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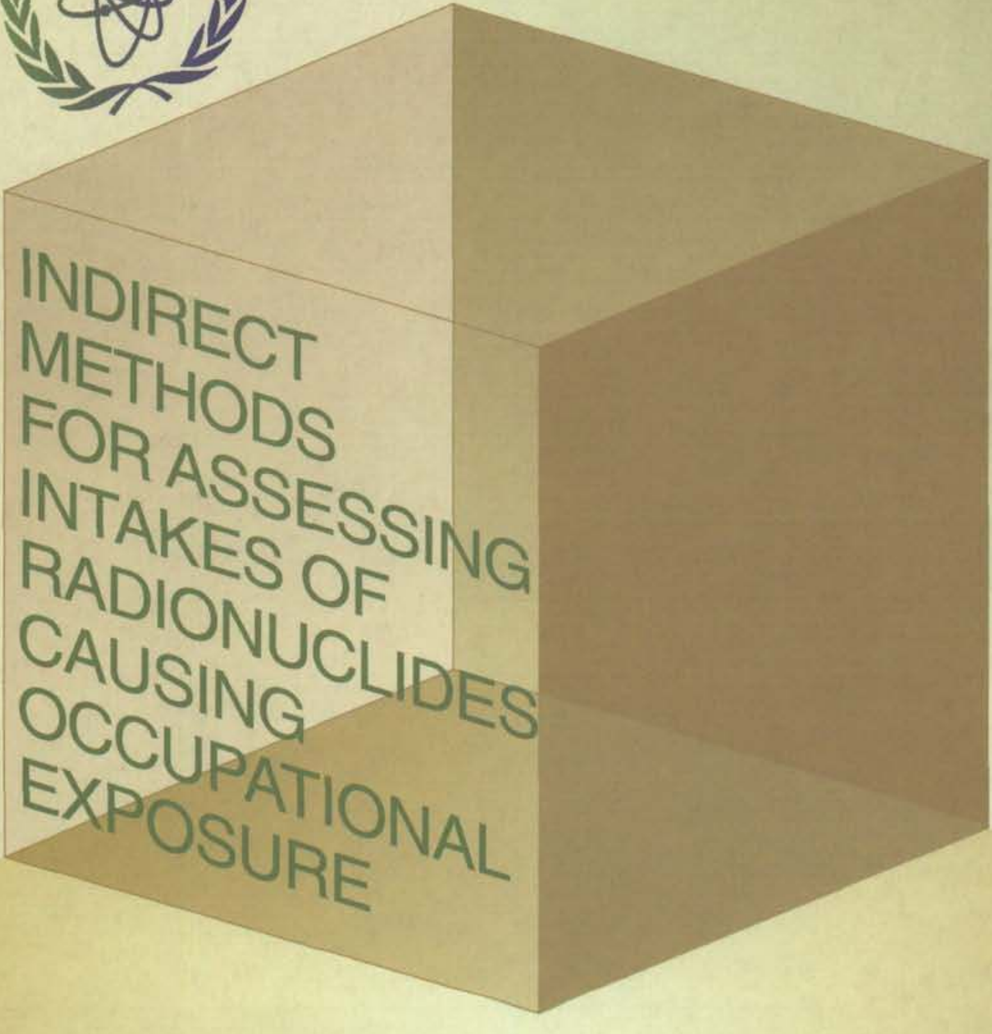

INDIRECT METHODS FOR ASSESSING INTAKES OF RADIONUCLIDES CAUSING OCCUPATIONAL EXPOSURE

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FOREWORD

Radioactive materials are used in many industries, and, whenever unsealed radioactive sources are present, intakes of radionuclides by workers can occur. Included here are the use of radioactive sources in medicine, scientific research, agriculture and industry, the operation of various facilities that are part of the nuclear fuel cycle, and work involving exposure to naturally occurring radionuclides at enhanced levels. Intakes can occur by a number of pathways, and the monitoring of workers and the workplace in such situations is an integral part of any occupational radiation protection programme.

Guidance on conducting assessments of intakes of radioactive material arising from occupational exposure is given in a Safety Guide, Assessment of Occupational Exposure due to Intakes of Radionuclides, published in 1999. This guidance depends generally on two approaches: *direct* and *indirect* methods of monitoring. To provide information on the establishment and operation of facilities for the estimation of intakes by direct methods, the IAEA published in 1996 a Safety Practice, Direct Methods for Measuring Radionuclides in the Human Body. The present Safety Report is the complementary report, providing technical details and practical information on indirect methods of monitoring for intakes of radionuclides.

This Safety Report is intended to assist in the setting up and operation of an indirect monitoring programme for workers, providing technical advice on the collection and analysis of biological and physical samples used to estimate intakes. The analytical methods described may, however, also be applied to the monitoring of patients following medical treatment or of members of the public following a release of radionuclides to the environment.

EDITORIAL NOTE

CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

Occupational intakes of radionuclides can occur as a result of various activities. These include work associated with the different stages of the nuclear fuel cycle; the use of radioactive sources in medicine, scientific research, agriculture and industry; and work which involves exposure to enhanced levels of naturally occurring radionuclides. The statutory requirements of most national authorities require that assessments be made of the doses resulting from such intakes. The IAEA Safety Guide, Occupational Radiation Protection [1], gives guidance and recommendations on setting up monitoring programmes for assessing exposure of the workforce. This guidance is based on the requirements contained in the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (BSS) [2]. A second Safety Guide, on Assessment of Occupational Exposure due to Intakes of Radionuclides [3], provides recommendations on the setting up and running of effective monitoring programmes for assessing intakes of radionuclides, and is supported by three Safety Reports of which this is one.

The present Safety Report gives technical details and practical information on indirect methods of monitoring for intakes. A companion Safety Practice, Direct Methods for Measuring Radionuclides in the Human Body [4], has also been published. Advice on the interpretation of both kinds of measurement data in terms of radiation doses to workers will be found in a third Report, Assessment of Radiation Doses from Intakes of Radionuclides by Workers, which is under preparation. Additional information on doses to the public is given in the Safety Report, Assessment of Doses to the Public from Ingested Radionuclides [5].

1.2. OBJECTIVE

The purpose of this Safety Report is to provide technical information on the collection and analysis of, mainly, biological samples used to estimate intakes of radionuclides, principally by measuring their rates of elimination from the body. It also provides information on the analysis of physical samples that can be used for estimating intakes, such as the measurements of airborne radioactive material. The Report is intended to assist in the setting up and operation of an indirect monitoring programme for workers who may come into contact with the radionuclides concerned. It is to be used in conjunction with the Safety Guide on Assessment of Occupational Exposure due to Intakes of Radionuclides [3] and the Safety Report on Assessment of Radiation Doses from Intakes of Radionuclides by Workers.

1.3. SCOPE

This Safety Report considers the sources of samples available for indirect monitoring of internal exposure, and gives guidance on the choice of appropriate samples. It also gives information on methods of sample collection. It discusses the various measurement techniques available and the methods of sample analysis. It gives advice on the methods to be used for reporting results. Finally, nine Annexes are provided that describe the detailed analytical procedures that have been developed by various laboratories for particular sources and samples, for which indirect methods are a primary means of estimating intakes.

The information given in this Safety Report is intended for application in occupational monitoring. The analytical methods described, however, may also be applied to monitoring the retention and excretion of radionuclides by patients following medical treatment, or by members of the public following the release of radionuclides to the environment. Analytical methods for measuring radionuclides in environmental samples are described in Technical Reports Series No. 295, Measurement of Radionuclides in Food and the Environment, A Guidebook [6].

1.4. STRUCTURE

A variety of sample sources is potentially available for indirect assessment of the content of radionuclides in the body. Section 2 of this Report reviews the criteria used in selecting suitable biological and physical samples for internal dose assessment, and appropriate methods of analysis. Table I is included to direct readers to Annexes that illustrate the use of particular methods. Section 3 of the Report considers in more detail the types of biological sample available and their merits both as indicators of an intake and as a basis for determining the rates of excretion of radionuclides following intakes of various chemical forms, such as are required in dose estimation. Section 4 then examines the use of physical samples such as air filters for monitoring the workplace.

Many techniques are available for measuring the levels of activity in biological and physical samples, depending on the nature of the emitted radiations and the physical/chemical form of the sample. Section 5 describes methods of sample preparation, as well as the main radiometric techniques for identifying and measuring the levels of radionuclides in samples. The use of non-radiometric techniques, such as mass spectrometry or fluorescence, is also considered. Section 6 provides a summary of the statistical considerations to be taken into account in interpreting these analyses.

Information is provided in Section 7 on the need for, and the use of, effective methods of quality assurance together with practical considerations in the application of these techniques. In Section 8 procedures for the reporting of results are considered.

Finally, Annexes I–IX give specific technical information on the analysis of selected radionuclides in a range of different sample matrices, on the basis of experience gained in various laboratories.

2. SELECTION OF SAMPLES AND METHODS OF ANALYSIS

2.1. INTRODUCTION

Indirect monitoring is based on the determination of activity concentrations in *biological* materials separated from the body, or in *physical* samples taken from the work environment. Indirect methods are most suitable for those radionuclides, such as tritium, which do not emit significant amounts of penetrating radiations. For other radionuclides, such as those which emit only low energy photons, the uncertainties and sensitivity of the direct monitoring measurement may be such that an indirect method can provide a more reliable estimate of the intake, despite its dependence for interpretation on biokinetic models of processes which may vary with time and between individuals. In other cases, although direct methods are available, indirect methods may be sufficiently accurate and more practical or accessible than direct monitoring. If in vivo monitoring facilities are not available, indirect methods have to be employed. The principal indirect methods which rely on biological samples for dose assessment are shown in Table I. This table also identifies Annexes in this Report which discuss representative methods for the analysis of selected radionuclides in biological samples.

2.2. BIOLOGICAL AND PHYSICAL SAMPLES

The biological samples used for the estimation of intakes and the assessment of internal exposures are most commonly urine and faeces, although breath and blood or other samples can be used in special cases. The choice of bioassay sample will depend not only on the major route of excretion, as determined from the biokinetic model for the element and the physico-chemical form of the intake, but also on such factors as ease of collection, analysis and interpretation. Urine samples are readily obtained and analysed, and generally provide information on the intake of chemical forms of radionuclides that are readily transferred to the blood. Intakes of insoluble

TABLE I. PRINCIPAL METHODS USED IN INDIRECT ASSESSMENT

	Radiometric methods									
Available emissions	α			β			γ		Non-radiometric methods	
Sample	Urine	Faeces or physical sample	Breath	Urine	Faeces or physical sample	Breath	Urine	Faeces or physical sample	Urine	Faeces or physical sample
Source preparation requirements	Isolation of radioelements from matrix; Chemical separation of elements		None	None or concentration	Separation from matrix	None	None		None or separation of elements	
Methods of detection	Gross α counting α spectroscopy			Liquid scintillation counting (limited energy discrimination)	Gross β counting		Gross γ counting γ spectroscopy		Fluorimetry ICP/MS Delayed neutron assay	
Examples in Annexes	IV: Ra-226 VI: thorium VII. plutonium, americium and curium			II: H-3, C-14 and higher energy mixed fission and activation products	III: strontium IV: Ra-228, Pb-210	II: H-3	I: γ emitting radionuclides		V: uranium VI: thorium	

material by either inhalation or ingestion can often be reliably assessed only from faecal samples.

The lung absorption types and the fractional absorption from the gastrointestinal (GI) tract, f_1, for ingestion and inhalation of various chemical forms of selected radionuclides are given in Tables II and III, respectively. These tables can be used to provide guidance on the choice of bioassay samples for indirect monitoring. In general, for chemical forms of radionuclides with inhalation types specified as V (very rapid lung absorption), F (fast lung absorption) or M (moderate lung absorption), urine monitoring should be used. For inhalation type S (slow lung absorption) faecal monitoring may be preferable, although systemic activity can only be monitored by urine measurements. For ingestion of radionuclides, urine monitoring is to be preferred, even for low values of absorption, as this will provide an estimate of the dose to body tissues for absorbed activity. Faecal monitoring may be needed to determine the total activity taken into the body.

The choice of sample and the method of analysis will also depend upon the physical half-life and the principal radioactive emissions. The radiological characteristics of the radionuclides that are generally of the greatest radiological significance are highlighted in Table IV.

Physical samples include air samples, surface wipes and smears, and examples of other materials from the workplace that can be used to identify the physico-chemical form of radionuclide contaminants. Assessments of doses due to internal exposures based on activity in physical samples are particularly uncertain because of the large variability in workplace conditions and the potential for intakes by individuals.

2.3. METHODS OF ANALYSIS

The choice of method for estimating intakes of radionuclides by workers depends on a number of factors. Where intakes and the corresponding committed effective doses are likely to be low, measurements of physical samples from the workplace may be sufficient. However, where intakes corresponding to doses that are a significant fraction of the dose limit may occur (e.g. effective doses greater than 1 mSv), individual — usually biological — samples should be analysed as far as possible.

Analysis of either biological or physical samples requires the detection and quantification of emissions from the radionuclides present, or other (non-radiometric) methods of estimating activities, by means of appropriate instrumentation. In many cases the radionuclides must first be separated from the sample matrix to allow sensitive and reproducible detection. In other cases limitations of the detectors prevent discrimination between radionuclides that have similar emissions, for

TABLE II. TYPE CHARACTERISTICS OF SELECTED RADIONUCLIDES — INGESTION

Element	Gut transfer factor f_1	Compounds
Hydrogen	1.00	Tritiated water
	1.00	Organically bound tritium
Carbon	1.00	Labelled organic compounds
Sulphur	0.80	Inorganic compounds
	0.10	Elemental sulphur
	1.00	Organic sulphur
Calcium	0.30	All compounds
Iron	0.10	All compounds
Cobalt	0.10	All unspecified compounds
	0.05	Oxides, hydroxides and inorganic compounds
Nickel	0.05	All compounds
Zinc	0.50	All compounds
Selenium	0.80	All unspecified compounds
	0.05	Elemental selenium and selenides
Strontium	0.30	All unspecified compounds
	0.01	Strontium titanate ($SrTiO_3$)
Zirconium	0.002	All compounds
Niobium	0.01	All compounds
Molybdenum	0.80	All unspecified compounds
	0.05	Molybdenum sulphide
Technetium	0.80	All compounds
Ruthenium	0.05	All compounds
Silver	0.05	All compounds
Antimony	0.10	All compounds
Tellurium	0.30	All compounds
Iodine	1.00	All compounds
Caesium	1.00	All compounds
Barium	0.10	All compounds
Cerium	5.0×10^{-4}	All compounds
Lead	0.20	All compounds
Polonium	0.10	All compounds
Radium	0.20	All compounds
Thorium	5.0×10^{-4}	All unspecified compounds
	2.0×10^{-4}	Oxides and hydroxides
Uranium	0.02	All unspecified compounds
	0.002	Most tetravalent compounds, e.g. UO_2, U_3O_8, UF_4
Neptunium	5.0×10^{-4}	All compounds
Plutonium	5.0×10^{-4}	All unspecified compounds
	1.0×10^{-4}	Nitrates
	1.0×10^{-5}	Insoluble oxides
Americium	5.0×10^{-4}	All compounds
Curium	5.0×10^{-4}	All compounds

Note: Excerpted from Ref. [2], Table II-IV.

TABLE III. TYPE CHARACTERISTICS OF SELECTED RADIONUCLIDES — INHALATION

Element	Absorption type	Gut transfer factor f_1	Compounds
Hydrogen	V[a]		Tritium gas, methane
	V[b]		Tritiated water, organic compounds
Carbon	V[a]		Monoxide
	V[b]		Dioxide, organic compounds (vapour)
Sulphur	F	0.80	Sulphides and sulphates: determined by combining cation
	M	0.80	Elemental sulphur. Sulphides and sulphates: determined by combining cation
	F[a]	0.80	Vapour
Calcium	M	0.30	All compounds
Iron	F	0.10	All unspecified compounds
	M	0.10	Oxides, hydroxides, halides and nitrates
Cobalt	M	0.10	All unspecified compounds
	S	0.05	Oxides, hydroxides, halides and nitrates
Nickel	F	0.05	All unspecified compounds
	M	0.05	Oxides, hydroxides and carbides
	[c]		Carbonyl
Zinc	S	0.50	All compounds
Selenium	F	0.80	All unspecified inorganic compounds
	M	0.80	Elemental selenium, oxides, hydroxides and carbides
Strontium	F	0.30	All unspecified compounds
	S	0.01	Strontium titanate ($SrTiO_3$)
Zirconium	F	0.002	All unspecified compounds
	M	0.002	Oxides, hydroxides, halides and nitrates
	S	0.002	Zirconium carbide
Niobium	M	0.01	All unspecified compounds
	S	0.01	Oxides and hydroxides
Molybdenum	F	0.80	All unspecified compounds
	S	0.05	Molybdenum sulphide, oxides and hydroxides
Technetium	F	0.80	All unspecified compounds
	M	0.80	Oxides, hydroxides, halides and nitrates
Ruthenium	F	0.05	All unspecified compounds
	M	0.05	Halides
	S	0.05	Oxides and hydroxides
	F[a]	0.05	Tetroxide
Silver	F	0.05	All unspecified compounds and metallic silver
	M	0.05	Nitrates and sulphides
	S	0.05	Oxides, hydroxides and carbides

TABLE III. (cont.)

Element	Absorption type	Gut transfer factor f_1	Compounds
Antimony	F	0.10	All unspecified compounds
	M	0.01	Oxides, hydroxides, halides, sulphides, sulphates and nitrates
Tellurium	F	0.30	All unspecified compounds
	M	0.30	Oxides, hydroxides and nitrates
	F[a]	0.30	Vapour
Iodine	F	1.00	All compounds, except:
	F[a,d]	1.00	elemental iodine
	V[a]	1.00	methyl iodide
Caesium	F	1.00	All compounds
Barium	F	0.10	All compounds
Cerium	M	5.0×10^{-4}	All unspecified compounds
	S	5.0×10^{-4}	Oxides, hydroxides and fluorides
Lead	F	0.20	All compounds
Polonium	F	0.10	All unspecified compounds
	M	0.10	Oxides, hydroxides and nitrates
Radium	M	0.20	All compounds
Thorium	M	5.0×10^{-4}	All unspecified compounds
	S	2.0×10^{-4}	Oxides and hydroxides
Uranium	F	0.02	Most hexavalent compounds, e.g. UF_6, UO_2F_2 and $UO_2(NO_3)_2$
	M	0.02	Less soluble compounds, e.g. UO_3, UF_4, UCl_4 and most other hexavalent compounds
	S	0.002	Highly insoluble compounds, e.g. UO_2 and U_3O_8
Neptunium	M	5.0×10^{-4}	All compounds
Plutonium	M	5.0×10^{-4}	All unspecified compounds
	S	1.0×10^{-5}	Insoluble oxides
Americium	M	5.0×10^{-4}	All compounds
Curium	M	5.0×10^{-4}	All compounds

[a] SR-1.

[b] SR-2.

[c] SR-1; instantaneous translocation to the bound state; removal from there with a retention half-time of 0.1 d.

[d] The absorption type for elemental iodine was erroneously given as V in the BSS (Ref. [2], Table II-IX, p. 276).

Notes: Excerpted from Ref. [2], Tables II-V and II-IX. Types F, M, S and V denote absorption types with fast, moderate, slow and very fast absorption from the lung, respectively.

TABLE IV. RADIOLOGICAL CHARACTERISTICS OF SELECTED INTERNAL CONTAMINANTS

Radionuclide	Half-life	Principal emission energies (MeV) and yields (fraction/transformation)[a]
H-3	12.3 a	$\beta\ 5.683 \times 10^{-3}$ (1.0)
C-14	5730 a	$\beta\ 4.945 \times 10^{-2}$ (1.0)
P-32	14.3 d	$\beta\ 6.947 \times 10^{-1}$ (1.0)
S-35	87.4 d	$\beta\ 4.883 \times 10^{-2}$ (1.0)
Ca-45	163 d	$\beta\ 7.723 \times 10^{-2}$ (1.0)
Ca-47	4.53 d	$\beta\ 2.411 \times 10^{-1}$ (0.819)
		$\beta\ 8.169 \times 10^{-1}$ (0.18)
		$\gamma\ 1.297$ (0.749)
Fe-55	2.7 a	Auger 6.045×10^{-4} (1.39)
		Auger 5.145×10^{-3} (0.495)
		Auger 5.806×10^{-3} (0.112)
		X ray 5.899×10^{-3} (0.166)
Fe-59	44.5 d	$\beta\ 8.095 \times 10^{-2}$ (0.456)
		$\beta\ 1.492 \times 10^{-1}$ (0.528)
		$\gamma\ 1.099$ (0.561)
		$\gamma\ 1.292$ (0.436)
Co-57	271 d	Auger 5.574×10^{-3} (0.854)
		Auger 6.302×10^{-3} (0.204)
		$\gamma\ 1.365 \times 10^{-1}$ (0.106)
		$\gamma\ 1.221 \times 10^{-1}$ (0.856)
Co-58	70.8 d	$\beta^+\ 2.011 \times 10^{-1}$ (0.15)
		$\gamma\ 8.108 \times 10^{-1}$ (0.994)
		$\gamma\ 5.11 \times 10^{-1}$ (0.3)
Co-60	5.27 a	$\beta\ 9.577 \times 10^{-2}$ (0.999)
		$\gamma\ 1.332$ (1.0)
		$\gamma\ 1.173$ (0.999)
Ni-59	75 000 a	X ray 6.93×10^{-3} (0.2)
		X ray 6.915×10^{-3} (0.102)
		Auger 6.817×10^{-3} (0.11)
		Auger 7.359×10^{-4} (1.3)
Ni-63	96 a	$\beta\ 1.713 \times 10^{-2}$ (1.0)
Zn-65	244 d	$\beta^+\ 1.430 \times 10^{-1}$ (0.0146)
		$\gamma\ 1.116$ (0.507)
		$\gamma\ 5.11 \times 10^{-1}$ (0.0292)
Ga-67	3.26 d	$\gamma\ 3.00 \times 10^{-1}$ (0.168)
		$\gamma\ 1.85 \times 10^{-1}$ (0.209)
		$\gamma\ 8.37 \times 10^{-2}$ (0.287)
		$\gamma\ 9.33 \times 10^{-2}$ (0.383)

TABLE IV. (cont.)

Radionuclide	Half-life	Principal emission energies (MeV) and yields (fraction/transformation)[a]
Se-75	120 d	$\gamma\ 4.006 \times 10^{-1}$ (0.113)
		$\gamma\ 2.795 \times 10^{-1}$ (0.252)
		$\gamma\ 2.647 \times 10^{-1}$ (0.549)
		$\gamma\ 1.360 \times 10^{-1}$ (0.606)
		$\gamma\ 1.211 \times 10^{-1}$ (0.177)
Se-79	65 000 a	$\beta\ 5.582 \times 10^{-2}$ (1.0)
Sr-85	64.8 d	$\gamma\ 5.140 \times 10^{-1}$ (0.980)
Sr-89	50.5 d	$\beta\ 5.833 \times 10^{-1}$ (1.0)
Sr-90	29.1 a	$\beta\ 1.957 \times 10^{-1}$ (1.0)
		$\beta\ 9.348 \times 10^{-1}$ (1.0)[b]
Y-90	2.67 d	$\beta\ 9.348 \times 10^{-1}$ (1.0)
Zr-95	64.0 d	$\beta\ 1.200 \times 10^{-1}$ (0.445)
		$\beta\ 1.088 \times 10^{-1}$ (0.548)
		$\gamma\ 7.567 \times 10^{-1}$ (0.55)
		$\gamma\ 7.242 \times 10^{-1}$ (0.445)
Nb-94	20 300 a	$\beta\ 1.659 \times 10^{-1}$ (1.0)
		$\gamma\ 8.711 \times 10^{-1}$ (1.0)
		$\gamma\ 7.026 \times 10^{-1}$ (1.0)
Nb-95	35.1 d	$\beta\ 4.332 \times 10^{-2}$ (1.0)
		$\gamma\ 7.658 \times 10^{-1}$ (1.0)
Mo-99	2.75 d	$\beta\ 4.426 \times 10^{-1}$ (0.82)
		$\beta\ 1.330 \times 10^{-1}$ (0.166)
		$\gamma\ 7.395 \times 10^{-1}$ (0.122)
Tc-99	213 000 a	$\beta\ 1.013 \times 10^{-1}$ (1.0)
Tc-99m	6.02 h	$\gamma\ 1.405 \times 10^{-1}$ (0.889)
Ru-103	39.3 d	$\beta\ 6.277 \times 10^{-2}$ (0.868)
		$\beta\ 2.386 \times 10^{-1}$ (0.0638)
		$\gamma\ 4.971 \times 10^{-1}$ (0.864)
		$\gamma\ 6.103 \times 10^{-1}$ (0.0528)
Ru-106	1.01 a	$\beta\ 1.003 \times 10^{-2}$ (1.0)
		β 1.508 (0.788)[b]
		$\gamma\ 5.118 \times 10^{-1}$ (0.206)[b]
Ag-108m	127 a	$\gamma\ 7.229 \times 10^{-1}$ (0.909)
		$\gamma\ 6.144 \times 10^{-1}$ (0.908)
		$\gamma\ 4.339 \times 10^{-1}$ (0.903)
		$\beta\ 6.289 \times 10^{-1}$ (0.0854)[b]
Ag-110m	250 d	γ 1.384 (0.243)
		$\gamma\ 9.375 \times 10^{-1}$ (0.343)

TABLE IV. (cont.)

Radionuclide	Half-life	Principal emission energies (MeV) and yields (fraction/transformation)[a]
Ag-110m	250 d	$\gamma\ 8.847 \times 10^{-1}$ (0.729) $\gamma\ 6.577 \times 10^{-1}$ (0.947) β 1.119 (0.0127)[b]
Sb-124	60.2 d	$\beta\ 9.189 \times 10^{-1}$ (0.226) $\gamma\ 6.027 \times 10^{-1}$ (0.958) γ 1.691 (0.477)
Sb-125	2.77 a	$\beta\ 2.155 \times 10^{-1}$ (0.135) $\beta\ 8.698 \times 10^{-2}$ (0.403) $\gamma\ 6.360 \times 10^{-1}$ (0.113) $\gamma\ 6.006 \times 10^{-1}$ (0.176) $\gamma\ 4.279 \times 10^{-1}$ (0.295)
Sb-126	12.4 d	$\beta\ 6.889 \times 10^{-1}$ (0.163) $\gamma\ 6.950 \times 10^{-1}$ (0.996) $\gamma\ 6.663 \times 10^{-1}$ (0.996) $\gamma\ 4.148 \times 10^{-1}$ (0.857)
Sb-127	3.85 d	$\beta\ 3.038 \times 10^{-1}$ (0.356) $\gamma\ 7.837 \times 10^{-1}$ (0.15) $\gamma\ 6.857 \times 10^{-1}$ (0.366) $\gamma\ 4.730 \times 10^{-1}$ (0.257)
Te-127m	109 d	ce 5.645×10^{-2} (0.416)[c] ce 8.332×10^{-2} (0.148) ce 8.392×10^{-2} (0.249)
Te-129m	33.6 d	$\beta\ 6.072 \times 10^{-1}$ (0.31)
Te-131m	1.25 d	γ 1.207 (0.0974) γ 1.125 (0.114) $\gamma\ 8.522 \times 10^{-1}$ (0.206) $\gamma\ 7.737 \times 10^{-1}$ (0.381)
Te-132	3.26 d	$\beta\ 5.940 \times 10^{-2}$ (1.0) $\gamma\ 2.282 \times 10^{-1}$ (0.882)
I-123	13.2 h	$\gamma\ 1.59 \times 10^{-1}$ (0.828) $\gamma\ 1.27 \times 10^{-1}$ (0.135)
I-124	4.18 d	$\beta^+\ 9.736 \times 10^{-1}$ (0.112) $\beta^+\ 6.859 \times 10^{-1}$ (0.112) $\gamma\ 1.691 \times 10^{0}$ (0.106) $\gamma\ 7.228 \times 10^{-1}$ (0.101) $\gamma\ 6.027 \times 10^{-1}$ (0.611) $\gamma\ 5.11 \times 10^{-1}$ (0.453)

TABLE IV. (cont.)

Radionuclide	Half-life	Principal emission energies (MeV) and yields (fraction/transformation)[a]
I-125	60.1 d	X ray 2.747×10^{-2} (0.741) X ray 2.720×10^{-2} (0.398)
I-129	1.57×10^{7} a	β 4.888×10^{-2} (1.0) X ray 2.978×10^{-2} (0.369)
I-131	8.04 d	β 1.915×10^{-1} (0.894) γ 3.645×10^{-1} (0.812)
I-132	2.30 h	β 8.413×10^{-1} (0.176) γ 9.545×10^{-1} (0.181) γ 7.726×10^{-1} (0.762) γ 6.677×10^{-1} (0.987)
I-133	20.8 h	β 4.406×10^{-1} (0.835) γ 5.299×10^{-1} (0.863)
I-134	0.876 h	β 4.628×10^{-1} (0.311) β 9.685×10^{-1} (0.118) γ 8.841×10^{-1} (0.653) γ 8.470×10^{-1} (0.954)
I-135	6.61 h	β 5.349×10^{-1} (0.241) γ 1.260 (0.286) γ 1.132 (0.225)
Cs-134	2.06 a	β 2.101×10^{-1} (0.701) γ 7.958×10^{-1} (0.854) γ 6.047×10^{-1} (0.976)
Cs-136	13.1 d	γ 1.235 (0.198) γ 1.048 (0.797) γ 8.185×10^{-1} (0.997)
Cs-137	30.0 a	β 1.734×10^{-1} (0.946) γ 6.616×10^{-1} (0.85)[b]
Ba-133	10.7 a	γ 3.560×10^{-1} (0.621) γ 3.029×10^{-1} (0.184)
Ba-140	12.7 d	β 3.396×10^{-1} (0.389) β 3.569×10^{-1} (0.229) γ 5.373×10^{-1} (0.244)
Ce-141	32.5 d	β 1.298×10^{-1} (0.7) β 1.809×10^{-1} (0.3) γ 1.454×10^{-1} (0.48)
Ce-144	284 d	β 9.023×10^{-2} (0.758) γ 1.335×10^{-1} (0.108)

TABLE IV. (cont.)

Radionuclide	Half-life	Principal emission energies (MeV) and yields (fraction/transformation)[a]
Tl-201	3.04 d	γ 1.674 × 10^{-1} (0.100)
Pb-210	22.3 a	ce 4.333 × 10^{-2} (0.136) ce 3.012 × 10^{-2} (0.52)
Po-210	138 d	α 5.297 (1.0)
Ra-224	3.66 d	α 5.686 (0.951)
Ra-226	1 600 a	α 4.785 (0.944)
Ra-228	5.75 a	β 9.865 × 10^{-3} (1.0) ce 6.670 × 10^{-3} (0.444) Auger 3.909 × 10^{-3} (0.556)
Th-228	1.91 a	α 5.423 (0.727) α 5.340 (0.267)
Th-230	77 000 a	α 4.688 (0.762) α 4.621 (0.234)
Th-232	1.4 × 10^{10} a	α 4.010 (0.768) α 3.952 (0.23)
Th-234	24.1 d	β 5.055 × 10^{-2} (0.725) ce 7.128 × 10^{-2} (0.103)
U-232	72 a	α 5.320 (0.685) α 5.264 (0.312)
U-233	1.58 × 10^{5} a	α 4.825 (0.844) α 4.783 (0.132)
U-234	2.44 × 10^{5} a	α 4.773 (0.723) α 4.721 (0.274)
U-235	7.04 × 10^{8} a	α 4.398 (0.56) α 4.366 (0.176)
U-236	2.34 × 10^{7} a	α 4.518 (0.738) α 4.470 (0.259)
U-238	4.47 × 10^{9} a	α 4.198 (0.768)
Np-237	2.14 × 10^{6} a	α 4.772 (0.25) α 4.789 (0.471)
Np-239	2.36 d	β 1.258 × 10^{-1} (0.526) γ 2.776 × 10^{-1} (0.141)
Pu-238	87.7 a	α 5.499 (0.716) α 5.456 (0.283)
Pu-239	24 100 a	α 5.156 (0.738) α 5.143 (0.152)

TABLE IV. (cont.)

Radionuclide	Half-life	Principal emission energies (MeV) and yields (fraction/transformation)[a]
Pu-240	6 540 a	α 5.168 (0.734) α 5.124 (0.265)
Pu-241	14.4 a	β 5.236×10^{-3} (1.0)
Am-241	432 a	α 5.486 (0.852) α 5.443 (0.128) γ 5.954×10^{-2} (0.357)
Am-243	7 380 a	α 5.276 (0.879) α 5.233 (0.107)
Cm-242	163 d	α 6.133 (0.741) α 6.070 (0.259)
Cm-244	18.1 a	α 5.805 (0.764) α 5.763 (0.236)

[a] For β^+ and β emissions, the values given are mean energies.

[b] Emission of daughter product.

[c] ce, internal conversion electrons.

Note: Data from ICRP Publication 38 [39].

example some actinides; in these instances the samples must be subjected to radiochemical separation of the element(s) before counting.

Because the particular method chosen for analysis depends on local requirements and resources, there are no internationally agreed procedures for the assay of samples obtained for indirect assessment of levels of radionuclides in the body. A large number of acceptable procedures are available, however, and in use at various laboratories. The preference for a given procedure will depend upon the equipment available, the samples to be analysed, their anticipated levels of activity and the experience of the staff. Even though a service laboratory may choose to use a particular procedure, it is still obliged to ensure that it is capable of meeting the criteria specified by the regulatory authority.

For a particular sample, the method of analysis depends primarily on the emissions available for detection. If only particulate emissions are available (α or β particles), or the yield or energies of any photon emissions are very low, these radionuclides may first need to be separated from the sample matrix to achieve a good and reproducible detection. In the case of urine (or other liquid) samples, however, direct dispersion of a small volume of the sample into a liquid scintillant may be sufficient. Radionuclides emitting penetrating photons can, in general, be quantified in bulk samples. Although these radionuclides may also be detectable in the body by direct counting, detection in excreta may be more practical, or useful for screening purposes.

To detect several radionuclides in a complex mixture additional methods must be applied. For photon emitters, γ spectroscopy is easily performed on a bulk sample. For β emitters, however, only limited resolution of the individual components can be achieved by energy discrimination in the detector output, for example in liquid scintillation counting, because of the continuous distribution of β energies. In addition, this discrimination is only effective when the activity of the higher energy emitter is much less than that of the lower: otherwise, masking may come into play.

Although α particles are monoenergetic, and although α spectroscopy can also be performed on radionuclides once separated from the sample matrix, many α emissions fall into a narrow region of the energy spectrum and cannot always be resolved. In these cases the respective elements must first be separated chemically before the activities are determined.

Non-radiometric methods for quantifying radionuclides are also used in special cases. In general, these methods will be used where they are more convenient for routine monitoring programmes, or have greater sensitivity.

Decisions about the choice of a monitoring programme will be influenced by whether routine or special monitoring is to be undertaken. For *routine or task related* monitoring, the conditions in the workplace should be established and the methods chosen for assessing exposure of the workforce will be based on a knowledge of the radionuclide(s) present and the chemical forms likely to be encountered. For *special* monitoring, which is carried out in actual or suspected abnormal situations, much less will be known about the characteristics of the exposures. Here, a comprehensive monitoring programme will normally be established using both direct and indirect methods until the most appropriate monitoring method emerges on the basis of the results obtained.

3. BIOLOGICAL SAMPLES

3.1. INTRODUCTION

Indirect measurement using biological samples involves the determination of radionuclide concentrations in materials separated from the body, usually in excreta, for the purpose of assessing intakes. The main sources of bioassay data are urine, faeces, breath and blood, although other sources such as hair and teeth have been employed in special cases. In addition, the analysis of activities in a nose blow or nasal swab can be used to provide an early estimate of the identities and relative levels of radionuclides in an inhalation mixture, even though these data can provide only a crude indication of potential intakes.

The choice of bioassay sample will depend not only on the major route of excretion, as determined from the biokinetic model for the particular physico-chemical form and route of intake, but also on such factors as ease of collection, analysis and interpretation. In general, urine samples are easy to collect and will be the basis for the determination of intakes of materials that are readily absorbed, and of the levels of systemic activity in body tissues. Intakes of materials that are poorly absorbed, after either inhalation or ingestion, are usually assessed from faecal samples, but these samples are not readily collected and their measurement results are difficult to interpret. In addition, due consideration must be given to the type of monitoring being undertaken. Special monitoring carried out to assess a specific intake may involve more comprehensive sampling than would be necessary for routine or task related monitoring [3].

For all such sources, contamination of the sample during collection must be avoided. In general, bioassay samples should be collected only outside contaminated work areas following thorough decontamination, especially, of the hands, so that activity in the sample is representative of activity within the body. Practical considerations on sample collection and handling are described in Section 3.7.

In many cases the activity in a single voiding will be insufficient for detection, or for providing an accurate estimate of the total daily excretion, and hence the intake. This problem could be addressed by pooling consecutive samples. In some cases, however, for example shortly after an intake or following an accident when the activity in consecutive samples changes rapidly, it may be preferable to analyse each sample individually and to conduct an appropriate statistical analysis of the data.

All samples of material from the body must be handled with due regard for the possible presence of bacteria, viruses or other biological hazards.

3.2. URINE

Urine contains waste and other materials, especially water, filtered by the kidneys from the blood and collected for periods of up to several hours in the bladder before excretion (Fig. 1, based on ICRP Publication 54 [7]). The nominal daily (24 h) output of urine from 'Reference Man' is 1.4 L [8], but this depends strongly on physiological and environmental conditions. In addition, there are significant diurnal variations in the excretion of many materials, so that in general total daily output must be collected to estimate accurately the daily excretion rate in urine. Ideally, 24 h samples should therefore be used for routine monitoring, but are especially recommended for special

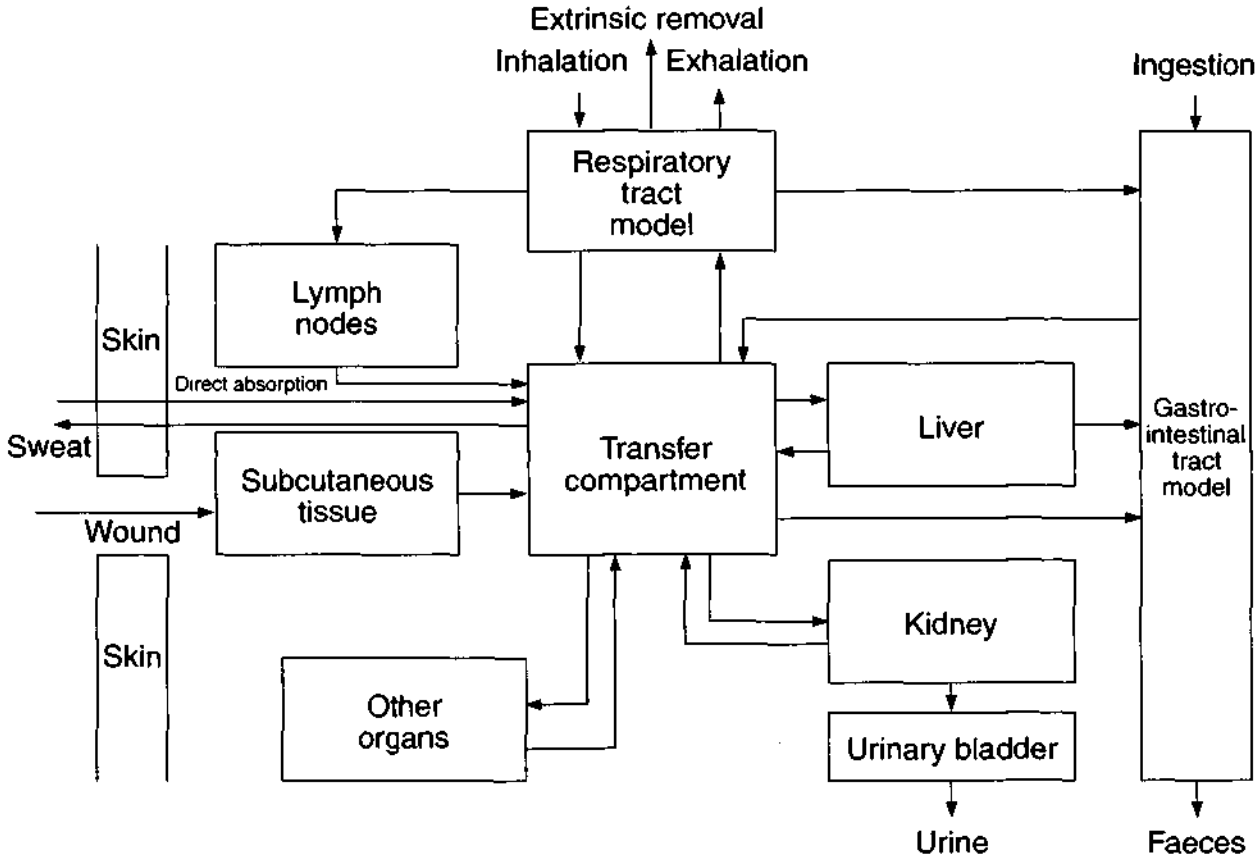

FIG. 1. Routes of intake, transfers and excretion [7].

monitoring, where the potential for significant doses may be greater. Where 24 h samples are not easily collected, the first morning voiding is preferable for analysis [9].

The daily excretion of creatinine, produced as a metabolic product in muscle metabolism, is typically less variable than the volume of fluid lost in urine, although some individuals may still exhibit wide daily variations. Measurements of creatinine levels in urine have therefore been used to estimate 24 h excretion of radionuclides from urine samples collected over part of a day. The use of creatinine to normalize excretion rates has been reviewed by NCRP [9], and its application in the estimation of 24 h excretion of uranium has been discussed by Jackson [10] and Karpas [11].

An exception to the need for 24 h collection is the excretion of tritiated water, for which any sample provides an accurate estimate of the concentration in total body water. In this case, frequent sampling improves the time resolution needed for biokinetic modelling.

Because of the times required for many materials to enter the blood and to be filtered out by the kidneys, together with the problem of the mixing of any excreted activity with the accumulated contents of the bladder, results from samples of urine obtained soon after an incident must be interpreted cautiously. It is recommended that a urine sample be collected as soon as possible after exposure, another after several hours and subsequently at intervals, in order to aid in this interpretation. The proper sampling interval depends on both the excretion rate and the physical half-life of the radionuclide(s) involved; specific guidance is provided in the Annexes to this Report for selected radionuclides.

As an example, because radioiodine levels in urine samples would be expected to decrease by about three orders of magnitude in the first week after an intake [12], the greatest sensitivity for detecting an intake would be obtained within the first 24 hours. In contrast the urinary excretion of plutonium decreases only slowly with time after intake; consequently, the systemic content of plutonium can be assessed from urine measurements for many years after the intake.

Some chemical forms of the radioisotopes in urine, such as bicarbonate and many biological substrates, can be degraded within the sample as a result of biological contamination and other processes. In addition, radioactive material can be lost from solution by adsorption onto the surfaces of some containers. For these reasons samples must often be stabilized until analysis by refrigeration or freezing, or by the addition of a carrier or of an acidic, basic or other preservative, such as thymol, as is appropriate for the particular situation.

3.3. FAECES

Faecal samples consist of waste products transported through the GI tract and cellular debris sloughed from the walls of the intestines. They contain materials cleared from the lung, systemic material excreted into the GI tract, primarily in bile, and material passing unabsorbed through the GI tract following ingestion (Fig. 1).

The nominal transit time for material passing directly through the GI tract is about two days [13], but this varies considerably with diet, health of the individual and other factors. In addition, the mass and composition of individual voidings is quite variable. For these reasons it is usually necessary to collect total faecal elimination over a period of three to four days, in order to obtain reliable estimates of daily excretion rates.

Where there is a requirement to collect faecal samples for the assessment of an intake by inhalation, it will be necessary to allow for the presence of any ingested materials, especially in early samples.

Faecal samples are particularly subject to biodegradation; therefore they should be analysed promptly, ashed or preserved by deep freezing. Such samples can also contain significant levels of potentially toxic biological contaminants.

Wet or dry ashing procedures can be used in the analysis of faecal samples. Radionuclides may then be analysed either in the volatile fraction or in the residual ash.

3.4. BREATH

After uptake into tissues, significant exhalation of radionuclides occurs only for intakes of a few materials which are either metabolized to gases or volatile liquids, or produce gaseous decay products. For these intakes, breath samples can provide a ready source of activity directly released from the blood, and free of most interfering radionuclides. For example, about one third of an intake of tritiated water is excreted in breath, whose specific activity rapidly reaches equilibrium with that in body water. Many forms of ^{14}C labelled organic materials are metabolized to carbon dioxide (CO_2), which is eliminated mainly in breath. Analysis of radon (^{222}Rn) or thoron (^{220}Rn) can provide a sensitive method of monitoring the body content of ^{226}Ra or ^{232}Th, respectively, although it is best used to complement other assessment methods.

Breath sampling requires the worker to breathe into a collecting apparatus for up to 30 min, depending on the volume required. Specific components of the breath can be extracted by appropriate techniques, for example the trapping of water vapour in a low temperature trap, or the absorption of CO_2 on ethanolamine based absorbers. These materials may then be counted directly, or the radionuclides extracted for analysis.

3.5. BLOOD

Blood samples provide the most direct source for estimating circulating internal contamination. The majority of radionuclides, however, are rapidly cleared from the blood. Because of this, and recirculation in the body, measurements of activity in the blood are generally poor indicators of the total systemic content. However, in a few exceptional circumstances blood samples may be useful for monitoring systemic activity. Examples include ^{51}Cr labelled erythrocytes (red blood cells), tritiated water and radionuclide complexes bound to carriers in the blood. Blood samples are not frequently used because of medical constraints on the sampling procedure; they should be collected only by qualified medical persons under carefully controlled hygienic conditions.

Depending upon the purpose of sampling, blood samples may need to be stabilized by the addition of preservatives to prevent clotting. Blood fractionation procedures are well established for separating the blood into its various constituents, which can then be analysed individually for activity.

In an accident situation involving potentially high exposures, blood samples are frequently collected for analysis of white blood cell counts, chromosomal aberrations or other indicators of radiation exposure. In such cases, collection of additional samples for activity analysis may provide a useful indicator of the extent of the exposure.

3.6. OTHER BIOLOGICAL SAMPLES

Other biological materials separated from the body, which include saliva, teeth, hair, nails and nose blows, can be used to demonstrate intakes of radionuclides, but owing to the uncertainties in the models which predict intake and dose from activity in these sources, these results are generally not used as a basis for dose calculation. Nose blows specifically are useful for the purpose of screening for intakes, and for identifying other radionuclides in an intake mixture that cannot be detected by direct methods. Nose blow samples are frequently taken to screen for α contamination. In this case samples are treated with acid and the solution obtained evaporated on a tray for α counting. However, γ contaminants can be measured in the same samples.

Samples of hair have been analysed for plutonium. Measurements have been made as a function of distance from the root [14], to infer a pattern of concentration in the bloodstream over a period of weeks. Caution is needed to ensure that hair care products have not resulted in contamination of the sample with naturally occurring radionuclides, such as uranium. Teeth incorporate many of the elements of bone, such as strontium, and may provide an indication of childhood exposures [15]. However, the surface of the teeth can be contaminated as a result of recent intakes. Tissue samples obtained as a result of surgical or autopsy procedures can also be used as a basis for estimating the body content of radionuclides [16]. Analysis of tissue excised from contaminated wounds can also assist in dose assessment [17]. The problem of skin contamination is considered in Section 4.5.

3.7. SAMPLE COLLECTION AND TRANSPORT

The collection of appropriate biological samples is an integral part of monitoring for the detection of intakes of many radionuclides. Consequently workers must be given training in the requirements for sample collection and the appropriate methods to be used.

In many cases where prompt excretion of an intake, or part of an intake, may occur it is useful to collect routine samples after a weekend or longer period away from workplace exposure, in order to separate the transient excretion of short term retained materials from the excretion of the more important long term retained components [18, 19]. Such a practice, for example, can separate faecal excretion as a result of recent intakes by ingestion from the slow clearance of intakes by inhalation of insoluble particulate material. Otherwise even small intakes by ingestion may mask the low levels of long term clearance from the lung, resulting even from large intakes by inhalation.

For collection of urine samples, workers should be provided with clean containers having tight fitting lids. The time of the collection period should be recorded, along with the appropriate sample identification. Unless samples are to be processed promptly, preservatives may need to be added to the sample containers, or the samples may be stored at reduced temperature.

For the scheduling of sample collection, due consideration should be given to whether medical interventions, such as chelation therapy, the administration of diuretics, blocking agents or laxatives, could have influenced the pathways or rates of excretion of radionuclides.

Clinical faecal sampling kits, such as those used by hospitals, are available from suppliers and can be provided to the worker to aid in sample collection. Ease of sample collection, and explanation of the importance of the sample for dose assessment to the worker, will greatly improve compliance with requests for samples. Faecal collection containers should be lined with plastic bags that can be ashed with the sample during the subsequent analysis.

Procedures for sample transport should be established which will need to take account of:

(a) Appropriate identification of the samples,
(b) Need for pre-treatment of the samples,
(c) Appropriate packaging and labelling of containers,
(d) Prevention of biological or radionuclide contamination,
(e) Specification of 'chain of custody' requirements (Section 7.2).

When samples are to be transported across national borders, differing regulations may need to be taken into account.

4. PHYSICAL SAMPLES

4.1. INTRODUCTION

Physical samples include air samples, surface swipes and smears, and other material samples from the workplace. They provide an indication of the radionuclides and their relative concentrations in the working environment. However, because models which predict intake from air or surface concentrations depend strongly on such factors as breathing rates and other behaviours, estimates of intake based on these measurements can be very uncertain. Moreover, physical samples cannot be used as a basis for determining individual biokinetic characteristics. Finally, these samples cannot always be repeated if the analytical result is suspect or lost and workplace conditions have changed.

Nevertheless, workplace samples can provide a useful basis for determining the need for individual bioassay monitoring (Ref. [3], Appendix I). For intakes of some radionuclides which emit no penetrating radiations, and which result in only very low levels in excreta, physical samples may provide the best estimates of intake, when combined with reasonable assumptions about the exposure conditions.

Following inhalation of radionuclides the correct interpretation of bioassay results and the consequent dose assessment will depend upon a knowledge of the particle size distribution, as this influences deposition in the respiratory system. If the particle size distribution is unknown, and exposures are low, the assumption of a default activity median aerodynamic diameter (AMAD) of 5 μm is sufficient (Ref. [3], para. 3.40). However, if the workplace exposure is expected to exceed the investigation level, then workplace and individual specific parameters may need to be developed to estimate the committed dose more accurately (Ref. [3], para. 3.33). On some occasions the choice of the appropriate bioassay technique and schedule may also depend on the particle size distribution. For example, early faecal sampling is indicated following inhalation of insoluble particles.

The particle size characteristics of airborne activity can be assessed by the use of either static (fixed) air samplers or personal air samplers [20, 21]. The most common equipment used for particle size characterization is the cascade impactor. Personal cascade impactors are also coming into widespread use [22]. Each stage of the cascade is analysed and a calibration factor is applied to obtain the particle size distribution.

4.2. STATIC SAMPLERS

Average values of radionuclide concentrations in the workplace air can be measured using static air samplers, which collect radioactive materials from air flowing through the instrument at a prescribed flow rate for a known period of time. These flow rates should be carefully calibrated. A fixed position impactor should be positioned at around 1.5 m from the ground, at the site where occupational exposures may occur. The measured values of interest, however, apply only at the site of sampling, and may differ from that in the breathing zone of the worker by up to an order of magnitude [23–25]. Moreover, static sampling data usually contain no information on chemical form or particle size, and may cover periods when the worker was not present. For these reasons, static air monitoring is commonly used only to provide information on workplace conditions or to assist in interpreting the results of individual monitoring, but, except for radon, is rarely used to estimate intakes. In setting up static sampling procedures, the sampling time should be determined in such a way as to avoid excessive saturation at any of the sampling stages.

4.3. PERSONAL AIR SAMPLERS

Personal air sampling is a form of individual monitoring whereby samples of airborne activity in the breathing zone of the worker are collected using self-powered equipment worn or carried by the worker. The sampler flow rates must be carefully calibrated particularly as intakes estimated by personal air sampling may result in assigned doses. Because activity concentrations can vary by an order of magnitude or more within short distances of the worker, particularly where the worker's own activities lead to resuspension of surface materials, personal air sampling can provide a much more reliable basis for estimating intake than static air monitoring. Moreover, personal air samplers (PASs) can be worn under some forms of protective equipment, and can thus be used to check on the adequacy of protective measures.

Some types of personal air samplers prevent particles of non-respirable size from reaching the collection medium [26]. In addition, personal cascade impactors provide information on determining particle size distributions [27]. However, the determinations of chemical form and solubility characteristics require separate analyses. Personal air sampling data, together with reasonable assumptions on such factors as chemical form, particle size and breathing rates, are now commonly used to estimate intakes of radionuclides which are difficult to assess by other techniques. PASs are also useful for providing assurance at the end of a potential exposure period that unexpected intakes by inhalation are unlikely to have occurred. It should be noted, however, that this may not be reliable for high specific activity particles ('hot particles'), where the sample collected may not be representative of the activity inhaled [28–30].

4.4. SURFACE CONTAMINATION MEASUREMENTS

The models that relate radionuclide concentration on working surfaces to intake are particularly uncertain and depend strongly on such factors as resuspension rates and particle size for inhalation, and hygiene practices for ingestion. Therefore, surface activity measurements are usually used only to indicate levels of activity in the work environment which justify other forms of monitoring, and not to estimate intakes.

Surface contamination monitoring employs a variety of techniques specific to the expected forms of contamination, but generally involves swiping a defined area of the surface to remove loose radioactive material. Material collected on the swipe is then analysed, either directly or following extraction from the matrix, using the same methods as are employed for biological samples. The efficiencies of collection of radioactive material should be determined for the particular combination of surface and swiping material for the particular procedure employed, but are typically about 10% for a moist swab on a semi-porous surface.

4.5. SUPERFICIAL CONTAMINATION OF WORKERS

In many cases, the first (and perhaps only) indication that an intake of radioactive materials may have occurred is a finding of superficial contamination on a worker's clothing or skin. There are no reliable models available for predicting what fraction of superficial contamination may have been inhaled, ingested or absorbed through intact skin. However, some 'rules of thumb' for relating contamination measurements to intakes have been published. One such example relates activity found on a nasal smear to that inhaled. The nostrils are swabbed with moistened cotton tipped applicators (one per nostril), which are then counted. If only one shows activity, it may be indicative of a transfer of contamination from the hand to the nose, rather than an inhalation. Heid and Jech [31] reported that for a 4 μm AMAD particle size about 10% of the total intake could be found in the nostrils, based on the ICRP Task Group lung model [32]. A similar value can also be derived from the new ICRP respiratory tract model [33]. However, such results must be interpreted with caution. The intake, relative to the activity found in the nostrils, may be affected by whether or not the person breathes by mouth, the time elapsed between the intake and the sample collection, whether or not the nose was blown or the person sneezed, and the particle size distribution. The finding of skin contamination about the nose and mouth can also be used as an indicator of a possible intake. However, such measurements are best used to indicate the relative magnitude, and therefore the possible severity, of an intake, rather than to quantify the intake precisely.

5. TECHNIQUES OF ANALYSIS

5.1. INTRODUCTION

Before analysing a biological or physical sample to determine its radionuclide content, decisions need to be taken as to the appropriate methods of detection and the need, if any, for processing of the sample prior to counting. These decisions will be based on the radionuclide(s) involved, the chemical and physical form(s), the possible presence of interfering radionuclides and the levels of detection sensitivity necessary to meet monitoring requirements.

In many cases, for example with routine samples obtained from a well characterized workplace, there may be a good knowledge of the radionuclides that are likely to be present in the sample. In these circumstances the available emissions can be readily predicted. In other cases it may be necessary to screen the sample to identify any emissions of α or β particles, or X or γ photons [9].

Possible sources of interference in biological samples may include the presence of naturally occurring radionuclides resulting from intakes in food and water, radionuclides from previous intakes, radiopharmaceuticals administered for diagnostic or therapeutic purposes, and prescription or non-prescription drugs, such as antacids, that affect the chemical processing of the sample. Workplace air samples may include naturally occurring radon and thoron progeny, or high levels of dust loading, which may also affect sample analysis.

The measurement sensitivity needed for a specific radionuclide will depend on the level of intake that must be detected to demonstrate compliance with regulatory requirements. In general it will be necessary that the minimum detectable activity (MDA) (Section 6.2) for an analysis be less than the derived recording level (DRL) for the specific radionuclide and monitoring programme. The DRL for a particular sample is equal to the concentration of the radionuclide in that sample that corresponds, through assumptions about the time and pattern of intake, and the use of biokinetic models, to an intake at the recording level (RL) [3]. Typical MDAs for techniques commonly used for measuring radionuclides in biological samples are included in the Annexes.

5.2. RADIOCHEMISTRY

For accurate measurement of α and β emitters, a source containing the analyte must in general be separated from the bulk of the sample matrix and from other interfering radionuclides. This usually involves:

(a) Sample preparation and preconcentration,
(b) Chemical separation of the radionuclides,
(c) Preparation of the source for counting.

However, for some pure emitters in a simple matrix, such as ^{3}H or ^{14}C in urine, very little preparation or preconcentration is needed before measurement.

5.2.1. Sample preparation and preconcentration

The usual aim of sample preparation is to reduce the sample to its inorganic constituents, unless organic components are to be analysed. Simple aqueous samples are frequently evaporated to dryness on a hotplate.

To remove all the organic materials which can interfere in the radiochemical procedure, biological samples are mineralized by dry ashing in a muffle furnace or microwave oven, and then treated by oxidation with strong acidic reagents (wet ashing), followed by evaporation to incipient dryness. These steps may need to be repeated. The criterion for successful mineralization is to obtain a white residue from successive wet and dry ashings. Care, however, must be taken to prevent the loss of volatile elements such as caesium and polonium during the ashing process.

Some complexes like Pu-DTPA need vigorous mineralization to destroy completely the chelate and to liberate the Pu^{4+} cation. For highly insoluble matrices, such as plutonium oxide fused at high temperatures, or samples on fibreglass filters, treatment with hydrofluoric acid may be needed.

Preconcentration is the process of separating the radionuclides of interest from the bulk of the other inorganic materials by co-precipitation techniques: for example, iron hydroxide or lanthanum fluoride in phosphate is used for actinides, calcium oxalate in phosphate for calcium, strontium and actinides, and bismuth phosphate for americium. Addition of the stable element as a carrier followed by a subsequent precipitation is commonly used for ^{32}P and ^{90}Sr. For the analysis of organic material containing ^{14}C and ^{3}H, specific techniques for the collection of these radionuclides during ashing procedures are needed.

5.2.2. Radionuclide separation

Radiochemical separation is carried out to isolate the analyte of interest. The ash or precipitate resulting from preconcentration is redissolved and chemical separation of the radionuclides is achieved by (a) solvent extraction, (b) ion exchange separation or (c) co-precipitation.

Solvent extraction procedures, which may be specific to oxidation state, can be used to separate the radionuclide into an organic phase for further analysis or for direct counting by liquid scintillation.

In ion exchange separation, including chromatography techniques, solutions of the analyte at a controlled pH, commonly acid, are passed through an ion exchange column or other medium which retains the radionuclide of interest. The retarded material can then be eluted or otherwise separated for counting. Different radionuclides can be separated sequentially by altering the characteristics of the eluent.

Co-precipitation can also be used to extract specific radionuclides from solution following the addition of a stable carrier. The precipitate may be assayed directly for radionuclide content, or used in further separation procedures. For example, ^{90}Sr may be co-precipitated with calcium and measured by β counting on planchettes.

Advanced methods of radiochemical separation have been developed based on mass spectroscopy, sometimes involving accelerators, laser induced resonance ionization or other ion selection methods. Although these methods offer extremely high sensitivities they are usually too expensive for routine use.

5.2.3. Preparation of the source for counting

The requirements for sample preparation depend upon the type of measurement to be performed and the radionuclide emissions. For gross α and β counting, the purified solution can be evaporated directly onto an appropriate substrate for counting. This could be a filter paper or a stainless steel planchette. Source preparation for β emitters can be achieved by precipitation of different anions such as hydroxide, oxalate, sulphate, carbonate and phosphate. The precipitate is separated and transferred to a filter paper or planchette for counting. For photon (γ or X ray) spectrometry, minimal source preparation is normally required, but due consideration should be given to determining the attenuation in the sample and providing a reproducible geometry.

Sources for α spectrometry must be very thin and planar (i.e. essentially 'massless') to preclude self-absorption and the resultant energy degradation. They can be prepared by the following techniques:

(a) Electrodeposition of the purified radionuclides onto polished stainless steel discs from different media;
(b) Co-precipitation of the actinide with microgram quantities of lanthanum, cerium or neodymium fluoride and collection onto a small filter paper for counting;

(c) Solvent extraction onto an organic film previously deposited onto a stainless steel disc [34];

(d) Solvent extraction followed by evaporation onto a planchette. (This method frequently gives results of lesser quality than those of other methods.)

5.3. DETECTION METHODS

The quantitive assessment of radionuclides in monitoring samples requires specialized equipment for α, β and γ counting. A number of non-radiometric methods may also be used.

5.3.1. Radiometric techniques

5.3.1.1. Alpha particle counting

When identification of the radionuclide is unnecessary, gross α counts can be performed with gas flow proportional counters or scintillation counters. Gas flow counters usually use a counting gas of 90% argon and 10% methane, and typically have detection efficiencies of around 45%. Scintillation counters employ a thin, transparent membrane coated with a scintillator such as ZnS that is placed in proximity to the sample. Scintillations in the ZnS are then viewed by a photomultiplier tube. These counters typically have efficiencies of about 45%, and usually have lower backgrounds than proportional counters.

At relatively high levels of activity, α emitters may also be detected by liquid scintillation counting, using techniques similar to those employed for β emitters (discussed later). A common technique for assessing plutonium is to extract the radionuclide from acid solution into an organic phase containing a scintillant. The extraction process would normally be carried out in the scintillation vial prior to counting [35].

When radionuclide identification is required, α spectrometry is employed. Alpha spectrometers are most often silicon surface barrier detectors in a vacuum chamber. Alpha spectrometers have low backgrounds, but when very low levels of activity (mBq) must be detected, long counting times (days to weeks) are needed. Such α spectrometers have excellent energy resolution, but some radionuclides, for example, ^{239}Pu and ^{240}Pu, have very similar α energies which cannot be distinguished, and therefore are normally reported together. Gridded ionization chambers can also be used for α spectrometry but they have poorer resolution.

In some circumstances ultra-sensitive detection of α emitters may be required. One method is fission track autoradiography, in which the sample to be analysed for fissile radionuclides (^{239}Pu or ^{235}U) is placed in contact with a fission track detector and irradiated by neutrons in a reactor. The density of fission tracks recorded on the detector (a thin sheet of polycarbonate or a nuclear emulsion, for example) is proportional to the concentration of the fissile nuclide in the sample. Because this technique cannot distinguish between fissile radionuclides, elaborate radiochemical pre-separation is required to avoid traces of other fissionable materials.

In some cases, this technique can be used to obtain information on the distribution of ^{235}U or ^{239}Pu in a biological sample by placing a thin section of the sample in contact with the detector prior to irradiation.

A less expensive autoradiography method is the use of an α-sensitive track detector, such as polyallyl diglycol carbonate (CR-39). The passing α particles create damage tracks in the detector, and when the entire detector is etched in sodium or potassium hydroxide solution, erosion proceeds more rapidly along the damage tracks than for the bulk material. Consequently, pits are created which are visible under a microscope and can be counted; again, the density of α tracks on the detector is proportional to the concentration of α emitters in the sample. In most practical circumstances this method is unable to distinguish between α emitters, and so radiochemical separation is necessary before use. Track detectors are commonly used to monitor radon in the workplace.

5.3.1.2. Beta particle counting

The liquid scintillation coincidence counter or spectrometer is the instrument of choice for the measurement of β emitters, particularly for low energy β emitters such as ^{3}H, ^{14}C, ^{35}S and ^{241}Pu. The radionuclide is dispersed in an organic liquid scintillator that fluoresces when exposed to α or β radiation, emitting photons in the visible or ultraviolet region of the electromagnetic spectrum. The numbers of photons emitted is proportional to the energy deposited in the solvent. The photons are detected by two photomultiplier tubes operated in coincidence to reduce background.

Counting efficiency is strongly dependent on quenching of the detection process, especially for low energy β emitters. Quenching may arise from interference with the fluorescence process because of chemical interactions between the sample and scintillator, or absorption of the emitted light because of strong colouration of the sample solution.

A number of techniques are used to address this quenching effect including:

(a) The use of internal standards;

(b) The use of a physico-chemical procedure, such as distillation or the addition of activated charcoal, to remove any quenching agents;

(c) Analysis of spectral shape, as quenching will progressively shift the counts to lower energy channels in the counting equipment;

(d) External standardization, where scintillations in the samples induced by an external source are compared with those induced in an unquenched sample. Analysis of the spectral shape may also be used as part of this technique.

Because β particle energy spectra are continuous up to a characteristic maximum energy, β liquid scintillation measurements have limited capability to identify radionuclides. Some discrimination is possible by dividing the spectrum into three regions of low (e.g. ^{3}H), medium (e.g. ^{14}C) and high (e.g. ^{32}P or ^{90}Y) energies. The presence of counts in a lower energy region (or 'window'), but not in a higher energy region, restricts the possible β emitters that may be present in the sample. However, the presence of high energy emitters in a sample can mask the simultaneous presence of lower energy emitters; consequently, for simultaneous determination, the concentration of a low energy emitter in a sample must be significantly greater than that of the higher energy emitters in the same sample.

Particular attention must be devoted to variation in background activity levels due to ^{40}K in a sample such as urine. Glass vials also contain ^{40}K, so plastic vials are often used when a low background is needed. The same counting system can also be used without a scintillator in the cocktail for measurement of high energy β particles, using the Cerenkov effect.

Gas flow, Geiger–Müller and proportional counters are commonly used for the measurement of samples deposited on planchettes, but only for β emissions with energies above several hundred keV. In this application, the counter must be calibrated for the energy of interest.

5.3.1.3. Photon counting

High energy photon emitters can be measured in bulk samples with a minimum of preparation. Measurements of low energy photon emitters, however, are not performed with bulk samples because of serious photon attenuation.

Most of the low level γ ray measurements for bioassay are performed with NaI(Tl) scintillation and Ge(Li) or high purity germanium semiconductor detection. NaI(Tl) detectors are commonly used for detecting photon emitters, and have a high counting efficiency but a low spectral energy resolution. They are best used for assessing samples containing a single radionuclide, or radionuclides with well separated photon energies.

The major advantage of semiconductor detectors such as Ge(Li) or high purity germanium is their good energy resolution, which allows an unambiguous identification of various photon emitting radionuclides. However, they must be operated at liquid nitrogen temperature, lack sensitivity in comparison with NaI(Tl) crystals, and are more expensive to purchase and maintain.

Low energy photons, usually X rays, are measured with semiconductor detectors such as Si(Li) or high purity germanium. These detectors must be well shielded using lead or steel that contains very low levels of radionuclide contaminants, and they should also be located in a low level background counting room. Further information on X or γ photon detectors is given in the Safety Series Practice, Direct Methods for Measuring Radionuclides in the Human Body [4].

5.3.2. Non-radiometric techniques

5.3.2.1. Luminescence methods

Routine measurements of natural or depleted uranium in urine are frequently made with fluorimetric techniques, which determine the mass of uranium present. Such methods may be perfectly adequate for bioassay monitoring where the isotopic content of the uranium is known and where chemical, rather than radiological, toxicity is an issue. Fluorimetric methods are faster to perform than α spectrometric methods, and so are particularly useful after an accident, when rapid assessment is important. However, levels of enriched uranium that are of radiological concern are below the mass limit of detection of fluorimetric methods.

(a) Alkaline fluoride fusion

This method involves taking an aliquot of less than 1 mL of urine, drying carefully to a red heat to remove carbon, melting the residue in the flux of an alkali (usually sodium) fluoride, and cooling to a glass bead that is read on a fluorimeter. The MDA of this method is about 3 μg·U/L.

(b) Kinetic phosphorescence analysis (KPA)

This method uses a computer controlled, laser induced, time resolved luminescence analyser. A 10 mL urine sample is wet and dry ashed to remove organics; the uranium is extracted into an ion exchange resin and then eluted; chlorides are removed; and the uranium solution is mixed with a complexing agent for KPA analysis. A laser is used to stimulate uranium ions in reference and sample cells; de-excitation of the ions results in emission of photoluminescence photons that are converted to an electronic signal by photomultiplier tubes; analysis of the light intensity as a function of time permits determination of the uranium ion concentration in the sample cell. The MDA of this method is about 7 ng·U/L. The analytical measurement itself is rapid, and so this method offers high throughput rates [36].

5.3.2.2. Delayed neutron counting

The uranium content of urine or other samples can be obtained by measuring the delayed neutrons resulting from fission of the uranium after irradiation by thermal neutrons in a nuclear reactor [37]. For interpretation in terms of total uranium content, the fraction of ^{235}U in the sample must be known.

5.3.2.3. Inductively coupled plasma/mass spectrometry (ICP/MS)

ICP/MS may be used for measuring plutonium, uranium, thorium and other radionuclides in bioassay samples. Because the analytical method is based only on the mass of the radionuclide, chemical separation of elements with isotopes of the same mass number (e.g. analysis for ^{238}Pu in the presence of ^{238}U) may be required before the measurement. The method has the advantage of permitting simultaneous measurement of isotopes of the same element with similar α particle energies (e.g. ^{239}Pu and ^{240}Pu).

In this method, an inductively coupled plasma ionization source is connected to a mass spectrometer so that a small fraction of the plasma is drawn into the spectrometer. Samples must be in solution with relatively low total dissolved solids, and so typical sample preparation methods may need to be used to prepare the solutions for analysis, as for α spectrometric measurements. Because very small sample volumes are analysed, care must be taken to ensure sample homogeneity. In addition, because of interference from water, argon and common solvents, the method is normally restricted to radionuclides of mass greater than 60.

The method has an MDA of about 25 pg ^{238}U/L, and about 200 Bq/L, for 93% enriched uranium. Although the method is in routine use for monitoring uranium workers at some facilities [38], the equipment is expensive, and has not come into widespread use.

5.4. DETERMINATION OF SAMPLE ACTIVITY

The various methods of sample analysis for specific radionuclides in biological and physical samples will yield measured levels of activity, for example as counts per minute. These sample counts, after correcting for any background counts arising either from the apparatus or from other sources of activity in the sample(s) under test, must then be converted into absolute levels of the radionuclide in the sample to give, for example, Bq·kg^{-1}. The method adopted for determining the absolute amount of

TABLE V. RADIOLOGICAL CHARACTERISTICS OF SELECTED TRACER RADIOISOTOPES

Radionuclide	Half-life	Principal emission energies (MeV) and yields
Y-88	107 d	γ 0.898 (0.934), γ 1.836 (0.993)
Th-229	7340 a	α 4.846 (0.563), α 4.902 (0.102)
Pu-236	2851 a	α 5.721 (0.318), α 5.768 (0.680)
Pu-242	376 300 a	α 4.857 (0.224), α 4.901 (0.775)
Am-243	7380 a	α 5.276 (0.879), α 5.233 (0.107)

Note: Data from ICRP Publication 38 [39].

the specific radionuclide(s) under analysis will depend upon the method of analysis adopted. Detailed examples are given in the Annexes, but the methods used include:

(a) Comparison of γ counts obtained from the sample with those from standards containing known amounts of the specific radionuclide, measured under the same counting geometry;
(b) Yield determination for α or β emitters using alternative isotopes (tracers) of the radionuclide under analysis;
(c) Internal and external standardization for β emitters analysed by liquid scintillation counting.

Non-radiometric techniques, such as fluorimetry or ICP/MS, will also give absolute levels of radionuclides in samples after calibration with suitable standards.

These methods generally depend upon the availability of accurately calibrated standards, which will commonly be obtained commercially but they can also be prepared 'in-house' by absolute counting techniques.

Many radiochemical techniques rely on separation procedures for which the recovery can be quite variable. It is then essential to use methods that allow determination of the yield obtained. To obtain a quantitative estimate of the amount in the original sample, a tracer must be added to the sample before the beginning of the analytical procedure. This tracer will generally be another isotope of the element being assayed. Some of the most commonly used radioactive tracers for α emitters are ^{229}Th, ^{232}U, $^{236/242}$Pu and ^{243}Am, and for β emitters ^{85}Sr and ^{88}Y (Tables IV and V). Alternatively stable isotopes can be used, with recovery of the tracer assayed by gravimetric methods. The sample preparation method must ensure complete isotopic exchange between the analyte and the tracer. The fraction of the tracer that is subsequently obtained at the end of the separation procedure is termed the chemical yield, or recovery, *Y*.

This technique can be applied to the sequential analysis of samples for a number of radionuclides, provided all the required tracers are added to the original sample before any analytical procedures commence.

6. ACTIVITY CALCULATION AND STATISTICAL CONSIDERATIONS

6.1. CALCULATION OF SAMPLE ACTIVITY

The activity in the sample is calculated as

$$A = \frac{1}{F}\left(\frac{N_s}{t_s} - \frac{N_b}{t_b}\right) \tag{1}$$

where N_s is the number of counts in the peak from the sample, N_b is the number of counts in the same region from the blank, F is the efficiency of the method and t_s and t_b are the counting times for the sample and the blank, respectively.

When a tracer is added to determine yield, F may be calculated using

$$F = \frac{1}{A_t}\left(\frac{N_t}{t_s} - \frac{N'_b}{t'_b}\right) \tag{2}$$

where N_t is the number of counts in the tracer region, N'_b is the number of counts in the tracer region from the blank, t_s and t'_b are the counting times for the tracer (and sample) and the blank, respectively, and A_t is the activity of the tracer added (Bq).

6.2. DETECTION LIMITS

In the reporting of an analytical measurement, it is important to specify the detection capability of the method. The minimum significant activity (MSA) is the smallest signal taken to be significantly in excess of the background response for the specific measurement method. The minimum detectable activity (MDA) corresponds to the level of activity which is required to ensure, with some chosen level of confidence, that the net signal will be detected.

The definitions and formulas are specified in the Safety Guide on Assessment of Exposure due to Intakes of Radionuclides, Appendix II [3]. When α, the probability of a type I error (false positive), and β, the probability of a type II error (false negative), are both set equal to 0.05,

$$\mathrm{MSA} = \frac{1.65}{F}\sqrt{\frac{n_b}{t_s}\left(1+\frac{t_s}{t_b}\right)} \tag{3}$$

and

$$\mathrm{MDA} = 2\mathrm{MSA} \text{ (for most cases)} \tag{4}$$

where n_b is the blank sample count rate.

When the Poisson distribution of counts is not adequately approximated by a normal (Gaussian) distribution, because the total counts are low (say ≤10),

$$\mathrm{MDA} = \frac{3}{Ft_s} + 2\mathrm{MSA} \tag{5}$$

For analytical processes that end in non-counting types of measurements, and have continuous output signals such as uranium fluorescence analysis,

$$\mathrm{MDA} = 2\mathrm{MSA} \tag{6}$$

For samples and blanks with equal counting times t,

$$\mathrm{MSA} = \frac{1.65}{F}\sqrt{\frac{2n_b}{t}} = \frac{2.33}{F}\sqrt{\frac{n_b}{t}} \tag{7}$$

where $\sqrt{n_b / t}$ is the standard deviation of the blank count rate, for a stable counting system, with purely Poisson variations.

When F varies significantly between measurements as, for example, when there are variations in chemical yield, the HPS Standard N13.30 [40] recommends using $F_{0.05}$, the lower bound value of F that represents a 0.95 probability that the value of F is higher than $F_{0.05}$,

$$F_{0.05} = \bar{F} - 1.65U \tag{8}$$

where $\bar{F}$ is the average value of F obtained from a series of calibrations and U is the overall uncertainty estimate, given at the 68% confidence level as

$$U = \left(\sum_i \sigma_i^2 + \frac{1}{3} \sum_i \delta_i^2 \right)^{1/2} \tag{9}$$

where δ_i are the estimated systematic uncertainties and σ_i are the random uncertainties at the 68% confidence level.

The calculated MDA becomes

$$\mathrm{MDA} = \frac{4.66 s_b}{F_{0.05}} + \frac{3}{F_{0.05} t} \tag{10}$$

This will ensure that the probability of a type II error would be less than 0.05, even when there are variations in calibration results affecting an entire batch of samples.

When the radioactive half-life of a nuclide is very short and there is a significant decay during the time of measurement, t_s should be replaced by

$$t_s = \exp(-\lambda t)[1 - \exp(-\lambda \Delta t)]/\,\lambda \tag{11}$$

where λ is the physical decay constant, t is the time interval between the time of interest for calculating the quantity of analyte present and the initiation of the measurement process, and Δt is the duration of the measurement.

6.3. ERRORS AND UNCERTAINTIES[1]

The result of a measurement is an estimate of the value of a particular quantity, and should be accompanied by the uncertainty associated with that estimate. The uncertainty in the result of a measurement characterizes the dispersion of values that could reasonably be attributed to the measured quantity. All components of the uncertainty, of random or systematic origin, contribute to the dispersion.

In general, a measurement has imperfections that give rise to an error in the measurement result. The error of measurement is by definition the result of a measurement minus the true value of the measured quantity. Since a true value cannot be determined in practice, error is an idealized concept. The difference between error and uncertainty is exemplified by the case of a measurement with a large uncertainty, but with a value very close to the true value. Here the error is small but the uncertainty is large.

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[1] This section is based on Ref. [41].

Frequently the quantity of interest Y is not measured directly, but is derived from other measured quantities, $X_1, X_2, X_3, \ldots, X_n$,

$$Y = f(X_1, X_2, X_3, \ldots, X_n)$$

The estimated standard deviation associated with the quantity Y is determined from the estimated standard deviation of each variable X_i. The standard uncertainty $u(y)$ of y, the estimate of Y, is obtained by appropriately combining the standard uncertainties $u(x_i)$ of x_i, the estimates of the variables X_i,

$$u^2(y) = \sum_{i=1}^{n} \left(\frac{\partial f}{\partial x_i} \right)^2 u^2(x_i) \tag{12}$$

This expression is known as the law of propagation of uncertainty. When the uncertainties of the variables are considered equal to the standard deviations of the probability distributions of the x_i and the uncertainty of y is taken to be equal to the standard deviation of the probability distribution of y, the equation of the propagation of uncertainty may be written as

$$\sigma^2(y) = \sum_{i=1}^{n} \left(\frac{\partial f}{\partial x_i} \right)^2 \sigma^2(x_i) \tag{13}$$

If some of the variables x_i are significantly correlated, the appropriate expression is

$$\sigma^2(y) = \sum_{i=1}^{n} \left(\frac{\partial f}{\partial x_i} \right)^2 \sigma^2(x_i) + 2 \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \left(\frac{\partial f}{\partial x_i} \right) \left(\frac{\partial f}{\partial x_j} \right) \sigma_i \sigma_j r_{ij} \tag{14}$$

where r_{ij} is the correlation coefficient of x_i and x_j.

A comprehensive list of uncertainties in the measurement of the concentration of a radionuclide in a sample is given in HPS Standard N13.30-1996 [40].

As an example consider (as in Section 6.1) the measurement of activity A, where

$$A = \frac{1}{F} \left(\frac{N_s}{t_s} - \frac{N_b}{t_b} \right) = \frac{1}{F} (n_s - n_b) \tag{15}$$

and

$$F = \frac{1}{A_t}\left(\frac{N_t}{t_s} - \frac{N'_b}{t'_b}\right) = \frac{1}{A_t}(n_t - n'_b) \tag{16}$$

then

$$A = \left(\frac{n_s - n_b}{n_t - n'_b}\right)A_t \tag{17}$$

In this example, assume that the errors in A_t and t are negligible in comparison with those due to random variation in the counts.
From (12),

$$\sigma_A^2 = \left(\frac{\partial A}{\partial n_s}\right)^2 \sigma_{n_s}^2 + \left(\frac{\partial A}{\partial n_t}\right)^2 \sigma_{n_t}^2 + \left(\frac{\partial A}{\partial n_b}\right)^2 \sigma_{n_b}^2 + \left(\frac{\partial A}{\partial n'_b}\right)^2 \sigma_{n'_b}^2 \tag{18}$$

then

$$\sigma_A^2 = \left(\frac{A_t}{(n_t - n'_b)}\right)^2 \left(\frac{n_s}{t_s} - \frac{n_b}{t_b} + \frac{(n_s - n_b)^2}{(n_t - n'_b)^2}\frac{n_t}{t_s} + \frac{(n_s - n_b)^2}{(n_t - n'_b)^2}\frac{n'_b}{t'_b}\right) \tag{19}$$

7. QUALITY ASSURANCE

7.1. QUALITY ASSURANCE PROGRAMME

An essential and integral element of any bioassay monitoring programme is an effective quality assurance (QA) programme to provide continuing evidence that exposures are being assessed correctly, to enable management and regulatory agencies to verify the adequacy of workplace controls and to provide workers with trustworthy results. Maintenance of quality assurance programmes should be a primary concern of all those managing and working in the bioassay laboratory. The design and implementation of such a programme is described in the Safety Guide on Assessment of Occupational Exposure due to Intakes of Radionuclides [3] and in the Safety Series No. 50-C/SG-Q: Quality Assurance for Safety in Nuclear Power Plants and other Nuclear Installations [42].

7.2. QUALITY ASSURANCE TECHNIQUES

The quality assurance programme includes the procedures describing the appropriate techniques that must be applied to the processing and analyses of samples. These procedures must demonstrate that sufficient safeguards are taken at each step to assure the validity of the results.

Responsibility for the quality of a particular operation should be delegated to the person actually performing the operation. Such persons should be actively involved in the development of QA procedures, and trained in methods of detecting non-compliance. Management should motivate staff to detect, report and correct non-conformances. Quality assurance built into a programme from the bottom up is more effective that QA imposed from the top down. For the QA programme to be effective, all personnel must be confident that management expects and encourages performance that meets its objectives.

The QA programme must be thoroughly documented. A QA plan should be prepared that contains general instructions on implementing the programme and the various steps in its operation. Written procedures should describe every task, and contain QA requirements. For example, an analytical radiochemistry procedure should contain acceptable limits for chemical yield. Quality assurance procedures should document the use of control charts and other methods for tracking instrument historical backgrounds, efficiencies and other performance measures, and should contain instructions for reporting and correcting deviations, as well as for taking account of changes in the operation. Procedures for documenting and reporting results should also be prepared, as should procedures for record preparation, maintenance and archiving. The documentation should provide sufficient information for an auditor to trace the operation from start to finish and assess its validity. Quality assurance training should be incorporated into the training, certification and recertification programmes for all personnel. Such training should include:

(a) Their particular responsibility within the quality system,
(b) The basic philosophy and strategy of internal dose assessment,
(c) The principles and details of the methods used,
(d) The technical details and potential problems of the processes in which they are involved,

(e) The relation their work has with other parts of the programme,
(f) Recognition and reporting of problems that arise,
(g) Knowledge of the overall quality system and its objectives.

Good quality results are difficult to achieve in substandard environments. Adequate laboratory and office space should be available to accommodate both required equipment and personnel. Equipment should be reliable and stable, and appropriate to the task for which it is intended. A preventative maintenance programme should be instituted to ensure that equipment will not fail at a critical time, such as in an emergency. Ventilation, fume hoods and bench space are required for radiochemical operations. Shielded facilities should be provided for detectors, including those in direct assessment facilities. Access control to all facilities is required, both to protect sensitive equipment and to maintain appropriate confidentiality for records.

The environmental control should be adequate to ensure that no equipment is subject to conditions likely to affect its performance. Factors that should be controlled include: temperature, relative humidity, light levels, dust and reactive chemical vapours. A stable power supply is required so that the voltage and current remain within the required specifications of the equipment in use. Stray electric and magnetic fields should be minimized to avoid affecting equipment.

In many countries there are legal requirements that must be met for the laboratory to be accredited to perform measurements that may be used as the basis for formal dose assessment. In some countries the authorities that oversee such accreditation assess laboratories for accreditation in other countries that do not have such a service, often for a fee. In addition to accreditation as an approved dosimetry service, it is advised that laboratories attempt to gain accreditation to the relevant standard of the ISO [43]. Again, several countries have a body that supervises the accreditation of laboratories to such a standard (e.g. the United Kingdom Accreditation Service (UKAS)[2] in the UK, the National Institute of Standards and Technology (NIST) in the USA and the Office de Protection contre les Rayonnements Ionisants (OPRI) in France), which may be able to provide a service to laboratories in other countries.

Quality assurance techniques include:

(a) Unique labelling which identifies the sample and the person concerned throughout the process from sampling to the recording of the measurement results.

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[2] Formerly the National Accreditation of Measurement and Sampling (NAMAS), which was an activity under UKAS.

(b) Chain of custody of all samples to preclude loss, contamination, tampering or incorrect analysis. Empty and full sample containers should be sealed and transfer of containers should be documented at each stage of the operation.

(c) Quality requirements for the acquisition of all chemicals, equipment and other materials used in monitoring.

(d) Regularly scheduled calibration of all chemical procedures and detection systems, and frequent background monitoring. Calibration and background measurements should typically represent 10% or more of all measurements performed by each method of analysis.

7.3. PERFORMANCE ASSESSMENT

As part of its QA programme, a measurement facility may establish performance criteria to be applied to the analysis of spiked samples, i.e. samples for which the radionuclide content is known beforehand. Performance criteria should be well defined limits on the acceptability of the results of the measurement, as functions of the radionuclide content in the sample compared with the MDA of the method. The US Health Physics Society has, for example, published performance criteria for radiobioassay [40]. Acceptable results for the analysis of $^{239/240}$Pu in faeces may be a relative bias within −25 to +50% of the true value, when the true value is at least five times the MDA of the method. Similarly, acceptable results for the direct measurement of ^{241}Am in the lungs of a phantom could have a relative bias within −25 to +50% of the true value, when the true value is ten times the MDA of the method. Performance criteria for the precision of replicate measurements should also be set, for example, at a variation of not more than 30% in successive measurements of the same sample, if the true content is five times the MDA of the method. If measurements are of low activity so that random statistical errors are dominant, performance criteria cannot be more restrictive than statistical fluctuations permit. Finally, analyses of samples intended for performance assessment should be performed in at least a single blind fashion. That is, the analyst should not know the true value beforehand, although the assessment sample may be identified as such. In a double blind assessment, the analyst is not informed that the sample is anything other than routine. Although a double blind assessment may give a truer picture of the laboratory's capabilities, such assessments can be difficult to perform.

Laboratories performing direct or indirect measurements for internal dosimetry should participate in intercomparison exercises. National intercomparison programmes for direct measurements are offered in many countries, and international intercomparisons are also being co-ordinated (e.g. Refs [44–46]). Likewise, for indirect measurement both national and international intercomparisons have been co-ordinated, for example, by the French Atomic Energy Commission (CEA) [47]. In addition, periodic audits or reviews should be performed to verify compliance with the QA programme and the effectiveness of the internal dosimetry programme. Guidance for the conduct of audits and reviews is given in the Safety Guide on Occupational Radiation Protection [1].

8. RECORDS AND REPORTS

8.1. LABORATORY RECORD KEEPING

The purpose of record keeping, the nature and scope of the records, and the extent of record keeping systems depend on national requirements. Because the records should include the results of individual monitoring for internal contamination, results of bioassay or PAS sample analyses should be recorded and maintained. The minimum information maintained in the records should include:

(a) Unambiguous identification of the sample, including type of sample and worker or location from which it was obtained.
(b) Documentation of the custody of the sample from collection to analysis.
(c) Dates and times of sample collection and analysis, sample mass and/or volume.
(d) Analytical procedure(s) employed and the corresponding MDA(s).
(e) Identification of the counter or analytical instrument used and calibration information.
(f) Raw measurement data including gross and background counts in regions of interest or complete spectra, chemical yield and calculation of final result.
(g) Measured activity for each radionuclide analysed, together with its estimated uncertainty. The uncertainty should specify whether it is due to counting statistics only or to a total propagated error, and the number of standard deviations (or errors) it represents. All measurement results should be recorded, even if less than MDA or negative.
(h) Any comments on the analysis that may be helpful in the interpretation of the results, such as difficulties encountered in the procedure or unusual properties or circumstances of the sample (e.g., whether or not the sample was collected for special monitoring, if an unusual interference was noted or if a more sensitive analytical method was requested).
(i) Whether or not the result exceeds a specified derived recording or investigation level.

(j) Identity of the analyst(s).
(k) Copies of reports issued.

Supporting documentation that is common to a number of samples and/or analyses should also be maintained, including:

(i) Working procedures and practices,
(ii) Training records and qualifications of analysts,
(iii) Quality assurance procedures,
(iv) Quality control data such as background trends and estimates of MDA,
(v) Equipment calibration procedures and records,
(vi) Traceability of standard sources.

Records should be maintained in accordance with national standards.

8.2. REPORTING OF RESULTS

The procedures and levels to be used for reporting individual monitoring results should be clearly specified by the management or regulatory authorities. Information reported should be clearly identifiable and understandable. Normally only final results are reported, and results which fall below the MDA or derived recording level may be reported as such. (However, the actual result, along with its uncertainty, should be retained in the records.) The analytical report should contain at a minimum the following information:

(a) Identification of worker,
(b) Type of sample, and its time of collection and analysis,
(c) Final result and its uncertainty,
(d) MDA of the analytical method,
(e) Whether the result exceeds a derived recording or investigation level,
(f) Identification of the individual providing the report.

Because of privacy considerations, reports should be safeguarded and the worker's identity protected, as for medical records.

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NOTE ON THE ANNEXES

Nine technical Annexes follow which describe in detail certain procedures in use in laboratories to measure selected radionuclides in biological and physical samples. The selection is intended to provide information that will, on its own, allow the analysis of a particular sample, but which may also be adapted to the analysis of other comparable samples or radionuclides. Together, these Annexes cover all of the common methods currently used in indirect monitoring.

The Annexes provided are as follows:

(i) Measurement of γ emitting radionuclides in biological samples;
(ii) Analysis for β emitting radionuclides in urine by liquid scintillation counting;
(iii) Analysis for strontium isotopes in urine;
(iv) Analysis for ^{226}Ra, ^{228}Ra and ^{210}Pb in biological samples;
(v) Analysis for uranium in urine by fluorimetry;
(vi) Analysis for thorium in samples;
(vii) Analysis for plutonium, americium and curium in urine and faecal samples by α spectrometry;
(viii) Analysis for tritium and ^{14}C by combustion techniques;
(ix) Determination of creatinine in urine samples.

The Annexes describe, as necessary, methods of sample preparation, radionuclide separation and activity counting or non-radiometric methods of radionuclide quantification. They also include requirements for quality assurance and cautions about safety. They do not, however, give guidance on dose interpretation, which will be provided in the Safety Report, Assessment of Radiation Doses from Intakes of Radionuclides by Workers, which is under preparation.

Annex I

MEASUREMENT OF GAMMA EMITTING RADIONUCLIDES IN BIOLOGICAL SAMPLES

Based on information from the National Radiological Protection Board, UK

I–1. INTRODUCTION

This Annex describes the use of γ ray spectrometry for quantitative assessments of intakes by measuring the photon emitting radionuclides in biological samples with the use of the minimum of chemical preparation and for which self-absorption of the photon signal in the sample is also insignificant. It will therefore concentrate on the measurement of low density uniform samples (e.g. in urine) containing radionuclides that emit photons with energies >60 keV at a reasonable emission probability (>1–100% per decay, depending on sample activity and the measurement system). This technique is applicable to the assessment of a wide range of photon emitting radionuclides (see Table IV in the main text).

The increase of attenuation as photon energy decreases will cause the MDAs to be higher for lower energy photons. Chemical separation of the radionuclide from the sample may be necessary in such cases to obtain a well calibrated γ ray measurement (by decreasing absorption and bringing the sample nearer the detector and so reducing the MDA). In such cases, however, one of the principal advantages of photon spectrometry has been lost and other methods of sample measurement may be just as simple to perform and more appropriate. However, for some such samples (e.g., those containing volatile radionuclides, such as ^{125}I, that might be lost through chemical processing), a bulk γ ray spectrometry measurement may still be the most appropriate method of measurement.

It should be noted that γ ray spectroscopy will not detect pure α ray emitting radionuclides and is not suitable for monitoring pure β emitters, even though the detector may be capable of detecting the bremsstrahlung radiation. The appropriate technique for α or β emitters should be used.

I–2. DETECTORS FOR γ RAY AND X RAY SPECTROMETRY

Intrinsic germanium (Ge) semiconductor detectors or thallium-activated sodium iodide (NaI(Tl)) scintillation detectors are the most commonly used detectors for γ ray spectrometry.

The advantages of NaI(Tl) detectors are that they are robust, needing little maintenance and auxiliary equipment, and are available in any size, cost being the limiting consideration. The use of large volume NaI(Tl) detectors gives such systems high photon detection efficiencies. The disadvantage of these detectors is that they have relatively poor energy resolution (~80 keV full width at half-maximum (FWHM) (Section I–4) at 1332 keV for a 150 mm dia. × 100 mm deep detector). They are most useful for bioassay assessment when used to monitor samples for known mixtures of a few (or preferably one) radionuclides which emit a simple γ ray spectrum at energies above 150 keV. Scintillation detectors are not preferred for monitoring more complex mixtures of radiations or mixtures of unknown radionuclides, as their poor energy resolution requires γ ray peaks to differ by several tens of keV before they can be satisfactorily resolved. Spectral deconvolution techniques can be used to unfold and analyse complex spectra but their use implies prior knowledge of what is and is not present in the sample. Erroneous results can be produced by such methods when the information on the radionuclides present, or the background count rate, is not accurate.

It should also be noted that the energy calibration of an NaI(Tl) detector tends to drift with time and is affected by changes in temperature and humidity, even with the use of air conditioning systems. Frequent, possibly daily, energy calibration of such detectors is therefore recommended.

The advantage of semiconductor germanium detectors for γ ray spectrometry is their superior energy resolution (~2 keV at 1332 keV), which enables greatly enhanced radionuclide identification capability. Semiconductor detectors are therefore suitable for assessing samples that contain mixtures of unidentified radionuclides or complex mixtures of radionuclides. While the photon detection efficiency of semiconductor detectors is currently limited by the size to which the crystals can be grown, better overall sensitivity and MDA can be achieved with a semiconductor detector system than with scintillation detectors. This is again an outcome of the semiconductor detector's superior energy resolution, due to the reduction in the area under the peak that needs to be defined as background to the peak.

The operational disadvantage of semiconductor detectors is that they have to be cooled to liquid nitrogen temperature when in use (i.e. a high tension (HT) voltage is applied). Intrinsic semiconductor detectors can be warmed to room temperature when an HT voltage is not applied. However, keeping the detectors cold as much of the time as possible and keeping warming/cooling cycles to a minimum usually results in better long term performance of intrinsic semiconductor detectors and reduces maintenance costs.

As well as intrinsic germanium detectors (Ge), silicon (SiLi) detectors are suitable for use in γ ray and X ray spectrometry. Their disadvantage is that they will be ruined if ever permitted to warm to room temperature. It is therefore absolutely essential to ensure that cooling is maintained at all times. Should such detectors need to be moved at any time, for example, to be sent for repair or to facilitate laboratory maintenance, the hazard from liquid nitrogen spillage and from the weight of the filled dewar need to be carefully considered.

Electrical cooling systems are now becoming available, but the size, cost, possible noise and need for an uninterrupted electricity supply need to be carefully considered in comparison with liquid nitrogen cooling. Semiconductor detectors therefore require either reliable regular supply of liquid nitrogen or a reliable electricity supply. The vibrations caused when filling the liquid nitrogen dewar can interfere with the pre-amplifier unit and hence the detector signals. Making measurements while the detector is being filled should therefore be avoided.

The use of specialized detectors is required for the spectroscopy of low energy (5–60 keV) X rays and γ rays and should be considered for the detection of photons with energies below 200 keV. Depending on exact monitoring needs, large volume n type intrinsic germanium, thin crystal n type intrinsic germanium or lithium drifted silicon detectors, all with thin beryllium windows to reduce signal attenuation, are appropriate for measuring low energy photons. If one detector is required to monitor photons from a few tens of keV to several hundred keV, a large n type detector should be considered, bearing in mind that such detectors are a compromise and better quality results may be obtained from two measurements with a conventional high energy photon (>100 keV) detector and a thin crystal, low energy detector. The use of a thin crystal reduces the signal from Compton scattering, thus reducing the background continuum and hence MDA. Dual phosphor (Phoswich) scintillation detectors have also been used for low energy photon spectroscopy, but are not commonly used for this now, due to the superior performance of semiconductor detectors.

Under reasonable operating conditions, with the high energy bias continuously applied, the energy and energy resolution calibrations of semiconductor detectors are usually stable. However, it is advisable to check and document instrument calibrations approximately once a week so that any drift that does occur is detected and rectified. Such measurements and their recording should be part of any laboratory accreditation scheme.

Cooling is conventionally done for 'small' sample measuring systems (as opposed to body monitoring) using a dewar of liquid nitrogen, typically of the order of 30 L capacity, which requires filling approximately once a week.

I–3. SHIELDING

The background radiation count rate measured by the detector(s) needs to be as low as achievable so as to maximize detection sensitivity. The detection system should be housed in a low background room in which no open radioactive sources are handled and no chemical preparation of samples takes place. Simple precautions such as the use of a low barrier, the removal of laboratory coats and the wearing of overshoes are appropriate to guard against radioactive contamination being brought into the measurement laboratory. In addition, the detectors should each have additional shielding equivalent of up to 10 cm or more of lead. Detectors used for measuring low energy photons (<100 keV) should have lead shields lined with a graduated shield of tin or cadmium, copper and steel to attenuate the characteristic X rays from lead, tin and cadmium respectively, so as to attain a low background count rate in the low energy region. The contribution from radon decay products should be minimized by filtering the conditioned air entering the laboratory through a carbon filter. These precautions are even more necessary for photon measurement systems than for α particle and β particle measurement systems because the penetrating nature of the radiation can cause measurements to be affected by a radiation source in another part of the room. Even with all these precautions, variations in background counting and detector efficiency will occur and necessitate frequent calibration of the counting system.

I–4. CALIBRATION

Quantitative γ ray spectrometry analysis requires a high quality calibration regime. The three parameters that require calibration for each detector are the energy response (energy per MCA channel), energy resolution (FWHM, which is the full width of the peak in keV at half the maximum peak height) and detection efficiency as a function of energy. Detection efficiency is a function not only of the detector but also of the sample being measured because of the geometrical effects from its size and shape, and self-absorption by the sample, all of which will vary with its size and density and elemental composition. Accurate calibration therefore requires that samples being quantitatively assessed must be of a standard calibrated geometry, density and composition. Normally a laboratory will calibrate its system for a range of standard counting geometries appropriate to the range of samples that is being measured. A standard geometry is the combination of the vessel holding the sample (e.g. a 250 mL Marinelli beaker) and the matrix being measured (e.g. urine) together with a set spatial relationship between the detector and the sample. Each detector should be calibrated for each standard geometry that is measured on it. Rather than preparing calibration standards of different matrices, laboratories may choose water as the matrix for all standards. Correction factors may then be needed for samples which differ greatly in density from water.

Calibration standards should be traceable to national or international standards (NIST (USA), NPL (UK), PTB (Germany), etc.). Complex calibration matrices with densities varying from 0.2 to 1.6 $g{\cdot}cm^{-3}$ and containing radionuclides to allow energy calibration from ~60 keV to 2 MeV are now being produced which will assist calibration, but such standards are expensive. The photon spectra from such standards contain many low energy photon emissions, as it is at low energies that detection efficiency changes most rapidly as a function of energy and sample composition. As these standards contain short lived radionuclides, they require frequent renewal. For NaI detectors only single radionuclides should be used as standards.

When preparing a sample in a standard counting geometry the following factors should be considered, as they can cause inaccuracies in calibration:

- Solid samples need to be packed to a standard density and volume.
- The volume of liquid samples needs to be accurate for the standard geometry.
- Liquid samples need to be sealed to prevent both spillage and evaporation of the sample.
- Care should be taken with any sample containing either a chemically volatile radionuclide (e.g. iodine) or a gaseous radionuclide (radon) that may evaporate from the sample and steps taken to prevent this happening or to calibrate for this.
- Settling and separation of samples into solid and liquid phases or into separate immiscible liquid phases containing different radionuclide fractions will affect detection efficacy and hence may bias results.

If a sample requires qualitative assessment that cannot be measured in a standard counting geometry, this should be recorded and calibration carried out using the most appropriate calibrated counting standards, preferably using two standards which have detection efficiencies that would bracket the probable detection efficiency of the non-standard geometry. To prevent any cross-contamination between samples it is good practice to place each sample entering the low background measurement laboratory in a new plastic bag. Each sample should be clearly labelled with a unique sample code and each spectrum measured should be saved to a uniquely named data file, with a clear record kept of each measurement of each sample.

I–5. URINE MEASUREMENTS

Ideally a urine sample should be a full 24 h sample, with the time period of the sample carefully recorded. A 2.5 L plastic container is usually adequate for collecting such samples. All urine samples should contain some ^{40}K, so that at least one γ emitting radionuclide should be detected in all urine samples by most systems.

For most isotopes the sample can be counted as being untreated; this has the advantage of requiring no estimate of chemical recovery of the element of interest. The addition of nitric acid does not necessarily prevent the formation of precipitates in storage. If a precipitate is evident in the sample it should be heated with nitric acid until a clear solution is formed, unless the radionuclides of interest could be lost by volatilization. The sample can then be transferred into the standard counting geometry container.

If a lower MDA is required, this can be obtained by simply concentrating the sample. This can be achieved simply by evaporation of the acidified sample, noting again the warnings about the loss of volatile radionuclides. For specific radionuclides, concentration can also be achieved by co-precipitation or absorption on a precipitate, for example caesium can be absorbed on to ammonium molybdate from acid solution. If such a method is used then some estimate of recovery will be necessary, either by an internal or an external standard (e.g. adding a known activity spike to the sample of another radionuclide of the same element that will not be naturally in the sample and correcting the results according to the fraction of the spike's activity that is actually measured). The concentrate or filtered precipitate can then be counted at higher efficiency than the untreated sample.

I–6. FAECAL MEASUREMENTS

The usual faecal sample is a single voiding. All excreta samples can carry hazardous organisms and the unsterilized samples must be handled with care, preferably following guidelines laid down for the handling of biohazardous materials by the relevant national health and safety authority. Preferably staff handling such samples should be inoculated against hepatitis A and B and polio. Full protection may not be obtained until a year after the commencement of an inoculation programme. These problems are avoided when samples are ashed, as for non-volatile radioactive materials.

For measurement it would be advisable to transfer the sample to a standard geometry container, making sure the sample is evenly distributed and well mixed. Seal the container and quickly refreeze it. The frozen sample can be counted, having first sealed it in a new plastic bag to prevent any possibility of radiological or biological contamination. Counting a frozen sample reduces the biohazard and also reduces the chance of the sample separating and becoming non-homogenous in the counting period. Counting frozen samples can create condensation on the outside of the detector and, for some germanium detectors, but not all, this can cause significant drift in the energy calibration which may persist for about a day before the system dries out. Frequently, faecal material is dried before measurement, provided that the radionuclides to be determined are not volatile. This makes it easier to pack and produce a well mixed sample. Efficiency calibration standards, with the same density as dried faecal material, can be purchased for preparing standards.

I–7. OTHER SAMPLES

Nose blow samples or other measurements may be used as an indicator to determine if other forms of bioassay measurements of an individual are required. The measurement itself is not normally used as part of the dose assessment. The accuracy of the quantitative radionuclide assessment therefore does not need to be as good as for those bioassay measurements which are directly used to assess dose. However, it would be advisable for laboratories to have a standard procedure for taking and measuring such samples. The procedure should include precautions against adventitious loss or contamination of the sample, the use of a standard counting geometry and a procedure by which each sample is uniquely identified. The standard counting geometry used should minimize the effects of non-uniform distribution of activity.

Annex II

ANALYSIS FOR BETA EMITTING RADIONUCLIDES IN URINE BY LIQUID SCINTILLATION COUNTING

Based on information from Ontario Hydro Nuclear, Canada

II–1. INTRODUCTION

Intakes of β emitting radionuclides that are excreted in urine can often be assessed most readily by direct liquid scintillation counting (LSC) of a sample of the urine. With this technique an aliquot of the native urine sample is dispersed in an organic liquid scintillator, and the light photons emitted as a result of the absorption of energy from the β decays are detected by photomultiplier tubes operated in coincidence to reduce background. Where large numbers of samples must be processed, for example in the routine monitoring of workers at heavy water reactors, or where the contaminant radionuclides such as ^{3}H, ^{14}C, ^{32}P or ^{241}Pu emit few penetrating radiations, or where direct methods are unavailable, LSC provides the most practical method of individual monitoring for activity excreted in urine.

However, although the intensities of the scintillant light pulses are proportional to the energies of the individual β decays, only limited identifications of the components of a source mixture are possible because of the continuous distribution of the β energy spectrum, and then only if the concentrations of the higher energy sources are much less than those of the lower energy co-contaminants. Nevertheless, with the use of energy discrimination methods on the counter and the application of careful calibrations, for many situations LSC can provide a useful screening function as part of a dose assessment programme for intakes of mixed β–γ emitters. This screening can provide important additional information, for example on the time of intake for radioiodine (see Section 7 in Ref. [II–1]), or on the need for additional measurements. Although the identification of other β emitters in the urine may contribute additional sources of dose to the assessments, corrections for them can also be essential for the accurate measurement of a primary, lower energy source, such as ^{3}H.

Liquid scintillation counting of urine can also be adapted to the assessment of intakes of some α emitting radionuclides.

The following procedure was developed for routine monitoring at a heavy water power utility, where workers are exposed chronically to low levels of tritiated water (HTO) (for which dose assessments are required), less frequently to soluble forms of ^{14}C, and occasionally to other mixed fission and activation products. The procedure was developed as a basis for the dose assessments for HTO, and as a form of screening complementary to workplace monitoring for identifying other potential intakes. The primary individual monitoring for intakes of high energy mixed fission and activation products is a programme of routine self-monitoring using a whole body counter.

Apparatus

Fume hood
Calibrated liquid dispenser
LSC vials
LSC counter with multichannel analysis capability.

Reagents

LSC cocktail.

Calibration sources

^{3}H, ^{14}C and (e.g.) ^{137}Cs
Picric acid for quench calibration.

II–2. DETERMINATION OF THE PROTOCOL

(a) From a knowledge of workplace sources, determine which β emitting radionuclides may be present in worker urine samples.

(b) From their radiological characteristics (Table IV) determine the most useful β energy discrimination regions ('windows'), to a maximum of about 3. In the example, programme windows were set corresponding to β energies 0–18.6 keV (region A: ^{3}H), 35–110 keV (region B: ^{14}C) and 180–2000 keV (region C: other). These choices were based on optimizing the detection efficiency for counting real urine samples and ensuring that even double events resulting from decays of the much larger activities of ^{3}H would not interfere with higher energy regions. Since the detected distributions are shifted to lower energies in highly quenched samples, when counting efficiencies are low there will be significant uncertainty in the interpretation of the upper region measurements.

(c) From the dosimetric and biokinetic models, and the requirements of the monitoring programme for the primary intake (HTO), determine the sensitivity requirement for detection of activity in the sample. Together with system characteristics, this determines the sample volume and count time, accepting that increasing sample volume increases quenching.

(d) In the example case, a value for the MDA of HTO in region A of 2 kBq/L was determined, based on the variation in a group of 'uncontaminated' urine samples. For a sample volume of 1 mL, the required count time is 0.4 min. An additional count is required for the quench correction. This allows the required ~800 samples per day to be processed on three counters.

(e) Ensure that the available time and equipment can meet the proposed protocol, and adjust as necessary.

II–3. SYSTEM CALIBRATIONS

(a) Calibrate the LSC for energy using a suitable unquenched standard. This calibration should be checked periodically according to the LSC operating manual.

(b) Counting efficiency is influenced by chemical and colour quenching from materials in the sample, and can vary by a factor of two or more. Because these are not constant across the different samples, a single correction is not sufficient and the assumed efficiency must be based on the characteristics of the particular sample. The most accurate method is the addition of a high activity internal standard source to each sample following the count, but this method is time consuming. Most LSCs include an automatic quench correction mechanism such as external standardization, whereby during a second count an adjacent external γ source is used to generate known numbers of electrons in the particular sample mixture, and the ratio between the known and measured numbers of counts is used to correct the sample measurement. Calibrations are usually provided with the system, but should be validated using real urine samples, or close surrogates. Picric acid has been found to simulate the chemical and colour quenching effects of urine components appropriately.

(c) Calibrate the energy regions using appropriate traceable standards under various quench conditions. In the present case region C is calibrated with ^{137}Cs, which is the most frequent radionuclide detected, but it is recognized that counts in this window are useful only as an indication of a source (other than ^{3}H and ^{14}C) in the urine sample significantly above the count rate background.

(d) Check volumetric dispensers periodically, depending on the stability observed. Corrections should be made when a discrepancy of greater than about 0.1% is found.

II–4. SAMPLE PREPARATION

(a) Samples should be dispensed in a fume hood or other source of protection from the biological, chemical and radiological hazards. Samples must be thoroughly mixed before counting. In the present case 1.0 mL aliquots of urine are drawn into an automatic dispenser and dispensed into the LSC vial, followed by the LSC cocktail to a total volume of 15 mL. With such a system, periodic checks must be made to ensure an acceptable level of carry-over of radioactive material between samples.

(b) For high volume applications, the dispensing system can be built into a robotic assembly which samples from the urines and dispenses them automatically into vials in the LSC counting tray. With such systems, additional quality checks must be introduced to ensure reliable sample identification.

(c) Most LSCs provide a temperature controlled and light sealed counting chamber to minimize chemiluminescence and ensure a stable count rate. Samples need to be introduced into the chamber for a short period before counting for stabilization, usually 20 min or so.

II–5. SAMPLE COUNTING

(a) In the example programme, samples are counted, initially, for 0.4 min and the LSC reports disintegration rates, suitably corrected for quenching (using an external standard) and background, for the three energy regions.

(b) These rates are then converted to activities of ^{3}H (region A), ^{14}C (region B) and others (region C), based on calibrations with the standard sources.

(c) Any activity in regions B or C above the MSA triggers a re-count of the same dispensed sample for 10 min, to improve the MDA.

II–6. INTERPRETATION OF COUNTING RESULTS

(a) Activities measured in region A should be corrected for the low energy contributions from the spectra of sources identified in regions B and C. Such a correction would require information about the shape of the spectrum of the higher energy source over region A, for example by identifying the radionuclide. Usually the activity of ^{3}H, however, is much greater than any activity in regions B or C, by several orders of magnitude, so that no correction is required. Failure to make a correction for other sources of counts in region A will result in an overestimate of the concentration of HTO. The activity of HTO is then applied directly in dose assignment.

(b) The MDAs for screening in regions B and C were determined to be 0.8 kBq of ^{14}C and 1 kBq of ^{137}Cs, respectively, for the 0.4 min count, and 0.15 kBq and 0.25 kBq, respectively, for the 10 min count. These values were again determined from the variation in a group of background urine samples that contained only normal levels of naturally occurring radionuclides, such as ^{40}K.

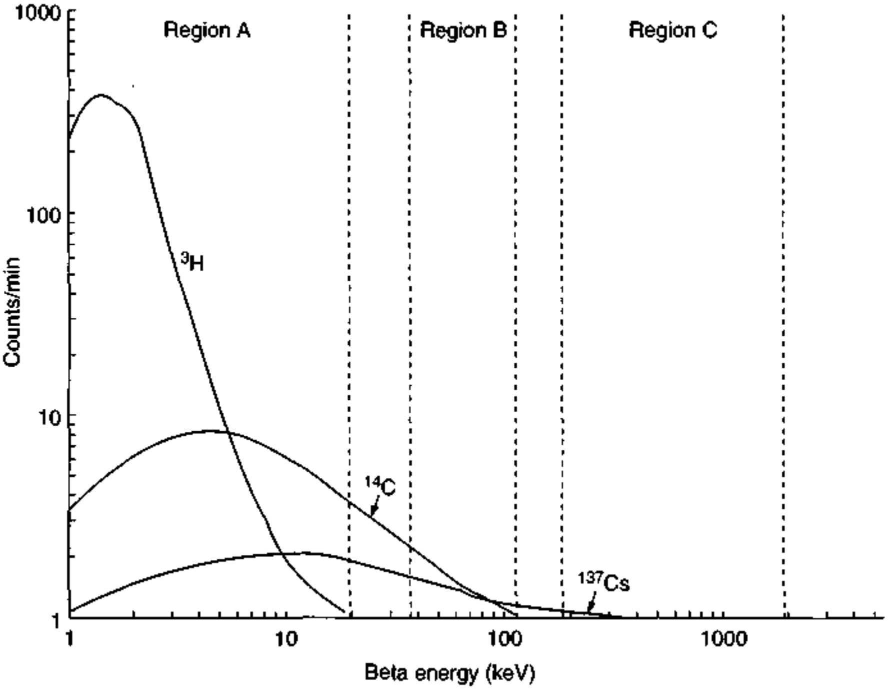

FIG. II–1. Example of the use of energy discrimination techniques in liquid scintillation counting of mixed β emitters in a urine sample.

Similar values for region C would apply to many other radionuclides, such as ^{131}I.

(c) When bioassay samples are provided frequently, as they are in a routine monitoring programme for HTO (usually at least biweekly), these levels are adequate as a basis for useful screening, although they would not in many cases be sufficient for dosimetry.

Figure II–1 illustrates the use of energy discrimation techniques in liquid scintillation counting for a urine sample containing ^{3}H, ^{14}C and ^{137}Cs.

REFERENCE TO ANNEX II

[II–1] INTERNATIONAL ATOMIC ENERGY AGENCY, Assessment of Occupational Exposure due to Intakes of Radionuclides, Safety Standards Series No. RS-G-1.2, IAEA, Vienna (1999).

Annex III

ANALYSIS FOR STRONTIUM ISOTOPES IN URINE

Based on information from Argonne National Laboratory, USA

INTRODUCTION

The material to be analysed is dissolved in nitric acid. After addition of the strontium carrier, strontium phosphates are precipitated from the urine. The strontium is separated from calcium by either the crown ether or the fuming nitric acid method. If the ^{89,90}Sr content is to be determined by liquid scintillation counting, the purified strontium solution can be used directly with the appropriate cocktail for liquid scintillation counting.

If the ^{90}Sr content is to be determined with a proportional counter based on the ^{90}Y count, the yttrium carrier is added to the purified strontium solution and, after a delay of at least seven days (to allow for ^{90}Y ingrowth), the yttrium is separated, mounted and counted. The storage period must be sufficient for significant ingrowth, and the appropriate growth factor applied. For the determination of ^{89}Sr, strontium carbonates are precipitated immediately after the yttrium precipitation, mounted and counted. ^{89}Sr is calculated from the difference between the total activity of the strontium carbonate and the activity calculated for ^{90}Sr.

If any ^{85}Sr is present it is best measured by γ spectrometry.

Apparatus

Fume hood
Hotplate or IR lamp
Glassware, beakers, centrifuge tubes
Buchner, fibreglass filters
Beta counter (liquid scintillation, gas flow, Geiger–Müller, or proportional counter)
Standard laboratory safety equipment (safety glasses, rubber gloves, protective clothing, etc.).

Reagents

Fuming nitric acid; 95–96% by weight
Nitric acid (8M)
Hydrophosphoric acid (H_3PO_4), concentrated
Oxalic acid solution, saturated

Ammonium hydroxide, concentrated
Acetic acid
Ammonium acetate (4.6M)
Sodium chromate solution (0.3M)
Sodium carbonate solution, saturated
Sodium hydroxide solution (6M)
Strontium carrier solution (5 mg strontium per mL as nitrate, standardized)
Yttrium carrier solution (10 mg yttrium per mL as nitrate, standardized)
Barium carrier solution (10 mg barium per mL)
Sr-SPEC pre-packed column (available from Eichrom Industries, Inc.)
Liquid scintillation cocktail (e.g., HISAFE III or equivalent).

Procedure

Preconcentration

(1) Transfer 250 mL of urine sample to a 500 mL beaker.
(2) Add 1 mL of strontium carrier.
(3) Add 2 mL of concentrated H_3PO_4.
(4) Stir solution for 1 h in a hot water bath.
(5) Add concentrated NH_4OH and adjust pH to a value of 10; leave solution for 1 h without stirring.
(6) Centrifuge solution; discard supernatant and calcine residue at 350–400°C.

Ca/Sr separation (crown ether method)

(1) Dissolve residue in 20 mL HNO_3 (8N).
(2) Transfer sample solution onto Sr-SPEC column.
(3) Rinse beaker with 3 mL HNO_3 (8M) and add to column after feed has passed.
(4) Rinse column three times with 3 mL HNO_3 (8M).
(5) Record the end time of the last rinse at the start of yttrium ingrowth (t_1).
(6) Elute the strontium with 10 mL of distilled water and collect it in a centrifuge tube.
(7) If the activity of ^{85}Sr is to be determined, the eluent should be counted in a γ spectrometer (the 514 keV line).

Ca/Sr separation (fuming HNO_3 method)

(1) Add 10 mL distilled water to the residue in the centrifuge tube in an ice bath, followed by 22.5 mL of fuming nitric acid (cautiously) to precipitate the strontium and stir for 30 min.

(2) Let stand overnight.
(3) Filter on fibreglass filter paper and wash with fuming HNO_3.
(4) Dissolve the crystals in distilled water and add 1 mL of yttrium carrier.
(5) Adjust the pH with concentrated NH_4OH; record time and date (t_1).
(6) Cool to room temperature, centrifuge and transfer supernatant to a new centrifuge tube.
(7) Add 1 mL of barium carrier.
(8) Adjust pH to 5.5 by addition of 1 mL of acetic acid and 2 mL of ammonium acetate.
(9) Heat in the water bath at 90°C.
(10) Add 1 mL Na_2CrO_4 solution, drop by drop, while stirring. The solution should have the colour of chromate ions.
(11) Stir vigorously until precipitation complete.
(12) Let cool, centrifuge and transfer the supernatant to a clean centrifuge tube.
(13) Discard the precipitate.
(14) If the activity of ^{85}Sr is to be determined, the supernatant should be counted in a γ spectrometer (the 514 keV line).

Source preparation

(1) Allow a 7 d period for ingrowth of ^{90}Y.
(2) After this period, add 1 mL of yttrium carrier and transfer the tube to a water bath at 100°C.
(3) Precipitate the yttrium present with NH_4OH; leave the tube in the hot water bath for 5 min after precipitation.
(4) Record the time of this precipitation (t_2).
(5) Cool the tube to room temperature and centrifuge for 15 min at 3000 rev./min.
(6) Keep the supernatant for strontium yield calculation and also for ^{89}Sr analysis if needed.
(7) Dissolve the residue in a few drops of concentrated HCl diluted to 35 mL with deionized water and place in the water bath.
(8) After 10 min, add a saturated solution of oxalic acid, stir for at least 5 min and allow the precipitate to form for 15 min.
(9) Cool the centrifuge tube to room temperature and filter the precipitate on a millipore filter.
(10) Dry the filter under an IR lamp, weigh, mount on a Lucite disc and count.
(11) Dilute the supernatant at 125 mL and add a few drops of NaOH.
(12) Boil to eliminate any trace of NH_4^+.
(13) Add 10 mL of saturated solution of Na_2CO_3 and stir vigorously. Let it cool and filter on fibrepaper, dry under an IR lamp and weigh the precipitates.

(14) Compute chemical yields as follows:

$$S = 0.1 \times \text{weight } SrCO_3 \times \frac{211.63}{147.63} \quad \text{(strontium yield)}$$

$$Y = 0.05 \times \text{weight } Y_2(C_2O_4)_3 \times \frac{274.92}{441.87} \quad \text{(yttrium yield)}$$

Calculation of results

Calculation of ^{90}Sr content based on the ^{90}Y count

The ^{90}Sr activity (Bq) is calculated by using the following expression:

$$^{90}\mathrm{Sr} = \left(\frac{N_s}{T_s} - \frac{N_b}{T_b}\right)\frac{C_1 C_2}{SYE}$$

where

N_s are the counts measured for the time T_s in the $Y_2(C_2O_4)_3$ precipitate,
N_b are the counts due to background for the time T_b,
T_s and T_b are sample and background counting times, respectively, in seconds,
C_1 is the growth factor of ^{90}Y as a function of its physical half-life of 64 h,

$$C_1 = 1 - \exp\left(-\frac{0.693}{64}(t_2 - t_1)\right) \quad (t_1 \text{ and } t_2 \text{ in hours})$$

C_2 is the decay factor of ^{90}Y since the second precipitation of $Y(OH)_3$ at time t_2 and the mean time of the counting period t_3,

$$C_2 = 1 - \exp\left(-\frac{0.693}{64}(t_3 - t_2)\right) \quad (t_2 \text{ and } t_3 \text{ in hours})$$

S is the strontium yield (%),
Y is the yttrium yield (%) and
E is the counter efficiency for ^{90}Y (%).

The detection limit for ^{90}Sr based on the ^{90}Y count as described above is 20 mBq/L. The above calculation yields the activity of ^{90}Sr in the sample (or aliquot) analysed. In order to convert the results to Bq/24 h of urine, the results are multiplied by the ratio of the 24 h urine volume excreted (either actual or assumed) to the sample (or aliquot) volume analysed.

^{89,90}Sr content determined by liquid scintillation counting

This technique is a direct measurement of ^{90}Sr activity that does not rely on ^{90}Y ingrowth. From the crown ether method, the eluted fraction of ^{90}Sr is collected directly in a vial containing the appropriate liquid scintillation cocktail. The vial is then counted in a liquid scintillation counter. By selecting two different energy regions on the same counter, say a low energy region from 20–50 keV and a higher energy region extending from 500 to 1400 keV, the ^{90}Sr activity of the sample can be calculated.

The net count rate of an energy region is given by

$$X - B = A_{Sr\text{-}90}\left\{E_{xSr\text{-}90}\exp(-\lambda_{Sr\text{-}90}t) + E_{xY\text{-}90}\left[1-\exp(-\lambda_{Y\text{-}90})t)\right]\right\}$$

where

X is the counting rate of the sample in the energy region,
B is the corresponding background counting rate,
$A_{Sr\text{-}90}$ is the activity of ^{90}Sr,
$E_{xSr\text{-}90}$ is the counting efficiency for ^{90}Sr in the energy region x,
$E_{xY\text{-}90}$ is the counting efficiency for ^{90}Y in the energy region x,
$\lambda_{Sr\text{-}90}$ is the decay constant of ^{90}Sr,
$\lambda_{Y\text{-}90}$ is the decay constant of ^{90}Y and
t is the time elapsed since the separation of ^{90}Sr.

If ^{89}Sr is also present, another term must be added to the above expression,

$$+A_{Sr\text{-}89}E_{xSr\text{-}89}\exp(-\lambda_{Sr\text{-}89}t)$$

where

$E_{xSr\text{-}89}$ is the counting efficiency for ^{89}Sr in the energy region x, and
$\lambda_{Sr\text{-}89}$ is the decay constant of ^{89}Sr.

This implies that for each isotope, in each energy region, the counting efficiency must be determined. During the first few hours after strontium separation, ^{90}Y is a very minor contributor to the spectrum and thus ^{90}Sr can be easily estimated. The ingrowth of ^{90}Y can be followed by re-counting the sample several times over successive days.

Annex IV

ANALYSIS FOR ^{226}Ra, ^{228}Ra AND ^{210}Pb IN BIOLOGICAL SAMPLES

Method provided by the staff of the Instituto de Radioproteção e Dosimetria – IRD/CNEN, Rio de Janeiro, Brazil

INTRODUCTION

Biological samples are put into solution through dry and wet ashing. The radium isotopes are co-precipitated with barium sulphate, and ^{210}Pb is precipitated as lead sulphate. The precipitates are then separated and transferred to filter papers, and chemical yields are determined gravimetrically. The filter papers are then counted with a proportional counter. Physical samples (workplace air monitor filters, etc.) may also be analysed using this method by following the procedure for faecal samples. Because the lead carrier may contain ^{210}Pb, careful determination of an appropriate blank is essential for ^{210}Pb analysis [IV–1].

Apparatus

Muffle furnace (400°C)
Hotplate
Laboratory oven (100°C)
Water bath
Desiccator
Centrifuge (3000 rev./min)
Laboratory glassware
Whatman GF/A, 40 and 44 filter papers
Counter sample holder
Proportional counter.

Reagents

HNO_3 (concentrated, 1M and 5M)
H_2O_2
HF (40%), 23.2M
$HClO_4$ (70%), 11.6M
NH_4OH (concentrated)
H_2SO_4 (0.1M and 3M)
NaOH (6M)

$(NH_4)_2SO_4$ (25 mg/mL)
KCl (0.1 g/mL)
Na_2S (1M)
Na_2CrO_4 (30%)
Ethanol (50%)
Citric acid (1M)
Glacial acetic acid
Ammonium acetate (40%)
Phenol (1%)
Methyl red
Barium carrier (20 mg Ba^{2+}/mL)
Lead carrier (20 mg Pb^{2+}/mL)
Titriplex-I (nitrilo-tri-acetic acid, NTA)
Titriplex-III (ethylenedinitrilo tetraacetic acid disodium salt dihydrate, EDTA)
Distilled water.

Procedure

1. Preparation of faeces samples

1.1. Ash the sample in a furnace (400°C for 24 h).
1.2. Weigh 1.5 g of ash and transfer it to a 250 mL volume Teflon beaker.
1.3 Add 1 mL of barium carrier and 1 mL of lead carrier.
1.4. Slowly add 68 mL of concentrated HNO_3 and 30 mL of H_2O_2. Cover the beaker with a watch glass and heat it on a hotplate until the solution becomes clear.
1.5. Filter the solution through a glass microfibre filter (Whatman GF/A) and reserve the filtered solution.
1.6. Transfer the fibre filter to a Teflon beaker and add 10 mL of HNO_3, 10 mL of HF and 2 mL of $HClO_4$.
1.7. Cover the beaker and heat it on the hotplate for 2 h or to incipient dryness.
1.8. Evaporate the solution, avoiding drying it out.
1.9. If necessary repeat steps 1.6 to 1.8.
1.10. Dissolve the residue with 30 mL of HNO_3 (1M).
1.11. Mix the solution from 1.10 with the filtrate from 1.5, and bring the volume to 1 L with HNO_3 (1M).

2. Preparation of urine samples

2.1. Measure the volume of the urine sample and transfer it to a beaker with a capacity twice the sample's volume.

2.2. Add 1 mL of barium carrier and 1 mL of lead carrier.

2.3. Add to the sample a volume of concentrated HNO_3 equal to half the original volume of the initial urine sample.

2.4. Evaporate the solution on a hotplate, adding HNO_3 until total destruction of the organic matter has been achieved. If necessary, add some drops of H_2O_2.

2.5. Transfer the residue to a beaker and complete the volume to 1 L by adding HNO_3 (1M).

3. *Separation of Pb and Ra*

3.1. To the solution from steps 1.11 or 2.5, add 5 mL of citric acid (1M), with 1% phenol and some drops of methyl red.

3.2. Add concentrated NH_4OH until the indicator changes colour (red/yellow). If any solids are not completely dissolved, add citric acid until complete dissolution is achieved.

3.3. Heat the solution on a hotplate until the boiling point and slowly add 50 mL of H_2SO_4 (3M), stirring the solution, until a pH of 1.5 is attained.

3.4. Decant the solution overnight.

3.5. Discard the aqueous phase and transfer the precipitate to a centrifuge tube with 100 mL capacity. Use distilled water to help in this transfer. Centrifuge at 3000 rev./min for 10 min and discard the supernatant.

3.6. Wash the precipitate with 20 mL of H_2SO_4 (0.1M). Centrifuge at 3000 rev./min for 10 min and discard the aqueous phase.

3.7. Add 2 g of Titriplex-I (NTA) to the precipitate, 30 mL of distilled water, some drops of methyl red and 7 mL of NaOH (6M). Heat in a water bath (60°C) until the precipitate is completely dissolved. If the solution is not clear, add more NaOH and Titriplex-I (NTA).

3.8. While still heating in the water bath, add 5 mL of $(NH_4)_2SO_4$ (25 mg/mL). Adjust the pH to 4.5–5.0, with glacial acetic acid, using methyl red as an indicator. Leave the solution in the water bath for 30 min, remove it and leave it standing until the next day.

3.9. Centrifuge the solution for 10 min at 3000 rev./min. Separate the supernatant and reserve for later lead analysis.

3.10. Wash the precipitate with ammonium acetate (40%). Centrifuge and separate the supernatant, mixing it with the previous solution for lead analysis. The precipitate will be used for the radium analysis.

4. *^{226}Ra and ^{228}Ra analysis*

4.1. Dissolve the precipitate with 2 g of Titriplex-III (EDTA), 30 mL of distilled water and four drops of methyl red. Add 5 mL of concentrated NH_4OH, in a heated water bath (80°C), until the indicator changes colour. If the precipitate is not completely dissolved, add more Titriplex-III (EDTA).

4.2. While the solution is still hot, add 5 mL of $(NH_4)_2SO_4$ (25 mg/mL). Adjust the pH to 4.5–5.0 with glacial acetic acid, observing the indicator changing colour.

4.3. Note and record the date and the time of the precipitation and continue the analysis on the next day.

4.4. Filter the precipitate with Whatman 44 (with a diameter of 2.5 cm) paper filter, using a filtration system. Wash with ethanol (80%) and dry it at 80°C for 15 min. Let it stand on a Petri dish until the following day. Weigh the filter to obtain the chemical yield (20 mg Ba^{2+} = 34 mg $BaSO_4$).

4.5. Transfer the filter paper containing the precipitate to a sample holder for counting, for example, a stainless steel disc, fixing it with a PVC ring.

4.6. Wait at least 21 d after the precipitation step to count the sample in a proportional detector. Do one count with the sample uncovered (to count α plus β particles), and then a second count with the sample covered with a filter paper to count only the β particles and not the α particles.

5. *Determination of ^{210}Pb*

5.1. To the solution that was obtained in step 3.9, add 1 mL of Na_2S (1M), and heat it in a water bath for 10 min.

5.2. Let the solution cool to room temperature, then centrifuge and discard the supernatant.

5.3. Wash the precipitate with distilled water and repeat the centrifugation.

5.4. Add to the precipitate, under heat, 5 mL of HNO_3 (5M) until the sulphide dissolves. Leave it for some minutes under heat.

5.5. After cooling to room temperature, filter the solution with Whatman 40 filter paper, using a 250 mL beaker.

5.6. Add methyl red to the solution as an indicator and adjust the pH of the solution to 4.5–5.0 with ammonium acetate (40%).

5.7. Heat the solution and precipitate with 2 mL Na_2CrO_4 (30%). Note and record the date and the time of the precipitation.

5.8. Filter the solution through Whatman 44 filter paper, of 2.5 cm diameter.

5.9. Wash the filter with ethanol (80%) and dry it at 100°C in a laboratory oven, for 15 min. Place the filter paper in a desiccator until room temperature is reached.

5.10. Weigh the filter to determine the chemical yield (20 mg Pb^{2+} = 31.2 mg $PbCrO_4$).

5.11. Cover the filter with adhesive plastic, wait 10 d, and count with a proportional detector.

6. *Calculations*

6.1. The ^{210}Pb activity can be found from

$$A_{Pb\text{-}210} = \frac{R_m - R_0}{60 R_q Q E_\beta [1 - \exp(-\lambda_{Bi\text{-}210} t)]} \text{(Bq/kg or Bq/L)}$$

where

R_m is the total counting rate (counts/min) of the sample,
R_0 is the counting rate (counts/min) of the background,
R_q is the chemical yield,
Q is the amount of sample used in the analysis (mass (kg) or volume (L)),
E_β is the counting efficiency (β of ^{210}Bi),
$\lambda_{Bi\text{-}210}$ is the decay constant of ^{210}Bi (=0.1383 d^{-1}), and
t is the time (d) elapsed between the precipitation and counting of $PbCrO_4$.

Note: The β efficiency of ^{210}Bi is obtained using three standard solutions of ^{210}Pb with around 4 Bq for each one. These standard solutions and the samples must be submitted to the same treatment from, and including, step 3.6. Then, the mean efficiency will be estimated.

6.2. The ^{226}Ra activity can be found from

$$A_{Ra\text{-}226} = \frac{R_m - R_0}{60 R_q Q E_\alpha \eta_{abs(Ra\text{-}226)} \{1 + K[1 - \exp(-\lambda_{Rn\text{-}222} t)]\}} \text{(Bq/kg or Bq/L)}$$

where

R_m is the total counting rate (counts/min) of the sample,
R_0 is the counting rate (counts/min) of the background,
R_q is the chemical yield,
Q is the amount of sample used in the analysis (mass (kg) or volume (L)),
E_α is the counting efficiency (α of ^{241}Am) using a source with $d = 2.5$ cm,
$\eta_{abs(Ra\text{-}226)}$ is the self-absorption coefficient,
$\lambda_{Rn\text{-}222}$ is the decay constant of ^{222}Rn (=0.181 d^{-1}),
t is the time (d) elapsed between the precipitation of radium and its counting, and
K is expressed by the following equation:

$$K = \frac{\eta_{abs(Rn\text{-}222)} + \eta_{abs(Po\text{-}218)} + \eta_{abs(Po\text{-}214)}}{\eta_{abs(Ra\text{-}226)}}$$

Note: The values of $\eta_{abs(Ra\text{-}226)}$ and K are obtained by α counting immediately after the $Ba(Ra)(SO_4)$ precipitate is dried. During the following weeks repeat the α counting with at least three ^{226}Ra standards, each with around 4 Bq, treated exactly like the sample in Section 4.

For each standard, plot $1 - e^{-0.181t}$ versus $(R_m - R_0)/60R_qE_\alpha A_{Ra\text{-}226}M$, where M is the mass (g) of the ^{226}Ra standard and $A_{Ra\text{-}226}$ is the specific activity (Bq/g) of the standard.

The point where the straight line intercepts the ordinate is the value of $\eta_{abs(Ra\text{-}226)}$. The slope of the line is the product of $\eta_{abs(Ra\text{-}226)}$ and K. The value adopted for both parameters is the mean value.

6.3. The ^{228}Ra activity can be found from

$$A_{Ra\text{-}228} = \frac{R_m - R_0}{60R_qQE_{Ra\text{-}228}} - \frac{E_{Ra\text{-}226}A_{Ra\text{-}226}\left[1-\exp(-\lambda_{Rn\text{-}222}t)\right]}{E_{Ra\text{-}228}} \quad \text{(Bq/kg or Bq/L)}$$

where

R_m is the total counting rate (counts/min) of the sample,
R_0 is the counting rate (counts/min) of the background,
R_q is the chemical yield,
Q is the mass or volume of sample utilized in analysis (mass (kg) or volume (L)),
$E_{Ra\text{-}228}$ is the counting efficiency (β of ^{228}Ra),
$E_{Ra\text{-}226}$ is the counting efficiency (β of ^{226}Ra),
$A_{Ra\text{-}226}$ is the activity concentration of ^{226}Ra (Bq/kg or L),
t is the time (d) elapsed between the precipitation of radium and its counting, and
$\lambda_{Rn\text{-}222}$ is the decay constant of ^{222}Rn = 0.181 d^{-1}.

6.3.1. Counting efficiency for the ^{226}Ra β particles

The three standards of ^{226}Ra with around 4 Bq for each, which had the same treatment of the sample (since step 4), are assembled. After 21 d, they are covered with a filter paper for β counting. The efficiency is calculated using the following equation:

$$E_{Ra\text{-}226} = \frac{R_m - R_0}{60R_qA_{Ra\text{-}226}M\left[1-\exp(-\lambda_{Rn\text{-}222}t)\right]}$$

where

R_m is the total counting rate (counts/min) of the standard,
R_0 is the counting rate (counts/min) of the background,

R_q is the chemical yield,
$A_{Ra\text{-}226}$ is the activity concentration of the ^{226}Ra standard (Bq/g),
t is the time (d) elapsed between the precipitation of radium and the counting,
$\lambda_{Rn\text{-}222}$ is the decay constant of ^{222}Rn (=0.181 d^{-1}), and
M is the mass (g) of the standard.

The efficiency to be used is the average of the three calculated values.

6.3.2. Counting efficiency for the ^{228}Ra β particles

The three standards of ^{228}Ra with around 4 Bq for each, which had the same treatment of the sample (since step 4), are assembled. After 21 d, they are covered with a filter paper for β counting. The efficiency is calculated using the following equation:

$$E_{Ra\text{-}228} = \frac{R_m - R_0}{60 R_q A_{Ra\text{-}228} M}$$

where

R_m is the total counting rate (counts/min) of the standard,
R_0 is the counting rate (counts/min) of the background,
R_q is the chemical yield,
$A_{Ra\text{-}228}$ is the activity concentration of the ^{228}Ra standard (Bq/g), and
M is the mass (g) of the standard.

The efficiency to be used is the average of the three calculated values.

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Annex V

ANALYSIS FOR URANIUM IN URINE BY FLUORIMETRY

Method provided by the staff of the Instituto de Radioproteção e Dosimetria – IRD/CNEN, Rio de Janeiro, Brazil

INTRODUCTION

Fluorescence is widely used for monitoring natural uranium in urine. Ultraviolet fluorimetry requires the counting of a fired glass bead prepared by addition of an alkali fluoride flux to a preconcentrated urine sample. Careful calibration of the UV fluorimeter is required.

Apparatus

Fume hood
Hotplate and IR lamp
Glassware (preleached)
Platinum crucibles
UV fluorimeter
Standard safety equipment (safety glasses, rubber gloves, protective clothing).

Reagents

Concentrated HNO_3
HNO_3 (1N)
H_2O_2 (30% solution)
Phosphoric acid (1M)
Alkaline flux (98% NaI, 2% LiF)
Uranium standard (100 µg/L) in HNO_3.

Procedure

Pre-leach all laboratory glassware to remove leachable uranium by filling with HNO_3 1:1, heating to 60°C on a controlled hotplate overnight, cooling and rinsing with deionized water.

Preconcentration

1. Transfer 50 mL of urine into a clean, preleached 100 mL beaker.

2. Add 25 mL of concentrated HNO_3 and heat on a hotplate.
3. Add small quantities of concentrated HNO_3 and H_2O_2 (30%) while heating and evaporate to dryness.
4. If a clear residue is not obtained, add 25 mL of concentrated HNO_3 and repeat step 3.
5. Once a clear residue is obtained (free of organic material), restore to the original volume (50 mL) with 1M HNO_3.

Fluoride fusion

1. Pipette 100 µL of the sample into a clean platinum crucible.
2. Pipette 100 µL of a uranium standard solution (100 (µg/L) into another clean platinum crucible. (The standard will be used to adjust the calibration of the fluorimeter.)
3. Analyse both the sample and the standard in triplicate.
4. Dry the sample and the standard under an IR lamp.
5. Add 400 mg of alkaline flux (98% NaF and 2% LiF) to each crucible containing a sample or the standard solution.
6. Add 400 mg of alkaline flux to each of the three empty and clean platinum crucibles for determination of the blank.
7. Place the sample, standard, blank and an empty crucible in a muffle furnace and heat to 1000°C for 20 min. The empty crucible is used to set the fluorimeter on the zero position.
8. Remove from furnace and cool the crucibles.

Measurement

1. Place the crucibles in a calibrated UV fluorimeter, for reading.
2. Establish the fluorimeter linear calibration using uranium standards with concentrations from 5 to 200 µg/L.
3. Determine the uranium concentration in the sample using the linear regression calibration curve of the fluorimeter.
4. The MDA of this method is approximately 5 µg/L.

Annex VI

ANALYSIS FOR THORIUM IN SAMPLES

Method provided by the staff of the Instituto de Radioproteção e Dosimetria –
IRD/CNEN, Rio de Janeiro, Brazil

INTRODUCTION

Urine and faecal samples and filters are ashed and dissolved. Thorium is separated by liquid–liquid solvent extraction [VI–1]. The source for α-spectrometry is prepared by deposition in a stainless steel disc covered with a thin film of tri-n-octyl phosphinoxide/vinnol/cyclohexanone [VI–2].

Apparatus

Stirring motor
Shaker
Muffle furnace
Quartz capsule, platinum crucible
Deposition cell
Alpha spectrometer
Hotplate
Glassware, beakers, centrifuge tubes.

Reagents

All reagents should be of analytical grade.

Vinnol (Wacker, Germany) (polyvinyl chloride powder)
H_3BO_3 (powder)
K_2CO_3 (powder)
HNO_3 (0.01M, 1M, 8M)
H_2O_2 (30%)
HNO_3 (65% (14.4M))
NH_4OH concentrated (13.3M)
$Ca(NO_3)_2$ (1M)
H_3PO_4 (25.4M)
$Al(NO_3)_3$ (2.8M)
Xylene
Alamine-336

HCl (7M)
Cyclohexanone
TOPO (tri-n-octyl phosphinoxide)
^{229}Th tracer in HNO_3 (1M).

Procedure

1. Preparation of urine samples

1.1. Measure the volume of urine *V* (L) and transfer it to a beaker.
1.2. Add approximately 100 mBq of the tracer (^{229}Th).
1.3. Add concentrated HNO_3 to the sample in the beaker, in a volume equal to 50% of the volume of the original urine sample.
1.4. Heat the solution on a hotplate, until almost dry. Add HNO_3 until a clear residue is obtained. If necessary add some drops of H_2O_2.
1.5. Transfer this residue to a 100 mL centrifuge tube, with 30 mL of 0.01M HNO_3.
1.6. Proceed to Section 4, Precipitation.

2. Preparation of faeces samples

2.1. Weigh the sample and transfer the frozen faeces sample to a quartz capsule and ash it for 24 h at 400°C.
2.2. Weigh the total mass of the ashes obtained and transfer 1 g of this to a 50 mL platinum crucible.
2.3. Add approximately 100 mBq of ^{229}Th tracer.
2.4. Fuse the sample with 2 g of H_3BO_3 and 2 g of K_2CO_3 and re-ash at 1000°C for 15 min.
2.5. Prepare a beaker with 2 L of cold water.
2.6. Take out the platinum crucible and plunge it into the cold water.
2.7. Decompose the melt in 60 mL of 1M HNO_3 in a 250 mL beaker.
2.8. Heat the beaker on the hotplate until a transparent liquid is obtained.
2.9. Transfer the solution to a 100 mL centrifuge tube.
2.10. Proceed to Section 4, Precipitation.

3. Preparation of filters

3.1. Fibreglass filters (PAS)

3.1.1. Put the filter in a Teflon beaker.
3.1.2. Add the ^{229}Th tracer.
3.1.3. Add 10 mL of concentrated HNO_3 and 5.0 mL of concentrated HF.

3.1.4. Heat the sample on a hotplate and evaporate it until almost dry (do not boil the solution).
3.1.5. Cover the beaker.
3.1.6. Cool the sample.
3.1.7. Add 5.0 mL of concentrated pericloric acid and 5.0 mL of concentrated HNO_3.
3.1.8. Evaporate this until dryness.
3.1.9. Add 10 mL of a solution of HNO_3 and HCl in the proportion of 1:3.
3.1.10. Evaporate until almost dryness.
3.1.11. Repeat steps 9 and 10.
3.1.12. Add 10 mL of 1N HNO_3, washing the beaker walls.
3.1.13. Evaporate until almost dryness.
3.1.14. Take out the sample from the hotplate and add 10 mL of 1M HNO_3 and 10 mL of 2.8M $Al(NO_3)$ and proceed to Section 5, Solvent Extraction.

3.2. Polycarbonate filters

3.2.1. Put the filter in a platinum crucible.
3.2.2. Add approximately 4 dis/min of ^{229}Th tracer.
3.2.3. Fuse the sample with 2 g of H_3BO_3 and 2 g of K_2CO_3 and re-ash at 1000°C for 15 min.
3.2.4. Prepare a beaker with 2 L of cold water.
3.2.5. Take out the platinum crucible and bathe it in the cold water.
3.2.6. Decompose the melt in 60 mL of 1M HNO_3 in a 250 mL beaker.
3.2.7. Heat the beaker on the hotplate until a transparent liquid is obtained.
3.2.8. Transfer the solution to a 100 mL centrifuge tube.
3.2.9. Proceed to Section 4, Precipitation.

4. Precipitation

4.1. Add concentrated NH_4OH to the solution from steps 1.5, 2.9 and 3.2.8 in the centrifuge tube raising the pH value to 7. If the precipitate has a volume which is less than a quarter of the sample volume, add 1 mL of concentrated solution of $Ca(NO_3)_2$ and 2 mL of H_3PO_4.
4.2. Centrifuge for 5 min at 3000 rev./min.
4.3. Take the precipitate and dissolve it with 4 mL of 8M HNO_3.
4.4. Add 10 mL of 1M HNO_3 and 10 mL of 2.8M $Al(NO_3)_3$ to the solution.

5. Solvent extraction

5.1. Transfer the solution to a 60 mL separation funnel.
5.2. Add 11 mL xylene and 1.25 mL alamine-336.

5.3. Shake the solution for 5 min and let it stand until complete phase separation.

5.4. Put the aqueous phase into another separation funnel and repeat the procedure.

5.5. Discard the aqueous phase and combine the two organic phases.

5.6. Wash the organic phase twice with 20 mL of 7M HCl, shaking it for 5 min to extract the thorium.

5.7. Combine the two aqueous solutions in the same beaker and dry the liquid on a hotplate.

5.8. Add 20 mL of concentrated HNO_3 and dry it again.

5.9. Dissolve the residue with 3 mL of 1M HNO_3.

6. *Deposition*

6.1. Transfer the solution to the deposition cell [VI–1], which contains a stainless steel disc covered with a thin film of tri-n-octyl phosphinoxide/vinnol/cyclohexanone [VI–2].

6.2. Stir the solution for 4 h at 500 rev./min.

6.3. Remove the steel disc and put it in a muffle furnace at 400°C for 1 h.

6.4. The deposition may follow any of the techniques described in Chapter 5, Section 5.2.3.

7. *Counting*

7.1. Count in an α spectrometry system.

8. *Calculations*

The calculations are carried out using the following formulas:

8.1. Determination of chemical recovery,

$$R_f = \frac{R_1 - R_2}{T_c E_d A_t}$$

where

R_f is the recovery factor for the tracer,
R_1 is the total counts in the tracer region,
R_2 is the total background counts in the tracer region,
T_c is the counting time (min),
E_d is the detector efficiency, and
A_t is the tracer activity (dis/min).

8.2. Determination of sample specific activity,

$$A = \frac{R_3 - R_4}{T_c 60 R_f E_d (W\ or\ Vol)}$$

where

A is the sample specific activity (Bq/g or Bq/mL),
R_3 is the total counts in the radionuclide region,
R_4 is the total background counts in the radionuclide region,
60 is the conversion factor from dis/min to Bq,
W or Vol is the mass of faeces ash or volume of urine (g or mL),
R_f is the recovery factor,
E_d is the detector efficiency, and
T_c is the counting time (min).

For urine measurements of thorium, in most situations the concentration of thorium will be below the MDA and other methods might be used, as for example the ICP-MS. The procedure for the ICP-MS is as follows:

1. Take 1 mL of the urine sample and transfer it to a beaker that was washed with bidistilled water (do not delay between urine collection and analysis).
2. Add 19 mL of 1N HNO_3, using analytical grade acid and distilled and deionized water.
3. Sample is ready to be analysed at the ICP-MS.

(MDA of the order of 10^{-12} g thorium).

The α spectrometry method is usually not appropriate to be used with the filters from the cascade impactor, since the mass collected at each stage is too small to allow measurements above the detection limits. In general, mylar films are used as impactor surfaces and are analysed by particle induced X ray emission (PIXE), neutron activation analysis or ICP-MS. The cut-off diameters of each stage and the number of stages of each cascade impactor vary. The particulates are separated by their aerodynamic diameters through the mass collected at each stage of the cascade. Using the PIXE method, the elemental composition of the mass impacted at each stage is quantified. The log–normal size distributions of airborne particulates is determined and the mass median aerodynamic diameter (MMAD) and the geometrical standard deviation are characterized. For ^{232}Th in radioactive equilibrium with its daughters, the thorium mass concentration is almost entirely due to ^{232}Th. The particulate size distribution thus has the same log–normal shape for both mass and activity. As a consequence, the determined MMAD is equal to the activity median aerodynamic diameter (AMAD).

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[VI–2] SACHETT, I.A., NOBREGA, A.W., LAURIA, D.C., Determination of uranium isotopes by chemical stripping and α-spectrometry, Health Phys. **46** (1984) 133.

Annex VII

ANALYSIS FOR PLUTONIUM, AMERICIUM AND CURIUM IN URINE AND FAECAL SAMPLES BY ALPHA SPECTROMETRY

Based upon procedures used at the National Radiological Protection Board, UK

INTRODUCTION

The analysis of plutonium, americium and curium in bioassay samples is normally achieved by radiochemical separation from samples to which tracers have been added. Samples of urine and faeces are ashed prior to separation of the actinides by ion exchange. The actinides are then plated onto stainless steel discs before assay by α spectrometry. A good recovery can be obtained using the detailed procedures described below.

Apparatus

Muffle furnace
Fume hood
Infrared lamp
Hotplate
Ion exchange columns
Alpha spectrometer.

Reagents

The reagents used should be of AnalaR quality or equivalent. Water used as a reagent in the analyses must be distilled, deionized or of AnalaR quality.

Except where specifically noted in the method, the solution strengths are only approximate and can be made up by dilution of commercial concentrated acids. No standardization is required.

The following are required:

16M, 12M, 8M and 2M HNO_3
5–10% HF in 8M HNO_3
Aluminium nitrate solution (0.5 $g \cdot mL^{-1}$)
Sodium nitrate
0.88 ammonia solution
12M, 1.5M HCl

12M HCl + 2% HI (freshly prepared)
12M, 1M H_2SO_4
Dowex or BioRad 1-X8 (50–100 or 100–200 mesh)
Bio-Rad AG-MPI macroporous anion exchange resin
Dibutyl-N,N-diethylcarbamylphosphonate (DDCP)
Calcium carrier (200 mg mL^{-1})
Phosphoric acid.

Special reagents needed for americium and curium separation

Prepare the following as needed:

Reagent A	90 mL methanol + 10 mL 10M nitric acid (90% methanol, 1 M nitric acid),
Reagent B	80 mL methanol + 20 mL 2.5M ammonium thiocyanate (80% methanol, 0.5M ammonium thiocyanate),
Reagent C	80 mL methanol + 5 mL 10M nitric acid + 15 mL water (80% methanol, 0.5M nitric acid),
Reagent D	60 mL methanol + 5 mL 10M nitric acid + 35 mL water (60% methanol, 0.5M nitric acid).

Do not store mixtures of nitric acid and methanol, concentrated acid–alcohol mixtures are used as rocket fuel. Keep all mixtures covered to minimize evaporation while not in use.

Procedure

Sequential methods for separation of plutonium, americium and curium in one sample are usually possible, but this should be confirmed for the sample concerned as recoveries in the later stages in a chain can be low. Where sequential analyses are to be performed they should be in the order plutonium first, americium second and curium third.

If only americium and or curium are to be determined, the initial steps in the plutonium method must be followed to remove any plutonium which might otherwise interfere with the α spectra.

Sample sizes and addition of tracers

The preferred tracers of Pu, Am and Cm are: $^{236 \text{ or } 242}Pu$, ^{243}Am and ^{244}Cm.

Pu-242 is the preferred plutonium tracer for low level work because it has a lower energy than the isotopes being determined. This eliminates ‘tailing’ of its spectra and interference with that of the isotope being determined.

Tracers should be added at the first possible opportunity. Where the whole of the sample is to be analysed, this would be before any dissolution or other treatment. In all analyses the tracer and the unknown isotope should be present in approximately equal quantities. If the expected sample activity is high, homogeneity requirements may require dissolution of a larger sample without tracers. Aliquots from this solution may then have tracer added and be used for analysis.

All other samples should have reagent blanks analysed alongside a sample batch at regular intervals. However, for samples expected to contain less than 5 mBq it is essential that a blank is analysed with every sample batch. This would usually be a reagent blank, but a 'blank' material sample could be used. If the chemical recovery of the tracer on any radiochemical measurement is less than 30%, then a repeat analysis should be performed if possible.

Except were specifically noted, reagent quantities are not precise. Volumes can be measured with a measuring cylinder, pipette or similar device. Mass need only be measured to the accuracy quoted in the method and can be estimated where the method uses the term 'approximate'.

Preparation and dissolution of urine samples

A 24 h urine sample is normally taken for radiochemical analysis.

1. After the addition of yield tracers and carriers the sample is digested with 8M nitric acid, cooled and the insoluble phosphates precipitated with ammonia. The precipitate is filtered off, ashed and dissolved in nitric acid ready for analysis.
2. Weigh the appropriate yield tracers into a 5 L beaker then add the sample (about 1.5 L). Wash the sample container with several portions of 8M nitric acid, adding the washings to the sample. Add a further 8M nitric acid to the sample until approximately 400 mL of nitric acid per litre of sample have been used. Then add 1 mL of 200 $mg{\cdot}mL^{-1}$ calcium carrier, 3 mL of phosphoric acid and a magnetic stirring bar.
3. Cover with a watch glass and place on a hotplate. Heat to boiling and simmer for about 30 min until the solution is of a pale straw colour.
4. Cool to room temperature. While stirring, add 0.880 ammonia solution until a precipitate forms. Remove the stirring bar.
5. After allowing the precipitate to settle, filter through a fan folded 24 cm Whatman No. 541 filter paper. Wash the beaker with water, transferring all precipitate into the filter. When dry, transfer the filter to a 150 mL Pyrex beaker labelled with a fireproof pen. Ash the precipitate and paper in a furnace, raising the temperature slowly to 500°C.
6. Cool to room temperature and add a minimum of nitric acid. Evaporate to dryness and re-ash at 500°C.

7. Cool to room temperature and dissolve the ash in 8M nitric acid. Cover the beaker with a watch glass and boil the solution gently on a hotplate for at least 20 min to hydrolyse the interfering condensed phosphates formed during the ashing process.
8. Continue the analysis as described below for the determination of plutonium. Even if only americium is to be determined, the initial steps of the plutonium method must be followed.

Preparation of faecal samples

Faecal samples are normally supplied in a polythene bag.

1. Weigh the sample including its bag into a beaker, if possible weigh a similar bag to provide an estimate of the true sample weight. Cut the bag open with scissors to prevent any gas pockets forming during ashing. As an alternative, a frozen sample can usually be easily and cleanly removed from its bag for a more accurate determination of the sample weight. Whatever the procedure chosen, a clear record shall be kept as to whether the bag was ashed with the sample or not.
2. Place the beakers in a furnace set at 110°C and leave at this temperature for several hours. Progressively raise the temperature at ~2°C·h^{-1} to 450°C and then at 25°C·h^{-1} to 500°C. This procedure may be automated.
3. After two days at 500°C turn off the furnace and let it cool. Remove and weigh the beakers. Add sufficient 8M nitric acid to moisten the sample and leave some free acid in the beaker. Evaporate the samples to dryness under a heat lamp. Control any tendency to foam by adding a few drops of octanol. When the sample is dry, rinse the watch glass with a minimum of 8M nitric acid and dry uncovered.
4. Place the beakers in the furnace and re-ash as described above but with an initial heating rate of 3–4°C·h^{-1}. A mixture of nitric acid and carbon approximates to gunpowder; this is not a threat to personal safety but can lead to ash being thrown out of the beaker with loss of the sample if uncontrolled combustion occurs.
5. After one or two days at 500°C switch off the furnace and allow to cool. Remove and weigh the beakers. Cover the beaker with cling film if the samples are not to be dissolved immediately.
6. Add sufficient 8M nitric acid to cover the ash, cover the beaker with a watch glass and boil gently for 20 min. If the ash in more than one beaker is to be combined do so at this point. If the sample is not completely dissolved, add acid until no further dissolution takes place. *If the sample is completely dissolved, proceed as indicated at step 12.*
7. Cool the sample to room temperature. Filter off any undissolved sample on a Whatman GFB fibreglass filter in a Buchner funnel. Use a filter size appropriate

to the volume of solution. Wash the beaker with 8M nitric acid, transferring any loose residues to the filter. Wash the filter with fresh 8M nitric acid. The residues may also be separated by centrifugation. Save the filtrate.

8. Carefully remove the filter along with the residue and transfer it to a PTFE beaker. Rinse the original beaker with a 5–10% hydrofluoric acid in nitric acid mixture. Transfer the wash solution to the PTFE beaker. There should not be prolonged contact of the hydrofluoric acid with the glass. Rinse the beaker(s) with nitric acid, adding the washings to the PTFE beaker. Discard the beaker(s) after this treatment.
9. Evaporate the solution in the PTFE beaker to dryness. Treat the residues by evaporating them to dryness three or four times with a minimum of a 3:1 hydrofluoric and nitric acid mixture. Expel the fluoride by evaporating the sample to dryness three or four times with a minimum of nitric acid.
10. Add sufficient 8M nitric acid to cover the residue, cover with a watch glass and boil gently for a few minutes. Cool to room temperature. It is difficult to see undissolved residues in the PTFE beaker. For this reason, transfer the solution to a centrifuge tube and centrifuge it even if it appears to be clear. Combine the supernatant with the filtrate from step 7. If any residues are present transfer them back to the PTFE beaker and repeat the treatment described in step 9.
11. Add 1 mL of 0.5 $g \cdot mL^{-1}$ aluminium nitrate solution to the combined sample.
12. If the sample is to be subdivided, the solution should be stored in a weighed plastic bottle and the required portion weighed out. If yield tracers have not already been added they should be weighed in to the analysis sample at this stage.
13. For plutonium or americium analysis, the solution must be low in phosphate. Add sufficient 8M nitric acid to make the ash concentration less than 2 g per 100 mL of solution.
14. Continue the analysis as described below for the determination of plutonium. Even if only americium is to be determined, the initial steps of the plutonium separation must be followed as its presence could interfere with the analysis of the americium and curium.

Determination of plutonium

This method is generally applicable for a solution of plutonium in 8M nitric acid arising from either urine or faecal samples. It can also be applied to 8M nitric acid solutions arising from virtually any matrix. The plutonium is separated from the 8M nitric acid phase by adsorption on anion exchange resin. Further purification is achieved by anion exchange from a concentrated hydrochloric acid phase. The separated plutonium is then electroplated on to a 25 mm stainless steel disc for analysis by α spectrometry.

1. The starting material is in solution in 8M nitric acid containing ^{242}Pu or ^{236}Pu tracer, and americium and curium tracers if necessary. The solution should be free from fluoride. As a safeguard against fluoride interference, add 1 mL of aluminium nitrate solution, 0.5 $g \cdot mL^{-1}$ if fluoride has been used in the dissolution procedure.
2. The solution must be low in phosphate, equivalent to less than 2 g of calcium phosphate per 100 mL of solution. The solution must also be low in chloride concentration.
3. Cool the solution to room temperature and add approximately 100 mg of sodium nitrite per 300 mL of solution. Dissolve the solid by gently swirling the contents of the beaker. This is an appropriate point in the method to leave the sample overnight if the solutions are being prepared one day and the columns run the next. Otherwise the sample should be allowed to stand for at least 15 min before proceeding.
4. Replace the beaker on the hotplate and boil the solution gently until no further brown fumes are evolved. Cool the solution to room temperature.
5. Put sufficient aqueous slurry of anion exchange resin (notes A and B at the end of this Annex) into a 1 cm diameter column to leave an 8 cm high column of settled resin. The resin volume is approximately 8 mL. Allow the liquid to drain. Surface tension prevents the resin from draining completely and so prevents cavitation. Wash the resin with three or four column volumes of concentrated hydrochloric acid and then condition it with three or four column volumes of 8M nitric acid. The resin is slowly degraded by nitric acid so the column should be prepared immediately before use.
6. Transfer the sample solution to the top of the column and allow it to drain through the resin. Wash the beaker with several small volumes of 8M nitric acid, passing the washes sequentially through the column. Then wash the column repeatedly with small portions of 8M nitric acid, until three or four column volumes have been used. The plutonium is absorbed on to the column while calcium, iron, phosphate and any strontium, americium and curium pass through. Collect the effluent (raffinate) if americium and/or curium are to be determined, otherwise discard it.
7. Wash the column repeatedly with small portions of concentrated hydrochloric acid until the equivalent of five column volumes has been used. Discard the eluate, which contains any thorium present.
8. Elute the plutonium into a clean beaker using small portions of concentrated hydrochloric acid containing approximately 2% by volume hydriodic acid (note C). Collect about 40 mL of eluate.
9. Evaporate the eluate to dryness on a low temperature hotplate or under a heat lamp, to prevent spraying. Remove iodide ions by evaporating the solution to dryness two or three times with several drops of concentrated hydrochloric

acid. Expel chloride and re-oxidize the plutonium to the tetravalent state by evaporating the solution to dryness two or three times with several drops of concentrated nitric acid. Use the acids to wash down the sides of the beaker.

10. Fill a 0.7 cm diameter disposable column (note D) with an aqueous slurry of anion exchange resin. The top of the settled resin bed should be just below the waist of the column. If necessary adjust the resin height as with the first column. The column volume is approximately 3 mL. Condition the resin with three or four column volumes of concentrated hydrochloric acid.
11. Convert the sample to the chloride by evaporating it to dryness twice with concentrated hydrochloric acid. Dissolve the sample in a minimum of concentrated hydrochloric acid and then transfer it to the top of the column. Wash the beaker with a few small portions of concentrated hydrochloric acid, passing the washes through the column, and continue to wash the column until five or six column volumes have been used.
12. Elute the plutonium from the column with small portions of concentrated hydrochloric acid containing 2% by volume of hydriodic acid (note C), continue until about 15–20 mL have been eluted.
13. Evaporate the eluate to dryness and eliminate halides as described in step 9.

Electrodeposition

Two methods of electrodeposition are available. Occasionally very fine beads of resin pass into the column eluate. In that case use method Pu 1 (steps 14 and 15), otherwise methods Pu 1 or Pu 2 (step 16) may be used.

14. *Pu 1 method:* Add a few drops of concentrated sulphuric acid to the sample and heat until white fumes are evolved. Cool to room temperature. If the acid is clear with no black grains in it proceed to step 15. A black coloration suggests the presence of ion exchange resin. Destroy the resin by evaporating to dryness and then heating the beaker to red heat in a Bunsen flame. After cooling, add more sulphuric acid and re-heat to fuming.
15. Cool. Add a few millilitres of water and one drop of methyl red indicator solution (note E). Add concentrated ammonia solution until the colour is yellow, and add 1M sulphuric acid dropwise until the colour just turns orange/red. Add one or two drops in excess. (Alternatively, adjust the pH with ammonia solution to 2–2.3 using a pH meter.) Transfer the solution with water to an assembled plating cell (note G and Fig. VII–1) and adjust the pH to 2–3 with ammonia solution or dilute sulphuric acid. Go to step 17.
16. *Pu 2 method:* Add a few millilitres of sulphate plating solution (note F) to the sample, warm the solution. Transfer the solution to an assembled plating cell (note G) with sulphate plating solution.

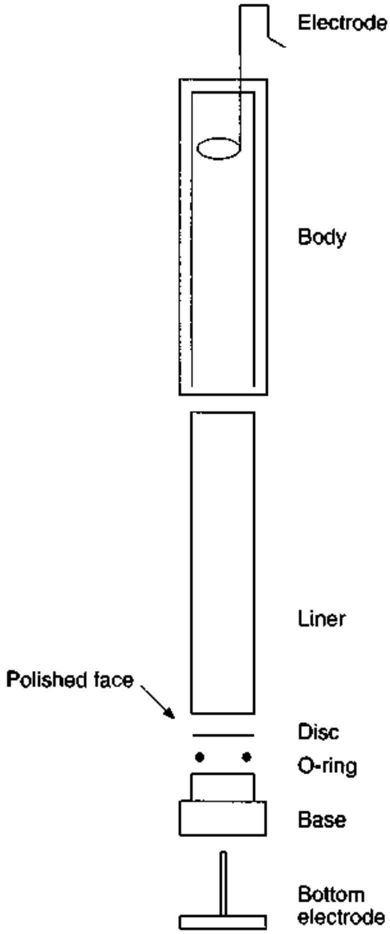

FIG. VII–1. Illustration of an electroplating cell.

17. Electroplate the plutonium at 500 mA, using the stainless steel disc (note I) as the cathode and a platinum wire anode. After 2.5 h, add about 1 mL of 0.880 ammonia solution and continue electroplating for about 1 min. Drain and quickly disassemble the plating cell. Wash the disc with water and then methanol.
18. Flame the stainless steel disc to a dull red heat in a Bunsen flame. This converts plutonium hydroxide to oxide and can also remove any polonium which may have carried through the method.
19. Count the disc in an α spectrometer until the uncertainty (sometimes called 'error') on the calculated result(s) is acceptable.
20. Calculate the result for the unknown isotope(s). Errors should be calculated at the 95% confidence level. If the result of the 'blank' sample is significant with

respect to the sample result, then the 'blank' value must be deducted. Where a 'blank' deduction has been made, this fact, and the 'blank' value, must be stated in the final report.

Determination of americium and curium

This method is generally applicable for a solution of americium or curium arising from samples of urine or faeces or virtually any other matrix. The starting material is the raffinate from the first anion exchange column used in the determination of plutonium (step 6). The removal of plutonium by anion exchange must be carried out even if the analysis of plutonium is not required. The americium and curium are separated from a 12M nitric acid phase by solvent extraction with dibutyl-N, N-diethylcarbamylphosphonate (DDCP). Further purification is achieved by adsorption on to Bio-Rad AG-MP1 macroporous anion exchange resin from a methanolic nitric acid phase. The separated americium and curium are then electroplated on to a 25.4 mm stainless steel disc for analysis by α spectrometry.

1. The starting material is the raffinate from an anion exchange column containing ^{243}Am and/or ^{244}Cm tracers (note J). Evaporate this solution to dryness and if the ash weight is unknown weigh the residue. The dried weight is approximately double the equivalent ash weight. Large samples should only be taken to near dryness, if baked on a hotplate the solids are difficult to redissolve.
2. Evaporate the sample to dryness (or near dryness) twice with concentrated hydrochloric acid and then twice with water. Dissolve the sample in a minimum of 1.5M hydrochloric acid. The size of column used for removal of any polonium depends on the volume of acid used. For less than 50 mL of solution use a 0.7 cm diameter column, for more use a 1 cm diameter column.
3. Fill a 0.7 cm diameter disposable column (note D) or a 1 cm diameter column (note B) with an aqueous suspension of anion exchange resin (note K). The settled resin bed should be 5.5 cm long for the small column and 8 cm for the large, the column volumes being approximately 3 and 8 mL, respectively. Surface tension will prevent the resin draining completely and so prevents cavitation. Condition the column with three to four column volumes of 1.5M hydrochloric acid.
4. Transfer the sample solution to the top of the column. Wash the beaker a few times with 1.5M hydrochloric acid and add to the column. Wash the column with small portions of 1.5M hydrochloric acid until the equivalent of five column volumes has been used. Collect the effluent from the column which contains the americium and curium.
5. Evaporate the sample to dryness (or near dryness). Expel chloride by evaporating the sample two or three times with a few millilitres of nitric acid. Use the acid to wash down the sides of the beaker.

6. Dissolve the residue in 12M nitric acid according to its ash weight:
   - For up to 2.5 g ash use 30 mL of 12M nitric acid.
   - For 2.5–5.0 g ash use 80 mL of 12M nitric acid.
   - For 5.0–7.5 g ash use 130 mL of 12M nitric acid.

   Use scaled proportions for larger samples.
7. Add approximately 100 mg of sodium nitrite to the solution. Cover the beaker with a watch glass and dissolve the solid by gently swirling the beaker. Heat the solution for approximately 15 min at 80–90°C. Cool the solution to room temperature.
8. Add the solution to one or more 250 mL separating funnels. Wash the beaker with two 10 mL portions of 12M nitric acid, adding these washes to the separating funnel.
9. Add 1 mL of DDCP per 50 mL of 12M nitric acid used. Shake the funnel for approximately 30 s. Allow the organic phase to separate; this will require a minimum of 3 h, preferably overnight.
10. Drain the lower aqueous layer into a beaker. (This solution can be retained if strontium is to be determined, otherwise discard it.) Wash the DDCP by adding 10 mL of 12M nitric acid per millilitre of DDCP and shaking for a few seconds. Allow the phases to separate for a few hours. Drain the lower aqueous phase and discard it.
11. Add to the separating funnel 5 mL of toluene and 20 mL of 2M nitric acid per millilitre of DDCP used. Shake the funnel for approximately 30 s. Allow the phases to separate. Drain and collect the lower aqueous phase, which contains the americium and curium.
12. Add to the separating funnel a further 20 mL of 2M nitric acid per millilitre of DDCP used. Shake the funnel for approximately 30 s and allow the phases to separate. Collect the lower aqueous layer, combining it with the solution from the first back-extraction. Discard the organic phase.
13. Evaporate the solution on a hotplate set at about 130°C or under a heat lamp. It is important not to char the organic residues left after the aqueous solution has evaporated.
14. Fill a 0.7 cm diameter disposable column (note D) with an aqueous suspension of Bio-Rad AG-MP1, 100–200 mesh (note K). The top of the settled resin bed should be just below the waist of the column. Surface tension will prevent the resin draining completely and so prevents cavitation.
15. Condition the column with 30 mL of reagent A (see **Reagents**). Resuspend the resin in this reagent after conditioning to eliminate bubbles and improve the flow rate.
16. Dissolve the residue from the DDCP extraction in a small amount of reagent A and add to the column. Wash the beaker with a few small portions of reagent A

and add to the column. Wash the column with small portions of reagent A until 30 mL have passed through it.

17. Wash the column with small portions of reagent B (see **Reagents**) until 15 mL have been used.
18. Add reagent C (see **Reagents**) to the top of the column, a few drops at a time until the pink coloration has started to move down the column. Wash the column with further small portions of reagent C until the pink coloration elutes (15–20 mL).
19. Elute americium and curium with small portions of reagent D (see **Reagents**), continue until 20 mL has been collected. Evaporate the eluate to dryness using gentle heat.

Methanol–nitric acid mixtures must be treated with care

20. Electrodeposit the sample as described in steps 14–18 of the method for plutonium.
21. Count the disc in an α spectrometer until the errors on the calculated result(s) are acceptable.
22. Calculate the result for the unknown isotope(s). Errors should be calculated at the 95% confidence level. If the result of the 'blank' sample is significant with respect to the sample result, then the 'blank' value must be deducted. Where a 'blank' deduction has been made, this fact, and the 'blank' value, must be stated in the report.

Notes

A. Dowex or Bio-Rad 1-X8 (50–100 or 100–200 mesh) resins may be used. The 50–100 mesh 1-X8 resin has a higher flow rate and therefore is particularly suitable for larger sample volumes. Before use the resins should be prepared by flotation. Half fill a screw cap 1 L bottle with dry resin, fill with water and shake to suspend the resin. Wait for the bulk of the resin to settle then decant off the fines. Refill with water and repeat until the resin settles to give a clear supernatant. Store in dilute (0.1M) hydrochloric acid.

B. 1 cm columns are made from 1.5 m lengths of 1 cm diameter soda glass tubing. Each is cut in half and then each piece has a waist drawn in it at its midpoint. This is cut to give four columns about 37 cm long with an approximately 3 mm jet drawn in one end. All cut edges should be flame polished and then the jet plugged with glass wool.

C. The hydriodic acid used should be from either a freshly opened vial or a bottle of analytical reagent grade that has been opened for less than three months. Always date a new bottle when opening for the first time.

D. Disposable polystyrene columns are 5.5 cm long by 0.7 cm diameter. They have a reservoir and a 70 µm filter. They are available from Bioconnections Ltd, Leeds, UK.

E. The methyl red indicator is prepared by dissolving 0.1 g of the free acid in 60 µL of methanol and diluted to 100 mL with water.

F. Sulphate plating solution is made by *carefully* adding 60 mL of concentrated sulphuric acid to 700 mL of water. Adjust the pH to 2.3 with ammonia solution, cooling will be needed. Make sure the solution is at room temperature before the final pH adjustment. Dilute to 1 L. The pH of this solution drifts and should be readjusted using a pH meter immediately before use.

G. The electroplating cell (Fig. VII-1) should be soaked in a detergent solution when not in use and rinsed well with tap water before use. Dry it and insert a clean new liner (note H). Wash the polished surface of a new 25 mm stainless steel disc with acetone to remove grease, then label the back of the disc with a scratched label and a felt tip marker. Place the disc in the cell with the polished surface facing inwards, back the disc with an O ring and screw on the base. Fill the cell with distilled water and place on a dry tissue to check for leaks. Discard the water before use.

H. Liners for the plating cell are moulded in polythene. Moss Plastics, Kidlington, UK, is a supplier.

I. The 25 mm stainless steel discs used for plating the plutonium and americium are 0.5 mm thick.

J. This method is applicable to both americium and curium but a tracer of the same element must be used. As this method can, with slight modification to the AG-MP1 column, separate americium from curium, it is possible that the yields of the americium and curium are not equal. Therefore it is not acceptable to use non-isotopic tracers.

K. The anion exchange resins are the same as those used for plutonium determination. The Bio-Rad AG-MP1 (100–200 mesh) is also prepared by flotation. Half fill a screw cap 1 L bottle with dry resin, fill with water and shake to suspend the resin. Wait for the bulk of the resin to settle then decant off the fines. Refill with water and repeat until the resin settles to give a clear supernatant. Store in dilute (~1M) hydrochloric acid.

Calculation of results

Refer to Section 6.1 of the main text for a calculation of results.

Annex VIII

ANALYSIS FOR TRITIUM AND ^{14}C BY COMBUSTION TECHNIQUES

Based on procedures used at the National Radiological Protection Board, UK

INTRODUCTION

Biological or physical samples containing the low energy β emitters tritium (E_{max} = 18.6 keV) and ^{14}C (E_{max} = 156 keV), in forms that are not readily soluble in hydrophilic solvents, must be isolated from their matrices to assure accurate and sensitive determination. The oxidation method of sample preparation allows the counting of solid or insoluble samples, provided they can be completely oxidized to tritiated water (HTO) and CO_2. These samples may include carbon in organic materials or as an elemental particulate, and tritium in organic bound forms (OBT). Following oxidation these products can be separated by selective absorption, and introduced into suitable LSC cocktails for liquid scintillation counting, as in Annex II. Such isolation also improves the analysis of other samples for which direct LSC would also be possible, as it eliminates colour and chemical quenching, and reduces the background count.

A variety of home built oxidation and collection systems have been constructed in various laboratories. In addition commercial systems, such as the Packard Model 307 Sample Oxidizer used at the NRPB, are available. All can provide a satisfactory analysis, provided the oxidation is complete and the HTO and CO_2 products are quantitatively collected.

Apparatus

Sample combustion chamber
H_2O condenser or cold trap
CO_2 trapping medium or cold trap
Liquid scintillation counter.

Reagents

Carbo-sorb, or other CO_2 absorbing material (e.g. ethanolamine)
LSC counting fluid.

Procedure

1. The sample should be characterized as to weight and the presence of other radionuclides, particularly those which may be oxidized.
2. The combustion system consists of a combustion flask, an ignition basket and continuous oxygen flow.
3. The sample is placed in a removable ignition basket, and a high electrical current is passed through the basket coil while the oxygen begins to flow, so that the sample ignites.
4. The combustion products exit to the collection systems through tubing that is pre-heated to prevent condensation.
5. Oxidized tritium (HTO) is collected on passage through an exchange column containing water.
6. Oxidized carbon (CO_2) is not trapped in the water trapping column, but is then collected by passage though a second column containing the CO_2 absorber.
7. After oxidation of the sample is complete, the isolation media are removed, as is the ignition basket containing any inert residue.
8. The HTO is introduced into an LSC cocktail in the usual way (Annex II), and the concentration of tritium determined. This concentration must then be related to the original sample through pre-determined yield factors.
9. The CO_2 is similarly quantified by LSC counting, and the activity in the original sample determined (close-up).

Annex IX

DETERMINATION OF CREATININE IN URINE SAMPLES

Based on information from the Japan Atomic Energy Research Institute, Japan

IX–I. INTRODUCTION

Creatinine in urine is determined using the classical picric acid method for creatinine by Jaffe's reaction. Creatinine in the urine sample is measured by the absorbance of photons in a spectrophotometer. The absorption is compared with a blank and standard solutions of creatine measured in the same way.

Apparatus

Water bath
Spectrophotometer
Beaker, volumetric flask, pipette, filter paper.

Reagents

Picric acid
Creatinine (reagent grade)
HCl (0.1M)
NaOH (1M).

Procedure

1. *Preparation of picric acid solution (0.04M)*

1.1. Dissolve 12.5 g of picric acid (Guaranteed Reagent) in 1 L of water.
1.2. Filter the solution through filter paper into a 1 L beaker.
1.3. Transfer 750 mL of the filtrate into the 1 L beaker and add 250 mL of water.

2. *Preparation of creatinine standard solution*

2.1. Stock solution 'C1' (1 mg/mL): dissolve 100 mg of creatinine (Guaranteed Reagent) in about 20 mL of 0.1M HCl solution, transfer to a 100 mL Pyrex volumetric flask and complete the volume with 0.1M HCl solution.
2.2. Standard solution 'S1' (2 μg/mL): dilute C1 as appropriate.
2.3. Standard solution 'S2' (5 μg/mL): dilute C1 as appropriate.

2.4. Standard solution 'S3' (10 μg/mL): dilute C1 as appropriate.
2.5. Standard solution 'S4' (15 μg/mL): dilute C1 as appropriate.

3. *Preparation of dilute urine sample solution*

3.1. Dilute 1 mL of urine sample to 100 mL in a volumetric flask using distilled water.

4. *Preparation of samples for measurement*

4.1. Pipette 5 mL of urine sample solution (diluted urine and each of the standards) into a 15 mL test tube.
4.2. Add 2 mL of picric acid solution.
4.3. Add 0.5 mL of 1M NaOH solution and mix well.
4.4. Warm in a water bath at 20°C for 40 min.

5. *Measurement*

5.1. Measure the absorbance at a wavelength of 517 nm with a spectrophotometer for each of the standards and the diluted urine solution.
5.2. Draw the calibration curve between concentration of creatinine and the absorbance based on the measurement for the creatinine standards.
5.3. Estimate the creatinine concentration in the urine sample from the calibration curve.
5.4. Calculate creatinine in the sample as a proportion of 24 h excretion, assuming 1.7 g of creatinine excreted in urine [IX–1]. Individual creatinine excretion rates can also be estimated [IX–2].
5.5. Calculate the 24 h excretion of radionuclides based on the amount of activity and creatinine in the urine sample.

REFERENCES TO ANNEX IX

[IX–1] JACKSON, S., Creatinine in urine as an index of urinary excretion rate, Health Phys. **12** (1966) 843.

[IX–2] BOENIGER, M.F., LOWRY, L.K., ROSENBERG, J., Interpretation of urine results used to assess chemical exposures with emphasis on creatinine: a review, Am. Ind. Hyg. Assoc. J. **54** (1994) 615.

GLOSSARY

AMAD. Activity median aerodynamic diameter. Fifty per cent of the activity in the aerosol is associated with particles of aerodynamic diameter (d_{ae}) greater than the AMAD. Used when deposition depends principally on inertial impaction and sedimentation, typically when the AMAD is greater than about 0.5 μm.

bioassay. Describes any procedure used to determine the kind, quantity, location and/or retention of radionuclides in the body by direct (in vivo) measurements or by in vitro analysis of material excreted or removed from the body.

biokinetic model. A mathematical model describing the intake, uptake and retention of a radionuclide in various organs or tissues of the body and the subsequent excretion from the body by various pathways.

deposition. The initial processes determining how much of the material in the inhaled air remains in the respiratory tract after exhalation. Deposition of material may occur during both inhalation and exhalation.

Derived Recording Level. The concentration of a radionuclide in a sample that corresponds, through assumptions about the time and pattern of intake, and the use of biokinetic models, to an intake at the recording level.

dose coefficient. The committed equivalent dose to tissue per unit intake at age t_0, $h_T(\tau)$, or committed effective dose per unit intake, $e(\tau)$, where τ is the time period in years over which the dose is calculated, i.e. 50 years for adults and $70 - t_0$ years for children. The unit of $h_T(\tau)$ and $e(\tau)$ is Sv/Bq.

fractional absorption in the gastrointestinal tract (f_1). The f_1 value is the fraction of an element absorbed from the gut to body fluids.

investigation level. The value of a quantity such as effective dose, intake or contamination per unit area or volume at or above which an investigation should be conducted.

Minimum Detectable Activity (MDA). That amount of radioactivity in a sample which produces a counting rate that will be detected (i.e. considered to be above background) with a certain level of confidence. In a sample containing activity equal to the MDA, random fluctuations will produce a counting rate less than the decision level with a certain probability β (normally taken to be 5%), thereby resulting in a false negative (type II error). This amount is also referred to as the Detection Limit or Lower Limit of Detection and the counting rate at this amount is also referred to as the Determination Level. A sample containing exactly the MDA will be taken to be free of activity only 5% of the time.

Minimum Significant Activity (MSA). That amount of radioactivity in a sample which produces a counting rate that may be reliably distinguished from background with a certain level of confidence. Random fluctuations in measurements of a suitable blank (including all sources of variation) will produce a counting rate at or above this level with a certain probability α (normally taken to be 5%). Thus, observation of a counting rate equal to that at the MSA level results in a false positive (type I error) in only 5% of the cases. This amount is also referred to as the Decision Limit and the counting rate at this amount is also referred to as the Critical Level. A sample containing exactly the MSA will be taken to be free of activity 50% of the time (i.e. the counting rate will be less than that corresponding to the decision level), but a true background sample will be taken to contain activity only 5% of the time.

physical half-life (T_p). The time taken for the activity of a radionuclide to decrease by 50% due to radioactive decay.

recording level. A level of dose, exposure or intake specified by the regulatory authority at or above which values of dose, exposure or intake received by workers are to be entered in their individual exposure records.

routine monitoring. A form of workplace or individual monitoring conducted on a fixed schedule for selected workers.

special monitoring. A form of monitoring prompted by an actual or suspected abnormal condition, usually being more detailed and comprehensive than routine monitoring.

types of materials. Categories of materials in the lung according to their rates of absorption from the respiratory tract to body fluids:

type F	Deposited materials that are readily absorbed into body fluids from the respiratory tract. (Fast rate of absorption.)
type M	Deposited materials that have intermediate rates of absorption into body fluids from the respiratory tract. (Moderate rate of absorption.)
type S	Deposited materials that are relatively insoluble in the respiratory tract. (Slow rate of absorption.)
type V	Deposited materials that are assumed, for dosimetric purposes, to be instantaneously absorbed into body fluids from the respiratory tract — applied only to certain gases and vapours. (Very rapid absorption.)
SR-1	Soluble or reactive gases and vapours — deposition may occur throughout the respiratory tract.
SR-2	Highly soluble gases and vapours — complete deposition assumed to occur in the extrathoracic airways.

uptake. The processes by which radionuclides enter the body fluids from the respiratory tract, gastrointestinal tract or through the skin, or the fraction of an intake that enters the body fluids by these processes.

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