INDIVIDUAL MONITORING FOR INTERNAL EXPOSURES IN EUROPE: CONCLUSIONS OF AN EURADOS ACTION

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Once the EC Directive 96/29 has been implemented into national regulation across Europe, the coordination of dosimetry laboratories for the monitoring of occupational exposures becomes the principal aim to achieve. Within this framework the European Radiation Dosimetry Group, EURADOS, carried out an Action on ‘Harmonisation of Individual Monitoring’ (2000–2004) to promote coordination in the field of individual monitoring of occupational exposures throughout Europe. With reference to internal exposures, the main aims were the completion of a catalogue of internal dosimetry services and an inventory of methods and techniques used for individual monitoring at European internal dosimetry facilities. At the end of this EURADOS Action, a report was published in *Radiation Protection Dosimetry* in 2004. The information collected related to various topics: the equipments used for the measurement of internal exposures, calibration and sensitivity data, the methods applied for the assessment of internal doses, Quality Control procedures, Quality Assurance Programmes in the facilities and legal requirements. The information to be presented here will give a general overview of the actual status of individual monitoring for internal exposures in Europe.

INTRODUCTION

One of the issues addressed in the Euratom Council Directive 96/29 of 13 May 1996⁽¹⁾ is that monitoring of the exposure should in general be performed by approved dosimetric services, but the Directive gives no criteria for the approval of these services. There is, however, a consensus among professionals that approved dosimetric services should perform in agreement with the relevant international standards [e.g. ISO (International Organisation for Standardisation) and IEC (International Electrotechnical Commission)] and recommendations (e.g. from the European Commission and the International Atomic Energy Agency).

The European Radiation Dosimetry Group, EURADOS, was created to be a scientific network of European laboratories (voting members) and Associated Members involved in scientific research in radiation dosimetry. Over the past 10 years emphasis has been put on the harmonisation of individual monitoring procedures in Europe. A working group funded by the European Commission in the 5th framework programme (2001–2004) was

established within the Concerted Action ‘Dosimetry Network’. Experts from EU member states aimed at assisting in the process of harmonisation of individual monitoring of internal and external exposures as part of the protection of occupationally exposed persons. In this instance the term harmonisation does not mean that the services have to follow exactly the same procedures but aim to meet the same requirements and as a consequence their results should be comparable.

One of the tasks of this Harmonisation Group was to study the integration of dosimetric methods for internal and external exposures. A subgroup of dosimetry scientists investigated how the results from personal dosimeters for external radiation, from workplace monitoring and from monitoring for internal exposure can be combined into a complete and consistent system of individual monitoring. A Dosimetry Network of 28 countries working together for harmonisation has been established. Important information about the performance of European dosimetric services has been collected, with the help of the contact-persons (one per country), who have actively collaborated with EURADOS in the distribution of Questionnaires among the individual monitoring facilities of each European country.

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INDIVIDUAL MONITORING OF INTERNAL EXPOSURES: EUROPEAN CATALOGUE OF METHODS AND SERVICES

One of the objectives of the EURADOS study was the completion of a catalogue of techniques and an inventory of methods applied at the internal dosimetry laboratories throughout Europe. The 'Eurados 2002 Questionnaire⁽²⁾' was sent to services, requesting information about the equipments used for the measurement of the retained/excreted/air concentration activity of radionuclides internally deposited in the body, and the methodology, available tools and legal requirements associated with the assessment of internal doses.

Seventy-three completed questionnaires were returned by e-mail from the internal dosimetry facilities of 26 countries. At the end of this EURADOS Action, a report was submitted for publication⁽²⁾. This report presents an important overview of the individual monitoring of internal exposures in Europe for workers with the risk of intake of radionuclides.

The Internal Dosimetry Catalogue contains all the materials collected from the services which completed the EURADOS questionnaire. This broad set of data includes information from techniques to performance criteria used in facilities dealing with the assessment of internal exposures. The interest was mainly focused on equipment, calibration procedures, sensitivity [minimum detectable activity (MDA) of the radionuclides of interest] and the methodology applied for the evaluation of effective doses. Reference levels, dosimetric tools, application of the latest International Commission on Radiation Protection (ICRP) biokinetic models and Quality Assurance of laboratories were also reported.

An inventory of the internal dosimetric services operating in each European state was also presented. Information about measurement techniques and the type of occupational internal exposures occurring by country was supplied by the contact-persons; other relevant information about individual monitoring requirements (approval of services, accreditation of laboratories, legal aspects, dose record keeping, etc.) was also included in the report.

A collaboration was carried out with other Internal Dosimetry Projects in Europe; for instance, the OMINEX⁽³⁾ projects (some UK internal dosimetry services agreed to share with EURADOS the information they supplied to OMINEX) and the 'Uranium and Thorium measurements related to internal exposures' Project⁽⁴⁾ supported by European Commission and coordinated by GSF (Germany).

CONCLUSIONS OF THE EURADOS HARMONISATION STUDY

Internal doses cannot be measured directly. The quantities adopted⁽⁵⁾ to express the doses received

from intakes of radionuclides for radiological protection purposes are the effective dose E (Sv) and the equivalent dose H_T (Sv) in tissue or organ T . The dose coefficient $e(g)_j$ (Sv Bq⁻¹) or committed effective dose per unit of intake for radionuclide j by ingestion or by inhalation is used to determine the committed effective dose E_{50} (Sv) from an estimated intake; for occupational exposures, the period of time over which the committed effective dose is assessed is 50 y.

The fraction of intake that remains in the body (activity obtained by direct measurement methods) or that is excreted from the body (evaluated by indirect methods) at a time t after the intake depends on the radionuclide, its chemical and physical forms, the route and the moment (t) of intake. In case of inhalation of radionuclides, the dose can also be inferred by the measurement of airborne activity concentration.

Workers undertaking tasks with the risk of internal exposure must be included in individual monitoring programmes for assessment of the activity retained in or excreted from the body. Type of technique (direct/indirect) and frequency of measurements are established after the complete characterisation of the potential intakes of the radionuclides present at the workplace.

The first conclusion taken from the Harmonisation Action is related to the type of internal exposures. Many workers monitored by the services included in the EURADOS study, undertake work in Nuclear Power Plants, research centres and in the medical field. Other workplaces with risk of internal contamination are non-nuclear industry facilities, nuclear fuel cycle laboratories and installations undergoing decommissioning. In a small number of cases monitoring of persons exposed to depleted uranium was also reported.

Intakes of radionuclides can be determined by either direct or indirect measurement methods. There are more workers assessed for internal exposures using direct methods [whole-body counting (WBC) or organ counting by gamma spectrometry] as compared with the number of persons exposed and evaluated using indirect techniques [determination of activity in excreta and personal air samplers (PAS) and static air samplers (SAS) monitoring]. Each type of measurement has advantages and disadvantages; *in vivo* (direct) counting offers the possibility of a rapid and convenient estimation of the activity of the radionuclide in the body, but suffers from important calibration uncertainties, especially for low-energy (X-ray and gamma-ray) photons. *In vitro* measurements of activity in biological samples require the use of a radiochemical analytical laboratory. All these special features were considered in the EURADOS study.

DIRECT TECHNIQUES: *IN VIVO* EVALUATION OF GAMMA EMITTERS RETAINED IN THE BODY

The direct technique most commonly used to monitor workers in Europe is WBC for *in vivo* determination of the activity of gamma emitters in total-body. The detection of radioiodine in thyroid is required for research and medical exposures; the assessment of the activity of radionuclides in lungs is generally associated with the exposure to Actinides (americium and uranium, also plutonium). Other techniques are available in a few facilities, for example the detection of Actinides in liver or in bone, wound counting and *in vivo* monitoring of beta emitters.

Whole-body counting: *in vivo* determination of gamma emitters in total-body

The sample of *in vivo* laboratories collaborating with EURADOS (63 whole-body counters from 52 internal dosimetry services) is representative of the direct methods applied in Europe for the evaluation of retained activity in the body after internal exposures. Five mobile units, 30 detector systems inside shielded room and 28 equipments with no shielded room were reported.

Harmonisation in relation to materials and size of room was found to exist in relation to the shielded rooms constructed in 50% of *in vivo* facilities. The aim is to reduce the background radiation and to decrease the limits of detection (maximum sensitivity). A typical European WBC would consist of a room constructed with walls of 16 cm of steel (outer shield), lined with 4 mm of Pb (inner shield) and Cu or Cd. This reference facility would meet recommendations established in the ICRU 69⁽⁶⁾ publication. Some laboratories were constructed with concrete (30–70 cm) or iron (few instances) instead of steel walls.

Different detector–subject arrangements are available in WBC determinations. The counting geometry selected by the facility usually looks for the performance of measurements making maximum efficiency accessible. Bed geometry (scanning and stationary) is the most widely used configuration, with chair geometry as an alternative arrangement, and the standing-up counting geometry being selected for quick monitoring.

The majority (53%) of European WBC facilities use thick NaI(Tl) detectors (status 2001) for the determination of gamma emitters in total-body (either single or multidetector systems). A large number of *in vivo* facilities installed high-purity Ge (HPGe) detectors (excellent spectral resolution) with different arrays.

In vivo calibration of whole-body monitoring requires the use of anthropometric phantoms with

photon transmission properties equivalent of human tissues. Plastic bottle phantoms, brick simulators and BOMAB units are preferred. A set of radionuclides is then introduced into the phantom with uniform distribution of activity in the simulated body. The EURADOS study shows that the mixture of isotopes selected by services for WBC calibration includes ^{137}Cs and ^{60}Co in most cases, with ^{109}Cd , ^{139}Ce , ^{203}Hg , ^{113}Sn and ^{88}Y being chosen less frequently. A group of facilities prefer the use of multilines emitters as ^{152}Eu or ^{133}Ba . In a few instances low-energy contribution is considered (^{57}Co or ^{241}Am), and only 15% of the WBCs perform total-body calibration with ^{40}K .

The MDA is the indicator of the sensitivity of *in vivo* detection systems. MDA represents the detection limit of the system expressed in terms of activity, and is calculated using a blank person or a phantom simulating the human body containing an expected amount of ^{40}K (~4500 Bq for the standard man). MDA mainly depends on background, counting efficiency and counting time.

The EURADOS Catalogue registered the values of MDA obtained by services for *in vivo* detection of ^{137}Cs and ^{60}Co in total-body. In general, there is a good agreement with the data supplied by those facilities using NaI(Tl) detectors for WBC. Differences were found in the cases of services using HPGe detectors, with lack of agreement in the sensitivity study. Optimisation of efficiency and sensitivity is achieved by using a mixture of detector systems (NaI + HPGe).

A typical European *in vivo* facility would use one thick NaI(Tl) detector for WBC, in either bed or chair geometry, for a counting time of 1200 s; the sensitivity supplied by the system would be ~60 Bq of ^{137}Cs and ^{60}Co in total-body. Lowest MDA data are found in a mixture of NaI(Tl) and HPGe detectors inside a shielded room. Here, for a counting time of 600 s, MDA values would meet the range 20–30 Bq. Quick monitoring is also performed with NaI(Tl) detectors, for a standing-up counting geometry and counting times of 1–2 min (MDA obtained are 100–250 Bq).

Arrays using HPGe detectors, in either bed or chair geometry, are used in a wide variety of working conditions. The performance of *in vivo* determinations of ^{137}Cs and ^{60}Co in total-body with HPGe detectors for a typical counting time of 1000 s was reported to have a sensitivity in the range of 50–200 Bq of MDA.

Thyroid counting: *in vivo* determination of ^{131}I and ^{125}I

Thirty-eight *in vivo* facilities (44 thyroid systems) collaborated with EURADOS in the harmonisation study. *In vivo* determination of ^{131}I (364.5 keV) is typically carried out with one NaI(Tl) detector at an

average distance of 10–15 cm from the thyroid area (50% of laboratories). MDA range covers values from 4 to 800 Bq, with a counting time range of 10–1800 s (typically 600 s). It was noted that HPGe detector are used in a small number of facilities also.

Thirteen of the *in vivo* facilities included in the catalogue determine ^{125}I retained in the thyroid gland. Those using semiconductor systems, HPGe or low-energy Ge detectors, obtained values of MDA ~ 5 Bq of ^{125}I (27 keV). In few instances NaI(Tl) detectors are selected, 25 Bq of ^{125}I being the lowest MDA reported; in this case analysis was carried out using the 27 keV (X ray) and 35 keV (gamma ray) emissions of this radionuclide.

Lung counting

In vivo determination of radionuclides in lungs is in general carried out for workers with risk of inhalation of Actinides with long retention time in lungs (U, Pu and ^{241}Am oxides). The difficulty of this type of measurements is related to the complex detection of the low-energy photons (10–200 keV) internally deposited in the body, going through the torso tissues and reaching the detector. Eleven laboratories participated in this part of the EURADOS Catalogue, the majority are using Ge detectors as lung counters. Lawrence Livermore National Laboratory (LLNL) and Alderson Phantoms are widely accepted for lung calibration; no services reported using the Japanese Atomic Energy Research Institute (JAERI) phantom. There was general agreement of the use of overlay chest plates with a composition of 50% muscle and 50% adipose for the efficiency study.

The assessment of the activity in lungs is carried out in routine monitoring from *in vivo* measurements of ^{241}Am , ^{235}U , $^{234}\text{Th}/^{238}\text{U}$, ^{239}Pu and ^{232}Th , with counting times covering the range 1200–3000 s. The sensitivity of the detector system is in general obtained for a typical chest wall thickness of 3 cm. Lowest MDA values (3–5 Bq) are obtained in the determination of ^{235}U (186 keV) in lungs (assessment of the intake of enriched uranium). The MDAs reported by services for *in vivo* determination of ^{241}Am are in the range 4–8 Bq. Forty-five becquerels of ^{234}Th is the value associated with the sensitivity of the detection system for the evaluation of ^{238}U in lungs (natural uranium).

INDIRECT TECHNIQUES: *IN VITRO* EVALUATION OF THE ACTIVITY EXCRETED FROM THE BODY

Forty-three *in vitro* laboratories participated in the EURADOS study answering the 2002-Questionnaire with information about the indirect techniques used for the determination of internal

exposures at workplace. The more important conclusions about methodology and performance features are presented as follows.

In vitro measurements of Actinides in excreta (19 laboratories)

Alpha spectrometry is the technique most commonly used by services for *in vitro* measurements of Actinides in excreta. ICP/MS (inductively coupled plasma mass spectrometry), high sensitivity technique for the measurement of uranium and thorium isotopes in urine, was available in three facilities included in the EURADOS Catalogue (2001), but no further information was supplied about its performance or procedures. Other indirect determinations of Actinides are carried out by Fluorimetry, Kinetic Phosphorescence Analyser (KPA) for uranium measurements, liquid scintillation counting (LSC) (application to alpha emitters), LABSOCS (Laboratory Sourceless Object Calibration Software) and Waterscintillator, and Gamma spectrometry.

Urine analysis of 24 h samples are performed by Alpha spectrometry with typical MDAs in the range of 0.1–0.5 mBq d⁻¹, with the counting time varying from 220 000 to 430 000 s. A second group of laboratories reported MDA values in the range of 0.1 (500 000 s) to 0.5 (250 000 s) mBq l⁻¹ of Actinides in urine. MDA values close to 0.1 mBq d⁻¹ are recommended for the determination of Actinides in urine. Passivated Implanted Planar Silicon (PIPS) detectors are widely used in Alpha spectrometry.

Normalisation of creatinine content in urine was only reported by two facilities. With reference to the collection of faecal samples, 3 d faecal samples or single-voiding are alternatives to 24 h samples in few instances. The radiochemical Actinides separation methods reported in some cases are extraction chromatography and ion exchange chromatography.

In vitro determinations of pure beta emitters in excreta (25 laboratories)

LSC (19 facilities) and proportional counters (six laboratories) are the techniques used by services for *in vitro* determination of beta emitters in excreta. Typical radionuclides to be evaluated are ^3H and ^{90}Sr with ^{14}C , ^{32}P , ^{35}S , ^{89}Sr , ^{90}Y , ^{125}I and ^{45}Ca also being of interest.

Agreement can be seen in relation to the volume of the sample required to measure ^{90}Sr in urine (24 h samples), but there is little harmonisation for the determination of ^3H in urine, with volumes varying from 10 ml to 1 litre. The preparation of the source for measurement (radiochemical separation, etc.) vary considerably among the laboratories. The sensitivity data reported by services show that

typical MDAs are in the interval 25–250 Bq l⁻¹ of ³H in urine using LSC technique; lowest MDA reported is 1.2 Bq l⁻¹ for a counting time of 100 000 s.

Gamma spectrometry determinations (17 laboratories)

HPGe detectors are widely used by services for *in vitro* determinations of gamma emitters in excreta. Ge(Li) and NaI(Tl) detectors are also employed. The volume of urine and faeces being collected covers a 24 h sampling period. The MDA values obtained by laboratories are ≤1 Bq for ¹³⁷Cs, ⁶⁰Co and ¹³¹I. Differences are found in the counting time selected by facilities with resulting MDAs varying from 0.01 to 1 Bq for typical gamma emitters.

PAS/SAS monitoring

PAS implies the collection of a sample representative of the activity concentration in the air inhaled by the worker. This allows an estimate of the intake of some typical radionuclides. To know or to make realistic assumptions about the particle size distributions in the inhaled material is required for the correct estimation of internal exposures.

SAS are commonly used to monitor workplace conditions, but can underestimate concentrations in air in the breathing zone of the worker [ICRP 78⁽⁷⁾].

Thirteen European laboratories (eight SAS and five PAS) included in the EURADOS Catalogue perform PAS/SAS monitoring. Some alpha, beta or gamma emitters can be evaluated by PAS/SAS measurements, particularly ¹³¹I, uranium, thorium and plutonium isotopes. The results are not always used for individual dose evaluation. Different MDA values are obtained in each laboratory depending on the radionuclide and type of monitoring selected. An important contribution was the information collected from UK Services, who agreed to share with EURADOS the information supplied to the OMINEX Project. A typical MDA value supplied for PAS monitoring, gross alpha is 0.02 Bq which applies to U, ²³²Th, ²²⁸Th and Pu-alpha.

ASSESSMENT OF INTERNAL DOSES IN EUROPE

The doses received by workers travelling and working at different European facilities have to be assessed in a similar way by internal dosimetric services. The design of individual monitoring programmes to evaluate occupational internal exposures depends on the annual dose limit in place. Of the 26 participating countries registered in the EURADOS Catalogue, 60% have implemented an annual dose limit of 20 mSv, and 40% established a limit of 100 mSv in 5 y with a maximum dose of

50 mSv y⁻¹. In this last instance, an operating value of 20 mSv as an annual average limit for 5 y was reported in some countries.

With reference to the record keeping of dose evaluations, 14 out of 24 countries have National Record of Doses (status in 2001). It is highly recommended that in the future each country sets up a central location for record keeping of internal doses. It is further proposed here that a Europe wide Record of Individual Doses be set up in the future.

The software available to European services for internal dose assessments (status 2001) includes the codes IMBA (Integrated Modules for Bioassay Analysis), LUDEP (Personal computer Program for Calculating Internal Doses), IMIE (Individual Monitoring of the Internal Exposure), Mondal/Moldes, MIRD (Medical Internal Radiation Dose) software, INDAC (Internal Dose Assessment Code), Cindy, Remedy and Retex. It is important to remark that not all of them operate with the latest ICRP recommendations as set out in the EU Directive 96/29. An important piece of work has been carried out by the IDEAS⁽⁸⁾ Project which includes the outcome of a Dose Intercomparison exercise. An objective of the IDEAS Project is the development of general guidelines for the estimation of committed dose from incorporation monitoring data.

In relation to the internal doses reported by services, a European reference level of 'lowest reported dose' of 0.1 mSv could be established as an average value of the data collected in the EURADOS Catalogue, but currently harmonisation does not exist. Some internal dosimetry laboratories calculate all doses obtained from measured activity above MDA. Upper limit of minimum reported dose is 2 mSv. The same lack of agreement occurs in the Investigation Level (IL) where European countries selected very different IL, from 0.1 to 6 mSv.

A brief summary of legal aspects, other matters and requirements employed in the management and operation of internal dosimetric services of some selected countries are reported as follows.

Sweden

EU Directive is implemented in Sweden since 1998 (SSI FS 1998:5/2000:10 Guide). The National Regulatory Body is SSI (Swedish Radiation Protection Authority) Institute. Scandinavian countries (Norway, Sweden, Denmark and Finland) are usually very well coordinated and harmonised in dosimetric matters.

With reference to the assessment of internal exposures, monitoring programmes in Sweden are established for three worker categories: Group of reference, Group of high risk of intake and Incidents with known intake. Twelve institutes are performing internal dosimetry (NPP, NFC, medical field, research and decommissioning), and no legal

approval of services is required. WBC monitoring shall be performed according to the documented procedure that is approved by SSI (SSI 2000:10).

The Radiation Protection Authority in Sweden is capable of evaluating internal doses, but the responsibility for that is, under normal conditions, given to services. Dose Recording Level is 0.25 mSv (minimum dose recorded by services) but the report to National Dose Register is carried out if $E_{50} > 1$ mSv. For doses $E_{50} > 5$ mSv, immediate communication of the incident to SSI is required.

Germany

Implementation of EU Directive in 2001 (Radiation Protection Ordinance). The term 'Regulatory Body' in Germany applies to federal and state authorities but the Accreditation of laboratories is carried out by state government. The more relevant guidelines for individual monitoring are 'Physical Rad. Monitoring (methods, intervals, action levels, etc.)', 'Determination of body doses caused by internal exposure' and 'Performance requirements for dosimetry laboratories'.

There are 25 institutes performing internal dosimetry in Germany and one National Dose Register. Monitoring programmes of workers for internal exposures have to ensure the detection of 1 mSv as annual dose. All the doses are reported, and the Investigation Dose Level in Germany is 6 mSv.

Hungary

Hungary is a new EU member state and has already implemented EU Directive. The Ministry of Health acts as Regulatory Body and the setting up of a National Dose Register is in progress. The laboratory performing the monitoring of internal exposure shall be accredited; there are three accredited internal dosimetry services in Hungary. One is legally commissioned for country record keeping.

Important institutes in Hungary for internal dosimetry matters are the National Research Institute for Radiobiology and Radiohygiene (NRIRR), which performs *in vivo* and *in vitro* measurements (monitoring of workers of Nuclear Power Plants, Nuclear Fuel Cycle, research and medical field), and the Atomic Research Institute (KFKI), with the capability of *in vivo* monitoring for the whole centre and upon request for other institutions as well as for the public. The lowest reporting dose in Hungary is 0.1 mSv and the IL is 6 mSv.

SUMMARY

An important network of services from 26 countries has been established for the harmonisation of individual monitoring of internal exposures. The

information collected from 73 internal dosimetry services provides an overview of the capability and actual status of techniques, methods and available tools for the assessment of internal exposures in Europe. The EURADOS Catalogue was published⁽²⁾, thus sharing with the dosimetric community the information collected.

Harmonisation is a reality in many aspects of internal dose assessments, but not in the overall process. Looking into the future, it is highly recommended the focus be placed on the lack of dosimetric tools and (methods used by many services) codes in many services to calculate effective doses using the latest ICRP models and effective regulation. The performance of a reference European internal dosimetry service has to guarantee adequate sensitivity of the measuring techniques and the accuracy of the results of Committed Effective dose. There is still a long way to go. As final proposal here it is suggested that a way forward might be the setting up of an European Accreditation Process for European Laboratories dealing with the assessment of internal exposures.

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