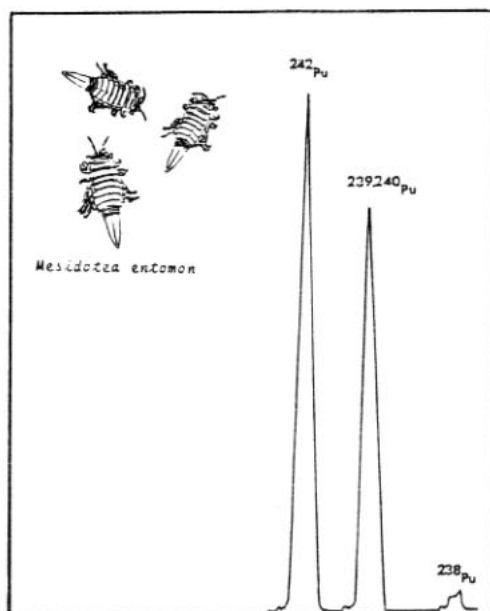


# The Sampling and Analysing Methods of Radionuclides used in The Nordic Countries for Environmental Samples



Edited by Tarja K. Taipale  
Finnish Centre for Radiation and  
Nuclear Safety

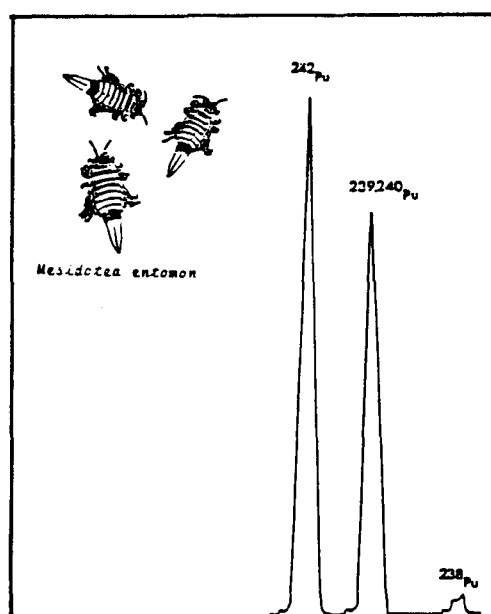
January 1985

This report forms a part of the program  
"Safety Research in Energy Production"

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**NKA** NORDIC LIASON COMMITTEE  
FOR ATOMIC ENERGY

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## SAMMANDRAG

Styrgruppen för radioekologi under det nordiska kontaktorganet för atomenergifrågor (NKA) har ansett det vara mycket viktigt att förbättra jämförbarheten för mätningar av omgivningsradioaktivitet i de nordiska länderna, då ett grundläggande krav för att utföra koordinerade forskningsprogram är resultatens jämförbarhet.

I detta syfte har flera undersökningar genomförts mellan laboratorier som mäter radioaktivitet i omgivningen. Dessa undersökningar har visat sig vara mycket nyttiga och har lett till en mera omfattande och systematisk överblick över den metodologi, som de nordiska laboratorierna använder för omgivningsmätningar.

Ytterligare har man ansett det nödvändigt att utarbeta rekommendationer eller till och med en överenskommelse om hur resultat bör presenteras för att göra en jämförelse av data lättare.

Denna rapport är baserad på enkätsvaren från de deltagande nordiska laboratorierna och sammanställd på strålsäkerhetscentralen i Helsingfors.

The participating nordic laboratories/  
De deltagande nordiska laboratorierna

Denmark/Danmark

Risø National Laboratory/Forsøgsanlaeg Risø  
Box 49, DK-4000  
Roskilde, DENMARK

National Institute of Radiation Hygiene/  
Statens Institut for Strålehygiene  
Frederikssundsvej 378  
Brønshøj, DK-2700  
DENMARK

Finland/Finland

Finnish Centre for Radiation and Nuclear Safety/  
Strålsäkerhetscentralen  
Box 268, SF-00101  
Helsinki, FINLAND

University of Helsinki, Department of Radiochemistry/  
Helsingfors Universitet, Radiokemiska institutionen  
Unioninkatu 35, SF-00170  
Helsinki, FINLAND

Norway/Norge

Institute for Energy Technology/Institutt for Energiteknik  
Box 40, N-2007  
Kjeller, NORWAY

National Institute of Radiation Hygiene/  
Statens Institutt for Strålehygiene  
Østerdalen 25, N-1345  
Østerås, NORWAY

Sweden/Sverige

National Institute of Radiation Protection/  
Statens Strålskyddinstitut  
Box 60204, S-10401  
Stockholm, SWEDEN

National Swedish Environment Protection Board/  
Statens Naturvårdsverk  
S-17011  
Drottningholm, SWEDEN

Studsvik Energiteknik AB  
S-61182  
Nyköping, SWEDEN

University of Lund, Radiation Physics Department/  
Lunds Universitet, Radiofysiska institutionen  
Lasarettet, S-22185  
Lund, SWEDEN

University of Uppsala, Radiobiological Institute/  
Sveriges Lantbruksuniversitetet,  
Institutionen för radioekologi  
Box 7031, S-75007  
Uppsala, SWEDEN

## CONTENTS

FOREWORD	1
SAMPLING	2
The purpose of the sample	2
Monitoring of the environment of the nuclear power plants	2
Monitoring of the global fall-out	3
Other surveying	3
Representativeness of sample	4
Selection of samples	4
Selection of sampling area	6
Selection of sampling frequency and time	8
Selection of sample size and sampling method	9
PRETREATMENT	10
Storage	10
Cleaning	11
Drying	11
Homogenizing and subsampling	12
Evaporation	12
Ion exchange	12
Dry and wet ashing	13
Hydrofluoric acid treatment and fusion	13
Leaching	14
DETAILED METHODS OF SAMPLING AND PRETREATMENT	14
Air	14
Deposition	15
Soil	17
Milk	18
Other foodstuffs	19
Terrestrial indicators	20
Sea water	21
Sediment and sedimenting material	22
Aquatic indicators	24

ANALYSING METHODS	25
γ-spectrometry	25
Preparation of the sample to the measuring geometry	25
Measuring	25
Analysing of spectrum	25
Calibration of detectors	26
Radiochemical determination of caesium	26
Methods for determination of strontium 89 and 90	27
Nitrate precipitation	27
Ion exchange	29
HDEHP-extraction	29
Methods for determination of transuranic elements	30
Determination of plutonium	30
Determination of americium and curium	31
Determination of tritium	33
Determination of radium 226	34
Determination of lead 210	35
Determination of polonium 210	35
TREATMENT AND REPORTING OF RESULTS	36
Recommendations	37
PAN-SCANDINAVIAN INTERCALIBRATIONS	38
Sediment intercalibration	38
Fucus intercalibration	39
Intercalibrations of detector systems for background radiation monitoring	39
Intercalibration of collecting and analysing of ground level air	40
Intercalibration of collecting and analysing of sedimenting material	40
REFERENCES	41



## FOREWORD

The Radioecology Group under the Nordic Liaison Committee for Atomic Energy has considered it to be of great importance to improve the comparability of environmental radioactivity measurements in the Nordic countries, a basic requirement for co-ordinated research programmes being comparability of results. In a case of emergency, good comparability between the results obtained will be required for mutual assistance.

Therefore several intercomparison exercises have been carried out between the laboratories measuring environmental radioactivity. The exercises have proved very useful and have led to a more comprehensive and systematic survey of the environmental measurement methodology used so far by the Nordic laboratories.

Furthermore it is considered necessary to make some recommendations or even to reach an agreement on how to present the results in order to make the comparison of, at least, monitoring data easier.

This report is based on the answers received from the participating laboratories to a questionnaire sent by the Finnish Centre for Radiation and Nuclear Safety, Helsinki. The laboratories are listed in an Annex to this report.

## SAMPLING

### The purpose of the sample

Monitoring in the environment of the nuclear power plants

The purpose of the sampling determines the types of samples, as well as the location and frequency of the sampling. The nuclear power plants or research reactors are monitored by studying the quantities and qualities of the radionuclides spreading in the environment. Some of the radionuclides can be analysed using direct measurements, but most of the environmental samples are analysed either chemically or radiochemically.

Environmental monitoring has several important aims:

- to supplement the discharge monitoring in order to verify the origin of radionuclides
- to contribute to the estimation of doses to the critical group to ensure adherence to the dose limits
- to determine the uptake of radionuclides by plants and animals for possible countermeasures in cases of unexpected release
- to contribute to a better understanding of the relationship between discharge and dose in the different dose pathways concerned
- to show the level and variation of environmental contamination and to provide basic data for informing the public.

The environment of nuclear power plants and research reactors is monitored in all Nordic countries.

## Monitoring of the global fall-out

The levels of different nuclides in the global fall-out and their transport pathways are also monitored using environmental samples. Generally, the global fall-out monitoring covers the whole country. Possible increases in levels can be observed and external and internal doses estimated. The results from the global fall-out monitoring programmes can be used for other purposes as well, e.g. to determine sedimentation rates. These samples are also used as reference samples for the nuclear power plant control.

The radionuclides most often studied are  $^{137}\text{Cs}$  and other long-lived  $\gamma$ -emitters,  $^{90}\text{Sr}$ , tritium and transuranic elements, especially  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$ . The global fall-out surveys are carried out in all Nordic countries.

## Other surveys

The results of the nationwide surveys and nuclear power plant monitoring programmes can be used for other research purposes as well. Transfer, sorption and accumulation of various elements and the effect of different environments on these phenomena are studied. The monitoring programmes are often supplemented by additional projects to obtain more detailed data.

The environments of planned uranium mines are studied by analysing natural radioactivity in various materials. This kind of work is done in Sweden, Finland and Greenland.

## Representativeness of sample<sup>1,2</sup>

### Selection of samples

The purpose of the sampling programme is to determine the types of samples. Some transport pathways of radionuclides are more significant than others.

Samples are selected according to some basic requirements. For the determination of short-lived radionuclides, samples which permit rapid measurement of nuclides, e.g. air and deposition, are selected. Long-lived radionuclides on the other hand are studied in the environment where they accumulate, e.g. in soil and sediment. It should also be ascertained that the sample type will be available during the entire research period which may include several years.

Air samples are collected for the monitoring of nuclear power plants and research reactors and of global fall-out. In the air, increased levels are quickest observable which makes air samples essential for the detection of short-lived radionuclides. The results are used for calculating inhalation and immersion doses. Air is also an important pathway for transporting and distributing radionuclides.

Deposition is collected in the environmental monitoring of nuclear power plants and global fall-out. It can also be used to detect increased activity levels. Results of activity measurements in deposition are used specially for external dose estimations and as a basis for calculating transfer factors of radionuclides.

Soil which is an important link in terrestrial food chains, reveals an accumulation of long-lived radionuclides. Soil samples are usually taken for the monitoring of global fall-out and nuclear power plants or reactors.

Milk and some other foodstuffs, e.g. meat, fish, cereals and vegetables, which are important parts of man's nutrition, are chosen for programmes monitoring fall-out and nuclear power plants. They contribute directly to the internal radiation dose to man.

Indicator organisms are collected for early detection of the accumulation of radionuclides in the biota. In the selection of the indicator organisms, the following features should be considered: occurrence, ease of sampling, endurance of the organisms under changing environmental conditions, and the marked accumulation ability of one or more radionuclides.

Grass and different kinds of moss and lichen are terrestrial indicators; slow-growing moss and lichen are integrative types of these indicators.

There are several aquatic organisms that can be used as indicators. In the Baltic Sea area, Fucus vesiculosus is a good indicator, because it exists everywhere except in the Bothnian Bay. It is also the most studied aquatic indicator in the Nordic countries, because it accumulates several trace metals and radionuclides. Other species of algae are also studied.

Mytilus edulis filters particles in seawater and can absorb radionuclides in particle form, e.g. transuranics. Macoma baltica is a special indicator in the Baltic Sea. It lives in the bottom sediment and is nourished by fresh sedimenting material. Mesidotea entomon, an interesting relict animal in the Baltic Sea, absorbs radionuclides effectively.

Sediment and sedimenting material are important research objects in the water ecosystem. Sedimenting material transfers and adsorbs radionuclides from the water phase. The absorption and accumulation of radionuclides in the bottom sediment are studied in sediment samples. Undisturbed sediment layers can be used to follow the development of the condition of the surrounding water area and possible migration of elements in the sediment.

Sea water in addition to air, is another important transfer agent of radionuclides. It is also an important research object, because all nuclear power plants in the Nordic countries are situated on the coast.

#### Selection of sampling area

The selection of the appropriate sampling area or point is important, each sample must represent the sampling time and area as well as possible.

In the monitoring programmes of nuclear power plants and research reactors, the sampling areas are selected to represent different distances to the reactor, being anything from hundreds of metres to tens of kilometres.

In the choice of the sampling point for example, wind conditions and especially the dominating direction of the wind are considered for monitoring air and deposition. In aquatic environments the knowledge of water currents is required.

In nation-wide surveying of the global fall-out the sampling areas are selected to represent the whole country. Measurements of samples then permit the fall-out level over the whole country to be evaluated.

The monitoring of nuclear power plants and reactors also requires the results of nation-wide monitoring programmes for comparison. When analysing a substance such as milk it is important to compare results from the vicinity of the reactor with the results of the nation-wide programme. Thus annual and seasonal variations in the fall-out level can be excluded from the releases from reactors.

The sizes of sampling areas vary depending on the type of sample. Rainwater samples are generally collected at the same fixed points by assuming that the composition of rainwater is constant in the area of interest. For plant samples, a more extensive area is chosen to ascertain a representative sample.

Air samples are primarily taken on the basis of meteorological conditions. The environment around a nuclear power plant or a reactor is monitored by selecting an area where the activity concentration in air can be supposed to be highest. The samples are usually taken where there is no interference from roads and other dusty place.

For the nation-wide global fall-out monitoring the most representative sampling sites are selected. When meteorological conditions vary greatly in different parts of the country, samples should be collected accordingly.

Rainwater collectors are usually placed according to the same principles as air collectors. Air concentrations usually show smaller variations than deposited activity which means that fewer air sampling stations are necessary than rainwater stations.

Soil samples are collected from undisturbed soil if possible. The collection area selected is generally flat and open terrain, without shadowing vegetation. The selection of sampling areas depends on the purpose of the soil sample. Smaller, exactly limited areas are selected for annual follow-up sampling. For an average areal concentration, a larger area is chosen to provide several parallel samples.

In milk sampling the use of larger dairies can make it difficult to find the area of production. When the area has been determined geographically, the proportion of milk of the total national production must also be known. This applies to other foodstuffs as well. Thus the foodstuff samples are collected from producers or wholesalers who know the production areas.

## Selection of sampling frequency and time

The frequency of the sampling also affects the representativeness of the samples. Some samples, e.g. air and deposition, are collected continuously. Samples are analysed daily, weekly, monthly or quarterly. Continuous sampling may sometimes be substituted by periodical collection of, say, milk samples in the environment of nuclear power plants or reactors. For example, milk samples are taken once a week for  $^{131}\text{I}$  determinations, and bulked weekly samples are analysed monthly for  $^{137}\text{Cs}$  and  $^{89}\text{Sr}$  and quarterly for  $^{90}\text{Sr}$ .

Cereals and other plant samples are mostly taken after harvesting, but quickly growing plants, such as grass, are collected several times during the summer. Meat samples are generally collected after the autumn slaughter and samples of game meat during the hunting season.

The prevailing conditions and their influence on the results are usually considered in the selection of sampling time. For example, in unfiltered sea water some radionuclide concentrations vary seasonally. Furthermore salinity, temperature and amounts of stable isotopes may have seasonal variations affecting the concentrations in aquatic samples. Also the concentrations of reactor-originating nuclides in sea water samples can vary significantly, depending on the sampling moment. If the sample happens to be taken just when the discharge plume is at the sampling site the concentrations of discharge nuclides will probably be high whereas on the next day they may be below the detection limit. These phenomena should be considered when results are compared.



### Selection of sample size and sampling method

Sample size depends essentially on the activity and ambition of the detection level and on such factors as the nuclide, the measuring equipment, and the need for parallel measurements. Samples with a high water content tend to be big, since there is little dry material.

Methods of sampling different materials also vary. Certain facts have to be taken into account when selecting the sampling methods.

Soil samples are usually collected to a depth of 25 to 50 cm, depending on the quality of the soil. The topmost layer, 0-5 cm, is sufficient for monitoring discharges from nuclear power plants. The upper layer is important, because most of the radioactivity is usually concentrated in that layer. Great care must be observed in sampling to ensure that undisturbed samples are taken.

The height of grass samples is taken into account. For instance when the food chain air-grass-cow-man is studied, the grass is cut at the same height as the cow bits it of.

In collecting vegetables, only the edible parts of the plant are used for analysis.

Sea water sampling is usually carried out at a depth of about 1 m when studying surface water and at about 1 m above the bottom when studying bottom water. Also the whole depth distribution of water can be measured. Samplers or plastic vessels are used for collecting. Sampling of near bottom water is done carefully, without mixing the bottom material.

Sediment is sampled with various gravity and box corers. The diameters of the samplers and the number of parallel samples taken for sufficient amounts of sediment material vary greatly. Samples are usually sliced into subsamples. It is important to take an undisturbed sample, without mixing the layers missing the uppermost layer of the sediment.

Sedimenting material is collected with traps, varying considerably in diameter and model but so that the ratio of the height to the diameter of the trap is more than 3 to avoid under or over trapping.

Seaweed sampling is carried out by divers, drags or directly by hands. When using drags, one cannot be sure of the quality of the sample. An advantage for the diver is that plants can be collected by hand, selecting them one by one at the sampling site. Various bottom samplers, crabs or dredges are used for sampling bottom animals.

The sampling methods of different environmental materials vary substantially. Each of the Nordic countries has its own methods for different sample types presented in connection with the different sample materials.

#### PRETREATMENT

The pretreatment methods vary significantly for each of the different materials and in each of the Nordic countries. There are however some general considerations for the pretreatment steps.

#### Storage

The samples must be stored without any risk of contamination, avoiding critical spoiling. The material of the containers for the liquid samples should be suitable for storing. Acid and carriers are added to water sample as soon as possible after sampling, avoiding adsorption of nuclides onto the walls of the containers. Before the sampling, acid is often added to the bottles placed in rainwater collectors. The longer the storing period the more important the acidifying of the water sample.

For short periods the biota and sediment samples can be stored in a refrigerator (+2-+4°C), whereas long-

term storing is better done by freezing. Several foodstuffs are also stored frozen. For short periods milk is usually stored in a refrigerator and occasionally a preservative e.g. sodium azide is added to prevent it from turning sour.

Several samples are stored dried or ashed, thus dry storage is needed.

### Cleaning

Cleaning of the sample from other materials involves pretreatment. For example, plant samples are freed of mould, parts of other plants and earth. Dead parts of the plant are also removed. Sea water samples can be filtered if the activity or amount of particles is measured separately. Soil samples are freed of plant roots etc.

The cleanliness of the collectors and containers may also affect the final result. When collecting deposition for strontium and caesium analyses, the collectors have to be rinsed thoroughly for otherwise the absorbed dry deposition on the surface is lost. It is important that the collectors are carefully cleaned for the next sampling period.

### Drying

Drying increases the amount of solid material in samples and permits longer storage. Before analysis solid samples are usually dried either in an oven or at room temperature<sup>3</sup>. It is important to prevent contamination during the drying procedure.

Samples are dried carefully and for a sufficiently long time at a fixed temperature to a constant weight. Measuring of both fresh and dry weight is required for the reporting of results.

Freeze-drying is a simple and reliable method that renders most samples easy to handle.

#### Homogenizing and subsampling

Homogenizing of the sample before subsampling for analysis is very important because dried samples contain grains or particles of different sizes. It is generally done after drying (or ashing). The subsampling is done carefully with a suitable apparatus or by hand. In spite of homogenizing, the subsample does not always represent the entire sample. Some nuclides may fasten or be adsorbed in different ways onto fine-grained and coarse-grained parts of materials.

#### Evaporation

Evaporation is the most commonly used concentrating pretreatment of liquid samples. Excessive evaporation temperature can cause the sample to boil and scatter. The evaporation bowl has to be made of such material that the evaporating material does not absorb into it. The walls are washed carefully after evaporation. Spectrometric measurements are often done directly from dried liquid material without measuring the yield, assuming it to be 100%.

#### Ion exchange

In some countries ion exchange is used in the collection of deposition. For these resins, the correct grain size and the method of regeneration are of great importance. The amount of resin also affects the adsorption of nuclides. It is essential that the nuclides studied are totally adsorbed in the resin, because no

yield determination is done<sup>4</sup>.

#### Dry and wet ashing

Solid samples are usually dry or wet ashed before radiochemical separation. The temperatures for dry ashing vary, most commonly being 450°C. The ashing time depends on the material. Dry ashing is only used for determining nuclides which do not evaporate at the temperature used. Even at 450°C sulphur and phosphorus can cause material to glow, resulting locally higher temperatures and loss of some nuclides. Carriers or tracers should be added before ashing to ensure that they are in the same chemical form as the nuclide to be analysed. However, they are generally added after dry ashing. Measuring of the ash weight is important for calculating results.

If dry ashing is not suitable the sample can be wet ashed. Carriers and tracers are usually added before a wet ashing procedure.

#### Hydrofluoric acid treatment and fusion

Hydrofluoric acid treatment may be necessary for some samples with a high content of silicates. With some biota samples, this is done after wet ashing before separation of plutonium and americium.

Samples difficult to melt or dissolve are fused. In strontium analysis, carbonate fusion is done for insoluble parts of rainwater and plant samples. Sodium hydroxide - sodium nitrate fusion is necessary for some air filters.

### Leaching

Acid leaching is done for soil and sediment samples. For transuranium analysis, combined wet ashing and acid leaching can compensate for dry ashing.

For strontium analysis in soil and sediment, leaching dissolves biologically available radionuclides for the sample. To ensure that the results are comparable the leaching period has to be long enough and the same for all samples of the same type.

## DETAILED METHODS OF SAMPLING AND PRETREATMENT

### Air

Air samples are collected for monitoring artificial radioactivity in the air near nuclear power plants and reactors. Global fall-out is also studied. The sampling is done for other purposes as well, e.g. as at the University of Lund in Sweden, where radon daughters are monitored in the environment of planned uranium mining sites.

The purpose of the sampling affects the choice of sampling site.

The selection of the type of sampler is principally a technical procedure. The amount of air is a parameter which affects the minimum detectable activity and varies between laboratories from  $300 \text{ m}^3 \text{ d}^{-1}$  to  $156000 \text{ m}^3 \text{ d}^{-1}$ .

Air samples are collected on glass fiber or on coal-impregnated glass fiber filters or on both successively. If the sample is ashed for analysis a paper filter must be used. Sampling time varies from one day to two weeks, being mostly one week. The height of the sampling point varies from one to three metres from the ground. In one laboratory the sampling point is on the roof of the laboratory, about 15 m above ground.

From air filters,  $\gamma$ -emitters are determined in all laboratories. Also,  $^{90}\text{Sr}$  is analyzed monthly or

quarterly on bulked samples. In one laboratory, total- $\alpha$  and - $\beta$  are also determined.

Pretreatment of filters is done in different ways. For total- $\alpha$  and - $\beta$  measuring no pretreatment is done. Filters are normally compressed to provide suitable counting geometry for radionuclide analysis by gamma-spectrometry. Paper filters are ashed before  $^{90}\text{Sr}$  analysis. Combined glass fiber filters representing the desired time period are ground and homogenized before subsampling for the  $^{90}\text{Sr}$  analyses. Filters are fused with sodium hydroxide and sodium nitrate before proper strontium separation.

Table I shows the data on the collectors and sampling and pretreatment methods used in the different laboratories.

Differences in sampling height and pretreatment methods can affect the comparability of results from different countries. The detection of short-lived  $\gamma$ -nuclides depends on the length of the sampling period and the interval between sampling and measurement.

### Deposition

Deposition samples are collected in all Nordic countries. The samples are used for monitoring both nuclear power plants or reactors and the global fallout level in the country concerned. Samples are also collected for special purposes, such as dose and model calculations. Also the environments of the planned uranium mining sites are studied.

Dry and wet deposits are collected together into open collectors.

Collectors vary from country to country. Their areas vary from  $0.05 \text{ m}^2$  to  $10 \text{ m}^2$ . This affects the detection limit. The material is either stainless steel, plastic coated glass-fiber or brass. The last is used for tritium sampling. The height of the collector from the ground varies from 1 m to 2.5 m.

Ion exchange resin is used as a sampling method in three laboratories. Both anion and cation exchange resins are used in the columns. Thus handling of large amounts of rainwater can be avoided.

The sampling period of deposition is usually one month. Ion exchangers are changed once a week and collectors are washed and cleaned monthly. Samples are analysed monthly or as combined samples of several months, e.g. quarterly.

$\gamma$ -nuclides are determined in all Nordic countries.  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ , total- $\alpha$  and total- $\beta$  are also determined. Tritium is measured from deposition in one laboratory and transuranic elements in another.

Rinsing and washing of the collectors are done in the field at the end of the sampling period. Collectors are rinsed with either acidic solution or distilled water or with both, adding the washings to the sample. Collectors for tritium samples are washed without taking the washing with. Carriers are added either before collecting into the collecting vessel or in the laboratory into the sample. No carrier is added to the tritium samples. Ion exchange resins are not usually pretreated in the field.

Rainwater samples are evaporated in the laboratories. In some laboratories  $\gamma$ -nuclides and possibly total- $\alpha$ , total- $\beta$  and  $^{89,90}\text{Sr}$  are measured or analysed directly from evaporated samples; in others, evaporated samples are ashed before the measurements. Ashing temperatures vary from 300 °C to 450 °C. The activity of ion exchange resin is measured directly or from the ashed sample as well. Tritium samples are pretreated by distilling.

Table II summarizes the various sampling and pretreatment procedures for rainwater.

Differences in sampling and pretreatment methods may affect the comparability of results between laboratories. These differences are probably caused by the types of collectors, the use of ion exchangers, and the different ways of rinsings and washings of collectors



and of ashing samples.

### Soil

Soil samples are collected for studying the transport of radionuclides, special attention being given to the slow accumulation of long-lived fall-out nuclides. The soil in the environments of nuclear power plants is also monitored. Moreover soil samples are analysed for radioecological survey of the transfer of radionuclides from soil to plants and for the effect different soils have on this transport. Natural radioactivity in the soil used for building materials is also studied.

The sampling areas are selected according to the purpose of the study. The sampling depth varies from 0 to 25 cm or to 50 cm. When monitoring nuclear power plants only, the topmost 0-5 cm layer of the soil is sufficient. The collecting is carried out with either a spade or a metallic auger. The diameter of the sampling auger varies from 6.5 cm to 7 cm and the size of the sampling area from 16 to several hundred square metres. The purpose of the sample affects the selection of the sampling area.

The sampling frequency of repeated collections varies from once a year to every five years. The sample is subsampled vertically into several slices, for example, 0-1 cm, 1-2 cm, or 0-5 cm, 5-10 cm.

The nuclides to be studied depend on the purpose of the sample.  $\gamma$ -emitters and specially  $^{137}\text{Cs}$  are measured in monitoring programmes. In some laboratories  $^{90}\text{Sr}$  and transuranic elements are also determined. Nuclides of natural series, particularly isotopes of radium, thorium and uranium, are measured for control of the natural radioactivity. In one laboratory the extractable parts of these nuclides are studied.

The pretreatment methods vary in the Nordic laboratories. Usually, samples are first freed of stuffs

and parts of plants and then weighed and dried. Drying temperatures vary from 20 to 105°C. Different methods are used in homogenizing the samples; in some laboratories, samples are milled and in the others sifted.  $\gamma$ -nuclides are usually measured from dried samples. The soil sample is ashed and leached before strontium analysis. Additionally, plutonium, uranium and radium are measured from the ashed sample in several laboratories, the temperature of the ashing varying from 450 to 550°C. In one laboratory, transuranium analysis is done from dried, wet ashed and leached samples.

Table III shows the summary of the soil sampling and pretreatment methods used.

The different pretreatment methods of the samples can affect the comparability of the results considerably. Factors of this kind can be the homogenizing, the temperature and the modes of ashing and the leaching time.

### Milk

Milk is an important foodstuff causing internal radiation dose of  $^{90}\text{Sr}$ ,  $^{131}\text{I}$  and  $^{137}\text{Cs}$  to man in the Nordic countries.

Milk samples are analysed in all Nordic countries. The most common purpose is monitoring of the nuclear power plants. Milk analyses are used for dose and model calculations as well.

Milk samples are collected from farms near nuclear power plants or reactors. Reference and fall-out samples are collected from either farms, dairies or transport tanks. Dry milk samples are taken from factories.

The frequency of sampling depends on the nuclides studied. For  $^{131}\text{I}$  analyses milk is usually taken once a week. Iodine is measured either directly from milk or by the ion exchange method.  $\gamma$ -nuclides especially  $^{137}\text{Cs}$ , and  $^{89,90}\text{Sr}$  are analysed once a month. For some purposes milk samples are taken quarterly. In one laborato-

ry uranium and radium are also analysed from some samples.

Volumes of samples vary from two to eight litres, 1-2 kg of dry milk is taken for analyses.

Pretreatment methods vary in the different laboratories. In two laboratories,  $\gamma$ -emitters are measured directly. In other laboratories, milk is ashed before analysing, the ashing temperatures being from 450°C to 490°C.

Table IV gives data on the milk sampling and pretreatment used in the Nordic laboratories.

Different modes of pretreatment can influence the comparability of the results and some radionuclides may be lost during evaporation and ashing, especially in studies on fresh fall-out.

#### Other foodstuffs

Radioactivity of foodstuffs other than milk is also studied. Meat, fish and cereals are the most analysed. Furthermore, eggs, roots and other vegetables as well as fruit and berries are collected.

Samples are collected for monitoring nuclear power plants and for other purposes as well, to follow the transfer of long-lived nuclides and doses and to make model calculations. Some foodstuffs, e.g. lettuce and elk meat, are used partly as indicators.

Sampling is carried out at private farms, central stores and abattoirs. Samples are usually taken once a year in connection with harvest; meat samples are taken once or twice a year, except reindeer meat samples, which are taken less frequently at present. Fish are obtained from local fishermen or research stations.

The pretreatment methods depend on the sample. Usually, the samples (especially meat and fish) are frozen first. The samples are cleaned, the edible part being retained for analysing. In some laboratories,  $\gamma$ -measurements are made from dried sample, especially

from plants, and in others from ashed sample. The dried samples are ashed at temperatures ranging from 450°C to 550°C.

$\gamma$ -nuclides, especially  $^{137}\text{Cs}$ , are analysed from all foodstuffs and  $^{239,240}\text{Pu}$  from some sample types. The  $^{90}\text{Sr}$  is determined in some foodstuffs by some laboratories. Table V summarizes the sampling and pretreatment methods of foodstuffs except fish used by the Nordic laboratories.

#### Terrestrial indicators

Terrestrial indicator samples are collected for monitoring the environments of nuclear power plants and uranium deposits. Grass is selected as an indicator sample and also because of its importance as a link in food chains. Various mosses and lichens are studied in the monitoring of nuclear power plants and for investigating food chains. Different species of lichen and moss are collected for monitoring uranium deposits.

In summer the sampling frequency varies from once to four times a year. Grass is usually collected at the same farms as the milk samples for studies on the air-grass-cow-man food chain. The sampling area varies from 0.25 to 8 m<sup>2</sup>. Normally moss and lichen are collected from areas of occurrence near the environments of nuclear power plants or uranium deposits. The samples weigh a few kilograms.

Pretreatment methods depend on the nuclides studied and vary in different laboratories.  $^{131}\text{I}$  can be measured directly from fresh or dried grass. In some laboratories the  $\gamma$ -nuclides are measured from ashed samples and in the others from dried samples. The ashing temperatures vary from 450°C to 550°C.  $^{89,90}\text{Sr}$  and transuranic elements are also analysed. U, Ra, Po and Pb isotopes are determined for the monitoring of uranium deposits.  $^{55}\text{Fe}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  are also determined in the investigation of

food chains.

Table VI summarizes the different indicator organisms as well as the sampling and pretreatment methods used by the laboratories.

The differences between laboratories in sampling and pretreatment may influence the comparability of the results.

### Sea water

Sea water is an important transfer agent for radio-nuclides in the environment.

Sea water samples are collected for monitoring the environments of nuclear power plants. Following the radioactive contamination of the Baltic Sea, the Danish Straits, the North Sea and the North Atlantic is also performed. Separate projects, such as coordinated research programme of the IAEA "Study of Radioactive Materials in the Baltic Sea" have been conducted.

Depending on the purpose, the sampling frequency varies from once to several times a year.

The sampling areas are also chosen according to the purpose. In the monitoring programmes of the nuclear power plants most of the water samples are collected near the coast. In studies of the condition of the sea area, the samples are taken over the whole of the area studied.

Both surface and near bottom waters are analysed.

The samples are taken either into a vessel direct or using a Ruttner-sampler. Pumping and filtering is used to study the amounts of particles and the distribution between particulate and the soluble forms. Near-bottom water is usually taken from one metre above the bottom. The volume of the sample varies from one litre to about two thousand litres depending on which nuclides are to be determined.

Y-nuclides are usually determined from all sea

water samples. Several nuclides, e.g. strontium, tritium, plutonium, americium, thorium, uranium, neptunium, polonium, technetium and lead are determined from sea water samples. Some of these nuclides are also studied from filtered water and from particles.

The pretreatment method depends on the nuclides studied.  $\gamma$ -nuclides are measured from evaporated samples or from sea water samples direct. Large samples are commonly pretreated by precipitation; AMP precipitation is usually done for caesium determination, and hydroxide precipitation for transuranics. Analyses are also done on evaporated samples. Samples for measuring tritium are first distilled.

All determinations can be done directly from unfiltered water. For some purposes such as the determination of the amount of the particles, the water samples can be filtered through 0.45 or the 0.3  $\mu\text{m}$  filters.

Table VII summarizes the sampling and pretreatment methods for sea water samples.

#### Sediment and sedimenting material

Sediment and sedimenting material are important research objects in the aquatic ecosystem. They are collected for the monitoring of the nuclear power plants. Additionally sediment samples are collected for monitoring global fall-out levels. Sediment is an important link in the studies of aquatic food chains. The age of sediment and the distribution of radionuclides in sediment are also investigated.

The sampling areas are selected according to the purpose of sampling. Large sedimentation basins of the sea area are chosen for sampling points, in studies of vertical distribution and levels of long-lived radionuclides. Additionally sediments of some lakes are studied. Sediment samples are usually taken once a year. During the open water period sedimenting material is collected

continuously at several points near the nuclear power plants. Special monitoring in the Biotest basin in Forsmark, Sweden, is performed. Sedimenting material is usually collected at a depth of one metre above the bottom; also depths of 5 and 10 metres from the surface are used. Before the measurements, the samples are combined into three-month samples.

Different apparatuses are used for collecting sediment. Usually, samples can be sliced into 1-5 cm subsamples.

Sedimenting material is collected in traps of various kind. The models and areas of sampling tubes vary greatly.

$\gamma$ -nuclides are measured from most samples, especially the  $^{137}\text{Cs}$  content is followed.  $^{90}\text{Sr}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  are also determined from some samples.

Before  $\gamma$ -measuring the samples are dried. Frozen samples can be dried with a vacuum freeze-drier. This makes the samples easier to handle. Samples are ashed and acid leached for strontium analysis. For plutonium and americium measurements dried samples are ashed or wet ashed.

Table VIII summarizes methods of sampling sediment and sedimenting material, as well as the pretreatment methods.

There are several factors, which may affect the comparability of the results. It is of utmost importance to keep the sample undisturbed during sampling. Mixing the sediment layers or missing the uppermost layer with a high water content leads to incorrect vertical distribution or/and total contents of the radionuclides. Different apparatuses with various diameters may also affect results.

### Aquatic indicators

Aquatic indicator organisms are useful monitoring objects when controlling environments of the nuclear power plants. They are also collected for studying fall-out levels in an aquatic environment.

Radioactivity measurements of biota samples are included also in the coordinated IAEA programme. In 1980-82 a Joint Nordic Fucus Project was carried out in Nordic countries to compare the contents of radionuclides in different sea areas.

The sampling areas are chosen according to the purpose of the study and the occurrence of the indicator species. Fucus vesiculosus is the most often studied species of algae in the Nordic countries. Additionally other species and bottom animals are studied. The sampling methods, diving, fishpots and several kinds of bottom samplers, depend on the sampling objects.

$\gamma$ -nuclides are usually determined from aquatic indicators. The short-lived discharge nuclides are of special interest in the vicinities of nuclear power plants.  $^{89,90}\text{Sr}$  is also analysed from most samples, plutonium, americium and technetium from some.

Bottom animal samples are preserved frozen. They are dried in an oven or in a freeze-drier, and ground and homogenized before  $\gamma$ -counting. Strontium analysis is preceded by ashing and transuranic analysis by ashing or wet ashing and acid leaching. Hydrofluoric acid treatment is necessary for some algae species, especially for the determination of americium.

Table IX summarizes the sampling and pretreatment methods of aquatic indicators.



## ANALYSING METHODS

### γ-spectrometry

γ-spectrometry is used by all Nordic laboratories.

#### Preparation of the sample for the measuring geometry

Solid samples are generally dried, ground and homogenized before measurements. In some laboratories chemical separation is carried out to concentrate and clean the samples. Liquid samples are evaporated to a smaller volume or to dryness; some samples are also ashed before γ-measurement.

The measuring geometry depends, among other things on the ambition for the detection limit, the amount of sample available for analysis, and the effectivity of pre-concentration. Different boxes and Marinelli beakers are generally used as measuring geometries; some samples are pressed into discs.

#### Measuring

The detectors most commonly used are Ge (Li) and high purity Ge or NaI detectors. The samples are measured in a background shield to reduce the background and are usually placed directly on the detector to increase the efficiency. Measuring times vary according to samples and nuclides.

#### Analysing of spectrum

The gamma spectra are almost always analysed by computer using the programs developed for this purpose. The programs generally contain partial factors like

- searching of peaks
- calculating of total peak area either according to manual calculation or applying

- adapting of energy and effectivity calibrations to the spectrum
- identifying of the nuclides in the spectrum
- making the half-life correction of the nuclide
- making background correction
- taking account of self absorption in the sample (some)
- making the coincidence correction (true coincidence, some)
- reporting of nuclide concentrations and the error estimations.

#### Calibration of detectors

The detectors are almost always calibrated for the same geometry as the measurement. Water is the most common basic matrix, but other matrices with a higher or a lower density are also used.

#### Radiochemical determination of caesium

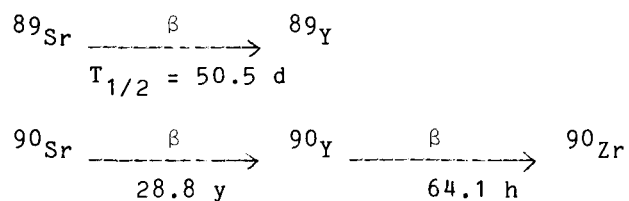
In one laboratory,  $^{137}\text{Cs}$  is measured after a radiochemical separation, which is used for rainwater, ashed grass and some milk samples.

After addition of the carrier, EDTA-solution is added to the dissolved sample. Caesium is separated from strontium by a cation exchange step, in which caesium is eluted with diluted hydrochloric acid before strontium elution. Caesium is precipitated as phosphomolybdate and purified by molybdate precipitation. After carbonate precipitation caesium is precipitated as perchlorate and the  $\beta$ -activity of the sample is measured with an  $\beta$ -analyser.

For fresh milk the above procedure is preceded by cation exchange.

The scheme of the analysis is presented in Figure 1.

Methods for determination of strontium 89 and 90



The amounts of  ${}^{89}\text{Sr}$  and  ${}^{90}\text{Sr}$  can be determined by measuring the combined amount of these nuclides and by subsequent separating of  ${}^{90}\text{Sr}$  and measuring its daughter nuclide  ${}^{90}\text{Y}$ . Strontium has to be separated by a chemical procedure from calcium and other matrices in environmental samples as well as from other radioisotopes.

Three different methods have been used in seven Nordic laboratories.

#### Nitrate precipitation

The most common method for separating strontium is the nitrate precipitation <sup>5-8</sup>. Pretreatment with drying or ashing is followed by different precipitations depending on the material; phosphate precipitation for milk samples, oxalate precipitation for sediment, soil, plant and rainwater samples, and both for sea water samples. The treatment methods, however, differ in the different laboratories.

Figure 2 shows the scheme for analysing soil and sediment samples.

After the first precipitations earth alkaline metals are isolated as carbonates (except in milk). Strontium can be separated from calcium and partly from other interfering ions by precipitating it as nitrate several times with 70-77 % nitric acid. Barium, radium and lead can be separated by precipitating them as chromates. Thorium, daughters of radium and rare earth metals, particularly yttrium, are removed by ferric hydroxide

scavenging. Strontium is then precipitated as carbonate.  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  can be measured from this precipitation, and calculated after  $^{90}\text{Sr}$  separation. Yttrium carrier is added to the diluted sample, and the solution is allowed to stand for at least fourteen days after ferric hydroxide precipitation.<sup>a</sup>  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  reach equilibrium during this period.

Yttrium is purified from strontium with hydroxide precipitation and precipitated after that as oxalate. The amount of  $^{90}\text{Sr}$  is determined by measuring its daughter nuclide  $^{90}\text{Y}$ . Yttrium oxalate is measured with a low background  $\beta$ -counter. It can also be measured as oxide after ignition.

After yttrium precipitation, the yield of strontium is measured from the solution; this can be done spectrometrically. The yttrium yield is measured from hydroxide or oxalate precipitation either titrimetrically or, after ignition to  $\text{Y}_2\text{O}_3$ , gravimetrically. The yield in Sr analysis can also be determined by  $^{85}\text{Sr}$  spike. The amount of stable strontium, which is determined separately, has to be taken into account in the yield measurement e.g. in grass.

With some modifications, this method, used originally by AERE, can be applied to all kinds of environmental samples and foodstuffs. The chemical yields vary, depending on the material. Strontium separates very well from calcium and other interferences.

Although time-consuming the method is reliable and safe for routine use. The reproducibility of the chemical analysis is about 5 %. The statistical error of  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  results depends on the activity of the sample, the error of  $^{89}\text{Sr}$  result being always greater than that of  $^{90}\text{Sr}$ .

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<sup>a</sup> The waiting time for  $^{90}\text{Sr}$ - $^{90}\text{Y}$  equilibrium to be reached (18 days) is not needed if less accurate results are adequate. A 10 % equilibrium can be reached in about one day.

### Ion exchange

One laboratory uses ion exchange method whereby strontium is first separated from calcium with a cation exchange step. Caesium is also removed from strontium with cation exchange in EDTA. Caesium can be eluted with diluted hydrochloric acid, strontium, which is retained in the resin, can be eluted with a stronger acid. Figure 3 shows the scheme of the method. Strontium is purified with chromate precipitation as in the nitrate precipitation method, and the analysis is continued according to it.

This method permits only ten day's waiting time for equilibrium, which has to be taken into account when calculating the results. This period, however, is too short, for it takes at least 14 days before the balance exists.

### HDEHP extraction

The third method used for strontium determination is the HDEHP extraction, used in one laboratory<sup>9,10,11</sup>. The methods for fresh and old fall-out differ slightly. The scheme of milk analysis for fresh fall-out nuclides is shown in Figure 4. After pretreatment, drying and ashing, the sample is diluted into 1M hydrochloric acid and extracted with HDEHP solution in the pH range 1.0 - 1.1. Strontium is retained in the water phase and yttrium is extracted to the organic phase. After addition of the yttrium-carrier and waiting for a balance between  $^{90}\text{Y}$  and  $^{90}\text{Sr}$  a second extraction is done with HDEHP. Calcium and other ions with an oxidation state of +2 are retained in the water phase. Yttrium is back-extracted with nitric acid, precipitated as oxalate and measured. The yield of strontium is measured from the calcium yield, which is determined gravimetrically.

The first extraction step is not required for old fall-out.

The detection limit for yttrium is 7.4 mBq/l per

600 ml milk.

This HDEHP method is shorter and thus quicker than the nitrate precipitation method in routine use. The intercalibration results have been good;  $^{89}\text{Sr}$  cannot be determined with this method, however.

The method has also been modified to permit rapid analyses (Fig. 5.)<sup>12</sup>. These are done without the waiting time and yttrium is analysed as in old fall-out samples. Strontium is precipitated as phosphate and purified with two nitrate precipitations. Then it is precipitated as carbonate and measured with a  $\beta$ -counter. The detection limit for this rapid method is 0.19 Bq l<sup>-1</sup> for  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  and 0.074 Bq l<sup>-1</sup> for yttrium per 50 ml milk sample.

#### Methods for determination of transuranic elements

##### Determination of plutonium

Ion exchange is the most commonly used method for plutonium separation<sup>13-20</sup>. It is used by five laboratories.

The isotopes studied are  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ . First, the sample is ashed or wet ashed and acid leached. For sea water samples, several hydroxide precipitations are done after evaporation or from the samples direct. After pretreatment, plutonium is oxidized to the +4 oxidation state. The oxidants used are perhydrol and sodium nitrite or hydroxylaminhydrochlorid. Anion exchange is done in either a nitrate or a chloride form. Washing is done with nitric acid and hydrochloric acid so that the heavy metals, uranium and thorium, can be removed. The first fraction from the exchange is collected for americium and curium analysis if needed. Plutonium is eluted by reducing to the +3 oxidation state; the reducing agents most commonly used are hydrochloric acid and ammonium iodide. Plutonium is electrodeposited

onto a stainless steel disc and measured with  $\alpha$ -spectrometry. The laboratories use different electrodeposition methods. Deposition can be done from diluted nitric acid or from sulphuric acid. Yields from electrodepositions are usually good.  $^{242}\text{Pu}$  tracer is added for the determination of chemical yield. Figure 6 shows the scheme for Pu analysis.

This method works well and gives pure spectra. Increasing of washing volumes and decreasing of elution rate can improve the separation. The ion exchange method is suitable for all kinds of samples: water, soil, sediment, plants, seaweeds and mussels, only the pretreatment stage differs. Intercalibrations have given good results.

The extraction method is used in one laboratory<sup>21</sup>. Samples are ashed and treated with acids. Figure 7 shows the scheme for determination. Dilution is followed by the lanthan fluoride precipitation. Plutonium is extracted into TTA from  $\text{Al}(\text{NO}_3)_3$  solution and then into 9M hydrochloric acid. During extraction  $\text{Pu}^{4+}$  separates from trivalent lanthanides and actinides. Uranium is separated with fluoride precipitation, ferric ions are separated with ether extraction. Plutonium is then electrodeposited.

The analysing methods differ from each other as regards the time when tracer is added. This can influence the final result. Further, the chemical state of the tracer added and of the plutonium in the sample have to be similar.

#### Determination of americium and curium

Three laboratories have reported their analysing methods for americium 241 and curium 242 and 244.

The HDEHP-extraction method is used in two laboratories<sup>20,22,23</sup>. The analysis continues immediately after plutonium separation. Both methods are very similar and are based on HDEHP extraction and several ion exchange steps (Fig. 8).

After plutonium ion exchange americium is in nitric acid or hydrochloric acid solution. Oxalate precipitation is done in one of the laboratories because of the high calcium and phosphate content in sediment and sea water. Calcium is separated with ferric hydroxide precipitation and ferric-ions with anion exchange. The most part of thorium and lanthanides can be eliminated with HDEHP extraction. The rest of the thorium and plutonium can be removed with cation and anion exchanges. The final purification is done using anion exchange resin in a nitric acid-methanol form. The remaining lanthanides can be separated with rodanide washing, which leaves americium and curium in the resin as complex ions. Two anion exchanges are required for soil and sediment samples. Americium and curium are electrodeposited in the same way as plutonium.

This method takes a long time, but it is suitable for soil, sediment, biota and sea water. Chemical yields have been 40 - 70% for sea water and 60 - 90% for the other samples. The detection limit depends on the measuring time, and is e.g.  $0.37 \times 10^{-3}$  mBq l<sup>-1</sup> for 100 l sea water sample for the measuring time of 7 - 10 days. The intercalibration results have been good.

Ion exchange method is used in one laboratory<sup>22-26</sup>. Analysis continues from plutonium separation. Calcium oxalate precipitation is done immediately after the pretreatment and before the plutonium separation. There are several ion exchange steps, including anionic and cationic steps. The separation of americium is based on elution with rodanide ions in cationic ion exchange resin. Purification from lanthanides and thorium is done with anion exchange and with rodanide ion solution as well. Americium is electrodeposited from diluted sulphuric acid at pH 2. Figure 9 shows the scheme for determination.

Several parallel determinations have given good results.



### Determination of tritium

Tritium is analysed from water samples; it is measured with liquid scintillation counting either immediately after distillation or after electrolytic enrichment.

The electrolytic enrichment method is illustrated in Figure 10<sup>27,28</sup>. The sample for tritium measurement is first distilled to dryness, after which 100 ml of each sample are taken for electrolysis. Sodium peroxide is added into the electrolysis cell for alkaline electrolysis. Iron and nickel are used for the cathode and the anode. The electrolysis is carried out at a temperature of +10°C first at a current of 3.0 amperes for three days and then at 0.3 amperes for seven days. The remaining volume about 3-5 ml is determined exactly. To prevent large amounts of hydrogen from binding in the sodium hydroxide, the enriched sample is neutralized with carbon dioxide to sodium carbonate. The sample is then distilled under reduced pressure at a low temperature in a dry ice-ethanol bath. Liquid scintillation counting is performed with dioxane-based scintillation solution. Calibration is carried out using the external standard channel-ratio method.

The detection limit in this method is 0.9 kBq m<sup>-3</sup> with an enrichment factor of 14 and a counting time of 200 minutes. The reproducibility error of the whole analysis ( $\pm 1\sigma$ ) is 10-20%.

The direct liquid-scintillation method is used for water samples if only a rough estimation of the tritium level is desired. First the sample is distilled to dryness. The water and scintillation solution are mixed at a ratio of 1:1 to obtain a homogeneous gel. The sample is measured with a low background liquid-scintillation counter. The calibration is done as in the electrolytic enrichment method.

The detection limit in this method is only about 7 kBq m<sup>-3</sup> and the reproducibility only 20-30% due to the great influence of the background fluctuations.

The method is very quick and can be used for monitoring tritium levels in the environment of nuclear power plants and reactors.

#### Determination of radium 226

Radium 226 determination is done for environmental samples in monitoring the environment of uranium deposits<sup>29</sup>. It is also determined when estimating internal doses from foodstuffs.

The size of the sample analysed is one litre of water or 5- 10 g of plant ash. The Ba-carrier is added and the ash is dissolved in hydrochloric acid. The hydroxide-carbonate fusion is done to the residue. After dissolution, the Pb-carrier is added. To the water sample, both Ba- and Pb-carriers are added directly. Radium, barium and lead are precipitated as sulphates. The precipitation is purified from polonium by washing with concentrated nitric acid. It is then dissolved in alkaline EDTA solution and coprecipitated with barium sulphate by adjusting the pH to 4.5. This slightly acidic solution keeps naturally occurring alpha emitters and lead carrier in solution as complex edetate ions, the barium complex ion is destroyed and barium-radium sulphate precipitate is formed. The time the precipitation takes must be noted. After washing with water the precipitate is centrifuged on a weighed stainless steel planchette, which is then dried and weighed. The alpha activity of precipitated radium and its daughter are measured. Figure 11 shows the scheme for the method.

The chemical yield is determined gravimetrically from the added barium carrier or with a  $^{133}\text{Ba}$  spike. The method is suitable for milk, foodstuffs and water.

If there is a lot of barium in the samples, e.g. in some seaweeds, the gravimetric yield determination is not suitable. In these cases, a tracer has to be used or the stable barium content in the sample has to be determined.

### Determination of lead-210

$^{210}\text{Pb}$  is determined from plant, foodstuff, water and sediment samples. The method is used to determine  $^{210}\text{Pb}$  indirectly through its high energy beta-emitting daughter  $^{210}\text{Bi}$  (1.2 MeV,  $T_{1/2} = 5\text{d}$ )<sup>30</sup>. After ashing the sample is stored for 30 days, during which time  $^{210}\text{Bi}$  reaches equilibrium with  $^{210}\text{Pb}$ . The ash is digested using both nitric acid and hydrochloric acid.  $^{210}\text{Bi}$  is isolated using DDTC (diethylammonium diethyldithiocarbamate) extraction in chloroform. Copper and iron can interfere with the extraction. After destroying the organic matter with nitric acid, hydroxide precipitation is done at pH 8. Bismuth is then precipitated as bismuth oxychloride. The precipitation procedure is specific for bismuth. The precipitate is filtered, covered with aluminium foil and counted in a low-background beta analyser. Figure 12 shows the scheme of the analysis.

The recovery of bismuth is calculated using the weight of bismuth in the final  $\text{BiOCl}$  precipitate and the amount of Bi-carrier added before separation.

The detection limit of the method is about 37 mBq/sample with a background lower than 1 cpm and a standard error ( $\pm 1\sigma$ ) about 10%.

The results of analyses have been reproducible.

### Determination of polonium-210

Polonium-210 is analysed from water, sediment and some indicator organisms in the environment of planned uranium mines<sup>31,32</sup>. It is also analysed from sediment to determine the ratio  $^{210}\text{Pb}$ - $^{210}\text{Po}$  for dating measurement.

After drying, the sample is wet ashed in nitric acid and finally in a mixture of nitric and hydrochloric acid. Wet ashing is necessary because polonium is a very volatile element. Undissolved silicates are filtered. The sample is neutralized and polonium is electrodeposited

onto a silver disc. Hydrazine is added to prevent the silver from darkening. The silver disc is measured by  $\alpha$ -spectrometry.

The yield is determined by  $^{209}\text{Po}$  or  $^{208}\text{Po}$  tracer added to the sample before wet ashing.

The method is quick and reliable.

Figure 13 shows the analysis scheme for polonium.

#### TREATMENT AND REPORTING OF RESULTS

The treatment and reporting of results, including units, vary significantly in the publications by the Nordic countries. Although the sampling and analysis procedures were comparable, the great variations in the processing and reporting would make the results difficult to compare. Some important points in the treatment and reporting of results are presented here.

Decay and yield correction usually have to be done first. Especially in studies of short-lived nuclides the correction for radioactive decay is very important. Generally, the sampling date is the reference date.

The reliability of the result must be confirmed before it is used. When parallel measurements have to be done, the results are compared; the average is usually taken as the final result. When calculating averages of results it should be weighed carefully which of the single results are to be included in the calculation. Moreover when concentrations are very low, the minimum detectable activity should be known, so that results below it can be rejected.

The error limits of the result should always be estimated. In addition to the statistical error, errors caused by the analysis and measuring apparatuses should be both estimated and reported as well. In many cases only the statistical error of the activity measurement is given.

Representativeness of a result or results should

also be considered; the more single results there are the better the representativeness. Difficulties are faced when conclusions have to be drawn from few results or from only one result.

If the average is reported, it would be useful to give the related range and the number of results used.

The reporting of background data, that is e.g. seasonal variation, salinity, pH, temperature of sea water, meteorological conditions and precipitation (air, deposition) makes it easier to compare the results. For foodstuff results, it is an advantage to know the part of production represented in the area reported.

### Recommendations

Some recommendations are needed to make it possible to compare results of analyses of environmental samples in the Nordic countries. The reporting of the results should include sampling date, period, place or area, amount and method. It is also useful to know the pretreatment and analysing methods. Table X summarizes these recommendations with special requirements for different sample types.

In the reporting of solid foodstuffs, results from the dry weight of plants and berries are more exact than those from the fresh weight, which varies with the humidity. Shaking and the time of collection can affect the fresh weights of plants. In contrast the fresh weight is more accurate for meat and fish, in which the end point of drying and the final dry weight are difficult to determine. Fresh weights are also used when calculating doses. In all results where the units  $\text{Bq kg}^{-1}$  dry weight or  $\text{Bq kg}^{-1}$  fresh weight are used, the ratio dry weight/fresh weight should be mentioned as well.

In reporting milk results constant K and Ca values

should be used and their concentrations in a litre of milk mentioned. When these values are known it is easy to calculate a value of Bq l<sup>-1</sup> milk. In milk results the knowledge of separation should be mentioned.

In reporting of results of sediment or sedimenting material, the water content gives information about the quality of the sediments. When reporting integral values for sediment, the unit of Bq m<sup>-2</sup> is used. In this case, the depth of sediment sample should also be mentioned.

#### PAN-SCANDINAVIAN INTERCALIBRATIONS

Pan-Scandinavian intercalibrations are carried out in analysing sediment, algae (Fucus vesiculosus) and sedimenting material and in comparing results of direct measurements of external radiation and air filters.

##### Sediment intercalibration

Sediment intercalibration was performed in May, 1979 at Barsebäck, Denmark<sup>33</sup>. The purpose was to calibrate both the sampling apparatuses and the analyses done by the laboratories. Five different devices were used for sampling and compared by measuring the dry weight of the samples. Samples were found to differ clearly from each other.

All five laboratories analysed the samples taken with each type of collector. <sup>40</sup>K, <sup>60</sup>Co, <sup>137</sup>Cs and <sup>239,240</sup>Pu were determined. In the results, differences in the activity levels are observed. For example, the relative standard deviation in the activity measurements of plutonium varied from 11% to 54% between the different laboratories and the samples.

This intercalibration showed that differences exist between both the collectors and analyses; intercalibrations

of this kind are therefore to be recommended.

#### Fucus intercalibrations

The sample was commercial dried seaweed from the Norwegian Westcoast. Five laboratories participated in the exercise. The  $^{137}\text{Cs}$  determinations showed a relative standard deviation of 6.5 %, while  $^{239,240}\text{Pu}$  varied by 26 % and  $^{226}\text{Ra}$  by 3 %.  $^{90}\text{Sr}$  was determined by only two laboratories, the relative standard deviation being 8.5 %. The determination of  $^{241}\text{Am}$  was unsatisfactory, the two results reported varying by nearly one order of magnitude.

#### Intercalibrations of detector systems for background radiation monitoring

Two intercalibrations of detector systems for background radiation monitoring were done<sup>34,35</sup>; the first one was carried out at Risø, Denmark, in June, 1980, with 22 participants and 23 instruments from 11 different laboratories; and the second in Helsinki, Finland, in May, 1982, where there were 12 participants and 13 instruments from 6 laboratories. High-pressure ionization chambers, NaI-scintillation counters, plastic scintillation counters, Geiger-Muller counters and Ge(Li) detectors as well as sets of TL dosimeter systems were the instruments to be calibrated.

Ionization chambers appeared to yield the most reliable results; the variations were larger in scintillators and GM instruments. Comparison of the calculated response-corrected terrestrial components, however, shows that the variations can largely be explained by variations in the response to components of natural gamma radiation.

Further intercalibrations and development of simple evaluation procedures are needed to improve the accuracy and comparability of environmental radiation measurements.

Intercalibration of collecting and analysing of ground level air

Intercalibration of air was done in May, 1982 at Nurmijärvi, Finland. Participants from four different laboratories used their own air samplers, and evaluated the artificial activity concentration in air by gammaspectrometry<sup>36</sup>.

Intercalibration of collecting and analysing of sedimenting material

An intercalibration of collection of sedimenting material was carried out in September - December 1981 at Loviisa, Finland. The aim was to compare the effectiveness of the different kinds of sediment traps used in the Nordic countries in the field of radioecology, as well as, the analyses of sedimenting material done in the participating laboratories.

Four different traps from Sweden and one from Finland were used to collect sedimenting material during a 12-week period in the water recipient area of the Loviisa nuclear power plant. Three of them represented cylindrical sediment vessels of different design and size; other two were identical cylinders with funnel bottoms. Marked differences were found in the amounts of sedimenting material collected by various traps when the results for dry weights per square metre were compared.

Three laboratories analysed all the samples collected gammaspectrometrically. Some differences were found in these results, too. Report of this intercalibration will be published in 1985.



## REFERENCES

1. Paakkola, O., Sample Collection and Preparation of Samples, Badan Tenaga Atom Nasional, Jakarta, 1975.
2. Paakkola, O., Insamling av miljöprov, Provinsamling och analysmetoder för radionuclider i långaktiva biösfärprover, Lund, Sverige, 1980.
3. Paakkola, O., Rantavaara, A., Några aktiva och inaktiva element i växter i Finland, Nordiskt radioekologiseminarium, Norrköping, Sverige, 1976.
4. Salo, A., Strontium-90 and Caesium-137 in Water in Finland, Report SFL-A4, Helsinki, 1966.
5. Bryant, F.J., Morgan, A., Spicer, G.S., The Determination of Radiostrontium in Biological Materials, AERE-R 3030, Harwell, 1959.
6. Osmond, R.G., Owers, M.J., Healy, C., Mead, A.P., The Determination of Radioactivity Due to Caesium, Strontium, Barium and Cerium in Waters and Filters, AERE-R 2899, Harwell, 1959.
7. Paakkola, O., Voipio, A., Strontium-90 in the Baltic Sea, Suomen Kemistilehti, B 38, 1965, pp. 11-17.
8. Determination of  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  in Aquatic Systems, Edited by M. Puhakainen, Finnish Centre for Radiation and Nuclear Safety, Helsinki, 1984, in Finnish, English Summary
9. Butler, Sr-90 Monitoring at the Savannah River Plant, Health Physics, 8, 1962, pp. 273-277.
10. Peppard, Mason, Moline, The Use of Dioctyl Phosphoric Acid Extraction in the Isolation of Carrier-Free Y-90, La-140, Ce-144, Pr-143 and Pr-144, Journ. Inorg. Nucl. Chem., 5, 1957, pp. 141-146.
11. Suomela, J., Halten av Sr-90 i mejerimjölk, 1965-1966, Intern basrapport från speciallab., SSI-1967-017.
12. Suomela, J., Snabb analysmetod för bestämning av Sr-89, Sr-90 och Y-90 företrädevis i mjölk, SSI-1973-010.
13. Procedure Manual, Health and Safety Laboratory Report, Radiochemical Determination of Plutonium in Soil by Leaching, HASL-300, 1972, pp. 89-114.

- 14 . Hakanen, M., Jaakkola, T., Distribution of Fallout Plutonium in Reindeer in Radioactive Foodchains in the Subarctic Environment, Contract CHE (11-1)-3011 of US, ERDA, Techn. Prog. Rep. for the period of Aug. 15., 1975 - Aug. 14., 1976, (Paper No 77), COO-3011-1.
15. Talvitie, N., Radiochemical Determination of Plutonium in Environmental and Biological Samples by Ion Exchange, Anal. Chem., Vol. 43, No 13, 1971, pp. 1827-1830.
16. Hakanen, M., Jaakkola, T., Distribution of Fallout Plutonium in Reindeer in Radioactive Foodchains in the Subarctic Environment, Contract No. CH E (11-1)-3011, US ERDA, Report for period Aug.15., 1976 - Nov. 14, 1977, Department of Radiochemistry, University of Helsinki, 1977.
17. Roberts, F., Brauer, F., Sequential Separation of Some Actinide Elements by Ion Exchange, HW-60552, 1959.
18. Suomela, J., Metod för bestämning av plutonium i omgivningsprover, SSI-1977-021.
19. Barring, N-E., Hellron, S., Kvantitativ elektrolytisk utfällning av plutonium ur sur lösning, AE-arbetsrapport, SSA-25, 1963.
20. Taipale, T. K., Tuomainen, K., Radiochemical Determination of Plutonium and Americium from Seawater, Sediment and Biota, STUK-B-VALO 26, Helsinki, to be published
21. Levine, H., Lamanna, A., Radiochemical Determination of Plutonium-239 in Low-Level Environmental Samples by Electrodeposition, Health Physics, 1965, Vol 11, pp. 117-125.
22. Bojanowski, R., Livingston, H., Schneider, D., Mann, D., A Procedure for Analysis of Americium in Marine Environmental Samples, in Reference Methods for Marine Radioactivity Studies 11, Techn. Rep. Ser. No. 169, Vienna, 1975, pp. 77-86.
23. Holm, E., Ballestra, S., Fukai, R., A Method for Ion-Exchange Separation of Low Levels of Americium in Environmental Materials, Talanta, Vol. 26., 1979, pp. 791-794.
24. Hakanen, M., Jaakkola, T., Rajamäki, S., Determination of  $^{241}\text{Am}$  in Sediment Samples in Radioactive Foodchains in the Subarctic Environment, Contract No. EY-76-C-02-3011, A003, US DOE, Final Report, Department of Radiochemistry, University of Helsinki, 1979.
25. Coleman, J., Asprey, L., Chisholm, R., Journ. Inorg. Chem. Vol. 31, 1969, p. 1167.
26. Talvitie, N.A., Electrodeposition of Actinides for Alpha Spectrometric Determination, Anal. Chem. Vol. 44, 1972, p. 280.

27. Östlund, H., Werner, H., The electrolytic enrichment of tritium and deuterium for natural tritium measurements, Tritium in the Physical and Biological Sciences, Vol. I, IAEA, Vienna, 1962.
28. Determination of Tritium from Water Samples with Electrolytic Enrichment Method, Edited by R. Saxén, Finnish Centre for Radiation and Nuclear Safety, Helsinki, 1984.
29. Goldin, A.S., Determination of dissolved radium, Anal. Chem. Vol. 33, 1961, p. 406.
30. Modified from: Radiochemical procedures for determination of selected members of the uranium and thorium series, Canmet Report 78-22, Canada Center for Mineral and Energy Technology, 1979.
31. Kauranen, P., Miettinen, J.K., in Radiecological Concentration Methods, Ed. B. Åberg and F.P. Hungate, Pergamon Press, Oxford, 1967.
32. Häsänen, E., Dating of Sediments, Based on  $^{210}\text{Po}$  Measurements, Radiochem. Radioanal. Letters, 31, 1977, pp.207-214.
33. Aarkrog, A., Nordisk Interkalibrering af sediment prøveindsamling og analyse, NKA's styringsgruppe for radioøkologi, 1980.
34. Böttger-Jensen, L., Nielsen, S., A Nordic Intercomparison of Detector Systems for Background Radiation Monitoring, Risø-M-2266, Denmark, 1981.
35. Arvela, H., A Nordic Intercalibration and Evaluation of the Detector Responses to Natural Environmental Gamma Radiation, Helsinki, 1983, STL-A46.
36. An Intercomparison of Airborne Dust Sampling and Sample Analysis between Nordic Countries, Joint Report Edited by Kari Sinkko, Helsinki, 1983, STL-B-TUTO-25.

Figure 1. Radiochemical determination of caesium 137.  
(Used in lab. no. 9. (Annex. 1.))

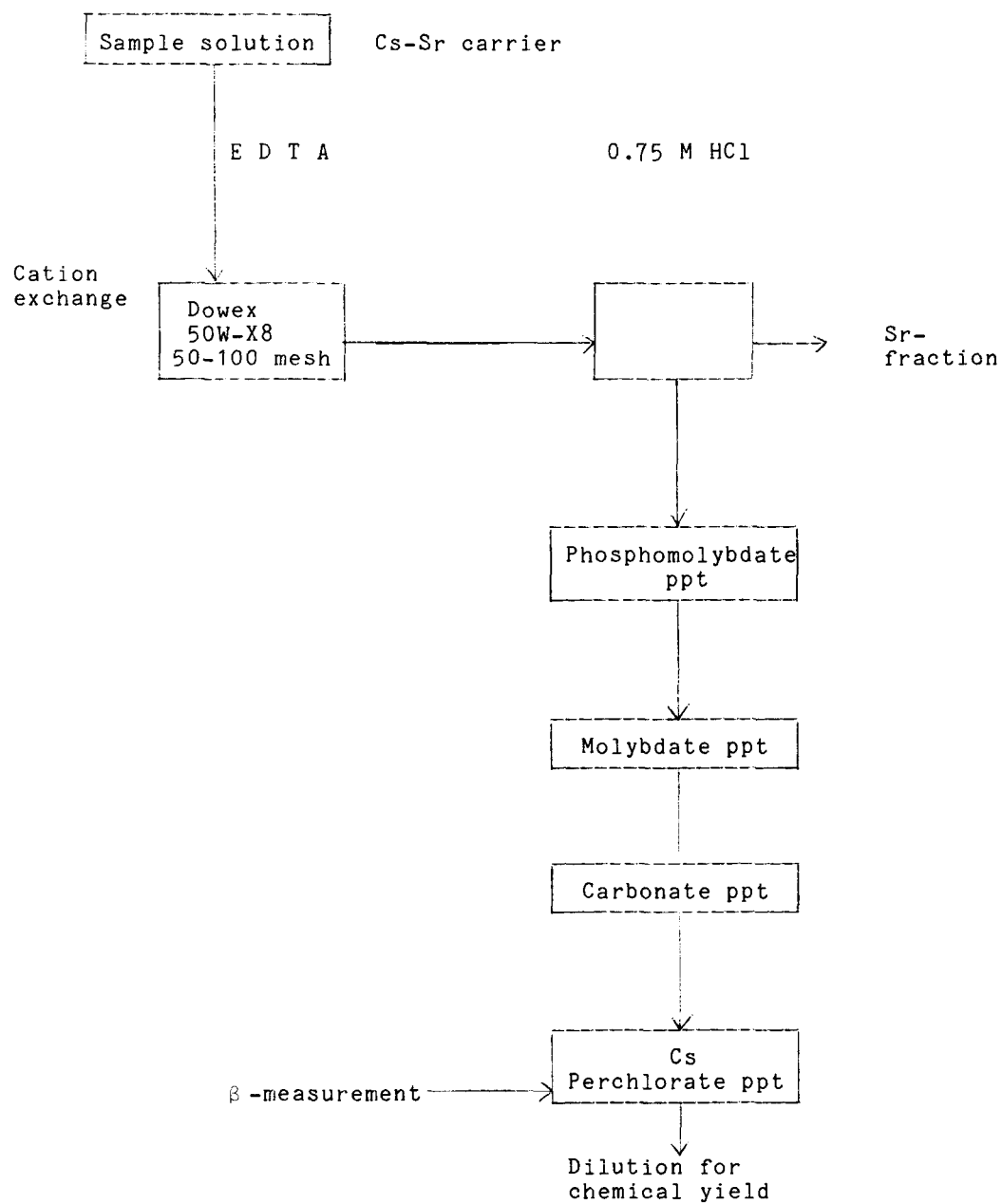


Figure 2.  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  determination from sediment and soil<sup>5</sup>.  
(Used in lab. no. 3., 5.)

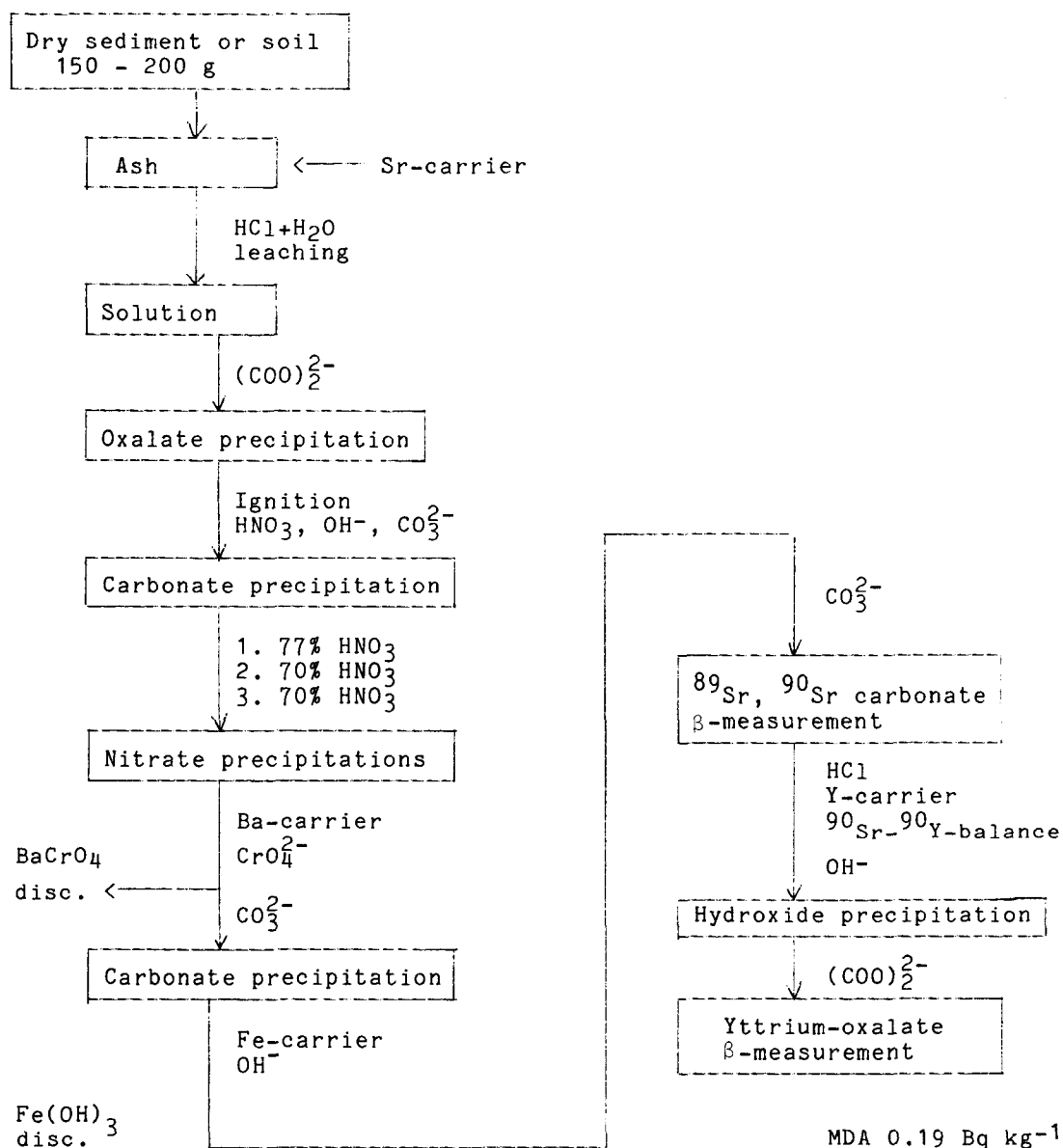


Figure 3. Determination of  $^{90}\text{Sr}$  ( $^{89}\text{Sr}$ ).  
(Used in lab. no. 9.)

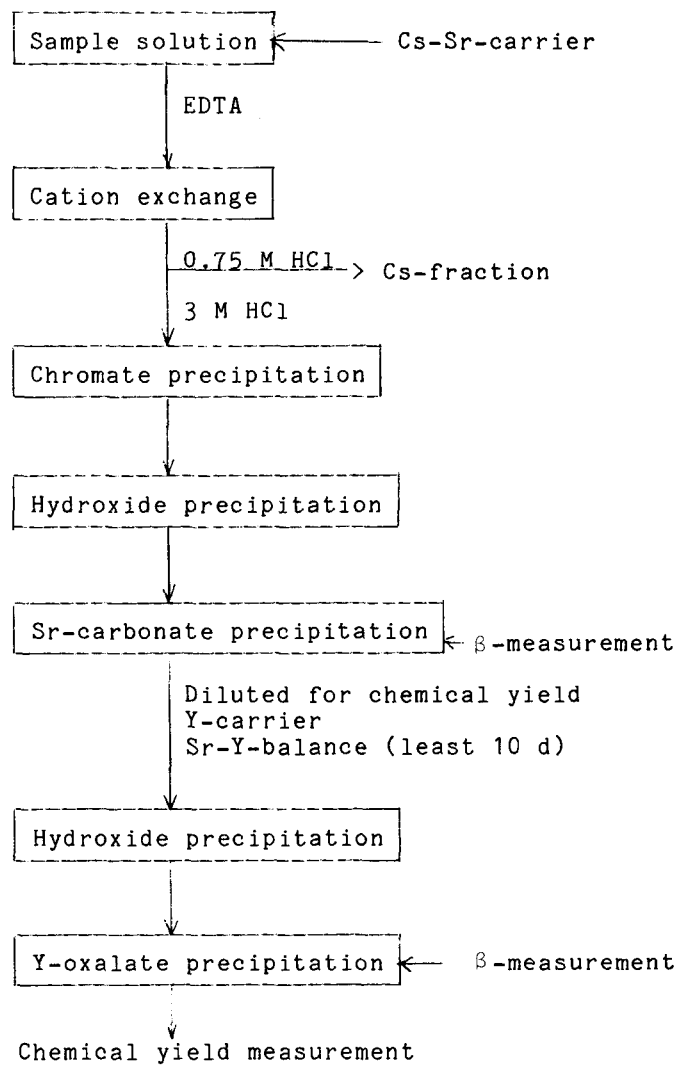
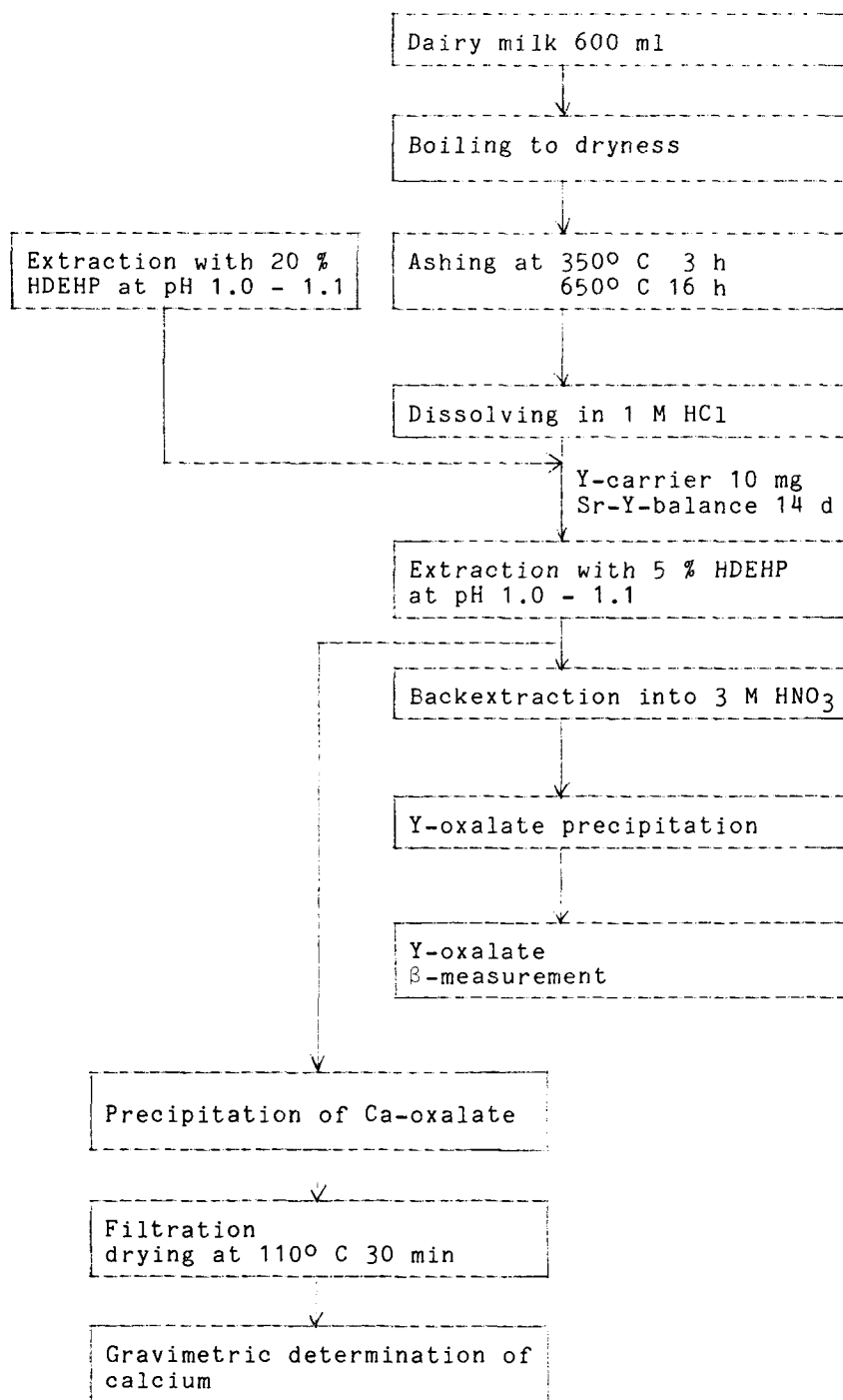


Figure 4. Determination of Sr 90 and calcium in milk<sup>9,10,11</sup>.  
 (fresh fallout)  
 (Used in lab. no. 7.)



MDA<sub>Sr</sub> 7.4 mBq l<sup>-1</sup>

Figure 5. Determination of Sr 89 and Sr 90 in milk<sup>12</sup>.  
(Rapid method)  
(Used in lab. no. 7.)

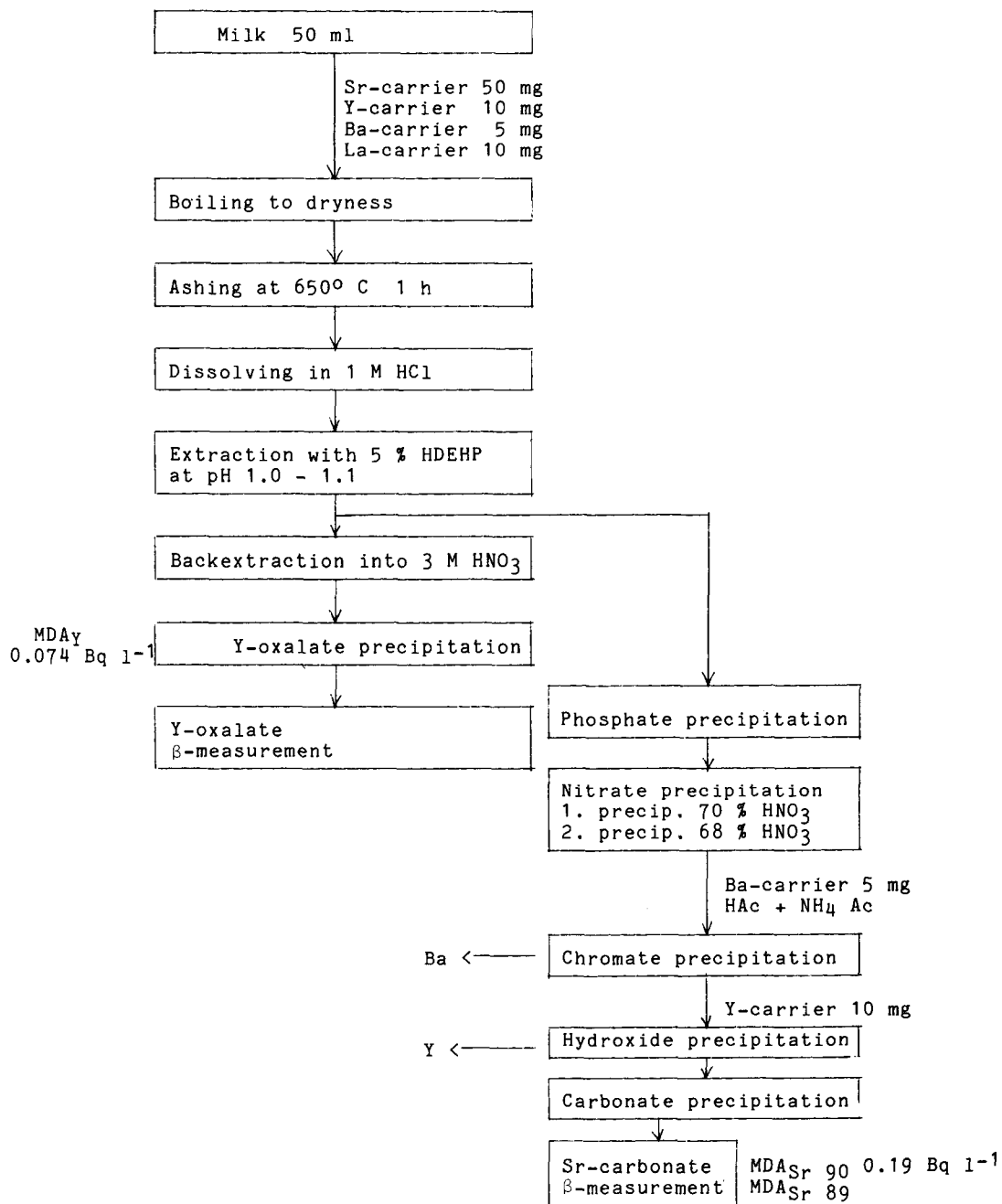




Figure 6. Determination of plutonium in soil, sediment and biota<sup>13-20</sup>.

(Used in lab. no. 3.,4.,7.,9.,10.)

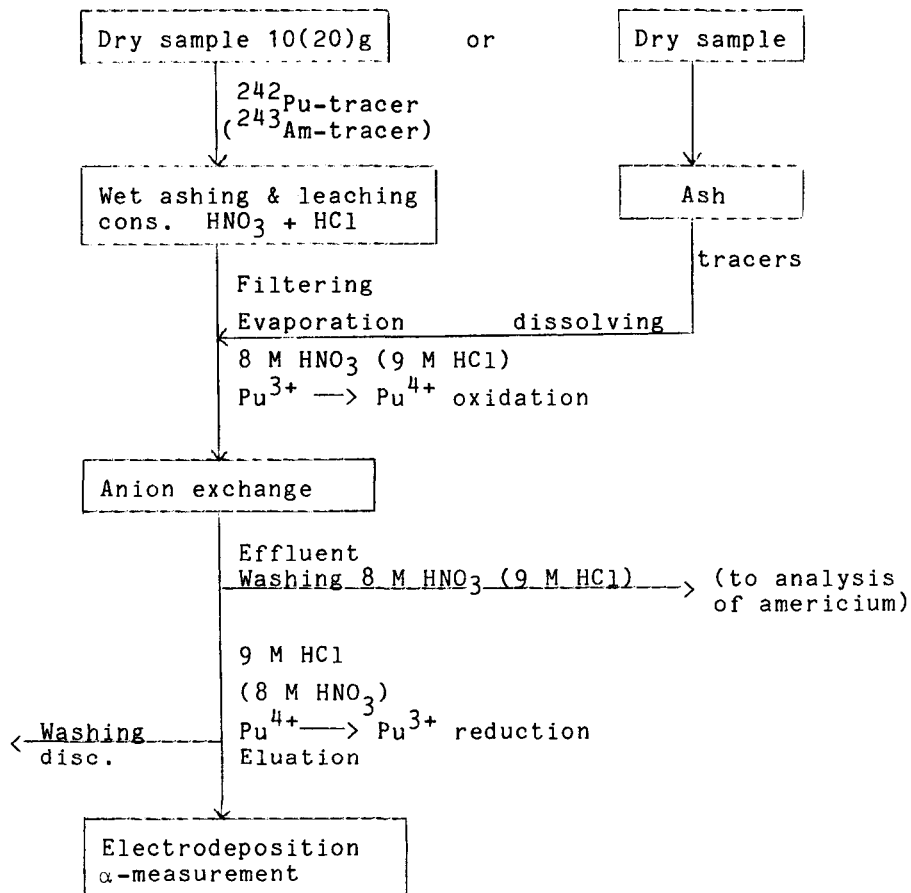


Figure 7. Determination of plutonium with TTA-extraction<sup>21</sup>.  
(Used in lab. no. 5.)

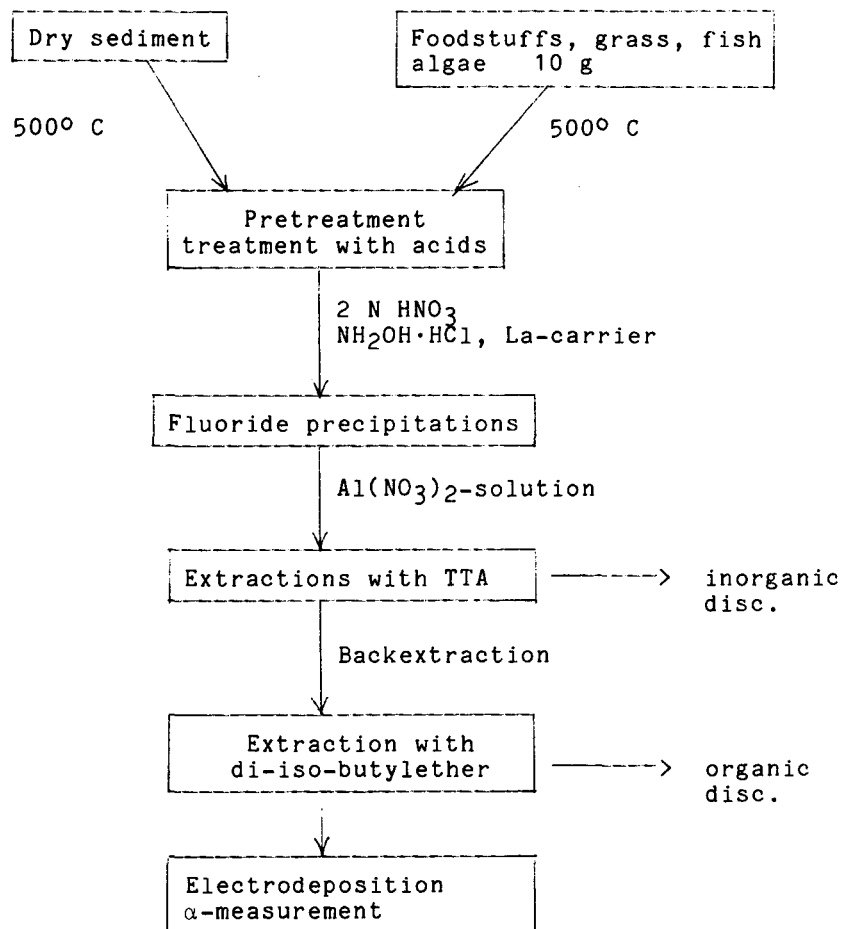


Figure 8. Determination of americium<sup>20,22,23</sup>.  
(Used in lab. no. 3., 10.)

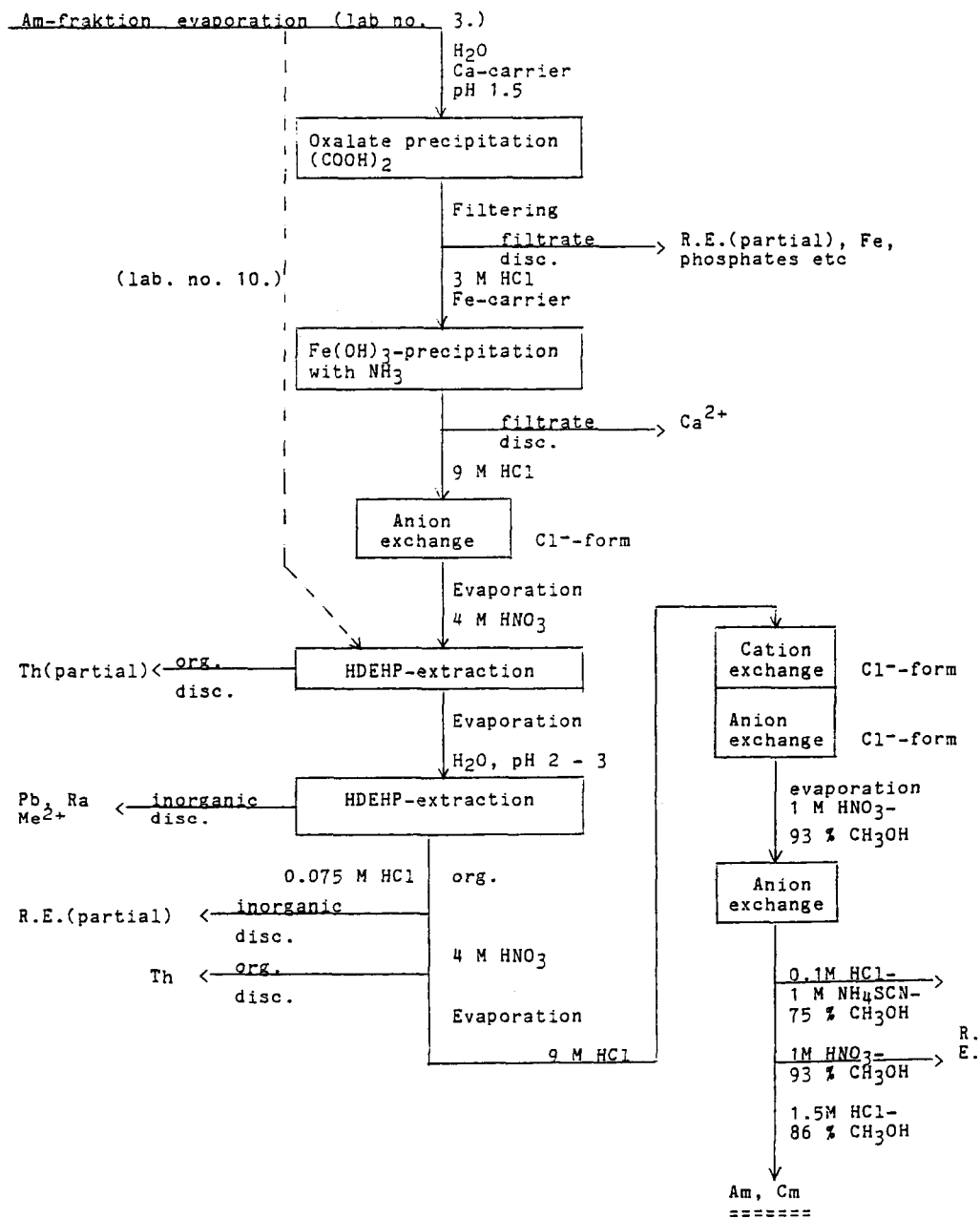


Figure 9. Determination of americium by ion exchange<sup>22,24,25,26.</sup>  
(Used in lab. no. 4.)

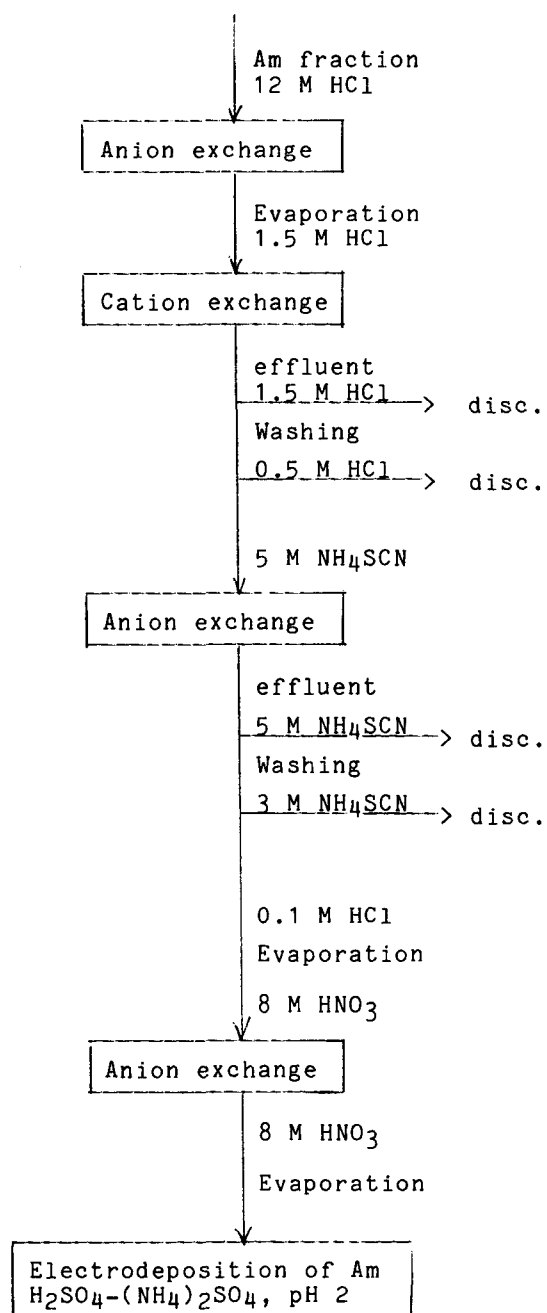


Figure 10. Determination of tritium from water samples<sup>27,28</sup>.  
(Used in lab. no. 3.)

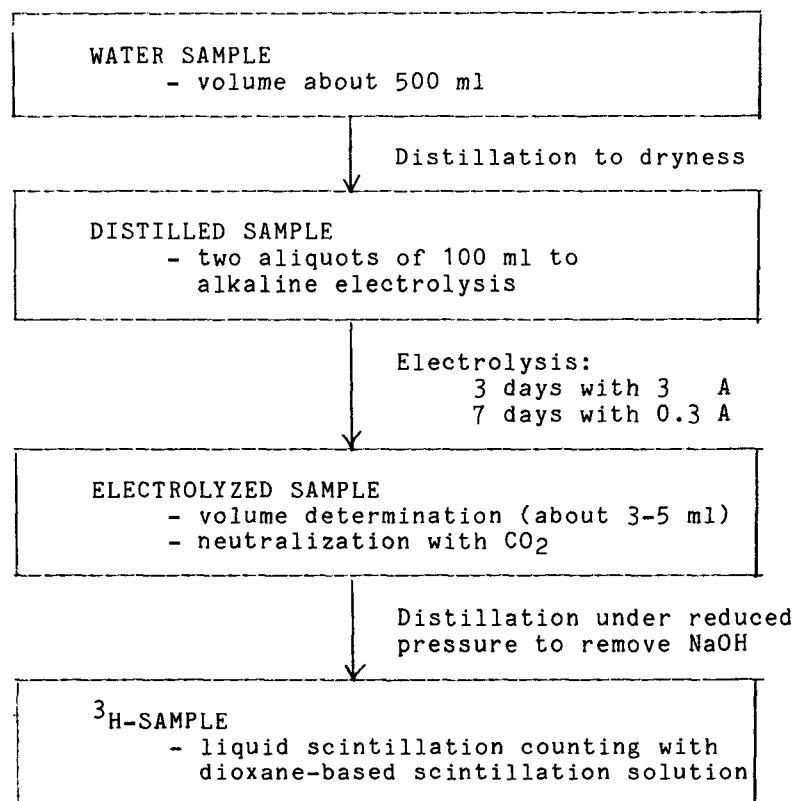


Figure 11. Determination of  $^{226}\text{Ra}$  from foodstuffs<sup>29</sup>.  
(Used in lab. no. 3., 7.)

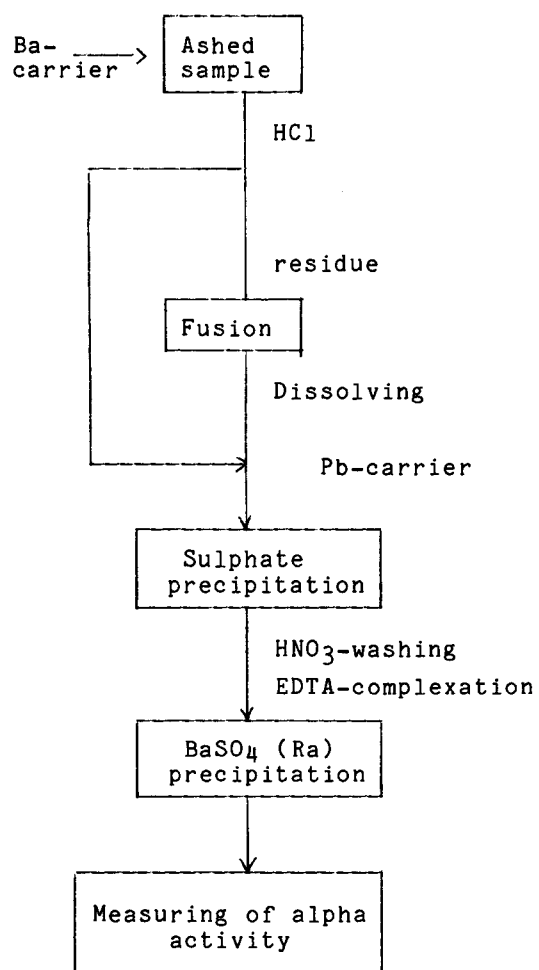


Figure 12. Determination of  $^{210}\text{Pb}$  from plants and food-stuffs<sup>30</sup>.  
(Used in lab. no. 3.)

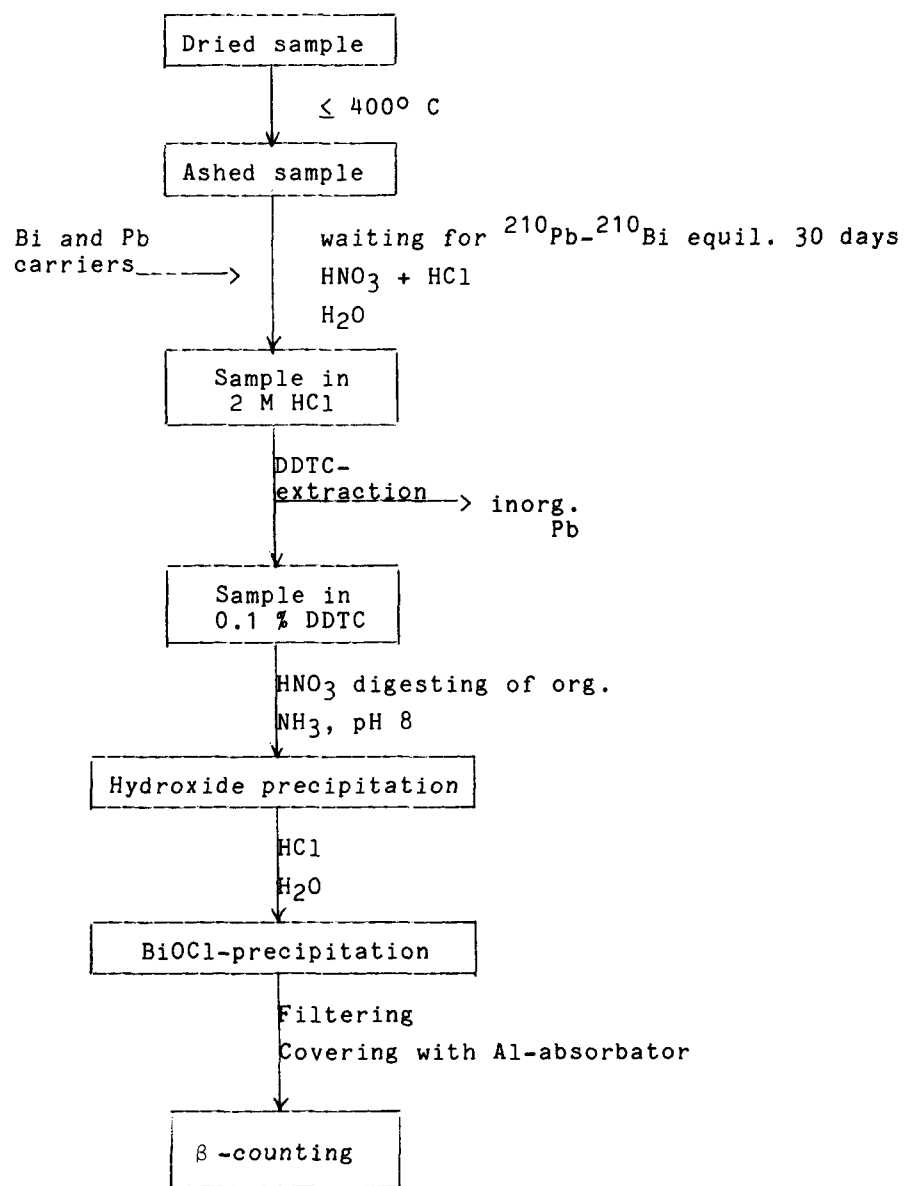


Figure 13. Determination of polonium  $^{210}_{31,32}$ .  
(Used in lab. no. 3., 4.)

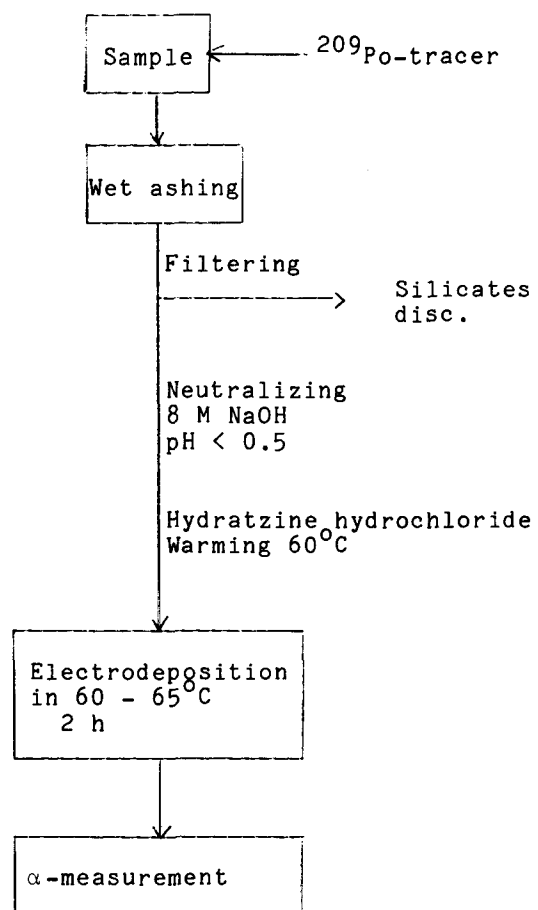




Table I. Sampling and pretreatment methods of air in the nordic laboratories.

Laboratory	Purpose of sampling	Type of collector	Sampling method amount filter	Frequency	Place (height)	Pretreatment	Analysis
Risø National Laboratory Denmark	-control of global and local fall-out	-compressor-centrifugal	-156000 m <sup>3</sup> /d - 40000 m <sup>3</sup> /d -paper -glass	-1 day -1 week	-3 m -1 m	-ashing -pressing	<sup>90</sup> Sr monthly γ-nuclides
Finnish Centre for Radiation and Nuclear Safety Finland	-control of global and local fall-out	-compressor -centrifugal	-18000 m <sup>3</sup> /d -glass -1500 m <sup>3</sup> /d -glass -carbon	-1 week -2 weeks	-1.5 m -1 m	-pressing	γ-nuclides <sup>90</sup> Sr quarterly
University of Helsinki Department of Radiochemistry Finland	-control of local and global fall-out		-2400 m <sup>3</sup> /d	-not conti- nuously	-15 m (on the roof)	-drying	

continues

Table I. continues

Laboratory	Purpose of sampling	Type of collector	Sampling method amount filter	Frequency	Place (height)	Pretreatment	Analysis
National Institute of Radiation Hygiene Norway	-control of global fall-out	-compres- sor	-18000 m <sup>3</sup> /d -glass	-1 week	-1.5 m	-pressing	Y-nuclides
National Institute of Radiation Protection Sweden	-control of global fall-out	-centrif- ugal	-24000 m <sup>3</sup> /d -72000 m <sup>3</sup> /d -glass -glass	-2-3 days -1 week	-1 m -1 m	-pressing	Y-nuclides
Studsvik Energiteknik AB Sweden	-control of local fall-out		-2800- 3600 m <sup>3</sup> /d -glass -carbon	-1 week -1 week	-1.5 m -1.5 m	-no -no	Y-nuclides monthly

Table II. Sampling and pretreatment methods of rainwater in the nordic laboratories.

Laboratory	Purpose of sampling	Sampling locations	Sampling method	Ion exchange	Pretreatment		Analysis
					in field	in laboratory	
Risø National Laboratory Denmark	-control of global and local fall-out -modelling	-10 +5 +2	-areal: 0.15 m <sup>2</sup>	no	-monthly sampling, combining	-evaporation	90Sr once in 2 months
		-1 in Risø	-areal: 10 m <sup>2</sup>	ion exchange		-ashing of the resin	γ -nuclides 90Sr from ash
National Institute of Radiation Hygiene Denmark	-control of global and local fall-out over Copenhagen -control of discharges of users of radioactive materials	-1 in Copenhagen	-indirect sampling	no	-1.5 kg of sewage sludge is pressed to 50-60 % water content		γ -nuclides

continues

Table II. continues

Laboratory	Purpose of sampling	Sampling locations	Sampling method	Ion exchange	Pretreatment		Analysis
					in field	in laboratory	
Finnish Centre for Radiation and Nuclear Safety Finland	-control of global and local fall-out -environmental studies near planned uranium mines	-18+5 in the whole country	-areal: -1 m <sup>2</sup> -0.05 m <sup>2</sup> (stainless steel)	no	-washing with 1% HNO <sub>3</sub> and distilled water	-evaporation	γ-nuclides monthly 89,90Sr monthly (from some samples)
			-0.05 m <sup>2</sup> (brass) -all collectors situated 1 m above ground	no		-ashing +450°C  -distilling	
Institute for Energy Technology Norway	-control of discharges and fall-out	-5 locations to cover all dominating wind directions	-standard meteorological equipment	no	-precipitation noted daily or monthly -no pretreatment	-evaporation for total-β	γ-nuclides total-β

continues

Table II. continues

Laboratory	Purpose of sampling	Sampling locations	Sampling method	Ion exchange	Pretreatment		Analysis
					in field	in laboratory	
National Institute of Radiation Protection Sweden	-control of the Test Ban Treaty -global and local fall-out studies and control	-7 locations over the whole country	-Ø2 m stainless steel funnel -2.5 m above ground	20 ml anion and 20ml cation exchange resins	-resins are changed once a week -washing and cleaning once a month	-ashing at 300° C about 20 h	γ-nuclides
Studsvik Energiteknik AB Sweden	-local control of the reactor	-1 location  -5 locations cover dominating wind directions	-1 m <sup>2</sup> stainless steel  -0.25 m <sup>2</sup> -1.5m above ground	no  anion and cation exchange resins	-washing with H <sub>2</sub> O once a week  -no pretreatment -changed once a month	-evaporation  -no	

Table III. Sampling and pretreatment methods of soil in the nordic laboratories.

Laboratory	Purpose of the sampling	Sampling locations	Sampling method	Pretreatment	Analysis
Risø National Laboratory Denmark	-following vertical transfer  -accumulation studies in the soil	-10 locations  -uncultivated soil mainly	-4x4 m <sup>2</sup> area -8 subsamples -auger Ø65mm -to 50 cm depth -every fifth year	-slices 0-10 cm, 10-20 cm etc. -drying -sifting	<sup>90</sup> Sr, <sup>137</sup> Cs <sup>239</sup> , <sup>240</sup> Pu <sup>241</sup> Am every fifth year
Finnish Centre for Radiation and Nuclear Safety Finland	-measuring total amounts of nuclides in soil per area and average concentration in surface layer	-uncultivated soil in the environment of nuclear power plants -clayey moraine	-auger Ø 7cm -0-25 cm -every third year	-slices 2 cm and 6-20 cm -drying at +85° C -sifting (2 mm) -combinating for Pu-analysis	<sup>90</sup> Sr, <sup>137</sup> Cs <sup>239</sup> , <sup>240</sup> Pu every third year
University of Uppsala, Radiobiological Institute Sweden	-measuring total concentrations -studies of transfer in different soil type -studies of the extractable parts of nuclides	-different farming environment -different farming areas in Skawaborg in western Sweden	-spade -20 subsamples from 50 m <sup>2</sup> area -0-25cm, 25-50cm -same firm places	-drying at 50°C -milling -sifting (2 mm)	Uranium  <sup>232</sup> Th, <sup>226</sup> Ra <sup>238</sup> U, <sup>234</sup> U <sup>230</sup> Th, <sup>232</sup> Th <sup>226</sup> Ra: extraction tests

continues

Table III. continues

Laboratory	Purpose of the sampling	Sampling locations	Sampling method	Pretreatment	Analysis
University of Lund Radiation Physics Department Sweden		-environments of planned uranium mines (Plentajok, Lilljuthatten)	-auger Ø 7cm to 30cm depth -about 15 samples in every area	-slices 3cm -drying	Uranium series U, Po, Ra γ-nuclides
National Swedish Environment Protection Board Sweden	-control of nuclear power plants	-the environments of nuclear power plants 2-3 areas/plant	-surface soil 0-5cm -once a year -2-3 litres	-cleaning -drying -ashing at 490° C	γ-nuclides (90Sr, 239,240Pu may become actual)
National Institute of Radiation Protection Sweden	-dose estimates from building material	-gravel pits	-occasional sampling from gravel	-crushing to small pieces	$^{226}\text{Ra}$ $^{232}\text{Th}$ $^{40}\text{K}$
Studsvik Energiteknik AB Sweden	-control of nuclear power plants -research projects	-2 locations according to monitoring programme	-once a year	-drying 105° C -homogenizing -ashing 550° C	

Table IV.

Sampling and pretreatment methods of milk in the nordic laboratories.

Laboratory	Purpose of sampling	Sampling locations	Sampling method	Pretreatment	Analysis
Risø National Laboratory Denmark	-measuring the doses of $^{90}\text{Sr}$ , $^{137}\text{Cs}$ caused by milk -modelling of radioactive contamination of foodstuffs	-7 drymilk factories over the whole country	-1 sample from the production of a month -2 kg	-ashing	$^{90}\text{Sr}$ , $^{137}\text{Cs}$
Finnish Centre for Radiation and Nuclear Safety Finland	-measuring of average concentration of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ in different areas of production over the whole country -dose estimates -control of nuclear power plants	-3 dairies -6 drymilk factories  -8 farms < 10km from nucl.p.plant  -local dairy < 40km	-1 sample weekly analysed once a month -1 sample monthly from production of a factory -5 litres or 1 kg -1 sample weekly from every farm -8 litres -1 sample weekly from large storing tank -8 litres from the whole production	-evaporation -ashing 450° C  -0.01-0.02% $\text{NaN}_3$ as preservative -evaporation -ashing	$^{131}\text{I}$ , $^{89}\text{Sr}$ (if needed) $^{90}\text{Sr}$ , Ca $^{137}\text{Cs}$ , K  $^{131}\text{I}$ weekly $^{137}\text{Cs}$ monthly $^{90}\text{Sr}$ monthly
Institute for Energy Technology Norway	-control of discharges from reactor	-2 farms in the environment of the institute	-1 sample weekly or -4 analysis yearly	-no pretreatment	$\gamma$ -nuclides: $^{131}\text{I}$ weekly $^{90}\text{Sr}$ quarter yearly

continues



Table IV continues

Laboratory	Purpose of sampling	Sampling locations	Sampling method	Pretreatment	Analysis
National Swedish Environment Protection Board Sweden	-control of nuclear power plants	-one farm near each power plant	-1 l unseparated milk from each farm	-no	$\gamma$ -nuclides specially $^{131}\text{I}$
National Institute of Radiation Protection Sweden	-control of $\gamma$ -nuclides in milk -average concentrations in the country	-5 dairies	-4 samples/dairy -yearly -2 litres	-ashing	$\gamma$ -nuclides, $^{137}\text{Cs}$ , K $^{90}\text{Sr}$
Studsvik Energiteknik AB Sweden	-control of the environment of the reactor	-1 farm	-1 sample weekly or -2 samples yearly (Sr, Cs)	-no	$\gamma$ -nuclides: $^{131}\text{I}$ , $^{125}\text{I}$ , $^{137}\text{Cs}$ every second week during the grazing period

Table V. Sampling and pretreatment methods of foodstuffs (other than milk) in the nordic laboratories.

Laboratory	Purpose of sampling	Sampling material	Sampling locations and method	Pretreatment	Analysis
Risø National Laboratory Denmark	-measuring of doses of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ -modelling	-cereals, vegetables bread, total diet -meat, egg	-yearly from different parts of the country -from Copenhagen	-ashing	$^{90}\text{Sr}$ , $^{137}\text{Cs}$
Finnish Centre for Radiation and Nuclear Safety Finland	-determination of doses to a man -determination of average concentrations of nuclides  -control of nuclear power plants	-cereals, 3 kg (wheat, rye) -leaf and root vegetables, fruits berries (5-7 kg) -meat, 2-3 kg (pork and beef)  -cereals, 5 kg (wheat, rye)  -lettuce, apple or currants -meat, 2 kg (beef)	-4 central-stores yearly from 5-8 product centers  -2 abattoirs twice a year  -5 farms near nuclear power plants (< 20km) -once or twice a year	-cleaning -drying $105^{\circ}\text{C}$ -milling -ashing $\leq 450^{\circ}\text{C}$	$^{90}\text{Sr}$ , Ca $^{137}\text{Cs}$ , K $\gamma$ -nuclides $^{89}\text{Sr}$ , $^{90}\text{Sr}$
University of Helsinki Department of Radiochemistry Finland	-following the transfer of long-lived radio-nuclides in food chains	-reindeer meat	-Lapland in the connection of slaughters -once a year	-freezing -(drying $+105^{\circ}\text{C}$ )	$^{137}\text{Cs}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$

continues

Table V. continues

Laboratory	Purpose of sampling	Sampling material	Sampling locations and method	Pretreatment	Analysis
National Swedish Environment Protection Board Sweden	-control of nuclear power plants	-meat (elg, goat, lamb) -cereals (wheat, barley) -vegetables	-the environments of nuclear power plants -yearly in the connection of harvest or slaughter	-cleaning -freezing -drying -ashing at 490° C	$\gamma$ -nuclides $^{90}\text{Sr}$ , $^{236}\text{Pu}$ (from some samples)
National Institute of Radiation Protection Sweden	-control of -nuclides in foodstuffs	-reindeer meat	-in the connection of reindeer slaughter, some samples as a average of some years	-freezing	$^{137}\text{Cs}$ , K
Studsvik Energiteknik AB Sweden	-control of nuclear power plants	-meat -cereals, vegetables	-the environments of nuclear power plants -once a year	-drying 105° C -homogenizing -ashing at 550° C	$\gamma$ -nuclides

Table VI. Sampling and pretreatment methods of terrestrial indicators in the nordic laboratories.

Laboratory	Purpose of the sampling	Indicator	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
Risø National Laboratory Denmark	-studies of food-chain: grass-cow-man, grass-air-foodstuffs	-grass	-the environment of Risø, 33 points <16 km -four times a year	-1 m <sup>2</sup> area -cutting 1-2cm over soil	-combinating -ashing	<sup>131</sup> I, <sup>90</sup> Sr γ-nuclides
Finnish Centre for Radiation and Nuclear Safety Finland	-control of the nuclear power plants -monitoring of uranium deposits	-grass -moss -lichen -beard moss	-in the environments of nuclear power plants -1) < 5 km 2) 0-10 km -2-4 times a year in summertime -the environment of uranium depositions in summer time	-grass from milk farms -cutting 1-2 cm over soil	-cleaning -drying -milling -ashing +450° C -for <sup>226</sup> Ra	<sup>131</sup> I <sup>89</sup> , <sup>90</sup> Sr γ-nuclides <sup>226</sup> Ra
University of Helsinki Department of Radiochemistry Finland	-study of food chains	-lichen	-Lapland and southern Finland -not continuously	-sampling by hand	-cleaning -drying	<sup>55</sup> Fe, <sup>210</sup> Po- <sup>210</sup> Pb, <sup>137</sup> Cs, <sup>90</sup> Sr, <sup>239</sup> Pu <sup>241</sup> Am
Institute for Energy Technology Norway	-control of the reactor in Kjeller	-grass	-the environment of Kjeller, 150-650m from reactor -4-5 times a year in summertime	-2 m <sup>2</sup> area -direct sampling without roots	-ashing	<sup>131</sup> I (no pre-treatment) γ-nuclides Pu

continues

Table VI. continues

Laboratory	Purpose of the sampling	Indicator	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
National Swedish Environment Protection Board Sweden	-control of nuclear power plants	-moss -lichen -grass -fern	-the environment of nucl.p.plants, -2-3 areas -once a year	-2 m <sup>2</sup> area	-drying 105°C -ashing 550°C or freeze-drying	γ-nuclides 239,240Pu
University of Lund, Radiation Physics Department Sweden		-moss -lichen	-the environments of planned uranium mines	-0.25-8 m <sup>2</sup>	-cleaning -drying -milling	U, Ra, Po γ-nuclides
Studsvik Energiteknik AB Sweden	-control of the environment of the reactor and nuclear power plants	-grass -moss -lichen -fern	-grass: 3 locations near Studsvik reactor weekly in summertime -others: twice a year	-grass: cutting to 2 cm, 50 x 50cm	-drying 150°C -ashing 550°C	γ-nuclides (Sr, Pu, U, Ra from some samples)

Table VII. Sampling and pretreatment methods of sea water in the nordic laboratories.

Laboratory	Purpose of sampling	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
Risø National Laboratory, Department of Health Physics Denmark	<ul style="list-style-type: none"> <li>-following the radioactive contamination of the Danish Straits</li> <li>-following the radioactive contamination of the North Sea, the North Atlantic and the Baltic Sea</li> </ul>	<ul style="list-style-type: none"> <li>-16 locations around Sealand surface and bottom water</li> <li>-half-yearly: summer and winter</li> <li>-occasional sampling with various ships - 100 samples/yr.</li> </ul>	<ul style="list-style-type: none"> <li>-pumping 50 litres</li> <li>-pumping 50-1800 litres</li> </ul>	<ul style="list-style-type: none"> <li>-acid and carriers added in the laboratory to the samples</li> <li>-Precipitation on board of Pu, Am, (OH) and radiocesium (AMP) in 200-1800 litres</li> </ul>	radiocesium $^{90}\text{Sr}$ , Pu, Am, $^3\text{H}$ .
Finnish Centre for Radiation and Nuclear Safety Finland	<ul style="list-style-type: none"> <li>-following the radioactive condition in the Baltic Sea</li> <li>-control of the nuclear power plants</li> </ul>	<ul style="list-style-type: none"> <li>-different parts of the Baltic Sea, 6+2 points</li> <li>-yearly</li> <li>-5-6 points near nuclear power plants</li> <li>-3-4 times a year</li> </ul>	<ul style="list-style-type: none"> <li>-1m under surface 35-130 litres</li> <li>-some samples also 1m above the bottom</li> <li>-with Ruttner from 1/2-1 m depth, 1 litre for tritium</li> </ul>	<ul style="list-style-type: none"> <li>-subsampling</li> <li>-adding of acids and carriers</li> <li>-evaporation</li> <li>-distilling (for tritium)</li> </ul>	$\gamma$ -nuclides $^{90}\text{Sr}$ , $^{239,240}\text{Pu}$ , $^{241}\text{Am}$ , $^3\text{H}$ (from some samples)

continues

Table VII. continues

Laboratory	Purpose of sampling	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
University of Helsinki, Department of Radiochemistry Finland	-study of the behaviour of Pu and Am -study of the distribution between particulate and soluble form	-constant points -1-2 times a year	-pumping through millipore filter (0.3 or 0.45 $\mu$ m) -0-200 m depth	-adding of acids and tracers -weighing of filters	$^{239,240}\text{Pu}$ , $^{241}\text{Am}$
Institute for Energy Technology Norway	-control of radioactivity and research work				
National Swedish Environment Protection Board Sweden	-control of nuclear power plants	-1-2 points near the outlet at each power plant	-1 l with Ruttner-sampler under the surface or 50 l sample	-adding of acids -adding of acids, carrier and AMP	$\gamma$ -nuclides 1 l Marinelli Cs-nuclides
University of Lund, Radiation Physics Department Sweden	-research	-North Sea, Arctic Sea, Baltic Sea -yearly expeditions	-100-1700 l  1-10 l	-precipitation with NaOH, pH9 -with AMP, -with $\text{NH}_3$ -no	Th, U, Np, Pu Am, cm  Cs Tc U, $^{210}\text{Po}$ , $^{210}\text{Pb}$

Table VIII. Sampling and pretreatment methods of sediment (1) and sedimenting material (2) in the nordic laboratories.

Laboratory	1	2	Purpose of the sampling	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
Risø National Laboratory, Health physics Department Denmark	x		-monitoring of global fall-out	-1-2 locations Danish Straits -annually	-HAPS samples 1-3 cm slices -13.5 cm Ø	-drying at 120°C homogenizing, -acidextraction for trans-uranics	γ-nuclides Pu, Am, occasionally <sup>90</sup> Sr
	x		-monitoring nuclear power plants	-3-5 locations Barsebäck and Ringhals -semi-annually	- " -		
	x		-relations between sediments and benthic radioactivity contest	-20 locations in Thule, Greenland -every 5th year	- " -		
University of Helsinki, Department of Radiochemistry Finland	x		-determination of the age of sediment -study of the distribution of nuclides in sediment	-the Baltic Sea -Lake Vanajavesi -Lake Saimaa -some small lakes		-slicing to subsamples -freezing	<sup>137</sup> Cs, <sup>210</sup> Pb, <sup>210</sup> Po <sup>239</sup> , <sup>240</sup> Pu, <sup>241</sup> Am

continues



Table VIII. continues

Laboratory	1	2	Purpose of the sampling	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
Finnish Centre for Radiation and Nuclear Safety Finland	x		-monitoring of global fall-out -determination of the balance of the Baltic Sea -research of food chains (Baltic Sea project of IAEA)	-5 points in the Baltic Sea, 150-170 samples -yearly	-box corer, 333 cm <sup>2</sup> : 5 cm slices to 30 cm -corer, 19.63 cm <sup>2</sup> : 1 cm slices to 15 cm	-freeze-drying -homogenizing -ashing (Sr) -wet ashing (transur.)	γ-nuclides 90Sr, 239,240Pu  241Am (from some samples)
		x	-monitoring of global fall-out -researching of food chains	-2 points -continuous sampling	-funnels: 4x66,5 cm <sup>2</sup> -collecting in 2 weeks periods -1 m above bottom (also 5 m and 10 m from surface in some points)	-combinating to 3 months periods -drying -wet ashing (Pu)	γ-nuclides 239,240Pu twice a year
		x	-control of nuclear power plants	-3-4 sampling points/ nuclear power plant -continuous sampling			
Institute for Energy Technology Norway	x		-control of the environmental radioactivity	-5 places in the Nitelva, once a year -10 places around the area of outlet of the waste (area 100 m <sup>2</sup> ) once a year	-ordinary grab, upper layer to 40 cm -volume of each sample ca. 5600 cm <sup>3</sup>	-drying -digesting 50°C -treatment with HNO <sub>3</sub> , HCl, HF and evaporation	γ-nuclides 90Sr, 239Pu

continues

Table VIII. continues

Laboratory	1	2	Purpose of the sampling	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
National Swedish Environment Protection Board Sweden (*Lunds Univ.)	x		-control of nuclear power plants	-1 sampling point/ nuclear power plant once/month	-Wilner corer 38.5 cm <sup>2</sup> 2 cm slices	-freeze-drying -homogenizing -ashing	Y-nuclides 239,240Pu*
			-monitoring of global fall-out -researching of food chains	-20 points every fourth year.	-corer, 19.63 cm <sup>2</sup> 2 cm slices to 15 cm		241Am (from same samples)*
		x	-control in the Biotest basin sewage sludge	-1/month	-2.5 x 12 cm	-freeze-drying homogenizing	Y-nuclides
			-control of nuclear power plants	-3-4 communal sewage works/power plant once a year	-500 ml sewage sludge	-drying -ashing	Y-nuclides
Studsvik Energiteknik AB Sweden	x	x					
University of Lund Department of Radiation Physics Sweden	x		-research	-sea and lakes -yearly expedition	-Box corer	-drying -dissolving	Y-nuclides Pu, Am, U, Po

Table IX. Sampling and pretreatment methods of aquatic indicators in the nordic laboratories.

Laboratory	Purpose of the sampling	Indicator	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
Risø National Laboratory, Health Physics Dept. Denmark	<ul style="list-style-type: none"> <li>-monitoring global fall-out</li> <li>-monitoring waterborn releases from nuclear installations (Sellafield)</li> </ul>	<ul style="list-style-type: none"> <li>-fucoids</li> <li>-<u>Mytilus edulis</u></li> </ul>	<ul style="list-style-type: none"> <li>-Roskilde Fjord</li> <li>-2-3 locations</li> <li>-semi-annually</li> <li>-Danish Waters</li> <li>-20 locations</li> <li>-annually</li> <li>-Faroes, Greenland</li> <li>-annually</li> <li>-Danish Waters</li> <li>-3-5 locations</li> </ul>	<ul style="list-style-type: none"> <li>-collecting from boat and from land by hand or by dredge</li> </ul>	<ul style="list-style-type: none"> <li>-drying 100°C</li> <li>-milling</li> <li>-ashing at 400°C</li> <li>-further ashing at 600°C for Sr, Pu, Am</li> </ul>	<ul style="list-style-type: none"> <li>Y-nuclides</li> <li>Pu, Am</li> <li>Tc</li> </ul>
Finnish Centre for Radiation and Nuclear Safety Finland	<ul style="list-style-type: none"> <li>-monitoring of global fall-out</li> <li>-researching of food chains</li> <li>-control of nuclear power plants</li> <li>-IAEA project (see seawater)</li> </ul>	<ul style="list-style-type: none"> <li><u>Fucus vesiculosus</u></li> <li><u>Cladophora sp.</u></li> <li><u>Enteromorpha sp.</u></li> <li><u>Mesidotea entomon</u></li> <li><u>Macoma baltica</u></li> <li><u>Mytilus edulis</u></li> </ul>	<ul style="list-style-type: none"> <li>-2 locations/nuclear power plant</li> <li>-1-2 times a year</li> <li>-3-5 locations near Finnish coast</li> <li>-once a year (IAEA)</li> </ul>	<ul style="list-style-type: none"> <li>-by diving or sampler (2 kg of <u>Fucus</u>, 200g of <u>Macoma</u> and <u>Mytilus</u>)</li> <li>-by fishpot (200 g of <u>Mesidotea</u>)</li> <li>-<u>Mytilus</u>: also meat and shell separately</li> </ul>	<ul style="list-style-type: none"> <li>-drying 105°</li> <li>-milling</li> <li>-ashing at 450° C</li> <li>-wet ashing (Pu, Am)</li> </ul>	<ul style="list-style-type: none"> <li>Y-nuclides 89,90Sr</li> <li>239,240Pu</li> <li>241Am (from some samples)</li> </ul>

continues

Table IX. continues

Laboratory	Purpose of the sampling	Indicator	Sampling locations and frequency	Sampling method	Pretreatment	Analysis
Institute for Energy Technology Norway	-control of environmental radioactivity	- <u>Potamogeton perfoliatus</u>	-Nitelva river (close to institute) -twice a year	-sampling by hand	-drying -ashing -treament with HNO <sub>3</sub> , HCl, HF	γ-nuclides 90Sr, 239Pu
National Swedish Environment Protection Board Sweden  (*Lunds Univ.)	-control of nuclear power plants  -researching of food chains	<u>Fucus vesiculosus</u> <u>Cladophora</u> <u>Littorina sp.</u> <u>Lymnea sp.</u> <u>Mytilus Edulis</u> <u>Macoma baltica</u>	-2-7 locations/ nuclear power plant  -once a year	-by diving or picking 2 kg <u>Fucus</u> <u>Cladophora</u> 200 g <u>Macoma</u> , <u>Mytilus</u> , <u>Littorina</u> , <u>Mytilis</u> , weat and shell separately	-drying 105°C -ashing at 550°C or freeze-drying -homogenizing	γ-nuclides 239,240Pu*  241Am from some samples
Studsvik Energi-teknik AB	-control programme	- <u>Fucus vesiculosus</u> - <u>Cladophora sp.</u> - <u>Enteromorpha sp.</u> - <u>Macoma baltica</u> - <u>Mytilus edulis</u>	-1-2 stations -once a year	-raking, dredge or diving	-drying -ashing	γ-nuclides
University of Lund Department of Radiation Physics Sweden	-research and monitoring	- <u>Fucus vesiculosus</u>	-monthly in 4 fixed stations -yearly expeditions world wide		-drying -ashing	γ-nuclides -Pu, Am, Tc

Table X. Recommendations for the use of units and additional information of samples.

Sample	Nuclide	Unit	Additional information
Air	all	Bq m <sup>-3</sup>	temperature
Deposition	all	Bq m <sup>-2</sup>	precipitation
Soil	all	Bq m <sup>-2</sup>	soil type and depth
Milk	Sr Cs others	Bq g <sup>-1</sup> Ca Bq g <sup>-1</sup> K Bq l <sup>-1</sup>	production area
Meat and fish	all	Bq kg <sup>-1</sup> fresh weight	production area
Foodstuffs other than milk, meat and fish	all	Bq kg <sup>-1</sup> dry weight	production area
All indicators	all	Bq kg <sup>-1</sup> dry weight	
Sea water	all	Bq m <sup>-3</sup>	depth and salinity (temperature)
Sediment and sedimenting material	all	Bq kg <sup>-1</sup> dry weight Bq m <sup>-2</sup>	depth

Annex. 1. The participating nordic laboratories.

Lab. No. Denmark

1. Risø National Laboratory  
Roskilde
2. National Institute of Radiation Hygiene  
Brønshøj

Finland

3. Finnish Centre for Radiation and Nuclear Safety  
Helsinki
4. University of Helsinki, Department of Radiochemistry  
Helsinki

Norway

5. Institute for Energy Technology  
Kjeller
6. National Institute of Radiation Hygiene  
Østerås

Sweden

7. National Institute of Radiation Protection  
Stockholm
8. National Swedish Environment Protection Board  
Drottningholm
9. Studsvik Energiteknik AB  
Nyköping
10. University of Lund, Radiation Physics Department  
Lund
11. University of Uppsala, Radiobiological Institute  
Uppsala