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IMPROVEMENT OF METHODS USED IN INTERNAL DOSIMETRY

PhD thesis booklet

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INTRODUCTION AND RESEARCH BACKGROUND

The determination of internal dosimetry requires the application of knowledge, tools and methodologies for the estimation of radiation exposure resulting from incorporated radioactivity. Radioactive material can enter the human body via inhalation, ingestion, or through intact or wounded skin. It is crucial to assess the resulting internal dose to occupational workers or members of the public. Although the committed effective dose cannot be measured directly, it can be estimated using mathematical models once the quantity of radioactive isotopes in the body has been determined through measurement. The level of intake depends on various factors, including the physico-chemical form of the radioactive material, as well as the time, mode and route of intake. To determine internal exposure, it is essential to use up-to-date and accurate knowledge available to assess the conditions of intake, the biokinetics (retention) of radionuclides, the energy transfer to organs and tissues by radiation, and the stochastic or deterministic effects of radiation.

The determination of the quantity and quality of radioisotopes in the human body using direct (*in vivo*) methods, primarily γ-spectrometry, is not fundamentally different from other measurements performed on prepared samples. However, standardising the size and shape of these samples poses a significant challenge. The generally unfavourable and individually variable measurement geometry and the limited measurement time often lead to a greater measurement uncertainty compared to that of inanimate samples. Despite these challenges, appropriate measurement of radioisotopes in the human body requires careful consideration of differences in body sizes and the location of internal organs of individuals. In recent years, there have been gradual improvements in both measurement equipment and methods. By accounting to anatomical differences between genders and age groups, physical phantoms are being developed to model the effects of different body geometries on the measurement results. This advancement allows for a more precise calibration of the measurements.

In general, the lower the energy of γ-radiation, the more important it becomes to consider the individual characteristics. When monitoring of uranium and transuranic isotopes (e.g. ^{239}Pu, ^{241}Am) in the body, both the design of the measurement technique and the quantification of these isotopes require thorough preparation. A key aspect of this preparation is the calibration of the measurement system using physical or numerical phantoms. Low-energy photons are significantly attenuated by materials between the source and the detector, including body tissues. Therefore, the use of realistic anthropomorphic phantoms that mimic the structure and density of human tissues or numerical models allows a more exact accounting for the photon's path through the body.

Part of the body measurements (e.g. lungs, skull, liver or thyroid) are used to identify and quantify the radioisotopes that reside in specific organs for extended periods. The aim of these measurement is to accurately determine the activity of the radioisotopes within the target organ, sometimes taking into account the contribution of signals from activity in other parts of the body. A substantial proportion of the calibration phantoms are designed based on an average body size and generally model a uniform distribution of radionuclides within each organ. However, in reality, the amount and distribution of radioactive material within each organ are not uniform and are influenced by complex physiological processes such as metabolism, inter-organ contact and tissue retention.

Age-related characteristics, such as the age-specific size of organs or the age-dependence of metabolic processes, must also be taken into account in these measurements. In recent years, particular emphasis has been placed on the monitoring and assessment of thyroid doses using the ^{131}I isotope. Since the size and position of the thyroid changes with age, calibration phantoms used to determine

measurement efficiency should be adjusted accordingly, especially when determining the organ dose of children.

In my research, I have shown in detail that to improve the accuracy of *in vivo* measurements for determining internal doses, calibration procedures must be developed that take into account several factors: the specific isotopes to be measured, the measurement geometry, the characteristics of the measurement device, the organs to be measured, and the age of the individual.

Indirect (*in vitro*) measurements of internal dosimetry involve the analysis of biological samples, such as excreta (urine, faeces), blood or nasal secretions, to determine the concentration of radioactive material present in the body, allowing the determination of the committed effective dose. Selecting the appropriate measurement method requires knowledge of the physical and chemical properties of the compounds carrying the radioisotopes to be tested, as well as their metabolic behaviour in the body. These measurements are either complementary to the results of whole-body and part-body counts or are necessary when dealing with short-range radionuclides (pure alpha or beta emitters, or isotopes emitting very low-energy X-ray or γ-rays), for which direct quantification is not feasible by other means. When dealing with complex biological samples in indirect dosimetry, several factors can affect the accuracy and reliability of measurement results. The chemical stability of the substances in samples may change during storage, transport and preparation. The sampling conditions also play a crucial role, as difficulties of sample collection and potential cross-contamination of the sample can impact the results. A further complicating factor is that the reproducibility of measurements is generally lower than that for other inanimate samples due to ongoing metabolic processes in the body.

An important factor that can affect the accuracy of measurement results in liquid scintillation (LSC) measurements is the proper knowledge of counting efficiency. A well-known difficulty in detecting tritium (^{3}H) in urine is the quenching effect, which leads to a reduction in scintillation light intensity and a shift in the measured spectrum. Several methods can be employed to obtain accurate efficiency in LSC, the most common ones being proper sample preparation, different calibration methods (such as standard addition and quench curve fitting), and the optimisation of the scintillator composition.

In addition to the measured data, additional information about other parameters characterising the intake situation is required for the calculation of intake and committed effective dose. The most essential ones are the time elapsed since exposure, the mode of exposure, the physical and chemical form of the isotope of the intake, and individual characteristics. It is important to emphasize that all this information and the quantities used in calculations are subject to uncertainties

The inaccuracy of parameter values in biokinetic models may also be due to model uncertainties and individual variability. One source of uncertainty is that these models were developed for *Reference Humans*, whose physical characteristics may differ substantially from those of the person being measured. Another source of uncertainty is that a person's biokinetic characteristics (such as the amount of urine or faeces excreted) can vary significantly over time. The reliability of the biokinetic models used is also affected by the uncertainty in the source data upon which the models are based. Ideally, biokinetic models are based on human data, but in cases where human information is not available, it is often supplemented or replaced by data from laboratory animals or chemical analogues. This substitution introduces further uncertainty, particularly due to challenges in extrapolating biokinetic data between mammalian species. Moreover, there is no empirical evidence that chemical analogues have similar physiological and biological properties for all isotopes. The latter adds to the uncertainty, particularly in the analysis of lanthanides and actinides.

The usefulness of measured data for dose estimation may also be affected by the use of decorporation treatments designed to accelerate the excretion of radioactive material after exposure. One type of such a treatment is the administration of DTPA (diethylene-triamine-penta-acetic acid), which is mainly used to treat internal contamination caused by actinides (e.g. ^{241}Am), due to its ability to form water-soluble complexes with actinides, thereby facilitating their more rapid excretion from the body. The treatment modifies the recommended retention function of radioactive material in the body and the default biokinetic models cannot be directly applied due to the faster excretion process. As a result, the calculation of the committed effective dose must take into account the modifications introduced by the DTPA treatment and should rely on a new retention function specific to the exposure case and the individual receiving the treatment. When interpreting data from urine measurements, it is crucial to account for the timing and frequency of DTPA treatment. After DTPA treatment, urine activity may initially increase significantly, reflecting the amount of radioactive material removed from the body and providing valuable insights into the treatment's effectiveness. The amount of radioactive material remaining in the body can only be determined by analysing samples taken some time after treatment, once the activity in the urine has decreased to approximately the same level as that without treatment.

Recent intercomparison exercises (ICIDOSE, IAEA-IDEAS) have also highlighted the difficulties of internal dosimetry. The use of biokinetic and dosimetric models may require expert decisions and judgements that may lead to differences in the estimated dose. The results of dose estimates from the same data set may in some cases exhibit significant variability, depending on different assumptions, the experience and skills of the analyst expert, and the hardware and software tools used.

The ongoing task of dose assessment experts is to harmonise dose estimation practices so that internal exposure results are comparable in terms of accuracy and reproducibility, similar to the standards achieved in external dose measurements. For instance, when two individuals are exposed to the same external radiation field, their dosimeters will expectedly yield consistent results, providing the best estimate of the dose received. A similar objective is pursued in the determination of internal contamination: starting from the same measurement results, the analyses should provide nearly identical results for the dose assessment. The reliability of the results should therefore be prioritized in the determination of internal dosimetry to the same degree as it is in external dosimetry.

OBJECTIVES

In developing methods suitable for internal dose assessment, special attention must be paid to two fundamental factors: the optimisation of the measurement methods and the investigation of the uncertainties of the parameters used for dose estimation. The optimisation procedures and the management of uncertainties are highly dependent on the specific isotope, the measurement methods and the tools used. For this reason, it is essential that the measurement and modelling methods are carefully adapted to the conditions and physico-chemical properties of the isotopes involved.

In the course of my work, the aim of my research is to further develop the measurement methods and models used in internal dosimetry by carefully studying the entire process, which includes several measurement techniques and the key aspects of dose estimation. With these advancements, I aim to enhance the sensitivity of measurements, reduce measurement uncertainties and improve the accuracy, reliability and reproducibility of dose estimates.

Lung measurements are particularly important for low-energy isotopes (e.g. ^{241}Am), as the radiation attenuation caused by the tissues in front of the organ is significant at this energy range. The aim of my work is to improve the accuracy and reliability of lung measurements for these isotopes, taking into account both the radiation attenuation effect of surrounding tissues and the influence of radioactivity present in other organs in the chest. To optimise the measurements, I looked for a measurement position that would minimise cross-effects during the chest measurements, particularly when the location of the radioactive material within the organ is unknown and only a single measurement can be performed.

In the event of an accident, the inhalation of radioiodine isotopes released into the atmosphere has the potential to result in their accumulation within the thyroid gland, where they may cause substantial radiation exposure. The size and location of the thyroid varies with age, and thus it is of particular importance to consider these factors when determining the organ dose in children. In order to ascertain the influence of each parameter on the measurement uncertainty and to optimise measurement procedures and a more precise estimation of the organ dose, sensitivity tests were conducted utilising diverse measurement geometries and phantoms on the available thyroid measurement equipment.

As I have already mentioned the ^{3}H isotope in urine is usually measured using a liquid scintillation measurement system, where one of the critical factors is the consideration of the quench effect. In my thesis, I investigated the effectiveness of different calibration methods aimed at eliminating or accounting for the quench effect without the need for time-consuming sample preparation of urine samples. My aim was to develop a method that would remain accurate in the presence of the quench effect, even for complex samples, thus improving the reliability and efficiency of measurement.

Based on the measurement data, biokinetic models must be used to determine the committed effective dose, taking into account the metabolism, absorption, distribution and excretion of the given isotope. However, suitable biokinetic models for the behaviour of the absorbed radioisotope in organic compounds may not be available in the literature. In the course of my work, I have investigated how the committed effective dose from internal exposure can be determined in such cases and have shown the extent to which various factors - such as inaccuracies in biokinetic models, unknown exposure times, and measurement uncertainties - influence the result obtained.

In some cases, the use of decontamination agents, such as DTPA, has the potential to mitigate the internal radiation exposure associated with specific isotopes. However, it should be noted that these treatments may potentially alter the correlations between successive measurements, which may in turn affect the fit of biokinetic models to the data. In my research, I examined the impact of DTPA treatment on the usability of urine measurement data following ^{241}Am incorporation. Additionally, I explored the "best estimation" for committed effective dose estimation by integrating diverse measurement outcomes and adjusting the specific absorption parameters associated with the intake.

The objective of this study was to enhance the precision and dependability of dose estimation models by optimising the measurement data to facilitate more accurate internal radiation exposure determination.

NEW SCIENTIFIC RESULTS

THESIS STATEMENT #1. In accordance with the characteristics of the instruments available in the laboratory, I developed a measurement protocol to determine the activity of the inhomogeneous distribution of the ^{241}Am isotope in the body. I have demonstrated established that in the case of partial body measurements of the lungs performed with an HPGe detector, after the intake of radionuclides emitting low-energy γ-rays (e.g. ^{241}Am), the counting efficiency determined by calibrations with phantoms of different designs (different isotope distribution, tissue-equivalent materials of different thicknesses) can deviate up to 10% at the given energy. I have shown that the thickness of the chest wall is a particularly critical factor, and that by accounting for this parameter, the resulting measurement uncertainty can be reduced by 15-50%, thereby improving the precision of dose estimation. [P1] [p1] [p2] [p3]

THESIS STATEMENT #2. I have determined that the radioisotopes in certain organs of the chest can have a significant cross-effect on measurements made above other organs: the radioactivity in lungs has a relative cross-effect of 20-40% on measurements made over the liver, whereas the activity in the liver can cause a cross-effect of less than 10% on measurements taken over the lungs. I have shown that positioning the measurement apparatus above the sternum reduces the sensitivity to uncertainties caused by the cross-effect. By applying the method I have proposed and developed for this specific measurement geometry, the accuracy of internal dosimetry-related measurements with a whole-body counter can be significantly improved, thus increasing the reliability of the measurements and the robustness of decisions concerning radiation exposure. [P1] [p1] [p2] [p3]

THESIS STATEMENT #3 Using my own measurements and Monte Carlo simulations of numerical models developed for detectors and thyroid phantoms available in the laboratory, I have shown that, in the case of thyroid measurements, phantoms with different physical designs used for efficiency calibration cause a difference of no more than 3% in the final result. I have also found that in the case of the thyroid gland, the efficiency calibration with adult-sized phantoms introduces a systematic error of about 10% in the results when applied to measurements for 5-year-old children's thyroids. The increase in tissue thickness per millimetre in front of the thyroid organ when measuring adult, adolescent and child thyroid phantoms causes a reduction in measurement efficiency of 1.5-2.5%, 1.0-2.5% and less than 1.0% respectively, depending on the distance between the detector and the neck. [P2] [P3] [P4] [p4] [p5] [p6] [p7] [p8]

THESIS STATEMENT #4 When determining the ^{3}H activity concentration in biological samples by liquid scintillation measurement, different methods are used to calculate the efficiency, of which standard addition and quench curve fitting are the most commonly used: I have shown that these methods can cause a difference of up to 10% in the efficiency result. Knowing the advantages of the two methods and the disadvantages experienced with complex samples, I have developed a new combined method for determining the efficiency of ^{3}H measurement. The essence of it is that the real measurement samples, which are assumed to have similar chemical properties, are first spiked by adding known activity according to the standard addition method, and then we produce pairs of tSIE - counting efficiency values from this group of samples. A

simple function (a regression line in the case I studied) is fitted to the pairs of values in a narrow tSIE range, so that we obtain a calibration function from which we can read the corrected counting efficiency for the tSIE value of the sample currently examined. I have demonstrated that the use of the combined method is advantageous because it compensates for the various sources of uncertainty, thereby increasing the overall measurement precision, especially in the case of complex samples. I verified the applicability of the method and its advantages in intercomparison exercises using real water samples. [P5] [p5] [p9] [p10] [p11]

THESIS STATEMENT #5 The models recommended in the literature for dose estimation of incorporated ^{14}C under indeterminate intake conditions are not always sufficiently accurate, though they provide a general indication of the need for further investigation. With the availability of a large amount of urine measurement data obtained in an incorporation event, I have developed models based on direct retention functions that allow more case-specific dose calculations. I showed that the committed effective dose obtained using current literature methods was nearly 50 times higher than the case-specific dose estimate for the studied scenario. I have pointed out that the case-specific approach remains sensitive to the appropriate choice of parameters that cannot be measured during the investigation. For example, selecting a urinary excretion fraction of 0.9 reduces the estimated effective dose by about 40%, while reducing the urinary excretion fraction to 0.1 increases the estimated effective dose more than fivefold compared to the committed effective dose obtained by a factor of 0.59, taken from the literature. [P6] [p12]

THESIS STATEMENT #6 I have determined the effect of the DTPA decorporation treatment performed on the recommendation of the doctor after the accidental incorporation of ^{241}Am on the urine measurement results and developed a model for the appropriate selection of the data. I have found that using the measurement results from urine samples collected within 50 days after DTPA treatment leads to highly inaccurate dose assessments. I have demonstrated that in order to refine the case-specific estimates, it is necessary to combine the lung and urine measurement data and apply the suitable absorption parameters, to ensure that the uncertainty of the dose estimate does not exceed 10%. [p13] [p14]

EXPLOITING RESULTS

My research in recent years has identified some critical points in the implementation of internal dosimetry and demonstrated that, among other things, increasing the precision of measurement alone does not always lead to a meaningful improvement in the dose estimation result. I have shown that developing optimised isotope- and device-specific strategies is crucial to address the major sources of uncertainty in a targeted way, while also ensuring practical applicability of solutions. Based on the knowledge gained from this research, dose estimation procedures can be developed that more accurately consider individual human factors and case-specific parameters. Thorough knowledge and appropriate methods are needed to ensure accurate measurement and calculation.

One important outcome of this work is the draft methodological guidance for monitoring of internal exposure to radiation, to be published by the Hungarian Atomic Energy Authority. The guide will assist the licensees in monitoring radioactive material incorporated by radiation workers in the course of their occupation and in the determination of the committed effective dose. The use of the

methodology will contribute to the harmonisation of internal exposure monitoring not just by providing a standardized approach but also clarifying the expectations of the authorities, therefore facilitating compliance with the radiation protection requirements laid down in current regulations. The guidance is intended primarily for the use by licensees but may also benefit internal radiation protection experts. The guidance also includes case studies to enhance its practical application. [op1] [op2] [op3]

SCIENTIFIC PUBLICATIONS RELATED TO THE THESIS STATEMENT

Journal publications related to the thesis statements

[P1] T. Pázmándi, A. Andrási, I. Fehér, A. Kocsonya, A. Pántya, P. Zagyvai, "Calibration of a whole body counter for ^{241}Am with the LLNL chest phantom", *Radiation Protection Dosimetry*, vol. 170, no. 1-4, pp. 225-230, Sept. 2016, https://doi.org/10.1093/rpd/ncv400.

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A tézispontokhoz kapcsolódó konferenciaközlemények

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További tudományos közlemények

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