Nordisk kernesikkerhedsforskning
Norrænar kjarnöryggis rannsóknir
Pohjoismainen ydinturvallisuustutkimus
Nordisk kjernesikkerhetsforskning
Nordisk kärnsäkerhetsforskning
Nordic nuclear safety research

NKS-B

The NKS-B RADSEM mini-seminar 2004

Risø, 19 August 2004



Programme

1. Introduction (practical arrangements, structure of NKS, call for proposals 2005), Sigurður Emil Pálsson

2. **RADCHEM**, Rajdeep Singh Sidhu
   Extended abstract: RADCHEM – Summary of a questionnaire on the determination of Sr, U, Pu and Am(Cm) (9 pages)

3. **NORCMASS**, Per Roos
   Extended abstract: Isotope ratio measurements with single collector ICP-MS (4 pages)

4. **LABINCO** - An intercomparison exercise on laboratory analyses of radionuclides in food and environmental samples, Sven P. Nielsen

Extended abstract: Intercomparison of laboratory analyses of radionuclides in environmental samples, 2004 (1 page)

5. **IRADES**, Tua Rahola / Maarit Muikku
   Extended abstract: IRADES - Assessment of Internal Doses in Emergency Situations (5 pages)

6. **CSKINETIC**, Christopher L. Rääf
   Extended abstract: Human Metabolism of Caesium (3 pages)

7. **REIN**, Lavrans Skuterud
   Extended abstract: REIN: Regional differences in reindeer radiocaesium contamination (4 pages)

8. **ECODOSES** - results and future plans, Astrid Liland

9. **INDOFERN**, Erkki Ilus
   Extended abstract: Status Report of the INDOFERN Project *(New indicator organisms for environmental radioactivity)* in August 2004 (11 pages)

10. Discussion (ideas for future work, status seminar 2005)

Eight NKS-B projects were presented, seven of these subsequently sent in an extended abstract. Links to each of these abstracts are given in the table above.

RADCHEM – Summary of a questionnaire on the determination of Sr, U, Pu and Am(Cm).

Sidhu, R.S., Holm, E., Roos, P., Lehto, J., Ikäheimonen, T.K. and Ramebäck, H.

1. General information

As part of the NKS-B project on radiochemical analysis, RADCHEM, a questionnaire on the determination of Sr, U, Pu and Am(Cm) in various samples was prepared and distributed to sixteen Nordic labs. All aspects from sample gathering to activity determination were highlighted in the questionnaire. Fourteen labs responded to the invitation and nine labs answered the questionnaire. Four labs do not perform the specified analysis. This report presents a preliminary summary of the response.

2. Sample

Collected according to laboratory quality manuals or received from contractors or research institutes. All samples, accept milk, analysed for actinides and Sr. Milk analysed only for Sr.

Water	Precipitation, effluent water, lake water, seawater collected by e.g. pumping in appropriate containers
Soil	Grab or corer: slicing or div. into e.g. litter, organic and mineral fraction
Sediment	Grab or corer (slicing)
Biota	Grass, peat: collection from restricted area (0.25-0.5 m^2), agricultural products and fish etc.
Filter/Swipe	Organic filters or swipes
Milk	Collection from farms
Urine	24 hour sample
Reactor fuel	
Nuclear waste	

3. Pre-treatment

Water	Filtration, acidification (pH1-2), tracer/carrier addition, redox treatment
Soil	Removal of stones, drying (60-105 ^oC) or freeze drying, sieving (2mm), tracer addition
Sediment	Removal of stones, drying (60-105 ^oC) or freeze drying, sieving (2mm), tracer addition
Biota	Carrier/tracer addition, drying (60-105 ^oC), preservation with $Na_2S_2O_5$, grinding, sieving (2mm)
Filter/Swipe	Tracer/carrier addition
Milk	Tracer/carrier addition
Urine	Acidification, tracer addition, heating

4. Enrichment

Most labs add tracer after dry ashing, but all add before wet dissolution/leaching

Water	Small low salt content waters: Evaporation Actinides: $Fe(OH)_3$ precip. using NaOH or NH_3, MnO_2 and NdF_3 precip. for uranium in small water samples Sr: carbonate or oxalate precip. of supernatant from actinide precip.
Soil	Dry ashing (400-600 ^oC) (one lab does not ash soil samples)
Sediment	Dry ashing (400-600 ^oC with the addition of oxalic acid) (one lab does not ash sediment samples)
Biota	Dry ashing (400-600 ^oC), Seeweed: 700 ^oC
Filter/Swipe	
Milk	Evaporation
Urine	Actinides and Sr: phosphate precipitation

Are samples covered during dry ashing?

5. Dissolution/leaching

Water	Hydroxides or evap.: HNO_3 or HCl depending on separation scheme MnO_2: HCl + H_2O_2 NdF_3: HNO_3 + HCl
Soil/sediment, biota	Aqua regia (plus H_2O_2, HF) followed by hydroxide precipitation of actinides and carbonate precipitation of Sr. Oxalate precipitation of actinides and Sr or just evaporation to dryness. Li-metaborate fusion Microwave oven treatment with HNO_3, HF, HCl
Filter/Swipe	Leaching w HNO_3 or aqua regia (plus H_2O_2), fusion, microwave oven treatment
Milk	Dissolution in HNO_3, HCl
Urine	Phosphate destruction using dry ashing, dissolution in HNO_3

6. Strontium determination

6.1. Separation

Fuming nitric acid precipitations (2 labs)

- Nitrate precipitation (Sr sep. from Ca)
- Hydroxide precipitation (Sr sep. from actinides and yttrium)
- Chromate precipitation (Sr separation from Ba, Ra and Pb)
- Carbonate precipitation

Used on all kinds of environmental samples.

Solvent extraction (1 lab, used only on milk samples)

- Extraction of Y-90 with TBP from 14 M HNO_3
- Elution of Y-90 with H_2O

Extraction chromatography (3 labs)

- Sr-Resin (0.7 grams vs. 3.0 grams)
- Load: 8 M HNO_3
  3 M HNO_3 – 0.1 M sulphamic acid – 0.1 M ascorbic acid – 0.3 M $Al(NO_3)_3$
  (sequential analysis using TRU- and Sr-Resin)
- Wash: 8 M HNO_3
- Wash: 3 M HNO_3
- Wash: 3 M HNO_3 – 0.05 M oxalic acid
- Sr elution: 0.05 M HNO_3

Two labs only use Sr-Resin for Sr-separations, while one lab uses both the traditional method and Sr-Resin.

6.2. Source preparation

Precipitation: Precipitation of Sr as carbonate (determination of ^{89}Sr, ^{90}Sr)
Addition of Y-carrier and ingrowth of ^{90}Y
Hydroxide precipitation (separation from Sr)
Oxalate precipitation and filtration (determination of ^{90}Y)

6.3. Activity determination

^{89}Sr and ^{90}Sr: Proportional counter
LSC: Cerenkov (^{89}Sr) and LSC (^{90}Sr)
Sr-yield determination using AAS on stable Sr
Y-yield determination: EDTA titration

^{90}Sr: Proportional counter
GM-counter
LSC
Sr-yield determination using either ^{85}Sr or AAS on stable Sr
Y-yield determination: EDTA ($ZnSO_4$) titration

Two labs have the possibility to analyse Sr directly using LSC. These labs use Sr-Resin and AAS for Sr-yield determination. Two labs count the ^{90}Y source after ingrowth. These labs use ^{85}Sr as yield determinant.

7. Uranium determination

7.1. Separation

Anion exchange: Resin: Dowex 1x4 or 1x8
Column dimension: 1x12 cm
Flow: Approx. 1 ml/min
Load: 9-12 M HCl
Th removal: 9-12 M HCl
Elute U: 0.1 M HCl

(One lab uses 9 M HCl load for rock samples and 12 M HCl load for water samples)

Resin: BIORAD AG-1x4 or Dowex 1x4
Column dimension: 1x12 cm
Flow: Approx. 1 ml/min
Load: 8 M HNO_3
Wash: 8 M HNO_3
Elute U: 0.1 M HCl

(One lab uses this procedure when the sample contains elevated levels of Fe)

Two labs use anion exchange for natural water, soil and sediment samples and UTEVA-Resin for filters and precipitation.

Solvent extraction: Water phase: 8 M HNO_3
Organic phase: 100% TBP
Wash: 8 MHNO_3 (repeated)
Dilute org. phase with xylene
Wash with 1.5 M HCl (Th) (repeated)
Elute U with H_2O (repeated)

Three labs use TBP extraction for uranium separation.

Extraction chromatography: Column: UTEVA-Resin (0.7 gram)
Load: 3 M HNO_3 – (0.5M) 1 M $Al(NO_3)_3$ – Fe-sulphamate – ascorbic acid
Wash: 9 M HCl
Wash: 5 M HCl – 0.05 M oxalic acid
U elution: 0.01 M HCl

Three labs use this procedure. In one lab the procedure is combined with TRU-Resin for Pu-determination, and in an other it is combined with both Sr-, TRU- and TEVA-Resin.

7.2. Source preparation

Electrodeposiiton: Catode: polised stainless steel (diameter 20 mm)
Anode: platinum
Distance: 5 (10) mm
Solution: 8-15 ml 2 M $(NH_4)_2SO_4$ or Na_2SO_4 plus H_2SO_4 ($NaHSO_4$),
pH = 2 (2.5) (slightly acidic)
0.75, 1.0, 1.8 ampere for 0.75, 1.5, 2, 4 hours

5 labs use only electrodeposition, while one lab uses both electrodeposition and micro co-precipitation.

Micro co-precipitation: Solution: 10 ml 4 M HCl, 4 ml 1 M HCl
50 µg Ce (Nd)
$TiCl_3$ (for U reduction)
0.6 (2) ml 40% HF
Filtration through 0.1 µm polypropylene filter (pre-treated with CeF_3 or NdF_3 substrate solution)

One lab uses only micro co-precipitation while another lab uses both this and electrodeposition.

7.3. Activity determination

Alpha-spectrometry using PIPS detectors (450 mm^2)
ICP-MS
Yield determination using ^{232}U (^{233}U)
Fluorescence analysis after fusion with NaF.

8. Plutonium determination

8.1. Separation

Anion exchange: Resin: BIORAD AG-1x4, 1x8 or Dowex 1x4
Column dimension: 1x12 cm, two labs uses 1x3-5 cm column
Flow: Approx. 1-2 ml/min
Load: 8 M HNO_3 added $NaNO_2$ (save for Am and Cm analysis)
Wash: 8 M HNO_3 (save for Am and Cm analysis)
Elute Th: 9-12 M HCl
Pu: 9 M HCl – NH_4I or 1.2 M HCl + H_2O_2 (two labs)

This method is used in 7 out of 8 labs that perform Pu-analysis. For determination of Pu in large samples, anion exchange is either repeated (one lab then uses anion exchange from HCl media for complete separation from Th) and/or combined with solvent extraction with TBP, TIOA or TTA, prior or after anion exchange. Most labs have good Pu-recoveries using anion exchange.

Solvent extraction: Liquid phase: 1.4 M HNO_3 added $NaNO_2$
Organic phase: 10% TIOA or 20% TBP in xylene
Pu elution: 1.2 M HCl containing 30% H_2O_2

Liquid phase: 1 M HNO_3
Organic phase: 0.5 M TTA (xylene or freon)
Pu elution: 8 M HNO_3

One lab has poor Pu-recoveries (less than 50%) when using TTA extraction.

Extraction Chrom.: TRU-Resin (0.7 gram)
Column dimension: 8 mm diameter
Flow: 1-2 ml/min
Load: 2 – 3 M HNO_3 added $Al(NO_3)_3$, sulphamic and ascorbic acid
Wash: 2 M HNO_3 added $NaNO_2$
Wash: 0.5 M HNO_3 (one lab does this)
Wash: 9 M HCl
Am: 4 M HCl
Wash: 4 M HCl + 0.1 M HF (one lab does this)
Pu: 4 M HCl added $TiCl_3$

The resin is used in 4 of 6 labs that perform Pu-analysis and is also combined with either UTEVA or Sr-Resin for uranium and Sr determinations, respectively.

8.2. Source preparation (alpha spectrometry)

As for uranium.

8.3. Activity determination

Alpha-emitters: Alpha-spectrometry using PIPS detectors (450 mm^2)
ICP-MS
Yield determination using ^{242}Pu

^{241}Pu: LSC on treated membrane filters after alpha-spectrometry
LSC on part of the Pu-solution after separation
Ingrowth of ^{241}Am followed by separation and alpha-spectrometry
Yield determination using ^{242}Pu

9. Americium and curium determination

9.1. Separation

Anion exchange (after Pu-separation using anion exchange)

Oxalate precipitation (pH 1-1.5, 4), dry ashing (450-600 ^{o}C), followed by $Fe(OH)_3$ precip. and/or anion exchange and/or liquid liquid extraction (TIOA) to remove traces of interfering actinides before final separation. One lab purifies the Am fraction further using HDEHP-TBP extraction followed by mixed cation-anion exchange before final purification.

Final purification:

Resin: BIORAD AG-1x4 or Dowex 1x4
Column dimension: 1x12 cm, one lab uses 1x3 cm column
Flow: Approx. 1 ml/min
Load: 1 M HNO_3 – 86% (93%) CH_3OH
Wash: 1 M HNO_3 – 86% (93%) CH_3OH
Lanthanide removal: 0.1 M HCl - 80% (75%) CH_3OH - 0.5M (1M) NH_4SCN
Elute Am/Cm: 1.5 M HCl – 86%CH_3OH

This procedure is used by three labs.

Extraction chromatography

If the sample does not contain interfering amounts of lanthanides, the Am fraction from TRU-Resin is pure enough for Am-determination. The interfering amount depends on follwing source preparation and activity determination procedure.

Three labs uses TRU-Resin followed by TEVA-Resin for the separation of Am and Cm from effluents from Pu-separation using anion exchange, or U-separation using UTEVA-Resin :

- Oxalate precipitation (pH 1.5)
- Ash at 600 ^{o}C
- 3M HNO_3 – 1M $Al(NO_3)_3$ – Fe-sulphamate – ascorbic acid
- TRU-Resin
    - Wash with 2 M HNO_3
    - Wash with 2 M HNO_3 – 0.1 M $NaNO_2$
    - Wash with 0.5 M HNO_3
    - Am and Cm elution with 4 M HCl
- Evaporate and dissolve in 2 M NH_2SCN – 0.1 M formic acid
- TEVA-Resin
    - Wash with 1 M NH_2SCN – 0.1 M formic acid
    - Am and Cm elution with 2 M HCl

One lab combines this procedure with UTEVA- and Sr-Resin for the sequential determination of Sr, U, Pu, Am and Cm in precipitation and filter samples.

9.2. Source preparation

As for uranium.

9.3. Activity determination

As for Pu alpha emitters.

10. Rapid procedures

^{241}Am: Addition of ^{243}Am followed by hydroxide, oxalate, fluoride etc. precipitation and activity determination using gamma spectrometry.
Seawater: $MnO_2/FeOH_3$ precipitation, TRU-Resin separation and source preparation using micro co-precipitation.

Pu-isotopes: TTA extraction of Pu from 1 M HNO_3 or 1 M HCl, elution with 8 M HNO_3. (small samples).

Uranium: ICP-MS determination without separation using ^{233}U as tracer.

Sr-isotopes: LSC counting (cerenkov and LSC).

Sequential analysis: Using combinations of separation schemes. One lab has a sequential procedure for the determination of Sr, U, Pu, Am and Cm in "low interference" samples.

Activity determination: LSC or ICP-MS

11. Quality assurance

Quality assurance protocol, intercomparison exercises, analysis of reference materials, cross check using independent procedure, blank determination, labs to handle different activity levels, detector calibration and background determination, uncertainty determination (counting and tracer activity), internal auditing etc.

12. Comments

Most labs are satisfied with the procedures they use.

- Developing individual matrix-dependant separation procedures can speed up the analytical methods, but this requires a more stringent supervision.
- Smaller ion exchange columns and reduced "washing and elution volumes" for the separation of actinides can be used to speed up the analysis without negative effects.
- Do not through away Pu-sources as they can be used to get valuable information on ^{241}Pu content.

- Problems with low Am-yields and poor Am-separation when using TRU- and TEVA-Resin.
- HCl-$TiCl_3$ elution of Pu from TRU-Resin eliminates Po-interference caused when Pu is eluted with ammonium oxalate.
- Not always necessary to separate trivalent actinides from lanthanides.
- One lab has occasional problem with poor separation of Pu from uranium when using TIOA extraction and anion exchange (2x) for Pu-separation (drainage water).
- Low Pu-recoveries when using TTA extraction.
- Several labs are working with the development of sequential procedures.
- Occasional high background when Sr-Resin is used for Sr separation (effluent water).
- Observe ^{228}Ac in the ^{90}Y fraction.
- Low yields when using the “fuming nitric acid” procedure for the determination of ^{90}Sr in seawater and seaweed.
- Indirect determination of ^{90}Sr in seawater using ^{90}Y separation and determination should be tested.
- Avoid the use of fuming nitric acid.

13. Conclusion

The detailed information provided by the labs on their practise regarding the specified analysis, is very valuable. The benefits are obvious as radiochemical analysis now can be studied and compared in detail. It is now 20 years ago such a study was last undertaken in the Nordic countries. Although, most of the techniques in use are still the same, some deviations can be seen: Besides Pu separation using anion ex. chrom., there is not a single procedure that is used in all labs. More labs are doing americium determination. Due to the commercial availability of extraction chromatographic resins, more labs are now using this technique. A comparison of the results provided by different labs in the NKS-B LABINCO excersise, will also provide a direct comparison of the different procedures in use.

Isotope ratio measurements with single collector ICP-MS

The ICP-MS technique has during the last 20 years become increasingly important in the analysis of long-lived radioisotopes. Although useful both with respect to rapid measurements as well as performing highly accurate isotope ratio determinations and analysis of extremely low levels the technique requires careful attention. The need for routine use in producing data often overshadows the need to look into the many parameters governing the quality of data. This is particularly important in the analysis of long lived radioisotopes due to the often extremely low concentrations and/or the need to perform isotopic ratio determinations with high accuracy of extreme ratios (U&Th isotopes).

When considering the need to measure isotope ratios of Pu and U isotopes in environmental samples there are thus a number of issues that deserves investigation in order to ensure quality of the data. These could roughly be grouped as follows:

- Sample introduction system
- Plasma instability
- Dead time corrections
- Mass fractionation
- Interferences
- Abundance sensitivity (peak tailing)

Sample introduction system

One of the main advantages with the ICP-MS technique which was early emphasized was the possibility to introduce the aqueous sample directly into the instrument without any pre-treatment. This was a significant step forward compared to for instance the sample introduction used in thermal ionization mass spectrometry (TIMS) where samples has to be loaded onto thin filaments after a very tedious chemical refinement of the sample atoms. However, whereas the TIMS sample just need heating (which may be controlled with high accuracy) to produce an ion beam the ICP-MS requires the aqueous sample to pass though a complex chain of operations in order to produce the ion beam to the mass spectrometer. This chain of events causes a much greater fluctuation in the ion beam intensity than what is seen in the TIMS signal. The sample introduction system in ICP-MS may be of very different types but for aqueous samples they all have the task of delivering the sample in a form which makes the plasma being able to efficiently transform it into a an ion beam. This often means producing an aerosol (nebulization) which in turn may be dried from water and thereafter introduced into the plasma where the sample is atomized and ionized. Since the amount of sample introduced to the nebuliser depends on pump movements and gas flows, signal fluctuations are present already at this point. The sample loading further influences the aerosol production both with respect to magnitude and aerosol size distribution, both parameters which influences the signal intensity due to variations in aerosol transport and in residence time in the plasma. In reality it is very difficult to remove the signal variations caused by the sample introduction system. Although the pump movements may be omitted by allowing for self-aspiration of the sample this noise are often of low frequency and thus plays a minor role in the precision of isotope ratio measurements provided the mass scanning is not in phase with the noise. Another way of minimising the effect of the sample introduction system is to minimise the sample

input rate and to use desolvating equipment to remove all water form the aerosol stream before reaching the plasma. Since measurements of radioisotopes at environmental levels often are proceeded by a radiochemical separation process there is a natural way of keeping the sample small (sub millilitre) and thus partly avoiding the sample introduction problems.

Plasma instability

One of the main problems with isotope ratio measurements in ICP-MS is the plasma instability. The plasma flickering causes a continuous signal variation due to variation in ionisation/atomisation efficiency and to variations in ion kinetic energy. Although there are no simple ways of avoiding the plasma flickering it may be reduced by the use of a shielded torch. Another way of reducing the influence of the plasma flickering is to optimise the mass scanning frequency with respect to the plasma flickering frequency so they are not in phase. This is often practically easy since the plasma flickering may show a rather continuous frequency distribution. Fast scanning usually however show better precision in isotopic ratios than slow scanning but this may also be due to avoiding the noise from the sample introduction system.

Dead time corrections

A factor sometimes of importance is the corrections for dead time. This is usually not necessary when considering Pu-isotopes in environmental samples but may certainly be important when measuring U and Th isotope ratios ($^{235}U/^{238}U$, $^{234}U/^{238}U$, $^{236}U/^{238}U$ or $^{230}Th/^{232}Th$) where the ratios may be extremely large. In such cases the ion beam intensity for the major isotope may reach millions of counts per second while it may be less than 1cps for the minor one. The methods frequently used to correct for the dead time may be either as in radiometric counting (observing the ratio of measured to introduced sample as a function of sample introduction rate) or by using samples having certified isotope ratios of the selected isotopes and using them in the same concentration as the actual sample(s). There are a number of well suited certified reference samples available. Figure 1 shows the frequently used U500 form NBL having a $^{235}U/^{238}U$ ratio of 1 and which also contains the low abundance isotopes ^{234}U and ^{236}U.

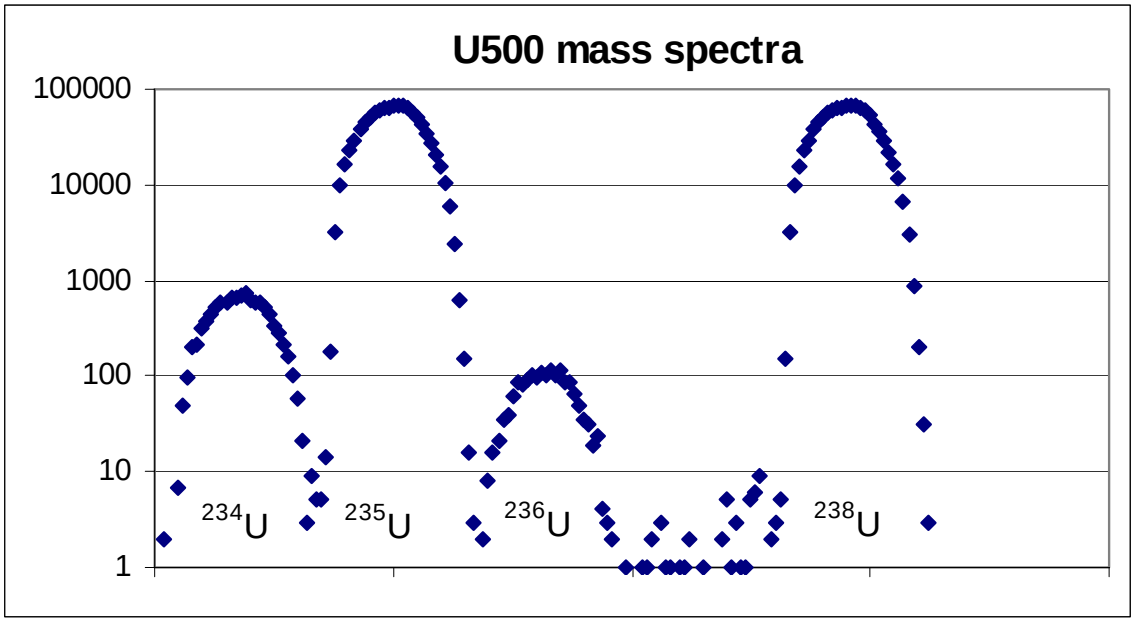


Figure 1: Typical mass spectrum of the certified reference material NBL U500. Obtained in low-resolution mode (R=300). PlasmaTrace 2 HR-ICP-MS, Risö National Laboratory.

Mass fractionation

Mass fractionation may occur between isotopes of different mass due to their different ability to be transported through the MS optics. Typically transmission of higher mass isotopes is somewhat larger than the lighter ones due to the fact that the lighter ones are more easily repelled away from the ion beam, especially in high intensity beams (samples with high concentration of the analyte or with a high salt content). The correction for mass fractionation is obviously a small problem for the heavy actinides since the relative difference in mass is rather small. Nevertheless, corrections may be necessary in high precision work and may be performed with the abovementioned U500 reference material with well known isotope ratios, provided the measurements are done at low count rates to minimise dead time.

Interferences

Interferences may originate from the sample or from the plasma or a combination of both. The interferences may be of polyatomic nature ($ArOH^+$) or simpler (UH^+) but all have in common that they interfere at mass position. The interferences may be avoided in different ways, either by removing the cause of them (eg removing the water also removes most hydrides and oxides) or by destroying them in collision cells filled with a gas of selected pressure and composition. They may also be avoided in high resolution MS by increasing the resolution. For the radioisotope work of U and Pu the most severe interferences are from hydrides since the UH^+ cause a significant contribution at mass 239. This is frequently avoided by desolvating equipment in the sample introduction system to remove the water. Further problems may arise due to combination of Pb and Bi isotopes with chloride isotopes all having masses in the 230-240 range. The strong interferences caused at several masses by chlorides is the main reason why HCl seldom is used in ICP-MS samples.

Abundance sensitivity (peak tailing)

A major limit in all ICP-MS instruments is the rather poor peak shape which means that the peak tail from major peaks disturb peaks of minor abundance. An example of how this may look is shown in figure 2. The strong tailing from the major isotope ^{232}Th makes the determination of the minor ^{230}Th very difficult. Unfortunately, the rather large spread in ion energy in ICP-instruments prevents the normal ion optics to produce better peaks. In high resolution instruments a combination of slits and energy filters may significantly improve the abundance sensitivity but the transmission is greatly reduced. Again, TIMS here shows a much better abundance sensitivity with some 0.01 ppm, compared to 5-50 ppm for ICP-MS, without compromising transmission.

The peak tailing is perhaps the largest problem in the analysis of extremely low levels of Pu-isotopes due to the presence of tailing from the 238U. For this reason the removal of uranium in the chemical separation procedure is of high priority and may ultimately determine to which level Pu may be measured. Without any separation it may be estimated that the detection limit for ^{239}Pu in an ordinary 1g soil sample (containing 20 mBq g^{-1} ^{238}U) is in the order of 100 mBq.

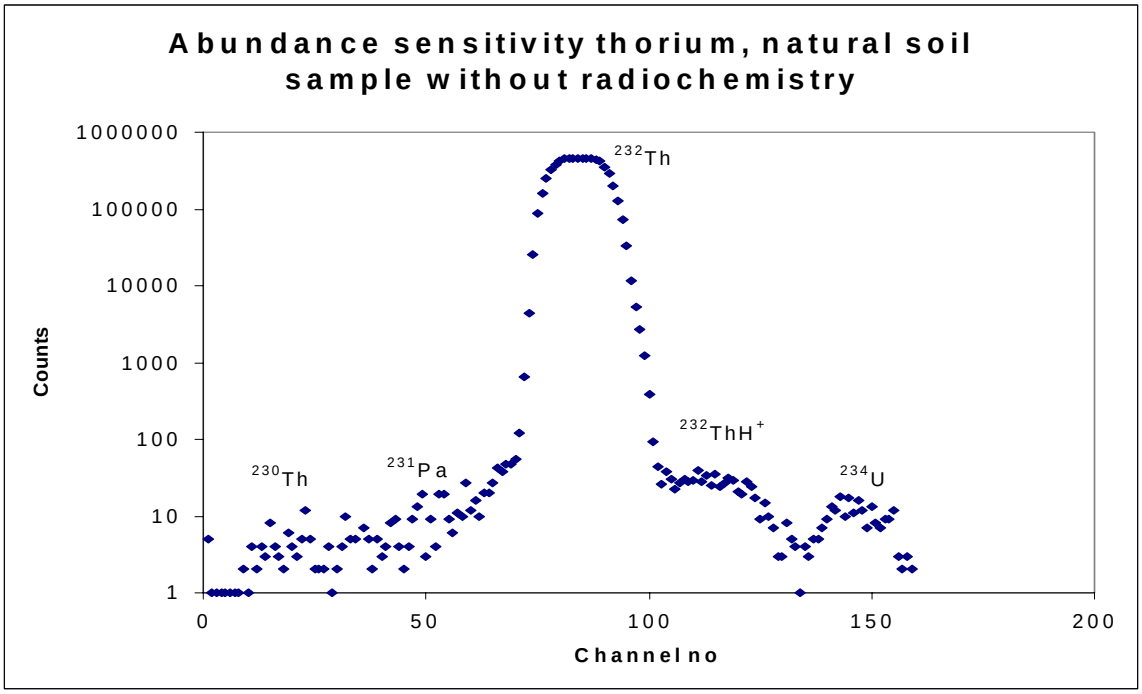


Figure 2:

Abundance sensitivity in low resolution mode (maximum transmission) for ICP-MS.

Concluions

Although ICP-MS has made it considerably easier to produce MS data the need to oversee parameters governing precision and transmission has not decreased. The use of certified reference samples with well known isotopic ratios, suitable sample introduction systems and a well designed chemical separation is of great help in minimising at least the most severe problems in isotope ratio measurements.

NKS-B Mini Seminar RADSEM 2004, 19 August 2004, Risø National Laboratory

NKS-B LABINCO Intercomparison
Intercomparison of laboratory analyses of radionuclides in environmental samples, 2004

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Extended abstract

The LABINCO intercomparison exercise involves laboratory analyses of radionuclides in environmental samples and food. Potential participants were invited in spring 2004 and feedback obtained on interest in sample types. Samples were distributed in July to 38 Nordic and Baltic laboratories and analytical results are requested by the end of October. The tables below list the sample types and sample amounts distributed and the radionuclides recommended for analysis. A final seminar in 2005 is planned for presenting and discussing results of the intercomparison and related radioanalytical issues.

Sample type, origin	Sample amount (kg)
Seawater - surface water from Danish waters, salinity about 8 ‰	50
Lake water - from a Swedish lake	25
Tap water - spiked with tritium	2 x 1
Sediment - from Roskilde Fjord	0.2
Seaweed - from Danish waters	0.15
Fish meal - Danish origin	2
Soil - Swedish origin	0.2
Dry milk - Danish origin	1
Cereal - Swedish origin	0.5
Lucerne/alfalfa pellets – Danish origin	2
Soil, Certified Reference Material	0.1

Nuclide	Sea-water	Lake water	Tap water	Sedi-ment	Sea-weed	Fish meal	Soil	Dry milk	Ce-real	Lu-cerne	Soil CRM
Cs-137	X	X		X	X	X	X	X	X	X	X
Sr-90	X	X		X	X	X	X	X	X	X	X
Tc-99					X						
Pu-isotopes				X	X		X				X
Am-241				X	X		X				X
Ra-226				X	X		X				X
Th-232				X	X		X				X
K-40				X	X	X	X	X	X	X	X
Po-210				X	X		X				X
Tritium			X								
Total alpha		X									
Total beta		X									
Other gamma				X	X		X				X

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IRADES - Assessment of Internal Doses in Emergency Situations

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EXTENDED ABSTRACT

Introduction

The need for assessing internal radiation doses in emergency situations have been demonstrated after accidents in Brasil, Ukraine and other countries. Studies in Ukraine, Belarussia and the southern parts of Russia after the Chernobyl accident showed an unexpected increase of thyroid cancers in children and young people. In the Nordic countries much work has been put down on quality assurance of direct measurements of radioactive substances in the body and on training of dose assessment calculations. This work has been partly funded by NKS. A good network of Nordic specialists has been created. But more needs to be done regarding the handling of an emergency situation. Nordic emergency preparedness exercises have not included training of direct measurements of people in the early phase of an emergency. There are not enough instruments or people to use them immediately available if suddenly needed. The aim of IRADES is to improve the situation especially for thyroid measurements which should be done as soon as possible after an alert to give support for decision making.

In a nuclear explosion more shortlived nuclides are distributed in the environment than in a nuclear power plant accident. In a nuclear explosion the radiation dose by inhalation is small compared to the external dose. In a nuclear power plant accident inhalation is often the dominating route of exposure at least during the first few days. Ingestion via food and drink become important after the radionuclides enter the foodchains.

Thyroid and whole-body counting measurements

To gain most from the thyroid measurements the procedure should be started without delay. Direct measurements give individual results and results for groups. An important protective measure is to administer stable iodine in case of exposure to radioactive iodine nuclides. But the protective effect is strongly dependent on time of administration. The effect decreases if the tablet is administered too early or too late. In an expected fallout situation the iodine tablet should preferably be taken 1-6 hours before exposure. For example 50 mg potassiumiodide KI given 30 minutes after exposure to ^{131}I decreases the dose to the thyroid to six percent of the dose without KI administration. When more than 12 hours has elapsed after intake administration of stable iodine no longer decreases the radiation dose to the thyroid. The iodine tablet is not any “wonderdrug” which would totally prevent any harmful effects on health.

In parallell with an earlier NKS project the five Nordic radiation protection institutes funded the phantoms to be used for calibrations and intercomparisons of both thyroid and whole-body counting systems.

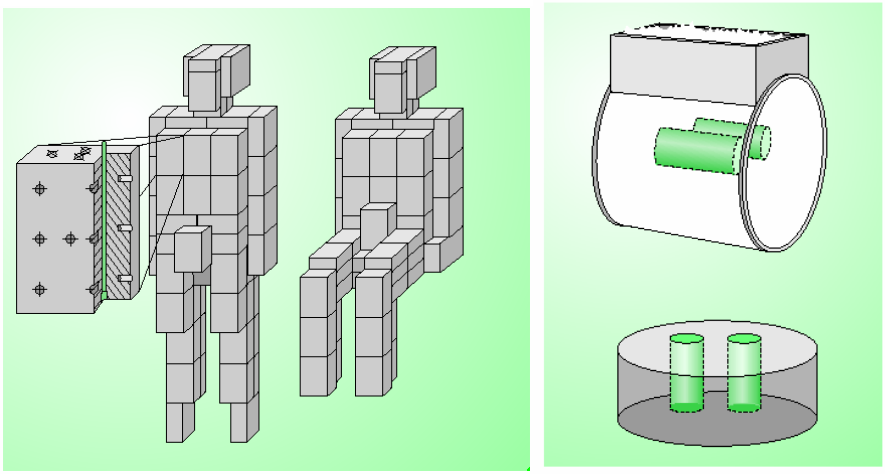


Figure 1. Whole-body and thyroid phantoms.

In the NKS-BOK-2.1.2 project a handbook listing instruments that can be used for thyroid and whole-body counting and the corresponding calibration factors was prepared. But no information on availability of instruments was collected. Nor does it say how many trained persons would be needed to do the measurements.

An inventory of available instruments for thyroid monitoring in the Nordic countries is underway within the IRADES project. Table 1 shows the questions asked. Some of the instruments are calibrated for ^{131}I others have to be calibrated. There are different types of measurement systems like thyroid monitors, instruments for uptake measurements of ^{131}I at hospitals, handheld instruments and whole-body counters that can be used. Some of them are designed for this purpose and others are routinely used for other purposes.



Figure 2. Examples of simple instruments and measurement geometries that can be used in emergency situations.

Calibration and intercomparison excercises will be offered to all Nordic organisations involved in activity determinations and internal dose assessments. For quality assurance and especially to fulfill accreditation requirements it is necessary to take part in intercomparison projects. Thanks to NKS it has been possible to arrange intercomparisons of direct measurements of people. A workshop on intercalibration and dose assessment will be arranged at the end of the IRADES project.

The inventory shows that there are tens if not hundreds of instruments that could be used in emergency situations. Some of them would naturally be needed for other measurements. Most of them are not gammaspectrometers but single channel instruments. The most sophisticated systems are whole-body counters with Ge-detectors and scanning options. In Denmark there are two whole-

body counting laboratories, in Finland two, in Norway four and in Sweden eleven. Whole-body counters are not suitable for measurements of large groups of people. At hospitals there are instruments for uptake measurements of I-131 in thyroidea which could easily be used in emergency situations. Gammacameras are better suited for whole-body ^{137}Cs measurements.

Plans for measurement strategies should be developed in each country taking into account national regulations, local circumstances and resources. In view of the close contact between people in the Nordic countries, it is very important to have the same approach in all countries. This will increase the public reassurance in case of a nuclear emergency situation. Also the bilateral agreements among the Nordic countries about assistance in emergency situations makes it important to cooperate.

End users of the IRADES report will be the radiation protection authorities. The biggest challenge will probably be to find and motivate enough people to be trained to handle the measurements in an emergency situation. These should not be the best experts in the field. The experts are needed for more demanding radiation protection tasks but should be there for advice, recommendations and support.

Technical description of a thyroid monitor

In Finland new equipment for thyroid monitoring has been obtained and preliminary plans for the monitoring strategy made. A short technical description of the Atomtex RKG-AT1320 monitor for detection of ^{131}I in field conditions is given below to illustrate the availability of light weight hand held instruments.
The thyroid monitor consists of a detector with 1”x1” NaI(Tl) crystal, control unit and lead collimator (Figure 3). The energy resolution of the crystal is better than 8 %. The monitor works as a spectrometer making it possible to do real time spectrum analysis in the field. The collected spectrum is shown on the display of the control unit. It is possible to store 299 spectra with size of 512 channels in the detection unit, from where they can be later on transferred to a PC. The energy stabilization is done before starting measurements using a ^{137}Cs source. Likewise the background control and sensitivity check (also with ^{137}Cs source) are carried out before measurements.



Figure 3. A RKG-AT1320 thyroid monitor: NaI(Tl) detector, control unit, charger, ^{137}Cs control source and cables

The aim of thyroid monitoring is to determine the ^{131}I activity in the thyroid with minimal interference from activity in the rest of the body and the environment. Thus it would be ideal to place a shielded or a collimated detector near the neck at the position of the thyroid. Assessment of the contribution to the detector response from radioiodine in blood and in surrounding measurement area can be made by placing the detector over a different area in of the body (e.g. thigh). However, the RKG-AT1320 thyroid monitors are planned to be movable and easy to carry. During the measurement the detection unit is placed to a stand, but it can also be held in hand. Thus, the weight of the detection unit cannot be more than a couple of kilograms. The collimator used in the present set-up is made of 5 mm thick lead (weight of the collimator roughly 1 kg). Handheld monitors without any collimator can also be used but the measurement uncertainty is higher.

The detection efficiency depends greatly on the distance between the detector and thyroid. To get good statistics in a short time the detector should be placed as close to the neck as possible. The exact position of the thyroid must be known in order to get right results in the close geometry. If the detection distance is longer, is not so crucial to place the detector exactly on the thyroid. Two measurement distances were used in the calibration of the RKG-AT1320 thyroid monitors, 7 cm and 20 cm (Figure 4). Three calibrations were done for both distances: adult, teenager and child. The St. Petersburg thyroid phantom and whole body phantom with ^{40}K rods were used in the calibration. Two capsules filled with ^{131}I solution were placed in the thyroid phantom. The minimum detectable activities (MDAs) when using the measurement time of 100 s in normal conditions are about 2000 Bq and 330 Bq for 20 cm and 7 cm detection distances, respectively. If longer time is chosen (for example 600s), the MDAs are significantly lower: 760Bq and 120 Bq for 20 cm and 7 cm distances. The set-up with 7 cm detection distance is about six times more efficient than that with 20 cm distance (Figure 4).

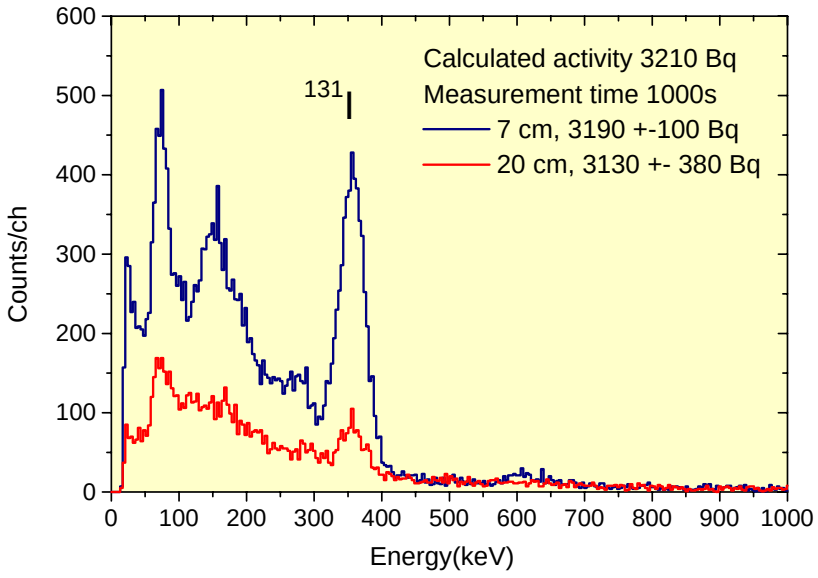


Figure 4. Gamma-ray energy spectra collected from a ^{131}I thyroid phantom (distances 7cm and 20 cm).

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Table 1.
Availability of instruments

	Stationary	Portable
How many instruments do you have for measurement and determination of ^{131}I-activity in thyroid?		
	Yes	No
Do you have personel able to train others to measure ^{131}I-activity in thyroid in an emergency situation?		
Are you interested in taking part in a intercomparison/calibration project with a thyroid phantom with a source of about 30 kBq (”mock-iodine”, ^{133}Ba + ^{137}Cs) ?		

Questions about instruments for measurement. (Copy for all instruments)

	Stationary	Portable
Is the instrument portable or stationary?		
	Yes	No
Is the instrument calibrated for measurement and determination of ^{131}I i thyroid?		
What is your estimation of MDA for a measurement of a couple of minutes? (for example 3 * SD for the background)		kBq

Extra information on the measuring instruments. (Of use to others with the same instrument but not absolutely necessary)

Manifacturer	Instrumenttype	Detectortype	Detectorsize	Calibrationfactor (µSv/h/Bq or cps/Bq ?)

Namn	
Adress	
Tel	
Fax	
E-Mail	

HUMAN METABOLISM OF CAESIUM

Status report on NKS-B prtoject Cskinetik

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Description — A study of the human metabolism of caesium incorporated in foodstuff compared with caesium in ion solution has been carried out mainly at the department of Rad. Physics in Malmö, starting in 2001 and ending recently, involving three human subjects who were subjected to intakes of contaminated fish and mushroom, together with a solution of ^{134}CsCl. After the intake a full sampling of all excreta during the first week after intake and whole-body counting during at least six months after were carried out. A continuation of that study, which was granted by the Swedish Radiation Protection Authority, is now undertaken by means of NKS funding. The aim is to, *i.)* investigate by means of literature surveys whether Scandinavian populations exhibit shorter biological half-time of radiocaesium than other populations, which has sometimes been hinted in the literature (ICRP 56); *ii.)* extend the kinetic experiments to more human subjects from the other Nordic countries. A shorter biological half-time in humans than the value of 110 d suggested by the ICRP, may in part explain the discrepancies observed between the measured ^{137}Cs body burdens in some Nordic populations and the estimated body burdens of ^{137}Cs in humans from detailed food surveys of the same populations (NKS-projekt).

In April 2004 the project CsKinetic was launched within the NKS-B programme. In August 18 a project meeting was held at Roskilde, where we discussed further steps in the project. It was concluded that the availability of human volunteers is still a problem, in terms of fixed dates for the on-set of experiments, but in Denmark it is most likely that the kinetic experiments can start in late September this year.

The following activities have now (August 30, 2004) been achieved:

i.) A literature survey containing more than 50 references on metabolic studies have been compiled by the participants in the project. Only studies conducted on adults involving both sampling of excreta as well as whole-body counting are included in the survey. The literature survey is yet to be complete, but a draft has been submitted to the NKS-B co-ordinator.

ii.) A survey of human candidates for participating in the ^{137}Cs and ^{134}Cs kinetic study during the coming autumn was done at a meeting shortly after the project was launched.

iii.) Collection of environmental samples that can be used for food ingestion to human subjects containing an amount of 137Cs that enables excretion samples to be detected within reasonable acquisition times by gamma spectrometry. A brief survey of what possible food samples usable to the study was done in late spring 2004, and it was thus concluded that reindeer meat in Norway and Sweden contains to little (less than 5 kBq kg-1) to be of use. Instead it was decided to return to the area outside of Gävle in Sweden, where the food samples of the previous study were collected. On August 11, 8 number of perch, of about 1 kg size were caught in the lake Hedsjön (Fig. 1.), together with a few hectograms of mushrooms in a forest area south of Gävle. The samples are temporarily at the SSI in Stockholm where preliminary ^{137}Cs activity determinations have been carried out (Fig. 2). The results show that the activity concentration is, on average, 7 kBq kg^{-1}, which means that it may require up to 5 hectogrammes of fish meat in order to have more than 2 kBq ^{137}Cs in edible form (taking into account of cooking losses of up to 30%).

Coming activities:

i.) Food preparation of the fish caught in Hedsjön is to be carried out in early September, in order to commence the first new human kinetic experiments on two volunteers from Denmark.

ii.) A review of the Nordic references of the Cs-kinetic studies, where an attempt to test whether a systematic difference in the biological half-time can be observed when compared with results from international studies.

iii.) A preliminary report of the literature study will be sent in to the NKS-B programme manager according to the contract in the beginning of October this year. There are also plans for application of renewal of funds for the project into the next year, in order to complete the kinetic experiments. The time schedule of further human experiments depends on various factors, availability of sampling and access to whole-body counters, etc. It is, however, expected that the other Nordic countries can contribute to additional human volunteers within a 1-year period.

Figure 1.

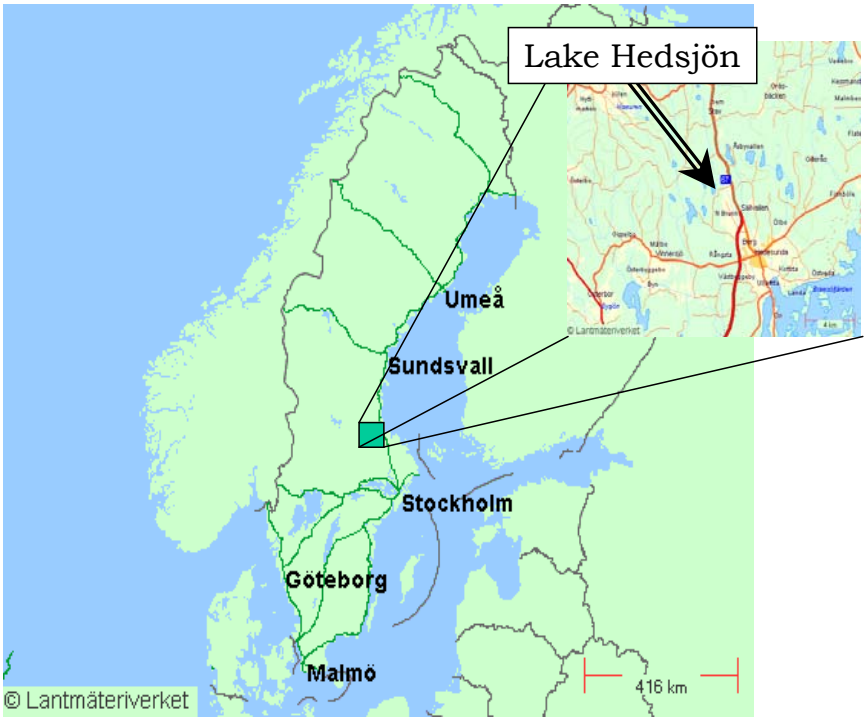
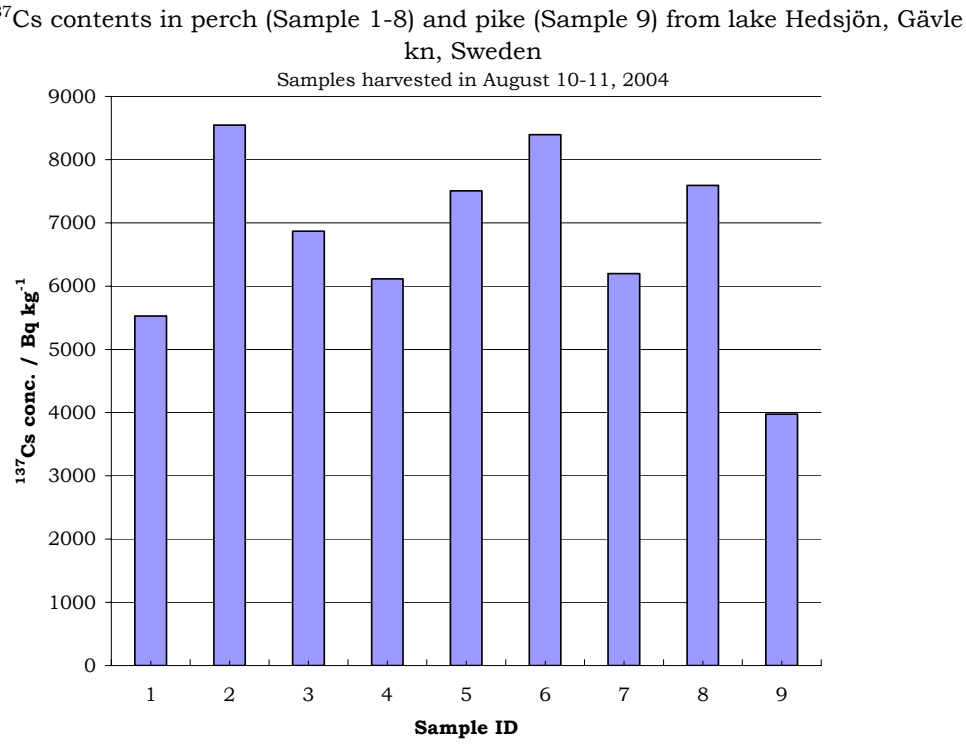


Figure 2.



REIN: Regional differences in reindeer radiocaesium contamination

L. Skuterud, Norwegian Radiation Protection Authority

Introduction

Reindeer is the part of Nordic food production most vulnerable to radioactive contamination. More than 15 y after the Chernobyl accident, countermeasures are still required in some Norwegian and Swedish reindeer herding districts before animals can be slaughtered for trade (i.e., contain less than 3000 Bq kg^{-1} in Norway and less than 1500 Bq kg^{-1} in Sweden). In autumn 2002, ^{137}Cs levels reached 9000 Bq kg^{-1} in a reindeer herd in central Norway due to grazing of an area kept aside due to high Chernobyl deposition density. Futhermore, radiocaesium concentrations in the southern Saami population are today comparable to, or higher than, those in people close to Chernobyl. A thorough understanding of reindeer radioecology is therefore an inevitable prerequisite in Nordic emergency preparedness.

Several radioecological studies of reindeer and reindeer meat consumers have been carried in the Nordic countries over the last 40 years. Nevertheless, there are still important areas of lacking knowledge, and changing environmental conditions like herding practices and habitat use alters factors once regarded as well-known. Despite all the reindeer radioecological studies and the recognised vulnerability of reindeer to radiocaesium fallout, no applicable model exists today. There is therefore a continuing need for Nordic cooperation in the area of reindeer radioecology and radiation protection, aimed at sharing the recent experiences and further developing the common basis for Nordic management and emergency preparedness.

Traditional reindeer husbandry was based nearly exclusively on winter slaughtering of animals, when lichen is the dominant dietary item. In this situation, radiocaesium deposition density and lichen availability will determine the radiocaesium concentrations in reindeer. Therefore the focus of previous radioecological studies was radionuclide transfer in the lichen – reindeer pathway. However, the availability and intake of lichen varies between areas due to differences in pastures, resulting in variable transfer of radiocaesium to reindeer. Historically, large changes have occurred in the number of reindeer, the intensity of grazing and therefore availability of lichen. The heavy grazing of lichen carpets taking place in some areas of northern Fennoscandia during the last decades may have added more variability to the previous situation. No radioecological assessment of the dietary differences between areas, nor the consequences of intensified grazing, has yet been carried out.

Modern reindeer husbandry in Sweden and Norway also includes a significant slaughtering in early autumn. This represents a challenge in the nuclear emergency preparedness, since there are relatively few relevant data sets available. Lichen intake can be significant also in early autumn (Gaare and Staaland, 1994), but other fodder vegetation, especially fungi, can potentially dominate the radiocaesium intake. During the last years there have been situations in Norway with ^{137}Cs concentrations in reindeer in early September exceeding the intervention level of 3000 Bq kg^{-1}, with the levels decreasing till December. Overgrazing of lichen will also increase the effect of other fodder vegetation on ^{137}Cs concentrations in reindeer.

Modelling transfer of radiocaesium to reindeer is a challenge compared to domestic animals, both due to seasonal differences in caesium retention and diet (both total quantity and composition), seasonal migration into areas of different deposition, and diversity in environments (e.g. forests, mountains and coastal areas). Some initial work on reindeer modelling has been carried out both in Sweden (Åhman and Nylén, 1998) and Norway (Skuterud, part of an ongoing PhD project).

The REIN project

The REIN project is the first NKS project on reindeer since the RAD projects in the early 1990s, except for a limited review of ecological half-times as part of NKS-B ECODOSES in 2003. REIN is a one year project with a budget of 85 kDKK. Project participants are Norwegian Radiation Protection Authority (coordinator), STUK – Regional laboratory in Northern Finland, Swedish University of Agricultural Sciences, Swedish Defence Research Establishment and Norwegian Institute of Nature Research.

The aims of the project are:

- Synthesize available information on concentrations and ecological half-times in reindeer (expansion and update of review in ECODOSES 2003).
- Use recent data, especially vegetation data, to improve estimates of uptake in vegetation and dietary intake in an existing draft reindeer model (Åhman and Nylén, 1998).
- Validate the model against long-term data series in Finland, Sweden and Norway.

Milestones and deliverables of REIN are:

- Report on effective ecological half-times for ^{137}Cs in reindeer in the Nordic countries — 30 September
- Workshop on modelling long term trends in radiocaesium contamination in reindeer — 12-13 October
- Model for radiocaesium in reindeer — 30 November
- Draft final report — 15 November
- Final report — 31 December

Results and discussion

The review of ^{137}Cs effective ecological half-times in reindeer in ECODOSES identified 2-3 fold differences in half-times between reindeer grazing areas, from 7-9 y in northern Fennoscandia to 3-5 y in central Norway and Sweden. Age of fallout and the proportion of Chernobyl fallout in the area can explain some of but not the whole difference (Åhman et al., 2001). Furthermore, a reduction in half-times of 1-2 y due to intensified grazing has been identified in Finland (Rissanen et al., 2003). Information from Greenland indicated similar half-times (7-10 y), while the Icelandic reindeer data indicate very slow decrease in ^{137}Cs concentrations with time.

Updated information from two reindeer grazing districts in central and south Norway (Østre Namdal and Vågå respectively) shows that ^{137}Cs concentrations in reindeer slaughtered in early winter declined with effective half-times of 3.5-4 y until 1998-1999, and that thereafter no significant decline has been observable. Fig. 1 shows the data from central Norway.

Furthermore, no decline in ^{137}Cs concentrations was detectable in reindeer slaughtered in the autumn since 1995. The seasonal differences in ^{137}Cs concentrations in reindeer previously observed have been less pronounced during the last years, and ^{137}Cs concentrations have occasionally been higher in the autumn than in winter.

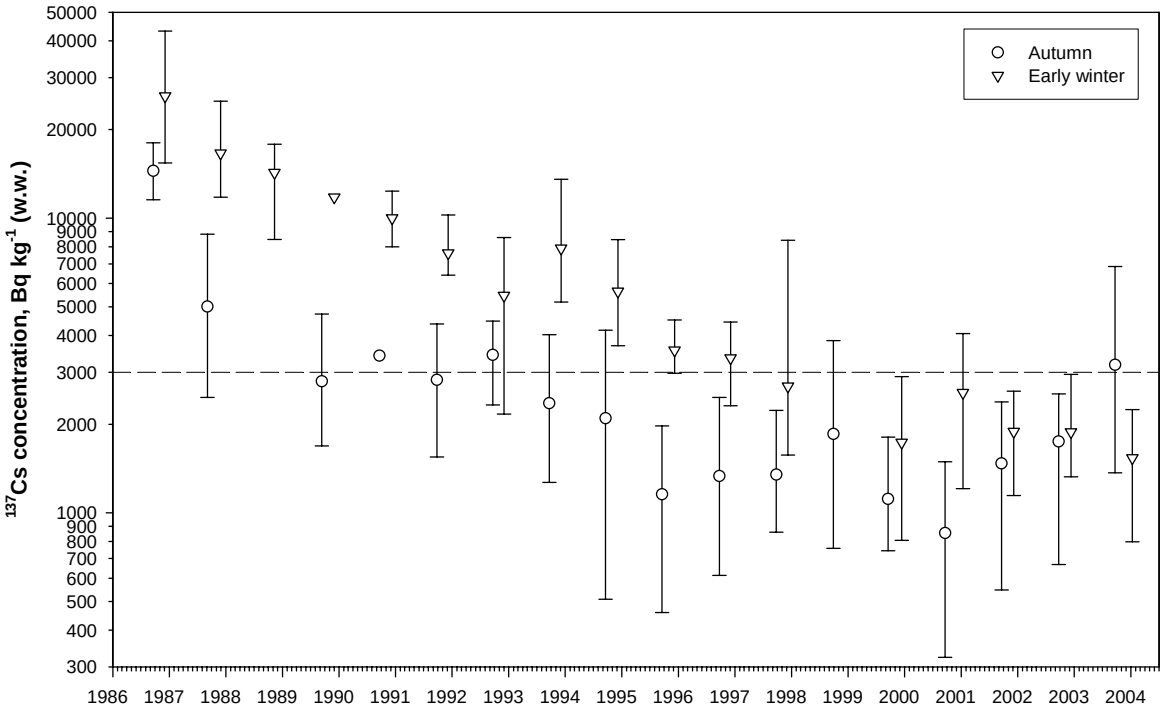


Figure 1 ^{137}Cs concentrations in reindeer calves in the autumn and early winter from Østre Namdal after the Chernobyl fallout: arithmetic mean values (Bq kg^{-1} w.w.) of 5 to 560 samples or live monitored animals (median 37; bars give ranges). The horizontal dashed line indicates the present intervention level for ^{137}Cs in reindeer meat in Norway (3000 Bq kg^{-1}).

Further comparison of regional data will be carried out in REIN in September 2004, and reasons for regional differences will be attempted elucidated. One potential reason for differences in half-times between north and south may be climatic influences on the rate of lichen growth, which will determine the rate of dilution of ^{137}Cs concentrations in lichen.

Decreasing seasonal differences in ^{137}Cs concentrations in reindeer have been expected due to the slower rates of decline in ^{137}Cs concentrations in other fodder vegetation than in lichen (Gaare and Staaland, 1994). ^{137}Cs concentrations in winter in the two districts studied in central and south Norway decreased to the level of autumn concentrations by 1998-1999. The lack of significant decrease in concentrations both in the autumn and winter thereafter indicate that vascular plants and fungi dominate as ^{137}Cs sources. Few studies on soil-to-plant transfer of radiocaesium have been continued in Fennoscandia after about 1995, but reviews of soil-to-plant transfer studies in other areas have shown insignificant reductions in transfer some years after fallout (e.g., Ehlken and Kirchner, 1996; Gastberger et al., 2000; Rigol et al., 2002). A similar slow or insignificant decline in concentrations in reindeer was not observed in the pre-Chernobyl monitoring data from northern Norway, possibly due to lichen being more dominant in the diet of reindeer in the northern more continental and arctic climate.

Rissanen and Ylipeiti (2003) presented results on ^{137}Cs concentrations in 28 fodder plant species from all Finnish reindeer herding districts, showing significant variability between species. Relatively high concentrations were found in some species, especially those growing on nutrient-poor mires. Similarly, lower nutritional quality of soils in Østre Namdal (Norway) due to a more oceanic climate (Moen, 1999) is hypothesized to be the cause of higher soil-to-plant transfer in this area than Vågå.

The studies in Østre Namdal and Vågå indicate that climatic influences on lichen growth and abundance (i.e. important factors in the reindeer's diet), and on soil properties, have a greater impact on long-term transfer of radiocaesium in the soil-plant/lichen-reindeer food chain than has been previously observed. Such climatic influences on soil-to-plant radiocaesium transfer will also be important for other grazing animals, e.g. sheep.

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Status Report of the INDOFERN Project *(New indicator organisms for environmental radioactivity)* in August 2004

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Objectives

The objective of the project is to identify new indicator organisms and biomarkers for assessment of environmental radioactivity in normal and emergency situations. The mechanisms of uptake and accumulation will also be studied. Indicator organisms are valuable monitoring objects in emergency situations because they accumulate effectively (and often very rapidly) radioactive substances from their medium. Thus, they are able to give information about nuclide composition and dispersion of radionuclides in early stage of emergency situations.

Traditionally, many organisms have been successfully used in radioecology (such as lichens, mosses, mushrooms, mussels, macro algae, especially the bladder-wrack *Fucus*). The objective of the project is to search new useful organisms accumulating effectively and specifically certain radionuclides in various Nordic ecosystems (terrestrial, fresh water, marine), and to compare their indicator value to those of the earlier known indicators. Since the Chernobyl accident, there is plenty of data on ^{137}Cs in biota but other radionuclides have been left to less attention. The aim of the project is to get more information about other long-lived nuclides (^{90}Sr, Pu and Am) and about the most abundant discharge nuclides from the nuclear power plants (e.g. ^{60}Co). In addition, the usability of different organs and tissues of the organisms as indicators will be studied.

The study yields new data on the occurrence, transport and concentrations of many important radionuclides in potential candidates of indicator organisms concerning a wide scale of Nordic ecosystems. The choice of candidates is based on references to literature concerning accumulation of radionuclides and stable elements to certain groups of organisms.

Collaborators and timetable

The project is participated by 8 laboratories representing all the Nordic countries: Risø National Laboratory (Denmark), University of Faroe Islands, STUK (Finland), Icelandic Radiation Protection Institute, Norwegian Radiation Protection Authority, Institute for Energy Technology (Norway), Agricultural University of Norway and Lund University (Sweden).

The planned duration of the project (and the expected period for funding from NKS) is 3.5 years (April 2002-September 2005). The annual schedule of the project work is highly specified due to the very short sampling periods during the summer months in the Nordic countries. The samples are taken during the summer months and the analysis of samples and the reporting of the results are carried out during the rest of the year. This is also the case considering the results from this year, 2004. The sampling period is still running and the results will be available at the end of the year at the earliest.

The results of the project will be presented through publications in open literature (some already published, some in print) and in an Open Final Seminar (NKS Report). In addition, the Project Group has decided to prepare a joint scientific publication based on the results. Each laboratory should contribute to this publication. This report should give a common overview about the use of indicator organisms in Nordic ecosystems, not only national results from different countries.

Collection of data

So far, data have been collected from about 150 organisms (species, family or group). The list of organisms consists of 44 mushrooms, 6 lichens, 5 mosses, 13 spore-bearing plants (*Pteridophytes*), 36 seed plants, 10 algae (including plankton and periphyton), 11 benthic animals, 11 fish species, 9 water fowls, 3 seals, 1 whale and the Polar Bear. A clear majority of the organisms (about 90 species) represent terrestrial environment; about 40-50 of them are from marine or brackish-water environments and about 10-20 from the fresh water environment. The most extensively studied individual organisms were from the marine/brackish-water environment, such as *Fucus vesiculosus* and *Mytilus edulis.*

The idea in sampling was to take the samples from relatively small areas where the environmental factors (type of soil etc.) and the amount of radioactive deposition are likely homogenous, which makes it possible to compare the indicator value of different organisms. In 2003 the sampling carried out by STUK was focused on fresh water organisms. About 40 samples were taken from lakes located in the areas which were most contaminated by the Chernobyl fallout in Finland.

Results

Terrestrial environment

In the terrestrial environment the activity concentrations of radionuclides (especially those of ^{137}Cs) show generally large internal variation within the same plant species and significant geographical variation between different sampling sites.

In many cases, mushrooms seemed to be the most effective collectors of ^{137}Cs. In 2002, the highest ^{137}Cs concentrations in Finland were detected in hedgehog mushrooms (*Hydnum* sp.), maximum 8500 Bq kg^{-1} dry wt., in ferns (*Polypodium vulgare)*, maximum 4300 Bq kg^{-1} dry wt., and then in lichens (*Hypogymnia [Parmelia] physodes*), 2600 Bq kg^{-1} dry wt. The ^{137}Cs concentrations were clearly lower e.g. in spruce and pine needles, birch leaves, and especially in juniper needles. In the twigs of blueberry and crowberry, the concentration was much higher than in the berries of crowberry, blueberry or lingonberry. The highest concentrations of ^{90}Sr were found in *Parmelia* lichens, ferns and the twigs of blueberry and crowberry (10-60 Bq kg^{-1} dry wt). The concentrations of ^{239,240}Pu and ^{241}Am were low in all the samples analysed (< 0.1 Bq kg^{-1} dry wt.) and those of ^{241}Pu were below the detection limit (1-2 Bq kg^{-1} dry wt.) (Ikäheimonen et al., 2003).

In Norway, the estimated ground deposition of ^{137}Cs (Bq m^{-2}) in each municipality and the corresponding activity concentrations of ^{137}Cs in different fungi species (n=1283) were used to calculate the ratio between the activity concentration in fungi and ground deposition (transfer factor). The transfer factors ranged from 0 to 5 (the maximum in a *Cortinarius* sample). The results of this study are summarised in Figures 1 and 2.

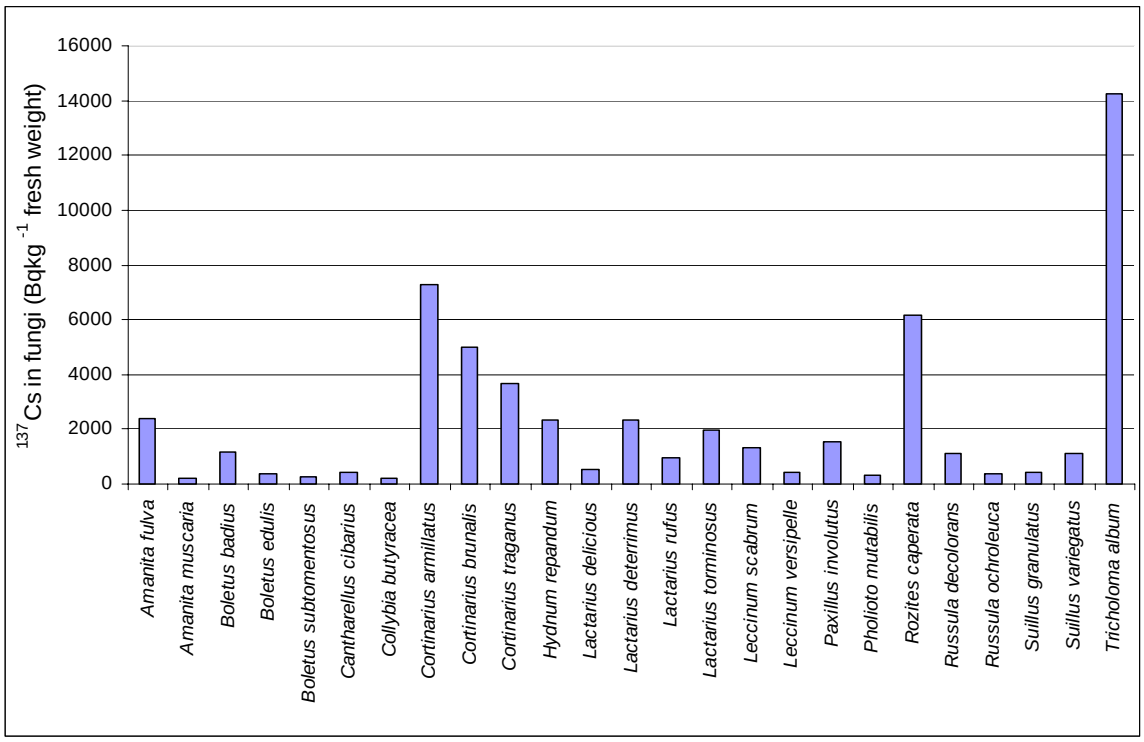


Figure 1. ^{137}Cs mean value (Bq kg^{-1} fresh weight) in different fungi species in Norway in 1990-2002.

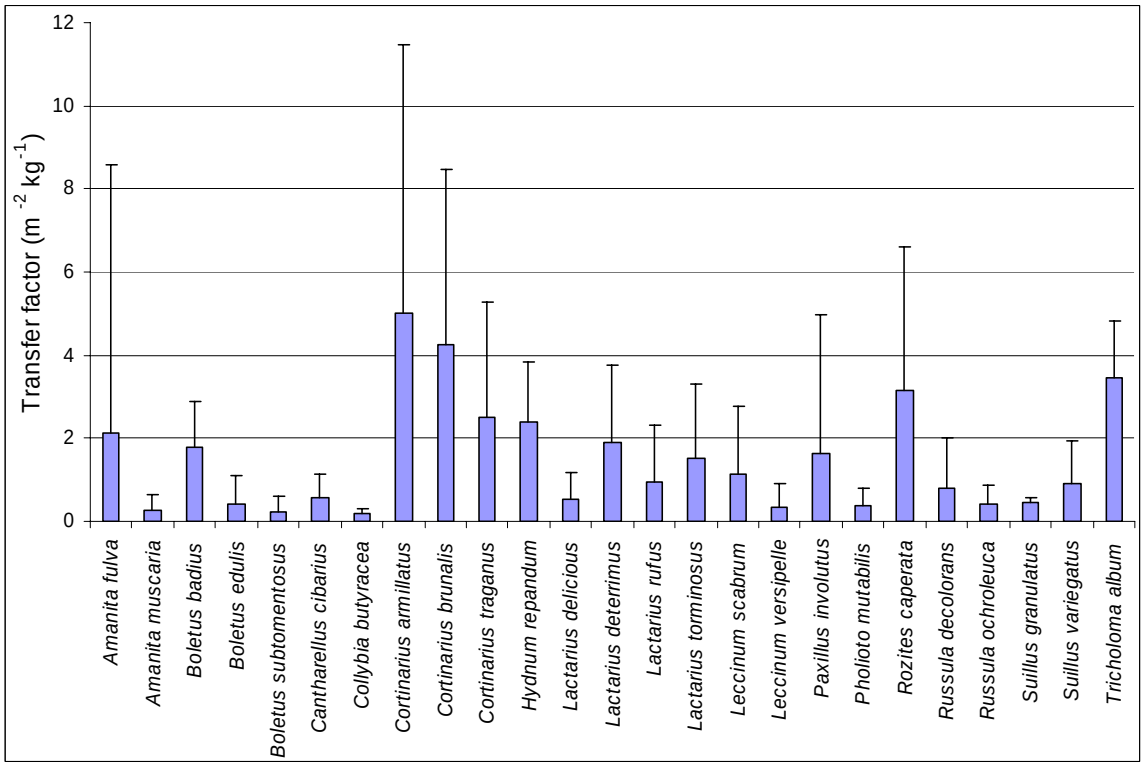


Figure 2. Mean transfer factor (m^2 kg^{-1}) with standard deviation in different fungi species (n=1283) in Norway in 1990-2002.

In a special vegetation-square-study carried out in Ringebu-Koppang region in Norway, activity concentrations of ^{137}Cs, ^{40}K, ^{90}Sr and ^{239,240}Pu were compared in five plant species. The ^{137}Cs concentrations were highest in Gypsy *(Rozites caperata),* Boletus and Reindeer lichen *(Cladina stellaris);* being lower but equal in Alpine fern *(Athyrium alpestre),* Dwarf birch *(Betula nana)* and in the twigs and berries of Northern crowberry *(Empetrum hermaphroditium).* The ^{40}K concentrations were highest in leaves of *Betula nana* and berries of *Empetrum hermaphroditium.* In the twigs of *Empetrum hermaphroditium,* the ^{40}K concentrations were lower and in *Cladina stellaris* generally below the detection limit. The activity concentrations of ^{90}Sr were higher in *Betula nana* and twigs of *Empetrum hermaphroditium* than in *Cladina stellaris* and berries of *Empetrum hermaphroditium.* The ^{239,240}Pu concentrations were highest in *Cladina stellaris* being lower and similar in *Betula nana,* twigs of *Empetrum hermaphroditium* and berries of *Empetrum hermaphroditium.*

The studies carried out by Lund University in terrestrial and fresh water environment have been focused on bracken (*Pteridium aquilinum*), water horsetail (*Equisetum fluviatile*), Rosebay Willowherb (*Epilobium angustifolium*), bulrush (*Typha latifolium*), bottle sedge (*Carex rostrata*) and wood horsetail (*Equisetum sylvaticum*). Also other species were analysed, such as common reed *(Phragmites australis),* broad-leaved pondweed *(Potamogeton natans),* common club-rush *(Schoenoplectus lacustris)* and white water lily *(Nymphaea alba).* The accumulation and aggregated transfer factors of ^{137}Cs were highest for bracken and horsetails, and they were the best indicator organisms among those analysed so far. The ^{40}K concentrations were highest in water horsetail. In the forest, the ^{137}Cs concentrations in needles from juniper were lower than in pine which in turn were lower than in spruce. ^{237}Np was studied in lichen *(Cladina stellaris)* at 18 sites in Sweden. The activity concentrations ranged from 0.1 to 2 mBq kg^{-1} with the highest concentrations in the areas where the Chernobyl fallout was highest as well.

In Icelandic terrestrial environment, the mosses of *Rhacomitrium* family were clearly the best indicators for ^{137}Cs. The internal variation of ^{137}Cs values within the organisms studied was considerable between the different sampling sites. The indicator value of *Empetrum nigrum* and *Cladonia arbuscula* was lower than that of *Rhacomitrium,* but the lowest ^{137}Cs concentrations were analysed in the leaves of tea-leaved willow (*Salix phylicifolia*) and in the needles of juniper (*Juniperus communis*).

Calluna vulgaris, Hylocomnium slendens and *Empetrum hermaphroditum* were, respectively, the most suitable indicators of radionuclides among the selected species in Faroe Islands. The measurements of ^{137}Cs and ^{90}Sr in Faroe Islands indicate a significant geographical variation of the activity concentrations in moss and lichen. The ^{137}Cs concentration in moss has decreased considerably since the Chernobyl accident (from 600 to 78 Bq kg^{-1} dry wt. at one sampling site).

In Finland, terrestrial plants were studied in 2001-2003 in three different sampling sites: Loviisa, Olkiluoto and Mänttä. The results are summarized in Figs 3-6. Mänttä represents the region most contaminated by the Chernobyl fallout in Finland. In general this can be seen in the ^{137}Cs concentrations in Mänttä, which are several times higher than in Loviisa and Olkiluoto. The highest concentrations in all the sites were found in mushrooms *(Hydnum., Cortinarius.* and *Hygrophorus)* and in ferns *(Polypodium, Dryopteris).* High concentrations were also detected in *Calluna vulgaris, Epilobium angustifolium* and in the twigs of *Vaccinium myrtillus,* as well as in the mosses. The highest ^{90}Sr concentrations in Loviisa and Olkiluoto were found in ferns and the twigs of *Vaccinium* and *Empetrum nigrum.* The Pu and Am concentrations were low: some mBq kg^{-1} dry wt. or less. The aggregated transfer factors of ^{137}Cs were highest for *Cortinarius, Hydnum* and *Hygrophorus* mushrooms, other mushrooms and ferns, and those of ^{90}Sr for the twigs, ferns and berries of *Empetrum nigrum.*

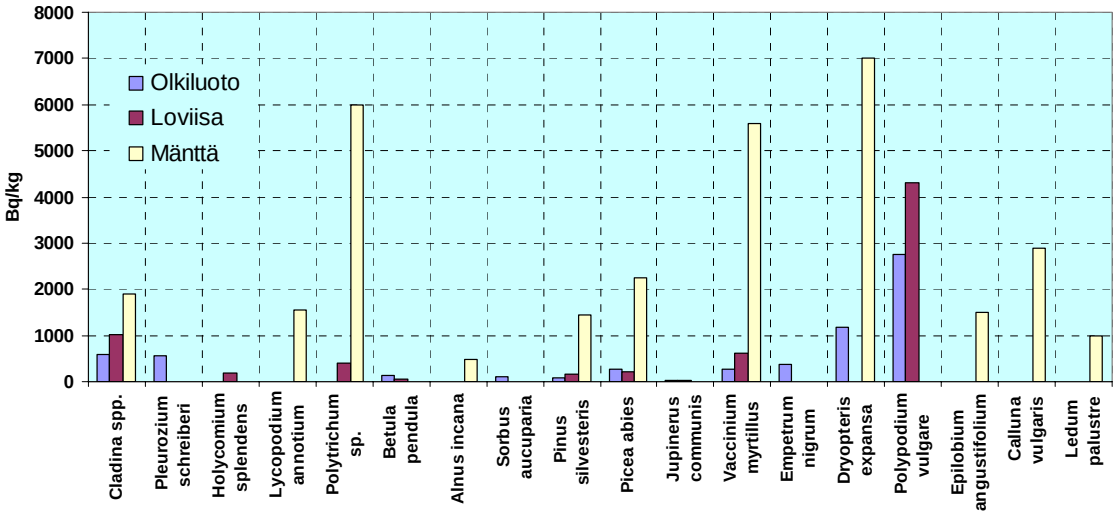


Figure 3. ^{137}Cs concentrations (Bq kg^{-1} dry wt.) in terrestrial plants in three different sampling sites (Olkiluoto, Loviisa and Mänttä) in Finland. Mänttä represents the area of the highest Chernobyl fall-out in Finland.

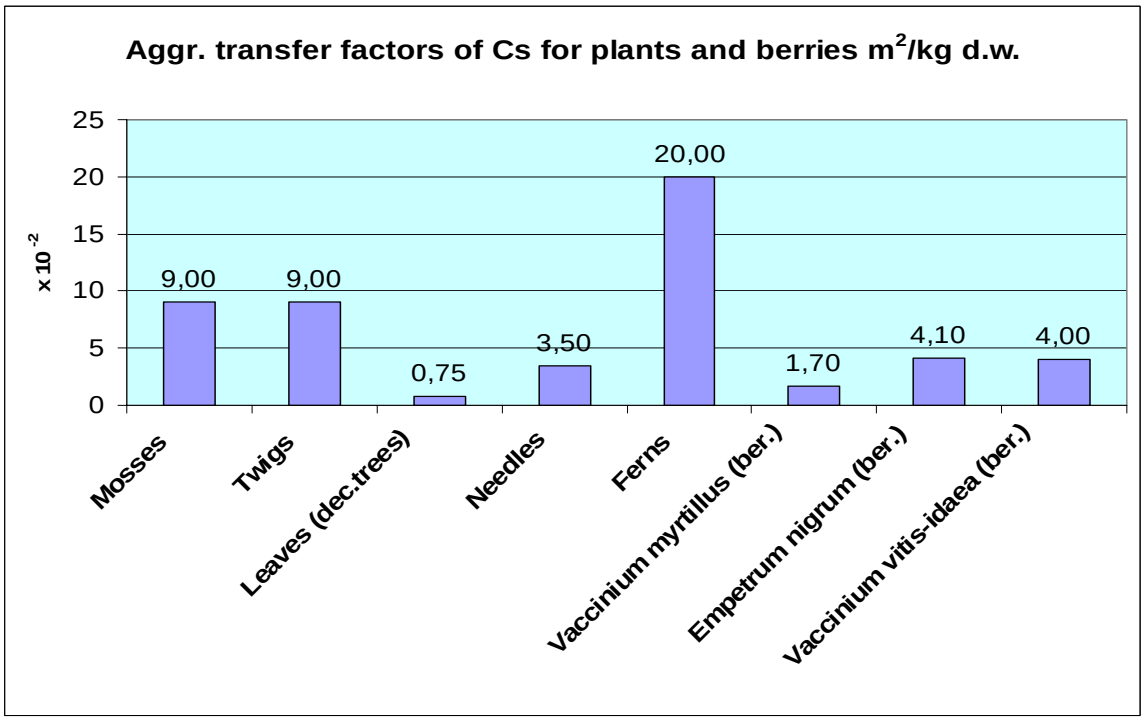


Figure 4. Aggregated transfer factors (m^2 kg^{-1} dry wt.) of ^{137}Cs for some vegetation groups in Finland (means of average values calculated separately for the three sampling sites).

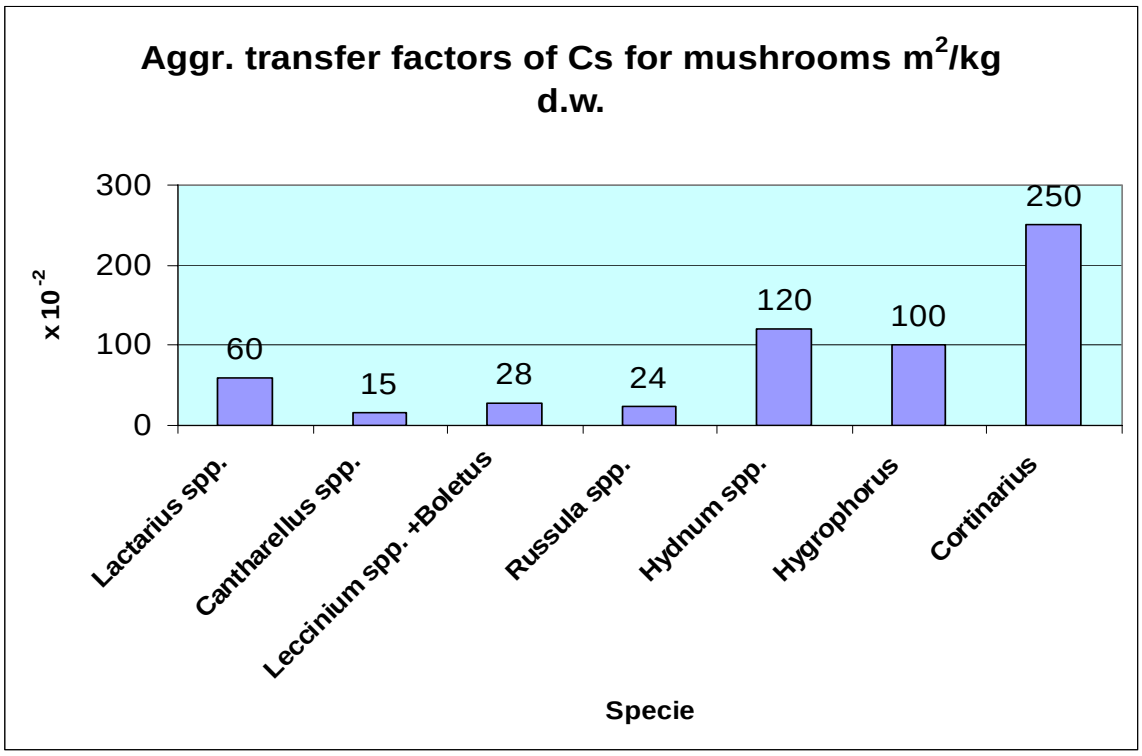


Figure 5. Aggregated transfer factors (m^2 kg^{-1} dry wt.) of ^{137}Cs for some mushrooms in Finland (means of average values calculated separately for the three sampling sites).

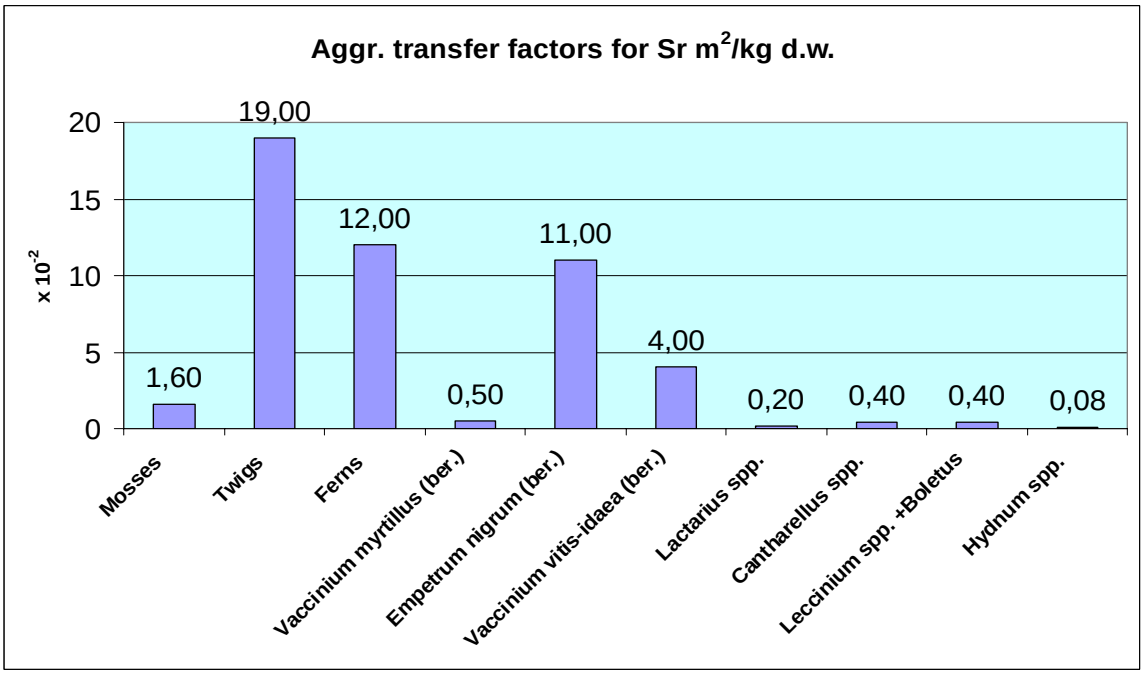


Figure 6. Aggregated transfer factors (m^2 kg^{-1} dry wt.) of ^{90}Sr for some plants, berries and mushrooms in Loviisa and Olkiluoto (means of local average values).

Aquatic environment

The results of a Marine Radioecology Study carried out in the sea area off the Olkiluoto Nuclear Power Plant (West coast of Finland) in 2001 are summarised in Figs 7-11. The aim of the study was to compare the indicator value of different members of the aquatic ecosystem for gamma emitting radionuclides in brackish water circumstances of the Bothnian Sea. The results are very well comparable because the samples were taken from a relatively small area.

The activity concentration of ^{40}K was clearly highest in a filamentous green alga *Cladophora glomerata*. The lowest concentrations were found in mussels, fish bones and egg shells (Fig. 7).

^{60}Co and ^{54}Mn were detected only at the lower trophic levels of the ecosystem. ^{60}Co was detected in phytoplankton, zooplankton, periphyton (maximum), macroalgae and other submerged aquatic plants, and in two mussel species (Common mussel, *Mytilus edulis*, and Baltic Tellin, *Macoma balthica*)(Fig. 8). ^{54}Mn was detected in zooplankton, periphyton, bladder-wrack (*Fucus vesiculosus*) and in *Ranunculus baudotii* (maximum). ^{60}Co and ^{54}Mn were not detected in fish or birds, or in their inner organs. This is in agreement with the results of the study carried out in the discharge area of Loviisa in 1988-1989 (INDOFERN Annual Report 2002).

The ^{137}Cs concentrations were highest in fish flesh (Fig. 9). The concentration factors (Fig. 10) ranged in fish from 540 in perch (*Perca fluviatilis*) to 83 in bream (*Abramis brama*). The concentration factors are based on fresh weight values. On the contrary, in fish-eating birds (Goosander, *Mergus merganser* and Great Black-backed Gull, *Larus marinus*), the highest concentrations were not in muscles but in liver and entrails. In birds' eggs the concentrations were very low; especially in egg shells. The concentration factors were calculated from ^{137}Cs concentrations in sea water also in the case of aquatic birds. Among the lower organisms, the best indicators of ^{137}Cs were the polychaete worm *Marenzelleria viridis*, periphyton, the bladder-wrack (*Fucus vesiculosus*) and the relict crustacean *Saduria entomon.*

Until 2001, most of the Chernobyl derived ^{134}Cs was already disappeared from the environment; only traces of this nuclide were detected in some fish samples, in zooplankton, periphyton and bladder-wrack.

The studies carried out in Norway show that for all the measured radionuclides except ^{90}Sr and ^{99}Tc, the activity concentrations were higher in Bladder-wrack *(Fucus vesiculosus)* than in Egg Wrack *(Ascophyllum nodosum)*. The activity concentrations of ^{137}Cs were on an average 50% higher and those of ^{40}K 20% higher in *Fucus* compared to *Ascophyllum*. On the contrary, the mass activity densities of ^{99}Tc were in average 60% higher for *Ascophyllum* than in *Fucus*, which indicates that ^{99}Tc is more effectively taken up in *Ascophyllum* than in *Fucus*. For ^{90}Sr the results do not show difference between the two species. The ^{7}Be concentrations were significantly higher in *Fucus* than in *Ascophyllum.*

The seasonal variations are more pronounced in *Fucus vesiculosus* than in *Ascophyllum nodosum.* In both species the ^{137}Cs concentrations were highest during the spring months at the south-eastern coast of Norway. The higher uptake in spring is probably due to increasing water temperatures, improving light conditions and larger outflow of Baltic Sea water at that period. The accumulation of ^{137}Cs and ^{40}K has been shown to be highest in the receptacles and the new vegetative tissue of *Fucus.* Because *Fucus* grows more rapidly and lives for a shorter time than *Ascophyllum* the ratio of the biomass of new vegetative tissue to the biomass of the whole plant is greater in *Fucus* than in *Ascophyllum.* The difference in growth rate between the two species may also explain the difference of seasonal variations in ^{137}Cs concentrations.

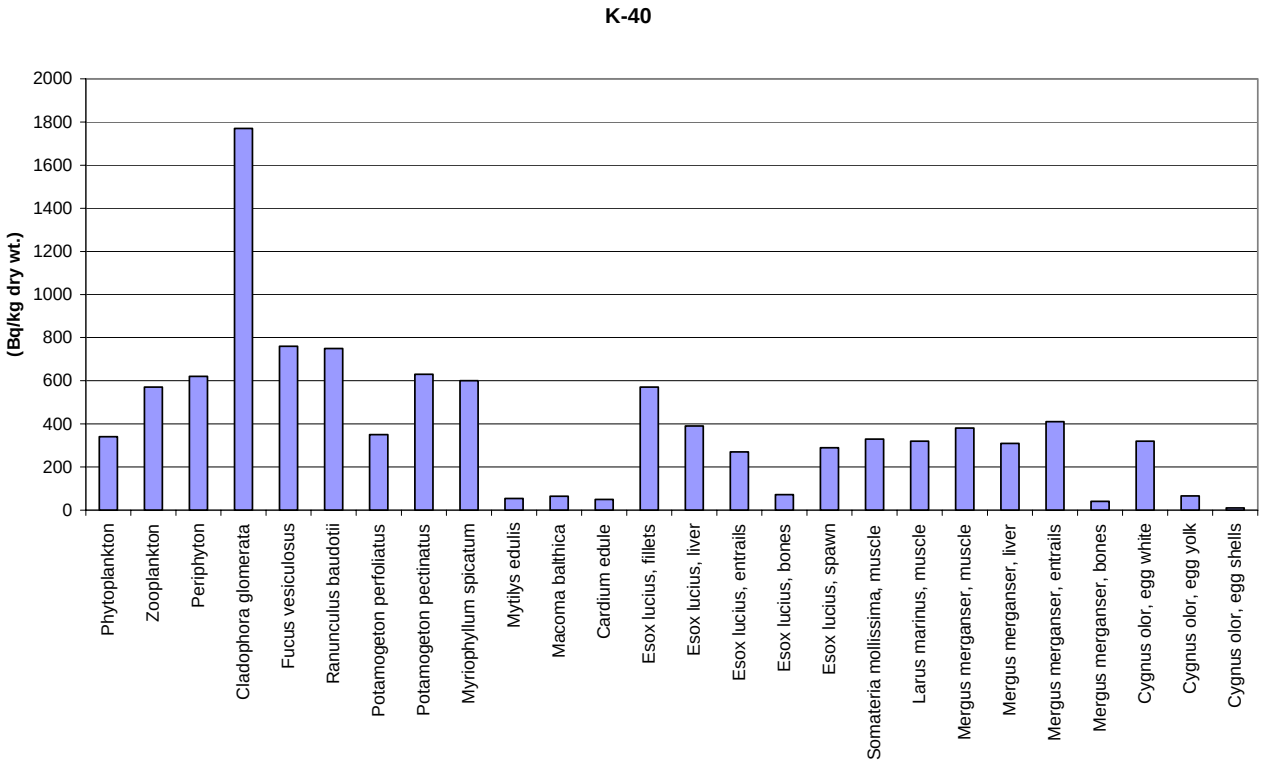


Figure 7. ^{40}K mean values (Bq kg^{-1} dry weight) in some indicator samples in the sea area off Olkiluoto in 2001.

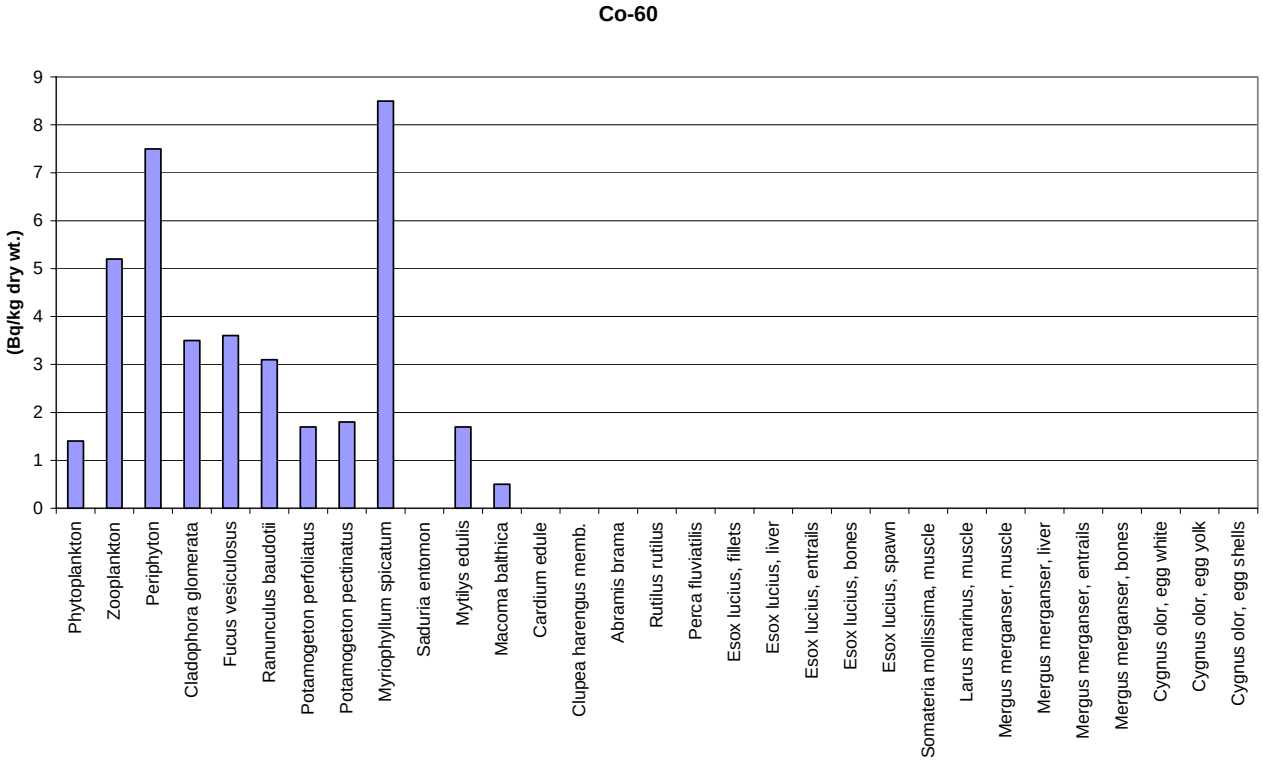


Figure 8. ^{60}Co mean values (Bq kg^{-1} dry weight) in some indicator samples in the sea area off Olkiluoto in 2001.

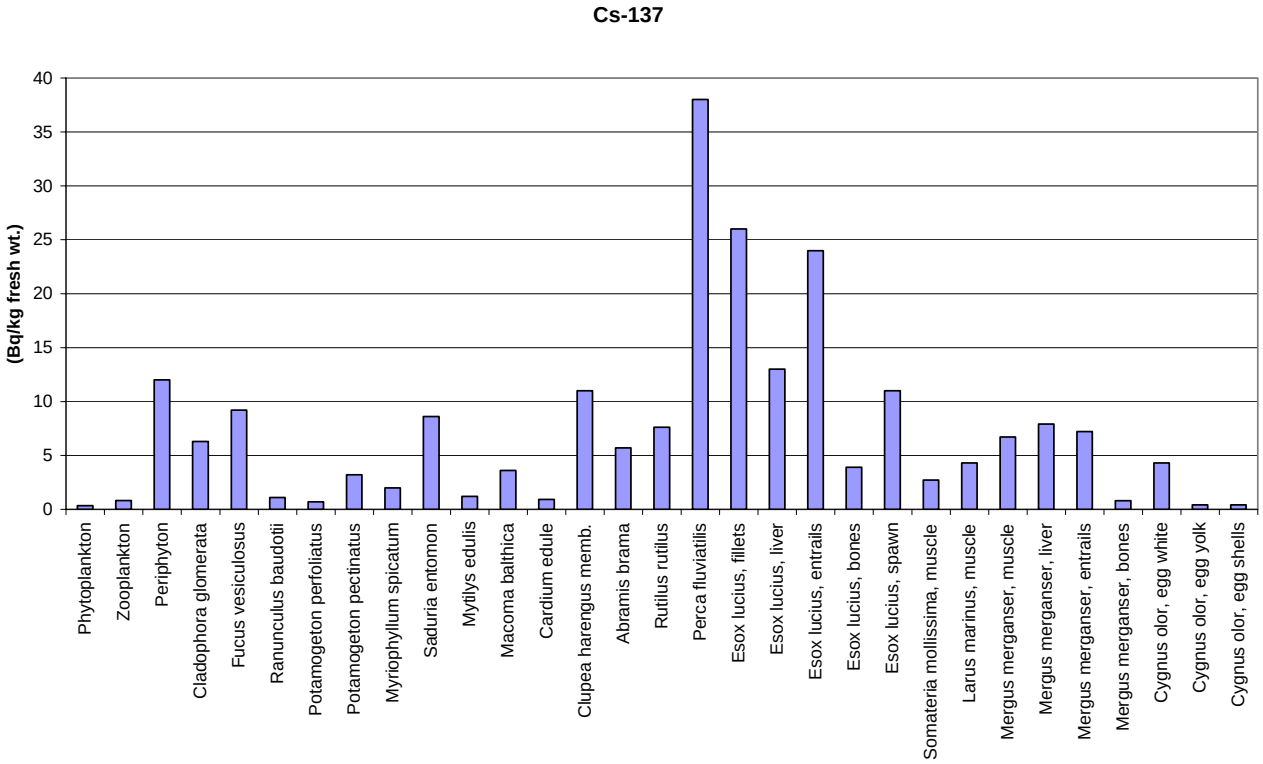


Figure 9. ^{137}Cs mean values (Bq kg^{-1} fresh weight) in some indicator samples in the sea area off Olkiluoto in 2001.

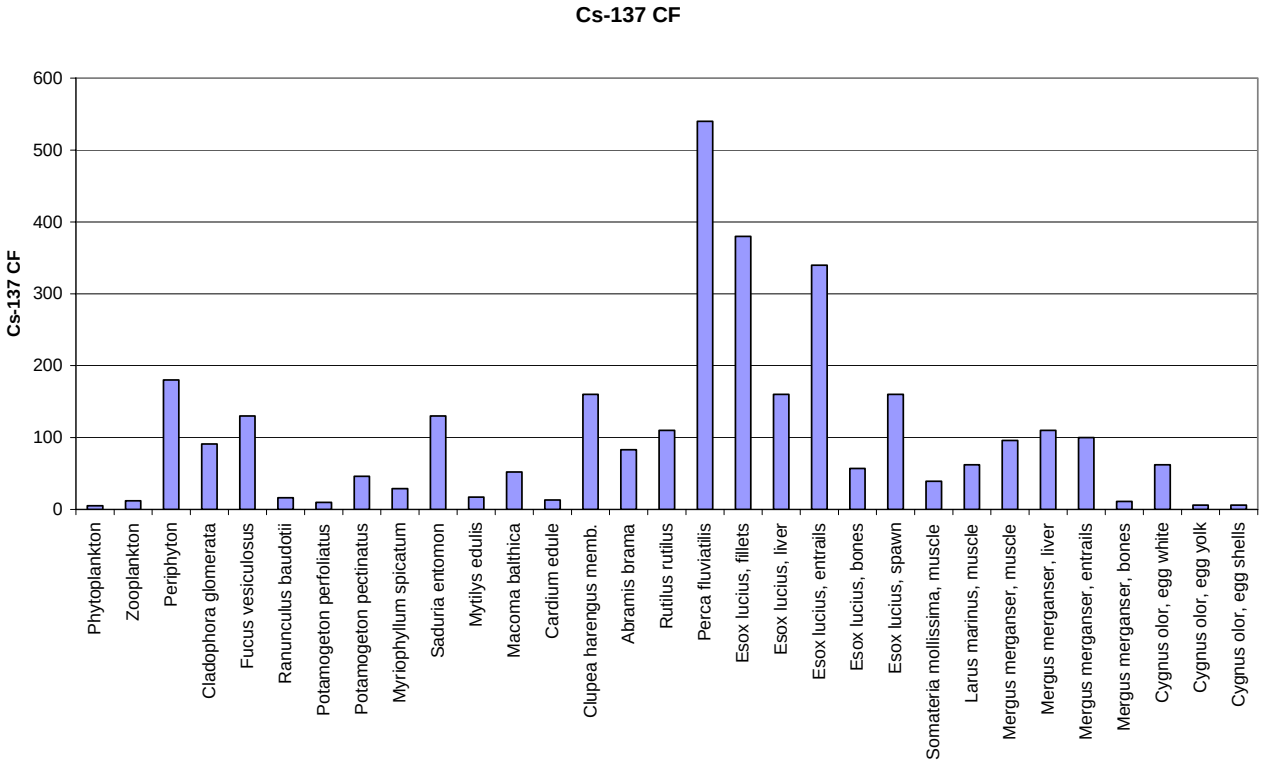


Figure 10. Mean concentration factors of ^{137}Cs (based on fresh weight values) in some indicator samples in the sea area off Olkiluoto in 2001.

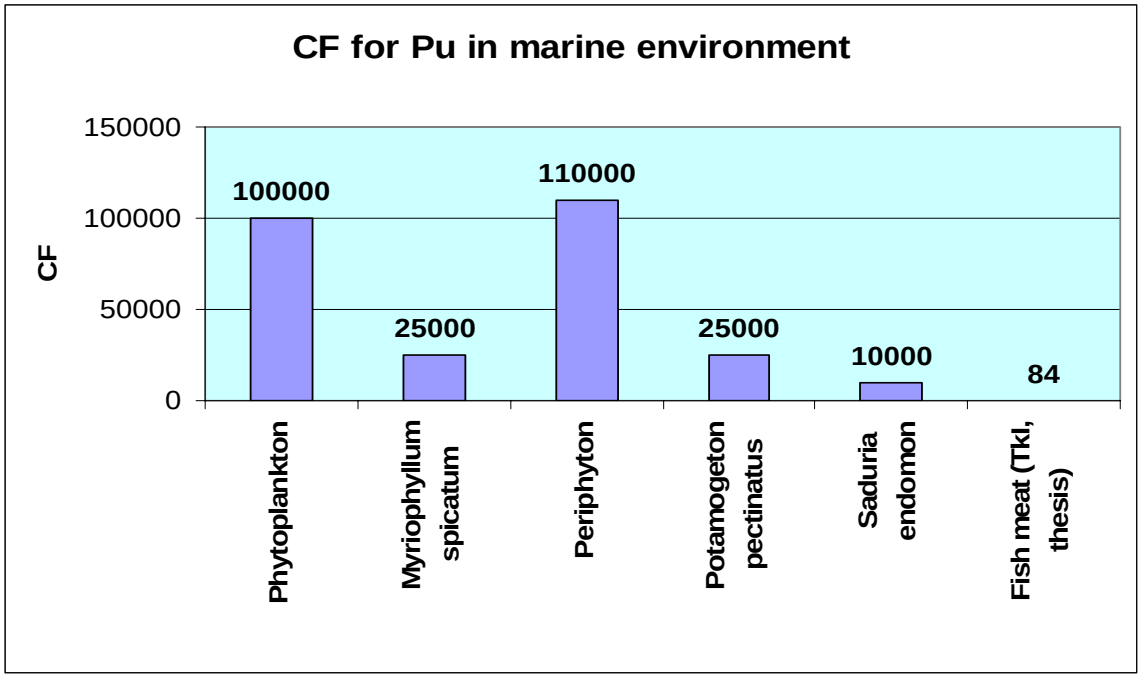


Figure 11. Concentration factors of Pu for some brackish water organisms in the Loviisa and Olkiluoto areas in 2001-2002.

The bladder-wrack, *Fucus vesiculosus*, has proved to be a suitable indicator for radioactivity also in the Faroese marine environment, as it reacts rapidly to ^{137}Cs contamination. The shorter post-Chernobyl half-life of ^{137}Cs compared to that from the 1960s, which has been demonstrated in the Faroese studies, is explained by the fact that the input from Chernobyl came as a short pulse, while the input to the atmosphere in the 1960s was distributed over a longer time scale.

In Iceland, samples of *Fucus vesiculosus*, *Ascophyllum nodosum*, mussels, crustaceans, aquatic birds, seals, a whale and a polar bear have been taken for indicator studies in the marine environment, but he radiocaesium concentrations have been very low or below the detection limit of the gamma-spectrometric measurements.

Different organs of seals from the Discofjord and Thule, Greenland, from west and south coast of Sweden and from the Baltic Sea have been analysed for ^{137}Cs and ^{210}Po by Lund University and Risø. The levels of ^{210}Po were high compared to average global values of ^{210}Po in fish, molluscs and crustaceans. The ^{210}Po concentrations were higher in liver and kidney than in meat, whereas radiocaesium is rather homogenously distributed in different organs. In Thule, the concentrations of ^{210}Po in seal *(Phoca groenlandica)* were found to be about 8 Bq kg^{-1} in flesh and 43 Bq kg^{-1} in liver. A clear salinity effect could be seen in radiocaesium levels; the levels were on the west coast of Sweden much lower than in the Baltic Sea. ^{210}Pb concentrations were very low, i.e. generally not detectable or 100 times lower than those of ^{210}Po.

At the Swedish west coast the concentration factor for ^{99}Tc in *Fucus vesiculosus* varied between 28 000 and 150 000. In Skagerrak, the concentration factor for $^{239+240}$Pu was estimated to be about 7000 and that for ^{237}Np about 400.

The Norwegian studies show that the concentrations of ^{99}Tc are higher in the stipes than the blades of Cuvie (*Laminaria hyperborea*), which probably reflects a long-term retention in this part of the plant. There are apparent seasonal fluctuations, with higher activity in winter/spring and lower in sum-

mer/autumn. Factors involved may include plant growth patterns, seasonal changes in ocean circulation and short-term variations in actual ^{99}Tc discharges from Sellafield.

Injection tests carried out in Norway show that almost all the injected $^{99m}TcO_4^{--}$ was found in the spleen (52%) and kidney (40%) of rainbow trout, while small traces were found in the brain and the eyes. Preliminary results from a pilot study show a surprisingly low retention of Pu with the highest fractions being found in liver, gills and kidney.

Another study carried out in Norway shows that the ^{99}Tc concentrations are clearly higher in female lobsters compared to males.

Publication within the INDOFERN Project

Lindahl, P. Studies of Long-lived Radionuclides in the Environment with Emphasis on ^{99}Tc, ^{237}Np and Pu isotopes. Doctoral dissertation, Lund University, Department of Radiation Physics, 2003, 50 p.

Ikäheimonen, TK, Klemola, S., Ilus, E. Behaviour of gamma-emitters, Sr-90 and transuranic elements in forest environment. Submitted to Journal of Analytical and Bioanalytical Chemistry.