

OMINEX: Development of Guidance on Monitoring for Internal Exposure

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Abstract. The aim of the OMINEX project was to provide advice and guidance on the design and implementation of internal dose monitoring programmes in the workplace in such a way that best use is made of available resources, while minimising costs. Topics addressed include choice of monitoring method(s), (eg excretion monitoring vs. *in vivo* monitoring), choice of measurement technique (eg alpha spectrometry vs. mass spectrometry), monitoring intervals, measurement frequency, required measurement sensitivity and accuracy, measurement parameters needed to achieve this performance, the resulting uncertainty in assessed intakes and doses, and minimum detectable doses. The underlying approach to optimisation was to consider costs versus “benefits”, the latter being quantified primarily by assessing the sensitivity or accuracy with which intakes and doses are determined from the results of particular monitoring methods. The aim of this paper is to present an overview of the results of the project. Some of the main results of surveys of current internal dose monitoring practice and the costs of monitoring programmes are presented. Recommendations on the optimisation of bioassay and *in vivo* measurement parameters are discussed. A novel method for the assessment of uncertainty in assessed intakes and doses is described, and the use of information on uncertainties in designing a monitoring programme is discussed using the example of tritium-in-urine monitoring. Recommendations are described for the monitoring of exposures to compounds of uranium, plutonium, thorium and caesium encountered in the nuclear industries.

1. Introduction

OMINEX (Optimisation of Monitoring for Internal Exposure) was an international project part-funded by the European Commission within the 5th (EURATOM) Framework Programme. The primary aim of the project was to provide advice and guidance on the design and implementation of internal dose monitoring programmes in the workplace. The target audience includes dosimetry service managers, regulators and senior medical staff in the nuclear industry. To ensure that the results of the project would be practicable and relevant to the needs of industry, OMINEX was conceived as a collaboration between research/advisory organisations (IRSN, NRPB, SCK-CEN, STUK) and nuclear industry organisations (EdF, TVONS, CEA). The project was carried out over a 3-year period, and work was completed in December 2003. Results were presented at a training course held in Paris in November 2003. This paper gives an overview, and presents some of the main results.

The primary aims of internal dose monitoring are: (a) to verify and document that workers are protected adequately against risks from radionuclide intakes; and (b) to verify and document that the protection complies with legal requirements [1]. The first aim is achieved by utilizing monitoring programmes that allow the assessment of internal doses with sufficient *accuracy* so that an acceptable estimate of risk can be made. This requires a consideration of the uncertainties in assessed intakes and doses that arise from different monitoring programme designs. For example, in ICRP Publication 78 [2], routine monitoring intervals are selected so that any underestimation in intake introduced by the

unknown time of intake is no more than a factor of three. The second aim is achieved by ensuring that doses can be assessed with adequate *sensitivity*, so that workers who have received doses above a particular level are reliably identified. An example of such a requirement is the need to demonstrate that annual doses are not in excess of 6 mSv for Category B workers, in order to confirm that they have been categorised correctly as required by Article 25 of EC Directive 96/29/EURATOM [3]. Since the results of monitoring need to be compared with dose limits, dose investigation levels or dose recording levels, sensitivity needs to be quantified in terms of the minimum detectable dose that a particular monitoring programme can achieve.

In the OMINEX project, the underlying approach to optimisation was to consider costs versus “benefits”, with the latter being quantified primarily by assessing the *accuracy* or *sensitivity* with which intakes and doses are determined from the results of particular monitoring methods. This type of approach is particularly appropriate for internal dose monitoring, where the economic costs of monitoring and control of internal exposures in the workplace are usually significantly greater than the equivalent costs for external exposures.

Advice has been developed on internal dose monitoring following exposure to a range of radionuclides and compounds that are of the most radiological interest, and that represent some of the most difficult problems in internal dose assessment. Topics on which advice is provided include choice of monitoring method(s), (eg urine or faeces (bioassay) monitoring, whole body or lung (*in vivo*) monitoring), choice of measurement technique (eg alpha spectrometry vs. mass spectrometry), monitoring intervals, measurement frequency, required measurement sensitivity and accuracy, measurement parameters needed to achieve this performance (detection efficiency, count times, etc.), resulting uncertainty in assessed intakes and doses, and minimum detectable doses. Advice on choice of monitoring methods has been limited to bioassay and *in vivo* monitoring; the use of personal air sampling or static air sampling has not been explicitly considered, although there is potential to extend the methods developed in OMINEX to these monitoring methods.

Work was organised into five distinct but inter-related “Work Packages”. It was necessary to have a reasonably comprehensive description of current internal dose monitoring practice in European Union (EU) countries together with information on associated costs of monitoring before providing advice on best practice. However, there has been little sharing of such information between different countries across the EU. Surveys of these topics were therefore carried out in the first two Work Packages. In the 3rd Work Package, surveys were carried out of bioassay and *in vivo* monitoring procedures and of measurement parameters affecting uncertainties in these measurements. This information was then used as an input to investigations into ways of exploiting available methods and techniques to reduce uncertainties in measurements. In the 4th Work Package, the major sources of uncertainty in internal doses assessed from the results of particular monitoring methods and measurement techniques were considered. A methodology was then developed to assess total uncertainty in assessed intakes and doses taking into account uncertainties in intake patterns, measurements, and respiratory tract and systemic model parameters.

Advice on monitoring for selected radionuclides/compounds was developed in the 5th Work Package. This made use of the results of the other Work Packages, particularly in respect of the assessment and optimisation of uncertainties in measured bioassay/*in vivo* quantities and assessed doses. In addition, previously-developed methods for the assessment of minimum detectable doses resulting from the use of different monitoring methods, monitoring intervals, etc. were utilized. These approaches are particularly important when considering monitoring for actinide exposures, where achieving adequate sensitivity is a significant issue.

2. Results and Discussion

There is space here to present only an overview of some of the main findings and results of the project. For a full account, reference should be made to published reports and papers (see References).

2.1. Surveys of current practice and costs

Responses to the survey on internal dose monitoring programmes were received from organisations in Austria, Belgium, the Czech Republic, France, Germany, Hungary, Italy, Spain, the Nordic countries, Russia and other former states of the USSR, and the UK. Information was collected on general aspects of monitoring such as type of operation, number of workers, monitoring practice and purposes of monitoring; on methods of monitoring of fission and activation products and actinides (uranium, thorium, plutonium, americium and mixed oxide (MOX) fuel); on calibrations and minimum detectable amounts (MDA); on chemical forms and solubility assumptions; on monitoring frequency and investigation levels; and on dose statistics. Tables I – III shows some examples of the information collected from the 25 organisations that responded to the main questionnaire.

Table I. Monitoring frequency used for routine monitoring: β, γ emitting radionuclides

Radionuclide(s)	Monitoring Method	Frequency (year ⁻¹)	For this radionuclide, monitoring method and frequency:	
			Number of organisations	Number of measurements
Fission and activation products (γ -emitters)	Whole body	1	12	6800
		2	7	2200
		4	2	480
		6	1	220
		12	1	1700
		52	1	8000
Iodine-131	Thyroid	1	4	5400
		2	2	-
		4	1	100
		12	4	2400
		26	1	-
		52	1	10
		Other	5	350
Tritium	Urine	1	2	20
		4	1	5
		6	1	200
		12	3	3000
		24	1	-
		52	4	> 3600
Strontium-90 / Yttrium-90	Urine	2	1	>1000
		4	1	100

Routine monitoring is usually carried out to confirm that doses are below a certain level (eg 6 mSv for Category B workers) or to identify unexpected exposures. The times of any potential exposures are usually not known, and so monitoring is carried out at fixed time intervals. Table I presents the data collected on the frequency of routine monitoring measurements for β - and γ -emitting radionuclides. For a particular radionuclide (or class of radionuclide), monitoring method and frequency, the table shows the number of organisations reporting that monitoring according to this specification is carried out, and the corresponding number of measurements performed. As can be seen, there is no clear consensus on monitoring intervals. For whole body monitoring, the largest number of organisations report a 1-year monitoring interval, but several organisations use much shorter intervals, and a significant number of measurements are carried out with a weekly interval. Similarly, for tritium-in-urine monitoring, five organisations use monitoring intervals of two weeks or less while seven organisations use longer monitoring intervals.

Table II shows the information collected on assumptions made about the solubility in the lung of different uranium compounds. In the ICRP Human Respiratory Tract model (HRTM) [4], default Absorption Types (F, M and S, representing fast, moderate and slow absorption) are specified for use

when insufficient material-specific information is available. It can be seen that there is a consensus in the use of default Types for some compounds (U_3O_8 , UO_2 , UF_6), but some organisations make questionable assumptions about other compounds (eg Type S assumed for UO_3 and UF_4). Table III shows the information collected on the number of cases with assessed doses in excess of 0.1 mSv, classified in dose bands. The large fraction of cases with doses less than 1 mSv demonstrates the success of internal dose monitoring and exposure control procedures among the responding organisations.

Table II. Absorption Type assumptions for uranium compounds

Compound	Type	No. of organisations
U_3O_8	S	6
UO_2	S	7
UO_3	M	2
	S	1
UF_4	M	2
	S	1
UF_6	F	2
UO_2F_2	F	2
	S	1
Metallic U	S	2

Table III. Internal dose statistics for the year preceding receipt of questionnaire

Dose range (mSv)	0.1 – 0.5	0.5 – 1	1 – 2	2 – 5	5 – 10	10 – 20	> 20
No. of cases	2017	216	105	17	6	0	0

Responses to the survey on costs were obtained from organisations in Austria, Belgium, Brazil, the Czech Republic, Finland, France, Germany, Greece, Rumania, Spain, and the UK. Table IV shows the information collected on bioassay and *in vivo* actinide measurements.

Table IV. Fraction of the total number of organisations reporting costs of measurements of actinides in urine and faeces, and of actinide-in-lung measurements, within the stated bands

Costs (euro)	No. of organisations			No. of organisations	
	Urine	Faeces		Costs (euro)	Lung
<300	55%	38%		<100	0%
300 – 400	33%	38%		100 – 200	75%
400 – 600	22%	25%		200 – 300	25%
>600	0%	0%		>300	0%

This data shows that the unit cost of a lung measurement is significantly less than that for an excretion measurement, and also that the costs of urine and faecal measurements are similar. While other factors such as dose sensitivity must also be taken into account, information on costs such as that shown in Table IV provides a valuable input to decisions on choice of monitoring methods.

2.2. Uncertainties in bioassay quantities and optimisation of measurement parameters

The survey of bioassay measurement procedures mainly considered the measurement of actinides in urine and faeces samples. Responses were received from 17 laboratories that carry out alpha spectrometric measurements, and from five laboratories that carry out inductively-coupled plasma mass spectrometric (ICP-MS) measurements.

The most important parameters affecting overall uncertainty and MDAs of alpha-spectrometric bioassay measurements are sample volume, tracer activity, chemical yield, counting efficiency, counting time, background count rate, and background count time. An example of the information collected is shown in figure 1. For α -spectrometric analyses, known amounts of tracers such as ^{242}Pu ,

^{243}Am , ^{232}U and ^{229}Th are added to the sample before chemical purification so that the chemical recovery of the measured radionuclide can be determined. Ideally the tracer activity should be of the order of the activity of the radionuclide to be measured in the sample. Figure 1 shows the spread of tracer activities used by the surveyed laboratories.

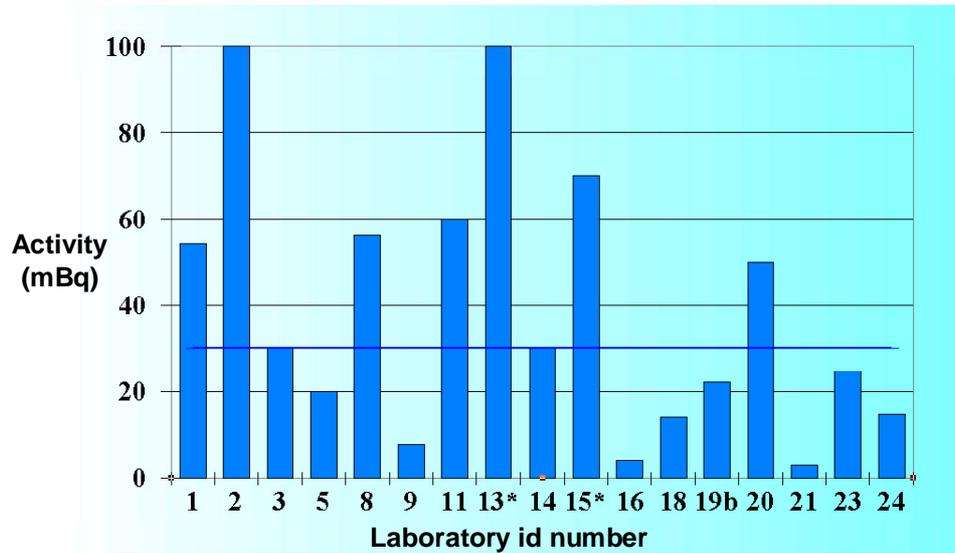


FIG. 1. Activity of ^{242}Pu tracer used in plutonium-in-urine analyses by the surveyed laboratories

Table V compares recommended values for the measurement parameters with averages of the values reported by the surveyed laboratories. The resulting improvement in predicted performance is shown by the values for measurement uncertainty and MDA; as can be seen, an improvement in MDA of a factor of about 3 could be expected. The results of the survey and the recommendations for optimized measurement parameter values are given in Hurtgen et al [5].

Table V. Recommended measurement parameter values and average values reported by laboratories

Measurement parameter	Laboratory Average	Optimized Values
Volume sample	24h (14 labs) ; 1 litre (4 labs)	24 hour sample
Tracer activity	32 mBq	6 mBq
Counting efficiency	30%	36%
Sample counting time	318000 s	250000 s
Background	6.2×10^{-6} counts s^{-1}	$<5 \times 10^{-6}$ counts s^{-1}
Background counting time	459000 s	500000 – 750000 s
Chemical yield	74%	85%
Uncertainty on 1 mBq/24h	35%	25%
Minimum detectable amount (MDA)	0.3 mBq/24h	0.1 mBq/24h

2.3. Uncertainties in in vivo quantities and optimisation of measurement parameters

The effect on *in vivo* measurement uncertainties and MDAs of choice of detector, detector volume and background level was considered [6]. Figure 2 shows the relationship between detector thickness and the decision level, L_c , at 60 keV (the main γ emission for ^{241}Am) and 20 keV (the approximate energy of X-ray emissions from plutonium). This demonstrates that, for measurements of low energy photon emitters, choice of detector type and thickness can have a significant effect on MDAs. For ^{241}Am measurements, the optimum detector thickness for Ge and Si detectors are 1.2 mm and 13.3 mm respectively.

Inhomogeneity in the spatial distribution of activity in the body can result in significant systematic errors. An example is inhomogeneity in the distribution of inhaled material after deposition in the lungs. Methods have been developed to quantify such errors using Monte-Carlo techniques to simulate the response of a detector to any specified distribution of activity [7]. The method has potential for use in

optimising detector geometries to minimise systematic errors. An example of the use of the method is shown in figure 3, which shows the errors arising if activity is concentrated in the periphery (margins) of the lungs rather than distributed uniformly.

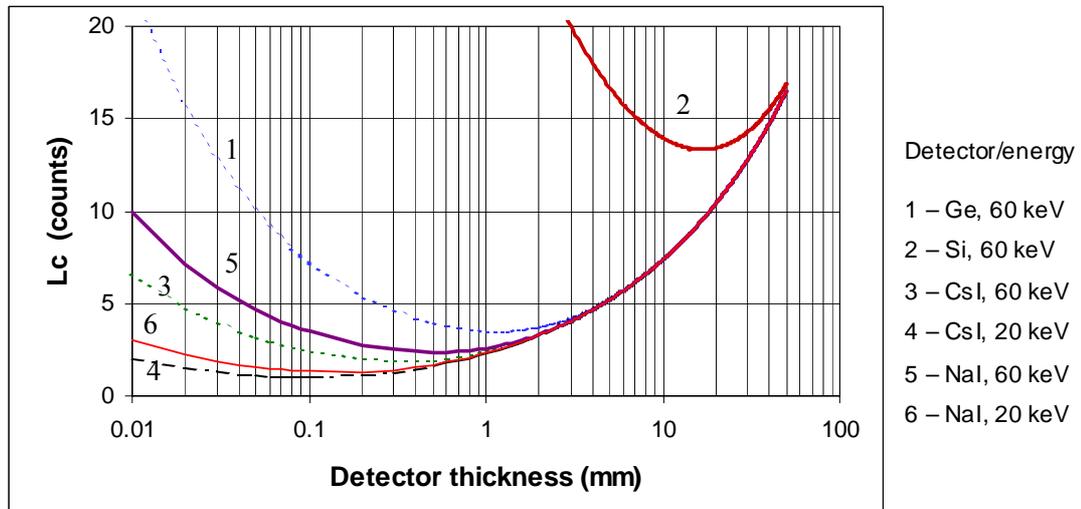


FIG. 2. Dependence of decision level, L_c , on detector thickness

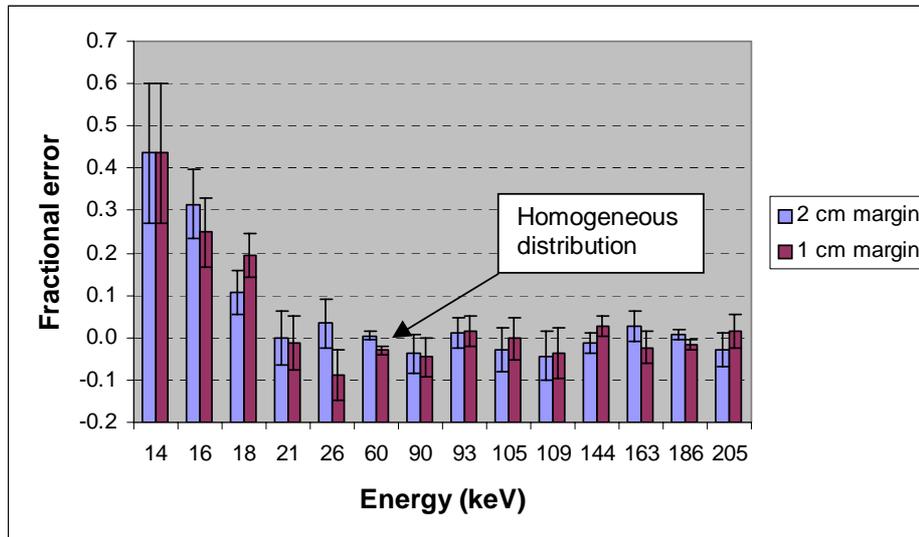


FIG. 3. Effect of distribution in the lungs on assessed activity (simulated $^{241}\text{Am}/^{235}\text{U}$ inhalation)

2.4. Uncertainties in assessed intakes and doses

A novel aspect of the OMINEX project was the development of a methodology for the assessment of total uncertainty in doses assessed from monitoring measurements. This takes account of uncertainties in intake patterns, measurement uncertainties, uncertainties in respiratory tract model parameters such as absorption parameters and particle size, and uncertainties in parameters describing retention of the radionuclide in organs of the body following uptake. An example of the information collected on variability in model parameters is shown in Table VI. This shows the variability in parameter values used in thyroid dose calculations and the resulting variability in the thyroid dose coefficient.

The methodology uses Monte-Carlo simulations to generate probability distribution functions (PDFs) for the assessed dose. The method was first implemented for the simple case of routine tritium-in-urine monitoring. Here, the only model parameter that needs to be considered is the biological half-time, which is usually assigned a nominal value of 10 days, but which is known to vary between 4 and 18

days [8]. Times of acute intakes within the monitoring interval were randomly chosen, and intakes before the monitoring interval were also simulated.

Table VI. Variability in model parameter values used in ^{131}I -in-thyroid dose calculations

	No. of cases	Median	Mean	Range	99 th percentile
Mass (g)	890	16.5	18.3	-	48.3
Uptake fraction	565	0.17	0.19	0.08 – 0.46	-
Biological half-time (d)	47	72	85	21 – 372	-
Dose equivalent / unit intake (Sv/MBq)	-	0.30	0.37	-	1.2

Figure 4 shows the uncertainty in the assessed dose for routine monitoring intervals in the range 7 – 90 days. The use of this information in formulating advice on tritium monitoring is discussed in section 2.5. The method has recently been extended to the more complex case of monitoring for ^{60}Co exposures, and a report on this work is currently in preparation.

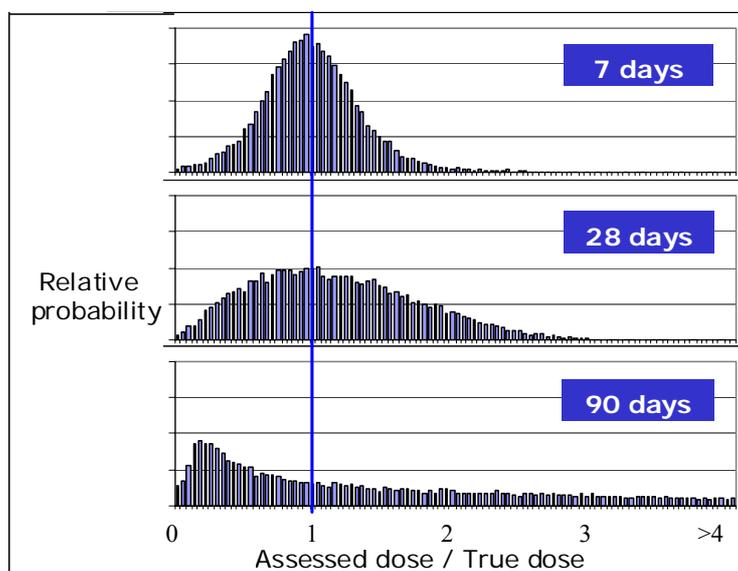


FIG. 4. Probability distribution functions describing the uncertainty in dose assessed from routine tritium-in-urine measurements made at the specified intervals

2.5. Advice on monitoring for specific radionuclides and compounds

Section 2.5.1 summarises advice on routine monitoring for tritium exposures, based on a consideration of uncertainties in the resulting assessed doses (section 2.4). Sections 2.5.2 – 2.5.5 summarise some of the advice on routine and special monitoring for actinide and ^{137}Cs exposures, which is based on considerations of minimum detectable dose. A full account is contained in published reports [9,10,11,12]. The annual dose levels considered are 20 mSv (the dose limit), 6 mSv (the upper dose level for Category B workers) and 1 mSv (the dose threshold defining an exposed worker). The advice for monitoring ^{137}Cs and actinide exposures is based on:

- assessment of dose from measurements at the end of the monitoring interval using realistic values for the MDA, or the reference level (RL) where variation in dietary excretion is important
- consideration in most cases of differences in AMAD¹ over the range 1-10 μm
- use of absorption parameter values (f_r , s_r and s_s) [4] which represent the reported range for the material specified
- known or unknown patterns of intake
- for ^{137}Cs , consideration of the variability in whole body retention half-times, and for uranium the relative importance of radiological and chemical toxicity.

¹ AMAD – Activity Median Aerodynamic Diameter [4]

2.5.1. Routine tritium-in-urine monitoring

PDFs such as those shown in figure 4 can be used to make judgements on factors such as the monitoring interval using a suitable criterion on maximum acceptable error. A reasonable criterion, which is adapted from the one used in ICRP Publication 78 [2], is that underestimates in dose, quantified using the 5% confidence level of the PDF, should be less than a factor of three. From a consideration of uncertainties in intake patterns, biological half-time and tritium-in-urine measurements, this criterion is met if urine sampling is carried out at 28 day intervals. More frequent sampling (cf Table 1) is not necessary.

2.5.2. Special monitoring for ^{137}Cs exposures

Absorption parameter values between default Types F and M, and whole body retention half-times between 50 d and 150 d, have been considered. These differences can affect the assessment of dose by an order of magnitude or more, as shown in figure 5. Whole body monitoring and urine assay are capable of assessing doses of 1 mSv y^{-1} if the MDAs are 100 Bq and 1 Bq d^{-1} respectively.

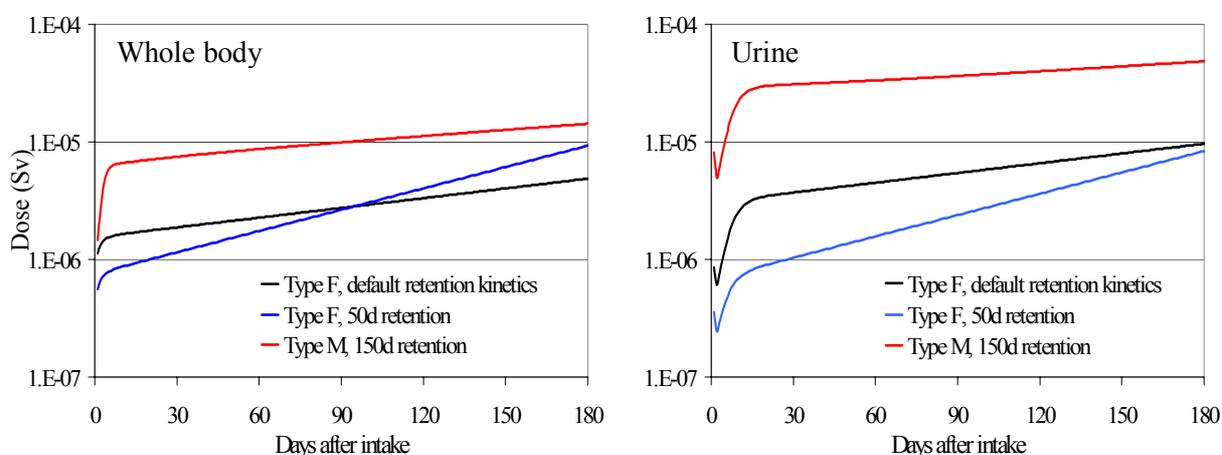


FIG. 5. Minimum detectable doses from acute ^{137}Cs exposures: whole body and urine monitoring

2.5.3. Routine monitoring for uranium

The compounds considered were uranyl nitrate, ammonium diuranate, uranium tributylphosphate, peroxide, trioxide, tetrafluoride, octoxide and dioxide of natural isotopic composition and present in industrial processes in the UK and France. For uranium octoxide and dioxide, intakes should be limited on the basis of dose; with an MDA of less than $3 \mu\text{g d}^{-1}$ in faecal samples, annual doses less than 6 mSv from repeated exposure can be confirmed [9,13]. For all the other compounds, chemical toxicity is the limiting factor; kidney content and concentration can be assessed from urine measurements [9,14]. The normally accepted 'safe' value is $3 \mu\text{g}$ uranium per gram kidney tissue, although recent evidence [14,15] suggests that this value should be reduced by about an order of magnitude. Recommended routine monitoring intervals are shown in Table VII.

Table VII. Recommended routine monitoring intervals for important uranium compounds

Compound	Lung	Urine	Faeces	Limit on intake ²
Nitrate				
Tributylphosphate	No	30 d	No	Chemical
Peroxide (UO_4)				
Ammonium diuranate	180 d	90 d	180 d	Chemical
Trioxide (UO_3)				
Tetrafluoride	180 d	90 d	180 d	Chemical/Radiotoxic
Octoxide (U_3O_8)	180 d	90 d	180 d	Radiotoxic
Dioxide (UO_2)				

² Based on a maximum concentration of $3 \mu\text{g}$ uranium per gram kidney tissue

2.5.4. Special monitoring for plutonium

The materials considered were nitrate, dioxide either alone or in combination with other metal oxides (including MOX), and Pu bearing dusts from a nuclear power plant (NPP). In general, faecal assay could confirm lower doses than urine assay although measurements of ^{137}Cs and ^{60}Co could be used with advantage for assessing low doses from the NPP dust. The examples shown in figure 6 illustrate the advantage of faecal assay (MDA 1 mBq d^{-1}) over urine assay (MDA 0.1 mBq d^{-1}) for assessing doses from inhaled ^{239}Pu . In figures 6 and 7, the shaded areas define the extremes in minimum detectable dose resulting from the ranges in model parameters shown in Table VIII. For other parameters, default values for occupational exposure were used.

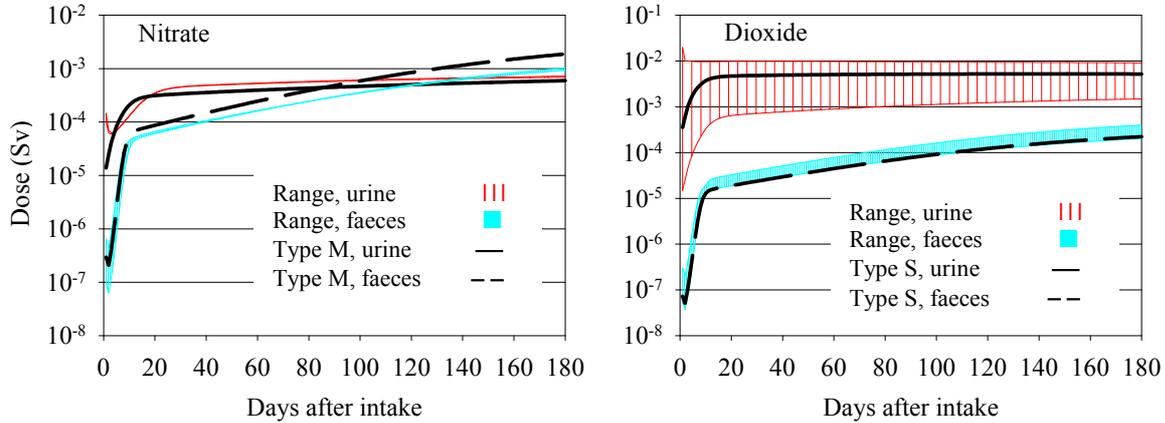


FIG. 6. Minimum detectable doses from plutonium nitrate and dioxide exposures

2.5.5. Special monitoring for thorium

The materials considered were nitrate and dioxide, both of natural isotopic composition. In general, faecal assay (RL 10 Bq d^{-1} or $2.5 \mu\text{g d}^{-1}$ ^{232}Th) resulted in lower minimum detectable doses than urine (RL 1 Bq d^{-1} or $0.25 \mu\text{g d}^{-1}$ ^{232}Th) for assessing doses from inhaled Th nitrate and dioxide (figure 7). For lower dietary excretion rates in faeces, the procedure could be used for assessing annual doses below 6 mSv . Thoron-in-breath measurements are also capable of demonstrating annual doses of $<6 \text{ mSv}$ for dioxide.

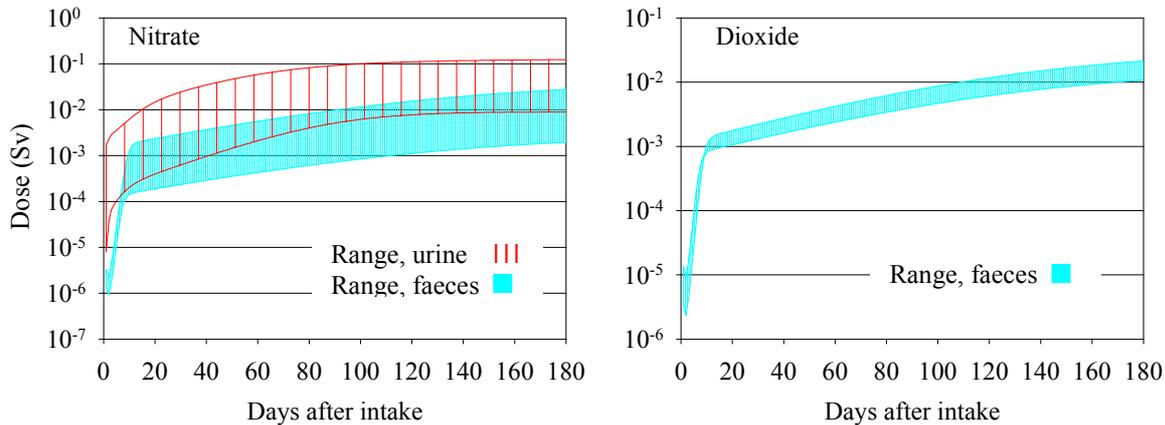


FIG. 7. Minimum detectable doses from thorium nitrate and dioxide exposures

Table VIII. HRTM parameter value ranges

		f_r	$s_r (\text{d}^{-1})$	$s_s (\text{d}^{-1})$	AMAD (μm)
Plutonium	Nitrate	0.21	0.23	2.4×10^{-3}	1 – 10
	Oxide	0.00005 – 0.05	0.2 – 20	10^{-4}	1 – 10
Thorium	Nitrate	0.03 – 0.3	0.2 – 20	10^{-4}	1 – 10
	Oxide	$10^{-6} - 10^{-2}$	0.2 – 20	10^{-6}	1 – 10

2.5.6. Monitoring for specific radionuclides and compounds: Conclusions

If all the factors that can affect the interpretation of monitoring data are taken into account (eg uncertainty in intake pattern, variability in particle size, absorption parameter values, differences in retention functions, and realistic MDAs) then clear judgements can be made on the most effective monitoring procedures, and on whether assessments of effective dose have sufficient sensitivity and accuracy to meet the appropriate legal requirements.

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