

ASSESSMENT OF OCCUPATIONAL EXPOSURE DUE TO INTERNAL RADIATION SOURCES

UNIT 6 INDIRECT METHODS FOR INDIVIDUAL AND WORKPLACE MONITORING

METHODS FOR INDIVIDUAL MONITORING AND WORKPLACE MONITORING OF INTERNAL EXPOSURES

- INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING
 In vitro measurements of activity concentration in biological samples
 Workplace monitoring Air sampling
- ADVANTAGES AND LIMITATIONS OF MONITORING TECNHNIQUES

• METHODS FOR INDIVIDUAL MONITORING AND WORKPLACE MONITORING OF INTERNAL EXPOSURES

The **doses due to intakes of radionuclides** can not be obtained directly from measurements but must be assessed from:

- In-vivo measurements of the retained activity M(Bq) in total body or organs, using whole/partial Body Counters
- ✓ In-vitro measurements of the activity concentration in excreta samples M(Bqd⁻¹, BqL⁻¹)
- ✓ Workplace monitoring Air sampling. Activity concentration in the air M(Bqm-³)

Or by a combination of these methods

- INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING
 - ✓ In vitro measurements of activity concentration of and gamma emitters in biological samples





EC Report 188 - Technical Recommendations for Monitoring Individuals for Occupational Intakes of Radionuclides (ec.europa.eu/energy/sites/ener/files/rp_188.pdf)

Measurements of

Workplace monitoring – Air sampling.
 activity concentration in the air with Personal (PAS)
 or Static (SAS) Air Samplers.

- INDIRECT METHODS In vitro Monitoring of Biological Samples
 - 1.- Excreta Samples (Urine and faeces)

✓ Urine samples. Information to take into account about urinary excretion:

- Daily urinary excretion is <u>1.6 Ld⁻¹ (reference man)</u> and <u>1.2 Ld⁻¹ (reference female)</u>
- Creatinine is excreted at an average rate of 1.7 g d⁻¹ (men) and 1.0 g d⁻¹ (women)

These values may be used for normalization (24h excretion)

- ✓ Faecal samples. Regarding faecal excretion:
 - Reference faeces weight for male is 150 g, and 120 g for female

- INDIRECT METHODS In vitro Monitoring of Biological Samples
 - 2.- Other biological materials
 - Nose blow or nasal swab analysis may be used after a suspected incident to detect a radionuclide intake through inhalation.
 - A positive result of the measurement (above the detection limit) gives an indication that an unexpected exposure may have occurred.
 - Excreta measurements or in vivo monitoring should follow, to confirm the intake and to provide a quantitative dose assessment.

Collection of excreta samples for individual monitoring

- ✓ Use single-use containers for collection and storage.
- Collection of samples in non-contaminated areas
- ✓ To avoid radioactive contamination of the sample.
- Acid reagents or other chemical agents should be added to sample containers to minimise precipitation and to prevent bacterial growth. Prompt analysis after reception of the sample is recommended (avoids deterioration by bacteriological action).
- The samples may be stored at reduced temperature. Refrigeration or freezing should be employed when appropriate
- ✓ A sample collected at the end of the work shift is the most sensitive indicator of exposure.
- A sample taken after a period of no exposure or after holidays confirms intake of radioactive material that is slowly excreted

Collection of excreta samples for individual monitoring

- Routine and special monitoring of occupational intakes: as general approach(*) the collection of 24-hour samples are recommended.
- Monitoring of ³H in urine: spot sample is provided in case of intakes of tritiated water, which is uniformly distributed in the body fluids
- ✓ When 24-hour samples are not collected, the first void in the morning is the preferable spot sample for analysis.
- Individual daily variations may occur in the excretions of some materials comparing with reference values
- ✓ Spot samples may not be representative after normalisation by volume or creatinine content.
- Analysis of a 24h-urine sample will result in a better dose assessment than using spot samples
- (*) Spot samples may be collected in case of direct analysis of actinides using ICP-MS or KPA techniques and for *in vitro* monitoring of beta emitters such as tritium (³H), ¹⁴C, ³²P and ³⁵S.

- Collection of excreta samples for individual monitoring
 - ✓ NORMALIZATION 24-hour excretion <u>Urine samples</u>
 - Normalization by creatinine content:
 - It is recommended to use creatinine measurements to estimate 24-hour excretion from urine samples collected over part of a day, or to confirm collection of 24h sample
 - Creatinine is excreted at an average rate of 1.7 g d⁻¹ for men and 1.0 g d⁻¹ for women. The ratio of this reference value to the measured creatinine content in the sample provides a correction to normalise the radionuclide amount measured in the sample to the equivalent of a true 24-hour collection.
 - Normalisation by volume- taking into account daily urinary excretion:
 - o 1.6 Ld⁻¹ (reference man)
 - 1.2 Ld⁻¹ (reference female)

Collection of excreta samples for individual monitoring

✓ NORMALIZATION – 24-hour excretion - Faecal samples

- Daily fluctuations in faecal excretion (large individual variations in transit time through the alimentary tract) may result in uncertainty in the interpretation of faecal monitoring data.
- Routine and Special Monitoring: to reduce the impact of these variations, sampling over a 3 day period is recommended.
- Important for accidental exposures: sampling over the 3 days following the intake
- Faecal samples are particularly subject to biodegradation; to be analysed promptly, ashed and stored, or preserved by deep freezing is highly recommended.
- **Reference faeces wet weight** for male is 150 g 120 g for female
- Faecal monitoring applies in investigation cases, especially for intakes of Type M, Type M/S or Type S materials

• INDIRECT METHODS - In vitro Monitoring of Excreta Samples (urine and faeces)

- In vitro bioassay is the measurement technique of choice to quantify internal contamination of pure alpha and beta emitters.
- **\checkmark Excreta monitoring** may be the only measurement technique available for radionuclides with **no** γ **ray emission or only low energy photon emissions.**
- ✓ The selection of the appropriate in vitro radiobioassay techniques are based on:
 - the information on the radionuclides involved,
 - the chemical and physical forms of incorporated radioactive material
 - the possible presence of interfering radionuclides,
 - the necessary sensitivity to meet monitoring requirements,
 - the availability of instrumentation and technical expertise in the laboratory.

- INDIRECT METHODS In vitro Monitoring of Excreta Samples (Urine and feces)
 - ✓ Alpha Spectrometry
 - This method is used for the monitoring of alpha emitters e.g. ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, ²⁴¹Am, ²⁴⁴Cm, ²³⁴U, ²³⁵U, ²³⁸U, ²²⁸Th, ²³⁰Th, ²³²Th,...
 - Radiochemical separation of the sample s required, prior to measurement
 - Semiconductor detectors or gridded ionisation chambers quantify individual radionuclides if energies are sufficiently different.
 - Long counting times Tc ~ 300000 s
 - Results in 2-3 weeks after reception of the sample





• INDIRECT METHODS - In vitro Monitoring of Excreta Samples (urine)

Mass Spectrometry methods provide information on isotopic composition

✓ Mass Spectrometry: ICP-MS (Inductively Coupled Plasma Mass Spectrometry)

- Allows direct analysis of long-lived radionuclides of uranium, thorium, plutonium and others in biological samples
- Rapid alternative vs. Alpha Spectometry, but much more expensive
- Analytical method is based only on the mass of the radionuclide. Chemical separation of elements with isotopes of the same mass number is needed.



- Easy sample treatment, diluted urine samples
- Results in 1-2 days

✓ Mass Spectrometry: TIMS (Thermal Ionisation Mass Spectrometry)

uranium and plutonium isotopic measurements on a variety of sample types

- INDIRECT METHODS In vitro Monitoring of Excreta Samples (urine)
 - ✓ Kinethic Phosphorescence Analyser (KPA) and Fluorimetry
 - Determination of total concentration of uranium in urine. samples
 - Rapid alternative vs Alfa Spectrometry but higher Detection Limits comparing with Alpha Spectrometry and ICP-MS
 - KPA analysis is a rapid method for screening

✓ Fluorimetry:

- This method quantifies total uranium in urine.
- It is a more rapid technique than alpha spectrometry
- The detection limit of fluorimetry is higher than that of KPA.



- INDIRECT METHODS In vitro Monitoring of Excreta Samples
 - ✓ Liquid Scintilation Counting (LSC)
 - The majority of beta emitters are monitored by LSC (especially low energy beta emitters)

 ³H (HTO, OBT),
 ¹⁴C, ³⁵S, ³²P,
 ⁹⁰Sr
 - Simultaneous determinations if the multiple radionuclides have different beta energies e.g. mixtures of ³H, ¹⁴C and ⁹⁹Tc, but it is not possible to measure ¹⁴C and ³⁵S simultaneously
 - Simple treatment of the sample, except for ⁹⁰Sr analysis which requires radiochemical separation
 - Counting time: Tc ~ 60 120 min
 - Results in 1 day (⁹⁰Sr in ~5 days)



INDIRECT METHODS - In vitro Monitoring of Excreta Samples

✓ Gamma Spectrometry

- Determination of radionuclides that emit X-rays or gamma radiation in biological samples by direct measurement with scintillation Nal(TI) or semiconductor HPGe detectors (gamma spectrometry)
- Samples in appropriate containers with the same geometrical configuration as the reference source used for the efficiency calibration.
- Rapid method. Simple sample treatment. Self-absorption of photons in the sample should be considered.
- Counting time is chosen according to the desired detection limit.
- Results in 1 day

INDIRECT METHODS - In vitro Monitoring of Excreta Samples

✓ Natural Background

- Background levels in excreta samples resulting from dietary intake should be taken into consideration if contribution from natural background has a significant effect on the assessed dose.
- If natural background levels are not taken into account, it should be demonstrated that their contribution to assessed dose is not significant.
- In vitro measurements before the start of the occupational exposure of a particular individual are highly recommended in order to quantify the individual background
- ISO 16638-1 and IDEAS Guidelines provide recommendations on how to handle monitoring data taking into account dietary intake. Where the occupational exposure is to natural uranium, a range of reference values must be set to distinguish between occupational exposures and natural background.
- In case of occupational exposure to either depleted or enriched uranium, the measurement of the isotopic content of a sample allows to know and to subtract the contribution from the natural uranium background

INDIRECT METHODS - In vitro Monitoring of Excreta Samples

✓ Natural Background

Occupational exposure to either depleted or enriched uranium:

- The reference value for an individual worker should be determined by measurements of blank bioassay samples before work with Uranium starts
- If this is not feasible/reliable, information on bioassay samples provided by a representative population of unexposed workers may be used to set background ranges and reference values.
- If this is not feasible, measurements of the uranium content in drinking water may be used to establish reference values.
- Published data may be used, e.g. reported in IDEAS Guidelines.
- According to ISO 16638-1: it must be demonstrated the reference value is representative of natural background level of the worker

- INDIRECT METHODS In vitro Monitoring of Excreta Samples
 - ✓ **Uncertainties** Type A and Type B uncertainty sources.

• Uncertainties **in measurements**: counting uncertainties, calibration procedures, possible contamination of the source or the measurement system, and fluctuations in background.

• Type B uncertainty for In vitro radiobioassay:

Quantification of the sample volume or weight; error in dilution and pipetting; evaporation of the solution in storage; stability and activity of standards used for calibration; chemical recovery; blank corrections; background contributions and fluctuations; electronic stability; environmental conditions; spectroscopy resolution and peak overlap; contamination of the sample and impurities; source positioning for counting; density and shape variation from the calibration mode; decay corrections; and assumptions about homogeneity in calibration.

INDIRECT METHODS - In vitro Monitoring of Excreta Samples

✓ Uncertainties - ISO 27048 describes and analyses the components of uncertainties in indirect measurements (Table B.3) and proposes quantification of measurement uncertainties by applying scattering factors.

Quantity	Type B scattering factor, K _{SFB}
True 24 h urine	1,1 ^a
Activity concentration of ³ H in urine	1,1
Simulated 24 h urine, creatinine or specific gravity normalised	1,7
Spot urine sample	2,0
Faecal 24 h sample	3
Faecal 72 h sample	1,9
Chest count	2

 Table B.3 — Default values for the lognormal scattering factor K_{SF} for various types of measurement from different studies (Type B errors) (derived from [21][24])

Taken from ISO 27048, Annex B, Table B.3

INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING

✓ Workplace monitoring – Air sampling.

Determination of Airborne Radionuclide Concentration M(Bqm⁻³).

Measurements are carried out in the working environment.

- Personal Air Samplers (PAS) and Static Air samplers (SAS) may be used for workplace monitoring of individual exposures.
- **PAS and SAS** may be appropriated where available *in vivo* and *in vitro* techniques can only quantify reliable doses above 6 mSv, as is the case of actinides (ISO 20553).
- Workplace monitoring of exposure to airborne naturally occurring radionuclides may be required

INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING

✓ Workplace monitoring – Sampling Methods

Determination of Airborne Radionuclide Concentration

- Personal Air Sampling PAS
 - PAS: portable device to collect sample representative of the activity concentration in the air inhaled by the worker;
 - PAS is most commonly used for the estimation of actinide exposures (e.g. to estimate inhalation intakes of Pu and U isotopes)
 - A sampling head with a filter is worn on the upper torso within the breathing zone, within 30 cm of nose and mouth. Sampling rates should be the same as breathing rates for a worker (~1.2 m³h⁻¹), but current devices often provide only about one fifth of this value.
 - Filter's activity at the end of sampling period can give a warning of unexpected high exposures.
 - Intakes are estimated using the ratio of sampling rate to typical breathing rate; doses are estimated using reference dose coefficients.

Protection

INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING

✓ Workplace monitoring – Determination of Airborne Radionuclide Concentration

- Personal Air Sampling PAS UNCERTAINTIES
 - Uncertainties arise from two main sources:
 - > Statistical variation of the number of particles in randomly sampled volume
 - Variability in the activity associated with each particle due to the variation of sizes of particles in the aerosol.
 - Sampler picking up non respirable particles can give false positive doses
 - For actinides with relativity high dose coefficients and higher specific activities (e.g. ²³⁹Pu), main source of uncertainty estimating intakes arises from the statistical variation in the radionuclide activity collected by the sampler during a defined sampling period.







INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING

✓ Workplace monitoring – Sampling Methods -

Determination of Airborne Radionuclide Concentration

Static Air Sampling – SAS

• Static Air Sampling (SAS) is used for monitoring workplace conditions, but can underestimate concentrations in air in the breathing zone of the worker up to several orders of magnitude.

 Where PAS is used together with SAS, PAS:SAS air concentration ratios can vary from <1 up to 100. Where air activity concentrations are reasonably homogenous in the workplace and multiple sample measurements are taken, ratios tend to be in the range of 1-10.

 Variability in the ratio arises from the spatial variation of aerosol concentration in the workplace, and depends on the relative positions of the SAS, the PAS, the source(s) of airborne contamination and the localised air flow patterns.



- INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING
- ✓ Workplace monitoring Sampling Methods

Determination of Airborne Radionuclide Concentration

- Static Air Sampling SAS
 - SAS can provide useful information on radionuclide composition and on particle size, if used with a size analyser such as a cascade impactor.
 - $\circ~$ SAS may be used to estimate intakes and doses for workers:
 - when expected doses are either low (ISO 20553) or for confirmation that workplace conditions do not require individual monitoring programmes.
 - Estimated intakes will need to account for potential underestimates of the SAS measurement by the application of correction factors



- A correction factor exceeding a factor of 10 is recommended for reviewing the reliability of intakes estimated in this manner.
- Further corrections are required to account for potential differences in the occupancy time of a worker in the area being monitored by SAS, the SAS sample time, and the aerosol retention characteristics within the local area.



INDIRECT METHODS: IN VITRO RADIOBIOASSAY AND AIR SAMPLING

✓ Workplace monitoring – Radon Exposure Monitoring

- Radon is an inert noble gas that is encountered in elemental form as a gas or dissolved in water.
- ²²²Rn, ²²⁰Rn and ²¹⁹Rn, are progeny radionuclides of radium isotopes (²²⁶Ra, ²²⁴Ra and ²²³Ra), which are members of the 3 natural radioactive decay series (parent radionuclides ²³⁸U, ²³²Th and ²³⁵U respectively).
- The isotopes ²²²Rn, ²²⁰Rn, ²¹⁹Rn are known as radon, thoron and actinon respectively.
- High concentrations of radon in air have been found in mines, waterworks, caves, underground stores and U/Th handling facilities.
- Information about monitoring and dosimetry of radon exposures: EC RP 188, Technical Recommendations for Monitoring Individuals for Occupational Intakes of Radionuclides (ec.europa.eu/energy/sites/ener/files/rp_188.pdf)

Chapter H "Radon Measurement and Dosimetry for Workers"



ADVANTAGES AND LIMITATIONS OF MONITORING TECHNIQUES

Methods	Advantages	Limitations
In vivo monitoring of radionuclides in an organ or in total body	Rapid measurement of the activity retained and deposited in the body, especially important in case of RN emergencies Rapid intake and dose assessments	Mainly $X_{ray} + \gamma$ emitter radionuclides Physical phantoms simulating internal contamination of organs or total body not always available Worse detection limit for actinide and NORM exposures when comparing with in vitro bioassay
In vitro radiobioassay of excreta samples	In vitro bioassay is the measurement technique of choice to quantify internal contamination of pure alpha and beta emitters. Alpha Spec. and ICP-MS: excellent detection limits	Alpha spectrometry: long time (~2 weeks) for estimating activity concentration in excreta samples. ICP-MS: better for NatU and Th in urine samples (and for other long lived radionuclides e.g. ²³⁹ Pu) but expensive technique
Workplace monitoring, Air Sampling	PAS and SAS may be useful when available <i>in vivo</i> and <i>invitro</i> techniques only quantify exposures reliably above 6 mSv, e.g. for monitoring actinides	High uncertainties may be associated when calculating intake, then difficult to use for dose assessment

REFERENCES - UNIT 6 - INDIRECT METHODS FOR INDIVIDUAL AND WORKPLACE MONITORING

EUROPEAN COMMISSION - RADIATION PROTECTION REPORT SERIES No.188 - Technical Recommendations for Monitoring Individuals for Occupational Intakes of Radionuclides (ec.europa.eu/energy/sites/ener/files/rp_188.pdf). EC RP 188 (2018).

EUROPEAN RADIATION DOSIMETRY GROUP [EURADOS] - IDEAS Guidelines (Version 2) for the Estimation of Committed Doses from Incorporation Monitoring Data. EURADOS Report 2013-01 ISBN 978-3-943701-03-6 (2013). http://www.eurados.org/~/media/Files/Eurados/documents/EURADOS%20Report%202013-01%20online%20version.pdf?la=en INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION, Basic Anatomical and Physiological Data for Use in Radiological Protection. Reference Values. ICRP Publication 89. Ann. ICRP 32 (3-4) (2002).

INTERNATIONAL ORGANIZATION FOR STANDARDIZATION. RADIATION PROTECTION – Monitoring of Workers Occupationally Exposed to a Risk of Internal Contamination with Radioactive Material. ISO 20553:2006. (ISO:Geneva) (2006)

INTERNATIONAL ORGANIZATION FOR STANDARDIZATION. RADIATION PROTECTION – Performance criteria for radiobioassay. ISO 28218:2010 (ISO: Geneva) (2010).

INTERNATIONAL ORGANIZATION FOR STANDARDIZATION. RADIATION PROTECTION – Dose assessment for the monitoring of workers for internal radiation exposure ISO 27048:2011 (ISO: Geneva) (2011).

INTERNATIONAL ORGANIZATION FOR STANDARDIZATION. RADIATION PROTECTION – Monitoring and internal dosimetry for specific materials. Part 1: Inhalation of Uranium compounds. ISO 16638-1:2015 (ISO: Geneva) (2015).

INTERNATIONAL ORGANIZATION FOR STANDARDIZATION. RADIATION PROTECTION – Monitoring and internal dosimetry for specific materials. Part 2: Ingestion of Uranium compounds. ISO 16638-2:2019 (ISO: Geneva) (2019).