Executive Summary

Contract No: FIKR-CT2001-00164 — **IDEA**

Title: **Internal Dosimetry — Enhancements in Application**

Introduction

This project is all about one idea. Numerous methods are well known and established in laboratories which could enhance internal monitoring techniques – in principle. However, these benefits are not available to occupational internal dose assessments, because the applicability of such methods for routine application has not been demonstrated sufficiently. These enhancements in application of internal dosimetry – both in accuracy and speed – were the overall objective of this project.

Therefore the project aimed to provide faster and more reliable, i.e. demonstrably proven, monitoring techniques (both in-vivo and bio-assay) for better operational monitoring. The project did not aim primarily at the development of new methods. In order to provide enhanced protection in the field of internal exposure it is of potential importance

- to verify and apply new measurement concepts,
- to enhance existing measurement concepts,
- to improve calibration techniques for individual assessment, and
- to apply better monitoring and calibration techniques to reduce uncertainties in the evaluation of whole and partial body activity and individual intakes for dose assessment.

Objectives

This project deals with the methodological improvements on monitoring techniques of incorporated radionuclides and with recommendations based on the results of works achieved in the frame of the EC-funded IDEA project FIKR CT2001 00164. This project gives a brief overview of the newly developed laboratory methods and provides information on the potential improvements to the currently used techniques. The main aims were to increase monitoring sensitivity and reduce uncertainty considering also the cost-benefit relations as well as the real need of the given application. The work was concentrated to the following three main areas:

- Optimisation of in-vivo monitoring techniques by selection of the most suitable detector and monitoring arrangement;
- Improvement of efficiency calibration technique in in-vivo radioactivity monitoring with special attention to numerical calibration methods by Monte Carlo simulation;
- Improvement in bioassay monitoring of excreta using IC-PMS methodology applied for selected radionuclides like uranium, thorium and actinides.

Before summarising the main conclusions on these three areas it has to be generally stated that the selection of measuring method and equipment should be based on realistic needs and on financial and technical resources available for the given monitoring purpose. From the point of view of the necessary cost, careful consideration should be given to the realistic requirements to avoid introducing more sophisticated methods and installing more complex facilities than the monitoring task and programme can justify.

Results

In-vivo monitoring technique

The conclusions for the selection of the most suitable detector for a certain purpose in in-vivo counting can be summarised in the followings. In the range of medium and low energy photons the detector should be as thin as possible considering the energy of interest. If the source is positioned in front of the detector the detector should have a large front surface as possible. The best spectral resolution can be achieved using semiconductor detectors like Si(Li), HPGe, pure Si and CdZnTe detectors, among which HPGe crystals can be produced with the largest surface. Especially in case of complex gamma spectra large NaI(Tl) detectors cannot compete with the Ge diodes.

In order to increase the monitoring sensitivity or in other words to reduce the Minimum Detectable Amount besides improving the counting efficiency it has primary importance to reduce the detector background namely to increase the signal/background ratio or the so called Figure of Merit. The background continuum has essentially no spectral feature while the photopeaks are due to specific radionuclides and represented by characteristic peaks in the background spectra. To be able to reduce the ambient background the application of proper shielding is required. The requirements for primary shielding materials are high attenuation of gamma rays, requiring high atomic number and density; with sufficiently low concentrations of natural or artificial radionuclides as impurities. Steel or lead is the most commonly used materials. If lead is chosen, the characteristic X rays induced by ambient radiation can be removed by a lining of 2 to 6 mm copper. For major installations typical thickness for the primary material is 50-100 mm lead or 100-200 mm steel. The most effective arrangement is a wholly shielded large enclosure comprising both the subject and the detector(s) used however there are also more open structures, which eliminate direct paths for radiation between the detector and the surrounding environment. While the former kind of shielding is to be used for low energy photon emitting radionuclides, the latter can be applied for nuclides emitting photons above 200 keV. Another way of reducing detector background is to apply active shielding. The so-called anticoincidence techniques can be an effective way to reduce those parts of background produced by photon scattering in the detector at self. The applicability of this technique depends very much on the decay scheme of the radionuclide in question (e.g. ^{60}Co).

Generally it is substantial prerequisite to make careful selection of shielding and constructional materials to achieve as low background contribution as the measuring task requires. In general routine monitoring of fission or activation products does not require high efficiency measurements and very effective shielding but high energy resolution capability might be needed, while to measure actinides for instance in the human lungs needs to apply as high efficiency detection arrangement as possible and large sophisticated shielding enclosure.

In-vivo calibration technique

Reliable efficiency calibration of measuring systems is one of the most crucial tasks in in-vivo monitoring. Therefore calibration technology is included in the important areas for reduction and optimisation of the systematic errors and for improving the accuracy in the interpretation of measured in-vivo data. Currently used phantoms are usable for common geometries and whole-body measurements but are limited in the fine structure and individual variations necessary for low energy application. In the frame of this project the main aim was to investigate the capability and applicability of numerical calibration method in routine in-vivo monitoring procedures, therefore this report gives a short overview on efficiency calibration methods using physical phantoms and deals with computer simulation in more details.

The traditional method applied for the calibration of body radioactivity monitors is based on physical phantom calibration. Some practical information on the most frequently applied phantoms are summarised in the report considering also that these phantoms can be regarded as references for the numerical phantom calibration method.

For uniformly distributed sources in the whole body the BOMAB type phantom consisting of 10 cylindrical and elliptic-cylindrical shaped vessels made of polyethylene are the most widely used phantom in the photon energy range exceeding 100 keV. These vessels can be filled with liquid containing known amount of radioactive solution that can be solidified by forming stable gel or other solid polymer inside the phantom. Based on similar principle the use of whole-body phantoms assembled with 1-2 litre commercially available bottles is very simple and practical. One can easily simulate very different body sizes and statures in arbitrary monitoring geometry. The cost of such bottles is very low so it is not necessary to refill them after a single use. The comparison measurements on counting efficiencies of bottle phantoms to those obtained with the use of BOMAB phantoms in corresponding sizes showed an agreement within 5-10% in the photon energy range above 100 keV. In the last years more and more whole-body counter laboratories especially in Europe are using the IGOR phantom for the calibration of uniformly distributed sources in the photon energy range above 60 keV. The phantom is made of polyethylene bricks and the radioactive sources are made of 2 cylindrical rods for each brick containing a solution stabilised in a special resin. The study on comparing the IGOR phantom with the BOMAB phantom showed good agreement, so their simulation of uniform activity distribution in the whole body is similar. By means of the IGOR phantom non-uniform activity distribution can also be simulated.

Livermore chest phantom is the most elaborated phantom that is used since long for in-vivo determination of transuranic radionuclides emitting low energy photons and deposited in the lungs, tracheo-bronchial lymph nodes and in the liver. The phantom is anthropomorphic and the different tissue substitutes were very carefully prepared. There are available chest overlays in different thickness and in three adipose-muscle tissue compositions in order to represent various chest wall thickness. The radioactivities are uniformly distributed within the organs. The phantom is especially useful for dual-detector systems in close counting geometry. The JAERI chest phantom was constructed on the basis of the experiences gained during the use of the Livermore phantom. The JAERI phantom represents the average adult Japanese male, but can also be applied for most ethnic groups in Asia. Similar to the Livermore phantom an overlay plate can also be applied to be able to fit better to the chest wall thickness of the person.

Because in reality the deposition is heterogeneous in the subject, this can result in large uncertainties in the assessed activity. In consequence, significant corrections may need to be made to phantom-based calibration factors in order to obtain absolute calibration efficiencies applicable to a given individual. The importance of these corrections is particularly crucial for in-vivo measurements of low energy photons emitting radionuclides, such as actinides, deposited in the lung. The best solution currently is the use numerical calibration method using Monte Carlo simulation.

A software system called OEDIPE has been developed and described in the report for covering all necessary steps required by the calculation procedure namely creation of numerical phantom (for voxel phantom by image processing after CT or MRI scan on the phantom), source description, detector characterization, phantom-detector positioning, detector response calculation by Monte Carlo code and comparison of efficiencies obtained by calculation and measurement.

The applicability of the numerical calibration method has been tested by detailed validation programme. The results of the validation procedures on the IGOR phantom family and whole-body Bottle phantom in the photon energy range of 500-1500 keV and 80-1350 keV respectively, showed less than 15% deviations between measurements and calculations. Similar results could be obtained in case of the Livermore chest phantom and in the photon energy range of 50-200 keV that is very promising for further applications.

Comparison of simulated counting efficiencies obtained on different chest phantoms and on a real patient resulted in different values that underlines the importance of uncertainties even in higher photon energies which can be reduced by using individually specified computational phantoms.

The major source of uncertainties using physical phantom calibration in measurement of actinides in the human lung is that neither the individually specific lung and chest wall geometry nor the real activity distribution can be simulated properly. The numerical calibration method provides essential improvement in the accuracy of these measurements.

There are several very promising potentials in the application of the numerical efficiency calibration method. One application possibility is to use the method for finding the optimum source-detector geometry in in-vivo counting. Promising results can also be expected in introducing the individual specific calibration of chest counting arrangements for low energetic photon emitters using voxelized phantom obtained from CT or MRI images of the human subject to be measured. The systematic investigation of the effect of non-uniform activity distribution within the lung on the counting efficiency can provide information on the possible level of uncertainties in internal exposure assessment. In this way the uncertainty in exposure assessment due to inhaled radionuclides with special attention to transuranic elements could be reduced.

A very promising application is expected for deriving the counting efficiency by numerical simulation considering the redistribution due to the biokinetics of the given radionuclide in the body. This will lead to the application of a time-dependent efficiency governed by the biokinetic behaviour of the radionuclide.

New possibilities are provided in the application of numerical reconstruction of any body part in order to localise and quantify radioactive contamination in wounds. The method is capable not only to the calculation of the activity but also to the assessment of received dose.

Bioassay monitoring technique

Bioassay monitoring techniques estimate the activity of the incorporated radionuclides in the body by analyzing biological samples excreted or removed from the body. Radiochemical procedures are used to reduce the various sample matrices to a form suitable for activity measurements. The most commonly used activity measurement technique for the determination of alpha emitting radionuclides is alpha spectrometry. However mass spectrometry, and especially inductively coupled plasma mass spectrometry (ICP-MS), has evolved as an attractive alternative to alpha spectrometry and beta counting in bioassay monitoring. The work carried out in this project provided acceptable guidelines for optimum performance of ICP-MS measurements of U, Th and certain actinides, including sampling procedure, operational parameters of the instruments, and interpretation of the measured data.

Unlike in alphaspectrometry, the lower limit of detection in activity concentration obtained with ICP-MS measurement depends on the half-life of the radioisotope, and in particular decreases as the half-life increases. This trend is due to the physical principle of ICP-MS. This very sensitive technique allows for the detection of atoms present during a given counting time.

The lower limit of detection for natural uranium isotopes is generally of the order of 20 pg or about 0.5 µBq. A great advantage of ICP-MS over the currently used standard bioassay monitoring methods is the low measurement time required. Measurements of urinary uranium by ICP-MS require relatively small sample volumes of a few ml, however based on such spot samples there are large uncertainties due to the individual differences in daily urinary excretions which is the basis of dose assessments. Therefore, in the frame of this project the daily urine excretion volume was investigated in detail on "normal" German subjects and found to be 1709 ± 665 ml/d and 1606 ± 675 ml/d for males and females respectively. It is recommended that 24-hrs urine sampling should be carried out and wherever feasible, three consecutive days urine sample may be collected to avoid large uncertainties in the quantitation of daily urinary excretion of U. When the concentrations in the sample are too low for a direct measurement of uranium by ICP-MS, selective separation and pre-concentration may be necessary. Results of validation measurements showed good agreement with those obtained by other methods. Monitoring occupational exposures requires the knowledge of baseline levels for non-exposed subjects. Urinary excretion measurements have been performed on more than 1300 non-exposed subjects of both genders and were found to be a variation within the range of 10-30 ng/d total daily excretion. This value may change from day to day by up to a factor of two or even more for a given individual. The results of this study demonstrate significant regional variations of daily urinary uranium excretion due to differences in environmental uranium concentrations and also due to dietary habits. ICP-MS measurements are simple, rapid and economical.

Similar to uranium, the measurements of thorium by ICP-MS require relatively small sample volumes, however 24-hrs urine sampling should be conducted. Though in principle, elemental concentrations in aqueous solutions can be measured directly by ICP-MS without any previous sample preparation, for the measurement of thorium by ICP-MS salt removal from the urine samples could be advantageous. During validation the results obtained on Th excretion values in the same urine samples using radiochemical neutron activation analysis, were similar to that obtained applying ICP-MS. Interpretation of thorium measurements of occupational exposures also requires the knowledge of baseline Th excretion levels for non-exposed subjects. The day-to-day variations in the urinary excretion of thorium were investigated on 15 non-exposed subjects. There are considerable individual differences found in the total daily excretion (in ng/d) and these values changed from one day to another by a factor of two or even more. An average value of (1.81 ±1.49) ng/d thorium baseline level could be derived from the study.

For reliable measurement of ^{239}Pu by ICP-MS, the radionuclide in the sample has to be pre-concentrated and then purified to be able to separate it from uranium. Therefore at least 24-hrs urine sampling should be conducted. In view of the requirement of very low quantity of ^{239}Pu to be measured in urine, which corresponds to 1 mSv exposure, therefore the concentrations in aqueous solutions can not be measured directly by ICP-MS, consequently the pre-concentration of ^{239}Pu from urine samples and its subsequent purification is required.

Summarising the findings on bioassay investigations the application of ICP-MS measurements are relatively simple, rapid and cost-saving. Applying new improved measuring techniques (HR-SF-ICP-MS), detection limits of 0.01 ng/L for ^{238}U, 0.25 ng/L for

^{235}U, 0.1 ng/L for Th and 0.001 pg/L for ^{239}Pu in urine samples can be achieved. The analytical capabilities of ICP-MS studied so far seem to provide sufficient proof that this method has potential to apply for the member of the public and to become as a routine technique to monitor workers for most of the radionuclides incorporated in the body.

Implications

The goal was to disseminate the findings to the routine monitoring community, which was done at the *European Workshop on Individual Monitoring* (Vienna 2005). It can be anticipated that some – if not all – recommendations for improvement will be implemented to improve sensitivity, accuracy, or cost/benefit of monitoring methods.

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