were done before sending the sample material to the participating laboratories. The material was proved to be homogeneous (Fig. 4).

The water samples were assumed to be homogeneous and only the homogeneity of Rn-222 in untreated water was tested by taking 8 samples when filling the bottles and scintillation vials. In addition, 5 glass bottles were taken and opened and analysed after each laboratory had confirmed they had received their samples. These tests showed that samples should be homogeneous and the result obtained for the samples stored in glass bottle showed that only 2–3% of Rn-222 in the sample had escaped during storage. However, at least one of the laboratories reported that the sample had leaked, so it is also possible that Rn-222 that escapes during the actual shipping is greater.

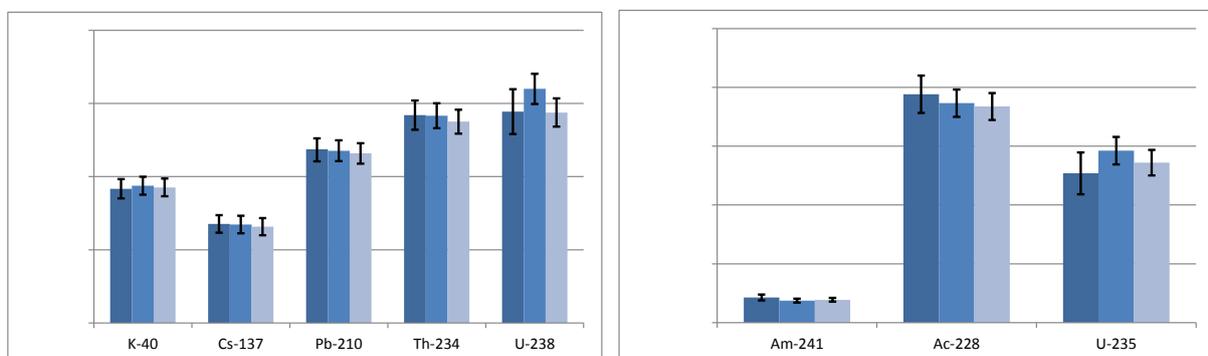


Figure 4. Results of the homogeneity tests from three different aliquots of the sediment sample material. The y-axis represents the activity concentrations (Bq/kg) of the nuclide in question present in the sediment. The uncertainties refer to the coverage factor $k = 1$.

3. Participating laboratories

Laboratories from nine different countries were involved in the intercomparison. In addition to the laboratories of the CAMNAR partners (laboratories N, P, U, V and Y in Table 1), 21 other laboratories participated in the intercomparison although not all of them reported the results. Their contributions were mainly in the framework of the following activities:

- NKS GammaSpec 2017 seminar (Nielsen et al., 2017).
- Joint Nordic-Baltic intercomparison organized in cooperation between the NKS CAMNAR project and the Council of the Baltic Sea States (CBSS), Expert Group on Nuclear Radiation and Safety (contact persons: Janusz Gąciarz (CBSS Secretariat) and Anna Nalbandyan, NRPA (coordinator of cooperation between gamma-spectrometric laboratories under CBSS EGNRS)).

The presence of other than CAMNAR laboratories in the intercomparison was highly appreciated since their contribution gives additional information, especially regarding the conclusions, but also for giving added value for the results in the project. Particular benefit is improvement of the international cooperation. On the other hand, possibility for this broad intercomparison provided a unique opportunity for comparison of methods and measurement results, and checking of the overall performance between broad spectrum of the laboratories (authorities, research institutions, industrial sites labs, national sanitary control services, etc.) from many countries.

Table 1. Laboratories participated in the intercomparison (in alphabetical order).

A	Bundesamt für Strahlenschutz / Federal Office for Radiation Protection, GERMANY
B	Center for Physical Sciences and Technology (FTMC), Metrology department, Ionizing Radiation Metrology Laboratory, LITHUANIA
C	Central Laboratory for Radiological Protection Dosimetry Department, POLAND
D	Central Mining Institute, Silesian Centre for Environmental Radioactivity, POLAND
E	Cyclife Sweden AB, SWEDEN
F	Dansk Dekommissionering, DENMARK
G	Ignalina Nuclear Power Plant, Laboratory of Environment Monitoring, LITHUANIA
H	Institute for Energy Technology, Health and Safe Department Environmental Monitoring Section, NORWAY
I	Institute of Marine Research, NORWAY
J	Landesmessstelle für Radioaktivität, Fachbereich Physik/Elektrotechnik, Universität Bremen, GERMANY
K	National Centre for Nuclear Research, Dosimetric Surveys Lab, LPD, POLAND
L	National food and veterinary risk assessment institute, Radiology group, LITHUANIA
M	Nature Research Centre, LITHUANIA
N	Norwegian Radiation Protection Authority, Østerås, Tromsø and Svanhovd, NORWAY
O	Polish Academy of Sciences, The Henryk Niewodniczanski Institute of Nuclear Physics, Nuclear Physical Chemistry Department, POLAND
P	Radiation and Nuclear Safety Authority, STUK, FINLAND
Q	Radiological Division of Environmental Protection Agency, LITHUANIA
R	SCK•CEN Labo LRM, BELGIUM
S	State Ltd, Latvian Environment, Geology and Meteorology Centre, LATVIA
T	Sundhedsstyrelsen, Strålebeskyttelse, DENMARK
U	Swedish Defence Research Agency, FOI, SWEDEN
V	Swedish Radiation Safety Authority, SSM, SWEDEN
X	Svensk Kärnbränslehantering, SKB, SWEDEN
Y	Technical University of Denmark, Center for Nuclear Technologies, DTU-Nutech, DENMARK

4. Description of the analytical methods used in the intercomparison

There are different techniques for analysing activity concentrations of natural and artificial radionuclides from environmental samples. High-resolution gamma-ray spectrometry is commonly used in the determination of both man-made and naturally occurring radionuclides. The advantage of this technique is that several radionuclides can be simultaneously determined in a sample and sample preparation is relatively simple.

The selected method is often based on the detection limit of the method, the amount of the sample available, the time available for analysing the sample and, sometimes, also the cost of the analysis. Gamma-ray spectrometry is often used as a preceding method for other radioanalytical methods. However, the detection limit compared to liquid scintillation counting (LSC) or alpha spectrometry is often higher. Although alpha spectrometry and LSC generally have low detection limits a considerable amount of laboratory work is necessary before the sample is in a liquid form and the radionuclides of interest are separated from other interfering nuclides. Inductively coupled plasma mass spectrometry is a superior method for long-lived radionuclides such as U-235 and U-238, but also for Ra-226 in water (Lagace et al., 2017).

In this section the analytical methods used by the CAMNAR partners are described. Other laboratories participating in the intercomparison analysed the samples mainly by using gamma-ray spectrometry and the methods used by them are not described here. One laboratory outside CAMNAR partners also performed radiochemical analyses to the water sample.

4.1 Radiation and Nuclear Safety Authority, STUK, Finland

4.1.1 Gamma-ray spectrometry

STUK has altogether 16 HPGe gamma-ray spectrometers for determining gamma-ray emitting radionuclides in environmental samples. Of these, four are electrically cooled and others are cooled by condensing liquid nitrogen cooling systems. All spectrometers have digital multichannel analysers for data acquisition.

STUK has three main measurement geometries of which two are simple cylindrical (diameters 42 mm and 74 mm, heights 0–26 mm and volumes 0–30 mL and 0–100 mL, respectively) and the third one is Marinelli (volume 0.5 L). All samples are measured on top of the detector end-cap. In the case of simple cylindrical samples the efficiency calibration is determined for the sample thickness of 0 mm. Analysis software (UniSampo-Shaman) corrects this for real sample thickness and density.

The sediment and the water samples were measured several times by using different detectors and cylindrical measurement geometries. The results presented in section 5 are from the detector (labelled as B6), which full-energy peak detection efficiency was determined with low uncertainty and from the spectrum that had the highest counting statistics.

One of the sediment and water samples were vacuum-packaged to obtain secular equilibrium between radon and its daughters in order to reliably determine the Ra-226 activity concentration.

4.1.2 Radiochemical analyses

Radon (Rn-222)

Radon is determined from untreated water. The sample is prepared by adding 10 mL of water into a glass vial (equipped with a cap containing an aluminium foil) pre-filled with liquid scintillation cocktail Ultima Gold™ XR (Packard). The sample is stirred and radon is measured in a homogeneous solution with a liquid scintillation spectrometer 1414 Guardian™ (PerkinElmer). The activity concentration of Rn-222 is calculated from the alpha spectrum in the window covering the alpha peak. The alpha counting efficiency of radon in the selected alpha window is $269\% \pm 4\%$.

Gross alpha and beta and Ra-226

The sample (38 mL) is prepared by evaporating water into dryness in a teflon-coated polyethylene vial (Zinsser). The residue is dissolved in small amount of 1M HCl and then scintillation cocktail Ultima Gold AB (PerkinElmer) is added. The sample is counted one month after the sample preparation. During that time Ra-226 attains equilibrium with Rn-222 and its short-lived daughters. Gross alpha and beta are determined with a low-background liquid scintillation spectrometer 1220 Quantulus™ (Wallac).

Uranium (U-234, U-235, U-238)

The water sample is concentrated by applying iron scavenging. The precipitate is dissolved in concentrated HCl and uranium is then separated from other radionuclides by ion exchange method by using Dowex (1x8, 50/100 mesh). Uranium is co-precipitating with CeF₃ for the alpha measurement. The sample is counted with Alpha Analyst spectrometer (Canberra). U-232 is used as a chemical yield tracer.

Polonium (Po-210)

Po-210 concentration was determined using spontaneous deposition of Po-210 on a silver disk and alpha spectrometric measurement (Alpha Analyst) of the Po-210 activity. Po-209 is used for the chemical yield tracer.

The activity concentration of Po-210 was decay corrected to the reference date and the activity concentration of Pb-210 was used to calculate the ingrowth of Po-210 from Pb-210 before the spontaneous deposition was performed and this fraction was reduced from the total Po-210 activity concentration.

Lead (Pb-210)

Lead is separated from other radionuclides using extraction chromatography. The water sample is concentrated by applying iron scavenging. Pb-210 is separated from other radionuclides by Eichrom's Sr-resin. Sample is counted with a low-background liquid scintillation spectrometer 1220 Quantulus™ (Wallac) after the 30-day period during which Bi-210 attains nearly 100% equilibrium with Pb-210. The result is calculated from Bi-210 and Pb-210 together. Inactive lead is used for the yield determination.

4.1.3 Determination of U-238 from the water sample by ICP-MS

STUKs Inductively coupled mass spectrometry device consists of quadrupole-based iCAP Q ICP-MS (Thermo Scientific) and SC-4 DX FAST (Elemental Scientific) autosampler.

Set up procedure: Before starting the routine measurements the ICP-MS will be warmed up for a minimum of 20-30 minutes. Depending on the status of the equipment mass and detector calibration will be checked using optimizing solution (Set up solution, 11 elements, Romil Ltd). Before every analysis performance test will be run with “Tune solution B for iCAP Q” (7 elements, Romil Ltd) to check maximum of ion signals and low oxide (<0.02%) and doubly charged ion rates (<0.03%).

Measurements: For calibration of the measurement three different uranium (U) concentration are used: 1, 10 and 100 µg/L (U, Romil Ltd.). For quality control (QC) of the measurements two control samples with concentrations of 5 and 50 µg/L (U, Romil Ltd.) will be included in every analysis. It is important that calibration standards and quality controls are produced from different patches of control stocks. Same concentration of nitric acid (Super Purity Acid, Romil-Spa™, 1%, v/v) should be used in samples, calibration and quality controls. Calibration solutions are prepared freshly before measurements. Every solutions to be measured by ICP-MS contain internal standard (Bi, Bismuth, Romil Ltd). The internal standard is added on-line by the autosampler in the final concentration of 2 µg/L.

The well water samples are acidified with 65% nitric acid (Super Purity Acid, Romil-Spa™) into final concentration of 1% (v/v). If estimated concentration is high the samples will be diluted with 1% nitric acid. Before analysis samples are centrifuged for ten minutes 2800 rpm to remove potential insoluble material. Calculation of the concentrations is done by the software of the ICP-MS instrument. The limit of quantification of uranium in water samples is 0.01 µg/L and measurement uncertainty is 20% (95% confidence limit).

4.2 Technical University of Denmark, DTU Nutech, Denmark

4.2.1 Gamma-ray spectrometry

For water sample, about 3L of acidified water sample was first evaporated to about 0.3 L, and transferred to a plastic standard container. The volume was measured in the container for geometry correction. The sediment sample was directly transferred to a standard plastic container, pressed using a specific metal cylinder to measure the height, which was used for geometry correction. The sample was also weighed to measure the density of the samples, which is used for the correction of self-adsorption effect. The activity concentration was calculated using the mass of the samples, considering the concentration factor for water sample.

The prepared sample was measured using HPGe detector for gamma emitters, each sample was measured for 3–5 days, the gamma spectroscopy was acquired and analysed using Genie 2000 software. The counting efficiency and sum coincidence correction were performed. The data presented are decay corrected to the reference date.

4.2.2 Radiochemical analyses

Radon (Rn-222)

Radon is determined from untreated water. Two types of sample preparation was used, directly field filling and laboratory preparation. STUK prepared one sample by filled 10 mL of water into a low diffusion plastic vial (TEFLON lined inside of vial and aluminium foil covered cap) pre-filled with 10 ml of Opti-Flour O scintillation cocktail (PerkinElmer) in the filed and sent to Risø for measurement. Another sample was prepared in Risø lab using one litre untreated water collected by STUK and sent to Risø. 10 mL of this untreated was taken to 20 mL low diffusion vial, and 10 mL of Opti-Flour scintillation cocktail was added in Risø Lab. 5-7 samples was prepared for two type of sample preparation method. The samples with scintillation cocktail was well mixed to extract radon to organic scintillation cocktail before measurement. Radon-222 with its short-lived daughter radionuclides (Po-218 and Po-214) was measured using a liquid scintillation spectrometer (Quantulus 1220, PerkinElmer) using alpha/beta discrimination function at SPA100 for 3 cycles and 30 min. of each cycle. The activity concentration of Rn-222 is calculated from the alpha spectrum in the window covering the alpha peaks of Rn-222, Po-218 and Po-214. The alpha counting efficiency of radon in the selected alpha window is $269\% \pm 5\%$.

Radium (Ra-226)

Acidified water was used for determination of Ra-226. About 300 mL of acidified water was weighed to a beaker, about 10 Bq ^{133}Ba tracer was spiked, then 15 mg barium carrier was added and well mixed. Then 3 mL of 5 mol/L H_2SO_4 was added and well mixed. The formed $\text{Ba}_2(\text{Ra})\text{SO}_4$ precipitate was separated by settling for overnight, siphoning the supernatant and centrifuge. The precipitate was washed with water to remove the acid. Finally, the precipitate was dissolved using 1 mol/L EDTA (pH8) in a water bath of 90°C . The separated Ra solution was measured using a NaI detector for ^{133}Ba to monitor the recovery of Ra in the separation. The solution was transferred to a 20 mL low diffusion plastic vial and diluted to 10 mL using water. 10 mL of Opti-Flour scintillation cocktail was added, and the vial was sealed. After 30 days ingrowth time, the activity of ingrown Rn-222 and its daughters was measured using Quantulus 1220 liquid scintillation spectrometer using alpha/beta discrimination function at SPA100 for 3 cycles and 30 min of each cycle. The activity concentration of Ra-226 is calculated from the alpha spectrum in the window covering the alpha peaks of Rn-222, Po-218 and Po-214. The alpha counting efficiency of radon and daughters in the selected alpha window is $269\% \pm 5\%$. The measured recovery was used for correction of activity.

Gross alpha and beta

Acidified water was used for determination of gross alpha and beta. 10 mL acidified water was directly taken to a low diffusion plastic vial, then 10 mL of scintillation cocktail Ultima Gold LLT (PerkinElmer) was added and well mixed. The sample was measured using Quantulus 1220 liquid scintillation spectrometer for 3 cycles and 30 min of each cycle after 3 weeks after collection.

Polonium (Po-210)

Acidified water was used for determination of Po-210. The water samples were first concentrated for 10 times by evaporation at 90 °C. 30 mL of concentrated water was transferred to a cell, Po-209 tracer was spiked, and HCl was added to a concentration of 0.5 mol/L HCl. A silver disc fixed in a special Teflon holder exposing one side of the silver disc, which was suspended in a vertical position in the solution, Polonium was spontaneously deposited on the silver disc surface at 90 °C under magnetic stirring for 3 h. The Po-210 deposited on the silver disc was measured using an alpha detector. Po-209 is used as chemical yield tracer for correction.

Lead (Pb-210)

Not determined

4.2.3 Determination of U-234, U-235 and U-238 from the water sample by ICP-MS

Uranium isotopes were measured in the acidified water sample using ICP-MS. The water samples were diluted using deionized water for 10 times and acidified with 65% nitric acid (high purity) into final concentration of 1% (v/v). U-234/U-238, U-235/U-238 ratios and U-238 concentration were measured, and In³⁺ was used as internal standard in the measurement. A U-238 standard series of 0.1–100 ppb was prepared from U-238 standard solution (IRRM-184) and used for calibration. The activity concentration of U-234 and U-235 was calculated based on the measured U-234/U-238, U-235/U-238 ratio and U-238 concentration.

4.3 Swedish Radiation Safety Authority, SSM, Sweden

4.3.1 Determination of dry content

Dry content was determined by drying an aliquot (4 g) at 80°C overnight immediately after the sample was opened. Correction for moisture was 6.3%.

4.3.2 Gamma-ray spectrometry

Two samples were prepared from the sediment sample. The first one was filled to 35 mL and the second sample to 30 mL in a cerbo 35 mL geometry. The 35 mL sample was vacuum-packed and stored for one month to obtain secular equilibrium between Ra-226 and its daughters. The sub sample of 30 mL was not vacuum-packed, since there will always be a gap between sample and lid where the radon daughters can be enriched. The water sample for gamma-ray spectrometry was prepared in a cerbo 60 mL geometry. The geometry is not radon-tight so Ra-226 was not reported. The second water sample for Rn-222 had leaked a little bit in the transport. It was poured into a radon tight 250 mL flask all to the rim.

All samples were measured on 2-4 HPGe detectors and analysis was performed using APEX/Genie 2000. The reported result is only from one of the detectors and not the mean of the measurements. The activity in the vacuum packed 35 mL sample ($d=0.48 \text{ g/cm}^3$) was determined using a 35 mL ($d=1.0 \text{ g/cm}^3$) calibration (polynomial fit). There isn't any detector

calibrated for a full geometry (35 mL) with density $d=0.50 \text{ g/cm}^3$, how come 35 mL ($d=1.0 \text{ g/cm}^3$) was used. The activity in the 30 mL sample ($d=0.36 \text{ g/cm}^3$) was determined using a 30 mL ($d=0.5 \text{ g/cm}^3$) calibration (polynomial fit). The activity of the 60 mL water sample was determined using a 60 mL ($d=1.0 \text{ g/cm}^3$) calibration (polynomial fit). All of these samples were measured on a holder 1 cm from the detector housing. The water sample for Rn-222 determination was measured directly on the detector housing and determined using a ($d=1.0 \text{ g/cm}^3$) calibration (interpolated fit).

Transmission measurements for correction of self-attenuation was done for Pb-210, and since the correction factor was found to be 1.0 transmission measurements was not applied for the Am-241 correction. Data for half-lives (Table 2) and emission probabilities were taken from ENDF/B-VII.1.

Table 2. The half lives and energies used for the activity calculation of each nuclide and calibration used for the determination of activity in the sediment sample.

Nuclide	Energy (keV)	Half life	Calibration
K-40	1460	$1.25 \cdot 10^9 \text{ y}$	30 mL ($d=0.5 \text{ g/cm}^3$)
Cs-137	661	30 y	30 mL ($d=0.5 \text{ g/cm}^3$)
Pb-210	46	1600 y	35 mL ($d=1.0 \text{ g/cm}^3$) + corr. transmission
Ra-226	295, 352, 1764	1600 y	35 mL ($d=1.0 \text{ g/cm}^3$)
Ra-228	338, 911, 969	5.75 y	30 mL ($d=0.5 \text{ g/cm}^3$)
Th-228	238, 583	5.75 y	30 mL ($d=0.5 \text{ g/cm}^3$)
U-235	185	$7.04 \cdot 10^8 \text{ y}$	35 mL ($d=1.0 \text{ g/cm}^3$)*
U-238 (Th-234)	63	$4.47 \cdot 10^9 \text{ y}$	30 mL ($d=0.5 \text{ g/cm}^3$)
U-238 (Pa-234m)	1001	$4.47 \cdot 10^9 \text{ y}$	30 mL ($d=0.5 \text{ g/cm}^3$)
Am-241	59	432 y	35 mL ($d=1.0 \text{ g/cm}^3$)*
Rn-222	609,1120,1765,768,1238, ...	3.82 d	250 mL ($d=1.0 \text{ g/cm}^3$) calibration, interpolated fit

* The nuclide was only detected with a detector that doesn't have a calibration for $d=0.5 \text{ g/cm}^3$

Since Pb-210 wasn't in secular equilibrium with Ra-226 corrections to the reference date was calculated after measurement. An assumption was made that Ra-226 decays to Pb-210 without loss of Rn-222 from the sample. The activity was calculated using this formula

$$A_{Pb210,ref} = A_{Pb210} \cdot e^{-\lambda_{Pb210} \cdot t} + \frac{A_{Ra226} \cdot \lambda_{Pb210}}{\lambda_{Pb210} - \lambda_{Ra226}} \cdot (e^{-\lambda_{Pb210} \cdot t} - e^{-\lambda_{Ra226} \cdot t})$$

$A_{Pb210,ref}$ = Activity concentration of Pb-210 at the reference date [Bq/kg]

A_{Pb210} = Activity concentration at the measurement date [Bq/kg]

λ_{Pb210} = Decay constant for Pb-210 [1/s]

λ_{Ra226} = Decay constant for Ra-226 [1/s]

t = time between the reference date and the measurement date

A_{Ra226} = Activity concentration of Ra-226 [Bq/kg]

Activity of Ra-226 was determined with LSC for the water sample and with gamma-ray spectrometry for the sediment sample.

4.3.3 Radiochemical analyses

Radon (^{222}Rn) in water

Two different aliquots of the water was measured. One sample was prepared by STUK according to the SSM procedures and another sample was prepared from the same water that was used for gamma-ray measurements. The second water sample was prepared by adding 10 ml water to a plastic vial (equipped with a cap containing an aluminum foil) pre-filled with 12 ml liquid scintillation cocktail Ultima Gold™ XR (Perkin Elmer). Sample was shaken and radon was measured in a homogeneous solution with a liquid scintillation spectrometer Quantulus 1220 (Wallac). The activity concentration of ^{222}Rn is calculated from the alpha spectrum in the window covering the most part of the alpha peak. The alpha counting efficiency of radon in the selected alpha window was 3,15 cps/Bq (unc. $\pm 3,1\%$ $k=2$)

Gross alpha, beta and Ra-226 analysis in water

A water sample of 38 ml was prepared by evaporating water into dryness in a teflon-coated polyethylene vial (Zinsser). The residue was dissolved in 1 ml 0.1M HCl and then 20,5 ml OptiPhase HiSafe 3 (Perkin Elmer) was added. The sample was counted one month after the sample preparation. During that time ^{226}Ra reached secular equilibrium with ^{222}Rn and its short-lived daughters. Gross alpha and beta was determined with a low-background liquid scintillation spectrometer 1220 Quantulus™ (Wallac). The gross alpha activity was calculated by withdrawing the activity from the short lived daughters Po-218, Rn-22 and Po-214. The activity of those are determined from the ROI for Pb-214 (~3,0* ROI for Pb-214). Like-wise the gross beta activity was calculated by withdrawing the activity for Bi-214 and Pb-214 (~2,4* ROI for Pb-210).

Uranium (^{234}U , ^{235}U , ^{238}U)

The water sample is concentrated by applying iron hydroxide precipitation. The precipitate was dissolved in 8 M HNO₃ and uranium was then separated from other radionuclides by liquid-liquid extraction with TBP. Uranium was electro-plated on stainless steel for the alpha measurement. The sample was counted with AlfaAnalyst spectrometer (Canberra). ^{232}U is used as a chemical yield tracer.

Polonium (^{210}Po)

^{210}Po concentration was determined using spontaneous deposition of ^{210}Po on a copper disk and alpha spectrometric measurement (AlphaAnalyst) of the ^{210}Po activity. Before the deposition the water sample is concentrated with evaporation. ^{209}Po is used for the chemical yield tracer.

4.4 Norwegian Radiation Protection Authority, NRPA, Norway

4.4.1 Gamma-ray spectrometry

Two sediment samples from STUK were sent to the NRPA laboratories in Tromsø and Svanhovd for gamma-spectrometry measurements. Samples were treated routinely, according to the SOPs used at NRPA.

Sediment samples were dried in the oven at 105 °C for 24 h (to determine the water %), then homogenized, placed at the W2 geometry (fill height 0–26 mm, volume 0–104 mL) and vacuum packed for 3 weeks to obtain secular equilibrium between radon and daughters.

For measurement, both laboratories used HPGe detectors (electrically cooled) from Canberra. The spectra were analysed by an in-house software Spec10.

4.4.2 Radiochemical analyses, LSC and alpha spectrometry

Radon (Rn-222)

Radon was determined at the NRPA laboratory in Østerås, in two ways:

1. From an untreated water sent by STUK in 1 L glass bottle: using 10 mL syringes, three water samples were extracted from the bottle; each sample was then analysed 3 times. First, on the reception date (27 April 2017), and then twice - the day after (28 April 2017). The middle value of the samples' analyses results was reported.
2. From a sample glass with a scintillation fluid sent by NRPA to STUK in order to collect the sample according to the NRPA methods and with the NRPA equipment: the sample was received at NRPA on 2nd of May 2017 and was analysed the same day. As only one sampling glass was sent, so only one sample was analysed.

Measurements of radon in the water samples mentioned above were performed with the WALLAC 1211 RACBETA liquid scintillation counter.

Uranium (U-234, U-235, U-238)

Uranium isotopes were analysed by alpha spectrometry after radiochemical separation. The method used is similar to the method described by Martin and Hancock, 2004. Uranium was initially co-precipitated with Fe(OH)₃. The precipitate was then dissolved in 8 M HNO₃ and extracted into TBP (tri-butyl phosphate) together with thorium. After dilution of TBP with xylene, thorium was removed by repeated washing with 1.5 M HCl. Uranium was finally back extracted with distilled water. After electrodeposition (Hallstadius, 1984) the sample was analysed by alpha spectrometry (Canberra Alpha Analyst and Genie-2000 software). U-232 was used as a yield determinant.

Polonium (Po-210) and Lead (Pb-210)

Polonium was analysed by alpha spectrometry (Canberra Alpha Analyst and Genie-2000 software) after it was spontaneously deposited on to silver discs. The method used is a slightly modified method described by Flynn (1968). Initially, the polonium was co-precipitated with MnO₂. The precipitate was then dissolved in diluted HCl and H₂O₂ under heating. Finally, polonium was auto-deposited on to polished silver discs from 5 M HCl at 65 °C for 3 hours. A small amount of ascorbic acid was also added to the sample. Po-209 was used as a yield determinant.

After auto-deposition, residual polonium was removed by ion exchange. New Po-209 tracer was added and the sample was stored for 6 months before it was analysed again.

4.5.1 Swedish Defence Research Agency, FOI, Sweden

4.5.1 Gamma-ray spectrometry

At FOI all detectors have semi-empirical calibrations that are free from systematic effects like true coincidence summing (TCS). Corrections for TCS and attenuation of the sample is calculated by using GESPECOR.

The sediment and the water samples were measured before and after vacuum packaging. After three weeks secular equilibrium is obtained between radon and its daughters and the Ra-226 activity can be determined. Other radionuclides were determined from the measurement of the sample geometries that were not vacuum-packaged.

For the determination of Pb-210 a broad-energy detector was used in an ultra-low background lead shield. Pb-210 was determined by the Cutshall method. There is an on-going work regarding the validation of an interpolative calibration.

4.5.2 Gross alpha and beta

Three subsamples of 40 mL each were prepared by evaporating water into dryness in a beaker. The residue was dissolved in a small amount of 1M HCl and then scintillation cocktail Ultima Gold AB (PerkinElmer) was added to the top of the scintillation vial to limit the void for the Rn-222, if present. The samples were then left to stand for three weeks prior to measurement. Gross alpha and beta are determined with a scintillation spectrometer 1220 QuantulusTM (Wallac).

4.5.3 Uranium (U-234, U-235, U-238) alpha spectrometry and ICP-MS

For alpha spectrometry the water sample was evaporated into dryness in a beaker and transferred into 3 M HNO₃. The samples were loaded to and separated from other radionuclides by using UTEVA resins and were counted with an Octète PLUS Alpha Spectroscopy workstation (OrtecAlfa). U-232 is used as a chemical yield tracer for alpha spectrometry whereas U-233 is used as a chemical yield tracer for ICP-MS measurements.

For ICP-MS, the water sample was diluted 100 times for the U-234 determination. For the U-238 and U-235 determination the sample was diluted 10000 times. The dilutions were done with 0,28M HNO₃. The sample was spiked with IRMM-040a (U-233) to an appropriate concentration. The samples were measured using an Element XR (Thermo Scientific, Bremen, Germany). The contributions from the spike solution was taken into account when calculating the uranium concentrations of U-234, U-235 and U-238.

4.5.4 Polonium (Po-210)

Po-210 concentration was determined using spontaneous deposition of Po-210 on a silver disk and alpha spectrometric measurement of the Po-210 activity. Po-209 is used for the chemical yield tracer. The samples were counted with an Octète PLUS Alpha Spectroscopy workstation (Ortec).

5. Results

In the following, the nuclide-specific results are presented separately for the sediment and the water samples. In all cases the combined uncertainties refer to the coverage factor $k = 1$.

Outliers of the data shown as red points in the set of the following figures were removed from the average values shown as a solid line. In the calculation of the average values, the data point is considered here as an outlier if the absolute value of the standard score $|z| > 2$. The standard score $z = (x - \mu)/\sigma$, where x is the value of the data point and μ and σ are the mean and standard deviation of the measured values.

5.1. Results of the sediment sample by using gamma-ray spectrometry

Intercomparisons are often organized in such a way that Cs-137 is present in the source material. Other radionuclides are not so often reported, which is also visible in the following set of figures (Figs. 5 and 6). Subsequently, the dispersion of the results is larger for other nuclides than in the case of Cs-137.

The half-life of U-238 was used for Th-234 and Pa-234m. Average values of the activity concentrations of Th-234 and Pa-234m were 269 ± 33 Bq/kg and 285 ± 35 Bq/kg, respectively. The average value of U-235 was 13.9 ± 2.8 Bq/kg, which gives the value of 19.9 ± 4.8 for the activity ratio U-238/U-235. The respective value for natural U is 21.5.

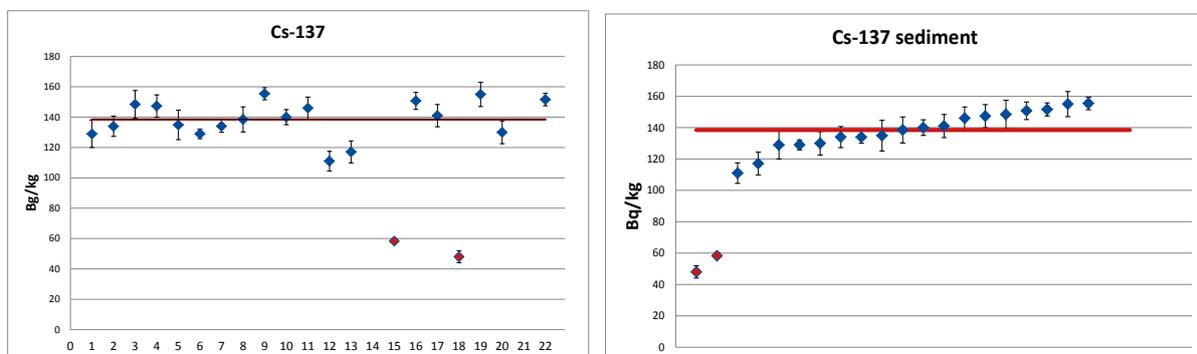


Figure 5. Cs-137 activity concentrations in the sediment sample reported by each laboratory (laboratory code is in the horizontal axis). The values in the leftmost figure are organized according to the individual laboratory codes of the participants whereas values in the right are organized according to the order of magnitude. Outliers denoted as red points are not taken into account in the average value (solid horizontal line).

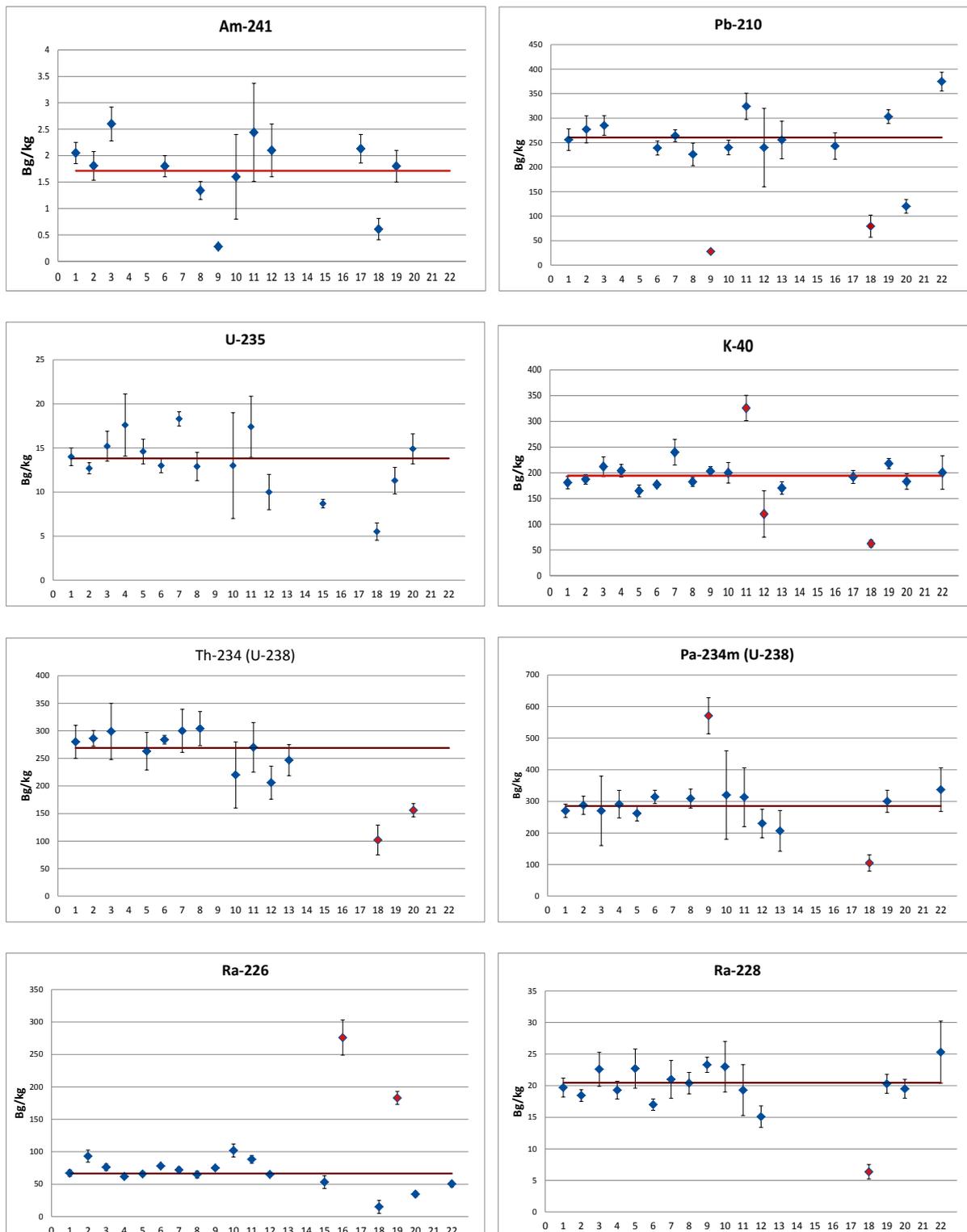


Figure 6. Activity concentrations of different radionuclides in the sediment sample reported by each laboratory (laboratory code is in the horizontal axis). Horizontal lines refer to the average values from which outliers (denoted as red points) are removed.

5.2. Results of the water sample by using gamma-ray spectrometry

Analysis results from water using gamma-ray spectrometry revealed considerable variation compared to those obtained from the sediment material (Fig. 7). The reason is unclear and could be investigated in a future project and intercomparison. As a comparison, average values obtained from alpha spectrometry and mass spectrometry are inserted in some of the figures.

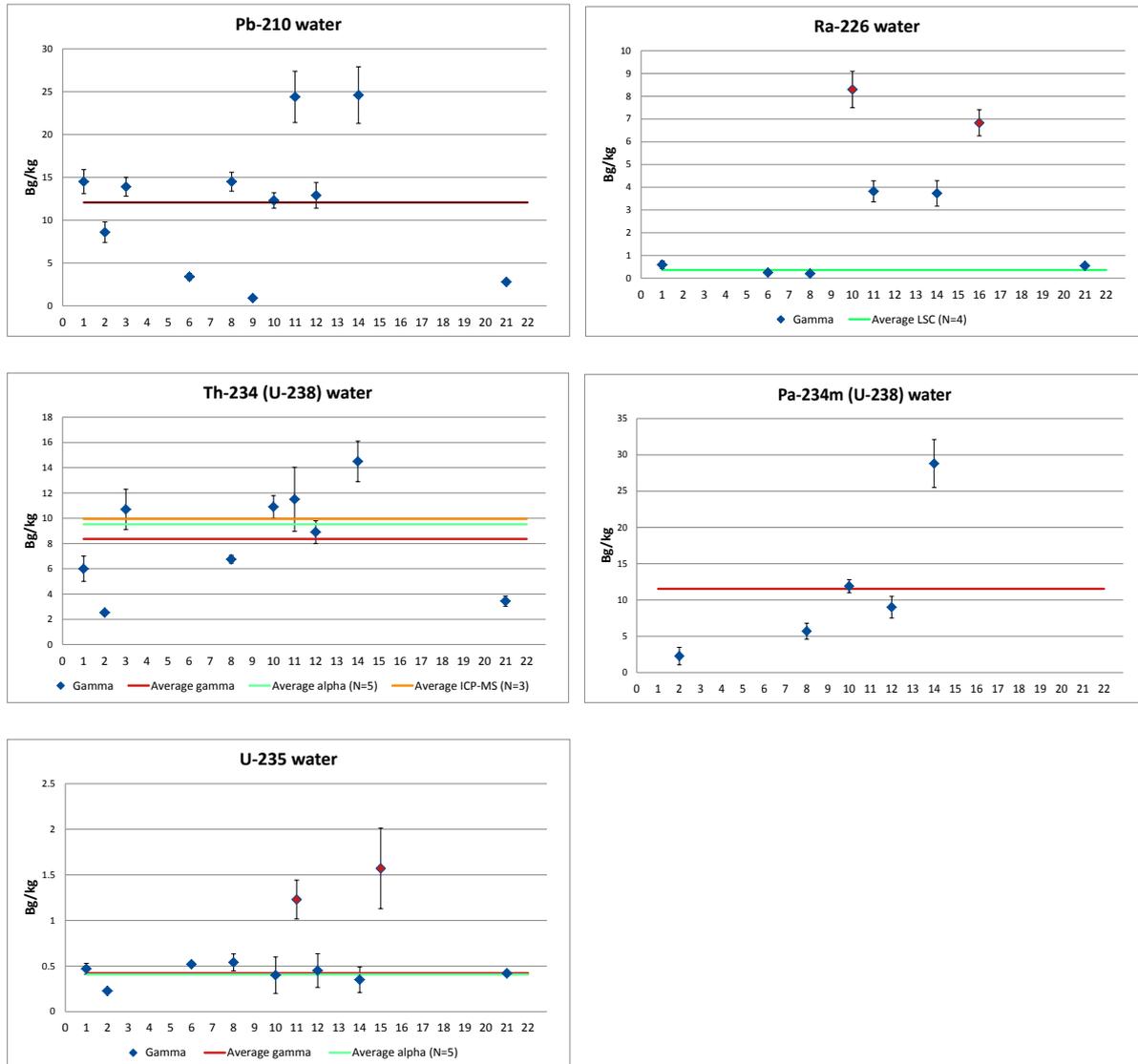


Figure 7. Activity concentrations of different radionuclides in water reported by each laboratory (laboratory code is in the horizontal axis). Horizontal lines refer to the average values from which outliers (denoted as red points) are removed. Average values from alpha spectrometry and mass spectrometry (N is the number of results from which the average value was calculated) are presented in some of the figures as a comparison.

5.3. Results of the water sample by using radiochemical means

In addition to gamma-ray spectrometry, the water samples were also analysed by alpha spectrometry, liquid scintillation counting and inductively coupled plasma mass spectrometry. Reported values are those reported by the participants, and are not rounded for the report.

Isotopes of U were determined by ICP-MS in three laboratories (Table 3) and with alpha spectrometry in five laboratories (Table 4). The results of U-238 and U-235 determined by ICP-MS and alpha spectrometry are compatible and agrees well with those obtained by gamma-ray spectrometry (Table 5).

Table 3. Activity concentration of U-238, U-235 and U-234 reported by different laboratories and determined by ICP-MS. Uncertainties refer to the coverage factor $k = 1$.

Laboratory number	U-238 ($\mu\text{g/L}$)	U-235 ($\mu\text{g/L}$)	U-234 ($\mu\text{g/L}$)
Laboratory 1	760 ± 75	-	-
Laboratory 2	793.4 ± 55.5	-	-
Laboratory 6	759.3 ± 8.7	5.533 ± 0.088	0.0732 ± 0.0014

Table 4. Activity concentration of U-238, U-235 and U-234 reported by different laboratories and determined by alpha spectrometry. Uncertainties refer to the coverage factor $k = 1$.

Laboratory number	U-238	U-235	U-234
Laboratory 1	9.5 ± 0.68	16 ± 1.1	0.37 ± 0.03
Laboratory 2	9.87 ± 0.69	16.86 ± 1.22	0.45 ± 0.03
Laboratory 3	9.59 ± 0.16	16.6 ± 0.28	0.4538 ± 0.0097
Laboratory 4	9.5 ± 0.4	16.7 ± 0.6	-
Laboratory 12	10.4 ± 0.6	17.6 ± 1	0.44 ± 0.07

Table 5. Comparison of the average values and respective standard deviation, σ , of the activity concentrations of U-238, U-235 and U-234 between different analysis methods. In the case of U-238 determined by gamma-ray spectrometry only the results obtained by Th-234 are taken into account in the average value. This was because of the notable variation on the results obtained by using the 1001 keV peak of Pa-234m. In the case of ICP-MS the reported average value was converted from $\mu\text{g/L}$ to Bq/L.

Analysis method	U-238	U-235	U-234
Alpha spectrometry (Bq/L) (N=4)	9.5 ± 0.3	0.41 ± 0.06	16.5 ± 0.3
Gamma-ray spectrometry (Bq/L) (N=11)	8.4 ± 4.0	0.42 ± 0.10	-
ICP-MS (Bq/L) (N=3)	9.6 ± 0.2	-	-

The results of gross α and gross β determination and Ra-226 activity concentration in water were determined by liquid scintillation counting (Table 6). In general, variation of the results is large especially in the case of gross β determination. The reason is unknown and should be investigated in a future intercomparison; standardization and calibration might be reasons for the variation.

Ra-226 was also determined by gamma-ray spectrometry (Fig. 7) but the results varied a lot. The gamma-ray results from laboratories 1, 6, 8 and 21 were in general compatible with those obtained by LSC.

Table 6. The results of Gross α , Gross β and Ra-226 determination in the water sample reported by different laboratories. Uncertainties refer to the coverage factor $k = 1$.

Laboratory number	Gross α (Bq/L)	Gross β (Bq/L)	Ra-226 (Bq/L)
Laboratory 1	28 ± 2	15 ± 2	0.28 ± 0.04
Laboratory 2	42.05 ± 6.69	65.42 ± 19.63	0.525 ± 0.061
Laboratory 3	36.8 ± 1.9	43.4 ± 2.5	0.377 ± 0.032
Laboratory 6	26 ± 0.5	28 ± 0.6	-
Laboratory 12	28.95 ± 0.995	31.68 ± 0.705	0.277 ± 0.039

Rn-222 results (Table 7) varied between 4130 Bq/L – 5180 Bq/L (prefilled vials) and results were generally lower for samples taken from glass bottle, varying between 4360 Bq/L – 4960 Bq/L. This might be caused by the evaporation of Rn from the sample during shipping and

handling. Also not all laboratories have practices for handling samples that are not taken directly from the source to the sample vials.

Table 7. The results of the Rn-222 concentration measurements in water using liquid scintillation counting (LSC) and gamma-ray spectrometry reported by different laboratories. Uncertainties refer to the coverage factor $k = 1$.

Laboratory number	LSC, prefilled vials Rn-222 (Bq/L)	LSC, glass bottle Rn-222 (Bq/L)	Gamma Spectr. Rn-222 (Bq/L)
Laboratory 1	5070 ± 260	4960 ± 260	-
Laboratory 2	4126.3 ± 152.8	4380.3 ± 151.6	-
Laboratory 3	4590 ± 120	4470 ± 110	4720 ± 260
Laboratory 4	5184 ± 1036.8	4364 ± 872.8	-

Due to high variation in Po-210 and the lack of more than two Pb-210 results, the results of these radionuclides are left out from the report and are not discussed here. Explanation for the large variations in the Po-210 results could be the use of different approaches for the decay correction of the data. In future intercomparison exercises it is essential to instruct participants to use the same reference dates and decay corrections.

6. Conclusions

In the CAMNAR project, lake sediment and water samples were analysed in different Nordic and European laboratories using a set of analytical methods. The aim of the CAMNAR project was to improve the quality and capabilities of the participating laboratories by conducting an extensive interlaboratory exercise on the analytical methods used to determine several radionuclides present in the samples. The techniques used in the intercomparison are gamma-ray spectrometry, alpha spectrometry, liquid scintillation counting and inductively coupled plasma mass spectrometry.

CAMNAR revealed that intercomparisons are essential for improving analytical capabilities of the laboratories. The laboratories taking part in the intercomparison are given the chance to evaluate their achievement and identifying improvements that can improve their quality and capability for future analyses. The strength of this intercomparison was the possibility to compare the results obtained from different analytical methods.

Gamma-ray spectrometry is a widely used technique to determine gamma-ray emitting radionuclides in the samples. However, present intercomparison revealed extraordinary variation between the results reported by different laboratories. Especially the results of the water sample are of concern and should be carefully investigated in a future intercomparison.

In general, alpha spectrometry, gamma-ray spectrometry and mass spectrometry gave consistent results for the radionuclides U-235 and U-238 in the water despite the large variation between gamma-ray spectrometry results.

The results of gross α and gross β determination in water by using liquid scintillation counting also revealed considerable variation, and this should be examined more closely by organizing a new intercomparison exercise concentrating on the analysis of these two parameters. Also, the analysis of Po-210 needs to be examined in future intercomparisons.

Organizing an intercomparison exercise for the analysis of Rn-222 in natural water is generally a difficult task. There are always considerable uncertainties in the whole process, e.g. sample collection, shipping and handling of the samples. The results show variations between the sample types (prefilled vial and sample in glass bottle) and because of the low number of participants in this exercise, no general conclusions can be drawn from the results.

Based on the outcome of the NKS CAMNAR project, we would like to underline that arranging comprehensive intercomparisons involving different analytical methods is important in the future in order to improve capabilities and quality of the analyses of different laboratories. Especially, exercises for determining artificial and naturally occurring radionuclides in natural water is recommended.

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Title	Comparison of the analytical methods used to determine natural and artificial radionuclides from environmental samples by gamma, alpha and beta spectrometry
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Abstract	In CAMNAR, an extensive interlaboratory exercise on the analytical methods used to determine several radionuclides present in the environmental samples was organized. Activity concentration of different natural radionuclides, such as Rn-222, Pb-210, Po-210, K-40, Ra-226, Ra-228 and isotopes of uranium, in addition to artificial Cs-137 and Am-241 were analysed from lake sediment samples and drinking water. The measurement techniques were gamma-ray spectrometry, alpha spectrometry, liquid scintillation counting and inductively coupled plasma mass spectrometry. Twenty six laboratories from nine Nordic and European countries participated in the intercomparison. Extraordinary variation between the results reported by different laboratories were revealed for some radionuclides indicating the need of future intercomparisons especially in the case of natural water samples.
Key words	Intercomparison, gamma-ray spectrometry, alpha spectrometry, liquid scintillation counting, inductively coupled plasma mass spectrometry