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Guidance Report on Screening and Rapid Methods for In Vitro Emergency Bioassay

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Editorial

In the last years a growing interest for radiological and nuclear emergencies has been expressed by many institutions and scientific associations like EURADOS network. In particular Working Group 7, Internal Dosimetry, expressed the need to evaluate the state of the art in emergency bioassay analyses. Actinides and ^{90}Sr were chosen as the "stars" of in vitro measurements, but as they normally require radiochemistry and long counting times, incompatible with fast emergency response, a questionnaire was sent to many laboratories all around Europe, related to PROCORAD Association (<http://www.procorad.org>), to test their preparedness level, develop new emergency procedures and to write a Guide, useful for everyone.

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Abstract

This report is intended to be a guide to indirect monitoring methods with special attention to actinides and ^{90}Sr intakes in an emergency scenario, focus on nuclear or radiological accident/incident scenario including malevolent one. Such situations may involve high doses and many affected people, needing a rapid response and demanding to quickly determine the number of exposed individuals and assess contamination levels.

In routine operations, bioassay techniques for activity quantification of these radionuclides require the application of radiochemical separation protocols and long counting times to reach very low minimum detectable activity values. In contrast to routine methods, screening and rapid methods focus on speed (rapid turnaround times, ease of use, and sufficient sensitivity for emergency decision-making) and practicality (minimal preparation to facilitate rapid and shorter analysis to meet the urgent needs of an emergency scenario). Both approaches are essential for comprehensive radiological protection and response.

This document outlines the minimum types and requirements for sampling and measurement methods, as well as the best screening or triage methods. It also offers different proposals for rapid quantification methods and includes observations and considerations related to the intakes of the above-mentioned radionuclides. This technical report can be considered as a practical and useful tool in radiological emergency scenarios for the main decision processes, required for intervention.

1. Introduction and Context

According to IAEA [IAEA 2015] or European Commission [EC2014], emergency is defined as a non-routine situation or event that needs prompt action, primarily to mitigate a hazard or adverse consequences for human life, health, property or the environment. Focus on nuclear or radiological emergency, agency defines as an emergency in which there is, or is perceived to be a hazard due to the energy resulting from nuclear chain reaction or from the decay of the products of a chain reaction and/or radiation exposure.

In a nuclear or radiological emergency, one of the objectives of the emergency response is to regain control of the situation and mitigate the consequences, to save lives and avoid or minimize severe deterministic effects, to provide first aid and critical medical treatment, and to manage the treatment of radiation injuries to reduce the risk of stochastic effects. It is very important to keep the public informed and to maintain public trust. Therefore, in this context, early and rapid estimates of radionuclide intakes are a high priority for appropriate intervention and medical decision-makers [Alves-2017]. The official decision-making depends on the ability to rapidly and accurately identify and quantify internal contamination by bioassays techniques.

Radionuclides can enter the body by inhalation and/or ingestion routes, absorption through the intact skin or through wounds (see Figure 1.1). Therefore, individual monitoring is necessary for the assessment of internal exposures, consisting in the measurement of the amount of radioactive material retained in the body of individual workers or members of the public (i.e. direct measurements or *in vivo* monitoring), and the measurement of the radioactive material excreted by exposed people (indirect measurements or *in vitro* monitoring).

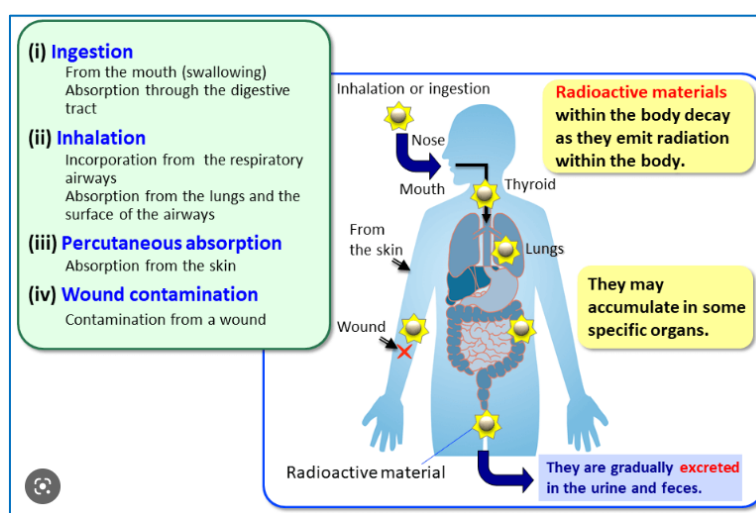


Figure 1.1: Possible pathways for internal contamination of the human body.

In this emergency scenario there are several key issues directly or indirectly related to each other that need to be considered: number of people exposed, type of exposure (acute, chronic), route of exposure, radionuclides involved, physical and chemical properties of the incorporated material, location of the incident, type of laboratories and their capabilities.

Depending on the radionuclide(s) involved, the number of exposed people, the distance between the event and the laboratories and the available measurements methods, the monitoring programs applied and the time and type of response are different. An emergency scenario may be associated

with high doses and many people affected, so it is necessary to give a prompt response and to know the number of persons exposed to internal contamination, to identify the radionuclides involved and to evaluate their contamination levels.

It should also be taken into consideration that the people responsible for the sampling of bioassay samples (nasal swaps, urine samples, faecal samples, blood samples ...) most likely will not be the people that arrive first at the scene of the incident. Law enforcement crews, medical emergency crews and fire brigade probably will already have arrived and set up perimeters. It is very important to contact first the person responsible for the co-ordination, let him or her know that your crew is present at the scene and ready to start their specific tasks, ask for a position to set up your post (or indicate the position of the laboratories involved in case of an on-site emergency) and get the permission to start sampling. Notify other emergency responders (such as medical teams) of your presence and intentions if needed.

Depending on the time elapsed after the incident, the actions taken and the goal of these actions will be different as well. The period of time after the occurrence of the incident can therefore be divided in three basic categories: triage and information gathering, medical aid and short term follow up and long term follow up.

1.1. Triage and information gathering

In the first hours after the incident little is known about the incident and its possible consequences. The first concern of the intervention teams is to gather as much information as possible about the impact of the contamination as fast as possible. This information is of crucial importance for the decision makers to define an adequate planning of the actions that need to be taken (such as evacuation of people, closing off of working areas, providing necessary first medical aid, samples to be taken, analyses to be performed, decontamination measures, etc.) [Chunseng-2012].

In the timeframe immediately following the incident, screening methods are the first solution to produce fast and relatively reliable results to estimate the severity of the event in terms of radiological risks and damages. The aim of these measurements is to gather information about the nature of the incident (which radionuclides are dispersed, what is the amount of radioactivity that was released, what is the surface and the boundaries of the area that was contaminated). This information will be used by the decision makers to decide which actions are required. Measurements that can be performed include (but are not limited to) surface monitoring of the area around the place of the incident by direct gamma-ray detection or contamination detection, swipes of contaminated material, soil samples, water samples or other relevant samples taken in the vicinity of the place of incident. However, these actions are not part of the scope of this document since they do not yield results that can be used for bioassay purposes.

Screening methods for bioassay measurements on the other hand focus more specifically on estimating the impact of the internal contamination of the people involved in the incident and the population living close to the place that is affected by the contamination. In the timeframe directly following the incident, the results of these screening tests are used to determine how many of the people possibly exposed to the radioactivity released can be classified as internally contaminated. The results of the screening tests should be available shortly after the incident occurred (within hours) and are primarily used as a triage instrument to identify contaminated people in need of follow up measurements. They are not meant for dosimetric purposes. Actions performed in this stage are external decontamination (shower or intensive washing), treatment of small wounds,

external contamination measurement, nasal swap analysis (internal contamination) and sampling of other types of excreta (urine, faeces).

1.2. Medical aid and short term follow up

As a direct consequence of the screening measurements a more detailed picture is obtained of the amount of exposed people that are contaminated (externally and/or internally) and the amount and type of radioactivity they are contaminated with. This allows the decision makers to set up a follow up plan of who should be subjected to which type of radiological measurement. The aim of these measurements is to obtain precise information about the contamination and the results should be sufficiently reliable to be used for the calculation of the dose received by each individual (dosimetric calculations). Both *in vivo* measurements (whole body counting, thyroid counting, lung counting) and *in vitro* measurements (urine analysis, faecal analysis) can be used for this goal. For the *in vitro* measurements (the scope of this document) sufficient samples have to be taken and analysed to obtain a good understanding of the source activity inhaled or ingested, the type of radioactivity involved and the excretion rate. Daily urine and faecal samples should be collected and kept for analysis and the collected samples must be analysed according to the follow up plan determined by the decision makers. Care has to be taken to consider the possible effects of complexing agents that might have been administered to the contaminated people (such as solutions of diethylenetriaminepentaacetic (DTPA) salts). This period of short term follow up starts as soon as possible (the day after the incident) and lasts several days up to a few weeks.

1.3. Long term follow-up

Finally, all people identified as being internally contaminated will be subjected to a long term follow up. This period stretches over months up to a year after the occurrence of the incident. Regular urine samples and faecal samples will be collected and analysed to obtain a more precise excretion model that will allow a reliable calculation of the dose received. The frequency of sampling depends on the type of contamination (radiological and biological half-life of the isotopes involved). The reliability of the results in this phase should be as good as possible, even if this means an increase in processing time (caused by more extensive separation methods and longer counting times). Since the amount of radioactivity excreted will normally decrease with time, only at this stage the concept of detection limit (DL) (or minimal detectable amount) becomes of interest.

1.4. Considerations in an emergency

1.4.1. People exposed

Following a radiological or nuclear emergency, workers, public, emergency responders may become contaminated with radionuclides. The number of persons considered as “emergency persons” could possibly be very large. Rapid activity answers and timely monitoring for internal contamination and assessing their intake and categorization (screening evaluation and distribution of the people exposed) are important issues of emergency response. Type of people exposed may vary greatly according to age, sex, physical conditions, state of healthy, etc. Moreover, in the nuclear radioactivity setting, exposed persons may not necessarily be familiar with radiation or requirements for measuring laboratories (type of sample, collection, types of containers, meaning of the results or outcome....) as well as quality control measures. So, these factors condition the type of response and require the application and adaptation of analysis and monitoring protocols to different

circumstances and they are a challenge in every specific emergency scenario. Sometimes, a large population affected may need to be quickly assessed for possible internal contamination (yes/no answer, semi-quantitative answer, or quick measurement with high DL) to provide information about contamination for dose assessment and necessary medical treatment.

1.4.2. Type of exposure/Radionuclides

Intakes of radionuclides can be evaluated through measurements of radionuclides present in organs and tissues, distributed inside the body, or in urinary and/or faecal excretion. Intakes of radionuclides are evaluated through in vivo and in vitro bioassays. Internal contamination with radioactive material is typically assessed by in vitro bioassay, such as urine/faecal measurement, or by in vivo bioassay, such whole body, organ, or wound counting. For emergencies where gamma or some beta emitters are involved, in vivo bioassay using portal monitoring or hand-held monitoring could be an excellent alternative and more suitable for in the field screening than in vitro bioassay, while for emergencies where alpha emitters or ^{90}Sr are involved, the laboratories based in vitro bioassay are required and, in the most cases, the unique possibility [Chunseng-2011].

Other important topic is the mixture of radionuclides involved in the emergency; it is advisable the maximum knowledge of isotopic composition: mixed alphas/betas or mixed pure beta emitters that may interfere the identification and measurement, may require radiochemical isolations which increase the difficulty of the procedures, the time to provide an answer and the number of required processes to be performed at the same time.

1.4.3. Type of laboratories/capabilities

One of the main problems in the Internal Dosimetry Services are to adapt laboratory routine protocols to an emergency scenario. It could appear a lack of capacity and capabilities for massive screening of large number of people exposed to alpha contamination. In an emergency situation the requirements and type of samples are different from routine, prioritizing of samples criteria must be established, techniques of measurement and the requirements in terms of detection limits are modified, methods of rapid analysis and their quality control are established but not necessarily well-trained.

This work has been developed by international in vitro bioassay laboratories that routinely perform radionuclide activity determinations in excreta for monitoring and surveillance of exposed workers and in exceptional incidents/accidents. Most of them are accredited laboratories according to a quality system (e.g. following ISO 17025_2017) and/or are authorised laboratories by the national regulatory bodies. They apply standardized, validated, and intercomparison methods and achieve very low detection limits using very specific quality conditions (tracers, low background levels...). Moreover, laboratories apply conditions and criteria for acceptance/rejection of samples received, specific containers are delivered and used for sample collection, there are specific sampling instructions according to the radionuclides to be analysed (spot, 24h, night sampling), and even the personnel exposed to in vitro measurements are fully aware of the instructions.

This guide focuses on a hypothetical emergency scenario involving intakes of actinides and/or Sr-89/90. In routine monitoring programme the main problem of these radionuclides is the quantification of their activity in biological samples because of the necessity to apply radiochemical treatment protocols and long counting time in order to "detect" acceptable levels of dose. These two conditions are opposite to the bioassay required in an emergency scenario. It is therefore imperative

to take into account and adapt bioassay laboratories (many of which are accredited for routine measurements and monitoring) to the emergency situation. This implies that the quality controls could be modified or suppressed, postponing deliverance to existing customers, acceptance of all type of samples.... etc. During the emergency, the demand for bioassay will probably exceed the capability and capacity of the laboratory, and as a consequence of the overload, it could be required bioassay to be performed in other type of laboratories. A global laboratory network would facilitate collective efforts in response to an emergency, but unfortunately, no such network for in vitro laboratories exists and would require an international assistance platform.

The aim of this report is to provide guidance, to update and compile current information on in vitro measurement techniques for the quantification of alpha emitters and $^{89/90}\text{Sr}$ in excreta (urine, faeces, mucus...). The minimum sampling requirements are presented as well as the best screening and/or triage techniques, rapid quantification methods, the most important warnings and considerations that allow a correct and more complete decision-making process to be carried out.

2. Type of samples

In an emergency situation, radionuclides may be incorporated into the human body by inhalation, ingestion, through absorption from intact skin or a wound. The intake can be estimated by measuring the concentration of the nuclides in the excreta using in vitro bioassays.

Given the limited time frame available for performing mainly screening measurements it is not practical to have the patients transported to the analytical facility for sampling, unless one has a (mobile) analysis laboratory available at the incident site. Sampling has to be done close to the incident site, but outside of the contaminated area. So, the first concern is to have an adequate team at the premises able to monitor people involved in the incident or affected by the contamination caused by it for external contamination and take the proper actions (removing cloths, washing of hands, face and hair). These people are also responsible for transferring affected persons from the contaminated zone to the not contaminated (sampling) area without spreading the contamination any further (containment).

At the sampling location all relevant information of the patients has to be registered. This includes but is not limited to their full name, address, phone number, age or date of birth. The sampling time should also be recorded as well as the conditions (did the patient just left the contaminated area, and if so: when? where was the patient at the time of the incident and what was he or she doing?). A unique identification number has to be given and registered to the patient as well as a unique identification number for each sample taken from the patient, including the type of sample (e.g. left nostril, right nostril, nose blow, face swipe). Data are preferentially registered electronically (spreadsheet or database) and communicated either directly (by means of a network) or indirectly (by storing the relevant data on an electronic carrier such as a flash drive) with the analysis lab.

Next, the main instructions of collection of the samples used in bioassays are explained.

2.1. Nasal swabs/Nasal mucus Samples

Nasal swabs can be a useful indicator of an inhalation intake however their interpretations are subject to very large uncertainties [NF EN ISO 20553 2017].

They should be collected as soon as the patient's condition permits, before showering or washing, preferably just after leaving the contaminated area and within 30 min of the contamination incident and in a radiological clean area. This procedure must be performed before an individual put a finger in, or blows his/her nose [HAS 2011].

It is recommended to clean the area around the nose and then take a nose-blow sample by letting the patient blow his/her nose in a paper tissue. Sampling can be performed using cotton tipped applicators or flag type applicators (which are inserted directly into the nose).

However, some laboratories use paper tissues to have the person blow his/her nose and collect the mucus excreted. In this case this nose-blow sample can be contaminated by radioactive dust that sticks to the skin around the nose (lips, cheeks, mouth) and could give a false positive result (it is however usually assumed that in case of screening a false positive result is better than false negative)

The typical nasal-swab procedure is to use a slightly moistened cotton tipped applicator and gently rotate the swab around the accessible surface of the nostril. A separate swab should be used for each nostril to prevent cross contamination of either the swabs or the nostril.

A variation on this technique is to wrap a small gauze pad or a 47 mm air filter paper common in occupational exposure settings around the applicator.

The use of the gauze pad or filter paper allows a flat geometry, which is particularly suited for counting for alpha activity that can be shielded by a thin film of water. Each swab should be put into its own container (e.g., test tube, envelope, or bag), labelled with personal identification (first and last name, date of birth and date and hour of collection).

The use of paper tissue is also possible.

2.1.1. Instructions of collection

General instructions of collection:

- Collect the sample as soon as possible, preferably within 30 min of the contamination incident.
- In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample (see remark on using paper tissues).
- Use a nasal swab slightly moistened with cotton tipped applicator and gently rotate around the accessible surface of the nostril.
- Proceed in the same way with other nasal swab in the other nostril to avoid cross contamination.
- Each swab should be put into its own container (e.g., test tube, envelope, or bag), labelled with personal identification: first and last name, date of birth and date and hour of collection and nostril (left, right).
- Depending on the number of samples to be analysed, the laboratory's criteria and its capabilities, both tips (or a single nose blowing tissue) can be combined to perform the measurement, as in case of an emergency the differences in activity between the left and right nostril should not be relevant.
- Nasal mucus can be collected with nasal swabs as above or with tissues. The patient blows in the tissue and the tissue should be put in a small plastic bag with personal identification (first name and last name, date of birth and date and hour of collection).
- A sheet to register administrative information is needed (phone number, e-mail adress...) to contact for extra samples and for follow up measurements

The samples will be sent to the laboratory as soon as possible.

2.1.2. Preservation and storage

There is no specific condition for storage and preservation (room temperature).

2.1.3. Transport

There is no specific condition for transport (room temperature).

2.2. Urine Samples

2.2.1. Instructions of collection

In an emergency scenario, spot urine will be collected to provide a quick dose assessment for contaminated individuals. Spot samples collect the total volume of a single voiding of urine and the typical volume is from 50 to 300 ml.

The collection should be done in a clean environment to minimize the potential for external contamination of the sample (single-use containers and clean hands is advisable) [ICRP 130].

The first urine voiding after an intake is likely to be not representative of the intake, because it will include urine accumulated in the bladder prior to the intake. It is recommended that the bladder be emptied as soon as practical after an intake, and the second voiding some hours later is used for analysis. The first urine is the model of the past just before incident. This is true only if the first sample is discarded less than 2-4 hours after incident. If the time is longer, it is not advisable because a little quantity of contamination could be excreted.

Urine should be collected preferably just after leaving the contaminated area, in clean containers that can be tightly sealed following sample collection and must be adequately identified with personal identification (at least first name, last name, date of birth, date and hour of collection).

General instructions of collection:

- Discard the first urine after the possible intake. Collect the sample immediately after.
- In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample.
- Identify the sample: first and last name, date of birth and date and hour of collection.
- An instruction sheet can be attached to provide more information (instruction for collection, impact on human body in case of nuclear incident, websites...)
- In case of an accident with actinides and/or 90Sr, it is recommended/suggested to collect a spot sample shortly after the accident and a urine sample 24 hours later. Each person is given a urine container and a proper sheet with instructions how to take a 24-hour urine sample at home (or at worst in the hospital). If the screening results of this person require so and the decision makers find it necessary, only then the available resources can be used to effectively analyse the urine sample.

2.2.2. Preservation and storage

In order to help to the preservation of the samples it is recommended to add HNO₃ concentrated (10 ml per litre of urine) and store it in a fridge.

Sodium benzoate could be used as preservative (1g per litre of urine), added in the container. HNO₃ concentrated can be added when the samples are arriving in the laboratory.

The samples will be sent to the laboratory as soon as possible.

2.2.3. Transport

- Place the sample in a zip lock bag, along with sufficient absorbent with precautions such as sealed container, zip bag, shipping container.
- Place specimens upright in the shipping container.

- Fill any residual space in the shipping container to prevent spillage during the transport.
- If it is possible, maintain refrigerated.

2.3. Faecal Samples

2.3.1. Instructions of collection

Due to its characteristics, this kind of sample is hardly usable in an emergency scenario, particularly when the number of involved people is high. Collection of faecal samples is not the sample of choice in emergency situations, but for further information even faecal samples are considered in this document. They are required as short-, medium- and long-term follow-up and/or confirmation samples for insoluble compounds, especially for suspected or confirmed high level of dose.

The samples collected for dose assessment need to be total voiding, not simply stool smears or swabs. The samples must be taken quickly after contamination.

It is recommended that stool samples are collected on a daily base, and at least up to 3 days following the incident [HAS 2011, NF EN ISO 20553]. This means taking 3 individual daily samples, not one single sample combining all stools.

The collection should be done in a clean environment to minimize the potential for external contamination of the sample (single-use containers and clean hands is advisable). Faecal samples should be collected in air-tight plastic containers and must be adequately identified with personal identification (first and last name, date of birth and date and hour of collection).

A leaflet with clear and succinct instructions will be given with the material to collect samples.

2.3.2. Preservation and storage

It is advisable to store the samples at least between 2°C and 8°C, and preferably to freeze them between -15°C and -20°C until pre-treatment by calcination, in order to limit inconvenience.

2.3.3. Transport

There are no special transport conditions but it is advisable to store the samples at least between 2°C and 8°C.

2.4. Saliva samples

The saliva sample is of interest, for the measurement of tritium, as an earlier indicator of exposure, as an indicator of the maximum level of contamination and can be easier to carry out.

It is used in incidental emergency situations involving tritium. [HAS 2011]

2.4.1. Instructions of collection

The saliva can be collected with different type of devices with swab or not. The most used sampling system is the Salivette® system.

- Collect the sample as soon as possible, preferably within 30 min of the contamination incident.

- In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample
- Remove the swab from the Salivette® container
- Place the swab in the mouth and chew it for about 60 seconds to stimulate salivation.
- Return the sawb with absorbed saliva to the Salivette® container and repalce the stopper

2.4.2. Preservation and storage

There is no specific condition for storage and preservation (room temperature).

2.4.3. Transport

There is no specific condition for transport (room temperature).

3. Techniques of measurement

Decisions and selection about the appropriate method of measurement and the pre-treatment of the sample prior to counting are very important. These decisions and flowcharts should be based on the information on the radionuclides involved (if they are known), the number of people exposed, the chemical and physical forms, the possible presence of interfering radionuclides, the sensitivity of technique, the availability of instrumentation and technical expertise in the laboratory or the time elapsed since the accident and last but not least the number of people involved.

Due to this guide which includes general lines in emergency situations, it must be understood that the main key is **time**. Therefore, it is important to place special emphasis on the fact that the routine methods of each laboratory may not be applicable. The most important aspect is to make an effort to minimize the time needed to get a result. This may result in not maintaining all the established steps in a routine procedure, e.g. performing radiochemical separation.

Depending on the techniques available in each laboratory, priority must be given to analysis with the fastest equipment or those that provide us more information in less time. An example of this would be the use of ICP-MS or gamma spectrometry, even if this means achievable detection limits are not reached.

Routine methods for measuring alpha radionuclides and Sr isotopes in biological samples specifically involve radiochemical processes in order to first be separated from the matrix to allow sensitive and reproducible detection.

Instrumentation for radiometric assessment is divided into two classes depending on the type of emission: alpha particles and beta particles.

Alpha emitters can be detected by a variety of techniques such as alpha spectrometry, mass spectrometry (ICP-MS: Inductively Coupled Plasma Mass Spectrometry and TIMS: Thermal Ionization Mass Spectrometer), KPA (Kinetic Phosphorescence Analyzer), fluorimetry, gas flow proportional counters or scintillation counters. For some alpha isotopes, such as ^{241}Am , gamma spectrometry could be performed when their activity is high.

Sr isotopes can be detected measuring beta activity by liquid scintillating counting or proportional counter. If you want to specifically measure $^{89/90}\text{Sr}$ it is necessary to perform a separation first, since the sample can contain a lot of other beta emitters.

Table 3.1 shows indications of the minimum detectable activity values achievable with the most common techniques used in radio-toxicology.

Techniques of measurement	Urine/Faeces/Nasal swabs
<i>Alpha spectrometry</i> (U, Th, Am, Pu, Cm, Np isotopes)	0.1-0.05 mBq/l (urine) 0.2-0.5 mBq/24h (faeces)
<i>ICP-MS</i> (^{238}U , ^{235}U , ^{234}U)	1 ng/l (urine)
<i>TIMS</i> (^{239}Pu , ^{238}U , ^{235}U , ^{234}U)	0.005 mBq/l (urine)
<i>Liquid scintillation counting</i> ($^{89-90}\text{Sr}$)	0.1 Bq/l (urine) 100 mBq/sample (nasal swabs/mucus) ⁽¹⁾
<i>Proportional counting</i> (alpha emitters & $^{89-90}\text{Sr}$)	0.05 Bq/l (urine) 50-100 mBq/sample (nasal swabs/mucus) ⁽¹⁾
<i>Phosphorimetry & Fluorimetry</i> (^{238}U)	0.05 - 1 $\mu\text{g/l}$ (urine)
<i>Gamma ray spectrometry</i> (^{241}Am)	100 mBq/l (urine) ⁽¹⁾

Table 3.1: Techniques of measurement and achievable detection limit values in **routine** bioassay methods [EURADOS Report 2013-01].

3.1. Alpha Spectrometry

3.1.1. Purpose and scope

Alpha spectrometry can be used for the measurement of actinides (such as Am, Np, Pu, Th, Cm, U) and Po. Spectra are collected in an alpha spectrometer with vacuum connected PIPS (Passivated Implanted Planar Silicon) detectors. Measurement equipment should be calibrated prior to any sample measurements. Energy calibration and efficiency calibration are performed using certified sources. Calibration and quality assurance in terms of efficiency, energy shape FWHM (Full Width at Half Maximum) and background must be checked in. Efficiency calibration can be controlled by the counting area obtained by measuring a single reference source (commonly ^{241}Am or ^{238}Pu are used). Energy calibration control is made in the whole range of the alpha particle energies emitted by isotopes of Am, U and Pu (4 MeV - 6 MeV).

The main advantage of alpha spectrometry technique is that it allows the identification and quantification of the radionuclides in the sample at a very low level, provided that a good radiochemical separation of the radioelements is achieved.

3.1.2. Conditions of use

To use this measurement technique, sample preparation and separation method are necessary (nuclides with unresolvable alpha energies such as ^{241}Am and ^{238}Pu , ^{237}Np and ^{234}U ...). In addition, it requires a source preparation by electrolytic deposition onto stainless steel planchets or by rare earth fluoride microprecipitation onto for example, polypropylene filters [ISO 13166:2020] [ISO 13167:2023] [UNE 73340-1:2021].

¹ Values from the experience of some laboratories that have participated in the guide

Many commercial solutions are available that include valid information and procedures for sample preparation and use of resin cartridges for actinide separation. The use of vacuum assisted flow and stacked cartridges allows sample preparation to be completed in as little as 10 hours at flow rates between 2-3 ml/minute.

Main interferences and important considerations:

- (1) ^{238}Pu and ^{241}Am energy interferences because of bad radiochemical separation
- (2) ^{242}Pu and ^{234}U energy interferences because of bad resolution
- (3) ^{226}Ra and ^{238}U energy interferences
- (4) When Cm is analysed together with Th using ^{229}Th as a tracer, it is necessary to remove its daughters ^{225}Ac and ^{221}Fr that interfere.
- (5) Presence of ^{210}Po interferes with ^{243}Am , used as a tracer, if not well eliminated
- (6) ^{212}Bi and ^{242}Cm
- (7) ^{232}U tracer should be cleaned from its daughters to avoid false positive measurement of ^{228}Th .
- (8) Problems of the interferences are enhanced when peak resolution is not good enough
- (9) Sodium nitrite is used to oxidize Pu(III) to Pu(IV) to enable the Pu/Am separation.
- (10) High levels of phosphate may lead to reduced recovery of actinides in calcium phosphate precipitation and column separation.

3.2. Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

3.2.1. Purpose and scope

ICP-MS technique may be used for long-lived radionuclides in biological samples. The aim of the technique is to focus on the mass of the radionuclide; radiochemical separation is only needed when isotopes of same mass are present in the sample. Depending on the sensitivity of the mass spectrometry equipment and the level of activity useful for the specific aim of the measurement, **thorium, plutonium, or other actinides** may also be analysed [ISO 17294-2:2016]. In the case of having a high resolution ICP-MS equipment and a sample de-solvating nebulizer, the signal can be increased by up to 10x. Otherwise, for some isotopes such as ^{238}Pu , a previous radiochemical separation will be necessary.

Uranium isotopes in urine. ICP-MS spectrometry allows quantifying values in pg/l – ng/l concentrations of the different isotopes (^{233}U , ^{234}U , ^{235}U , ^{236}U and ^{238}U) by mass [ASTM C1844-16]. The ratio of ^{235}U to ^{238}U (enrichment measurement) may in turn be used to estimate levels of minor uranium isotopes, in particular ^{234}U , that are more important for calculating the internal radiation dose received by an individual exposed to uranium, especially if enriched.

3.2.2. Conditions of use

Uranium calibration can be done by external calibration or isotope dilution. Samples that have been shown to contain levels of uranium that are sufficiently high to allow enrichment to be specified and that the user of the data considers to be significant are diluted by a factor of 5-20 with 0.1-0.4M nitric acid. ICP-MS has been calibrated for mass bias parameter using a uranium reference material with certified enrichment level less than 5% for the ^{235}U value. Analysis times per sample can be reduced to 1 or 2 minutes per sample depending on the equipment sensitivity. The main advantage in emergency situation is that the sample turnaround time is much shorter (compared with other techniques to quantify actinides, such as alpha spectrometry).

Interferences and important considerations:

- (1) Ultra high purity (UHP) concentrated nitric acid and water to reduce and control the blank signal.
- (2) The positive bias is most likely caused by polyatomic interferences on the minor U isotopes (^{235}U and ^{236}U). Urine digestion improved isotope ratio accuracy over dilution-only methods, but chemical separation of U from the urine matrix provided the most reliable results. In addition, accurate measurement of $^{236}\text{U}/^{238}\text{U}$ requires use of a desolvation system to reduce interference from $^{235}\text{UH}^+$ on ^{236}U . It will also be possible to eliminate the interference by analysing the samples in medium resolution and/or using an ICP-MS with a dynamic oxygen reaction cell.

3.3. Liquid Scintillation Counter (LSC)

3.3.1. Purpose and scope

Liquid-scintillation counting can be applied to measure Sr isotopes and alpha emitters in urine, faecal and nasal swabs/mucus samples. Separation between beta and alpha emitters is possible if the instrument is equipped with an alpha/beta discrimination option. With very high counting efficiencies and less concern about sample solids content, this method can attain low detection limits reliably, especially for gross alpha.

If you are performing a direct measurement on urine samples, for instance, you are limited to the volume of sample you can add to the LSC cocktail (which is usually only a few ml), but also you can concentrate the sample or add a distillation step if the radionuclides to be measured allow it.

It can provide more accurate measurements of gross alpha and beta activity because corrections for efficiency are much smaller than those that need to be applied for gas-proportional counting methods.

The liquid-scintillation counting technique ensures uniform source geometry with liquid samples (a nasal swap into LSC cocktail does not ensure uniform source geometry) and avoids the potential of uneven distribution of solids encountered in the source preparation for proportional counting.

3.3.2. Conditions of use

Strontium in urine is separated using Sr-resin prior to liquid scintillation counting. Cation exchange resin or calcium phosphate precipitation is used to concentrate strontium from samples. Stable strontium and/or strontium-85 tracer are added to monitor method yields and correct results in order to improve precision and accuracy [ISO 13160:2021].

Alpha emitters in nasal swabs and mucus samples can be measured the same way as other samples. It only requires the nasal swab material to be dissolved and put in a mild acidic solution (pH 1 - 2). The alpha activity can be detected in the beta spectrum or in a separate portion if the instrument is equipped with alpha/beta separation option. Counting efficiency is near to 100% [ISO 11704:2018].

Addition to liquid scintillation cocktail is required before counting in an energy and efficiency calibrated counter.

Interferences and important considerations

Strontium

- (1) If it is suspected that natural strontium is present in the sample, its concentration should be determined by a suitable means and the yield calculation appropriately modified.

- (2) After separation of Sr from the sample ^{90}Y will start to grow in again. The results obtained by LSC have to be corrected for the contribution of ^{90}Y .

3.4. Proportional Counting

3.4.1. Purpose and scope

The thin-window, heavily shielded, gas-flow, anticoincidence-circuitry proportional counter is the recommended instrument for counting gross alpha and beta low radioactivity.

These include a very low background (lower than in liquid-scintillation spectrometry) and a high sensitivity to detect and count an alpha and beta radiation range.

3.4.2. Conditions of use

Strontium in urine is separated using Sr-resin. Cation exchange resin or calcium phosphate precipitation is used to concentrate strontium from samples. Stable strontium and/or strontium-85 tracer are used to monitor method yields and correct results to improve precision and accuracy. Also, the counter used should be calibrated using a ^{90}Sr calibration source. If another calibration source has been used (e.g. ^{40}K or ^{204}Tl) then the results have to be communicated as equivalent to the isotope used for calibration (e.g. ^{90}Sr activity = xx Bq/l ^{40}K -equivalent) [ISO 13160:2021].

Alpha emitters in nasal swabs and mucus samples. Sample is pre-treated in a glass vial with concentrated HNO_3 and hydrogen peroxide and the sample heated to dryness. This step is repeated as necessary, and the residue is dissolved in dilute HNO_3 . The wet-ashing is required due to organic impurities or suspended materials.

In both cases, the evaporation of the sample on planchet is required before counting in a geometry and efficiency calibrated counter [ISO 9696:2017] [ISO 10704:2019].

Interferences and important considerations

Strontium.

- (1) If it is suspected that natural strontium is present in the sample, its concentration should be determined by a suitable means and the yield calculation appropriately modified.
- (2) It is necessary to remove ^{140}Ba and ^{40}K isotopes as well as other matrix interference's.
- (3) After separation of Sr from the sample ^{90}Y will start to grow in again. The results obtained have to be corrected for the contribution of ^{90}Y .

Alpha emitters in nasal swabs and mucus samples.

- (1) If the method of measurement used is based on the thin layer method, it is important that the thickness on the planchet is low to avoid high self-absorption that penalize result accuracy and detection limits.

3.5. Other Techniques

3.5.1. Cherenkov

Can be used for the measurement of ^{89}Sr . The solution obtained after separation of strontium from the sample is put into a LSC vial (no scintillation cocktail added) and measured. This measurement only obtains the effect of the high energy beta particles emitted by ^{89}Sr . Once the Cherenkov

measurement is completed, LSC cocktail can be added and a regular LSC measurement can be performed to also measure the activity of the ^{90}Sr present in the sample [ISO 13160:2021].

Cherenkov counting can be used also for a measurement of ^{90}Sr through beta emission of ^{90}Y once activity equilibrium is reached.

3.5.2. Thermal ionisation mass spectrometry (TIMS)

One of the applications is to measure Sr isotopes. Typically, the abundance of ^{90}Sr has been measured using radiometric techniques, such as liquid scintillators or gas ionization detectors. The major disadvantage of these techniques is the total analysis time. Mass spectrometry has provided a means to dramatically reduce analysis time, improving sample throughput. However, the peak tailing from the highly abundant ^{88}Sr has limited the ability of conventional mass spectrometry to be able to resolve samples with low radioactivity. Typical abundance sensitivities for the $^{90}\text{Sr}/^{88}\text{Sr}$ ratio for inductively coupled plasma mass spectrometry with a dynamic reaction cell was in the order of 10^{-9} , whereas thermal ionization mass spectrometry (TIMS) was in the order of 10^{-10} , using a TIMS with a Wide Aperture Retardation Potential (WARP) energy filter, which has no effect on peak tailing on the high mass side of the peak. As a result, TIMS has the ability to precisely and accurately determine $^{90}\text{Sr}/^{88}\text{Sr}$ ratios of 9.1×10^{-12} . This enables samples with very low activities to be measured.

3.5.3. KPA (Pulsed-Laser Phosphorimetry)

This test method covers the determination of total uranium, by concentration, in urine, $0.05 \mu\text{g/l}$ or greater [ASTM D5174-07(2013)]. Samples with uranium by mass, levels above the laser phosphorimeter dynamic range (analytical range of $400 \mu\text{g/l}$ or greater) are diluted to bring the concentration to a measurable level.

A sample aliquot is pipetted into a pre-treated glass vial. Concentrated HNO_3 and hydrogen peroxide are added, and the sample heated to dryness. This step is repeated as necessary, and the residue is dissolved in dilute HNO_3 . A complexant provides phosphorescent lifetime, stronger complexing, and low background level of uranium is added to the sample. The wet-ashing is required due to organic impurities or suspended materials.

Interferences and important considerations

- (1) Absorption (inner filter effect). Ferric iron and oxy-anions such as nitrate and organic acids are examples. Visible excitation (425 nm) may be absorbed by yellow solutions, for example, chromate. The consequences of this may be reduced signals and low analysis results.
- (2) Lumiphors. Many organic substances, such as humic acids and organic degradation products from incomplete ashing emit luminescence of varying lifetimes after excitation. Also possible medication taken by the person can interfere.
- (3) Quenching. Shortened triplet-state lifetime and reduced phosphorescence intensities of the excited uranyl complex.
- (4) Competing reactions. The uranyl ion must be protected from various intermolecular mechanisms which rapidly quench the uranyl luminescence. Complexation fulfils this need and examples of effective agents are phosphoric acid, polyphosphates and Uraplex.

3.5.4. Fluorimetry (Fluorimetric Method)

The method covers the determination of total uranium, after a co-precipitation step or direct analysis, in urine, from 0.1 or 1.0 µg/l.

The uranium is concentrated by co-precipitation with aluminium phosphate. Then is dissolved in diluted nitric acid containing magnesium nitrate as a salting agent and the co-precipitated uranium is extracted into ethyl acetate. After the ethyl acetate is removed by evaporation, the extracted residue is dissolved and diluted to volume in a small flask. Aliquots are transferred to each of two fusion dishes and dried. Flux containing sodium fluoride is added to each of the dishes, fused at a prescribed temperature (625°C), cooled, and read in a fluorimeter.

Interferences and important considerations

- (1) Quenching. The fluorescence of uranium in a fluoride matrix can be quenched or enhanced by either cations or anions. These interferences can be eliminated by coprecipitation with aluminium phosphate.
- (2) Carbonate ions form soluble uranium complexes which prevent the coprecipitation on aluminium phosphate. Carbonates are removed by acidification and expelled from solution as volatile carbon dioxide.

3.5.5. ²⁴¹Am by gamma spectrometry

This method describes the use of gamma spectroscopy, using germanium (Ge) diodes crystals, for the measurement of gamma photons emitted from ²⁴¹Am present in urine sample [ISO 20042: 2019].

Determine energy and efficiency calibrations for each detector at several energies between 50 and 2000 keV for the geometries of interest (59.5 keV gamma photons of ²⁴¹Am).

Because gamma spectroscopy is non-destructive, it is possible to analyse for gamma-emitting radionuclides without separating them from the urine matrix. This technique makes it possible to identify and quantify ²⁴¹Am when the activity is higher than 0.1 Bq/l.

4. Screening Methods (Triage)

Triage is defined as a rapid method with simple procedures to sort persons into groups according to their activity levels and/or injury or disease for the purpose of expediting clinical care and maximizing use of the available clinical services and facilities.

4.1. Purpose and Scope

Since wounded people will receive immediate medical aid and will be prone to proper registration and medical follow up, they should also be subjected to measurement of the longer-term impact of a possible internal contamination through the collection of urine and/or fecal samples. Therefore, it serves little use to spend extra time in trying to take blood samples or other samples related to the wounds for screening and probably even taking nasal swabs can be omitted.

Secondly, ingestion of radioactive matter without simultaneous inhalation of the same matter is highly unlikely in the case of a nuclear incident. Therefore, except in case of tritium, as already exposed in chapter 2, taking saliva samples to estimate the amount of radioactivity ingested by the people involved might not be the best approach for immediate measurements, although it might be considered as an alternative method depending on the nature of the incident.

Because of the above considerations measuring the possible inhalation of radioactive contamination by taking nasal swabs (also called nose blows) seems to be the best approach to get as much information as possible in a short period after the occurrence of the nuclear incident.

4.2. Detection Limits (Sensitivity)

Sensitivity and with that the obtainable detection limit of the screening methods is of minor importance. Since the patients being monitored are considered to be possibly contaminated due to a nuclear incident, the radioactivity levels expected to be found in nasal swabs will be high, probably in the order of tens of Becquerels or higher. With the methods described further a detection limit well below 1 Bq can easily be obtained.

Results of screening analyses should never be used to calculate doses nor as a basis to determine medical treatment [Klumpp-2017]. Screening methods serve one sole purpose: to obtain an answer to the question whether or not there is an indication of internal contamination. At best one can try to find an additional answer to the question if the contamination is caused by alpha emitting, beta emitting and/or gamma emitting radionuclides and to give a rough indication on the level of contamination (did the person inhale Becquerel, kilobecquerel or even higher amounts of radioactive matter?). In the latter case the swabbing efficiency has to be taken in to consideration. Usually only about 10% or less of the contaminant retained on the nostril hairs is removed by the swab [Yunlong-2017].

4.3. Minimum requirements of screening methods

The focus of the screening method selected for use should lie on the number of individual samples that can be processed and the speed at which the results are available to the decision makers. Other parameters such as accuracy, precision and sensitivity are far less important than throughput time and capacity.

Therefore, the screening method has to be able to process dozens if not hundreds of samples and deliver the results the same day the incident occurred or at the latest the day after. This timeframe includes taking the samples at the site of the incident, registering the necessary information, transporting the samples to the analysis lab (if needed), preparing the samples for measurement, performing the measurement, retrieving the measurement data, calculating the results and reporting the findings to the decision takers.

To assure a smooth operation and a sufficiently high measurement capacity this requires a number of persons with different responsibilities: taking the nasal swaps, register the necessary data about the sample, label the sample, package it in such a way that it can be transported without the danger of contaminating the environment to where the sample is taken. In the analysis laboratory the sample needs to be received, converted to a measurable object and measured. Finally, the measurement data has to be retrieved, the result calculated, registered in the database, linked to the patient's information and communicated to the decision makers. The preparation lab should be equipped with a sufficient large working area to allow the preparations of several samples simultaneously and have sufficient storage for the contaminated waste produced (packaging material, empty sample containers, gloves, pipettes ...).

The instrument used to measure the contamination of the converted samples needs to be equipped with a sample changer and be able to perform the analysis in a few of minutes (e.g. counting time limited to about 15 to 30 minutes depending on the capacity at hand and the number of samples expected to be measured).

4.4. Precautions and requirements when sampling

The main type of collection for triage are nasal swabs or mucus. Samples are taken by inserting a swab or a paper flag (moistened with a drop of distilled water) in each of the nostrils and wiping the inner surface of the nostril by turning the swabs or flags. Each sample is then transferred to an adequate container (e.g. small plastic, sealable bag or plastic vial with a screw cap) and labelled with the unique identification code of the sample and the patient. When using a paper tissue (e.g. ash free filter paper), have the patient blow his/her nose into the tissue. If required use two paper tissues and have the patient blow each nostril individually. Put the tissue(s) in an adequate container, preferably the same container that will be used later on in the analysis lab for the chemical destruction of the sample (e.g. a Pyrex round bottom flask). Seal the container and label it with the unique identification codes given to the sample and the patient. It is very important to avoid contamination of the sample by radioactive matter deposited outside the nose (e.g. around the mound and on the skin of the face). Clean these areas prior to sampling if necessary.

Samples and the related information should be transferred in batches to the analysis laboratory as soon as possible so a frequent transport between the sampling site and the analysis lab has to be provided (unless a mobile analysis laboratory is available).

4.5. Groups of methods/approaches by methodology or by isotope

Several options are available for measuring nasal swabs. First of all, there is the choice of sampling material. Simple cotton swabs can be used, or special swabs with low porosity to prevent incorporation of radioactive material in the swab material (and consequently absorption of alpha particle energy), paper flags or paper tissues (see Figure 4.1).

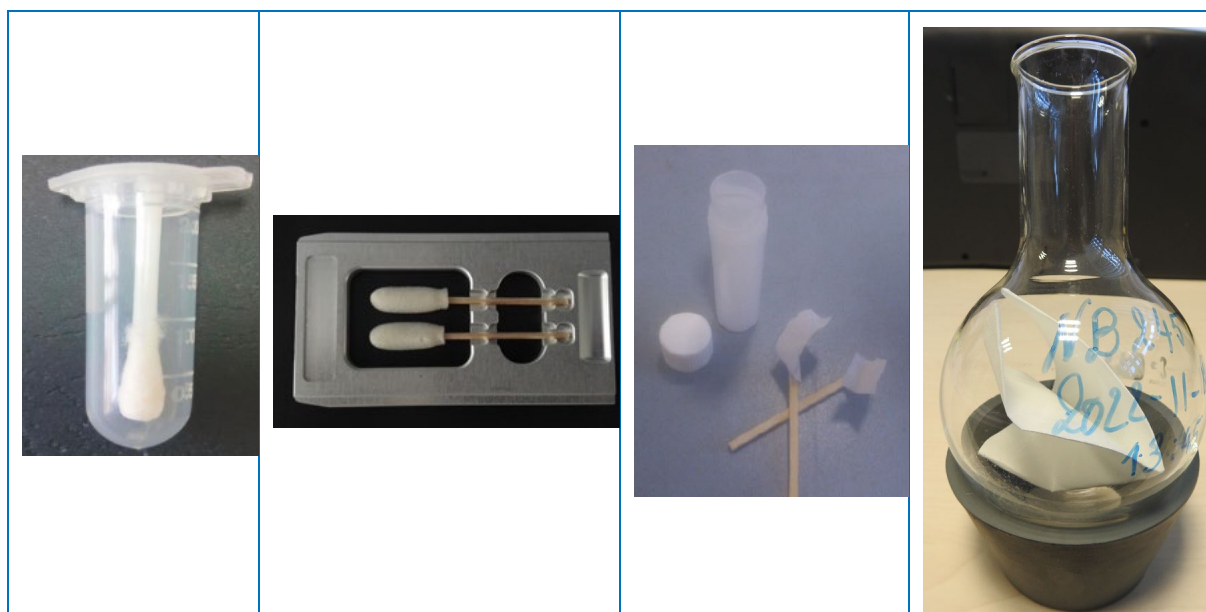


Figure 4.1: Different sampling materials for nasal swabs. Left: single cotton swab, middle left: double non-porous swab, middle right, paper flag and right: ash free paper tissue.

Secondly, the preparation of the samples can be different. Samples can be measured directly, after leaching or after full destruction using wet acid mineralization.

Finally, different measuring techniques can be used such as gamma-ray spectroscopy, alpha spectroscopy, liquid scintillation or solid-state scintillation (ZnS counting) or gas proportional counting. The choice in sampling, sample preparation and measuring technique has to be adapted to the type of radioactive decay to be measured.

4.5.1. *Measuring gamma-ray activity in nasal swabs*

When gamma emitters are present in the contamination (alone or in a mixture), it is recommended to perform their measurement by methods that allow a reliable evaluation of the intake, i.e. simplified in vivo methods (Whole Body Counter) or, alternatively, gamma spectrometry of small samples of urine (such as spot sample). This is especially useful for a mixture of radionuclide because the above-mentioned intake evaluation can be extended to alpha and beta emitters by means of the scaling factor obtained by measurement of environmental samples (likely the first samples where the radiological characterization of the contaminant material will be determined).

There are different options for methods for internal contamination screening:

Case A.- When the presence of gamma emitter is previously verified (this can be argued even by simple measurements of dose in air).

The best solution is performing an in vivo simplified measurement of a collection of nose mucus and (after voiding bladder) a collection of a small urine sample. In case of in vivo measurement positive it will be necessary to activate and specific in vitro rapid methods for alpha and beta emitters in urine.

In case of negative in vivo measurement, it will be essential a confirmation of non-significant contamination by alpha and beta emitter by scintillation counting of nasal mucus sample (both total beta and total alpha activity evaluation) (this step should even be avoided).

In both cases, if the results of gamma emitters are positive, a dose assessment and a decision on activation of medical treatments will perform. However, if alpha activity in nose mucus is detected (positive), an activation of medical treatments regardless gamma outcomes.

Case B.- When there is not possible to realize an in vivo measurement, a contemporary collection of nose mucus and (after voiding bladder) a collection of a small urine sample are recommended. Total alpha and total beta measurement of nose mucus by scintillation counting and gamma spectrometry of urine are carried out. In case of presence of alpha or beta contamination in nose mucus or gamma in urine (even with negative mucus) activation of alpha and beta emitters specific in vitro rapid methods.

The evaluation of positive dose by gamma emitters leads to a decision on activation of medical treatments. Likewise, positive alpha activity in nose mucus implies the activation of medical treatments regardless the gamma outcome.

Although not really relevant for internal contamination screening, measuring gamma-ray contamination of nasal swabs or spot urine samples might be required. The results provide information about the composition of the contamination with regards to gamma emitting isotopes and alpha or beta emitting isotopes that decay to gamma emitting daughters. No sample preparation is necessary and the nasal swabs or sport urine samples can be measured immediately after reception in the analysis laboratory. Preferably a high-resolution detector (such as a high purity Germanium detector) is used. The counting time should be set to acquire sufficient counts to have a 5 to 10% statistical accuracy at the 2σ confidence level.

4.5.2. Measuring beta particle activity in nasal swabs

Measuring the beta activity of nasal swabs requires either some extra sample preparation or extra care in selecting the correct combination of sampling material and detector. While direct measurement of high energetic beta particles is possible, the benefits usually do not outweigh the risks. The only real advantage of a direct measurement is the shorter processing time which in turn will enable to process more samples in a given period of time. This strategy could be selected if a large number of samples, contaminated with only beta emitters of high energy, has to be processed. The preferred choice of detector should be liquid scintillation counting by putting the nasal swabs directly in to the scintillation cocktail. Only porous nasal swabs should be used as they allow the cocktail to penetrate into the swab material and to come in direct contact with the beta emitting material to ensure proper signal generation.

Note that by using liquid scintillation detectors equipped with alpha/beta discrimination options it is possible to obtain both the result for alpha contamination and beta contamination in a single measurement. Depending on the radiochemical composition of the contamination this can possibly even be achieved with liquid scintillation counters without an alpha/beta discrimination option. If such equipment is available measuring the nasal swabs by liquid scintillation for both alpha and beta contamination is the preferred method of measurement.

Start each series of measurements with a background check (blank swab in scintillation cocktail) and measure this blank for about 30 minutes. This will give information on possible contamination of the detector or the counter, a possible correction factor for the background contribution (if needed) and, at the same time, give the waiting samples some time for the cocktail to penetrate the swab material. Again, count the samples sufficiently long to obtain a 5 to 10% statistical accuracy at the 2σ confidence level. Make sure the counter is equipped with a sample changer. It is recommended to

use scintillation vials to package and transport the nasal swabs after collection, as this avoids the need to transfer swabs from a container to the actual counting vial.

When paper flags or paper tissues are used to collect the samples, the preferred way to measure possible beta contamination is to treat the sample in a similar way as samples to be measured for alpha activity.

The use of direct measurement of non-porous swabs (surface contamination only) and proportional counting is not recommended. The risk of cross-contamination and contamination of the detector is simply too high, unless specifically designed counters for this purpose are used.

4.5.3. Measuring alpha particles activity in nasal swabs

Overall, the measurement of alpha activity (possibly combined with the measurement of beta activity) should be the main focus for internal contamination. As alpha particles have a very limited penetration into liquid or solid matter, care must be taken to avoid the partial or total absorption of alpha particle energy during measurement. For that reason, sample preparation is strongly recommended if not obligatory.

The best way to prepare the samples is to make sure that the sampling material used contains only acid-soluble substances in (e.g. no glass fibre reinforced paper materials present). In this way the samples can be easily and completely dissolved in the analysis laboratory by treating them with a mixture of concentrated nitric and sulphuric acid and heating them. Once the sample is completely solubilized, the liquid has to be eliminated by evaporating it to dryness. The residue can then be dissolved again in a dilute nitric acid solution (1 M or less) to prepare the counting object. Sample preparation can be done in a time frame of about 1 hour and multiple samples can be treated simultaneously. This method allows the processing of regular swabs, paper flags and paper tissue.

The diluted solution can be added to liquid scintillation cocktail for direct measurement by liquid scintillation counting. If simultaneous measurement of alpha and beta activity is required, the counter should be set up to measure the samples in an alpha/beta separation mode. Count the samples sufficiently long enough to obtain a 5 to 10% statistical accuracy at the 2σ confidence level. Make sure the counter is equipped with a sample changer.

Another option is to evaporate the diluted solution on to a metal disk (flat or curved) and to measure these disks by either alpha spectroscopy, zinc sulphide counter or gas proportional counter. The advantage of the latter method is that individual results for both alpha and beta activity can be obtained simultaneously while alpha spectroscopy and zinc sulphide counting only deliver a result for the alpha contamination present in the sample. The benefit of alpha spectroscopy is that it also gives an indication of the isotopic composition of the contamination, which might be useful information but is not really the main purpose of the screening. The main disadvantage of this technique is the increased risk of contamination of the counting chain (detector and alpha chamber) and the difficulty of producing the counting disks (especially if using flat disks). Also, the absence of a sample changer requires additional human intervention.

When using the method described above it is possible to obtain a constant stream of results after an initial delay of a couple of hours. The number of results generated per hour is related only to the counting time used and the capacity in terms of detectors.

4.6. Reporting of the results

Results should by preference be communicated as soon as they are available and have been validated. The preferred format of the report is in an electronic form (e.g. spreadsheet, database, CSV or XML document) in such a way that the decision makers receiving the report can directly process the results. Exchanging reports is preferably done by e-mail attachments or by direct access of the decision makers to the database of the analysis laboratory (or a shared copy of it). Encryption of the email and the attachments is prone to local legislation and has to be considered up front. However, it is not recommended.

Each result should, at least, contain the identification of the patient, the identification of the sample, the time of sampling and the sampling conditions, the time of measurement, the type of measurement (beta total, alpha total, alpha specific, ...), method used (liquid scintillation counting, alpha spectrometry, gas proportional counting, ...), the calculated result with an uncertainty indication (specifying the coverage factor related to the uncertainty, e.g. $k = 2$ for about a 95% uncertainty interval) and the unit used for reporting the result (counts, Bq/sample, Pu-equivalent activity, ...).

4.7. Post processing requirements

All documentation received with the samples has to be kept for later investigation and control.

All prepared samples (counting vials or metal disks) have to be kept for later investigation. They have to be stored in such a way that they are not susceptible to deterioration and cannot cause cross contamination of the working environment.

All waste produced during the preparation of the samples (packaging materials, pipettes, gloves, ...) has to be discarded as radioactive waste.

Regular checks on possible contamination of the working environment and the counters used have to be performed (e.g. by using wipes of the inner surface of fume hoods or measuring blanks in the detectors used). The results of these measurements have to be kept for future reference as well.

4.8. Caveats and considerations

While analyzing nose swabs might seem to be the best option for quick data gathering right after a nuclear incident, one should be aware that the usefulness of nose swabs diminishes strongly with passing time. The general agreement is that swabs collected more than an hour after a possible contamination will likely produce unreliable results because the radioactive matter inhaled will have been mostly cleared away by the normal mucociliary activity taking place in the nostrils. For that reason, care has to be taken to properly register the time of sampling and to process only samples that are taken within a timeframe of up to 1 hour after the contamination. It is also important to truly understand the difference between **the time of the incident** and **the time of contamination**. Airborne particles can remain in the air for extended periods of time after the incident that caused the release of these particles. People being evacuated from the incident area are thus still possible candidates for nose swab analysis, even if the incident occurred several hours ago.

If the measurement of nasal swabs is focused on the determination of contamination by alpha emitting particles, additional precautions must be taken to avoid the absorption of the alpha particles or part of their energy in the sampling tissue used. Most of the commercially available swabs

are made of cotton, sponge, polyester or non-woven fabric. These materials tend to be very porous and the radioactive substance will end up deep inside the tip of the swab and probably surrounded by moisture or water. When measuring the swabs directly without sample preparation two things have to be taken into consideration. The first is to make sure the swabs are completely dry as the water present in the tips will absorb the energy of alpha particle. Secondly, it must be ensured that material of the detector used can come into direct contact with the alpha particles emitted. A good example of this is the use of liquid scintillation counting where the dried tips are immersed into the liquid scintillator cocktail. Measuring a highly porous swab using a solid-state detector (such as a Zinc Sulphite counter or a proportional counter) will yield very unreliable and greatly underestimated results.

When using swabs, always take sample from both nostrils (i.e. use two swabs per person) and try to measure the swabs separately. If the results of both swabs show a tenfold greater difference or more in activity level between the left and the right nostril, this might indicate that the contamination of the higher level nostril has been biased by the contaminated person himself (for instance by the patient's hands or fingers when wiping his or her face or nose).

5. Rapid Methods

5.1. Purpose and scope

The general purpose and scope is to identify techniques for measuring internal contamination by radioactive material that can be effectively used for a reliable estimate of the internal dose in situations where the need to quickly monitor a large or very large number of people is actual. In this frame the term "monitoring procedure" must be understood as the association of predefined dosimetry safety conditions (dose levels, minimum number of individual monitoring measurements to be completed in a predefined period of time, time useful for collection of sample) with specific measurement methods capable to satisfy them. Given that, in the most cases a series of acute contaminations is the most likely event to be faced, the advantages offered by the implementation of an "appropriate system for addressing this challenge" are almost self-evident; among these we recall in particular the possibility:

- to estimate the individual risk linked to the incorporation of radioactive material for all the people involved and to identify the subjects for whom actions should be taken;
- to provide useful information to the medical staff to evaluate the opportunity to adopt pharmacological therapies, to carry out tissue ablation or removal of radioactive fragments,
- to carry out a timely check even on the personnel involved in the rescue intervention,
- to limit the social and psychological impact of the event by providing the members of the general population concerned with correct and reliable information on the real extent of their personal risk and demonstrating the interest and active commitment of the Public Institutions in protecting every single citizen.

The application of a monitoring procedure may or may not follow a triage phase which enables the selection of people with the highest probability of risk (see chapter 4). In any case, it must respond to a fundamental aim: identifying as soon as possible all the cases where pre-established dose levels are exceeded so that measures can be taken to mitigate the effects, whether this involves applying pharmacological therapies to reduce the effects of radiations or to enhance "de-corporation" of radioactive contaminants, or more simply to insert people into a long-term follow-up program.

From an operational point of view, the objective is to be able to quickly provide data, that are sufficiently reliable enough to be immediately compared with (one or more) pre-established reference dose values deemed "significant", in order to implement appropriate interventions. Therefore, it is necessary to adopt an emergency operational scheme based on specific reference levels, L_R .

An example of a single level scheme and of a two-levels scheme (L_A = action level, L_I = investigation level) is presented respectively in Figure 5.1 and Figure 5.2, respectively.

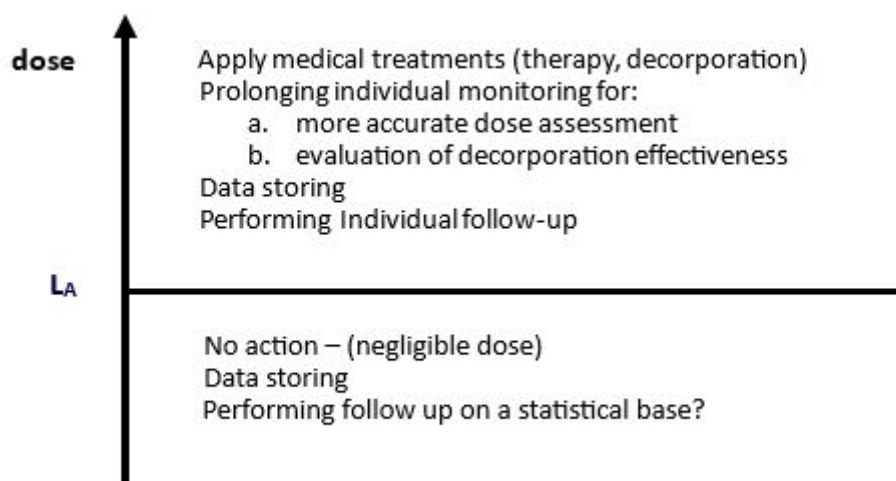


Figure 5.1: Single level operational scheme for internal contamination monitoring in emergency scenarios

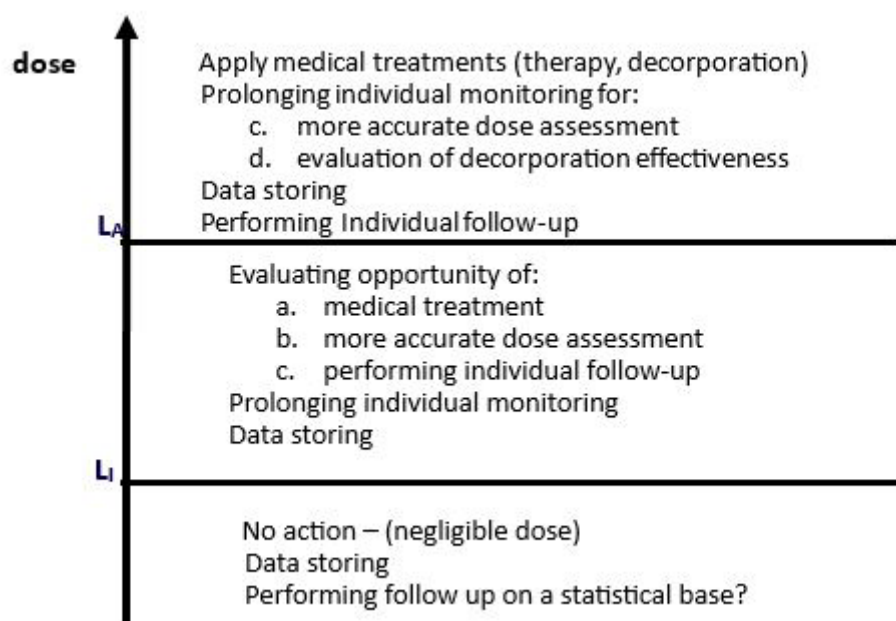


Figure 5.2: Two-level operational scheme for internal contamination monitoring in emergency scenarios

It is very important to point out that the adopted operational scheme adopted, even if it is based on a single reference level, strongly influences the possible measurement methods that can be used. Conversely, the limits placed by the available measurement methods realistically applicable in the specific situation, deeply influence the values that can be adopted for the reference levels.

In compliance with the ICRP indications for emergency situations [ICRP 109 and 146], the condition that must be guaranteed in all cases is that the reference levels, expressed in terms of committed effective dose, must be included at least within the range 20 mSv - 100 mSv, whether one or several (lower values are obviously well accepted, if possible).

It is also almost obvious that none of all this can't ignore careful considerations and feasibility assessments made in relation to:

- type of emergency to be managed
- pollutant dispersion modalities (i.e. prevalent route of intake)
- number of people to be monitored (potentially or actually)
- dose levels involved with respect to reference levels
- resources available in terms of personnel, instrumentation/laboratories and logistics.

Summarizing, the identification of appropriate measurement techniques should be generally performed in compliance with the following indication:

- the technique should be quick and easy and to use and should allow a large number of measurements in a short time (order of magnitude: dozens or hundreds per day)
- measurement results should be easily interpretable in terms of dose
- as far as possible, the following should be avoided:
 - samples that are difficult to be manage (faeces) or require long sampling periods or the storage of a large quantity of material to be stored (daily urinary excretion)
 - measurement methods requiring long sample processing period
 - monitoring must allow the detection of internal contamination levels that are lower ((preferably significantly lower) than those corresponding to the pre-established reference levels
- uncertainties associated with the measurement data shall be such they do not give rise to an excessive number of cases of doubt as to whether or not the reference levels have been exceeded.

5.2. Measurement Techniques: general requirements

Given the close relationship existing between the suitability of the measurement techniques adopted and the pre-established reference levels (one or more), whose choice also pertains to aspects of both a scientific and political nature, the following considerations will refer to a single reference level set at a committed effective dose of 10 mSv of (with possible further limitation, in specific cases, at a committed equivalent dose of 100 mSv to a specific organ). Taking into account the possible different choices that can be made, information is provided on the scaling factors for the various parameters in order to comply with other choices of reference values.

First of all, it should be underlined that the following treatment is based only on monitoring using in vitro techniques on small samples of urine (typically from 50 to 100 ml) assuming an average daily urinary excretion of 1600 ml, and considering that the collection and management of faeces is practically not applicable in emergency situations, while the possibility of using in vivo measurements has been shortly treated in a specific chapter on the monitoring of gamma emitting radionuclides.

In terms of timing, analytical techniques will be only considered useful if they allow to confirmation or exclusion of exceeding the adopted reference dose level, L_R (in mSv):

- providing the result of each individual measurement (in vitro measurement) within a maximum of 48 hours from sample collection or, in the case of accumulation of an extremely large number of samples to be monitored, from the activation of the monitoring procedure to the specific sample,

- making possible a working line (obviously with all the necessary equipment and staff) to complete the measurement of no less than 20 samples in 72 hours.

5.3. Minimum detectable activity (concentration) and minimum detectable dose

In the field of radioactivity measurement, the minimum detectable activity (*MDA*) (here considered to have the same statistical significance of the Detection Limit expressed in counts and indicated in ref. [ISO 11929]) , whether expressed in absolute terms (total activity) or relative terms (activity concentration), is the parameter that summarizes and defines, more than any other, the potential of a specific measurement technique in relation to the specific needs of the field of application in which one must operate. In radiation protection, and more specifically in the context of measures to assess the dose from internal contamination, this parameter, whose mathematical definition and practical determination can be found in the specific ISO publications [ISO 11929], constitutes in fact the basis for the definition of appropriate monitoring programs to guarantee that predefined risk levels are not exceeded.

In contrast to external exposure dosimetry, the measurement data in internal dosimetry, whether obtained from *in vitro* or *in vivo* measurements, cannot be directly compared with pre-established reference levels but must first necessarily be interpreted in terms of intake and effective committed dose. To this end, especially in a situations where the comparison should be immediate or, at least very fast, it's useful to introduce the concept of "minimum detectable effective dose", $MDA_E(\tau)$, that means the committed effective dose due to a specific radionuclide corresponding to an activity measured in the specific sample equal to the *MDA* value, where for *in vitro* measurements τ is the time (days) elapsed between contamination and sample collection, with MDA_{24h} expressed in terms of daily excretion (Bq/24h), and for *in vivo* measurements the time elapsed between contamination and actual measurement (with *MDA* expressed in Bq in the whole body); limiting to *in vitro* measurements of urine, in formula:

$$MDD_E(\tau) = \frac{e_{50} \cdot MDA_{24h}}{m(\tau)} \quad [5.1]$$

where: τ is the time elapsed between intake and collection of the urine sample (days);

$MDD_E(\tau)$ is the value of minimum committed effective dose in mSv due to the specific radionuclide investigated;

e_{50} is the dose coefficient in mSv/Bq;

MDA_{24h} is the value of the minimum detectable activity of the investigated radionuclide normalised to 24-hour excretion (Bq in 24h);

$m(\tau)$ is the proper urinary excretion function for the specific radionuclide, its chemical and physical form and the route of intake.

Indicating with QU the quantity of urine (in ml) used in the analysis, with R_U the ratio between the volume of daily urinary excretion for a Reference Man (1600 ml/day) and QU and with MDA_{QU} the minimum detectable activity obtained by analysing a quantity QU (ml) of urine, we obtain:

$$MDA_{24h} = MDA_{QU} * R_U = MDA_{QU} * 1600/QU \quad [5.2]$$

In order to consider the type of measurement useful for the purposes of complying with the reference level, the minimum condition is therefore expressed by:

$$MDD_E(\tau) = e_{50} * MDA_{24h} / m(\tau) = e_{50} * MDA_{QU} * R_U / m(\tau) < L_R \quad [5.3]$$

which allows to determine, in the given situation, the value of MDA_{QU} to be achieved with the specific technique adopted. As we will see, this condition is strictly dependant on the time spent between intake and sample collection, bringing to the necessity to define a time interval, $\Delta\tau_{UC}$, useful for sample collection.

Taking into account the parameters involved in the calculation of MDA for the specific measurement technique adopted, the possibility to make the monitoring procedure useful in a specific situation must therefore be based on the combination of the following parameters:

- > quantity of sample (i.e. volume of urine) used in the measurement, QU
- > counting time, Tm
- > detection efficiency, if changeable
- > time elapsed between contamination and collection of urine sample, τ

Given the aims of the measurements and the scenario, intersubjective variability of the metabolism of the radioelement and related uncertainties (i.e. values of retention/excretion functions and dose coefficients) are not considered in this treatment.

5.4. Selection and verification of a monitoring procedure

In order to identify and project a **monitoring procedure** responding to the conditions previously discussed and that allows, as already underlined, to carry out a suitable number of measurements in a sufficiently short time (at least a few dozens in 2 or 3 days from the contamination), a condition expressed in terms of dose can be reported in terms of counts.

Focusing the discussion on monitoring by *in vitro* measurements of urine samples, in order to identify a procedure complying to the condition posed by [5.3] for a specific radionuclide, (RN), one must first start from the value adopted for the Reference Dose Level, L_R , and convert it into terms of intake $I(L_R)$ of the radionuclide investigated, obviously considering the type of compound and the route of intake. This is easily achieved by applying the following formulas for committed effective dose:

$$I(L_R) = L_R / e(50) \quad (\text{in Bq}) \quad [5.4]$$

Multiplying $I(L_R)$ by the appropriate value of the urinary excretion function, $m(\tau)$, the activity of the radionuclide $A_{U,RN}(\tau)$ expected in the urine daily excreted at τ time from the contamination is:

$$A_{U,RN}(\tau) = I(L_R) * m(\tau) \quad [5.5]$$

By indicating with QU the volume of the urine sample used in the measurement and with R_U the ratio between the daily urinary excretion volume for the Reference Man (1600 ml/day) and QU , the activity of the radionuclide $A_{U,RN,S}(\tau)$ expected in the sample to be analysed is got easily from the following formula:

$$A_{U,RN,S}(\tau) = A_{U,RN}(\tau) / R_U = A_{U,RN}(\tau) * QU/1600 \quad [5.6]$$

Multiplying $A_{U,RN,S}(\tau)$ by the adopted counting time, T_m , and the yield, Y , of the specific radioactive emission selected for the measurement we obtain:

$$D_{U,RN,S,Tm}(\tau) = A_{U,RN,S}(\tau) * T_m * Y \quad [5.7]$$

where $D_{U,RN,S,Tm}(\tau)$ is the expected number of radiations of the type considered emitted during a measurement of duration T_m due to activity $A_{U,RN,S}(\tau)$ of the radionuclide, RN, present in the urine sample, S, used for the analysis and collected at τ days after the intake.

Considering also the counting efficiency for the selected radioactive emission (ε) obtainable with the instrumentation used and the specific measurement set up adopted, as well as the chemical recovery, ρ , typical of the method used for the pre-treatment of the measurement sample, the following equation is obtained:

$$C_{U,RN,S,Tm}(\tau) = D_{U,RN,S,Tm}(\tau) * \varepsilon * \rho \quad [5.8]$$

where $C_{U,RN,S,Tm}(\tau)$ is the expected number of counts recorded by measuring the specific radionuclide in the given conditions which are hereafter summarized:

- > internal contamination is due to a specific radionuclide, RN, in a known chemical form and through a known route of intake
- > the occurred intake, $I(LR)$, gives a committed effective dose corresponding to the adopted reference level, LR
- > monitoring is performed by in vitro measurement of the activity concentration in urine
- > the quantity of urine (QU) used for the measurement is a small fraction of the daily urinary excretion collected at day τ after the contamination.
- > adoption of a specific measurement technique performed in predefined set up which leads to a defined counting efficiency ε for the considered radiation
- > counting time, T_m , is pre-established
- > the sample pre-treatment is characterized by a stable (and possibly high) chemical recovery, ρ

Bringing everything back into a single formula we have:

$$C_{U,RN,S,Tm}(\tau) = I(L_R) * m(\tau) * (QU/1600) * T_m * Y * \varepsilon * \rho \quad [5.9]$$

Therefore, indicating with LD the detection limit [ISO 11929] for the measurement technique expressed in terms of total counts collected in a measurement time T_m , the technique is suitable if it satisfies the following minimum criterion:

$$C_{U,RN,S,Tm}(\tau) \geq LD \quad [5.10]$$

that is:

$$I(L_R) * m(\tau) * (QU/1600) * T_m * Y * \varepsilon * \rho \geq LD \quad [5.11]$$

where LD must be calculated using as a blank a sample of urine of volume QU , surely exempt from any contamination, to be measured applying exactly the same method and set up (counting time, source to detector distance....) taken for the evaluation of $C_{U,RN,S,Tm}(\tau)$.

From a practical point of view, the condition set in Eq. 5.11 must be satisfied for urine samples collected several days after the intake, especially for events involving a large number of people.

Given the dependence of the first term of the equation on $m(\tau)$, which obviously decreases with time, the duration of the time interval for a useful urine collection ($\Delta \tau_{UC}$) is limited. $\Delta \tau_{UC}$ is assumed here to be 5 days as a minimum²

From these equations it is highlighted that the parameters on which it is possible to directly intervene to define a proper measurement procedure with respect to the adopted values for the reference levels mainly are:

- > the value of $L(R)$ (i.e assuming a different value of the reference level, LR)
- > the quantity of urine analysed (QU),
- > the counting time (T_m);
- > the chemical recovery ρ (through the choice of a proper pre-treatment method)

where the value of the excretion function, $m(\tau)$, essentially limits the time interval useful for sample collection, while the counting efficiency, ε , which must obviously be maximized, essentially depends on the instrument type and relative set up used for the measurements.

Since the goal is to simplify and shorten as much as possible the monitoring method in order to perform the maximum number of measurements per day, while respecting the above-mentioned conditions (equation 5.10), it should be underlined that:

- > increasing the reference level, LR , is the simplest, most effective and fastest way to achieve the objective, given that no changes in the measurement method would even be necessary; however, this choice needs to be carefully evaluated and balanced because if it is abused to the point of substantially modifying the pre-established monitoring conditions, it could have serious repercussions both from a strictly technical-health point of view and from that of the political management of the event;
- > increasing the quantity of urine analysed, QU , should be a way to obtain a gain in terms of expected counts (value of $C_{U,RN,S,Tm}(\tau)$); conversely it could have an unfavorable impact on the absolute value of the detection limit, LD , because of variations on measurement parameters, and especially on the productivity (increasing of the duration of the procedure, lowering of the number of samples that can be processed simultaneously by a single operator);
- > counting time, T_m , is a parameter that can only really be used to optimize the procedure when it lasts a few hours or less; in fact it should be noted that its increase has a proportional effect on the duration of the procedure, whereas the gain in terms of the ratio between $C_{U,RN,S,Tm}(\tau)$ and LD has rather the typical trend of the square root of the relative increase in T_m itself;
- > chemical recovery, ρ , can only be a significant factor when it is very low and variable; in the case of small urine samples, with appropriate pre-treatment it is generally very high (above 70%) and stable, and is hardly to work on to optimize the procedure.

At least, it's important to point out that all decisions must be taken having in mind the need to be able to carry out as many useful measurements as realistically possible in a short period of time. For this reason, in the planning phase the whole procedure must be carefully projected and evaluated, not only in terms of duration and capacity to achieve reliable results, but also in terms of the number

² It's important to point out that $\Delta \tau_{UC}$ must not be confused with the duration of urine collection which can be a spot collection or extended up to 24 hours as a maximum

of samples that can be processed by a single operator in a fixed, reasonable time (for example, the duration of a work shift). It is therefore clear that there is a need to set up proper “*ad hoc* rapid measurement methods”.

5.5. Monitoring procedures

Monitoring procedures based on rapid measurement methods are here presented for a list of radionuclides of major radiation protection interest, complex to be monitored and for which, considered the field of application (emergencies), the use of in vitro techniques applied to small urine samples is practically mandatory; specifically: ^{90}Sr , ^{241}Am , ^{244}Cm and ^{239}Pu (as a reference for alpha emitting Plutonium isotopes).

The procedures and methods here presented are designed on the basis of the dosimetry considerations set out in the previous paragraph and should be simply taken as examples. They can obviously be replaced with alternative solutions according to each laboratory’s capabilities (procedures, equipment, personnel...) and preferences, as long as they meet the monitoring needs according to any indications in terms of dose (reference levels) established for the specific situation.

The presentation is organised exposing for each radionuclide considered a sheet containing:

- the list of the characteristic parameters of the proposed monitoring procedure, including implementation time and number of measurements that can be performed simultaneously by a single operator such as the possible productivity³ in 3 days of a single “working line” (obviously it is given that labs involved are equipped with all the necessary materials and instrumentation). The Detection Limit (LD) value and the corresponding MDA value achievable with the method are also reported,
- the list of the instruments, equipment, and reagents necessary for the measurement of a single sample (or a set of samples),
- the description of the measurement method, from the pre-treatment of the sample to the final radioactivity measurement,
- a list of the chemical form in which the specific radionuclide can be realistically inhaled or ingested,
- a dosimetry evaluation in which the applicability of the procedure with respect to the adopted reference level is tested on the basis of the parameter $C_{U,RN,S,Tm}(\tau)$ (equation 5.10), for all the listed significant chemical forms,
- for inhalation and ingestion of all chemical forms assumed by the specific radionuclide respecting condition (5.10 and 5.11), “dosimetric tables” containing, for the first seven days after the intake (always supposed as an acute intake), the most significant dosimetric data in the hypothesis that the measured activity in urine is equal to MDA value (minimum condition for detecting the presence of an actual contamination); the last day from the intake useful for urine collection (upper limit of $\Delta\tau_{UC}$) is also provided,
- for all cases where the monitoring procedure is not applicable as proposed (not respecting equation 5.10), the possible solutions or choices to be adopted, if any, to get the monitoring procedure useful and the respective “dosimetry tables”.

³ Number of analysis completed in a fixed time interval

It is important to point out that:

- reported *LD* and *MDA* values have been calculated on the basis of the outcomes of experimental tests specially carried out by applying each specific measurement method to a set of 20 blank samples (urine samples surely not contaminated) to which only the proper tracer was added, when necessary,
- data provided allow to plan the number of measurements that can be performed by a laboratory in a defined period, taking into account availability of materials and instrumentation, the number of operators available, their experience and skill, the possibility they have of working simultaneously and/or with rotation of shifts,
- the time interval for useful collection, $\Delta\tau_{UC}$, is a parameter related to the condition of applicability of the method with respect to the Reference Level adopted (equation 5.11). It indicates the time interval during which, using a specific monitoring procedure, sample collection is appropriate to achieve pre-established performance, that is: all contaminations giving a committed effective dose above the reference level shall be detected; in other words: the minimum detectable dose of the procedure is below the reference level if the sample is collected in the interval $\Delta\tau_{UC}$. It may not be significant or appropriate for the management of the event. However, in view of the possible need to use the monitoring procedure for mass contamination events, $\Delta\tau_{UC}$ should anyway be long enough to allow urine to be collected from as many people as possible, taking into account the almost inevitable inconveniences and errors that this situation would certainly entail, especially when dealing with members of the public. For this reason, the conditions of acceptability of the monitoring procedure will also include a condition on $\Delta\tau_{UC}$. The condition assumed here is that it must not be less than 5 days, if possible, although this value is not to be understood as mandatory, but as a prudential indication to be considered and evaluated on the basis of the actual situation to be faced.

Finally, it is especially important to underline that the Monitoring procedures hereafter presented, including the use of guide radionuclides, have been projected based on the last ICRP recommendations [ICRP 130] and the most update dosimetry and metabolic data for the radionuclides investigated [ICRP 134, ICRP. 137, ICRP 141]. To this end OIR Viewer (Occupational Intake of Radionuclides data viewer) has been used which, at the moment, is the only tool that provides the excretion functions and dose coefficients based on the last ICRP (https://journals.sagepub.com/doi/suppl/10.1177/ANIB_48_2-3). **For this reason, data are referred to occupational exposure and thus to the reference man.**

Strictly speaking the excretion functions available in OIR Viewer are not suitable for the population, but when ICRP will propose excretion and retention functions for all the categories of the population, the approach here proposed will still be valid and could be the object of a second edition of this document.

5.6. Strontium (^{90}Sr)

The standard methodologies [Horwitz 1992, ISO/FDIS 13160] commonly adopted in the usual monitoring of internal contamination by ^{90}Sr are based on the chromatographic extraction and separation of the radionuclide in urine samples (volumes of a few hundred ml, typically 500 ml) and on the evaluation of its activity by measuring the ingrowth of the daughter, ^{90}Y [ISO/FDIS 13160, Ridone 2017], or following its decay after it has reached equilibrium in activity with the parent. Both

processes require many days to obtain reliable measurements and are therefore not suitable for use in emergency situations or any case where a large number of people are involved.

The procedure proposed here is based on the measurement of urine samples performed by counting the total beta activity immediately after their treatment, in the hypothesis that the entire activity present, net of the blank, can be attributed to ^{90}Sr . This hypothesis needs that the above-mentioned treatment leads to a complete purification of the sample from other beta-emitting radionuclides, in particular radio-caesium and ^{40}K , and an almost total elimination of the ^{90}Y that should be present because of its excretion and its ingrowth due to ^{90}Sr decays from the time of urine collection. In any case, the method involves an overestimation of the activity due to:

- > the ^{90}Y possibly not eliminated by treatment
- > the quantity of ^{90}Y formed in the sample from the end of ^{90}Sr extraction and separation procedure to the completion of the counting (no more than 7 hours)
- > the use of the counting efficiency for ^{90}Sr even for the interfering ^{90}Y whose appropriate efficiency is actually much higher.

In total, the overestimation has been evaluated to be no more than 10%, on average, where the maximum contribution is due to the new formed ^{90}Y (about 6%). This overestimation is judged as an acceptable price to pay in favour of the possibilities given by the method, both in terms of number of measurements that can be performed per day, and in terms of validation of the results obtained. It is quite clear that it is however possible to subsequently repeat the counting of the sample, even several times, to evaluate the regrowth of ^{90}Y , in order to be able to perform a more accurate evaluation of the intake and dose. This may be necessary especially when the level of contamination appears as critical with respect to the adopted Reference Level.

With adequate supplies of expert personnel and the organisation of a proper work shift, this method, or an alternative one with comparable performances, allows a single, properly equipped "working line", to complete up to 90-100 measurements of ^{90}Sr in 3 days.

5.6.1. Measurement Method

Equipment

- 1 centrifuge tube (50 ml)
- 1 centrifuge (6 places rotor)
- 1 beaker 250 ml volume
- 1 Hot plate dim. 30 x 40 cm at least.
- 1 Sr-spec column with reservoir
- 1 stainless steel dish for proportional counter
- 1 Proportional counter

Reagents (analytical grade)

- Water, complying with ISO 3696, grade 3
- Stable Sr-carrier ($\text{Sr}(\text{NO}_3)_2$)
- Concentrated HNO_3 (> 65%)
- 30% H_2O_2
- 3M $\text{Ca}(\text{NO}_3)_2$ solution
- Concentrated H_3PO_4 (85%)
- Concentrated NH_4OH (28%)
- 4M NH_4OH solution
- 8M HNO_3 solution
- 0.05M HNO_3 solution

5.6.2. Pre-treatment and preparation of the measurement sample

After the addition of 2 mg of Sr carrier as a tracer to a 30 ml urine sample directly in a 50 ml centrifuge tube, 1 ml of 3M $\text{Ca}(\text{NO}_3)_2$ and 5 ml of concentrated H_3PO_4 are added, and the pH is adjusted to 8.5 – 9 with 10 ml of concentrated NH_4OH to allow the formation of calcium phosphate precipitate. The precipitate is let to settle down for 10 minutes and the sample is centrifuged at 3000 rpm for 15 min. The supernatant is discarded, the precipitate is rinsed with 4M NH_4OH and centrifuged again for 15 minutes. Finally, it is dissolved and transferred in a glass beaker with 5 ml of concentrated HNO_3 together with 3 rinses (5 ml concentrated HNO_3 each) of the centrifuge tube and it is evaporated to dryness on hot plate with some drops of H_2O_2 . The residue is dissolved in 10 ml of 8M HNO_3 and loaded on a Sr-spec column previously conditioned with 5 ml of 8M HNO_3 . After the sample is drained, a rinse solution of 8M HNO_3 (5ml) is used to wash the sample beaker and the column. Strontium is then eluted with 10 ml of 0.05M HNO_3 . An aliquot of 9 ml (90%) of this solution is quantitatively transferred to a stainless-steel disc, heated to dryness on hot plate, transferred into a proportional counter and measured for 6 hours. The remaining aliquot of 1 ml is used to evaluate the chemical recovery in Atomic Absorption Spectroscopy (AAS)⁴, to be performed preferably just during gross beta measurement in order not to prolong the overall time of the procedure.

5.6.3. Measurement of activity and evaluation of chemical recovery

The measurement of activity is performed using a 10-channel low-level planchet Gas Proportional Counter for 6 hours.

The activity A (Bq/l) of each sample is given by the following formula:

$$A_{90\text{Sr},S} = \frac{CR_S - CR_B}{\epsilon_{90\text{Sr}} \cdot \rho \cdot V \cdot k_V}$$

where: CR_S is the count rate of the sample (cps)

- > CR_B is the mean count rate of the blank (cps)
- > $\epsilon_{90\text{Sr}}$ is the counting efficiency for ^{90}Sr
- > ρ is the chemical recovery
- > V is the volume of the sample (L)
- > k_V is the correction factor for the reduction of analyzed sample volume due to the aliquot of eluate used for the chemical recovery evaluation (in case of AAS $k_V=0,9$)

The daily urinary excretion of ^{90}Sr activity (with the approximation that all counts are due only to strontium) is given by:

$$A_{90\text{Sr},d} = R_U \cdot A_{90\text{Sr},S} = 59,3 \cdot A_{90\text{Sr},S}$$

where R_U is the ratio between the volume of daily urinary excretion for a Reference Man (1600 ml/day) and the volume of urine effectively analyzed (30 ml x 0,9). The evaluation of the

⁴ This quantity is enough for AAS, in case of ICP-MS a lower amount of the eluate is sufficient for chemical recovery evaluation.

chemical recovery is made in AAS by comparison of the concentration of Sr-carrier measured in two standard solutions prepared for the calibration of the spectrophotometer.

5.6.4. Chemical forms assumed by ^{90}Sr

The alimentary tract transfer factor, f_A , used in this report is defined in ICRP Publication 100 as the fraction of activity entering the alimentary tract that is absorbed to blood, taking no account of losses due to radioactive decay or endogenous input of activity into the tract.

Inhalation

- > aerosol type F – chloride, sulphate and carbonate – $f_A = 0.25$
- > aerosols type M – fuel fragments, all unspecified forms - $f_A = 5.0 \cdot 10^{-2}$
- > aerosols type S – FAP (Fused Aluminosilicate Particles), PSL (Polystyrene) - $f_A = 2.5 \cdot 10^{-3}$

Ingestion

- > titanate - $f_A = 1.0 \cdot 10^{-2}$
- > all other chemical forms - $f_A = 0.25$

5.6.5. Evaluation of the reference monitoring procedure

Reference monitoring procedure parameters

- > reference level for committed effective dose, $L_R = 10 \text{ mSv}$
- > time interval for useful collection of urine sample, $\Delta\tau_{UC} : 5 \text{ days}$
- > activity measurement technique: total beta counting
- > monitoring sample: urine
- > volume of urine analysed: 30 ml
- > recovery tracer: stable Sr (2 mg/sample)
- > chemical recovery: 70% (a conservative value is applied)
- > counting efficiency: 34% ($\pm 1\%$)
- > counting time: 22000 s (6 hours)
- > yield: 100%
- > typical LD value for ^{90}Sr : 35 counts
- > typical MDA value for ^{90}Sr : 0.4 Bq/24h
- > duration of the pre-treatment for a single sample: 4.5 hours
- > number of sample pre-treatment performable simultaneously by a single operator: 10
- > duration of the simultaneous pre-treatment of 10 samples: 6-8 hours
- > time to complete the determination of ^{90}Sr activity in 10 samples by a single operator: 12-13 hours
- > estimated productivity in 3 days by a single “working line”: 90-100 measurements completed (including final results)

5.6.6. Condition of acceptability of the procedure:

$$C_{U,RN,S,Tm}(\tau) > LD=35 \text{ counts}$$

$$\Delta\tau_{UC} \geq 5 \text{ days}^5$$

In order to test the acceptability of the procedure the values of $C_{U,RN,S,Tm}(\tau)$, calculated for all strontium compounds listed above, are reported in Table 5.1 for urine collected at the first seven days following the intake, both by inhalation and ingestion.

Table 5.1: Values of the parameter $C_{U,90Sr,S,Tm}(\tau)$ obtained for ^{90}Sr applying the proposed monitoring procedure to urine collected in the first seven days following the intake

<i>$C_{U,RN,S,Tm}$ (counts) - intake of ^{90}Sr</i>								
τ	Inhalation of aerosol						Ingestion	
(days from intake)	Type S		Type M		Type F		Titanate	other compounds
(d)	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm		
1	776	1560	197000	352000	1630000	1520000	1420000	1700000
2	434	954	113000	212000	694000	759000	599000	687000
3	274	607	72300	130000	451000	461000	386000	434000
4	201	421	52000	96300	330000	325000	284000	322000
5	158	325	43400	72300	257000	260000	221000	253000
6	130	265	34700	62600	208000	211000	181000	206000
7	110	221	29000	53000	173000	176000	150000	170000

From the analysis of data in Table 5.1 it follows that the proposed procedure for the detection of ^{90}Sr contaminations giving committed effective dose values equal to the proposed reference level (10 mSv) is largely acceptable both in case of inhalation of all chemical forms and ingestion of all compounds.

In the following [dosimetry tables](#) the main dosimetry data (excretion function $m(\tau)$, intake, minimum detectable effective dose (MDD_E) are shown in the hypothesis that the activity of the radionuclide measured in urine sample (A_m) is equal to MDA. Data are given for collection of urine performed up to the seventh day after the contamination (the fifth day as a minimum) and allow to establish the maximum internal dose attributable when no contamination is detected applying the monitoring procedure. The last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$) is also reported.

5.6.7. Dosimetry tables for ^{90}Sr (reference monitoring procedure)

Reference data:

- > subject: reference man
- > route of intake and compounds:
- > inhalation of aerosol (AMAD: 1 μm , 5 μm)- compounds: type F (chloride, sulphate and carbonate), type M (fuel fragments, all unspecified forms), type S (FAP, PSL)
- > ingestion of: strontium titanate, all other chemical forms

⁵ Condition not mandatory but strongly recommended

- > Reference Level for committed effective dose: 10 mSv
- > measurement method: total beta counting
- > sample analysed: 30 ml of urine
- > minimum detectable activity: 0.4 Bq (value normalised to 24 h excretion)

Table 5.2: ^{90}Sr : inhalation of type F aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 1 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$4.7 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$8.5 \cdot 10^1$	$2.1 \cdot 10^{-4}$	< 1%
2	$2.0 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$2.0 \cdot 10^1$	$5.0 \cdot 10^{-4}$	< 1%
3	$1.3 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$3.1 \cdot 10^1$	$7.6 \cdot 10^{-4}$	< 1%
4	$9.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$4.2 \cdot 10^1$	$1.0 \cdot 10^{-3}$	< 1%
5	$7.4 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$5.4 \cdot 10^1$	$1.3 \cdot 10^{-3}$	< 1%
6	$6.0 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$6.6 \cdot 10^1$	$1.7 \cdot 10^{-3}$	< 1%
7	$5.0 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$7.9 \cdot 10^1$	$2.0 \cdot 10^{-3}$	< 1%
5000 ^(c)	$1.0 \cdot 10^{-6}$	$4.0 \cdot 10^{-1}$	$4.0 \cdot 10^5$	$9.9 \cdot 10^0$	99%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose.

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.3:** ^{90}Sr : inhalation of type F aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ (AMAD = 5 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$5.6 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$7.1 \cdot 10^0$	$2.3 \cdot 10^{-4}$	< 1%
2	$2.8 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$1.4 \cdot 10^1$	$4.5 \cdot 10^{-4}$	< 1%
3	$1.7 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$2.3 \cdot 10^1$	$7.5 \cdot 10^{-4}$	< 1%
4	$1.2 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$3.3 \cdot 10^1$	$1.1 \cdot 10^{-3}$	< 1%
5	$9.6 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$4.1 \cdot 10^1$	$1.3 \cdot 10^{-3}$	< 1%
6	$7.8 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$5.1 \cdot 10^1$	$1.6 \cdot 10^{-3}$	< 1%
7	$6.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$6.1 \cdot 10^1$	$2.0 \cdot 10^{-3}$	< 1%
5000 ^(c)	$1.30 \cdot 10^{-6}$	$4.0 \cdot 10^{-1}$	$3.1 \cdot 10^5$	$9.8 \cdot 10^0$	98%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.4:** ^{90}Sr : inhalation of type M aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 5 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$6.8 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$5.8 \cdot 10^1$	$1.8 \cdot 10^{-3}$	< 1%
2	$3.9 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$1.0 \cdot 10^2$	$3.1 \cdot 10^{-3}$	< 1%
3	$2.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$1.6 \cdot 10^2$	$4.8 \cdot 10^{-3}$	< 1%
4	$1.8 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$2.2 \cdot 10^2$	$6.6 \cdot 10^{-3}$	< 1%
5	$1.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$2.6 \cdot 10^2$	$7.9 \cdot 10^{-3}$	< 1%
6	$1.2 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$3.3 \cdot 10^2$	$9.9 \cdot 10^{-3}$	< 1%
7	$1.0 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$4.0 \cdot 10^2$	$1.2 \cdot 10^{-2}$	< 1%
2300 ^(c)	$1.2 \cdot 10^{-6}$	$4.0 \cdot 10^{-1}$	$3.3 \cdot 10^5$	$9.9 \cdot 10^0$	99%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.5: ^{90}Sr : inhalation of type M aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ (AMAD = 5 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$7.3 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$5.4 \cdot 10^1$	$9.8 \cdot 10^{-4}$	< 1%
2	$4.4 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$9.0 \cdot 10^1$	$1.6 \cdot 10^{-3}$	< 1%
3	$2.7 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$1.5 \cdot 10^2$	$2.6 \cdot 10^{-3}$	< 1%
4	$2.0 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$2.0 \cdot 10^2$	$3.6 \cdot 10^{-3}$	< 1%
5	$1.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$2.6 \cdot 10^2$	$4.8 \cdot 10^{-3}$	< 1%
6	$1.3 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$3.1 \cdot 10^2$	$5.5 \cdot 10^{-3}$	< 1%
7	$1.1 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$3.6 \cdot 10^2$	$6.5 \cdot 10^{-3}$	< 1%
2800 ^(c)	$7.2 \cdot 10^{-7}$	$4.0 \cdot 10^{-1}$	$5.5 \cdot 10^5$	$9.9 \cdot 10^0$	99%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.6:** ^{90}Sr : inhalation of type S aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 1 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$3.4 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$1.2 \cdot 10^3$	$4.4 \cdot 10^{-1}$	4%
2	$1.9 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$2.1 \cdot 10^3$	$7.9 \cdot 10^{-1}$	8%
3	$1.2 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$3.3 \cdot 10^3$	$1.3 \cdot 10^0$	13%
4	$8.8 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$4.5 \cdot 10^3$	$1.7 \cdot 10^0$	17%
5	$6.9 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$5.8 \cdot 10^3$	$2.2 \cdot 10^0$	22%
6	$5.7 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$7.0 \cdot 10^3$	$2.6 \cdot 10^0$	26%
7	$4.8 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$8.3 \cdot 10^3$	$3.1 \cdot 10^0$	31%
22 ^(d)	$1.6 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$2.5 \cdot 10^4$	$9.4 \cdot 10^0$	94%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.7:** ^{90}Sr : inhalation of type S aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ (AMAD = 5 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$3.6 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$1.1 \cdot 10^3$	$2.2 \cdot 10^{-1}$	2%
2	$2.2 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$1.8 \cdot 10^3$	$3.6 \cdot 10^{-1}$	4%
3	$1.4 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$2.8 \cdot 10^3$	$5.7 \cdot 10^{-1}$	6%
4	$9.7 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$4.1 \cdot 10^3$	$8.2 \cdot 10^{-1}$	8%
5	$7.5 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$5.3 \cdot 10^3$	$1.1 \cdot 10^0$	11%
6	$6.1 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$6.5 \cdot 10^3$	$1.3 \cdot 10^0$	13%
7	$5.1 \cdot 10^{-5}$	$4.0 \cdot 10^{-1}$	$7.8 \cdot 10^3$	$1.6 \cdot 10^0$	16%
36 ^(d)	$8.0 \cdot 10^{-6}$	$4.0 \cdot 10^{-1}$	$5.0 \cdot 10^4$	$9.9 \cdot 10^0$	99%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.8: ^{90}Sr : ingestion of Strontium titanate, $f_A = 1.0 \cdot 10^{-2}$

τ (days)	$m(\tau)$ (titanate)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.8 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$2.2 \cdot 10^2$	$2.4 \cdot 10^{-4}$	< 1%
2	$7.6 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$5.2 \cdot 10^2$	$5.7 \cdot 10^{-4}$	< 1%
3	$4.9 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$8.1 \cdot 10^2$	$8.9 \cdot 10^{-4}$	< 1%
4	$3.6 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$1.1 \cdot 10^3$	$1.2 \cdot 10^{-3}$	< 1%
5	$2.8 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$1.4 \cdot 10^3$	$1.6 \cdot 10^{-3}$	< 1%
6	$2.3 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$1.7 \cdot 10^3$	$1.9 \cdot 10^{-3}$	< 1%
7	$1.9 \cdot 10^{-4}$	$4.0 \cdot 10^{-1}$	$2.1 \cdot 10^3$	$2.3 \cdot 10^{-3}$	< 1%
4000 ^(c)	$5.4 \cdot 10^{-8}$	$4.0 \cdot 10^{-1}$	$7.4 \cdot 10^6$	$8.1 \cdot 10^0$	81%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.9:** ^{90}Sr : ingestion of all chemical forms (but titanate), $f_A = 0.25$

τ (days)	$m(\tau)$ all forms ^(a)	$A_M = \text{MDA}^{(b)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(c)}$ (mSv)	Ratio E_{50}/L_R
1	$4.7 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$8.5 \cdot 10^1$	$2.0 \cdot 10^{-4}$	< 1%
2	$1.9 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$2.1 \cdot 10^1$	$5.0 \cdot 10^{-4}$	< 1%
3	$1.2 \cdot 10^{-2}$	$4.0 \cdot 10^{-1}$	$3.3 \cdot 10^1$	$7.9 \cdot 10^{-4}$	< 1%
4	$8.9 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$4.5 \cdot 10^1$	$1.1 \cdot 10^{-3}$	< 1%
5	$7.0 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$5.7 \cdot 10^1$	$1.4 \cdot 10^{-3}$	< 1%
6	$5.7 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$7.0 \cdot 10^1$	$1.7 \cdot 10^{-3}$	< 1%
7	$4.7 \cdot 10^{-3}$	$4.0 \cdot 10^{-1}$	$8.5 \cdot 10^1$	$2.0 \cdot 10^{-3}$	< 1%
5000 ^(d)	$9.5 \cdot 10^{-7}$	$4.0 \cdot 10^{-1}$	$4.2 \cdot 10^5$	$1.0 \cdot 10^1$	100%

(a) all chemical form but titanate

(b) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(c) Committed effective dose

(d) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

5.7. Americium and Curium (^{241}Am , ^{244}Cm)

Given that in internal dosimetry americium and curium are treated as having the same chemical properties and thus similar metabolic behaviour, the procedure proposed here (reference rapid monitoring procedure) applies a widely used standard measurement method based on the column extraction of americium and curium isotopes and preparation of the measurement sample by electrodeposition on a steel plate to be measured by alpha spectrometry [Arginelli 2016].

Compared to the procedure applied for routine monitoring, the only substantial difference is the use of small urine samples (reference: 50 ml), and a sufficiently short counting time (up to 80000 s), which makes it possible to achieve useful results in approximately 30-35 hours from the start of sample treatment.

With adequate supplies of expert personnel and the organisation of a proper work shift, this method, or an alternative one with comparable performances, allows a single, properly equipped "working line" to complete up to 45-50 measurements of ^{241}Am and ^{244}Cm in urine in 3 days.

5.7.1. Measurement Methods

Equipment

- > 2 beaker 150 ml volume
- > 1 Hot plate dim. 30 x 40 cm at least.

- > 1 30 ml reservoir in polypropylene for each sample analysed
- > 3 centrifuge tube (50 ml)
- > Vacuum box with internal rack for 50 ml centrifuges tubes, 3 outer tips in polypropylene and 3 inner tips in polyethylene for each sample analysed
- > 1 Normal DGA (N, N, N', N'-tetra-n-octyldiglycolamide) 2 ml cartridge (50 -100 μm)
- > 1 electrodeposition system
- > 1 alpha spectrometry system equipped with 450 mm² PIPS detector

Reagents (analytical grade)-

- > Water, complying with ISO 3696, grade 3
- > ²⁴³Am tracer solution (10 mBq/ml) certified by a metrological institute
- > Concentrated HNO₃ (> 65%)
- > 30% H₂O₂
- > 5M HNO₃ – 1M Al(NO₃)₃ solution
- > 5M HNO₃ solution
- > 0.1M HNO₃ solution
- > 0.5M HCl solution
- > Na₂SO₄ 15%_w solution
- > NaHSO₄ 5%_w solution
- > (NH₄)₂C₂O₄ 2%_w solution
- > KOH 25%_w solution
- > 95% Ethanol

5.7.2. Pre-treatment and preparation of the measurement sample

After the addition of 10 mBq of ²⁴³Am as a tracer to a 50 ml urine sample, 10 ml of concentrated HNO₃ and 5 ml of 30% H₂O₂ are used to destroy the organic matter. The solution is evaporated to dryness and the residue is mineralized several times, until a white residue is obtained, with 4 ml of concentrated HNO₃ + 2 ml of 30% H₂O₂ and finally dissolved in 15 ml of 5M HNO₃ – 1M Al(NO₃)₃ solution.

This solution obtained is loaded into a Normal DGA 2 ml cartridge placed on a vacuum box and preconditioned with 10 ml of 5M HNO₃; after the sample is drained, a rinse solution of 5M HNO₃ (10 ml) is used to wash the sample beaker and the cartridge. Then, another rinse step with 0.1 M HNO₃ (20 ml) is used to lower the nitric concentration and to elute uranium isotopes and other matrix components that could interfere with the source preparation ⁽⁶⁾. Finally, americium is stripped from DGA using 20 ml of 0.5M HCl. The eluate is then transferred in a 150 ml beaker, evaporated to dryness and mineralized three times with 4 ml of concentrated HNO₃ + 2 ml of 30% H₂O₂.

The residue is dissolved by adding 2.5 ml of 5%_w NaHSO₄, 2 ml of water and 5 ml of 15%_w Na₂SO₄. Then the solution is transferred into the electrodeposition cell with three portions of 1 ml water, to rinse the beaker, 1 ml of (NH₄)₂C₂O₄ 2%_w is added, then the anode is set up and the electrodeposition carried out at a constant current (about 1.0 A) for 60 minutes.

Finally, 2 ml of 25%_w KOH are added to stop the reaction; after 1 minute the power is turned off, electrode is removed, and the electrodeposition cell is emptied. The electrodeposition disk, which

⁶ Experimentally verified.

constitutes the real measured sample, is removed from the cell, washed first with water and then with ethanol and left air dry.

5.7.3. Measurement of activity and evaluation of chemical recovery

The measurement of activity is performed by alpha spectrometry using ion implanted silicon detectors (PIPS) of 450 mm² active surface. The measured sample (electrodeposition disk) is positioned inside a vacuum chamber at a distance of 1 mm to the detector's active surface and counted for 80000 s.

Taking into account that alpha ray counting efficiency in vacuum does not depend on alpha ray energy, the evaluation of ²⁴¹Am and ²⁴⁴Cm activity in the sample is obtained by comparison to ²⁴³Am applying these simple formulas:

$$A_{241\text{Am},S} = A_{243\text{Am},S} \times NC_{241\text{Am}} / NC_{243\text{Am}}$$

$$A_{244\text{Cm},S} = A_{243\text{Am},S} \times NC_{244\text{Cm}} / NC_{243\text{Am}}$$

where: $A_{241\text{Am},S}$ is the activity of ²⁴¹Am in the sample (Bq)
 $A_{244\text{Cm},S}$ is the activity of ²⁴⁴Cm in the sample (Bq)
 $A_{243\text{Am},S}$ is the activity of ²⁴³Am added to the sample (Bq)
 $NC_{241\text{Am}}$ is the net count in the ROI of ²⁴¹Am
 $NC_{244\text{Cm}}$ is the net count in the ROI of ²⁴⁴Cm
 $NC_{243\text{Am}}$ is the net count in the ROI of ²⁴³Am

The daily urinary excretion of ²⁴¹Am activity and ²⁴⁴Cm activity ($A_{241\text{Am},d}$, $A_{244\text{Cm},d}$ in Bq/24h) are then given respectively by:

$$A_{241\text{Am},d} = R_u \times A_{241\text{Am},S} = 32 \times A_{241\text{Am},S}$$

$$A_{244\text{Cm},d} = R_u \times A_{244\text{Cm},S} = 32 \times A_{244\text{Cm},S}$$

where R_u is the ratio between the volume of the daily urinary excretion for a Reference Man (1600 ml/day) and the volume of urine analyzed (50 ml).

Chemical recovery, ρ , is evaluated applying the following formula:

$$\rho = NC_{243\text{Am}} / (A_{243\text{Am},S} \times T_m \times Y \times \varepsilon)$$

where T_m is the counting time, ε the detection efficiency and Y the yield of the alpha emissions considered.

5.7.4. Chemical forms assumed by ²⁴¹Am

Inhalation

- > aerosol type F – citrate – $f_A = 5.0 \cdot 10^{-4}$
- > aerosols type M - oxide, chloride - $f_A = 1.0 \cdot 10^{-4}$
- > aerosols type S - associated with plutonium oxide - $f_A = 5.0 \cdot 10^{-6}$
- > aerosol - nitrate - $f_A = 3.0 \cdot 10^{-4}$

Ingestion

- > all compounds - $f_A = 5.0 \cdot 10^{-4}$

5.7.5. Evaluation of the reference monitoring procedure

Reference monitoring procedure parameters

- > reference level for committed effective dose, $L_R = 10$ mSv
- > time interval for useful collection of urine sample, $\Delta\tau_{UC}$: 5 days
- > activity measurement technique: alpha spectrometry
- > monitoring sample: urine
- > volume of urine analysed: 50 ml
- > recovery tracer: ^{243}Am (10 mBq/sample)
- > chemical recovery: 90% (a conservative value is applied)
- > counting efficiency: 33% ($\pm 1\%$)
- > counting time: 80000 s (22 h, less than 1 day)
- > typical LD value for ^{241}Am : 7.5 counts
- > typical LD value for ^{244}Cm : 4.0 counts
- > typical MDA value for ^{241}Am : $1.0 \cdot 10^{-2}$ Bq/24h ⁽⁷⁾
- > typical MDA value for ^{244}Cm : $5.0\text{E-}03$ Bq/24h ⁽⁶⁾
- > duration of the pre-treatment for a single sample: 6 hours
- > number of sample pre-treatment performable simultaneously by a single operator: 8
- > duration of the simultaneous pre-treatment of 8 samples: 8-9 hours
- > time necessary to complete the measurement of 8 samples by a single operator: 30-35 hours
- > estimated productivity in 3 days by a single “working line”: 45-50 measurements completed (including final results)

Condition of acceptability of the procedure

$$C_{U,RN,S,Tm}(\tau) > LD=7.5 \text{ counts}$$

$$\Delta\tau_{UC} \geq 5 \text{ days}^{(8)}$$

In order to test the acceptability of the procedure the values of $C_{U,RN,S,Tm}(\tau)$, calculated for all americium compounds listed above, are reported in table 5.2.0 for urine collected at the first seven days following the intake, both by inhalation and ingestion.

⁷ MDA value experimentally obtained for ^{241}Am : $9.5 \cdot 10^{-3}$ Bq/24h; for ^{244}Cm : $4.2 \cdot 10^{-3}$ Bq/24h

⁸ Condition not mandatory but strongly recommended

Table 5.10: Values of the parameter $C_{U,241Am,S,Tm}(\tau)$ obtained for ^{241}Am applying the proposed monitoring procedure to urine collected in the first seven days following the intake

$C_{U,241Am,S,Tm}(\tau)$ (counts)									
τ	Inhalation of aerosol								Ingestion
(days from intake)	Type F		Type M		Type S		Nitrate		all compounds
(d)	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	
1	860	878	244	251	6	6	603	589	3780
2	782	743	228	213	5	5	557	498	579
3	586	540	180	167	4	4	427	375	277
4	430	392	138	121	3	3	316	276	164
5	317	284	106	93	2	2	232	199	116
6	234	209	80	74	2	2	176	153	92
7	176	155	64	59	1	1	135	115	79

From the analysis of data in Table 5.10, it is immediately clear that with respect to detection of ^{241}Am contaminations giving committed effective dose values equal to a reference level of 10 mSv, the monitoring procedure proposed:

- is (widely) acceptable for inhalation of Type F, compounds of Type M and nitrates and for ingestion (all compounds)
- is not acceptable for inhalation of aerosol of highly insoluble compounds (type S) associated with plutonium oxides. In this case an alternative procedure must be used.

For all cases for which this procedure is applicable, in the following dosimetry tables the main dosimetry data (excretion function $m(\tau)$, intake, minimum detectable effective dose (MDD_E)) are shown in the hypothesis that the activity of the radionuclide measured in the urine sample (A_m) is equal to MDA. Data are given applying the procedure for collection of urine performed up to the seventh day after the contamination (fifth day as a minimum) and allow to establish the maximum internal dose attributable when no contamination is detected. The last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$) is also reported.

5.7.6. Dosimetric tables for ^{241}Am (reference monitoring procedure)

Reference data:

- subject: reference man
- route of intake and compounds:
 - inhalation of aerosol (AMAD: 1 μm , 5 μm) – compounds: type F, type M and nitrate
 - ingestion - all compounds
- Reference Level for committed effective dose: 10 mSv
- measurement method: alpha spectrometry
- sample analysed: 50 ml of urine
- counting time: 80000 s
- minimum detectable activity: 10 mBq (value normalised to 24 h excretion)
- estimated productivity in 3 days by a single “working line”: 45-50 measurements

Table 5.11: ^{241}Am : inhalation of type F aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 1 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.2 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$4.5 \cdot 10^0$	$8,6 \cdot 10^{-2}$	0.9%
2	$2.0 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$5.0 \cdot 10^0$	$9,5 \cdot 10^{-2}$	1.0%
3	$1.5 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$6.7 \cdot 10^0$	$1,3 \cdot 10^{-1}$	1.3%
4	$1.1 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$9.1 \cdot 10^0$	$1,7 \cdot 10^{-1}$	1.7%
5	$8.1 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.2 \cdot 10^1$	$2,3 \cdot 10^{-1}$	2.3%
6	$6.0 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.7 \cdot 10^1$	$3,2 \cdot 10^{-1}$	3.2%
7	$4.5 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2.2 \cdot 10^1$	$4,2 \cdot 10^{-1}$	4.2%
40 ^(c)	$1.90 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$5.3 \cdot 10^2$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.12:** ^{241}Am : inhalation of type F aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.3 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$7.7 \cdot 10^0$	$8,5 \cdot 10^{-2}$	0.8%
2	$1.1 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$9.1 \cdot 10^0$	$1,0 \cdot 10^{-1}$	1.0%
3	$8.0 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1,2 \cdot 10^1$	$1,4 \cdot 10^{-1}$	1.4%
4	$5.8 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1,7 \cdot 10^1$	$1,9 \cdot 10^{-1}$	1.9%
5	$4.2 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2,4 \cdot 10^1$	$2,6 \cdot 10^{-1}$	2.6%
6	$3.1 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$3,2 \cdot 10^1$	$3,5 \cdot 10^{-1}$	3.5%
7	$2.3 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$4,3 \cdot 10^1$	$4,8 \cdot 10^{-1}$	4.8%
37 ^(c)	$1.1 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$9,1 \cdot 10^2$	$1,0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.13:** ^{241}Am : inhalation of type M aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 1 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$4.6 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2,2 \cdot 10^1$	$3,0 \cdot 10^{-1}$	3.0%
2	$4.3 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2,3 \cdot 10^1$	$3,3 \cdot 10^{-1}$	3.3%
3	$3.4 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2,9 \cdot 10^1$	$4,1 \cdot 10^{-1}$	4.1%
4	$2.6 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$3,9 \cdot 10^1$	$5,4 \cdot 10^{-1}$	5.4%
5	$2.0 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$5,0 \cdot 10^1$	$7,0 \cdot 10^{-1}$	7.0%
6	$1.5 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$6,7 \cdot 10^1$	$9,3 \cdot 10^{-1}$	9.3%
7	$1.2 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$8,3 \cdot 10^1$	$1,2 \cdot 10^0$	12%
250 ^(c)	$1.4 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$7,1 \cdot 10^2$	$1,0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.14: ^{241}Am : inhalation of type M aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ (AMAD = 5 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.7 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$3.7 \cdot 10^1$	$3.0 \cdot 10^{-1}$	3.0%
2	$2.3 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$4.4 \cdot 10^1$	$3.5 \cdot 10^{-1}$	3.5%
3	$1.8 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$5.6 \cdot 10^1$	$4.4 \cdot 10^{-1}$	4.4%
4	$1.3 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$7.7 \cdot 10^1$	$6.2 \cdot 10^{-1}$	6.2%
5	$1.0 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.0 \cdot 10^2$	$8.0 \cdot 10^{-1}$	8%
6	$8.0 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$1.3 \cdot 10^2$	$1.0 \cdot 10^0$	10%
7	$6.4 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$1.6 \cdot 10^2$	$1.3 \cdot 10^0$	13%
220 ^(c)	$8.1 \cdot 10^{-6}$	$1.0 \cdot 10^{-2}$	$1.2 \cdot 10^3$	$9.9 \cdot 10^0$	98.8%

- (a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion
(b) Committed effective dose
(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.15: ^{241}Am : inhalation of nitrate aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 1 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.3 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$7.7 \cdot 10^0$	$1.2 \cdot 10^{-1}$	1.2%
2	$1.2 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$8.3 \cdot 10^0$	$1.3 \cdot 10^{-1}$	1.3%
3	$9.2 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.1 \cdot 10^1$	$1.7 \cdot 10^{-1}$	1.7%
4	$6.8 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.5 \cdot 10^1$	$2.3 \cdot 10^{-1}$	2.4%
5	$5.0 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2.0 \cdot 10^1$	$3.2 \cdot 10^{-1}$	3.2%
6	$3.8 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2.6 \cdot 10^1$	$4.2 \cdot 10^{-1}$	4.2%
7	$2.9 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$3.4 \cdot 10^1$	$5.5 \cdot 10^{-1}$	5.5%
150 ^(c)	$1.6 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$6.2 \cdot 10^2$	$1.0 \cdot 10^1$	100%

- (a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion
(b) Committed effective dose
(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.16: ^{241}Am : inhalation of nitrate aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ (AMAD = 5 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$7.7 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.3 \cdot 10^1$	$1.3 \cdot 10^{-1}$	1.3%
2	$6.5 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$1.5 \cdot 10^1$	$1.5 \cdot 10^{-1}$	1.5%
3	$4.9 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2.0 \cdot 10^1$	$2.0 \cdot 10^{-1}$	2.0%
4	$3.6 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$2.8 \cdot 10^1$	$2.7 \cdot 10^{-1}$	2.7%
5	$2.6 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$3.8 \cdot 10^1$	$3.7 \cdot 10^{-1}$	3.7%
6	$2.0 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$5.0 \cdot 10^1$	$4.8 \cdot 10^{-1}$	4.9%
7	$1.5 \cdot 10^{-4}$	$1.0 \cdot 10^{-2}$	$6.7 \cdot 10^1$	$6.5 \cdot 10^{-1}$	6.5%
100 ^(c)	$9.9 \cdot 10^{-6}$	$1.0 \cdot 10^{-2}$	$1.0 \cdot 10^3$	$9.8 \cdot 10^0$	98%

- (a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion
(b) Committed effective dose
(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.17: ^{241}Am : ingestion (all compounds), $f_A = 5.0 \cdot 10^{-4}$

τ (days)	$m(\tau)$ all compounds	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$3.0 \cdot 10^{-5}$	$1.0 \cdot 10^{-2}$	$3.3 \cdot 10^2$	$2.0 \cdot 10^{-2}$	0.2%
2	$4.6 \cdot 10^{-6}$	$1.0 \cdot 10^{-2}$	$2.2 \cdot 10^3$	$1.3 \cdot 10^{-1}$	1.3%
3	$2.2 \cdot 10^{-6}$	$1.0 \cdot 10^{-2}$	$4.5 \cdot 10^3$	$2.7 \cdot 10^{-1}$	2.7%
4	$1.3 \cdot 10^{-6}$	$1.0 \cdot 10^{-2}$	$7.7 \cdot 10^3$	$4.5 \cdot 10^{-1}$	4.5%
5	$9.2 \cdot 10^{-7}$	$1.0 \cdot 10^{-2}$	$1.1 \cdot 10^4$	$6.4 \cdot 10^{-1}$	6.4%
6	$7.3 \cdot 10^{-7}$	$1.0 \cdot 10^{-2}$	$1.4 \cdot 10^4$	$8.1 \cdot 10^{-1}$	8.1%
7	$6.3 \cdot 10^{-7}$	$1.0 \cdot 10^{-2}$	$1.6 \cdot 10^4$	$9.4 \cdot 10^{-1}$	9.4%
53 ^(c)	$5.9 \cdot 10^{-8}$	$1.0 \cdot 10^{-2}$	$1.7 \cdot 10^5$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

5.8. Alternative monitoring procedure for ^{241}Am

The inadequacy of the reference monitoring procedure for internal contamination by inhalation of highly insoluble compounds (S type) of ^{241}Am associated with plutonium oxide, requires the implementation of an alternative procedure to be applied.

Taking into account the values of parameter $C_{U,241\text{Am},S,Tm}(\tau)$ of Table 5.10, and the consideration above-mentioned on chemical recovery and counting time, the solution proposed here is based on the adoption of a reference level for committed effective dose of 20 mSv and the use of 100 ml urine samples to be prepared using a chemical treatment identical to that proposed for 50 ml samples, obviously with the necessary adjustments to the quantity of reagents to be used.

This solution, decided on the basis of a series of experimental tests carried out to evaluate the possible use of volumes up to 200 ml, it allows the acceptability condition (equation 5.10) to be respected for samples collected within 5 days after the contamination with a slight reduction (no more than 15 % less) of productivity (number of samples processed in a pre-fixed time interval) on the basis of a reference levels still absolutely adequate for an emergency situation [ICRP 96, ICRP 109]

The following are the dosimetry tables for the alternative monitoring procedure compiled on the basis of the same criteria used for the reference one.

5.8.1. Dosimetric tables for ^{241}Am (alternative monitoring procedure)

Reference data:

- > subject: reference man
- > route of intake and compounds: inhalation of aerosol (AMAD: $1 \mu\text{m}$, $5 \mu\text{m}$) – compounds: type S
- > Reference Level for committed effective dose: 20 mSv
- > measurement method: alpha spectrometry
- > sample analysed: 100 ml of urine
- > counting time: 80000 s
- > minimum detectable activity: 5.5 mBq (value normalised to 24 h excretion)
- > estimated productivity in 3 days by a single “working line”: 38-42 measurements

Table 5.18: ^{241}Am : inhalation of type S aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ (AMAD = 1 μm)	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.2 \cdot 10^{-5}$	$5.5 \cdot 10^{-3}$	$2.5 \cdot 10^2$	$7.3 \cdot 10^0$	36%
2	$2.1 \cdot 10^{-5}$	$5.5 \cdot 10^{-3}$	$2.6 \cdot 10^2$	$7.6 \cdot 10^0$	38%
3	$1.6 \cdot 10^{-5}$	$5.5 \cdot 10^{-3}$	$3.4 \cdot 10^2$	$1.0 \cdot 10^1$	50%
4	$1.2 \cdot 10^{-5}$	$5.5 \cdot 10^{-3}$	$4.6 \cdot 10^2$	$1.3 \cdot 10^1$	66%
5 ^(c)	$9.0 \cdot 10^{-6}$	$5.5 \cdot 10^{-3}$	$6.1 \cdot 10^2$	$1.8 \cdot 10^1$	89%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.19:** ^{241}Am : inhalation of type S aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-3}$	$4.2 \cdot 10^2$	$7.2 \cdot 10^0$	36.0%
2	$1.1 \cdot 10^{-5}$	$5.5 \cdot 10^{-3}$	$5.0 \cdot 10^2$	$8.5 \cdot 10^0$	42.5%
3	$8.4 \cdot 10^{-6}$	$5.5 \cdot 10^{-3}$	$6.5 \cdot 10^2$	$1.1 \cdot 10^1$	55.7%
4	$6.3 \cdot 10^{-6}$	$5.5 \cdot 10^{-3}$	$8.7 \cdot 10^2$	$1.5 \cdot 10^1$	74.2%
5 ^(c)	$4.7 \cdot 10^{-6}$	$5.5 \cdot 10^{-3}$	$1.2 \cdot 10^3$	$2.0 \cdot 10^1$	100%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

5.9. Chemical forms assumed by ^{244}Cm (ICRP 141, OIR)

Inhalation

- > aerosol type F – citrate – $f_A = 5.0 \cdot 10^{-4}$
- > aerosols type M – $f_A = 1.0 \cdot 10^{-4}$
- > aerosols type S – $f_A = 5.0 \cdot 10^{-6}$
- > aerosol – oxide, nitrate and chloride – $f_A = 2.5 \cdot 10^{-4}$

Ingestion

- > all compounds – $f_A = 5.0 \cdot 10^{-4}$

5.9.1. Evaluation of the reference monitoring procedure

Relevant parameters

- > Dose Reference level. $L_R = 10 \text{ mSv}$ (committed effective dose)
- > Volume of urine analysed: 50 ml
- > Chemical recovery: 90%
- > Detection efficiency; 33%
- > Counting time. t_m : 80000 s
- > Detection limit. $LD = 4 \text{ counts}$

Condition of acceptability of the procedure:

$$C_{U,RN,S,Tm}(\tau) > LD=4 \text{ counts}$$

$$\Delta\tau_{UC} \geq 5 \text{ days}^{(9)}$$

In order to test the acceptability of the procedure the values of $C_{U,RN,S,Tm}(\tau)$, calculated for all curium compounds listed above, are reported in table 5.2.0 for urine collected at the first seven days following the intake, both by inhalation and ingestion.

Table 5.20: Values of the parameter $C_{U,244Cm,S,Tm}(\tau)$ obtained for ^{244}Cm applying the proposed monitoring procedure to urine collected in the first seven days following the intake

$C_{U,244Cm,S,Tm}(\tau)$ (counts) - Intake of ^{244}Cm -									
τ	Inhalation of aerosol								Ingestion
(days from intake)	Type S		Type M		Type F		oxide/nitrate/chloride		all compounds
(d)	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	
1	8	8	342	352	1361	1379	743	791	5712
2	8	7	319	300	1238	1167	675	669	876
3	6	5	252	234	928	849	540	511	419
4	5	4	193	169	681	615	405	377	248
5	3	3	149	130	501	446	304	280	175
6	3	2	111	104	371	329	230	219	139
7	2	2	89	83	278	244	182	170	120

Analysis of data in Table 5.20 shows immediately that with respect to detection of ^{244}Cm contaminations giving committed effective dose values equal to the reference level of 10 mSv, the proposed procedure:

- is acceptable for inhalation of aerosol of compounds of Type F, compounds of Type M, oxides, nitrates, chlorides and for ingestion (all compounds);
- is not acceptable for inhalation of aerosol of highly insoluble compounds (type S). In this case an alternative procedure must be used.

For all cases for which the reference monitoring procedure is applicable, in the following [dosimetry tables](#) the main dosimetry data (excretion function $m(\tau)$, intake, minimum detectable effective dose (MDD_E) are shown in the hypothesis that the activity of the radionuclide measured in the urine sample (A_m) is equal to MDA. Data are given for collection of urine performed up to the seventh day after the contamination (fifth day as a minimum) and allow to establish the maximum internal dose attributable when no contamination is detected by this procedure. The last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$) is also reported.

⁹ Condition not mandatory but strongly recommended.

5.9.2. Dosimetric tables for ^{244}Cm (reference monitoring procedure)

Reference data:

- subject: reference man
- route of intake and compounds:
 - inhalation of aerosol (AMAD: 1 μm , 5 μm) – compounds: type F (citrate), type M, oxide, chloride, nitrate
 - ingestion - all compounds
- Reference Level for committed effective dose: 10 mSv
- measurement method: alpha spectrometry
- sample analysed: 50 ml of urine
- counting time 80000 s
- minimum detectable activity: 5.0 mBq (value normalised to 24 h excretion)
- estimated productivity in 3 days by a single “working line” : 45-50 measurements

Table 5.21: ^{244}Cm : inhalation of type F aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.2 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$2.3 \cdot 10^0$	$2.7 \cdot 10^{-2}$	0.3%
2	$2.0 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$2.5 \cdot 10^0$	$3.0 \cdot 10^{-2}$	0.3%
3	$1.5 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$3.3 \cdot 10^0$	$4.0 \cdot 10^{-2}$	0.4%
4	$1.1 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$4.5 \cdot 10^0$	$5.5 \cdot 10^{-2}$	0.5%
5	$8.1 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$6.2 \cdot 10^0$	$7.4 \cdot 10^{-2}$	0.7%
6	$6.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$8.3 \cdot 10^0$	$1.0 \cdot 10^{-1}$	1.0%
7	$4.5 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.1 \cdot 10^1$	$1.3 \cdot 10^{-1}$	1.3%
1020 ^(c)	$6.0 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$8.3 \cdot 10^2$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.22: ^{244}Cm : inhalation of type F aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.3 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$3.8 \cdot 10^0$	$2.7 \cdot 10^{-2}$	0.3%
2	$1.1 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$4.5 \cdot 10^0$	$3.2 \cdot 10^{-2}$	0.3%
3	$8.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$6.3 \cdot 10^0$	$4.4 \cdot 10^{-2}$	0.4%
4	$5.8 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$8.6 \cdot 10^0$	$6.0 \cdot 10^{-2}$	0.6%
5	$4.2 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.2 \cdot 10^1$	$8.3 \cdot 10^{-2}$	0.8%
6	$3.1 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.6 \cdot 10^1$	$1.1 \cdot 10^{-1}$	1.1%
7	$2.3 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$2.2 \cdot 10^1$	$1.5 \cdot 10^{-1}$	1.5%
900 ^(c)	$3.5 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$1.4 \cdot 10^3$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.23: ^{244}Cm : inhalation of type M aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$4.6 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.1 \cdot 10^1$	$1.1 \cdot 10^{-1}$	1.1%
2	$4.3 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.2 \cdot 10^1$	$1.2 \cdot 10^{-1}$	1.2%
3	$3.4 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.5 \cdot 10^1$	$1.5 \cdot 10^{-1}$	1.5%
4	$2.6 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.9 \cdot 10^1$	$1.9 \cdot 10^{-1}$	1.9%
5	$2.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$2.5 \cdot 10^1$	$2.5 \cdot 10^{-1}$	2.5%
6	$1.5 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$3.3 \cdot 10^1$	$3.3 \cdot 10^{-1}$	3.3%
7	$1.2 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$4.2 \cdot 10^1$	$4.2 \cdot 10^{-1}$	4.2%
980 ^(c)	$5.0 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$1.0 \cdot 10^3$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.24:** ^{244}Cm : inhalation of type M aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.7 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.9 \cdot 10^1$	$1.1 \cdot 10^{-1}$	1.1%
2	$2.3 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$2.2 \cdot 10^1$	$1.2 \cdot 10^{-1}$	1.2%
3	$1.8 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$2.8 \cdot 10^1$	$1.6 \cdot 10^{-1}$	1.6%
4	$1.3 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$3.8 \cdot 10^1$	$2.2 \cdot 10^{-1}$	2.2%
5	$1.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$5.0 \cdot 10^1$	$2.9 \cdot 10^{-1}$	2.9%
6	$8.0 \cdot 10^{-5}$	$5.0 \cdot 10^{-3}$	$6.3 \cdot 10^1$	$3.6 \cdot 10^{-1}$	3.6%
7	$6.4 \cdot 10^{-5}$	$5.0 \cdot 10^{-3}$	$7.8 \cdot 10^1$	$4.5 \cdot 10^{-1}$	4.5%
830 ^(c)	$2.9 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$1.7 \cdot 10^3$	$9.8 \cdot 10^0$	98%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.25:** ^{244}Cm : inhalation of oxide, nitrate and chloride aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.1 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$4.5 \cdot 10^0$	$5.0 \cdot 10^{-2}$	0.5%
2	$1.0 \cdot 10^{-3}$	$5.0 \cdot 10^{-3}$	$5.0 \cdot 10^0$	$5.5 \cdot 10^{-2}$	0.6%
3	$8.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$6.3 \cdot 10^0$	$6.9 \cdot 10^{-2}$	0.7%
4	$6.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$8.3 \cdot 10^0$	$9.2 \cdot 10^{-2}$	0.9%
5	$4.5 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.1 \cdot 10^1$	$1.2 \cdot 10^{-1}$	1.2%
6	$3.4 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.5 \cdot 10^1$	$1.6 \cdot 10^{-1}$	1.6%
7	$2.7 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.9 \cdot 10^1$	$2.0 \cdot 10^{-1}$	2.0%
1000 ^(c)	$5.5 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$9.1 \cdot 10^2$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.26: ^{244}Cm : inhalation of oxide, nitrate and chloride aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$6.5 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$7.7 \cdot 10^0$	$4.7 \cdot 10^{-2}$	0.5%
2	$5.5 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$9.1 \cdot 10^0$	$5.5 \cdot 10^{-2}$	0.6%
3	$4.2 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.2 \cdot 10^1$	$7.3 \cdot 10^{-2}$	0.7%
4	$3.1 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$1.6 \cdot 10^1$	$9.8 \cdot 10^{-2}$	1.0%
5	$2.3 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$2.2 \cdot 10^1$	$1.3 \cdot 10^{-1}$	1.3%
6	$1.8 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$2.8 \cdot 10^1$	$1.7 \cdot 10^{-1}$	1.7%
7	$1.4 \cdot 10^{-4}$	$5.0 \cdot 10^{-3}$	$3.6 \cdot 10^1$	$2.2 \cdot 10^{-1}$	2.2%
900 ^(c)	$3.1 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$1.6 \cdot 10^3$	$9.8 \cdot 10^0$	98%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.27:** ^{244}Cm : ingestion of all compounds, $f_A = 5.0 \cdot 10^{-4}$

τ (days)	$m(\tau)$ all compounds	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$3.0 \cdot 10^{-5}$	$5.0 \cdot 10^{-3}$	$1.7 \cdot 10^2$	$6.5 \cdot 10^{-3}$	0.1%
2	$4.6 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$1.1 \cdot 10^3$	$4.2 \cdot 10^{-2}$	0.4%
3	$2.2 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$2.3 \cdot 10^3$	$8.9 \cdot 10^{-2}$	0.9%
4	$1.3 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	$3.8 \cdot 10^3$	$1.5 \cdot 10^{-1}$	1.5%
5	$9.2 \cdot 10^{-7}$	$5.0 \cdot 10^{-3}$	$5.4 \cdot 10^3$	$2.1 \cdot 10^{-1}$	2.1%
6	$7.3 \cdot 10^{-7}$	$5.0 \cdot 10^{-3}$	$6.8 \cdot 10^3$	$2.7 \cdot 10^{-1}$	2.7%
7	$6.3 \cdot 10^{-7}$	$5.0 \cdot 10^{-3}$	$7.9 \cdot 10^3$	$3.1 \cdot 10^{-1}$	3.1%
1400 ^(c)	$2.0 \cdot 10^{-8}$	$5.0 \cdot 10^{-3}$	$2.5 \cdot 10^5$	$9.8 \cdot 10^0$	98%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

5.10. Alternative monitoring procedure for ^{244}Cm

The inadequacy of the reference monitoring procedure for internal contamination by inhalation of highly insoluble compounds (S type) of ^{244}Cm , requires the implementation of an alternative procedure to be applied.

Taking into account the values of the parameter $C_{U, 241\text{Am}, S, Tm}(\tau)$ of Table 5.20, the consideration reported above on chemical recovery and counting time and the fact that in the majority of cases contamination by ^{244}Cm is associated with ^{241}Am , the solution here proposed is the same adopted for ^{241}Am : reference level of 20 mSv and use of 100 ml urine samples.

This solution allows the acceptability condition (equation 5.10) to be respected for samples collected within 9 days from the contamination on the basis of a reference levels still absolutely adequate for an emergency situation [ICRP 96, ICRP 109], even if a slight reduction of productivity (no more than 15 % less) must be registered.

The following are the dosimetry tables for the alternative monitoring procedure compiled on the basis of the same criteria used for the reference one.

5.10.1. Dosimetric tables for ^{244}Cm (alternative monitoring procedure)

Reference data:

- > subject: reference man
- > route of intake and compounds: inhalation of aerosol (AMAD: 1 μm , 5 μm) – compounds: type S
- > Reference Level for committed effective dose: 20 mSv
- > measurement method: alpha spectrometry
- > sample analysed: 100 ml of urine
- > counting time: 80000 s
- > minimum detectable activity: 3.0 mBq (value normalised to 24 h excretion)
- > estimated productivity in 3 days by a single “working line” : 38-42 measurements

Table 5.28: ^{244}Cm : inhalation of type S aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.2 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$1.4 \cdot 10^2$	$2.7 \cdot 10^0$	14%
2	$2.1 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$1.4 \cdot 10^2$	$2.9 \cdot 10^0$	14%
3	$1.6 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$1.9 \cdot 10^2$	$3.8 \cdot 10^0$	19%
4	$1.2 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$2.5 \cdot 10^2$	$5.0 \cdot 10^0$	25%
5	$9.0 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$3.3 \cdot 10^2$	$6.7 \cdot 10^0$	33%
6	$6.9 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$4.3 \cdot 10^2$	$8.7 \cdot 10^0$	43%
7	$5.4 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$5.6 \cdot 10^2$	$1.1 \cdot 10^1$	56%
10 ^(c)	$3.1 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$9.7 \cdot 10^2$	$1.9 \cdot 10^1$	97%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.29:** ^{244}Cm : inhalation of type S aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.3 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$2.3 \cdot 10^2$	$2.8 \cdot 10^0$	14%
2	$1.1 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$2.7 \cdot 10^2$	$3.3 \cdot 10^0$	16%
3	$8.4 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$3.6 \cdot 10^2$	$4.3 \cdot 10^0$	21%
4	$6.3 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$4.8 \cdot 10^2$	$5.7 \cdot 10^0$	29%
5	$4.7 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$6.4 \cdot 10^2$	$7.7 \cdot 10^0$	38%
6	$3.6 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$8.3 \cdot 10^2$	$1.0 \cdot 10^1$	50%
7	$2.8 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$1.1 \cdot 10^3$	$1.3 \cdot 10^1$	64%
9 ^(c)	$1.9 \cdot 10^{-6}$	$3.0 \cdot 10^{-3}$	$1.6 \cdot 10^3$	$1.9 \cdot 10^1$	95%

(a) Both values of measured activity (AM) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)5.11. Plutonium ($^{239+240}\text{Pu}$, ^{238}Pu)

Plutonium may be encountered in a large variety of chemical and physical forms, including metal, carbides, hydroxides, oxides (PuO_2 , plutonium-uranium mixed oxide (MOX)), chlorides, oxalates, nitrates, citrates, tri-butyl-phosphate and even in form of $\text{Pu}(\text{NO}_3)_4$ in aqueous solution (widely encountered in nuclear fuel fabrication and reprocessing). This fact joined to its high radio-toxicity makes it probably the most complex radioelement to be monitored for internal dosimetry purposes, especially in emergency situations where it could be difficult to rapidly individuate the actual compound and its solubility and thus to establish a proper monitoring procedure, if any.

As for americium and curium, the reference rapid monitoring procedure proposed here applies a standard analytical method based on column extraction of plutonium isotopes and followed by electrodeposition of Pu isotopes on a steel plate to be measured by alpha spectrometry, the only difference compared to the routine monitoring procedure is the use of small urine samples (reference: 50 ml), and a sufficiently short counting time (up to 80000 s), which makes it possible to achieve useful results in approximately 30-35 hours from the start of sample treatment.

The procedure will be referred only to ^{239}Pu , given that all the results can be immediately transferred to the other two most significant alpha emitting plutonium isotopes (^{238}Pu and ^{240}Pu) with no significant differences.

With adequate supplies of expert personnel and the organisation of proper work shifts, this procedure, or an alternative with comparable performances, allows, when applicable, a single adequately equipped "working line" to complete up to 45-50 measurements of ^{239}Pu in urine in 3 days.

5.11.1. Measurement Methods

Equipment

- > 2 beaker 150 ml volume
- > 1 Hot plate dimension 30 x 40 cm at least.
- > 1 30 ml reservoir in polypropylene for each sample analysed
- > 3 centrifuge tube (50 ml)
- > Vacuum box with internal rack for 50 ml centrifuges tubes, 3 outer tips in polypropylene and 3 inner tips in polyethylene for each sample analysed
- > 1 TEVA 2 ml cartridge (50 -100 μm)
- > 1 position available in an electrodeposition system
- > 1 detection chamber equipped with a 450 mm^2 PIPS detector in alpha spectrometric system.

Reagents (analytical grade)

- > Water, complying with ISO 3696, grade 3
- > ^{242}Pu tracer solution (10 mBq/ml) certified by a metrological institute
- > Concentrated HNO_3 (> 65%)
- > 30% H_2O_2
- > 3M HNO_3 solution
- > NaNO_2
- > 9M HCl solution
- > 2M HCl solution
- > Na_2SO_4 15%_w solution
- > NaHSO_4 5%_w solution
- > $(\text{NH}_4)_2\text{C}_2\text{O}_4$ 2%_w solution
- > KOH 25%_w solution
- > 95% Ethanol

5.11.1.1. Pre-treatment and preparation of the measurement sample

After the addition of 10 mBq of ^{242}Pu as a tracer to a 50 ml urine sample, 10 ml of concentrated HNO_3 and 5 ml of 30% H_2O_2 are used to destroy the organic matter. The solution is evaporated to dryness and the residue is mineralized several times until a white residue is obtained, with 4 ml of

concentrated HNO_3 + 2 ml of 30% H_2O_2 and finally dissolved in 15 ml of 3M HNO_3 solution. The sample is gently warmed on hot plate and 0.3 g of NaNO_2 are added under magnetic stirring until brown fumes disappear.

This solution obtained is loaded onto a TEVA 2 ml cartridge placed on a vacuum box and preconditioned with 10 ml of 3M HNO_3 ; after the sample is drained, a rinse solution of 3M HNO_3 (5 ml) with 2 drops of H_2O_2 30%⁽¹⁰⁾ is used to wash the sample beaker and the cartridge. Then a rinse of 3M HNO_3 (25 ml) and a rinse of 9M HCl (20 ml) are used to elute uranium and thorium isotopes and other matrix components that could interfere with the source preparation. Finally, plutonium is stripped from TEVA using 20 ml of 2M HCl .

The eluate is transferred in a 150 ml beaker, evaporated to dryness and mineralized three times with 4 ml of concentrated HNO_3 + 2 ml of 30% H_2O_2 .

The residue is dissolved by adding 2.5 ml of 5%_w NaHSO_4 , 2 ml of water and 5 ml of 15%_w Na_2SO_4 . Then, the solution is transferred into the electrodeposition cell with three portions of 1 ml water, to rinse the beaker, 1 ml of $(\text{NH}_4)_2\text{C}_2\text{O}_4$ 2%_w is added, then the anode is set up and the electrodeposition carried out at a constant current (about 1.0 A) for 60 minutes.

Finally, 2 ml of 25%_w KOH are added to stop the reaction; after 1 minute the power is turned off, electrode is removed, and the cell is emptied. The electrodeposition disk, which constitutes the real measured sample, is removed from the cell, washed first with water and then with ethanol and left air dry.

5.11.1.2. Measurement of activity and evaluation of chemical recovery

The measurement of activity is performed by alpha spectrometry using ion implanted silicon detectors (PIPS) of 450 mm² active surface. The measurement sample (electrodeposition disk) is positioned inside a vacuum chamber at a distance of 1 mm to the detector's active surface and counted for 80000 s.

Taking into account that alpha ray counting efficiency in vacuum does not depend on alpha ray energy, the evaluation of ^{239}Pu activity in the sample is obtained by comparison to ^{242}Pu applying these simple formulas:

$$A_{239\text{Pu},S} = A_{242\text{Pu},S} \times NC_{239\text{Pu}} / NC_{242\text{Pu}}$$

where: $A_{239\text{Pu},S}$ is the activity of ^{239}Pu in the sample (Bq)
 $A_{242\text{Pu},S}$ is the activity of ^{242}Pu added to the sample (Bq)
 $NC_{239\text{Pu}}$ is the net count in the ROI of ^{239}Pu
 $NC_{242\text{Pu}}$ is the net count in the ROI of ^{242}Pu

The daily urinary excretion of ^{239}Pu activity ($A_{239\text{Pu},d}$ in Bq/24h) is then given by:

$$A_{239\text{Pu},d} = R_u \times A_{239\text{Pu},S} = 32 \times A_{239\text{Pu},S}$$

where R_u is the ratio between the volume of the daily urinary excretion for a Reference Man (1600 ml/day) and the volume of urine analyzed (50 ml).

Chemical recovery, ρ , is evaluated applying the following formula:

¹⁰ Hydrogen peroxide ensures that any U reduced to U(IV) by NaNO_2 is re-oxidized to U(VI) and is not retained on TEVA resin.

$$\rho = NC_{243\text{Am}} / (A_{243\text{Am},S} \times T_m \times Y \times \varepsilon)$$

where T_m is the counting time, ε the detection efficiency and Y the yield of the alpha emissions considered.

5.11.2. Chemical forms assumed by ^{239}Pu (ICRP 141, OIR)

Inhalation

- aerosol type F – $f_A = 5.0 \cdot 10^{-4}$
- aerosols type M – citrate, tri-butyl-phosphate, chloride – $f_A = 1.0 \cdot 10^{-4}$
- aerosols type S – $f_A = 5.0 \cdot 10^{-6}$
- aerosol – dioxide, mixed oxide – $f_A = 2.0 \cdot 10^{-6}$
- aerosol – dioxide 1-nm nanoparticles – $f_A = 3.5 \cdot 10^{-4}$
- aerosol – nitrate – $f_A = 1.0 \cdot 10^{-4}$

Ingestion

- Insoluble forms – $f_A = 1.0 \cdot 10^{-5}$
- Soluble forms – nitrate, chloride, bicarbonate, unidentified chemical forms, $f_A = 5.0 \cdot 10^{-5}$

5.11.3. Evaluation of the reference monitoring procedure

Reference monitoring procedure parameters

- > reference level for committed effective dose, $L_R = 10 \text{ mSv}$
- > time interval for useful collection of urine sample, $\Delta\tau_{UC}$: 5 days
- > activity measurement technique: alpha spectrometry
- > monitoring sample: urine
- > volume of urine analysed: 50 ml
- > recovery tracer: ^{242}Pu (10 mBq/sample)
- > chemical recovery: 80% (a conservative value is applied)
- > counting efficiency: 33% ($\pm 1\%$)
- > counting time: 80000 s
- > typical LD value for ^{239}Pu : 8.5 counts
- > typical MDA value for ^{239}Pu : $1.3 \cdot 10^{-2} \text{ Bq/24h}$
- > duration of the pre-treatment for a single sample: 6 hours
- > number of sample pre-treatment performable simultaneously by a single operator: 8
- > duration of the simultaneous pre-treatment of 8 samples: 8-9 hours
- > time to complete the measurement of 8 samples by a single operator: 32-35 hours
- > estimated productivity in 3 days by a single “working line”: 45-50 measurements

Condition of acceptability of the procedure:

$$C_{U,RN,S,T_m}(\tau) > LD = 8.5 \text{ counts}$$

$$\Delta\tau_{UC} \geq 5 \text{ days}^{(11)}$$

¹¹ Condition not mandatory but strongly recommended

In order to test the acceptability of the procedure the values of $C_{U,RN,S,Tm}(\tau)$, calculated for all plutonium compounds listed above, are reported in table 5.4.0 for urine collected at the first seven days following the intake, both by inhalation and ingestion.

Table 5.30: Values of the parameter $C_{U,239Pu,S,Tm}(\tau)$ obtained for ^{239}Pu applying the proposed monitoring procedure to urine collected in the first seven days following the intake

<i>$C_{U,RN,S,Tm}$ (counts)- Inhalation and Ingestion of ^{239}Pu</i>														
	Inhalation of aerosol												Ingestion	
τ	Type F		Type M		Type S		Dioxide/ mix. oxide		Dioxide. nanopart.		Nitrate		insoluble	soluble
(d)	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm	1 μm	5 μm		
1	33	35	10	10	0.4	0.4	0.1	0.1	26	28	10	11	162	160
2	66	66	19	19	0.8	0.8	0.2	0.2	53	50	20	20	132	132
3	68	66	20	19	0.8	0.8	0.2	0.2	53	50	21	20	72	72
4	58	56	17	17	0.7	0.7	0.2	0.2	46	43	18	17	41	41
5	47	45	15	14	0.6	0.5	0.1	0.1	37	35	15	14	27	28
6	37	35	12	11	0.4	0.4	0.1	0.1	31	28	12	11	20	20
7	29	27	10	9	0.4	0.3	0.1	0.1	24	22	9	9	16	17

From the analysis of data in Table 5.30 it is immediately clear that with respect to detection of ^{239}Pu contaminations giving committed effective dose values equal to the reference level of 10 mSv, the reference procedure proposed here:

- is acceptable for inhalation of aerosol of compounds of Type F, compounds of Type M, dioxide 1-nm nanoparticles and nitrates and for ingestion of all compounds,
- is not acceptable for inhalation of aerosol of insoluble compounds (type S) and highly insoluble compounds (plutonium dioxide and mixed oxides). In this cases, taking into account data from table 5.4.0, separate alternative procedures should be used.

For all cases for which the reference monitoring procedure is applicable, in the following dosimetry tables the main dosimetry data (excretion function $m(\tau)$, intake, minimum detectable effective dose (MDD_E) are shown in the hypothesis that activity of the radionuclide measured in the urine sample (A_m) is equal to MDA. Data are given for collection of urine performed up to the seventh day after the contamination (fifth day as a minimum) and allow to establish the maximum internal dose attributable when no contamination is detected. The last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$) is also reported.

5.11.4. Dosimetric tables for ^{239}Pu (reference monitoring procedure)

Reference data:

- subject: reference man
- route of intake and compounds:
- inhalation of aerosol (AMAD: 1 μm , 5 μm) – compounds: type F, type M, dioxide 1 nm nanoparticles and nitrate,
- ingestion - all compounds
- Reference Level for committed effective dose: 10 mSv
- measurement method: alpha spectrometry

- sample analysed: 50 ml of urine
- counting time: 80000 s
- minimum detectable activity: 13 mBq (value normalised to 24 h excretion)
- estimated productivity in 3 days by a single “working line” : 45-50 measurements

Table 5.31: ^{239}Pu : inhalation of type F aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.7 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$7.6 \cdot 10^1$	$2.6 \cdot 10^0$	26%
2	$3.4 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$3.8 \cdot 10^1$	$1.3 \cdot 10^0$	13%
3	$3.5 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$3.7 \cdot 10^1$	$1.3 \cdot 10^0$	13%
4	$3.0 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$4.3 \cdot 10^1$	$1.5 \cdot 10^0$	15%
5	$2.4 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$5.4 \cdot 10^1$	$1.8 \cdot 10^0$	18%
6	$1.9 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$6.8 \cdot 10^1$	$2.3 \cdot 10^0$	23%
7	$1.5 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$8.6 \cdot 10^1$	$2.9 \cdot 10^0$	29%
15 ^(c)	$4.6 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.8 \cdot 10^2$	$9.5 \cdot 10^0$	95%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.32:** ^{239}Pu : inhalation of type F aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.0 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$1.3 \cdot 10^2$	$2.4 \cdot 10^0$	24%
2	$1.9 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$6.8 \cdot 10^1$	$1.3 \cdot 10^0$	13%
3	$1.9 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$6.8 \cdot 10^1$	$1.3 \cdot 10^0$	13%
4	$1.6 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$8.0 \cdot 10^1$	$1.5 \cdot 10^0$	15%
5	$1.3 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$9.9 \cdot 10^1$	$1.9 \cdot 10^0$	19%
6	$1.0 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$1.3 \cdot 10^2$	$2.4 \cdot 10^0$	24%
7	$7.9 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.6 \cdot 10^2$	$3.1 \cdot 10^0$	31%
15 ^(c)	$2.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$5.2 \cdot 10^2$	$9.8 \cdot 10^0$	98%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.33:** ^{239}Pu : inhalation of type M aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$3.6 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.6 \cdot 10^2$	$8.9 \cdot 10^0$	89%
2	$7.3 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.8 \cdot 10^2$	$4.4 \cdot 10^0$	44%
3	$7.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.7 \cdot 10^2$	$4.3 \cdot 10^0$	43%
4	$6.6 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.0 \cdot 10^2$	$4.9 \cdot 10^0$	49%
5	$5.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.3 \cdot 10^2$	$5.9 \cdot 10^0$	59%
6	$4.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.9 \cdot 10^2$	$7.2 \cdot 10^0$	72%
7	$3.8 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.4 \cdot 10^2$	$8.5 \cdot 10^0$	85%
8 ^(c)	$3.2 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$4.0 \cdot 10^2$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.34: ^{239}Pu : inhalation of type M aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.2 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$5.9 \cdot 10^2$	$8.2 \cdot 10^0$	82%
2	$4.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.1 \cdot 10^2$	$4.4 \cdot 10^0$	44%
3	$4.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.1 \cdot 10^2$	$4.4 \cdot 10^0$	44%
4	$3.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.7 \cdot 10^2$	$5.2 \cdot 10^0$	52%
5	$2.9 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$4.4 \cdot 10^2$	$6.2 \cdot 10^0$	62%
6	$2.4 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$5.4 \cdot 10^2$	$7.5 \cdot 10^0$	75%
7 ^(c)	$2.0 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$6.4 \cdot 10^2$	$9.0 \cdot 10^0$	90%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.35:** ^{239}Pu : inhalation of dioxide 1-nm nanoparticles aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$1.2 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$1.1 \cdot 10^2$	$3.2 \cdot 10^0$	32%
2	$2.4 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$5.4 \cdot 10^1$	$1.6 \cdot 10^0$	16%
3	$2.4 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$5.4 \cdot 10^1$	$1.6 \cdot 10^0$	16%
4	$2.1 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$6.1 \cdot 10^1$	$1.8 \cdot 10^0$	18%
5	$1.7 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$7.6 \cdot 10^1$	$2.3 \cdot 10^0$	23%
6	$1.4 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$9.2 \cdot 10^1$	$2.8 \cdot 10^0$	28%
7	$1.1 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$1.2 \cdot 10^2$	$3.5 \cdot 10^0$	35%
14 ^(c)	$3.9 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.3 \cdot 10^2$	$9.9 \cdot 10^0$	99%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.36:** ^{239}Pu : inhalation of dioxide 1-nm nanoparticles aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$7.3 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.8 \cdot 10^2$	$3.0 \cdot 10^0$	30%
2	$1.3 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$9.9 \cdot 10^1$	$1.7 \cdot 10^0$	17%
3	$1.3 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$9.9 \cdot 10^1$	$1.7 \cdot 10^0$	17%
4	$1.1 \cdot 10^{-4}$	$1.3 \cdot 10^{-2}$	$1.2 \cdot 10^2$	$2.0 \cdot 10^0$	20%
5	$9.0 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.4 \cdot 10^2$	$2.4 \cdot 10^0$	24%
6	$7.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.8 \cdot 10^2$	$3.1 \cdot 10^0$	31%
7	$5.7 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.3 \cdot 10^2$	$3.8 \cdot 10^0$	38%
13 ^(c)	$2.2 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$5.9 \cdot 10^2$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.37: ^{239}Pu : inhalation of nitrate aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$3.5 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.7 \cdot 10^2$	$8.5 \cdot 10^0$	85%
2	$7.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.8 \cdot 10^2$	$4.2 \cdot 10^0$	42%
3	$7.2 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$1.8 \cdot 10^2$	$4.1 \cdot 10^0$	41%
4	$6.2 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.1 \cdot 10^2$	$4.8 \cdot 10^0$	48%
5	$5.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$2.5 \cdot 10^2$	$5.8 \cdot 10^0$	58%
6	$4.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.1 \cdot 10^2$	$7.2 \cdot 10^0$	72%
7 ^(c)	$3.3 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.9 \cdot 10^2$	$9.0 \cdot 10^0$	90%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.38:** ^{239}Pu : inhalation of nitrate aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$6.1 \cdot 10^2$	$8.0 \cdot 10^0$	80%
2	$3.9 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.3 \cdot 10^2$	$4.3 \cdot 10^0$	43%
3	$3.9 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.3 \cdot 10^2$	$4.3 \cdot 10^0$	43%
4	$3.3 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$3.9 \cdot 10^2$	$5.1 \cdot 10^0$	51%
5	$2.7 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$4.8 \cdot 10^2$	$6.2 \cdot 10^0$	62%
6	$2.1 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$6.1 \cdot 10^2$	$8.0 \cdot 10^0$	80%
7 ^(c)	$1.7 \cdot 10^{-5}$	$1.3 \cdot 10^{-2}$	$7.6 \cdot 10^2$	$9.8 \cdot 10^0$	98%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection

Table 5.39: ^{239}Pu : ingestion of soluble forms, $f_A = 5.0 \cdot 10^{-5}$

τ (days)	$m(\tau)$ soluble forms	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$2.9 \cdot 10^{-6}$	$1.3 \cdot 10^{-2}$	$4.4 \cdot 10^3$	$5.31 \cdot 10^{-1}$	5%
2	$2.4 \cdot 10^{-6}$	$1.3 \cdot 10^{-2}$	$5.4 \cdot 10^3$	$6.41 \cdot 10^{-1}$	6%
3	$1.3 \cdot 10^{-6}$	$1.3 \cdot 10^{-2}$	$9.9 \cdot 10^3$	$1.2 \cdot 10^0$	12%
4	$7.5 \cdot 10^{-7}$	$1.3 \cdot 10^{-2}$	$1.7 \cdot 10^4$	$2.1 \cdot 10^0$	21%
5	$5.0 \cdot 10^{-7}$	$1.3 \cdot 10^{-2}$	$2.6 \cdot 10^4$	$3.1 \cdot 10^0$	31%
6	$3.7 \cdot 10^{-7}$	$1.3 \cdot 10^{-2}$	$3.5 \cdot 10^4$	$4.2 \cdot 10^0$	42%
7	$3.0 \cdot 10^{-7}$	$1.3 \cdot 10^{-2}$	$4.3 \cdot 10^4$	$5.2 \cdot 10^0$	52%
15 ^(c)	$1.6 \cdot 10^{-7}$	$1.3 \cdot 10^{-2}$	$8.0 \cdot 10^4$	$9.7 \cdot 10^0$	97%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.40: ^{239}Pu : ingestion of insoluble forms, $f_A = 1.0 \cdot 10^{-5}$

τ (days)	$m(\tau)$ insoluble forms	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$5.9 \cdot 10^{-8}$	$1.3 \cdot 10^{-2}$	$2.2 \cdot 10^5$	$5.2 \cdot 10^{-1}$	5%
2	$4.8 \cdot 10^{-8}$	$1.3 \cdot 10^{-2}$	$2.7 \cdot 10^5$	$6.4 \cdot 10^{-1}$	6%
3	$2.6 \cdot 10^{-8}$	$1.3 \cdot 10^{-2}$	$5.0 \cdot 10^5$	$1.2 \cdot 10^0$	12%
4	$1.5 \cdot 10^{-8}$	$1.3 \cdot 10^{-2}$	$8.6 \cdot 10^5$	$2.1 \cdot 10^0$	21%
5	$9.9 \cdot 10^{-9}$	$1.3 \cdot 10^{-2}$	$1.3 \cdot 10^6$	$3.1 \cdot 10^0$	31%
6	$7.3 \cdot 10^{-9}$	$1.3 \cdot 10^{-2}$	$1.8 \cdot 10^6$	$4.2 \cdot 10^0$	42%
7	$5.9 \cdot 10^{-9}$	$1.3 \cdot 10^{-2}$	$2.2 \cdot 10^6$	$5.2 \cdot 10^0$	52%
15 ^(c)	$3.2 \cdot 10^{-9}$	$1.3 \cdot 10^{-2}$	$4.0 \cdot 10^6$	$9.7 \cdot 10^0$	97%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Last day from intake useful for urine collection

5.12. Alternative monitoring procedure for ^{239}Pu : inhalation of aerosol of type S compounds

The procedure proposed here is based on the adoption of a reference level of committed effective dose of 100 mSv and the use of 100 ml urine samples to be prepared applying a chemical treatment identical to that used for 50 ml samples, just making the necessary adjustments to the quantity of reagents to be used. This solution, allows the acceptability condition (equation 5.10) to be respected for samples collected within 5-6 days from the contamination, first day excluded, maintaining a satisfactory productivity (30-35 measurement in 3 days) on the basis of a reference level which is surely high but still acceptable for an emergency situation. In order to make useful even urine produced within 24 hours after the contamination, a value for L_R of 120 mSv and 110 mSv should be respectively adopted for AMAD of $1 \mu\text{m}$ and $5 \mu\text{m}$.

The following are the dosimetry tables for the alternative monitoring procedure compiled on the basis of the same criteria used for the reference one.

5.12.1. Dosimetric tables for inhalation of aerosol of type S compounds of ^{239}Pu

Reference data:

- > subject: reference man
- > route of intake and compounds: inhalation of aerosol (AMAD: $1 \mu\text{m}$, $5 \mu\text{m}$) – compounds: type S
- > Reference Level for committed effective dose: 120 mSv
- > measurement method: alpha spectrometry
- > sample analysed: 100 ml of urine
- > counting time: 80000 s
- > minimum detectable activity: 7.0 mBq (value normalised to 24 h excretion)
- > estimated productivity in 3 days by a single “working line” : 35-40 measurements

Table 5.41: ^{239}Pu : inhalation of type S aerosol, AMAD = 1 μm , reference level = 120 mSv

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1 ^(c)	$1.8 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$3.9 \cdot 10^3$	$1.2 \cdot 10^2$	100%
2	$3.6 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$1.9 \cdot 10^3$	$6.0 \cdot 10^1$	50%
3	$3.6 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$1.9 \cdot 10^3$	$6.0 \cdot 10^1$	50%
4	$3.1 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$2.3 \cdot 10^3$	$7.0 \cdot 10^1$	58%
5	$2.6 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$2.7 \cdot 10^3$	$8.3 \cdot 10^1$	70%
6 ^(d)	$2.1 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$3.3 \cdot 10^3$	$1.0 \cdot 10^2$	86%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Acceptable only for reference level of 120 mSv

(d) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)**Table 5.42:** ^{239}Pu : inhalation of type S aerosol, AMAD = 5 μm , reference level = 110 mSv

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1 ^(c)	$1.1 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$6.4 \cdot 10^3$	$1.1 \cdot 10^2$	98%
2	$2.0 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$3.5 \cdot 10^3$	$6.0 \cdot 10^1$	54%
3	$2.0 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$3.5 \cdot 10^3$	$6.0 \cdot 10^1$	54%
4	$1.7 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$4.1 \cdot 10^3$	$7.0 \cdot 10^1$	63%
5	$1.4 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$5.0 \cdot 10^3$	$8.5 \cdot 10^1$	77%
6 ^(d)	$1.1 \cdot 10^{-6}$	$7.0 \cdot 10^{-3}$	$6.4 \cdot 10^3$	$1.1 \cdot 10^2$	98%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Acceptable only for reference level of 110 mSv

(d) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Another choice could be to increase the volume of urine to be analysed, although a real improvement in terms of reducing the minimum detectable doses requires moving to larger urine volumes (200 ml or more) which would lead to significantly longer treatment durations, notable drop in productivity (40-50% less) and higher variability in chemical recovery.

5.13. Alternative monitoring procedure for ^{239}Pu : aerosol inhalation of Pu dioxide or mixed oxides

Using applying a monitoring procedure based on alpha spectrometry measurement of urine samples, the possibility to obtain useful results for intake of these compounds in a short time (no more than 2 days) makes it mandatory to use a much higher quantity of urine (200 ml at least) and to adopt a committed effective dose reference level significantly over 100 mSv.

However, such a monitoring procedure set up in this way would however be difficult to accept, first because of the possibility of neglecting excessively high dose levels (it should be noted that such choices would even lead to reference levels well above 1 Sv for the dose equivalent to a specific organ), but also from a practical point of view, taking into account the low productivity that can be achieved and the limited time available for useful urine collection.

The following dosimetry tables show the results obtained by adopting these choices on the base of the same measurement method already used for ^{239}Pu in the previous conditions and properly adapted for a higher volume of urine.

5.13.1. Dosimetry tables for inhalation of aerosol of ^{239}Pu dioxide or mixed oxides

Reference data:

- > subject: reference man
- > route of intake and compounds: inhalation of aerosol (AMAD: 1 μm , 5 μm) – compounds: dioxide and mixed oxides
- > Reference Level for committed effective dose: 200 mSv
- > measurement method: alpha spectrometry
- > sample analysed: 200 ml of urine
- > counting time: 80000 s
- > minimum detectable activity: 4.2 mBq (value normalised to 24 h excretion)
- > estimated productivity in 3 days by a single “working line” : 20-25 measurements

Table 5.43: ^{239}Pu : inhalation of dioxide/mixed oxide aerosol, AMAD = 1 μm , reference level = 200 mSv

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1 ^(c)	$6.9 \cdot 10^{-7}$	$4.2 \cdot 10^{-3}$	$6.1 \cdot 10^3$	$2.7 \cdot 10^2$	137%
2	$1.4 \cdot 10^{-6}$	$4.2 \cdot 10^{-3}$	$3.0 \cdot 10^3$	$1.4 \cdot 10^2$	68%
3	$1.4 \cdot 10^{-6}$	$4.2 \cdot 10^{-3}$	$3.0 \cdot 10^3$	$1.4 \cdot 10^2$	68%
4 ^(d)	$1.2 \cdot 10^{-6}$	$4.2 \cdot 10^{-3}$	$3.5 \cdot 10^3$	$1.6 \cdot 10^2$	79%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Sample collection time not acceptable

(d) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

Table 5.44: ^{239}Pu : inhalation of dioxide/mixed oxide aerosol, AMAD = 5 μm , reference level = 200 mSv

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1 ^(c)	$4.2 \cdot 10^{-7}$	$4.2 \cdot 10^{-3}$	$1.0 \cdot 10^4$	$2.5 \cdot 10^2$	125%
2	$7.7 \cdot 10^{-7}$	$4.2 \cdot 10^{-3}$	$5.5 \cdot 10^3$	$1.4 \cdot 10^2$	68%
3	$7.6 \cdot 10^{-7}$	$4.2 \cdot 10^{-3}$	$5.5 \cdot 10^3$	$1.4 \cdot 10^2$	69%
4 ^(d)	$6.4 \cdot 10^{-7}$	$4.2 \cdot 10^{-3}$	$6.6 \cdot 10^3$	$1.6 \cdot 10^2$	82%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion

(b) Committed effective dose

(c) Sample collection time not acceptable

(d) Last day from intake useful for urine collection (upper end of $\Delta\tau_{UC}$)

5.14. Additional considerations on rapid monitoring of internal contamination by inhalation of ^{239}Pu in form of insoluble or highly insoluble compounds

The analysis just presented, highlighted that the proposed reference procedure, and its various options, is more than satisfactory for all forms of plutonium, except for inhalation of insoluble S-type compounds or highly insoluble oxides. For them, it is in fact necessary to move to a quantity of urine to be analysed that is intermediate between a small monitoring sample and a standard sample and, above all, to L_R values that are probably too high in terms of risk assessment, raising serious problems of acceptability, especially if they have to be applied to the general population. This problem is particularly significant given that this type of compounds is very common in nuclear waste, and could therefore easily be involved in accidents or threatening events.

An alternative for monitoring inhalation of high insoluble plutonium compounds should be found in the use of mass spectrometry, ICP-MS. The procedure still requires a pre-treatment for extraction of the analyte investigated and a purification of the sample, in particular from ^{238}U . For committed effective dose reference levels of 10 mSv, the type of mass-spectrometer to be used must give the possibility to achieve a detection limit, expressed in terms of mass concentration of ^{239}Pu in urine, of the order of 5.0×10^{-14} g/l (50 fg/l), that is a daily urinary excretion of ^{239}Pu of the order of 0.20 mBq/24h (obviously all these values must be corrected by a factor “n” if a reference level of $n \times 10$ mSv is adopted).

From this perspective, it must be observed that the use of ICP-MS type mass spectrometry is not feasible. The typical “sensitivity” of the instrument for elements with atomic mass of this type is in fact of the order of 10^{-10} g/L (i.e. 1000000 fg/L), widely suitable for monitoring internal contamination from Uranium (^{238}U and ^{235}U) and natural Thorium (^{232}Th), but evidently far insufficient for radionuclides with much higher specific activity such as ^{239}Pu .

A proper solution can be found in the use of much more sophisticated mass spectrometers like QICP-MS [Xiao 2023], ICP-SFMS [M.V. Zoriy-2004], multi-collector ICP-MS [Parrish 2006]. However, these instruments are much more expensive, less widespread and, above all, not commonly used in radio-toxicology laboratories, a fact which raises many doubts about their use in emergency situations, where familiarity with the analytical methods and the instrumentation used, in relation to the type of the matrices and the purpose of the measurements themselves, is fundamental to be able to operate with the rapidity and speed, that a very particular situation like this requires.

The key point of the whole issue lies in the already argued need to use urine samples, when by far the most suitable sample for the monitoring of this type of contaminations, especially in special or emergency situations, a real “golden standard”, would be faeces, even a small fraction of their total daily excretion.

As a proof of this, in the following dosimetry tables 5.45 and 5.46, using the same approach as for urine, the performances achieved with the use of alpha spectrometry on a very small faeces sample are reported for the most complex case (inhalation of ^{239}Pu aerosol in the form of dioxide or oxide mixture) In this regard it is important to underline that the treatment method (not specified here) used for the purpose of preparing the “measurement sample” must guarantee a stable chemical recovery of no less than 50% (a value easily obtainable with any standard method specifically adapted to the quantity of sample to be analysed) and good purification from any interfering materials. The specifications to outline an appropriate and feasible monitoring procedure are reported below.

Reference data:

- > subject: reference man
- > route of intake and compounds: inhalation of aerosol (AMAD: 1 μm , 5 μm) – compounds: dioxide and mixed oxides
- > Reference Level, LR, for committed effective dose: 10 mSv
- > measurement method: alpha spectrometry of faecal ashes
- > daily faecal excretion (ashes obtain by calcination in a muffle furnace): 4.0 g
- > sample analysed: 100 mg of faecal ashes
- > chemical recovery: 50% (treatment not described)
- > counting time: 36000 s
- > counting efficiency: 33%

- > Detection Limit, LD: 12 counts
- > minimum detectable activity: 0.81 Bq (value normalised to daily fecal excretion)

Table 5.45: ^{239}Pu (by faeces): inhalation of dioxide/mixed oxide aerosol, AMAD = 1 μm

τ (days)	$m(\tau)$ AMAD = 1 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$4.4 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$1.8 \cdot 10^1$	$8.3 \cdot 10^{-1}$	8.3%
2	$1.4 \cdot 10^{-1}$	$8.1 \cdot 10^{-1}$	$5.8 \cdot 10^0$	$2.6 \cdot 10^{-1}$	2.6%
3	$9.2 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$8.8 \cdot 10^0$	$4.0 \cdot 10^{-1}$	4.0%
4	$3.6 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$2.2 \cdot 10^1$	$1.0 \cdot 10^0$	10.1%
5	$1.2 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$6.7 \cdot 10^1$	$3.0 \cdot 10^0$	30.3%
6 ^(c)	$4.1 \cdot 10^{-3}$	$8.1 \cdot 10^{-1}$	$2.0 \cdot 10^2$	$8.9 \cdot 10^0$	89%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion (4 g faecal ashes)

(b) Committed effective dose

(c) Last day from intake useful for faeces collection (upper end of $\Delta\tau_{UC}$)

Table 5.46: ^{239}Pu (by faeces): inhalation of dioxide/mixed oxide aerosol, AMAD = 5 μm

τ (days)	$m(\tau)$ AMAD = 5 μm	$A_M = \text{MDA}^{(a)}$ (Bq/24h)	Intake (Bq)	$E_{50} = \text{MDD}_E(\tau)^{(b)}$ (mSv)	Ratio E_{50}/L_R
1	$8.5 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$9.5 \cdot 10^0$	$2.4 \cdot 10^{-1}$	2.4%
2	$2.7 \cdot 10^{-1}$	$8.1 \cdot 10^{-1}$	$3.0 \cdot 10^0$	$7.5 \cdot 10^{-2}$	0.7%
3	$1.8 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$4.5 \cdot 10^0$	$1.1 \cdot 10^{-1}$	1.1%
4	$6.7 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$1.2 \cdot 10^1$	$3.0 \cdot 10^{-1}$	3.0%
5	$2.0 \cdot 10^{-2}$	$8.1 \cdot 10^{-1}$	$4.0 \cdot 10^1$	$1.0 \cdot 10^0$	10%
6	$5.7 \cdot 10^{-3}$	$8.1 \cdot 10^{-1}$	$1.4 \cdot 10^2$	$3.5 \cdot 10^0$	35%
7 ^(c)	$2.0 \cdot 10^{-3}$	$8.1 \cdot 10^{-1}$	$4.0 \cdot 10^2$	$1.0 \cdot 10^1$	100%

(a) Both values of measured activity (A_M) and MDA are normalised to 24h excretion (4 g faecal ashes)

(b) Committed effective dose

(c) Last day from intake useful for faeces collection (upper end of $\Delta\tau_{UC}$)

It is very clear that the use of faeces in any case allows a much more conservative monitoring approach in terms of risk assessment and thus much more appropriate for managing the psychological and social impact of the contamination event, even assuming very precautionary values on the daily faecal excretion, on the chemical recovery and on Detection Limit (with respect to the short counting time). Moreover, despite the long time required for the first phase of calcination of the samples, the very significant reduction of the counting time with respect to urine samples allows a productivity to be similar to that obtained by analysing 200 ml of urine (20-25 samples in 3 days). From this point of view, it's important to underline that, with an appropriate number of large furnace-muffles available (at least three), the number of analyses completed by a single working line can increase significantly up to a hundred per week, if the monitoring is prolonged over time.

The real problem, which makes the use of faecal samples extremely difficult, or even quite impossible, is mainly linked to the huge problems associated with the collection, storing, transport, and management of this kind of samples, especially when it has to be produced in a very large numbers by members of the public. The use of faeces could be probably feasible if the contamination event is restricted to people employed in "nuclear activities" (in a broad sense) or, in any case, if the number of people to be monitored does not exceed a few dozen (at maximum a hundred or a little more). In these cases, it is still necessary to have several working lines readily available, such as several prepared and adequately equipped laboratories (personnel included), to have a real chance of successfully dealing with the scenario.

As will be shown in the following paragraph, a possible solution, albeit “indirect”, to solve the inherent problems of monitoring intakes of insoluble plutonium compounds, can be found when these contaminants are part of a mixed source. Furthermore, this solution is suitable in most cases where standard measurement methods are difficult to adapt to the needs of emergency situations.

5.14.1. Mixed sources

In principle, it should be the most complex case to deal with, since, at least on a theoretical level, it requires the implementation and execution of different measurement methods specific to each radionuclide, or group of radionuclides listed in the mixture. This could make it very difficult, if not impossible, to carry out monitoring in a timely manner, especially when the number of people to be controlled is very high.

This issue can be greatly simplified if a single radionuclide, or a restricted group of those present in the mixture, is responsible for most of the dose, making the others practically negligible in terms of dose, or if one of them, which is easy to be measured, can be used as a "guide" for the entire set of radionuclides involved, or for a group of them, at least for an initial dose assessment [EC RP 188].

The possibility of identifying and selecting a "guide radionuclide" (or more) can not only reduce the type and number of measurements to be carried out, simplifying, and lightening the monitoring procedure, but can also allow a reduction of the minimum detectable dose levels for those radionuclides, or some of their specific compounds, for which rapid measurement methods can only be applied by adopting very high reference levels. To apply this strategy, it is obviously strictly necessary to have knowledge of the qualitative and quantitative composition of the radionuclides mixture, which can be verified, or directly obtained, through radiological characterization measurements of environmental samples such as material deposited on inert surfaces or airborne particulate, if it can be sampled promptly.

Taking into account the peculiarities of each component with respect to the actual dispersion way of contamination in the environment, the aforementioned characterization should be coupled with a series of proper measurements of each specific radionuclide present, to be carried out on a restricted number of people involved (control group). This allows (or should allow) to have an information on the stability of the activity ratio (in percentage, AR%) between each radionuclide involved and the guide radionuclide, as well as the correspondence between the original composition of the radionuclides mixture and that actually "available" for incorporation at the receptor. In this regards it is important to point out that this last evaluation must be done not directly through results of the measurements performed on the biological samples, but through the intakes estimated by them. Given these values are strongly affected by intersubjective variability of metabolic behaviour, only statistical values (mean value, sample standard deviation) must be used for this kind of evaluations and outcomes should anyway be taken very carefully.

A test on AR% aimed to identify and verify the possible use of guide radionuclides (one or more) in a mixed source should be structured as it follows:

- execute a series of measurements on environmental samples in order to have a first experimental information on the qualitative and quantitative composition of the mixed source (action in any case mandatory to plan the monitoring)
- verify data obtained through the characterisation by comparison with the nominal composition of the contaminating material (when available);

- on the basis of the chemical and physical characteristics of the radionuclides present in the mixture, identify the possible guide radionuclide, RNG, taking also into account the complexity of its (rapid) method of measurement together with its weight in the mixture in terms of activity and radiotoxicity (contribution to total dose);
- on the basis of the set of measurements performed on environmental samples, for each radionuclide found (RND1, RND2, ..., RNDn) for which RNG will be the guide, evaluate the mean value of AR% with respect to the guide radionuclide and its associated sample standard deviation;
- once monitoring of the people involved by the contamination is started by the measurement of RNG, perform on a restricted number (Np) of persons (control group) also the measurement of RND1, RND2, ..., RNDn, each one by its specific method (it is suggested to limit this step only to radionuclides most significant in terms of dose);
- calculate intakes of RND1, RND2, ..., RNDn, by the results obtained applying the specific measurement, taking into account the possible chemical forms of each of them in light of the nature and origin of the mixture. In case of lack of information, firstly assume the most conservative hypotheses;
- evaluate the individual values of AR% for radionuclide RND1, RND2, ..., RNDn with respect to RNG and calculate for each set of Np data the mean value and the sample standard deviation;
- for each RNDi compare the mean values of AR% obtained respectively by environmental measurements and by intakes evaluated through internal contamination measurement, if necessary evaluating the possibility to minimize differences by changing the choices previously made on chemical form for the various radionuclides;
- after this:
 - if the residual difference between mean values is not significant at level 20%, prosecute the monitoring only by measurement of RNG - intake of RNDi will be obtained on the basis of the intake of RNG evaluated by measurement, corrected by AR% equal to the arithmetic mean of the 2 mean values previously compared
 - if the residual difference between mean values is greater than 20%, but the use of the more conservative AR% results in a total dose below the reference level, prosecute the monitoring only by measurement of RNG - intake of RNDi will be calculated on the basis of intake of RNG, obtained by measurement, corrected by the more conservative mean value of AR%
 - if the difference between mean values is greater than 20%, and the use of the more conservative AR% results in a dose above the reference level, RNG must be considered not eligible as a guide for RNDi - prosecute the monitoring performing also the direct measurement of RNDi (intake of RNDi and the corresponding dose will be obviously calculated on the basis of the result of this measurement).

For a more immediate understanding on the possible role of guide radionuclides in monitoring internal contamination by mixed sources and the strategies that can be adopted for an appropriate use of them, a series of example cases are presented below. This presentation will be preceded by a short list of radionuclides, or categories of radionuclides, for which easy and rapid measurement methods useful for monitoring widespread contamination are already available (such as gamma emitters or tritium). When applicable, these methods, briefly described here, can obviously generally be used to perform monitoring, but possibly also to identify and select a specific radionuclide as a guide to effectively address mass contamination from mixed sources

5.14.2. Gamma emitting radionuclides.

From a technical point of view, gamma emitting radionuclides present the minor difficulties for a mass internal contamination monitoring. Both in terms of sensibility and measurement capability, very simple mobile in vivo counters [Castagnet 2007 - Ciciani 2021]) provide in-fact suitable solutions for a large part of these radionuclides. These instruments apparatuses have limited set-up costs, and composed of a gamma spectrometric chain connected to a single detector (type NaI(Tl) or HPGe) which is commonly mounted on a mechanic stand proper for its positioning. Their ease of transport also allows for a rapid set up of field laboratories and can be mounted on mobile units making it possible to conduct in situ measurements without the enormous logistical obstacle of needing a fixed whole-body counter, which is likely to be far from the event site. The simple apparatuses shown in Figure 5.1, for example, make it possible to obtain MDA values approximately of 10 kBq in the whole body for ^{137}Cs and ^{60}Co and 0.5 kBq in the thyroid for ^{131}I with a counting time of 100 s.



Figure 5.1: Example of mobile in vivo counters (whole body counter on the left, thyroid counter on the right)

These MDA values allow for the detection of intakes corresponding to committed effective dose of the order of 0.5 mSv for each of the three listed radionuclides and for each route of intake by measurements performed within 10 days from the contamination. Adopting a conservative approach which takes into account inter-subject variability of retention functions and assuming as reasonable to complete a single measurement in 3 minutes, it follows that by using just one *in vivo* monitoring system like this, 200-300 people per day can be monitored on the base of a Reference Level of 1 mSv.

Alternatively, the monitoring of gamma emitters can be performed by the measurement of urine samples by gamma spectrometry. The measurement is still very simple given that no treatment of the sample is necessary but to put it in a proper plastic container reproducing already calibrated standard measurement geometry. The most common of them is the so called “Marinelli beaker”, available for volumes ranging from 200 ml to 2l. Especially for small urine samples (up to 100 ml), simple cylindrical containers can be used, possibly with a large base surface (not exceeding detector diameter) in order to increase counting efficiency.

Using a common p-type HPGe detector having 35-40% relative efficiency and assuming as a reference geometry the 500 ml Marinelli beaker, a minimum detectable activity of the order of a few

Bq/l of ^{137}Cs can be easily achieved by 60 minutes counting time (more or less the same for ^{60}Co , ^{131}I and in general all radionuclides emitting gamma radiation of energy higher than 200 keV having a yield above 50%). The corresponding committed effective dose levels strictly depend on the solubility of the contaminant (inhalation type) and can vary for the same radionuclide even by orders of magnitude. For the most common situations, the Minimum Detectable Dose values obtainable by this method for ^{137}Cs and ^{131}I are of the order of a few μSv for soluble compounds (type F for inhalation) that are largely the most probable, while for ^{60}Co they are of the order of tenths of μSv for type M compounds and of 1 mSv for type S compounds.

If it is appropriate to the specific case, gamma spectrometry of urine appears as a very good method to be adopted in emergency situations and it is surely a powerful alternative especially when organisation and logistical problems make it difficult or quite impossible the use of mobile in vivo counters. The only weakness of the method could be found in the limited number of urine sample executable per day by a single spectrometric chain with respect to in-vivo counting.

5.14.3. Uranium (natural, depleted or enriched)

For uranium, be it natural (NU), enriched (EU) or depleted (DU), a monitoring method commonly used in normal radioprotection practice can be adopted. It consists in the measurement by ICP mass spectrometry of a few millilitres of urine pre-treated through a simple dilution with acidified water. With the exception of the quantity of sample collected (normally the urine analysed is a portion of the 24-hour collection), the procedure does not require any particular modifications to be used in emergency situations and allows dozens of samples per day to be measured with Minimum Detectable Concentration (MDC) of about 4.5 ng/24h for (i.e. $5.5 \cdot 10^{-5}$ Bq/24h) and 0.65 ng/24h for ^{235}U (i.e. $5.2 \cdot 10^{-5}$ Bq/24h)) with quadrupole ICP-MS. These values are about one order of magnitude lower than the corresponding MDC obtainable by standard alpha spectrometry and would theoretically be suitable for detecting contamination levels corresponding to a committed effective dose well below 1 mSv for any compound and route of intake, even when the urine sample is collected more than a week after an acute intake.

Given that, in general, uranium appears unlikely as a significant source of radioactive contamination involving a large number of people because of its very low specific activity and its high density that strongly obstacles a wide diffusion in the environment, the first problem that in any case arises in uranium monitoring uranium is given by the dosimetry interpretation of the concentration data measured in excreta. It is in fact necessary to distinguish the contribution deriving from contamination (contamination fraction) from that due to naturally occurring uranium (background fraction), almost always present in measurable and extremely variable quantities, mainly due to the composition and origin of the foodstuff, drinking water included. Given that uranium concentration in urine due to natural background normally ranges between 1 and 100 ng/l (somewhere up to 3-4 hundred of ng/l), a value of 150 ng/l of NU in urine is here conservatively taken as the reference background.

In all cases of contamination by uranium mixtures with not-natural composition (EU and DU), the discrimination and quantification of the two fractions can be obtained by determining the ratio between mass concentration of ^{235}U and ^{238}U ($R_{M,235/238}$) in the monitoring sample and comparing it with respect to the characteristic value of UN ($R_{M,235/238} = 0.711\%$) [UNSCEAR 2016]. From this point of view, it is obviously essential the prior knowledge of the degree of enrichment, or depletion of ^{235}U in contaminating mixture, information that can be easily obtained from smear test measurements of the contaminating material available as a deposition at the place involved by the contamination.

In the impossibility of considering all cases, in this treatment reference values of 3% enrichment and 0.2% depletion of ^{235}U [UNEP, 2001] are assumed for EU and DU, respectively. Correspondently ^{234}U is taken as enriched by a factor of 10 and depleted by a factor of 5.

By these assumptions and taking into account the uncertainties associated to ^{235}U value measured by ICP-MS, for DU an absolute reliable discrimination of the contamination fraction can be conservatively put at 100 ng/l (2 mBq/24h of ^{238}U). This choice allows to apply for DU a Reference Level of 3 mSv up to 7 days after contamination.

As long as EU is concerned, ICP-MS measurement allows through the determination of the concentration of ^{235}U and ^{238}U and their mass ratio, to highlight, contributions to the excretion of a few tens of nanograms of EU also for enrichments of 3-4% (in this regard it is important to highlight that, all other conditions being equal, the higher the enrichment, the smaller the amount of EU contribution that can be discriminated). Limiting the evaluations to an enrichment of 3%, in presence of a background fraction up to 100 ng/l of NU, the minimum contribution by EU to the excretion can be estimated at a level of 20 ng/l ($4.0 \cdot 10^{-4}$ Bq/24h of ^{238}U). Taking into account that for dosimetry evaluations it is also essential to know the enrichment of ^{234}U , which in such cases can give by far the main contribution to the dose, assuming an enrichment process that leads to a $^{234}\text{U}/^{238}\text{U}$ activity ratio equal to 10 (i.e. $4.0 \cdot 10^{-3}$ Bq/24h of ^{234}U), in the worst, but most probable hypothesis (inhalation of insoluble compounds), the contribution to the excretion of 20 ng/l of EU at 3%, corresponds to committed effective doses lower than 4 mSv for about one week. For emergency monitoring of internal contamination by EU a Reference Level of 5 mSv can thus comfortably be adopted.

For NU, being not feasible the use of the composition of the isotopic mixture (particularly the $^{235}\text{U}/^{238}\text{U}$ ratio) as a parameter for discriminating contamination fraction from the background fraction, in mass screening it can be assumed, for simplicity, to determine the concentration of total uranium in daily urine taking a value of 150 ng/l (3.0 mBq/24h of ^{238}U) as a threshold for ascertaining the presence of an actual contamination. This choice allows us to considerably limit the number of cases of false positive and at the same time highlight all cases of intake corresponding to doses above 5 mSv up to 8 days after contamination even in the worst case (inhalation of type S compounds).

5.14.4. Tritium

Tritium is the one radionuclide for which the use of very limited urine samples is also foreseen by normal monitoring protocols since, at least in inorganic form (tritiated water, HTO), in a few hours it is distributed in an almost homogeneous way in the body water and urine, essentially without distinction between the different routes of intake, transcutaneous absorption included [Hill, 1993]. if the time of intake is known and the appropriate retention model for sex and age group is applied, a reliable dosimetry evaluation can be made from the simple determination of the current concentration of the radionuclide in body fluids (considered to be uniformly distributed in all organs and tissues for internal irradiation), of which even a small quantity of urine is considered to be sufficiently representative. The measurement method commonly adopted consists in counting 1-4 ml of urine by liquid scintillation (Liquid Scintillation Counting, LSC) after a simple dilution with bi-distilled water to limit the quenching effect (signal attenuation caused by reduced transparency to light of the sample). Adopting counting times of 180 s, it allows MDA levels of the order of 2 kBq/l to be easily obtained even using very common liquid scintillation counters. This concentration corresponds to committed effective doses well below 1 mSv even for urine samples collected several

weeks after contamination. Through the use of automated sample counting and replacement devices, which are normally supplied with LSC instruments, even if they are not particularly sophisticated, this method allows the monitoring of over 300 people per day for each available measurement system.

Although tritium appears to be the perfect guide radionuclide when it is present in a contaminating mixture, it is unlikely to be able to play this role, due to its very specific chemical-physical characteristics, which make it extremely unlikely that its behaviour in the environment will be related to that of other radionuclides.

5.14.5. Example cases on the use of guide radionuclides for contaminations by mixed sources

Three cases of possible use of a guide radionuclide for internal contamination monitoring will be examined:

- a) mixture of pure beta and beta-gamma emitters
- b) mixture of alpha emitters for which alpha spectrometry is the one way to perform monitoring measurements
- c) mixture of alpha emitters with the presence of radionuclides for which monitoring measurements can be performed by ICP-MS

5.14.5.1. Contamination by inhalation following an event that causes the dispersion in the air of a mixture of ^{137}Cs , ^{131}I and ^{90}Sr

Preliminary analysis of the case

^{137}Cs and ^{131}I are easily measurable through their gamma emission either by in vivo measurement (whole body counting for ^{137}Cs , thyroid counting for ^{131}I) or simultaneously by gamma spectrometry of urine (no treatment of the sample is required), whereas ^{90}Sr is a pure beta emitter whose measurement, even simplified by the rapid method previously exposed, is anyway complex and time spending. By the other hand the dispersion in air of ^{137}Cs and ^{90}Sr occurs in form of airborne particulate while iodine will very probably diffuse in part as airborne particulate, in part as a gas and thus it's not eligible as a guide for the other two contaminants (nor the inverse).

On this basis, a simplified monitoring procedure should be structured as it follows:

- identification and use of ^{137}Cs as a guide radionuclide for ^{90}Sr on the basis of a reference dose level (committed effective dose) of 10 mSv
- use of gamma spectrometry of urine (alternatively in vivo counting) for direct determination of ^{137}Cs and ^{131}I daily urinary excretion (in vivo activity)
- evaluation of intake and dose due to ^{137}Cs and ^{131}I by daily urinary excretion data (in vivo activity data)

- evaluation of the intake of ^{90}Sr on the basis of intake of ^{137}Cs using AR% value obtained by radiological characterisation of proper environmental samples
 - evaluation of dose due to ^{90}Sr
 - if it is negligible with respect to that due to ^{137}Cs and ^{131}I , continue the monitoring without changes
 - if it is not negligible, (i.e. > 10%) execution of the test on AR% (see above) for the use of ^{137}Cs as a guide radionuclide for ^{90}Sr
- continue the monitoring on the basis of the result of the test on AR%

5.14.5.2. Contamination by inhalation following an event that causes the dispersion in the air of a mixture of ^{241}Am and ^{239}Pu in the form of insoluble mixed oxides (AMAD $1\mu\text{m}$)

Preliminary analysis of the case

This is a typical mixture present in nuclear waste with an activity ratio between the two radionuclides that depends on the type of plant of origin, but which commonly tends to favor the presence of ^{241}Am . For this type of contaminant, it can be hypothesized a first mode of diffusion in the environment which tends to keep the activity ratios substantially unchanged in the fraction bio-available for inhalation.

For both radionuclides, internal contamination monitoring measurements are commonly carried out by alpha spectrometry of excreta using techniques which are substantially equivalent in terms of complexity and duration, despite some differences in chemical treatment. In emergency situations, or in any other case where rapid measurements are required, it is preferable to use ^{241}Am as a guide radionuclide due to its metabolic characteristics which make it possible to adopt reference levels definitely lower especially for insoluble compounds. As we will see, this choice, adequately supported by appropriate verification measurements, could significantly lower the estimable dose values due to ^{239}Pu if compared to values achieved by its direct measurement.

On this basis a simplified monitoring procedure should be structured as it follows:

- identification and use of ^{241}Am as a guide radionuclide for ^{239}Pu on the basis of a reference dose level for ^{241}Am of 20 mSv (committed effective dose)
- use of alpha spectrometry of small sample of urine (100 ml) for direct determination of ^{241}Am daily urinary excretion
- evaluation of intake and dose due to the inhalation of a type S compound of ^{241}Am (AMAD = $1\mu\text{m}$) by daily urinary excretion data
- evaluation of the intake of ^{239}Pu on the basis of ^{241}Am intake of using AR% value obtained by radiological characterisation of proper environmental samples
- evaluation of dose due to ^{239}Pu

- execution of the test on AR% for the use of ^{241}Am as a guide radionuclide performing measurement of ^{239}Pu in small samples both of urine and faeces for all the subjects of a proper control group:
 - verification of the acceptability of the value of plutonium intake deduced from the use of ^{241}Am as a guide radionuclide, by comparing it with the average of the intake values obtained from the measurement of plutonium in faeces
 - comparison of plutonium activity in urine and faeces in order to verify the class of solubility of the plutonium compounds actually present in the mixture
- continue the monitoring on the basis of the test result on AR%

Assuming a mixture of ^{239}Pu and ^{241}Am with a value of $\text{AR}\%=100\%$ and using ^{241}Am as a guide radionuclide, minimum detectable doses for ^{239}Pu calculated on the basis of intakes value reported in tables 5.18 and 5.19 for ^{241}Am , are always lower than 30 mSv for urine collected within 5 days from contamination ($\Delta\tau_{\text{UC}}$ for ^{241}Am), whereas for insoluble mixed oxides of ^{239}Pu , values of minimum detectable dose are between 140 and 250 mSv by direct measurement of ^{239}Pu in small (100 ml) urine sample collected within 4 days from the contamination (see tables 5.43 and 5.44). The comparison clearly shows that, in this very important case (given the frequency of its occurrence in nuclear activities), the use of ^{241}Am as a guide radionuclide, if permitted, not only allows to definitely simplify the monitoring procedure, but also to significantly improve its performances, guaranteeing much more conservative reference dose levels, at least when activity of ^{239}Pu is up to 3 times higher than ^{241}Am ($\text{AR}\%=300\%$) in the contaminating material.

5.14.5.3. Contamination by inhalation following an event that causes the dispersion in the air of a mixture containing ^{241}Am , ^{239}Pu and DU (depleted uranium)

Preliminary analysis of the case

The case seems very similar to the previous one, except for the additional difficulty due to the presence of depleted uranium, DU, here supposed at 0.2% for ^{235}U and 0.0011% of the total for ^{234}U by mass. However, it is precisely this fact that should offer the chance of significantly simplifying the monitoring procedure, given the easiness and speed of measuring uranium in urine by ICP-MS and the performance of this technique in terms of minimum detectable concentration. As reported above, MDC reported in terms of activity is in fact more or less one order of magnitude lower than that obtained by standard alpha spectrometry (sample of 500-1000 ml urine – counting time of several days) using a method that needs very small urine samples and takes no more than a few minutes to complete.

This is obviously conditioned on the actual possibility of using, in the specific situation, depleted uranium (^{238}U and ^{235}U) as a guide "radionuclide" for plutonium and americium; this implies:

- AR% value of both plutonium and americium with respect to DU must be stable (in time and space) in the contaminating mixture as it is incorporated into the body
- the order of magnitude of the activity concentration of DU in the mixture must be not too low compared to plutonium and americium isotopes.

The real problem, already analysed (see paragraph "Uranium" in the section "Mixed Source"), is given by the regular presence of a significant background of natural uranium in urine, which increases the levels of MDC actually achieved for DU in urine by ICP-MS up to 70-100 ng/L, corresponding to 1.5-2 mBq/24h for ^{238}U . If we suppose inhalation of type S compounds with AMAD = $1\mu\text{m}$, in terms of

intake this means the inhalation of 2.8 Bq, 20 Bq and 77 Bq of DU when urine is collected respectively in the first, in the second or in the fifth day after the contamination.

Assuming this minimum detectable intake in the hypothesis of identical activity concentration of DU, ^{239}Pu and ^{241}Am in the mixture and taking into account the committed effective dose coefficients of uranium isotopes, ^{239}Pu and ^{241}Am for inhalation of compounds of type S (all of the order 20-30 $\mu\text{Sv/Bq}$), the Minimum Detectable Effective Dose, MDD_{E} , by inhalation of the mixture as a whole is equal to 0.22 mSv at day one, 1.4 mSv at day two and 5.6 mSv at day five, with a contribution of about 30% attributable to DU. Therefore, if the Reference Level is fixed at 20 mSv, the use of DU as a guide radionuclide could be proposed for monitoring the internal contamination of such a mixture collecting urine within 5 days from the intake, until the sum of the concentrations of ^{239}Pu and ^{241}Am is up to 4.5 times higher than that of DU (proportionally for other L_{R} values).

On this basis, a simplified monitoring procedure for such a mixture should be structured as it follows:

- characterization of the contaminating mixture through measurements of proper environmental samples (composition of DU included)
- identification and use of DU (^{238}U and ^{235}U) as a guide for the whole mixture on the basis of a reference dose level of 20 mSv (committed effective dose)
- use of the standard methodology for uranium measurement in urine by ICP-MS
- evaluation of intake of DU on the basis of daily urinary excretion data
- evaluation of the intake of ^{239}Pu and ^{241}Am using AR% value obtained by radiological characterisation of proper environmental samples
- evaluation of dose due to ^{239}Pu and ^{241}Am
- execution of the test on AR% for the use of DU as a guide performing measurement of ^{239}Pu and ^{241}Am in small samples both of urine and faeces for all the subjects of a proper control group:
 - verification of the acceptability of the value of intake of ^{239}Pu and ^{241}Am deduced by DU comparing them with the average intakes value obtained by measurement of ^{239}Pu and ^{241}Am in faeces,
 - comparison of plutonium activity in urine and faeces in order to verify the class of solubility of the compounds actually present in the mixture
- continue the monitoring on the basis of the test result on AR%.

6. Conclusions

6.1. Gaps

- A global laboratory network would facilitate collective efforts in response to an emergency. Internal dosimetry laboratories and environmental laboratories should work and coordinate together in order to harmonize protocols for an emergency scenario. Unfortunately, no such coordination neither network for in vitro laboratories exists and would require an international assistance platform.

6.2. Main recommendations

- In an emergency scenario precise information and quick results of internal contamination is the main goal to calculate the dose received by each individual. Coordination of both in vivo measurements (whole body scan, thyroid scan, lung scan) and in vitro measurements (urine analysis, faecal analysis) should be used for this goal.
- The collected samples have to be analysed according to the follow up plan determined by the decision makers.
- Nasal swabs or nasal mucus samples are an useful indicator of an inhalation intake. It is recommended to collect them as soon as the people exposed condition permit, preferably within 1-2 hours of the contamination incident, depending on the emergency scenario.
- The collection of spot urine samples are recommended collected to provide a quick response and dose assessment for contaminated individuals.
- All documentation received with the samples has to be kept for later investigation and control.
- All prepared and analyzed samples should be kept and stored for later investigation.
- All waste produced during the preparation of the samples (packaging materials, pipettes, gloves, ...) has to be discarded as radioactive waste.
- Regular checks on possible contamination of the working environment and the counters used have to be performed.
- Other monitoring procedures and measurement techniques can be adopted and performed by in vitro laboratories but in any case they should be based on minimum detectable dose levels established in advance.
- Given that panic is the first effect of emergency situations, the design, planning and logistical organization are fundamental for the implementation of any extended monitoring, whether performed with in vitro methods (correct collection of samples) or with in vivo measurements (access of people to the place of acquiring measurement and their preparation).
- From this point of view, communication to the population on how to collect biological samples (urine) is of primary importance, in particular to avoid false contamination or unsuitable samples.
- In case of contamination by mixtures of radionuclides it is always advisable to consider a "guide radionuclide" for an initial evaluation of the contamination and of the individual dose.
- The individual monitoring design for a large crowd of people must be carried out on the basis of a predefined interpretative scheme. In particular, one or more reference dose levels must be indicated for the purpose of implementing specific actions.

- The adopted measurement techniques should respond to this scheme. In particular, they must allow the detection of all cases in which the dose from internal contamination exceeds the reference level with the lowest value. To this end, once defined the procedure, the result must take into account the most relevant parameters to get the minimum detectable dose. Specifically: detection efficiency, chemical yield, quantity of sample use and counting time.
- If the available measurement techniques are not suitable in relation to the adopted reference levels, the possibility of adopting higher reference levels (doses) must be considered.

7. Annexes

7.1. Register administrative information

Date:/...../.....

PERSONAL DATA

Name:

Surname:

ID/Passport:

Address:

Phone number

Email:

Age:

Sex: Male ☐ Female ☐

Female: Are you or do you think you may be pregnant? Yes ☐ No ☐

Incident location:

Time during the emergency scenario:

Minutes

Hours

Days

Do you have any of the following symptoms?

Nausea, vomiting, fever

Severe asthenia

Digestive disturbances

Skin signs

Other

TYPE OF SAMPLES YOU COLLECTED:

Nasal swabs/nasal mucus ☐

Spot sample ☐

24h urine sample ☐

Faecal sample ☐

7.2. Instructions for collection of NASAL SWABS/NASAL MUCUS SAMPLES

GENERAL CONSIDERATIONS

The collection should be done in a clean environment (OUT OF CONTAMINATED AREA), as soon as possible and preferably within 30 minutes of the contamination incident

Don't leave the containers in areas at risk of contamination.

Collect samples as soon as the conditions permits (better within 30 minutes of the contamination incident)

Samples can be taken using cotton tipped applicators or flag type applicators.

Clean your hands and the area around the nose. If you are undergoing radioisotope treatment or on special medication, inform the laboratory when submitting the urine.

Instruction of collection

- Collect the sample as soon as possible, preferably within 30 min of the contamination incident. In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample. Wash the area around the nose.
- Take the sample of the nasal mucus (either by swaps or by a nose blow tissue). Use a nasal swab slightly moistened with cotton tipped applicator and gently rotate around the accessible surface of the nostril. Proceed on the same way with the other nasal swab in the other nostril to avoid cross contamination.
- Each swab should be put into its own container (e.g. test tube, envelope, or bag), labelled with personal identification: first and last name, date and hour of collection and nostril (left. right).
- Nasal mucus can be collected with nasal swabs as above or with tissues. The patient blows in the tissue and the tissue should be put in a small plastic bag with personal identification (first name and last name, and date and hour of collection).
- Make sure that the container is properly closed before shipping or delivering it to the laboratory
- Use, preferably, porous nasal swabs to allow cocktail works and ensure the proper signal.

7.3. Instructions for collection of SPOT URINE SAMPLE

GENERAL CONSIDERATIONS

The collection should be done in a clean environment (OUT OF CONTAMINATED AREA)

Don't leave the containers in areas at risk of contamination.

Women should not collect urine during menstruation. Inform laboratory staff to assign new dates.

If you are undergoing radioisotope treatment or on special medication, inform the laboratory when submitting the urine.

Instruction of collection

- In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample.
- Discard the first urine after the possible intake. Collect the sample immediately afterwards directly into the clean container.

- Identify the sample: first and last name, and date and hour of collection.
- Make sure that the container is properly closed before shipping or delivering it to the laboratory

7.4. Instructions for collection of 24H URINE SAMPLES

GENERAL CONSIDERATIONS

The collection should be done in a clean environment (OUT OF CONTAMINATED AREA)

Don't leave the containers in areas at risk of contamination.

Women should not collect urine during menstruation. Inform laboratory staff to assign new dates.

If you are undergoing radioisotope treatment or on special medication, inform the laboratory when submitting the urine.

Instruction of collection

- In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample.
- Discard the first urine after the possible intake. Collect the sample immediately afterwards directly into the clean container. All urination shall be collected for 24 hours, including overnight.
- Identify the sample: first and last name, and date and time at sampling begins and ends.
- Make sure that the container is properly closed before shipping or delivering it to the laboratory

7.5. Instructions for collection of FAECAL SAMPLES

GENERAL CONSIDERATIONS

The collection should be done in a clean environment (OUT OF CONTAMINATED AREA)

Don't leave the containers in areas at risk of contamination.

If you are undergoing radioisotope treatment or on special medication, inform the laboratory when submitting the urine.

The faecal samples collected need to be total voiding, not simply stool smears or swabs.

Instruction of collection

- In a clean room and previous hand washing, take the sample as hygienically as possible to avoid external contamination of the sample.
- Faecal samples must be taken quickly after contamination directly into the clean and air-tight plastic container.
- It is recommended that stool samples are collected on a daily base, and at least up to 3 days following the incident. This means taking 3 individual daily samples, not one single sample combining all stools.
- Identify the sample: first and last name, and date of collection.
- Make sure that the container is properly closed before shipping or delivering it to the laboratory

Preservation and storage

It is advisable to store the samples at least between 2°C and 8°C, and preferably to freeze them between -15°C and -20°C until pre-treatment by calcination, in order to limit inconvenience.

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