REVIEW AND DEVELOPMENT OF AN URANIUM INTERNAL DOSIMETRY AND MONITORING PROGRAMME AT AN URANIUM PLANT

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DECLARATION

I declare that this research report is my own, unaided work. It is being submitted for the Degree of Masters of Science in the University of the Witwatersrand, Johannesburg. It has not been submitted before for any degree or examination in any other University. Information used in this report has been obtained while employed by Necsa

(Signature of candidate)

29 day of January 2013

ABSTRACT

Monitoring for internal exposures to uranium and calculating the corresponding Committed Effective Dose (CED) can be complex. Several contributing parameters such as the differences in the physiochemical nature of the uranium compound, the nature of the exposure scenario, variances in human metabolic behaviour and the capabilities of available bioassay techniques add uncertainty in developing an Internal Dosimetry and Monitoring Programme (IDMP). Necsa's IDMP was reviewed and found to be in line with best international practices and adequate for monitoring routine exposures to Type M uranium. As found in literature and shown in the present study, the monitoring for Type S uranium is problematic. The present study recommends continuance with the current Type S monitoring programme, however, the need for faecal analysis was identified. A combination of bioassay techniques can assist in determining the unknowns in the abovementioned contributing parameters. Analysis done to quantify the effect of differences in the contributing parameters has brought an understanding on how these parameters can influence and IDMP and knowledge gained from the present study will further enhance the programme and assist in developing the necessary documentation, providing the technical justification for Necsa's uranium IDMP.

DEDICATION

Dedicated to my wife

Suzette Beeslaar

2013

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LIST OF SYMBOLS

α	:	Alpha Particle
β	:	Beta Particle
λ	:	Gamma or Photon Wave
Bq	:	becquerel [Disintegration.s^{-1}]
Gy	:	gray
Sv	:	sievert [$J.kg^{-1}$]

NOMENCLATURE

Acronyms

ALI	:	**A**nnual **L**imit of **I**ntake
ADU	:	**A**mmonium **Di****u**ranate
AMAD	:	**A**ctivity **M**edian **A**erodynamic **D**iameter
ANSI	:	**A**merican **N**ational **S**tandard **I**nstitute
CAM	:	**C**ontinuous **A**ir **M**onitor
CED	:	**C**ommitted **E**ffective **D**ose
DAC	:	**D**erived **A**ir **C**oncentration
DCF	:	**D**ose **C**onversion **F**actor
DOE	:	**D**epartment **O**f **E**nergy of USA
DNC	:	**D**elayed **N**eutron **C**ounting
EU	:	**E**uropean **U**nion
EURADOS	:	**E**uropean **R**adiation **D**osimetry **G**roup
FAP	:	**F**ission and **A**ctivation **P**roducts
GI	:	**G**astro**i**ntestinal
GM	:	**G**eiger-**M**üller
HIRA	:	**H**azard **I**dentification and **R**isk **A**ssessment
HRTM	:	**H**uman **R**espiratory **T**ract **M**odel
IAEA	:	**I**nternational **A**tomic **E**nergy **A**gency
ICP-MS	:	**I**nductively **C**oupled **P**lasma-**M**ass **S**pectrometry
ICRP	:	**I**nternational **C**ommission on **R**adiation **P**rotection
ID	:	**I**nternal **D**osimetry
IDEAS	:	**I**nternal **D**ose **E**quivalent **A**ssessment **S**ystem
IDMP	:	**I**nternal **D**osimetry and **M**onitoring **P**rogramme
IMBA	:	**I**ntegrated **M**odules for **B**ioassay **A**nalysis
ISO	:	**I**nternational **S**tandards **O**rganisation
IVL	:	**I**nter**v**entional **L**evel
LET	:	**L**inear **E**nergy **T**ransfer
IL	:	**I**nvestigation **L**evel

LLD	:	**L**ower **L**imit of **D**etection
DL	:	**D**ecision **L**evel
MDA	:	**M**inimum **D**etectable **A**ctivity
ME	:	**M**ass **E**nrichment
MP	:	**M**onitoring **P**rogramme
MPe	:	**M**onitoring **P**eriod
NAA	:	**N**euron **A**ctivation **A**nalysis
Necsa	:	South African Nuclear Energy Corporation
NRC	:	**N**uclear **R**egulatory **C**ommission of the USA
NRPB	:	**N**ational **R**adiological **P**rotection **B**oard of the UK
OMINEX	:	**O**ptimisation of **M**onitoring for **In**ternal **Ex**posure
PAS	:	**P**ersonal **A**ir **S**ampler
PNNL	:	**P**acific **N**orthwest **N**ational **L**aboratory in the USA
PMT	:	**P**hoto**m**ultiplier **T**ube
RL	:	**R**ecording **L**evel
RPP	:	**R**adiological **P**rotection **P**rogram
SA	:	**S**pecific **A**ctivity
SAS	:	**S**tatic **A**ir **S**ampler
WBLC	:	**W**hole **B**ody and **L**ung **C**ounter

Technical Terms

Acute Intake	:	An intake occurring within a time period short enough that it can be treated as instantaneous for the purposes of assessing the resulting committed dose.
ALI	:	Annual Limit of Intake: The activity of a radionuclide which taken alone would irradiate a person, represented by reference man, to the annual dose limit of 20 mSv.
AMAD	:	Activity Median Aerodynamic Diameter – a measure of the particle size distribution of radioactive aerosols. Physical parameter for the description of the particle size of radioactive aerosols. Fifty percent of the activity in the aerosol is associated with particles of aerodynamic

		diameter greater than the AMAD. The AMAD is used for particle sizes for which deposition depends principally on inertial impaction and sedimentation: typically those greater than about 0.5 μm. For smaller particles, deposition typically depends primarily on diffusion, and the activity median thermodynamic diameter (AMTD) - defined in an analogous way to the AMAD, but with reference to the thermodynamic diameter of the particles.
Bioassay	:	Any procedure used to determine the nature, activity, location or retention of radionuclides in the body by direct (*in vivo*) measurement or by indirect (*in vitro*) analysis of material excreted or otherwise removed from the body.
Biokinetic model	:	A mathematical model describing the intake, uptake and retention of a radionuclide in various organs or tissues of the body and the subsequent excretion from the body by various pathways.
CED	:	Committed Effective Dose[$E(\tau)$]: The sum of the products of the committed equivalent doses in organs or tissues and the appropriate organ or tissue weighting factors (w_T), where τ is the integration time (50 year for workers) in years following the intake.
Chronic Intake	:	An intake over an extended period of time, such that it cannot be treated as a single instantaneous intake for the purposes of assessing the resulting committed dose.
Clearance	:	The net effect of the biological processes by which radionuclides are removed from the body or from a tissue organ or region of the body
Contamination	:	The activity of radionuclides present on surfaces, where the presence of such material is unintended and undesirable.
DAC	:	Derived Air Concentration – the ALI for a specific radionuclide divided by the air inhaled by a reference man

		in a working year (2400 m^3 for a 2000 h working year).
Effective Dose (E)		The tissue-weighted sum of the equivalent doses in all specified tissues and organs of the body, given by the expression: $E = \sum_{T} w_T \sum_{R} w_R D_{T,R}$ where w_R $D_{T,R}$ is the equivalent dose in a tissue or organ, T, and w_T is the tissue weighting factor. The unit for the effective dose is the same as for absorbed dose, J kg^{-1}, and its special name is sievert (Sv).
Excretion function	:	The fraction of an intake excreted per day after a given time has elapsed since the intake occurred
LLD	:	Limit at which it can be certain at a predetermined confidence level that a measurement will lead to detection.
HIRA	:	Hazard Identification and Risk Assessment is the systematic examination of activities and processes in the workplace, to determine the probability of harmful events, and the extent of the potential damage. This evaluation comprises a careful evaluation of the level of risk (i.e. the evaluation of the probability that the adverse event may occur, and the consequences of these events)
Intake	:	The activity of radioactive material entering the body, the principal routes being inhalation, ingestion or through intact or wounded skin (note in the case of inhalation of aerosols the intake is greater than the amount which is deposited in the body, because a part is exhaled).
Intake Retention Function	:	IRF is a function describing the amount of activity excreted per bioassay compartment (e.g. urine Bq/day) per unit intake (per Bq intake)
In-vitro	:	Indirect determination of intake of radionuclides. Measurement to determine the presence of or to estimate the amount of radioactive material in the excreta or in

		other biological material removed from the body.
In-vivo	:	Direct determination of intake of radionuclides. The measurement of radioactive material in the human body utilising instrumentation that detect radiation emitted from the radioactive material in the body.
Radiobioassay	:	The measurement of amount or concentration of radionuclide material in the body or in biological material excreted or removed from the body and analysed for the purpose of estimating the quantity of radioactive material in the body
Uptake	:	The processes by which radionuclides enter the body fluids from the respiratory tract, gastrointestinal tract or through the skin, or the fraction of an intake that enters the body fluids by these processes.

Uranium Compounds

U_3O_8	:	Triuranium Octoxide (Solid)
UF_6	:	Uranium Hexafluoride (Solid)
UF_4	:	Uranium Tetrafluoride (Solid)
UO_2F_2	:	Uranyl Fluoride (Solid)
UO_2	:	Uranium Dioxide (Solid)
UO_3	:	Uranium Trioxide (Solid)
UO_4	:	Uranium Peroxide (Solid)
$UO_3.H_2O.NH_3$	:	Ammonium di-uranyl (Solid) also known as ADU
$UO_2(NO_3)_2$	:	Uranyl Nitrate (Liquid)

CHAPTER 1

INTRODUCTION

A facility that handles uranium in significant quantities requires a programme to ***monitor*** internal contamination and to ***quantify*** the effect of the internal exposures.

Regulatory requirements compel such a facility to ensure exposures to workers and the public are justified, optimised and limited. A programme of monitoring the environment and workers is required to demonstrate that exposures are optimised and below regulatory limits. An Internal Dosimetry and Monitoring Programme (IDMP) forms part of the abovementioned requirements. The primary objective of an IDMP is the calculation of committed effective doses from intakes of radioactive material encountered in the workplace. Such an IDMP has to be technically justified and should comply with international best practices.

Raabe [RA94] defines Internal Dosimetry (ID) as *"...the scientific methodology used to measure, calculate, estimate, assay, predict, and otherwise quantify the radioactive energy absorbed by the ionization and excitation of atoms in human tissues as a result of the emission of energetic radiation by internally deposited radionuclides"*. The National Radiological Protection Board of the United Kingdom [NR04] defines a Monitoring Programme (MP) as *"…a combination of the monitoring strategy (task, routine, special), the location of the monitoring (individual, workplace monitoring) and monitoring technique"*.

A very basic structure of an IDMP is as follows:

Step 1 Worker internally exposed to radionuclides (e.g. uranium).

Step 2 Determine uptake amount of radionuclides in the body by means of an appropriate individual monitoring method (e.g. bioassay) and/or workplace monitoring programme (e.g. airborne contamination).

Step 3 Calculate Committed Effective Dose (CED) from the uptake using biokinetic models published by the International Commission on Radiation Protection (ICRP).

An IDMP forms an integral component of ensuring radiation safety within a facility. Workers at the South African Nuclear Energy Corporation (Necsa) who handle nuclear materials such as uranium are potentially at risk of inadvertent intakes of radioactive material. The most significant intakes of radionuclides usually occur when there is an accidental release of radioactive material in the workplace or when there is a loss of containment. Workplace and individual monitoring programmes are designed to provide the data needed (uptakes) to assess organ and tissue dose equivalents as well as determining the CED.

1.1 Statement of the problem

Uranium intake resulting in internal contamination represents an occupational risk to workers involved in the uranium industry. A monitoring programme capable of detecting internal exposures should be planned and implemented whenever an employee is potentially exposed to intakes of uranium. The objective of monitoring is to verify that workers are adequately protected against risks arising from intakes and to document this monitoring. Prompt and accurate assessment of internal exposures is important in determining each individual's CED and in establishing an accurate historical record.

Necsa's workforce is exposed to a wide spectrum of possible internal intakes from various uranium compounds, thus requiring a comprehensive IDMP. Recently, a state of the art Whole Body and Lung Counter (WBLC) was purchased. This has significantly improved the measuring methods that are now available and also resulted in an enhanced sensitivity for lung counting. Dose calculating software, known as Integrated Modules for Bioassay Analysis (IMBA), was also purchased which has improved the accuracy (best estimate) of reported results. Calculating doses is also less cumbersome with the new software. Current documentation at Necsa lacks a holistic overview of an IDMP. Necsa's policy and guiding principles on monitoring are clearly stated but the technical justification for dose calculations methodologies and monitoring programmes are not clearly documented.

Internationally, there have been significant developments regarding IDMP in recent years. However interlaboratory comparisons have revealed that different laboratories can obtain quite different estimates of intakes and doses when provided with the same monitoring data [DO07]. This was due to several reasons such as level of expertise of participants, errors in transcripts, assumptions made about time of intake and biokinetic models. Rahola [RA03] pointed out the lack of common strategies for monitoring programmes and found differences in routine monitoring for uranium. In terms of mitigation, a lot of effort has gone into the many studies needed to harmonise and optimise internal dosimetry practices, especially in the European Union (EU). The need for harmonisation of assessment procedures and monitoring protocols has been recognised in a research project carried out under the EU 5th Framework Programme. This has also prompted the International Standards Organisation (ISO) and European Radiation Dosimetry Group (EURADOS) to publish guidance documents such as [ID06] and [NR04], the Internal Dose Equivalent Assessment System (IDEAS) and Optimisation of Monitoring for Internal Exposure (OMINEX), respectively.

Taking into consideration recent international development as well as new equipment and software purchased and shortcomings in Necsa's documentation, a

need has thus been identified to evaluate and optimise Necsa's monitoring programme and to evaluate the dose calculating methods in order to ensure it is in line with and harmonised with international best practices.

1.1.1 Importance of the problem

The current scale of handling uranium and its compounds in fuel fabrication together with the possibility of future increases in volume indicates the importance of ensuring effective occupational monitoring of personnel. The South African government has indicated in the Integrated Resource Plan for Energy [IR11] that it plans to increase the nuclear component of its energy mix. Monitoring programmes and dose calculation methodologies which are not properly developed and in line with best international practices could lead to unreliable CED assessments and to the possibility of not detecting intakes of radionuclides. Personal monitoring provides an indication of the effectiveness of physical design features and administrative controls in controlling exposure to radioactive material and is an integral part of demonstrating compliance to regulatory requirements.

1.2 Study objectives

It is the primary objective of the present study to ensure that Necsa's IDMP for uranium at a recovery and purification plant complies with national and international standards, legislation and best practices. The IDMP should be adequate to demonstrate compliance with the performance criteria for monitoring and dose calculation.

Information and knowledge should be obtained in order to:

- assist in the design of monitoring programmes for individuals,
- provide guidance on the interpretation of monitoring data and doses,
- provide guidance on the estimation of associated doses and
- assist in the development of technical basis documentation.

The results of the present study should contribute to the health and radiological protection of workers by improving the methods for assessing internal dose from measurements and by implementing a common, harmonised approach to the design and implementation of internal dose monitoring programmes. The monitoring programme should be the best measurement, reflecting actual exposures. This will result in more reliable estimate of doses.

1.3 Limitation and scope of study

The scope of the present study is to review international practices and methodologies in IDMP and to compare with Necsa's programme. A knowledge base will be built up containing best practices and these best practices will be presented in the study. The present study will not include detailed descriptions of the biokinetic models, bioassay protocols and dose calculation methodologies. It will rather focus on the development of guidelines to optimise internal dosimetry calculations and monitoring programmes and to ensure Necsa's IDMP is in line with best international practices. Also included will be recommendations on changes needed in Necsa's IDMP.

The project entails studying and understanding exposure scenarios within a uranium plant, and specifically the physiochemical nature of the uranium compounds found in a uranium plant. A uranium monitoring programme, i.e. monitoring techniques and its capabilities, as recommend in literature will be described. Necsa's programme will be explained, its shortcomings highlighted and compared against cases found in the literature. This includes a focus on important parameters used during dose calculations and the effect it has on the calculated CED. Specifically, the study will include calculations and validation of limits of detection and performance criteria of monitoring techniques.

The study will be limited to only uranium intake within the context of a uranium recovery and purification plant, explicitly the chemical processing and recovery, thus excluding e.g. fuel plate manufacturing. Whenever the term uranium plant is

used, it refers to the chemical processing, recovery and purification of uranium and the manufacturing of uranium alloy metal.

The chemical hazards of uranium intake will not be considered and the present study will only identify the internal radiological hazard of uranium. External exposures from uranium will thus not be studied. Elevated radon concentrations can occur in poorly ventilated uranium storage areas and the internal radiological hazards of radon and its short lived products are excluded in this study. The study excludes criteria and conditions under which workers are to be included in a bioassay programme. This study will be broadly based on a literature study and does not include actual experiments to determine the physiochemical nature of exposures.

It is limited to routine monitoring of individuals exposed to Type M (medium solubility class) uranium. Monitoring for exposures to Type S (slow solubility class) uranium, task related and special monitoring (including additional monitoring after an action level has been exceeded) is excluded. The exclusions will be discussed, but not addresses specifically. The focus will be on individual monitoring, and workplace monitoring programmes will be excluded. Thus, it will not investigate the characteristics of workplace monitoring programmes (e.g. placement of workplace monitors etc.) in order to determine intakes.

1.4 Report structure and study design

The objective is to study available literature and provide a summary of the literature. Literature from published journals will be reviewed, including reports from international organisations (IAEA, ICRP etc), and reports from open literature such as presentations at conferences and reports from institutions available on the internet.

The three main chapters are:

Chapter 2 – Theoretical considerations this chapter provides the scientific basis for the study. It presents an overview and an understanding of an IDMP and its

requirements. In particular it addresses the radiological decay characteristics of uranium and the biokinetic behaviour of uranium. Knowledge gained in this chapter will be applied specifically to an uranium IDMP.

Chapter 3 – Development of a uranium monitoring programme: results, discussion and recommendations in order to apply the requirements of an IDMP, one needs to investigate and analyse the specific characteristics of an uranium IDMP. This entails first a detailed literature study on uranium's physiochemical and radiological (particularly enrichment) properties as it is applicable specifically in the environment of an uranium plant and an estimate of the uncertainties associated with these properties. The effect of these uncertainties is quantified. Further, literature research is also done on exposure scenarios and the associated uncertainties. Again specifically in the context of an uranium plant. The effect of altering physiochemical and biokinetic parameters are thus quantified. This will provide an understanding of the influence of these parameters and how it should be used in calculating doses. Recommendations are made on addressing these unknowns.

The second part of Chapter 3 investigates bioassay techniques and the various advantages and disadvantages of each technique. An uranium monitoring programme, i.e. monitoring techniques and its capabilities, as recommended in literature will be described. Necsa's programme will be explained, its shortcoming highlighted and compared against literature. Recommendations for improvements will be made. Recommendations from Chapter 3 will improve Necsa' capability to monitor for uranium and will also ensures calculated doses are the best estimate.

CHAPTER 2

THEORY

The purpose of this chapter is to provide a basic scientific overview of the theory and principles of an internal dosimetry and monitoring programme. The physics and biology of radiation protection and the principles of dosimetry are discussed first (Section 2.1). This includes the radiological characteristics of uranium and the ICRP biokinetics models. Workplace characterisation (Section 2.2), which is used as a basis for a monitoring programme, is reviewed thereafter. This entails the physiochemical nature of the intake compound and the characteristics of an exposure scenario. The monitoring programme (Section2.3) and its characteristics, which ensures exposures are adequately monitored, is reviewed. The principles of internal dose calculation (Section 2.4) in order to ensure doses are the best estimate, is discussed. This information will be applied in developing an IDPM specifically tailored for the situation at Necsa's Uranium Plant.

2.1 The physics and biology of radiation protection and dosimetry

In order to practice radiation protection, one needs an understanding of both the physics and the biology of radiation protection. The interaction between these two disciplines forms the basis for radiation protection principles.

2.1.1 Radiological characteristics

The radiological features of a material can be described by various physical characteristics. The activity, half-life and the type of decay characteristics are described below.

2.1.1.1 Activity

Radioactivity is the process through which nuclei spontaneously emit subatomic particles. The unit of activity is called the becquerel (Bq) and is defined as the

quantity of radioactive material in which one atom is transformed per second. Alternatively, it is defined as the number of disintegrations per atom per second. It should be noted that it is not the number of particles emitted per second, e.g. ^{60}Co emits 3 particles (one β^--particle and two γ-rays per transformation).

2.1.1.2 Half-life

Half-life is defined as the time required for the activity of the sample to reduce by one half and is given by the following equation:

$$T_{1/2} = \frac{ln(2)}{\lambda_d} \quad , \tag{2.1}$$

Material is also cleared from the human body at a specific rate, which is characterised by the biological half-life of the specific nuclide. The biological half-life is analogous to a physical (radioactive) half-life and can also be described by Eq. (2.1). The effective half-life (T_e) is a combination of the physical (T_p) and biological (T_b) half-life and is given by the following equation:

$$T_e = \frac{T_b \times T_p}{T_b + T_p} \quad . \tag{2.2}$$

For radionuclides that decay into stable progeny, or into progeny with radioactive half-lives very much longer than the human lifespan, only the radiation emitted by the parent radionuclide need to be considered for dosimetry. However, many radionuclides decay into short-lived progeny, or into progeny with an intermediate half-life. For these radionuclides, the amount of in-growth of activity of the progeny must be taken into account (over the period for which the dose is to be integrated). It should be noted that for calculation purposes the Integrated Modules for Bioassay Analysis (IMBA) software has various mathematical solutions to take this into consideration [BI94].

Half-lives need to be considered when deciding on monitoring periods. This is the case for radionuclides with a physical and or effective half-life within the same order as the monitoring period. This is discussed further in Section 2.3.3.

2.1.1.3 Types of radioactive decay

When nuclei emit subatomic particles, their configuration, state, and even identity may change. Typical decay mechanisms are shown below:

alpha decay: $${}^{A}_{Z}\mathrm{X} \rightarrow {}^{A-4}_{Z-2}\mathrm{Y} + {}^{4}_{2}\alpha$$

β^-decay: $${}^{A}_{Z}\mathrm{X} \rightarrow {}^{A}_{Z+1}\mathrm{Y} + \beta^- + \overline{\nu}_e$$

electron capture: $${}^{A}_{Z}\mathrm{X} + e^- \rightarrow {}^{A}_{Z-1}\mathrm{Y} + \nu_e$$

β^+-decay: $${}^{A}_{Z}\mathrm{X} \rightarrow {}^{A}_{Z-1}\mathrm{Y} + \beta^+ + \nu_e$$

gamma decay: $${}^{A}_{Z}\mathrm{Y} \rightarrow {}^{A}_{Z}\mathrm{Y} + \gamma \quad ,$$

where ${}^{A}_{Z}\mathrm{X}$ represents the initial nuclear species X with Z the atomic number (number of protons) and A the mass number (total protons and neutrons). Here, Y represents the final nuclear species after decay. There are two kinds of beta decay *viz.* β^--decay (electron) and β^+-decay (positron). A positron has all the properties of an electron, but has a positive charge. Electron capture occurs when a nucleus captures one of the orbital electrons and is competing with β^+-decay. During electron capture a proton is transformed into a neutron:

$$p + e^- \rightarrow n + \nu_e \tag{2.3}$$

which is also the case for β^+-decay:

$$p \rightarrow n + \beta^+ + \nu_e \quad . \tag{2.4}$$

However, for B^--decay a neutron is changed into a proton:

$$n \rightarrow p + \beta^- + \overline{\nu_e} \quad . \tag{2.5}$$

In the above equations ν_e represent an electron neutrino and $\overline{\nu_e}$ represents an anti-electron neutrino. It should be noted that following β^--decay or electron capture the daughter nucleus may be left in an excited state and which subsequently de-excites by emitting a γ-ray(s). In addition, electron capture leaves the daughter nucleus with a hole in the K or L electron shell orbits and a characteristics X-ray is produced when the hole is filled by a higher lying electron.

2.1.2 Radiological characteristics of uranium

Uranium is a radioactive element that occurs naturally in varying but small amounts in soil, rocks, water and plants. It has an atomic number of $Z = 92$ (number of protons). In nature, uranium exists as several isotopes *viz.* ^{238}U, ^{235}U and ^{234}U being the most prevalent. For naturally occurring uranium the abundance, half-life and specific activity (SA) is given in Table 2.1.

Table 2.1: Radiological parameters for naturally occurring uranium [SH92].

Uranium isotope	Abundance (%)	Half-life (y)	SA (Bq/g)
^{234}U	0.0055%	2.45×10^5	2.31×10^8
^{235}U	0.72%	7.04×10^8	8.00×10^4
^{238}U	99.27%	4.47×10^9	1.24×10^4

All three naturally occurring isotopes are radioactive, emitting alpha particles and γ-rays. The ^{235}U isotope is capable of fission leading to the release of considerable energy. It should be noted that ^{238}U and ^{234}U are part of the uranium decay series and ^{235}U is part of the actinium decay series. Here, ^{238}U is the parent nucleus of the uranium decay series ending in the stable nuclide ^{206}Pb. In addition, ^{234}U is a daughter of ^{238}U and is found in nature in radioactive equilibrium (secular equilibrium) with its mother nucleus. The daughter product radon (^{222}Rn) is an inert gas and can easily migrate away from the source and is rarely found in equilibrium. Also of note is that ^{234}U does not emit any significant number of γ-rays. Indeed, all the γ-rays emitted are with very low energies and with very low abundance. The uranium series decay primarily by emission of alpha and β^- particles (see Table 2.2).

The actinium decay series (see Table 2.3) starts with the parent nucleus ^{235}U and ends with stable ^{207}Pb. It also decays primarily by alpha and β^- particles. Of note is the 186 keV photon emitted by ^{235}U, which is used in the detection of uranium. The ^{238}U isotope does not emit any γ-rays, but the immediate daughter, ^{234}Th, does emit a 93 keV and a 63 keV γ-ray, but with very low abundance (< 5%).

Table 2.2: Uranium decay series (taken from [CE96]).

Nuclide	Half-life	Energy, MeV		
		Alpha[a]	Beta	Gamma, photons/ trans.[b]
$^{238}_{92}$U	4.51×10^9 years	4.18		
$^{234}_{90}$Th (UX_1)	24.10 days		0.193, 0.103	0.092 (0.04) 0.063 (0.03)
$^{234m}_{91}$Pa (UX_2)	1.175 min		2.31	1.0 (0.015) 0.76 (0.0063), I.T.
$^{234}_{91}$Pa (UZ)	6.66 h		0.5	Many weak
$^{234}_{92}$U (UII)	2.48×10^5 years	4.763		
$^{230}_{90}$Th (I_0)	8.0×10^4 years	4.685		0.068 (0.0059)
$^{226}_{88}$Ra	1,622 years	4.777		
$^{222}_{86}$Em (Rn)	3.825 days	5.486		0.51 (very weak)
$^{218}_{84}$Po (RaA)	3.05 min	5.998 (99.978%)[c]	Energy not known (0.022%)[c]	0.186 (0.030)
$^{218}_{85}$At (RaA′)	2 s	6.63 (99.9%)[c]	Energy not known (0.1%)[c]	
$^{218}_{86}$Em (RaA″)	0.019 s	7.127		
$^{214}_{82}$Pb (RaB)	26.8 min		0.65	0.352 (0.036) 0.295 (0.020) 0.242 (0.07)
$^{214}_{83}$Bi (RaC)	19.7 min	5.505 (0.04%)[c]	1.65, 3.7 (99.96%)[c]	0.609 (0.295) 1.12 (0.131)
$^{214}_{84}$Po (RaC′)	1.64×10^{-4} s	7.680		
$^{210}_{81}$Tl (RaC″)	1.32 min		1.96	2.36 (1) 0.783 (1) 0.297 (1)
$^{210}_{82}$Pb (RaD)	19.4 years		0.017	0.0467 (0.045)
$^{210}_{83}$Bi (RaE)	5.00 days		1.17	
$^{210}_{84}$Po (RaF)	138.40 days	5.298		0.802 (0.000012)
$^{206}_{82}$Pb (RaG)	Stable			

[a]Only the highest-energy alpha is given. Complete information on alpha energies may be obtained from Sullivan's *Trilinear Chart of Nuclides*, Government Printing Office, Washington, D.C., 1957.

[b]Only the most prominent gamma photons are listed. For the complete gamma-ray information, consult T. P. KOHMAN: Natural radioactivity, in H. Blatz (ed.): *Radiation Hygiene Handbook*. McGraw-Hill, New York, 1959, pp. 6–13. With permission.

[c]Indicates branching. The percentage enclosed in the parentheses gives the proportional decay by the indicated mode.

Table 2.3: Actinium decay series (taken from [CE96]).

Nuclide	Half-life	Energy, MeV		
		Alpha[a]	Beta	Gamma, photons/ trans.[b]
$^{235}_{92}$U	7.13×10^8 years	4.39		0.18 (0.7)
$^{231}_{90}$Th (UY)	25.64 h		0.094, 0.302, 0.216	0.022 (0.7) 0.0085 (0.4) 0.061 (0.16)
$^{231}_{91}$Pa	3.43×10^4 years	5.049		0.33 (0.05) 0.027 (0.05) 0.012 (0.01)
$^{227}_{89}$Ac	21.8 years	4.94 (1.2%)[a]	0.0455 (98.8%)[c]	
$^{227}_{90}$Th (RdAc)	18.4 days	6.03		0.24 (0.2) 0.05 (0.15)
$^{223}_{87}$Fr (AcK)	21 min		1.15	0.05 (0.40) 0.08 (0.24)
$^{223}_{88}$Ra (AcX)	11.68 days	5.750		0.270 (0.10) 0.155 (0.055)
$^{219}_{86}$Em (An)	3.92 s	6.824		0.267 (0.086) 0.392 (0.048)
$^{215}_{84}$Po (AcA)	1.83×10^{-3} s	7.635		
$^{211}_{82}$Pb (AcB)	36.1 min		1.14, 0.5	Complex spectrum, 0.065 to 0.829 MeV
$^{211}_{83}$Bi (AcC)	2.16 min	6.619 (99.68%)[c]	Energy not known (0.32%)[c]	0.35 (0.14)
$^{211}_{84}$Po (AcC′)	0.52 s	7.434		0.88 (0.005) 0.56 (0.005)
$^{207}_{81}$TI (AcC″)	4.78 min		1.47	0.87 (0.005)
$^{207}_{82}$Pb	Stable			

[a]Only the highest-energy alpha is given. Complete information on alpha energies may be obtained from Sullivan's *Trilinear Chart of Nuclides*, Government Printing Office, Washington, D.C., 1957.

[b]Only the most prominent gamma photons are listed. For the complete gamma-ray information, consult T. P. KOHMAN: Natural radioactivity, in H. Blatz (ed.): *Radiation Hygiene Handbook*. McGraw-Hill, New York, 1959, pp. 6–13. With permission.

[c]Indicates branching. The percentage enclosed in the parentheses gives the proportional decay by the indicated mode.

2.1.3 Interaction with matter

Radiation carries energy and whenever it interacts with a medium it may deposit some or all of it to the atoms in the medium. The result is some form of excitation or ionisation. Excitation occurs when an electron is raised to a higher-lying orbit (higher energy state) within the absorbing atom. Ionisation happens when the energy transferred exceeds the binding energy and the orbital electron is completely removed from the absorbing atom. This excitation or ionisation can form a basis of signal formation for a detector and can be measured by the processing electronics.

2.1.3.1 Interaction of particles

Heavy particles, such as alpha particles, interact with matter through Coulomb forces between the positive charge of the alpha particles and the negative charge of orbital electrons within the absorbing atom. The alpha particle interacts with several of the electrons and, depending on the proximity of the alpha particle, the electrons may either be ionised or excited. These interactions decrease the energy (velocity) of the alpha particle, resulting in a specific range (depth of penetration) depending on the absorber material.

Energy is converted into electromagnetic radiation (photons) during the interaction of energetic (fast) electrons with matter. The fraction of electron's energy converted increases with electron energy and is also dependent on the atomic number of the absorbing material. The spectrum of photons produced is continuous and is called Bremsstrahlung radiation. Backscatter occurs when low-energy incident electrons undergoes large-angle deflection along their tracks. Not all of the backscattered electron's energy is deposited in the absorbing matter.

A positron is emitted during β^+-decay and its range is relatively short and at the end of the range, when the energy is low it combines with an electron. The positron and the electron disappear and are replaced by two oppositely directed photons (due to conservation of momentum), each with an energy of 0.511 MeV

(the mass-energy equivalent of the electron). This process is known as annihilation.

2.1.3.2 Interaction of photons

Photons can primarily interact with material in three different ways: photoelectric absorption, compton scattering and pair production. All of these processes lead to the partial or complete transfer of photon energy to electron energy [AH07].

During the photoelectric effect, all of the energy of the photon is absorbed and transferred to an orbital electron. The atom is ionised (electron vacancy) when the electron is removed from the atom. The kinetic energy of the removed electron is equal to the original energy of the photon minus the binding energy. The vacancy is quickly filled by either a free electron or electron from a higher orbital shell resulting in the release of a characteristic X-ray. Fig. 2.1 depicts the photoelectric absorption interaction.

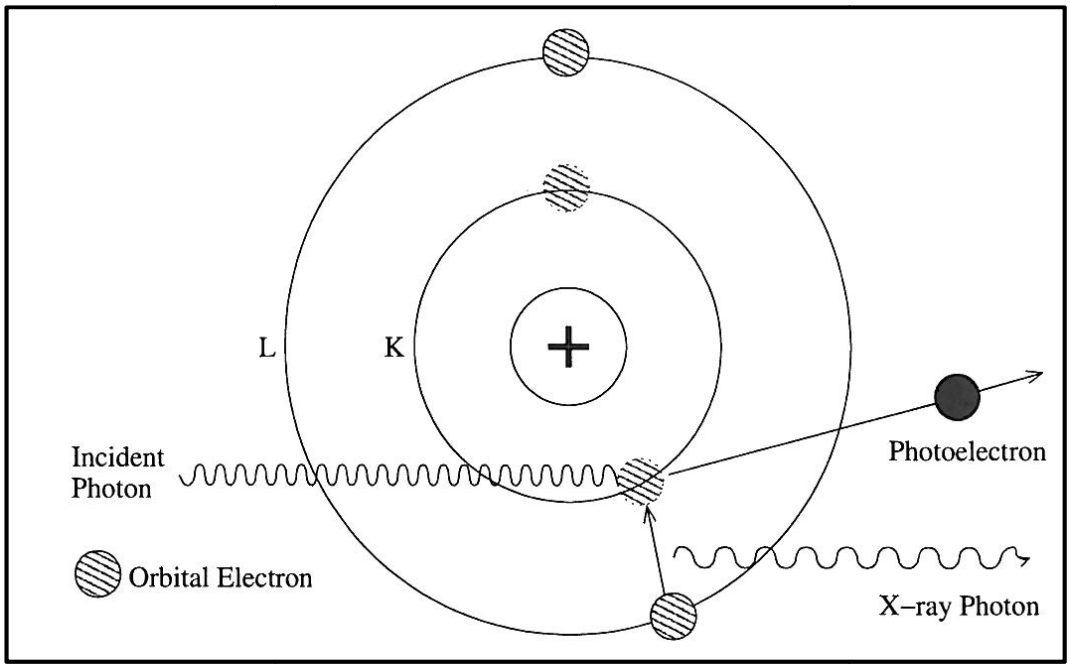

Figure 2.1: Photoelectric absorption (taken from [AH07]).

Compton scattering occurs when the incident photon interacts with an outer orbital electron, resulting in the ejection of the electron and scattering of the lower

energy photon. The kinetic energy of the ejected electron (recoil electron) is dependent on the energy of the incident photon and the angle of the photon scattering. The energy of the electron increases as the angle of the deflection increases and the scattered photon may continue with more compton interactions or photoelectric absorption. The ejected electron deposits its energy through excitation or ionisation. Fig. 2.2 depicts compton scattering.

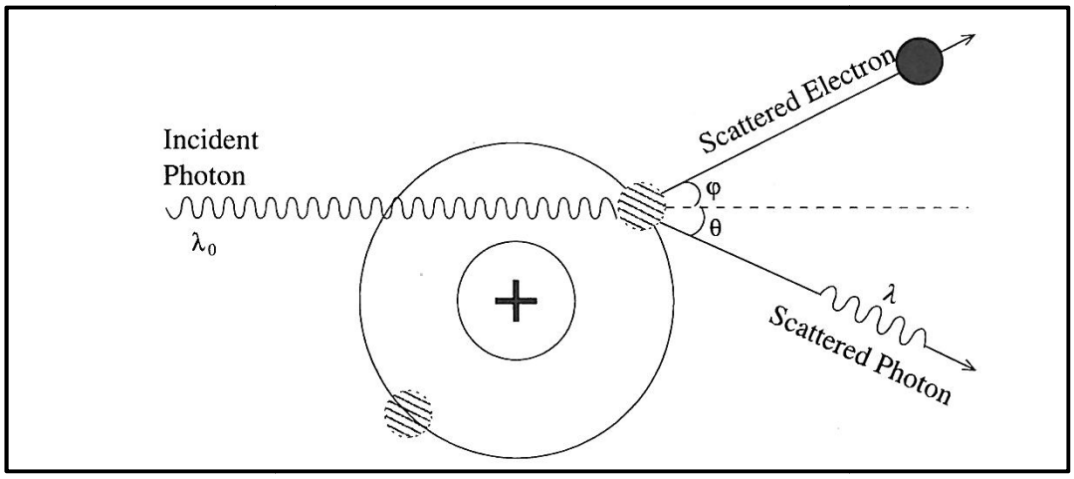

Figure 2.2: Compton scattering (taken from [AH07]).

Pair production may occur with high energy photons. The energy of the photon should be more than twice the rest-mass energy (>1.02 MeV) of an electron. An electron-positron pair is created instantaneously when the energy of the photon is converted under the influences of the Coulomb field of a nucleus. The excess energy (above 1.02 MeV) is shared as kinetic energy between the electron and positron. When the positron slows down it will interact with an electron, resulting in annihilation and the emission of two 0.51 MeV photons.

The relative importance of the above three interactions is graphically depicted in Fig. 2.3. It is depicted as a function of the *Z* number of the absorbing medium and the energy of the incident photon. The probability of a specific interaction depends on the energy of the incident photon and medium in which the absorption occurs. The line at the left represent the energy and *Z* number combination where the photoelectric absorption and the compton scattering are equally probable. The

line on the right represents where pair production and Compton scattering are equally probably. Thus, three distinct areas are defined where each of the three interactions are prominent.

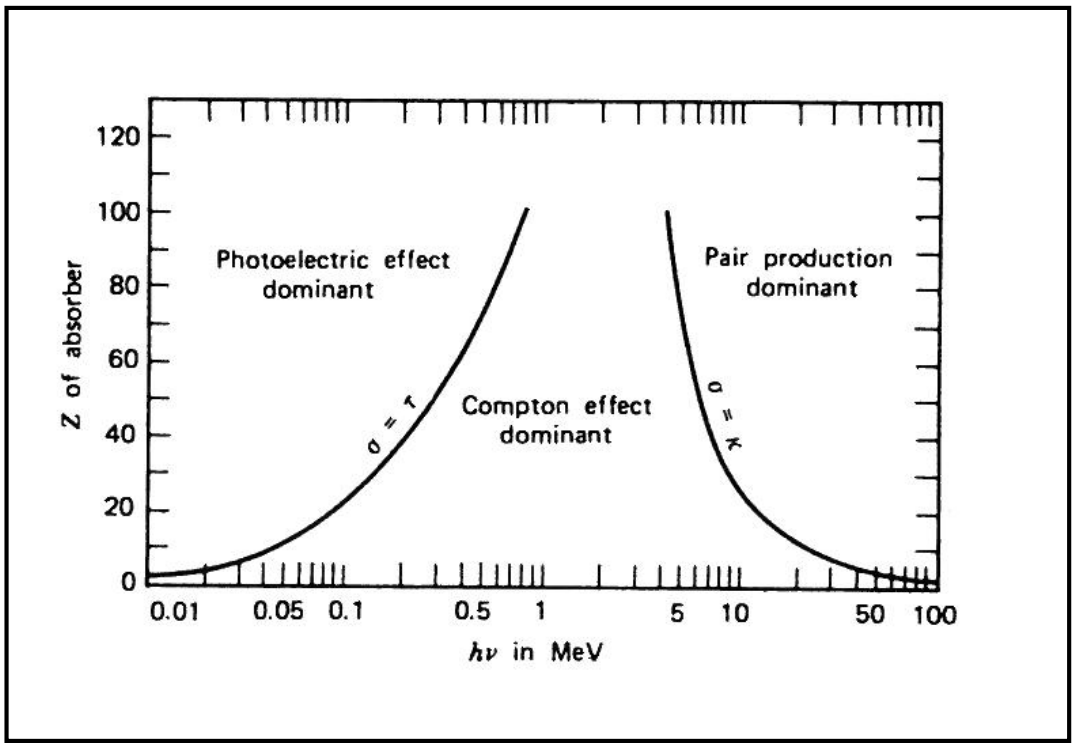

Figure 2.3: Probability of the three main interactions of photons with matter (taken from [KN99]).

2.1.4 Dosimetry quantities

Radiation measurements and the investigation of radiation effects require various specifications of the radiation field at the point of interest. Radiation dosimetry is the quantitative determination of energy deposited in a given medium by directly or indirectly ionizing radiations. Various quantities and units have been defined for describing the interaction of radiation with tissue, and the most commonly used dosimetric quantities and their units are defined below.

2.1.4.1 Fundamental quantities

Kerma (*K*) is defined as the sum of the initial kinetic energies of all the charged particles liberated by uncharged ionizing radiation (i.e., indirectly ionizing radiation such as photons and neutrons) when travelling through a tissue of mass

dm [ST08]. Not all of the kinetic energy is absorbed through the processes described in Section 2.1.3 only some of the energy is lost. As such, kerma is different from absorbed dose, according to the energies involved. At low energies it is almost equal, however, kerma is much larger than absorbed dose at higher energies since some of the energy escapes from the absorbing volume in the form of bremsstrahlung or fast moving electrons. The main parameter of interest is absorbed dose, which is measured as energy deposited per unit mass. Kerma (K) and absorbed dose (D) are given in Eq. (2.6) and (2.7).

$$K = \frac{dE_{\mathrm{Tr}}}{dm} \tag{2.6}$$

$$D = \frac{d\bar{\varepsilon}}{dm} \tag{2.7}$$

where $d\bar{\varepsilon}$ is the mean energy absorbed by matter of mass dm and dE_{Tr} is the sum of the initial kinetic energy per unit mass dm. The unit of absorbed dose is the gray (Gy), where 1 Gy = 1 J/kg.

2.1.4.2 Linear Energy Transfer

The biological damage created by ionising radiation relates to chemical alteration of the biological molecule caused by ionisation and excitation. This is dependent on the local rate of energy loss along the particle's track. There are differences in the energy deposited per unit path length for the various types of ionising radiation. For example, an alpha particle has a relatively short range in tissue and will thus have a high ionising intensity (energy deposited per unit length). This characteristic is described by the Linear Energy Transfer (LET) of the various types of ionising radiation, as seen in the Table 2.4 [RA94].

The LET is defined as the average instantaneous rate of energy loss to ionisations and excitations per unit path length. As can be seen the LET's of nuclear particles are significantly higher than those of electromagnetic radiation and electrons/positrons and tends to result in greater biological damage. This is of specific interest in the present study where the main nuclide of interest is uranium

which decays predominantly by an alpha emission. Alpha particles are easily absorbed by the dead layer of skin (external exposure) and are subsequently mainly a hazard with internal exposures where it deposits its energy over a very short range. The internal doses received from uranium's daughter build-up in the body is small (< 10%) compared with the doses received from the alpha decay of uranium. This is because the short-lived daughters are emitting mostly beta and gamma (photon) radiation which has significantly lower LET values.

Table 2.4: LET for different types of ionising radiation (taken from [RA94]).

Type of Radiation	**LET (keV/µm)**
X-rays, γ-rays, β particles, electrons	0.2 – 2
protons	15 – 25
neutrons	20 – 80
alpha particles	60 - 200

2.1.4.3 Effective dose

To express the relative effectiveness of different radiation, the absorbed dose (D) is multiplied by a radiation weighting factor, w_R. The weighing factor is summed if there are several types of radiation. To take into consideration the difference in sensitivity of the various tissues and organs in the human body, the absorbed dose is also multiplied by a tissue weighing factor w_T, which is also summed to take into account all the different tissues and organs. The resulting effective dose (E) has the unit sievert (Sv) and is defined as:

$$E = \sum_T w_T \sum_R w_R D_{T,R} \quad . \tag{2.8}$$

The radiation protection dose quantity, effective dose (E), is related to the human body and is not measurable. For external dosimetry the operational dose quantity

is the personal dose equivalent (H_{p10}). For internal dosimetry the operational dose quantity is the Committed Effective Dose (CED), E_{50}. The internal exposure is assessed over a period of 50 years.

2.1.5 Detection equipment

A detector responds to various types of radiation due to the interactions as described in Sections 2.1.3. Energy from the interactions is converted into an electrical charge within the detector's active volume. The electrical charge is collected to form the basis for an electrical signal. Typical detectors include:

- ionisation chambers,
- proportional counters,
- Geiger-Müller counters and
- scintillation counters.

The characteristics of each of them are dealt with in turn below.

2.1.5.1 Ionisation chambers

Ionization chambers, like proportional counters and Geiger-Müller counters, are gas filled detectors. An ionisation detector can operate with different gases and gas pressures but often air is used at atmospheric pressure and temperature. The gas absorbs the radiation's energy which ionises and excites the gas atoms along the interacting particle's track. The number of ion pairs created (and collected) is proportional to the energy deposited by the ionising radiation in the gas volume during interactions. Alpha particles produces more ion pairs than β^- particles and thus the chamber makes it possible to distinguish between the types of radiation with the pulse size being independent on the applied voltage. This enables ionisation detectors to distinguish between different types of radiation *viz*. alphas, betas and gammas. The ions move under the influence of the potential difference applied across the sensitive volume and are collected by the electrodes to form an electrical current which forms the basis of the measured electrical signal. A typical ionisation chamber detector system is schematically shown in Fig. 2.4 with the essential components indicated as:

A gas filled container

E physically separated electrodes (**E1** - positive anode; **E2 -** negative cathode) with a fixed potential difference (voltage) between electrodes

P power supply (battery)

D display.

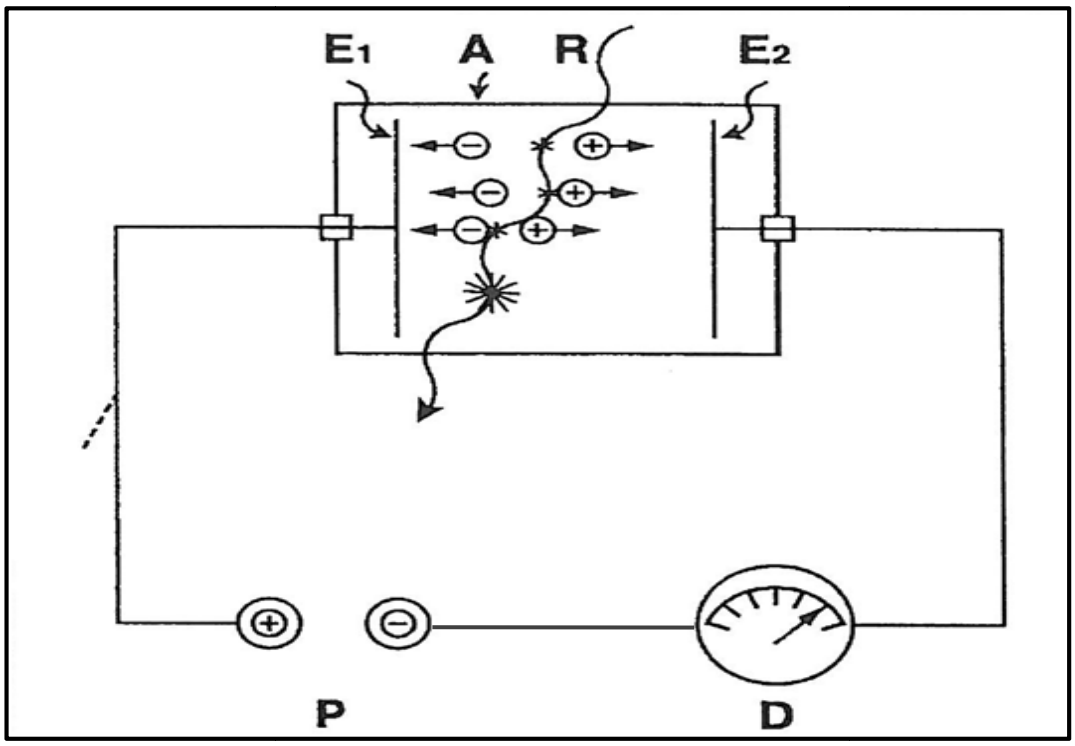

Figure 2.4: Typical ionisation chamber (taken from [IA04a]).

2.1.5.2 Gas multiplication and proportional counters

Proportional counters typically contain a mixture of inert and organic gases. Counters filled with boron trifluoride (BF_3) enable the detection of neutrons. The proportional counter's electrical signal is based on the generation of pulses and the detectors may operate at elevated gas pressures. The elevated gas pressure provides higher detection efficiencies for X-rays, γ-rays and β-particles. Sensitivity to low dose rates is increased due to the higher applied voltage. The higher voltage accelerates the ions produced and the fast moving negative ions in turn cause further ionization (see Fig. 2.5). This process is known as gas multiplication. Gas multiplication also occurs in Geiger-Müller counters.

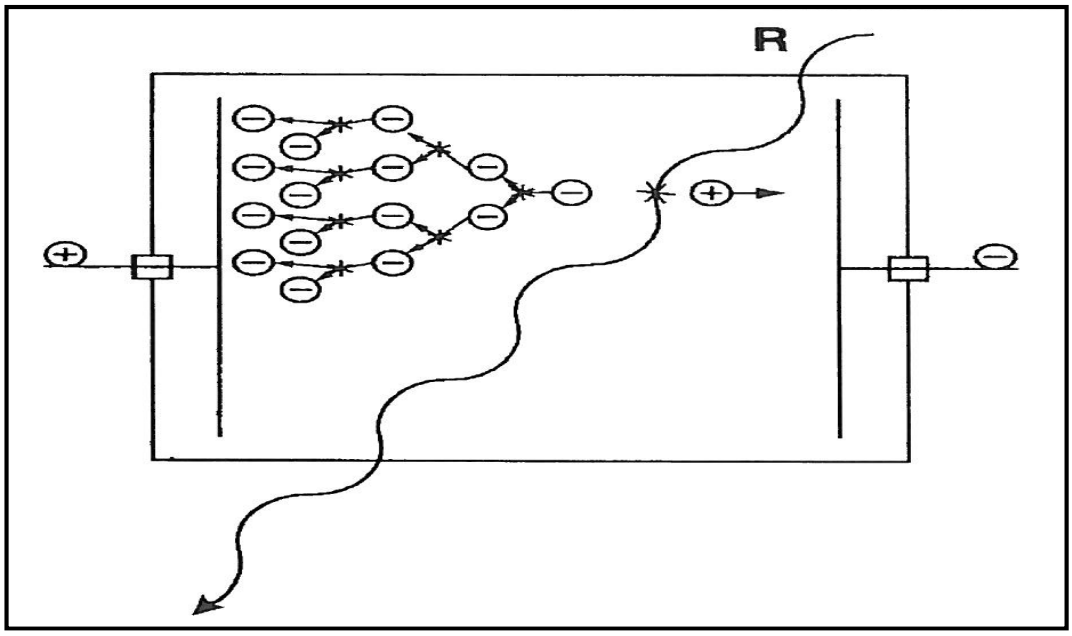

Figure 2.5: The effect of gas amplification in proportional counters (taken from [IA04a]).

The number of ions collected by the electrode will be proportional to the number of ions produced by the radiation (similar to ionisation detectors). A pulse of charge is generated when the ions are collected on the electrode and the subsequent pulse is proportional to the energy absorbed by the detector which thus still allows it the ability to distinguish between different types of radiation. In the case of proportional counters, the pulse height is larger (improved sensitivity), in comparison with ionisation chambers, due to the increased voltage. The pulse height is also proportional to the voltage across the electrodes.

2.1.5.3 Detector regions and Geiger-Müller counters

Geiger-Müller (GM) counters contain a low-pressure inert gas and traces of an organic or halogen gas called the quenching agent. The potential difference between a GM's electrodes is higher than proportional counters and large enough to cause a complete ionization of the detector gas. This complete ionisation from the gas amplification is caused by a single primary ionization. The size of the pulse is thus independent of the type of radiation. A GM's output differs from that of other gas filled detectors in that the pulses of electrical charge occurs at a count rate which is related to the radiation fluence (intensity).

The relative response of gas filled detectors to a single ionising particle is illustrated in Fig. 2.6. The number of ions collected, *n*, per ionising particle is plotted against the detector voltage, *V*. The difference between radiations types is also shown with the upper curve representing an alpha particle (more ion pairs created) and the lower curve a β^- particle (less ion pairs created). The different detector regions indicated are for

A ionization chambers: the range of voltage is above a minimum to prevent the recombination of ion pairs but large enough to collect the ions at the electrodes. The voltage is also low enough to prevent gas multiplication.

B proportional counters: the region starts where secondary ionisation is being produced. The detector's response (sensitivity) increases with increasing ionisation due to the secondary ionisation.

C Geiger-Müller counters: an avalanche of ions is produced and collected. The pulse height (number of ions collected) is independent of the primary ionisation.

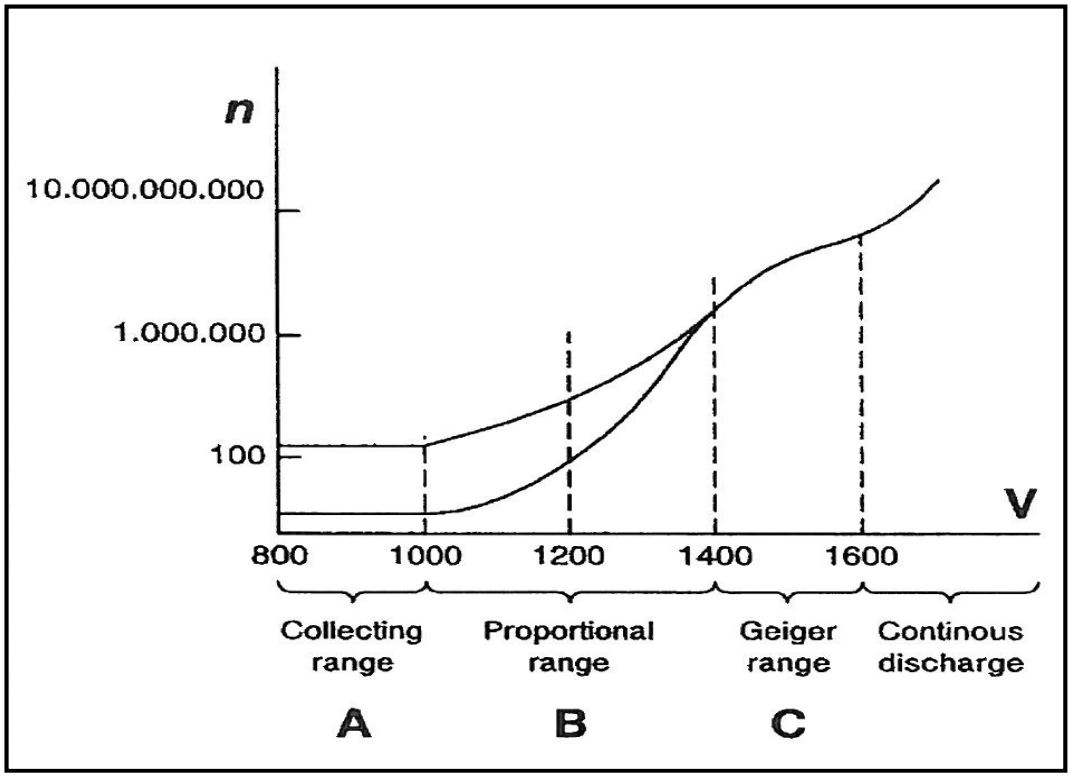

Figure 2.6: Typical output from gas filled detectors (taken from [IA04a]).

2.1.5.4 Scintillator counters

The kinetic energy of an ionising particle is changed into a light photon during the fluorescence process. Fluorescence is the prompt emission of visible light from a substance following its excitation. This process occurs in a scintillation crystal. Such scintillation phenomena form the basis of a sensitive radiation detection system. The scintillator crystal consists of a transparent crystal, usually a phosphor, plastic, or organic liquid that fluoresces when struck by ionizing radiation. A sensitive photomultiplier tube (PMT) measures the ultra violet light from the crystal. The PMT is attached to an electronic amplifier and other electronic equipment to count and quantify the amplitude of the signals produced by the PMT. When a charged particle strikes the phosphor, a light photon is emitted. This photon strikes the photocathode in the scintillator, releasing an electron. This electron accelerates towards the first dynode, causing multiple secondary electrons to be emitted, which accelerate towards the second dynode. More electrons are emitted and the chain continues, multiplying the effect of the first charged particle. This process is called photo-multiplication. At the last dynode, a voltage pulse appears across the external resistors. This voltage pulse is amplified and recorded. The intensity of the light produced is directly proportional to the energy of the ionising particle and subsequently it can be used as a spectrometer with the appropriate electronics. This process and the different components of a scintillator detector are depicted in Fig. 2.7.

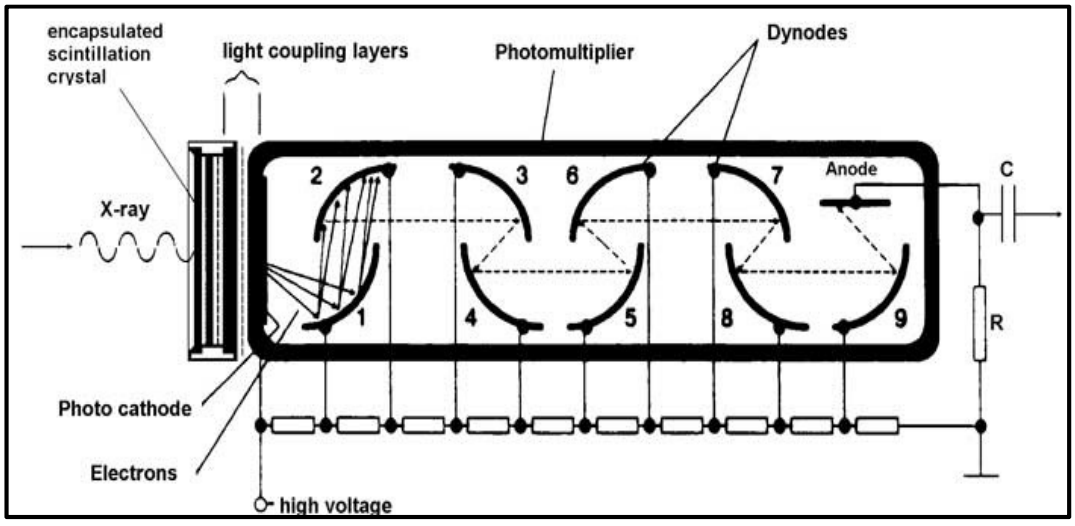

Figure 2.7: Scintillation counter (taken from [BR11]).

2.1.6 Sensitivity

The sensitivity of bioassay techniques is dependent on the sensitivity of the instrument used. Sensitivity is a measurement of the detection capability (level of activity measurable) of the technique and thus is an indication of the adequacy of a bioassay technique. It should be noted that in statistics, the probability of a false positive and a false negative error is designated respectively by an alpha and beta probability and should not be confused with alpha- and β-particles.

2.1.6.1 Decision level (L_C)

The decision level provides a way of distinguishing the difference between the result from an analyte and the result from an appropriate blank. It answers the question whether a positive result has been detected with an alpha probability of Type I (false positive) and is calculated in Refs. [AN96] and [CU68] as:

$$L_C = 2.33\sigma_B \,, \tag{2.9}$$

where

σ_B = standard deviation of net count of a subject with no added analyte (standard deviation of background sample)

Type I = error set at 5%.

2.1.6.2 Minimum detectable activity or detection limit

The detection capability of a technique is characterised by its Minimum Detectable Activity (MDA) and is defined using an appropriate blank phantom or sample. The MDA is also known as the detection limit or Lower Limit of Detection (LLD). MDA is defined in Refs. [AN96] and [KR05] as the smallest amount (activity or mass) of an analyte in a sample that will be detected with a probability beta of non detection (Type II error) while accepting a probability alpha of erroneously deciding that a positive (non-zero) quantity of analyte is present in an appropriate blank sample (Type I error) and is calculated as follows (substituting L_C):

$$\text{MDA} = 2.71 + 2\,L_C\,, \qquad (2.10)$$
$$\text{MDA} = 2.71 + 4.65\sigma_B\,,$$

where

σ_B = standard deviation of net count of a subject with no added analyte (standard deviation of background sample)

Type I, II = error for both set at 5%.

2.1.7 ICRP biokinetic models

The particle size being inhaled by a person and the person's breathing pattern (mouth or nasal breather, heavy or normal breather) will determine how and where particles are deposited on the respiratory system [DO08]. The size of the aerosol particles will determine the region of the respiratory tract where most of them will be deposited. These processes are described by the deposition model in ICRP 66 [IC94]. Mechanical and absorption processes determine the rate of clearance from the lungs. Particles are cleared from the respiratory system through three clearance pathways using two processes: physical (mechanical) and absorption [RA94]. The three pathways are external (extrinsic), blood and the stomach.

Absorption in the fluid of the lung and the subsequent transportation into the bloodstream is a two stage process: the dissociation of particles into material that can be absorbed in the blood (dissolution) and the absorption of the dissociated particles and soluble particles in the blood (uptake). This absorption process (depicted by solubility class) is affected mostly by the chemical properties of the inhaled material (see Refs. [EI94] and [DO08]). Physical properties such as particle size are secondary to chemical properties with respect to dissolution. Thus, the outcome of inhaled particles is dependent on their physicochemical properties [AN99]. After absorption into the blood it is cleared via the kidneys into the urine. The solubility class is further described in Section 2.2.1.2.

Mechanical processes are ciliary motion (particle transport) and extrinsic means (e.g. nose blowing). Some are cleared through the airways (ciliary motion), swallowed, and excreted in the faeces. Ciliary clearance leads to transfer to the stomach by swallowing.

The different areas in the respiratory tract are depicted in Fig. 2.8. The ICRP Human Respiratory Tract Model (HRTM) consists of four regions: Extrathoracic, Thoracic Bronchial, Bronchiolar and Alveolar Interstitial, denoted ET, BB, bb, AL, respectively.

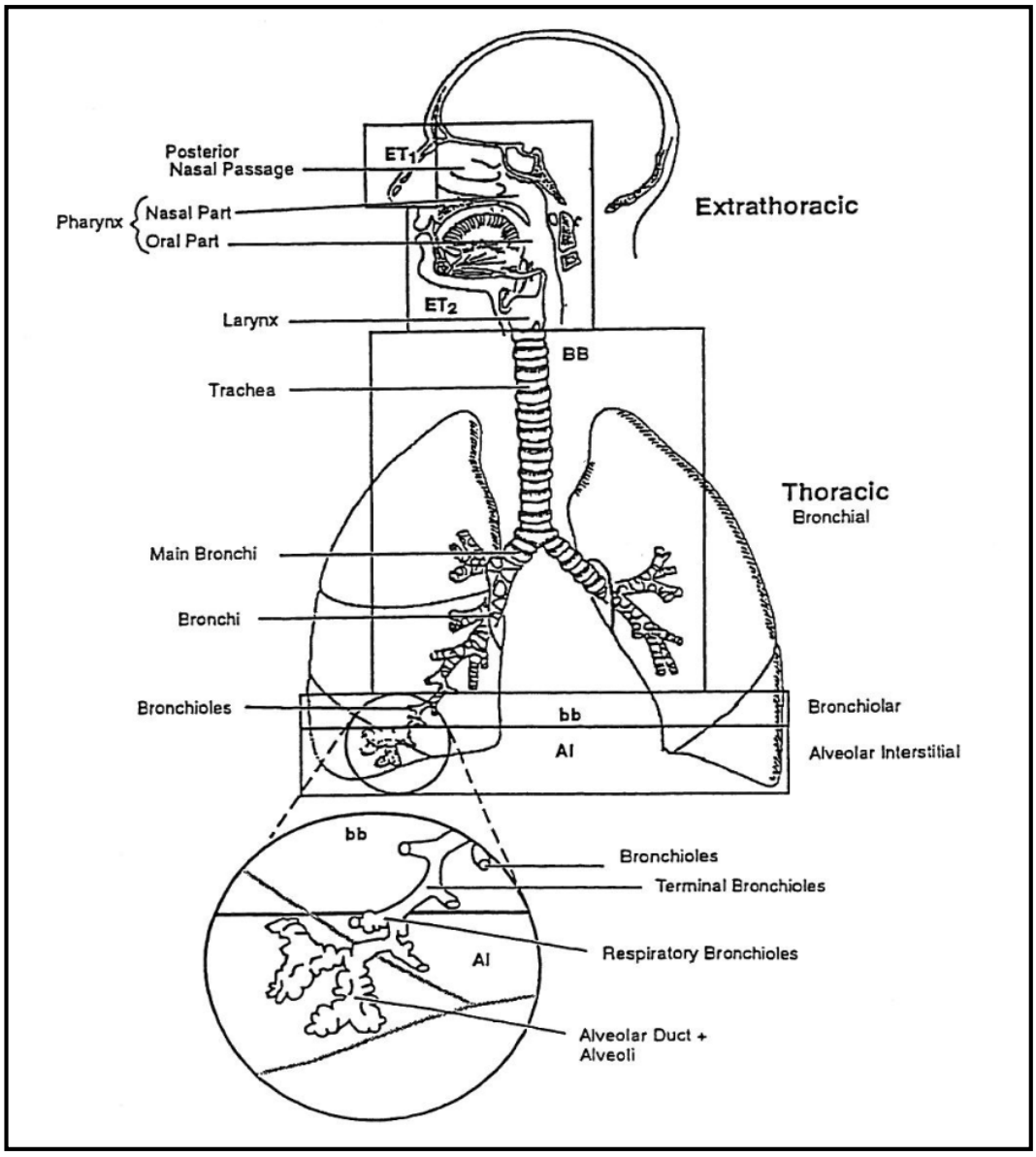

Figure 2.8: Respiratory tract regions for HRTM (taken from [IC94]).

The transfer of radioactive material through the gastrointestinal (GI) tract and the uptake in the blood is depicted by Fig. 2.9, the Human Alimentary Tract Model (HATM).

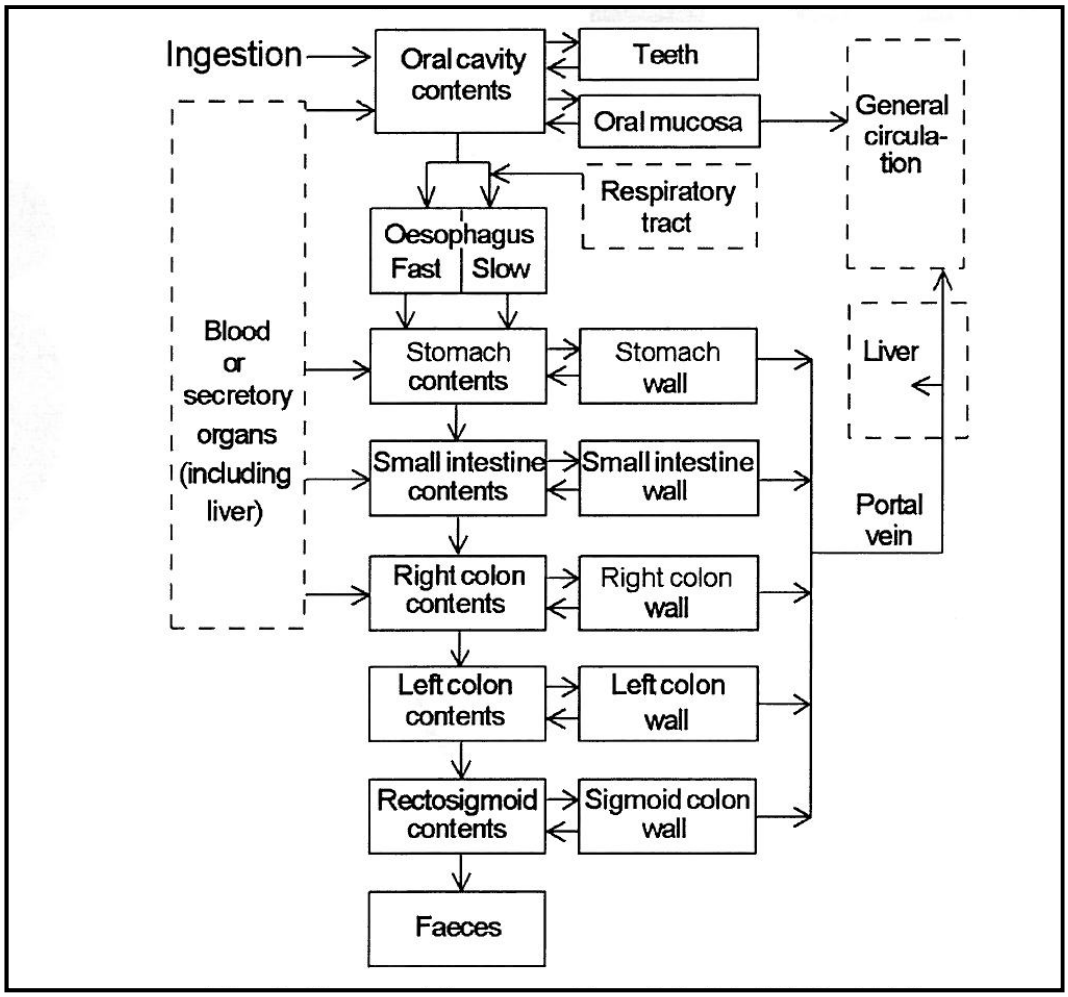

Figure 2.9: Human alimentary tract model (taken from [IC06]).

Material ingested or cleared from the lungs (ciliary motion) passes through the stomach, small intestine, upper large intestine and lower large intestine. The mean transit time (staying time) in the GI tract is very short (1 to 2 days); consequently only a small fraction of the ingested material is taken up in the blood, mostly from the small intestine. The uptake fraction depends strongly on the solubility of the ingested compound. Ingested material follows the same route as inhaled material (after it has been ingested via ciliary motion). The above

processes are represented by models of uptake and removal with a basic schematic depiction given in Fig. 2.10.

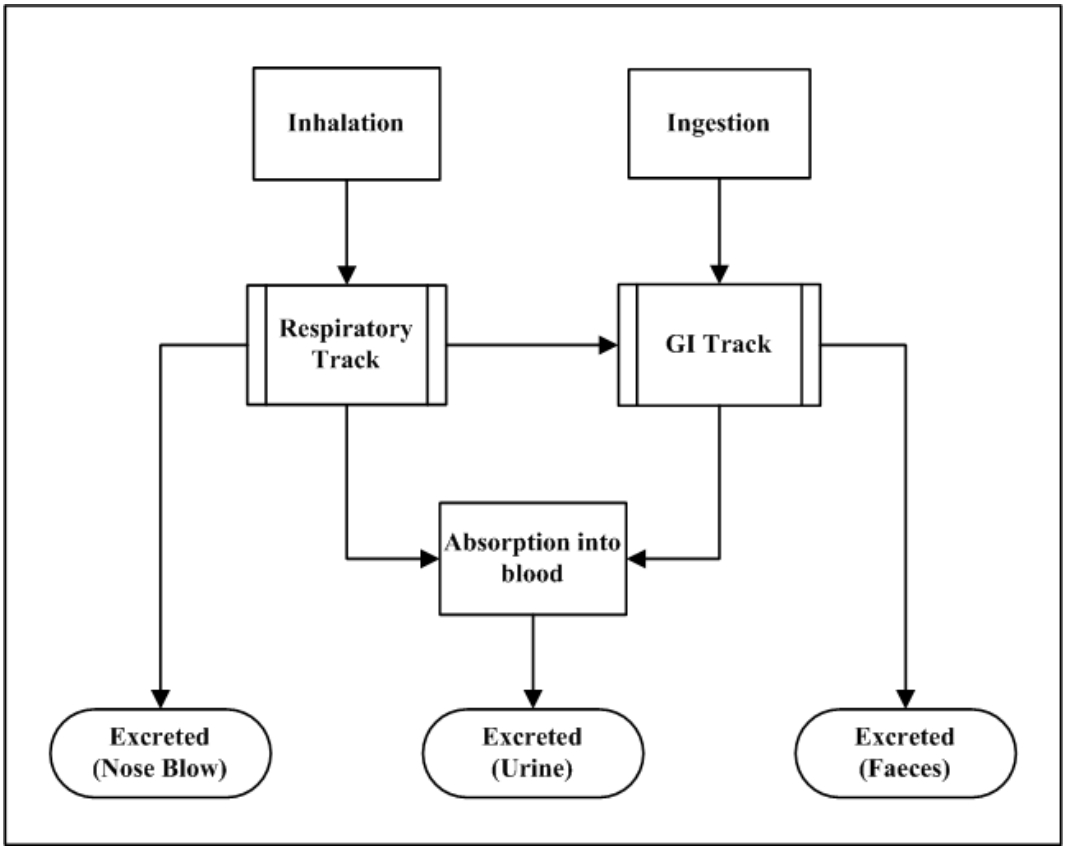

Figure 2.10: Basic routes after ingestion and inhalation of radioactive material.

2.1.8 Uranium biokinetics

The basic biokinetic model as modelled by ICRP is explained in the above section. Of specific interest for the present study is uranium and how it behaves in the human body. Inhaled soluble uranium, as described in Ref. [AN07], is swiftly transported from the lungs to the blood with most of it being excreted via the urine within the first day. A fraction of the uranium in the blood is deposited on bone surfaces where it is retained for several years. Significant deposits are also found in the kidney. Uranium, like most actinides, is bound in the bloodstream by the protein transferrin. It is deposited primarily in the skeleton and the liver. Some amounts are also deposited in the bone marrow, the spleen, muscle and gonads. The fractional deposition is 22% for the skeleton, 12% for the kidneys and 12% on other soft tissues [RA94]. Uranium is a heavy metal and

will subsequently damage the kidney tissue due to its chemical toxicity [EI94]. Uranium uptake may induce renal damage, which leads to renal failure in acute uranium poisoning [LE89]. Table 2.5 indicates default solubility classes according to ICRP [IC97] for typical compounds found within the uranium industry.

Table 2.5: Solubility class according to ICRP (taken from [IC97]).

Solubility Class	Compound
F	Most hexavalent compounds, e.g. UF_6, UO_2F_2 and $UO_2(NO_3)_2$
M	Less soluble compounds, e.g. UO_3, UF_4, UCl_4 and most other hexavalent compounds
S	Highly insoluble compounds, e.g. UO_2 and U_3O_8

The uptake of uranium by the blood from the lungs is significantly slower for insoluble uranium, leading to increased radiation exposure (compared to soluble) to specifically the respiratory tract. Subsequently doses to the liver, bone and kidneys will be less due to the slower uptake. The uranium particles remaining in the lung constitute a larger potential radiological hazard from the alpha decay energy absorbed in the surrounding tissue than particles that are cleared into the bloodstream and deposited in the bones and kidneys. This is due to the longer stay time in the lungs and the more sensitive tissue in the lungs [EI94].

The Activity Median Aerodynamic Diameter (AMAD) values depended on the specific uranium compounds, the chemical impurities in the compound and the manufacturing process. ICRP recommends using defaults values of 5 μm AMAD [IC97]. A review of literature by Rucker [RU01] concluded that the solubility rate constant (or rate of dissolution) increases rapidly as the particle size is reduced.

2.2 Workplace characterisation

The monitoring programme should be determined with consideration of the magnitude, possible fluctuations and likelihood of exposure levels [NR04]. Thus, is essential that the workplace and the job should be characterized before any decisions can be made on a monitoring programme. A workplace is characterised according to:

- the physicochemical nature of radionuclide compounds found in the workplace and
- the exposure scenarios (spatial, temporal and route) by which the human body is internally contaminated.

2.2.1 Physicochemical nature of intake compounds

The physical (specifically particle size) and chemical nature of a compound will dictate where, how much of it and for how long it will be deposited in the human body. The physical nature, specifically it radiological characteristics, will determine the radiological burden. This is one part of the workplace characterisation and is essential information in order to develop a monitoring programme. The physicochemical nature of the compound comprises largely of the following features:

- particle size,
- solubility class and
- radiological characteristics.

Each of these features is dealt with in turn below.

2.2.1.1 Particle size (AMAD)

Activity Median Aerodynamic Diameter (AMAD) is a measure of the particle size distribution of airborne radioactive aerosols and particles. It is that diameter of which fifty percent of the aerosol's activity is greater than the AMAD. Particle size determines the pulmonary pattern and, therefore, the associated radiological hazard from inhalation [CE96]. The AMAD combined with the breathing rate and whether the person is a nose or mouth breather determines the fractional deposition in each region of the respiratory tract.

Reference [DO95] stresses the importance of having realistic parameters for use in radiation dose assessments and thus recommends that particle size distribution should be measured for individual work practices. This type of study is outside the scope of the present research project. Default particle sizes which are characterized by AMAD values should only act as a guide if detailed information is not available. Inhaled aerosols could be due to particles generated from a process or from resuspended surface contamination. Resuspended aerosols tend to have larger AMAD values typically in the order of 6 µm.

2.2.1.2 Solubility class

Inhaled aerosol material is classified according to their clearance from the lungs. These classifications refer to the length of time particles from inhaled aerosols are retained in the pulmonary region [IC97]. Inhaled material is classified either as Type S, M or F. The stay time of inhaled particles has a significant impact on the committed effective dose. Chemical form determines solubility and consequently transportability in body fluids. The transportability of inhaled or ingested material determines its fate within the body and, therefore, the resulting radiation dose.

The monitoring of inhaled radiological contaminants is based on an understanding of the lung solubility of inhaled contaminants that are present at a facility [RU01]. Literature recommends that solubility studies be performed, but have been shown to be complicated to measure [DO04]. This type of study is again outside the scope of the present research project. The three classifications for absorption rates expressed as half times (see Section 2.1.7) are defined as follows [IC97]:

Type F (Fast): 100% absorbed with a half-time of 10 minutes

Type M (Medium): 10% absorbed with a half-time of 10 minutes and 90% is absorbed with a half-time of 140 days

Type S (Slow): 0.1% absorbed with a half-time of 10 minutes and 99.9% is absorbed with a half-time of 7000 days.

As can be seen, these three classes are very broad and range from minutes to days to several years. Therefore, knowing the solubility of intake particles to which exposure is possible is of vital importance when determining how to protect workers, with less soluble materials posing a greater radiological exposure hazard.

2.2.1.3 Radiological characteristics

The radiological characteristics of an isotope are defined amongst others by its type of decay (including energy of decay radiation), half-life and activity (including activity composition: mass enrichment). These characteristics play an important role in the calculated CED and the monitoring programme. Radiological characteristics are discussed in Section 2.1.

2.2.2 Exposure scenario

The exposure scenario provides information that will assist in determining the monitoring programme and assist in calculation of CED. It can be described according to the following attributes:

- spatial (sources of intake),
- pathway (routes of intake) and
- temporal (time of intake)

Each of these attributes is discussed in turn below.

2.2.2.1 Sources of intake (spatial)

Information such as where intake occurs and what the sources are represents the essential basic information needed. This will aid in developing control measures to prevent intake and to characterise intake. Based on the processes involved, one can determine which nuclides workers are exposed to and what will be the possible route of intake. Processes will determine whether there is a significant risk of internal intake and whether it should be monitored. Dry processes most probably will lead to material becoming airborne. If these processes are not contained it could result in inhalation of the material. Loose contamination being resuspended and becoming airborne is also a major source of exposure.

Contamination control plays an important role in the overall effectiveness of a radiological control programme in respect of internal exposures. Loss in containment will result in surface contamination and airborne dispersion that could lead to uptake of nuclides and subsequently to internal exposures. Airborne contaminants are continuously removed from the work place with the aid of a ventilation system and filtration. Contamination control is also provided by protective clothing and respiratory protection.

Information gained from determining the spatial characteristics of the exposure will aid in determining the monitoring programme, specifically the monitoring technique.

2.2.2.2 Routes of intake (pathway)

The route of intake can be by inhalation, ingestion, through a wound or injection or a combination these pathways. In Ref. [LE10] it was found that significant errors occur if only one intake route is assumed and that there were significant variations in the CED if different routes of intake are assumed. Thus, it is important to know the most likely route of intake and in cases of a significant intake is expected one should be as specific as possible. For routine monitoring, one intake path is usually assumed. This assumption can lead to high uncertainties.

In routine monitoring situations the pathway will most likely be inhalation, but it could also be ingestion or a combination of inhalation and ingestion. However, in Ref. [ID06] it was proposed that ingestion should be assumed only in those cases where there is clear evidence for this pathway (well established and documented). Otherwise the inhalation pathway should be assumed.

Information gained from determining the route of intake will aid in determining the monitoring programme (specifically the monitoring technique) and aid in the CED calculations.

2.2.2.3 Time of intake (temporal)

In Ref. [ID06] time of intake is categorised and defined as follows:

Acute intake: An intake occurring within a time period short enough that it can be treated as instantaneous for the purposes of assessing the resulting committed dose.

Chronic intake: An intake over an extended period of time, such that it cannot be treated as a single instantaneous intake for the purposes of assessing the resulting committed dose.

Information gained from determining the time of exposure will assist during CED calculations. Dose conversion factors for routine exposures in Ref. [IC97] are based on the assumption of an acute intake in the middle of the monitoring period. An intake in the middle of the monitoring period is also the default recommendation if the time of intake is unknown or if the exposure is chronic. Latest software available is capable of modelling true chronic intakes, thus negating the default recommendation of the ICRP.

2.3 Monitoring programme

A monitoring programme is divided into three monitoring strategies *viz*. routine, task related or special monitoring. The location of the monitoring is at the workplace (e.g. static air samplers) and at an individual level (e.g. passive air monitors, bioassay protocols). Workplace and individual monitoring are complementary to each other. Workplace monitoring will provide basic information needed to develop a programme of individual monitoring. Individual monitoring consists mainly of bioassay protocols, which are supported by personal air monitoring and workplace air monitoring. The bioassay technique and frequency will be decided by physiochemical nature (decay method, effective half-life, solubility etc.) of the nuclide that needs to be detected, its biokinetic behaviour within the human body and the sensitivity of the measurement technique.

The objective of the OMINEX Project [ET03] is to enable users to optimise the design and the implementation of a monitoring programme. This includes, amongst other possibilities, the choice of monitoring methods, measurement techniques monitoring intervals, monitoring frequencies and measurement sensitivity.

2.3.1 Monitoring strategies

There are three types of monitoring strategies which are described in Ref. [IA99b] as follows:

1) Routine or Confirmatory Monitoring

> *"It is associated with continuing operations and is intended to demonstrate that the working conditions, including the levels of individual dose, remain satisfactory, and to meet regulatory requirements. It is thus largely confirmatory in nature, but underpins the overall operational monitoring programme. Routine monitoring programme are made at pre-determined times which is not related to known intakes, and therefore it is necessary to make some assumptions about the pattern of intakes."*

2) Task Related Monitoring

> *"It applies to a specific operation. It provides data to support the immediate decisions on the management of the operation. It may also support the optimization of protection. In these cases, the time of intake or potential intake is known and workplace monitoring may provide additional information on the physical and chemical nature of any contamination."*

3) Special Monitoring

> *"It is investigative in nature and typically covers a situation in the workplace for which insufficient information is available to demonstrate adequate control. It is intended to provide detailed information to*

elucidate any problems and to define future procedures. It should normally be undertaken at the commissioning stage of new facilities, following major modifications either to facilities or procedures, or when operations are being carried out under abnormal circumstances such as an accident."

The focus of the present study is on routine monitoring. Thus, it is essential to note that this monitoring strategy is made at predetermined times which are not related to a known intake. Assumptions with respect to time of intake are made and the impact thereof will be discussed in the latter part of the study. Task related and special monitoring are excluded from the present study.

2.3.2 Location of monitoring techniques

Monitoring is done at two distinct locations. In Ref. [IA99b] the locations are described as follows:

1) Workplace Monitoring

"Workplace monitoring comprises measurements made in the working environment."

2) Individual Monitoring

"Individual monitoring is taken to mean measurement by equipment worn by individual workers, or measurement of quantities of radioactive materials in or on their bodies, and the interpretation of such measurements."

Typical workplace monitoring techniques are airborne contamination monitoring such as Static Air Samplers (SAS) alternatively known as Continuous Air Monitors (CAM) and surface contamination monitoring. Typical equipment worn includes personal air samplers and measurement of material in the body is grouped as bioassay techniques. The focus of the present study is on individual monitoring.

2.3.2.1 Workplace monitoring

Workplace monitoring should be used to trigger a programme of individual monitoring. It includes requirements such as the placing strategy of air samplers. Air samplers should be strategically placed; *viz.* placed where it is representative of the air breathed by workers (if it is to be used for internal dose calculations) or placed where it is representative of the average air concentration of a workplace. Workplace monitoring and an IDMP are complementary to each other. However, the requirements for workplace monitoring are not within the scope of the present study.

2.3.2.2 Individual monitoring

Bioassay is defined as any procedure used to determine the nature, activity, location or retention of radionuclides in the body by direct (*in vivo*) measurement or by indirect (*in vitro*) analysis of material excreted or otherwise removed from the body [IA99a]. *In-vivo* and *in-vitro* monitoring can thus also be grouped as individual monitoring. Other methods to determine internal contamination are by monitoring the airborne contamination. This includes personal air sampling (also classified as individual monitoring) and static air sampling (classified as workplace monitoring).

The order of preference for bioassay is direct body activity measurement (*in-vivo*), excreta analysis (*in-vitro*) and then personal air sampling [IC97].

In-Vivo monitoring

In-vivo monitoring is the direct monitoring of γ-ray emitting radionuclides taken up in the whole body or certain organs [SC03]. Counting activity in a body is only possible for nuclides emitting radiation that can escape the body. These are usually gamma emitters such as fission and activation products (e.g. ^{131}I, ^{137}Cs, ^{60}Co and ^{99}Mo/^{99m}Tc). These methods can also be used to determine the uptake of low-energy photon emitters such as long-live alpha emitters (^{235}U, ^{239}Pu and ^{241}Am). The types of monitoring techniques available for body activity counting are Whole Body Counting, Lung Counting, Thyroid Counting and Skeleton

Counting (knees and skull). Advantages are that it can be used for routine monitoring since it is direct, quick and a convenient measurement. Monitors (usually scintillator based detectors) with energy discrimination capability are necessary in cases with a mixture of radionuclides. *In-vivo* techniques are generally more accurate than *in-vitro* techniques because they do not rely on metabolic information that is required to determine excretion rates of nuclides. They are also more able to monitor for insoluble nuclides that are not readily dissolved and excreted. It has the disadvantage that equipment could be expensive (e.g. massive shielding, large detectors) and external contamination can be mistaken as a true intake.

In-Vitro monitoring

In-vitro monitoring is the indirect monitoring of radionuclides taken up in the body. The activity in the body is inferred from analysing material excreted or removed from the body. This method is used for radionuclides that emit no γ-rays or very low energy γ-rays that cannot be detected with *in-vivo* counting [SC03]. It is in most cases not interpreted quantitatively, but rather used as confirmation of satisfactory conditions [IC97]. Uncertainties associated with *in-vitro* monitoring are very high since the excretion functions for many radionuclides are not well known and there are large variations in excretion rates between individuals [SC03]. Typical excreta that are analysed are:

- faeces,
- urine,
- blood and
- other e.g. nose blows, breath, hair and nails etc.

In-vitro monitoring has a further disadvantage since it is unpleasant and requires samples of urine and faeces.

The chemical and physical form (physiochemical nature) of the material determines its behaviour on intake and its subsequent biokinetic behaviour in the human body, which will determine the route of excretion and subsequently the *in-vitro* technique to be used.

Faeces excretion has two components (see Refs. [IA99a], [ID06]):

- systemic faeces excretion which represents removal of systemic material via the Gastro Intestinal (GI) tract and
- direct faeces excretion of the material passing unabsorbed through the GI tract.

It is used for monitoring insoluble material such as Type-S or Type-M material ingested from the respiratory tract. Faeces should be collected over a period of several days, which could be impracticable. Uncertainties are high due to the daily fluctuations in faeces excretions. Consequently, the accuracy is mostly larger than a factor of 2. This technique is used in combination with urine analysis and lung counting. It is not usually done routinely and more often used in special monitoring due to above mention impracticalities and uncertainties.

Urine analysis is based on the removal of material from the plasma and extra cellular fluid. It is mostly used for soluble material, but can also be used for insoluble material. Typical radionuclides monitored are tritium (β^- emitter) and uranium compounds (see Refs. [NR04] and [ID06]).

The recommendation in Ref. [IA99] is to analyse blood in accident situations in which large intakes are suspected. This can provide data on the solubility and the biokinetic of the material involved, but has limited value for providing quantitative estimates of the intake at low doses.

Other excreta analysis includes nose blows, nose smears and exhaled breath. Typical nuclides monitored are ^{14}C, or ^{232}Th daughters, ^{220}Rn, ^{226}Ra and ^{228}Th. Measuring nose blows and nasal smears can be used as screening techniques in suspected incidents [IC97]. By using regional deposition fractions (fraction deposited in nose during inhalation), an estimation of the intake can be made. Analysis of hair and nails can provide additional information with respect to temporal distribution of intake [KA01].

Personal Air Samplers (PAS)

In Ref. [LO07] an internal dosimetry programme based on air sampling is described as the collection of a representative air sample. The air sample is representative of the activity concentration in the air inhaled by a worker. This allows the estimation of CED based on time exposed to the measured air concentration. Air samples can be taken with either personal air samplers or workplace samplers.

2.3.3 Monitor frequency and sensitivity

The frequency of monitoring will depend on the rate of retention and excretion of the radionuclide (biological half-life), the physical half-life, the sensitivity of the measurement technique and the acceptable uncertainty in the CED.

The chemical and physical form (physiochemical nature) of the material determines its behaviour on intake and its subsequent biokinetic in the human body, which will determine the stay time within the human body and subsequently the rate of excretion. This is characterised by the biological half-life of the nuclide. Effective half-life is defined (see Section 2.1.1) as the combination of physical half-life (radioactive decay) of the nuclide and the biological half-life (biological clearance).

In setting a schedule, one should ensure that an intake above a predetermined level is not 'missed'. An intake could be missed if the measured activity were to decline to a level below the sensitivity of the measurement technique during the time interval between the intake and the measurement. This is determined by the effective half-life of the nuclide. Thus, frequency of monitoring will also be driven to a great extent by the sensitivity of the measurement technique.

In Ref. [IA99a] it is proposed that the intervals of measurement are typically set so that intakes corresponding to >5% of the projected annual dose limit are not missed. The annual dose limit for whole body exposure is 20 mSv, as set by the ICRP [IC07]. The projected annual missed dose should thus not be more than 1

mSv (5% x 20 mSv). This is similar to the recording level set in Ref. [IC88] at 10% (2 mSv) of the annual dose limit. In Ref. [AN01] it is recommended that screening levels be significantly lower than this. The recommendation in Ref. [NR04] is to use 1 mSv, since a worker is only defined as a radiological worker if the annual dose exceeds 1 mSv. These levels can be seen as the maximum Decision Level (DL). This required sensitivity or required frequency can be calculated as follows:

$$\text{Sensitivity (mSv)} = 1 \text{ mSv} \times \frac{\text{MPe (days)}}{365 \text{ (days)}} \tag{2.11}$$

and

$$\text{MPe (days)} = 1 \text{ mSv}^{-1} \times \frac{\text{Sensitivity (mSv)}}{365 \text{ (days)}} \quad , \tag{2.12}$$

where

MPe = Monitoring Period (frequency)

l mSv = Maximum DL (missed dose)

Derived sensitivity values (derived decision levels) can be calculated based on the equivalent of 1 mSv *viz.* µg/ℓ or Bq/ℓ for urine monitoring (total uranium) and Bq for lung monitoring (only ^{235}U).

Routine bioassay measurement periods longer than five effective half-lifes are generally not recommended, because the potential individual specific deviations from assumed retention or excretion patterns can substantially affect associated doses. Five effective half-lifes imply that the original intake has decreased to 3% (0.5^5) of its original value. Furthermore, the sampling schedule (monitoring period) should also minimize the uncertainty in the estimated intake due to the unknown time of an intake within the monitoring period. In Ref. [IC97] it is recommended that monitoring periods should generally be selected so that assuming an intake to have occurred at the mid-point of the monitoring period it would not lead to an underestimation of the intake by a factor of more than three. This is in relation to the most conservative intake, *viz.* an intake on the 1st day of

the monitoring period. A solution for this is suggested in Ref. [ST03] and is discussed further in Section 3.3.3.

In order of preference, the criteria to be used for determining the frequency for a routine monitoring strategy are set out as follows:

- projected annual missed effective dose to 1 mSv,
- underestimation of calculated CED less than a factor 3 and
- practicality.

2.4 Internal dosimetry

Internal dosimetry is the scientific methodology used to calculate the activity retained in the human body and the resultant committed effective dose. Different doses can be calculated from the same set of measurement date due to several physical and human metabolic parameters that can vary significantly.

2.4.1 Committed Effective Dose (CED) calculation steps

In general a CED is calculated as follows:

Step 1: Bioassay measurement. Typical units are Bq in lungs, whole body or thyroid, and Bq/ℓ for urine. In essence, this is representative of the uptake. The uptake in a body part (organ) is determined from direct measurement of the organ (e.g. lung counting) or derived indirectly from excreta measurement (e.g. uptake in blood derived from activity measurements in urine).

Step 2: Calculate intake. Assume a route of entry and using the biokinetic behaviour of the nuclide, back calculate the intake from the measurement point (e.g. urine, lungs). Correct also for time of measurement, *viz.* correct for physical and biological decay.

Step 3: Calculate CED. Using the biokinetic behaviour of the intake nuclide, determine the distribution in the body and stay time in the various organs. Calculate CED using Eq. (2.8).

2.4.2 The philosophy of internal dosimetry

The basic philosophy for internal dosimetry was formulated by Project IDEAS [ID06]. It states:

> *"internal dosimetry, as in external dosimetry, the philosophy is that if two persons have the same internal exposure then the results of internal monitoring in terms of committed dose should be consistent (**harmonised**) with each other, and the results should be considered to be the best estimate (**accurate**). This entails a consistent approach to dose evaluation and the approach should lead to a best estimate. In cases of uncertainties it is more important to ensure that the committed dose is unlikely to exceed a specific level."*

For a given set of internal monitoring data in terms of body/organ activity and/or urine/faeces activity there should be one standard estimate for the intake and the committed effective dose. This standard estimate is defined by the monitoring data, the biokinetic models, dosimetric models, and some additional information, such as time of intake, route of intake, particle size, respiratory tract absorption type (solubility), etc. Recent previous internal exposures should also be taken into account.

The aim of Project IDEAS is thus to develop general guidelines for standardising assessments of intakes and internal doses [DO07]. Project IDEAS [ID06] proposed the following guiding principles:

1) Harmonisation

Any two assessors should obtain the same estimate of dose from a given data set.

2) Accuracy

The best estimate of dose should be obtained from the given data set.

3) Proportionality (Graded Approach)

The effort applied to the evaluation should be proportionate to the dose. The higher the dose, the more complex the approach should be and the lower the dose, the simpler the process.

Together the three principles will guide the formation of a standard estimate. This is detailed below.

2.4.2.1 Graded approach

The effort applied to the evaluation should be proportional to the dose [DO07]. As doses increase in magnitude, sufficient measurement results and workplace characterization data (physiochemical nature and exposure scenario) should be obtained in order to make adjustments to the models. This is to account for the unique behaviour of the radionuclides in the individual's body. Reference dose levels should be established which require enhancement of data collection and individual specific dose assessment efforts. A graded approach, with increasing level of complexity, was developed by Project IDEAS [ID06] and is described below.

2.4.2.2 Harmonised and accurate

Harmonising is achieved by following proper written procedures. Project IDEAS has developed a standard approach when estimating a dose. This approach is described in detail in the following section. In the case of routine monitoring, usually based on a single measurement, the procedure is simple. In instances where there are several measurements from different techniques, it can lead to different ways of handling data and subsequent different doses. In these cases, harmonisation is achieved by a systematic approach in handling data and changing parameters.

This standard and systematic approach becomes more complex as the dose increases [ID06]. Such a multifaceted approach ensures higher level of accuracy as the dose increases. Uncertainties associated with assessed internal dose can be significant. This is especially the case for actinides which are difficult to detect in the body and which have relatively high dose coefficients ($SvBq^{-1}$). Thus, it is important to make best use of the available information.

The ICRP has recently developed more realistic internal dosimetry models, namely the HRTM, HATM and recycling systematic models for actinides. However, these guidelines leave many assumptions open for interpretation, i.e. when to use assumed default values [DO07]. The biokinetic models, although realistic, are based on standard metabolic models and, therefore, do not necessarily reflect the true effect of uranium uptake in a real person. An individual's metabolism will not necessarily agree with this model. This leads to significant differences from using defaults values when calculating doses. A warning was given in Ref. [DO07] that these default values may not be valid in specific situations. Thus, it is important to understand the effect of using default parameters and the uncertainties associated with them. Preferably, assumptions should be more individual and exposure specific, especially when intakes have a significant dose impact. Guidance is needed on which ICRP default parameters are reasonable to change. This guidance is provided by Project IDEAS [ID06].

The aim is to change default values until a best fit is found. A measure of the "Goodness of fit" (GOF) and the criteria for deciding that the fit is good enough are critical. There may be conflict between "harmonisation" and "accuracy" [ID06]. The more generic an approach is (to ensure harmonisation) the less accurate it may be. Generally, the better the data (quality and quantity) the more likely it is that a statistical test will show that the data are inconsistent with the default model. Less data will probably fit the model easier. In the case of a single measurement any model will fit. Thus, it is important to ensure there are sufficient data for assessment of a significant dose. The default parameters are

changed and subsequently the default model. This is done in a systematic way till the model fits the data. Several statistical methods (maximum likelihood method, the Bayesian approach) for data fitting are available [IA04b]. Typical default parameters include the physiochemical properties (AMAD, solubility) of the intake nuclide and the exposure scenario of the intake (route and time of intake). It is the aim of the present study to investigate the effect of these parameters on the CED.

2.4.3 The standard estimate for internal dosimetry

Together the three principles studied above guide the formation of a standard estimate. This standard estimate follows a graded approach ensuring accurate and harmonised calculation of CED. In Ref. [ID06] the following standard estimate is recommended:

Level 0: Annual projected CED most likely < 0.1 mSv

- CED need not be calculated;
- measured value should be recorded (could be needed for further assessments in the future);
- implies typical measured doses for the following monitoring period: 1 month (~8 μSv); 3 months (~25 μSv) and 6 months (~250 μSv).

Level 1: Annual projected CED between 0.1 – 1 mSv;

- uncomplicated CED calculation based on ICRP default parameters;
- *a priori* information such as AMAD can be used if available.

Level 2: Annual projected CED between 1 – 6 mSv;

- perform a realistic assessment of dose;
- typical parameters to be adjusted relates to the material (AMAD and absorption type) and exposure scenario (time of intake);
- obtain a reasonable fit with predicted data.

Level 3: Annual projected CED > 6 mSv;

- comprehensive set of measurement data is needed;
- typically parameters to be adjusted relates to the individual e.g. HRTM particle transport rates in case of inhalation (including parameters relating to the material and the exposure scenario);
- systematically change default model parameters until fit is acceptable to all of the data;
- provide valid justification for rejected data.

The annual projected dose is accumulated over a period of 12 months assuming similar intakes in each monitoring period (MPe) and is given by.

Annual projected CED = Measured dose x Number of MPe per year. (2.13)

The above levels should be used for doses for a specific period as well as accumulated doses for several consecutive periods. Thus, re-evaluate a previous period's dose according to the next level should the accumulated dose for successive periods be in a higher level.

CHAPTER 3

DEVELOPMENT OF URANIUM MONITORING PROGRAMME. RESULTS, DISCUSSION AND RECOMMENDATIONS

Airborne workplace exposure to radioactive substances should be characterised by taking into consideration all available information regarding the physical and chemical (physiochemical) nature of the airborne contamination. This includes the spatial and temporal distribution of the exposure, where and when the intake occurs and the route of intake (exposure scenarios). Based on such information, for the present study a monitoring programme for uranium will be developed. To this end, in the first part of this chapter simulated bioassay measurements are generated in order to evaluate the effect of changing input parameters and default values. The aim is to quantify the sensitivity of the Committed Effective Dose (CED) calculation for changing parameters and to determine which parameters are important (sensitivity analysis). In addition, it will provide an understanding of the uncertainties associated with reported CED values and, when using a graded approach, one will also know which parameters will lessen uncertainties and provide a more accurate estimate of the CED. Based on the above and a literature survey, this chapter will provide recommended physicochemical values and aims to describe the complexity of exposure scenarios within a uranium plant. It is not within the scope of the present study to experimentally determine the physicochemical nature of the uranium exposures. As reported in the literature research it has been shown that these types of experiments can be extremely complex and complicated and requires a study on its own. Therefore, it was decided to pursue a literature study for similar exposure situations and use the literature recommended values as applicable. As such, typical processes and uranium compounds found within a uranium recovery and uranium alloy manufacturing plant will be described. In order to develop a site-specific uranium

monitoring programme, a literature study was done on various bioassay techniques used in various other programmes. The results were then compared with bioassay techniques used by Necsa and the limitation of the various techniques determined and discussed. The specific characteristics (e.g. sensitivity) of the various techniques were investigated. Using the above information as input, including literature study of uranium specific bioassay programmes, an individual monitoring programme for Type M uranium was developed for Necsa based on the guiding principles in Chapter 2.

3.1 Physiochemical nature of intakes

The physicochemical nature of the compounds found within a uranium plant can be described by the following characteristics:

- particle size,
- solubility class and
- radiological characteristics.

Each of the above are dealt with in turn below.

3.1.1 Particle size (AMAD)

Default particle sizes, characterized by Activity Median Aerodynamic Diameter (AMAD) values should only act as a guide if detailed information is not available. Actual determination of AMAD values is not part of the present study. As such, a literature survey for recommended AMAD values was undertaken and these values were used as applicable. The effect of AMAD on CED and intake values will be shown first where after the variances in typical AMAD values will be discussed. For the sake of intercomparison, all figures and tables are shown together at the end of this section.

3.1.1.1 Effect on CED per unit intake

Hegyi has shown the considerable effect of AMAD on CED [HE08]. For Type S ^{234}U, the ratio of CED between 0.4 μm and the default of 5 μm AMAD is 1.5 and the ratio of CED between 13 μm and the default of 5 μm AMAD is 0.5. This is for a fixed unit intake values (1 Bq/day) irrespective of the monitoring technique.

This can be seen in Fig. 3.1 and Table 3.1, as confirmed by calculations done by the author. Similar values were found by Dorrian [DO95]. A calculated CED can thus change up to 200% as the AMAD is changed. The same effect was found for Type M uranium as can also be seen in Fig. 3.1 and Table 3.1. The overall trend is that the radiological risk and resultant CED decreases as the AMAD increases, as illustrated in Fig. 3.1. The CED initially decreases with increasing particle size up to about 0.6 µm [KI05]. Thereafter, it slightly increases with particle size up to about 2.5 µm and decreases again [HE08]. Particles that dissolve easily into the bloodstream pose a lower radiological hazard due to the smaller stay time in the lungs and is in agreement with the statements in Section 2.1.8 concerning uranium biokinetics. The solubility rate constant (or rate of dissolution) increases rapidly as the particle size is reduced [RU01]. It should be noted that the particle size of interest for a uranium plant is between 4 - 9 µm.

3.1.1.2 Effect on intake value per unit bioassay measurement

The previous section has shown the variance in the calculated CED (for a fixed unit intake) as the AMAD is changed. This section describes the variance in the calculated intake value as the AMAD is changed. The extent of the variance is also dependent on the type of monitoring technique. The intake is calculated from the measured bioassay value (using biokinetic models) and thus is dependent on the monitoring technique (see the basic steps for CED calculations in Section 2.4.1). This is especially so for monitoring methodologies that depend on lung counting and was shown to be the case for Type S ^{239}Pu and Type M ^{241}Am by Fujita [FU03]. In the present work, similar calculations were done for Type M uranium for urine analysis and lung counting and the results are shown in Fig. 3.2 and Table 3.2. The results corroborate with literature values, indicating a significant dependence for both urine analysis and lung counting with lung counting the most sensitive to changes to changes in AMAD.

3.1.1.3 Effect on CED per unit bioassay measurement

As can be seen above, the CED calculations are strongly negatively dependent on AMAD for a fixed intake value. The CED values decrease with increasing

AMAD values. In contrast, the intake calculation is strongly positive dependent on the AMAD for a fixed bioassay measurement. The combined effect of these two calculations is shown in Fig. 3.3 and Table 3.3.

This dependence is less severe for the combined calculation, urine analysis and lung counting. It should be noted that the CED is significantly less dependent on AMAD for monitoring based on urine analysis except for very small AMADs (1 μm) and the over and under estimation is ~ 4% (submicron particles are not expected in a typical uranium plant). The largest deviation from the default AMAD (5 μm) is 0.93 for AMAD of 6.5 μm. As such, AMAD does not need to be considered for monitoring programs based on urine analysis and the ICRP default value of 5 μm can be used.

In the case of lung counting, the over estimation is significant for AMADs larger than 6 μm. For AMADs between 4 μm and < 8 μm the effect on CED is within ± 5%. Very small (< 4 μm) or large (> 9 μm) AMADs are unlikely in a uranium plant environment, as can be seen in Table 3.5. Large AMAD values close to 8 μm are found for U_3O_8, which can lead to severe underestimation (~ 33% as from Table 3.3) if the default ICRP value of 5 μm is used. Although submicron particles are not expected in a typical uranium plant, there is a possibility that high temperature operations may produce submicron aerosols [DO95]. As such, AMAD does not need to be considered for monitoring programs based on lung counting and the ICRP default value of 5 μm can be used, except in cases where U_3O_8 is used or in high temperature operations. In these instances it is advised to determine the actual AMAD values.

3.1.1.4 Determining AMAD values from the lung/faeces ratio

AMAD values can be determined from the lung/faeces ratio. This was done for an intake of Type M ^{241}Am [FU03]. The ratios were calculated from lung and faecal measurements taken on day 5 after an acute intake. Similar calculations were done for Type M uranium and are shown in Fig. 3.4 and Table 3.4. Faeces excretion shows strong variation between individuals for the 1st week after an

intake. As such, the lung/faeces ratio should be determined with measurements taken 5 or more days after intake. This ratio should be used as an indication of significant deviations from the ICRP defaults value of 5 μm.

3.1.1.5 Typical AMAD values

The AMAD values depend on the specific uranium compounds, the chemical impurities in the compound and the manufacturing process. ICRP recommends using defaults values of 5 μm AMAD [IC97]. A study done by Ansoborlo [AN02] found AMAD values varied between 1.1 μm and 8.5 μm with an average at 5.7 μm (± 1.9 μm). This finding is for all uranium compounds found in various uranium processing industries in France. Typical AMAD values for compounds found in French uranium plants are given in Table 3.5 [AN02].

In addition, Ansoborlo [AN02] also compared the AMAD average of 5.7 μm (± 1.9 μm) with two other studies (4 μm and 5.5 μm) and found reasonable similarities (see Refs. [AN97] and [DO95]). Consequently, Ansoborlo supported the AMAD default value of 5 μm as recommended by the ICRP. Dorrian [DO95] did a literature research on average AMAD value published for various work environments and found a median of approximately 4 μm for nuclear fuel-handling industries and subsequently also supported the ICRP default AMAD.

The importance of AMAD has been indicated in the above mentioned literature as well as in calculations done by the author. Following a graded approach, the default ICRP AMAD value will be acceptable for routine Type M monitoring (lung counting and urine analysis). The recommended default ICRP values are representative of the inhaled aerosol and do not significantly under- or over-estimate dose for AMAD values between 4 μm and 8 μm. However, more accurate AMAD values should be used for intakes of U_3O_8 if the intakes are close to or exceeding reference levels (in the case of lung counting).

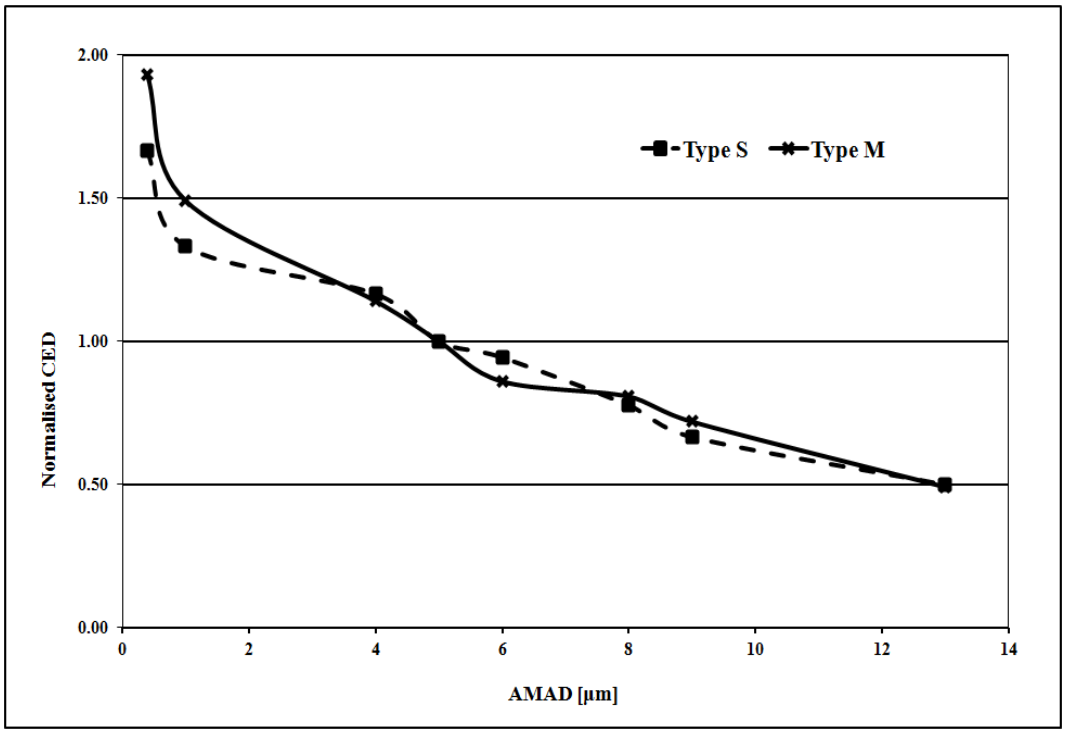

Figure 3.1: CED dependence on AMAD (fixed intake).

Table 3.1: CED dependence on AMAD (fixed intake).

	Normalised CED [a])	
AMAD (µm)	**Type M**	**Type S**
0.4	1.93	1.67
1	1.49	1.33
4	1.14	1.17
5	**1.00**	**1.00**
8	0.81	0.78
13	0.49	0.50

[a]) Calculations were done with IMBA assuming:

- chronic intake over 28 days,
- CED normalised to ICRP default AMAD value of 5 µm,
- low enriched (IMBA Values: 3.5%) and
- fixed intake of 1 Bq/day intake.

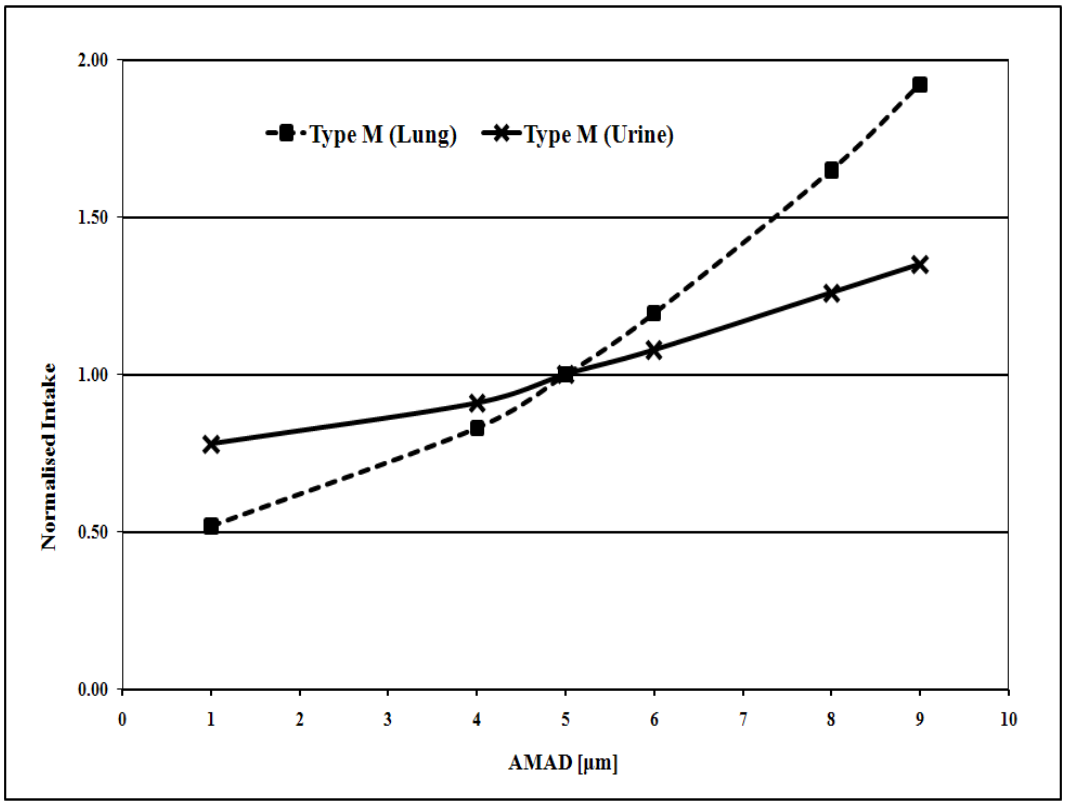

Figure 3.2: Intake dependence on AMAD (fixed bioassay measurement).

Table 3.2: Intake dependence on AMAD (fixed bioassay measurement).

	Normalised Intake [a])	
AMAD (µm)	**Lung Counting**	**Urine Analysis**
1	0.52	0.78
4	0.83	0.91
5	**1.00**	**1.00**
6	1.19	1.08
8	1.65	1.26
9	1.92	1.35

[a]) Calculations ware done with IMBA assuming:

- chronic intake over 28 days,
- Intake normalised to ICRP default AMAD value of 5 µm,
- 1 Bq measured in lungs on day 30 and
- 1 Bq/day measured in urine sample on day 30.

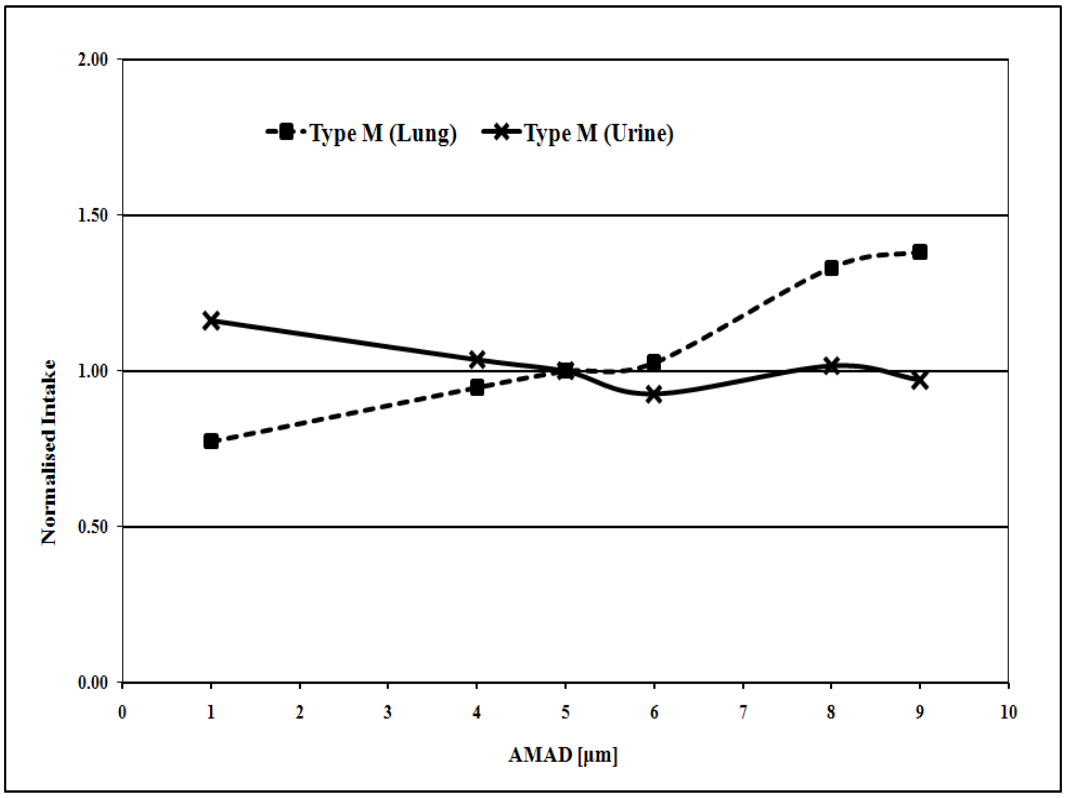

Figure 3.3: CED dependence on AMAD (fixed bioassay measurement).

Table 3.3: CED dependence on AMAD (fixed bioassay measurement).

	Normalised CED [a])	
AMAD (µm)	**Lung Counting**	**Urine Analysis**
1	0.77	1.16
4	0.95	1.04
5	1.00	1.00
6	1.03	0.93
8	1.33	1.02
9	1.38	0.97

[a]) Calculations were done with IMBA assuming:

- chronic intake over 28 days,
- CED normalised to ICRP default AMAD value of 5 µm with
- 1 Bq measured in lungs and 1 Bq/day measured in urine sample on day 30.

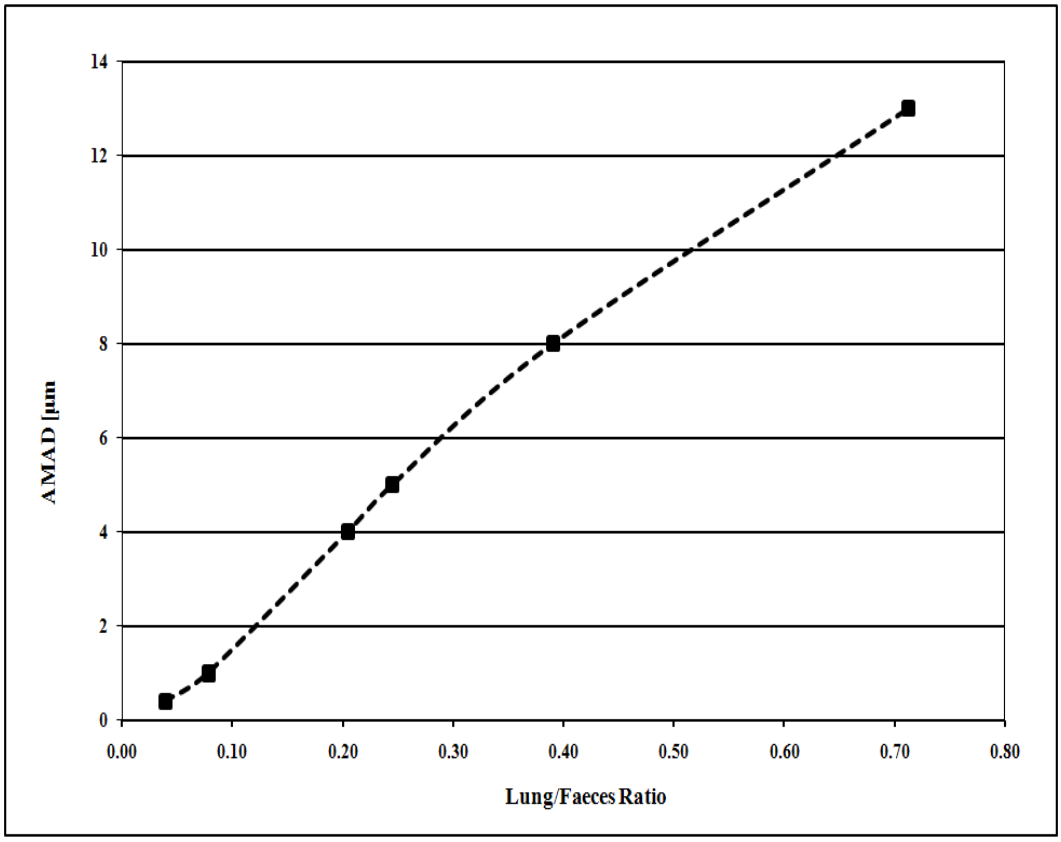

Figure 3.4: Faeces excretion *versus* lung content ratio for different AMAD.

Table 3.4: Faeces excretion *versus* lung content ratio for different AMAD.

AMAD (µm)	**Faeces Excretion / Lung Content at day 5** [a])
0.4	0.04
1	0.08
4	0.20
5	0.25
8	0.39
13	0.71

[a]) Calculations were done with IMBA assuming:

- acute intake 5 days prior,
- low enriched (IMBA values: 3.5%) and
- 1 Bq intake.

Table 3.5: Typical AMAD values (taken from [AN02]).

Compound	AMAD (µm)
U_3O_8	5.6 / 5.9 / 6.7 / 8.3 / 8.5
UF_4	4.2 / 5.1 / 5.8

3.1.2 Solubility class

The recommendation in literature is that solubility studies should be performed, but it has been shown difficult to measure [DO04]. Since this type of study is outside the scope of this research project, a literature survey was undertaken with a view to using determined values as applicable. Here solubility class and the subsequent stay time in the lungs plays a significant role in the calculated CED as described in Section 2.1.8 concerning uranium biokinetics.

3.1.2.1 Default solubility classes

Table 3.6 indicates default classes according to ICRP [IC97] for typical compounds found within the uranium industry. Normally found in a uranium plant are UAl_3, UO_3, $UO_2(NO_3)_2$, UF_4, UO_2, $UO_3.H_2.O.NH_3$ and U_3O_8. As described in Section 2.2.1, the absorption rate varies significantly between the three classes, from minutes to days to several years

Table 3.6: Solubility class according to ICRP (taken from [IC97]).

Solubility Class	Compound
F	Most hexavalent compounds, e.g. UF_6, UO_2F_2 and $UO_2(NO_3)_2$
M	Less soluble compounds, e.g. UO_3, UF_4, UCl_4 and most other hexavalent compounds
S	Highly insoluble compounds, e.g. UO2 and U3O8

3.1.2.2 Effect on CED

Here it should be noted that between solubility classes there are significant variances in CED. This is due to the large variances in absorption rate between the solubility classes and the effect on CED can be seen in Table 3.7. An incorrect solubility class for routine monitoring based on only urine samples can lead to under- or over-estimation by a factor of 1600 (0.0006/1) between Type F and S, a factor of 15 (0.0006/0.01) between Type F and M and a factor of 100 (0.01/1) between Type M and Type S. The under- or over-estimation is less severe with lung counting. It is a factor 2.1 (1/0.48) between Type M and S. Type F is not considered for lung counting due to the fast clearance from the lungs. The large variances in CED for urine monitoring can mainly be attributed to variances in calculating the intake from a unit bioassay measurement (1 Bq/day in urine) and not so much in calculating the CED from a unit intake (1 Bq/day intake). Thus, it is clear that information regarding solubility class is specifically important for urine monitoring.

Table 3.7: CED for different solubility classes.

Solubility class	**CED normalised (urine)** [a])	**CED normalised (lungs)** [a])
F	0.0006	-
M	0.010	0.48
S	1.000	1.00

[a]) Calculations were done with IMBA assuming:

- chronic intake over 28 days for urine and 180 days for lung,
- ICRP default values,
- low enrichment (IMBA values: 3.5%),
- 1 Bq/day measured in urine sample on day 30 and
- 1 Bq measured in lungs on day 180.

3.1.2.3 Factors to consider when evaluating solubility class

Within a specific solubility class, there can also be a wide range of dissolution rates. Most uranium compounds are actually a mixture of different chemical forms. This can result in a compound having fractional characteristics of F, M

and S at the same time. For UF_4, the dissolution rate varies because of a significant fraction of the UF_4 are Type F and S solubility classes and an appreciable fraction is Type M [EI94]. However, Type M is the recommended solubility class since the solubility of UF_4 is not strongly dependant on process history [EI94]. To an extent, lower Type S fractions were found for UO_3 which has the same recommendation as UF_4 [EI94] and indeed UO_3 may even display characteristics of Type F compound [RU01]. As such, the ICRP default solubility class for both UO_3 and UF_4 is Type M.

Another factor that need to be taken into account comes from the reviews done by Eidson [EI04] and Rucker [RU01] where it was found that the solubility of uranium oxides is very dependent on heat treatment. High temperature treatment involves temperatures that are close to or in excess of 800°C. The solubility class is strongly dependant on the materials process history according to Ref. [AN99]. Processing (thermal treatment) has an effect on the specific surface area of particles and subsequently the particle size distribution (AMAD) and also the dissolution. Heat treatment decreases the specific surface area, making the particle less soluble, thus closer to Type M solubility [RU01].

The rate of oxidation of uranium metals (UO_2) may also affect the solubility. Studies from actual exposure intakes have shown that some of the compounds indicated different solubilities, especially for compounds with a default solubility of Type S [EI94]. In particular, U_3O_8 is highly sensitive to process history, but most are consistent with Type S solubility class. Uranium oxides (e.g. U_3O_8, UO_2) will rather behave like Type M, except in cases of long term oxidation. Uranium metal alloy (e.g. UAl_3) is classified as Type S [RU01]. The ICRP default class for uranium oxides and metals are Type S. As can be seen, the literature recommendation is that these default values should be used with caution.

3.1.2.4 Recommendations regarding solubility class

Incorrect solubility class can lead to significant under- or over-estimation of CED, as can be seen in Table 3.7. Default values are usually on the conservative side,

resulting in the highest possible CED. Following the principle of a "graded approach", the resultant uncertainty in CED is acceptable for low doses. But in instances where doses approach investigation levels, it is recommended that further information is gained regarding the solubility to ensure a best estimate of the dose. This is especially so in the case of Type S compounds and preferably solubility studies should be performed to characterise the actual materials present at Necsa. Excretion rates can also be used to determine the solubility class (see Section 3.3.1). When setting up a monitoring programme or deciding on radiation protection control measures (e.g. ventilation, confinement, etc.) it is recommended to use the conservative default values, but the actual solubility class should be used for dose calculations if the doses approach investigation levels.

Normally found in a uranium plant are the compounds UAl_3, UO_3, $UO_2(NO_3)_2$, UF_4, UO_2 and U_3O_8. The solubility classes of compounds at Necsa's uranium plant, based on [RU01] and [EI94], can be classified as Type M, except where there is high temperature treatment of UO_2 or long term oxidisation of uranium metal (UO_2, U_3O_8). Long term oxidation does not occur within the uranium plant, but high temperature treatment of UO_2 does occur. This process is in an enclosed glove box, thus minimising the possibility of intake. Uranium metal (Type S) exposure could also occur during the reduction of UF_4 to uranium metal and during the melting of uranium metal and production of uranium shavings. But these either occur in a glove box or are done with the necessary respiratory protective equipment. Some processes are done on an *ad-hoc* basis, enabling task monitoring (see Section 3.2.1 for description of exposure scenarios). Thus, one can recommend a routine monitoring programme assuming exposure to Type M, while task monitoring for Type S is recommended (*ad-hoc* processes).

3.1.3 Radiological characteristics

Within a uranium recovery and fuel production facility, one only needs to consider uranium and its daughter products. The radiological characteristics of uranium are described in Section 2.1.2.

3.1.3.1 General radiological characteristics

The three major uranium isotopes mainly decay by emitting alpha and β^- particles. Subsequently, dose conversion factors (DCF) for the different isotopes of uranium will be the same because of similar radiological characteristics. Of concern is the high LET value for the alpha particles, thus posing a significant internal radiological hazard (see Section 2.1.8). The inhalation dose conversion factors for Type F, M and S are listed in Table 3.8.

Table 3.8: Dose conversion factors for inhalation (taken from [IC97]).

Uranium isotope	**DCF (Sv/Bq uptake)**		
	Type F	**Type M**	**Type S**
^{234}U	6.4×10^{-7}	2.1×10^{-6}	6.8×10^{-6}
^{235}U	6.0×10^{-7}	1.8×10^{-6}	6.1×10^{-6}
^{238}U	5.8×10^{-7}	1.6×10^{-6}	5.7×10^{-6}

The physical half-life of the isotopes is significantly longer than a typical monitoring period (see Table 2.1). Thus, the physical half-life does not need to be considered when deciding on a monitoring frequency. The biological half-life and subsequent effective half-life is important and is discussed in Section 3.3.3. Due to the long half-lives, the uranium daughter nuclides do not build-up in the body and subsequently do not pose a significant radiological risk. The internal dose contribution of the daughters is less than 10%. For radionuclides that decay into stable progeny, or into progeny with radioactive half-lives very much longer than the human lifespan, only the radiation emitted by the parent radionuclide needs to be considered for dosimetry. The IMBA software has various mathematical solutions to take this into consideration [BI94].

As mentioned, the three major uranium isotopes decay mainly by emitting alpha and β^- particles. As such, uranium is monitored usually with *in-vitro* techniques

such as urine and faecal analysis. Lung counting is also done, using the 186 keV photon emitted by ^{235}U. This is discussed further in Section 3.3.1.

3.1.3.2 The effect of mass enrichment on committed effective dose

Most of the mass of uranium consists of the isotopes ^{234}U, ^{235}U and ^{238}U in various mass combinations depending on the enrichment process. The mass distribution between the different isotopes for various enrichment grades for the Pelindaba enrichment process is listed in Table 3.9. One should take note that the expression "enrichment grade", indicates the mass enrichment of the ^{235}U isotope since ^{235}U is the fissionable isotope of importance during fuel production.

Table 3.9: Uranium mass distribution shown as a percentage at different enrichment grades for the Pelindaba enrichment process.

^{235}U	^{234}U	^{238}U
0.72%	0.0055%	99.275%
1%	0.0062%	98.994%
2%	0.0127%	97.990%
5%	0.0318%	94.970%
10%	0.0644%	89.940%
20%	0.132%	79.870%
30%	0.208%	69.790%
40%	0.299%	59.700%
50%	0.415%	49.590%
60%	0.571%	39.430%
70%	0.769%	29.230%
80%	1.050%	18.950%
93%	1.518%	5.482%

The effect of enrichment on the CED for a unit intake is shown in Table 3.10. As can be seen, enrichment has a minimal effect on the CED since the dose

conversion factors are almost the same for the different uranium isotopes (see also Table 3.8). The ^{234}U isotope is the largest contributor to the dose since most of the activity in the uranium is due to ^{234}U. This is discussed in the next section.

Table 3.10: CED per enrichment normalised to 5% enrichment.

Mass enrichment - ^{235}U	**CED normalised** [a])
5%	1
20%	1.02
90%	1.03

[a]) Calculations were done with IMBA assuming:

- Chronic intake over 28 days,
- ICRP default values and
- 1 Bq/day intake.

The Specific Activity (SA), of the different uranium isotopes is listed in Table 2.1 (page 11). The SA for ^{234}U is significantly larger than for the other isotopes. The total activity in the uranium mix is thus mostly due to ^{234}U; subsequently the CED will mostly depend on the ^{234}U mass content in the mixture. The activity contribution of ^{234}U is significantly lower at enrichments less than 2%.

The SA of a mixture is calculated from an assumed ^{235}U Mass Enrichment (ME) distribution. The SA of the compound (mixture) is the sum of the Weighted Specific Activity (WSA) for each nuclide. The WSA is a specific nuclide's contribution to the total activity of the mixture and is calculated as follows:

$$SA_{compound} = \sum_{i_U} WSA_{i_U} \tag{3.1}$$

$$WSA_{i_U} = SA_{i_U} \times ME_{i_U} \quad , \tag{3.2}$$

where

$SA_{compound}$ = specific activity of the mixture for a specific ^{235}U ME

SA = specific activity for each nuclide

WSA = weighted specific for each nuclide

ME = mass enrichment for each nuclide

i = uranium isotopes, ^{234}U, ^{235}U and ^{238}U respectively.

T

he weighted specific activity for ^{234}U, the % contribution of ^{234}U to the total activity and the change in the SA for various enrichment grades can be seen in Table 3.11. As expected, the SA changes significantly due to the change in ^{234}U mass content in a given mixture.

Table 3.11: Specific Activity of a mixture per enrichment.

Mass enrichment: (^{235}U mass contribution)	**^{234}U WSA (Bq/g)**	**SA of mixture (Bq/g)**	**^{234}U activity contribution**
0.72%	1.27×10^4	2.56×10^4	50%
1%	1.43×10^4	2.74×10^4	52%
2%	2.93×10^4	4.31×10^4	68%
5%	7.35×10^4	8.92×10^4	82%
10%	1.49×10^5	1.68×10^5	89%
20%	3.05×10^5	3.31×10^5	92%
30%	4.80×10^5	5.13×10^5	94%
40%	6.91×10^5	7.30×10^5	95%
60%	1.32×10^5	1.37×10^6	96%
70%	1.78×10^5	1.84×10^6	97%
80%	2.43×10^6	2.49×10^6	97%
93%	3.51×10^5	3.58×10^6	98%

3.1.3.3 Determination of ^{234}U activity

As can be seen above, the isotope of importance for internal dose is ^{234}U. Most monitoring techniques can only measure the ^{235}U activity. The total activity in a given uranium mix (and specifically the ^{234}U activity) is usually derived from the measured ^{235}U activity using specific activity and mass distribution [RU98]. This

implies that the mass distribution (enrichment) should be known during dose calculations. The exact enrichment is usually not known since workers can be exposed to different enrichments during different plate productions or during the initial feed batch production when various different enrichments are blended together. Errors in deriving the ^{234}U activity and subsequent total activity can thus lead to significant errors in the CED [FI94]. The ^{234}U isotope is the predominant contributor to SA of the uranium mixture and subsequently the predominant isotope for internal dosimetry [AN07].

Assuming that a unit activity (1 Bq/day ^{235}U for urine and for lungs a 1 Bq ^{235}U) is measured then the resultant total activity of uranium in the measurement is shown in Fig. 3.5 and Table 3.12. The total activity is directly proportional to the calculated CED. As can be seen, the total activity decreases sharply for low enrichments where after it increases as the enrichment increases. The total activity is calculated as follows:

$$\text{Total Activity} = \frac{1\ \text{Bq}^{235}\text{U}}{\text{SA}_{235_\text{U}}} \Big/ \text{ME}_{235_\text{U}} \times \text{SA}_{\text{compound}} \quad , \tag{3.3}$$

Workers at Necsa's uranium plant can be exposed to a range of enriched uranium products (mostly between 5% and 50%). The specific enrichments that are used are not mentioned here due to confidentiality reasons. An uncertainty in enrichment can lead to significant under or over estimation of the total activity, as shown in Table 3.12. It is recommended that for routine monitoring a default enrichment of 30% is assumed for all exposures. This implies a maximum underestimation of 2% (0.98 at 10% ME) and maximum overestimation of 17% (1.17 at 50% ME). For enrichments larger than 50% and less than 5% the under- and over-estimation is significantly larger. Should it be that exposure is expected at these extremes, i.e. low enrichment (depleted or natural uranium) or high enrichment, it is recommended that the actual enrichment should be used. Using the actual enrichment is also recommended if an unexpected intake has occurred or if the calculated CED exceeds a predetermined dose.

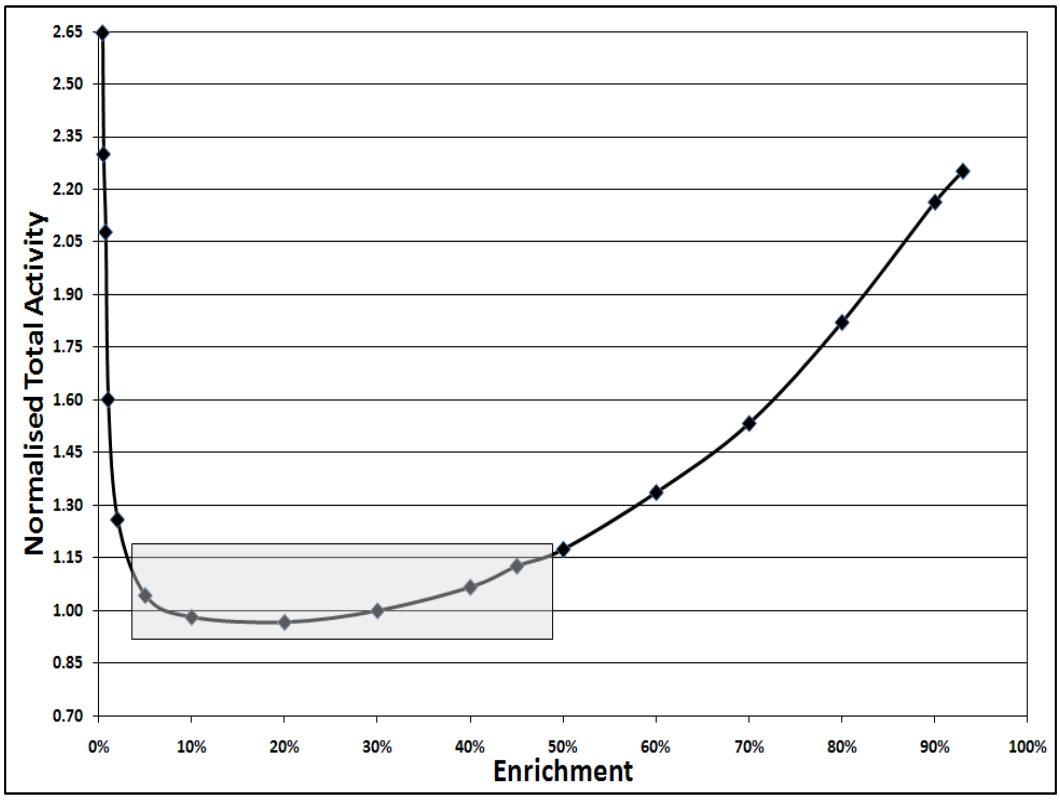

Figure 3.5: Total activity for different MEs per unit ^{235}U measured.

Table 3.12: Total activity for different MEs per unit ^{235}U measured.

ME ^{235}U	Total uranium activity normalised [a])
0.72%	2.08
2%	1.26
5%	1.04
10%	0.98
20%	0.97
30%	1.00
40%	1.07
50%	1.17
60%	1.34
70%	1.53
93%	2.25

[a]) Normalised to 30% enrichment.

3.1.3.4 Variances in mass enrichment

Matters are further complicated since the mass contribution ratio between the different uranium isotopes could also differ from those stated in Table 3.9. The values in Table 3.9 are specifically for the historic enrichment process used by Necsa. Since the ^{234}U activity is derived from the mass distribution, it adds further uncertainty and inaccuracy to the dose calculation. A plant could produce various blends of uranium isotopes (due to up or down blending) that results in a product having a SA that could differ from the predicted calculations [AN07]. The $^{235}U/^{234}U$ mass ratio is enrichment process specific and can differ significantly between enrichment processes [FE04]. Literature review indicates average mass ratios that differ from each other. Average mass ratios of 112 and 105 are cited in Ref. [FE04]. The average for the Necsa enrichment process (between 5% and 50%) is 141, as can be seen Table 3.13. This differs significantly from reported values. In recent times Necsa purchased enriched uranium for which the mass ratios differ from those of Necsa. Purchased uranium forms currently a small portion of the uranium being processed by Necsa, but this contribution can increase in the near future and a more representative mass distribution should be obtained.

Table 3.13: $^{234}U/^{235}U$ mass ratio for different MEs.

ME ^{235}U	**$^{234}U/^{235}U$ Ratio**
5%	157
10%	155
20%	152
30%	144
40%	134
45%	126
50%	120
Average	**141**
Standard Deviation	**15**

3.2 Exposure scenarios

The exposure scenario provides information that will aid in determining the monitoring programme and in calculation of CED and can be characterised according to:

- sources of intake (spatial),
- routes of intake (pathway) and
- time of intake (temporal).

Each of the contributing factors to the exposure scenario is discussed in detail below together with discussion of the software modelling of these scenarios.

3.2.1 Sources of intake (spatial)

A range of uranium physical forms and compounds, with different physical and chemical properties and behaviour, are used and produced at Necsa's uranium plant. Plates are manufactured to be used as fuel plates for the research reactor or as target plates to be used for isotope production. Scrap originating from various processes (reclamation of uranium waste) within the plant or U_3O_8 is used as feeding material to produce UF_4 and subsequently uranium metal. Uranium metals are cut into shavings in a glove box with negative pressure relative to the room. The shavings are dissolved in a nitric acid solution to produce a uranyl nitrate solution ($UO_2(NO_3)_2$). The uranyl nitrate is further concentrated and purified in fume cupboards and glove boxes. Ammonium di-urinate (ADU, $UO_3.H_2O.NH_3$) is produced during an ammonia precipitation stage.

At this stage all of the processes are wet chemical processes, except for uranium metal shavings. The probability of airborne contamination is thus the highest during the metal shavings stage. The ADU is placed in an oven for fluorination. The ovens are enclosed in a perspex box at a negative pressure relative to the room. The ADU is converted to UO_3 during heating and are then reduced to UO_2. Hydrogen fluoride (HF) is added and UF_4 is formed. The UF_4 is milled and magnesium oxide is added were after it is placed in an oven and heated to form UO_2. The UO_2 and aluminium is either melted separately or together to form pure

uranium ingots or a uranium alloy (UAl_3). This is a well known process and the same processes are described by [RU01] and [EI94].

Workplace surveillance has indicated that the areas surrounding the ovens are high airborne contamination areas and pose a high risk for intakes. Exposure to workers is minimised and controlled primarily by engineered methods, i.e. containment (glove box), fume hoods and ventilation. Additional respiratory protective equipment is prescribed in instances where containment is breached or procedures include working with furnaces (ovens). Contamination control (housekeeping) is also strictly enforced. Workers routinely work in all of the areas except for the uranium metal melting-area and the area where uranium metal scraps and shavings are produced. Work in these areas is on an *ad-hoc* basis and is controlled by task monitoring.

3.2.2 Routes of intake (pathway)

The most frequent route of uranium intake for workers is by inhalation according to Refs. [FI94] and [DO04]. For routine monitoring it is assumed that the intake is only inhalation and none is ingested directly through the mouth. Inhaled particles are deposited throughout the respiratory tract and a small fraction of those deposited is swallowed and excreted through the GI tract. The ratio of excretion between urine and faeces as well as lung counts can be used as indicator of the solubility class of the uranium and as an indicator of the route of intake. This is discussed in Section 3.1.2 and Section 3.3.1.4.

A portion of the uranium is ingested directly through the mouth during routine exposures and the assumption of only inhalation can lead to potential errors in the CED. Ingestion can happen due to poor occupational hygiene, e.g. workers not washing their hands and subsequently contamination is ingested during smoking or eating and incidents have been found where a worker contaminated a writing pen and placed it into the mouth.

A study by Lee [LE10] found that the exposure pathway (see Section 2.2.2) plays an important role and has a significant effect on the CED for intakes of ^{241}Am. For uranium intakes the effect is just as significant. This is substantiated in Table 3.14 indicating the variation in CED based on the fraction inhaled *versus* fraction ingested.

Table 3.14: Normalised CED - inhalation *versus* ingestion, Type M and S.

Route of Intake (inhalation : ingestion)	**CED normalised to only inhalation** [a])	
	Type S	**Type M**
1:0	1.00	1.00
3:1	0.75	0.76
2:1	0.67	0.68
1:1	0.50	0.53
1:2	0.33	0.37
1:3	0.25	0.29
0:1	0.00	0.05

[a]) Calculations were done with IMBA assuming:

- chronic intake over 30 days,
- ICRP default values,
- low enrichment (IMBA values) and
- 1 Bq/day measured in urine sample on day 30.

As can be seen, the impact on CED is significant, but a significant fraction of ingestion is highly unlikely during routine exposures. An increasing fraction of ingestion leads to lesser doses and as such an assumption of only inhalation will be a conservative (maximum) value and is acceptable for routine urine monitoring. It is advisable to take ingestion into consideration if a predetermined investigation level (e.g. 1 mSv) has been exceeded, if ingestion is highly likely or suspected or if there was an incident (acute intake).

The route of intake should also be considered for routine monitoring that is based only on faecal analysis or lung counting [LE10]. An AMAD value different from the ICRP default value will affect the fraction ingested from the respiratory tract. This effect is less severe for urine monitoring (see Section 3.1.1). Again, as such, the route of intake does not need to be considered for routine urine monitoring except in the cases mentioned above. However, it should be noted that faecal analysis can also aid in the determination of the route of entry (see Section 3.3.1).

3.2.3 Time of intake (temporal)

Due to the routine nature of the work done in a uranium plant, where there is seldom *ad-hoc* tasks for the majority of the workers, one can assume that exposures will be distributed evenly over the monitoring period. Workplace surveillance will indicate significant increases in airborne and surface contamination levels, thus indicating a possibility of an acute intake or an uneven distribution of airborne contamination of a monitoring period. A review of workplace surveillance data has confirmed this.

However, this is not true for maintenance workers who perform *ad-hoc* tasks and significant variances in airborne levels were found when containment had been breached. During the majority of maintenance operations intakes are not expected since tasks are well controlled and the necessary radiological protection equipment is worn. Maintenance workers are monitored routinely (monthly urine samples). Experience has shown that unexpected intakes can usually be traced back to an *ad-hoc* tasks and time of intake could have a significant impact on the actual dose. In some instances are pre- and post-task monitoring prescribed, consisting of lung counting and/or urine sampling which is time consuming and costly. As an alternative, nose blows or nasal swabs are used as screening tests. Nose blows are less time consuming, cost less and give an instant indication of a very recent intake i.e. during the task (see Refs. [DO99] and [DO04]).

Dose conversion factors for routine exposures are based on the assumption of an acute intake in the middle of the monitoring period [IC97]. Newer versions of

dose calculation software, such as IMBA, make it possible to simulate chronic exposures and have a more representative model of actual exposure scenarios. Previous software used by Necsa (LUDEP) was not capable of simulating chronic intakes [NR96]. Simulating chronic intakes rather than using the ICRP default of an acute intake is preferred and is thus consistent with the best estimate approach of IDEAS. The CED for an acute intake *versus* a chronic intake is compared and the effect on the calculated CED is shown in Table 3.15.

Table 3.15: Chronic *versus* acute normalised to chronic intake.

Monitoring period (Days)	**Acute CED / Chronic CED** [a])	
	Type M	**Type S**
12	1.05	1.06
28	1.14	1.16
58	1.28	1.30
88	1.34	1.36

[a]) Calculations were done with IMBA assuming:

- acute intake on day 6, 14, 29 and day 44,
- chronic intake over 12, 28, 58 and 88 days,
- ICRP default values,
- low enrichment (IMBA values) and
- 1 Bq/day measured in urine sample on day 14, 30, 60 and 90.

In all instances, the CED for an acute intake is larger than the CED for a chronic intake over the same period. The difference increases as the length of the monitoring period increases and is significant for monitoring periods larger than a month (> 16%). These values are consistent with what was found by Birchall [BI06]. Assuming a chronic intake is acute leads to an overestimation between 5% to 40% depending on the monitoring period. Newer versions of software for calculating CEDs are more capable of simulating exposure scenarios and thus it is recommended that the chronic option should be used for routine exposure calculations as suggested by Refs. [BI06] and [DO08]. Chronic intake can also be assumed for maintenance workers but should be used with caution.

3.2.4 Software modelling of exposure scenarios

The latest commercial internal dose calculating software available enables simulating exposure scenarios much more representative of the actual scenario, especially simulating different routes of intake and various times of intake combinations. Software for calculating CED was reviewed in Ref. [AN03]. Criteria used were, amongst others, practical use of codes, graphical capabilities, parameters available for change, data fitting, choice of biokinetic models etc. Necsa recently purchased IMBA (Integrated Modules for Bioassay Analysis) which is developed by the NRPB of the United Kingdom. IMBA was found in Ref. [AN03] to be a comprehensive code which uses the latest ICRP biokinetic and dosimetric modules. A CED was calculated for two reference cases and results were found to be homogeneous and accurate. The IMBA code is highly flexible and includes the modelling combination of intake pathways and various exposure scenarios. It also allows for variability in several of the biokinetic and physiochemical parameters. Using IMBA, by a relative expert user, will thus further ensure that results are the best estimate (harmonised and accurate) and proportional (graded approach).

3.3 Individual monitoring

Existing regulatory requirements are sometimes difficult to meet due to technical challenges related to speed, accuracy, detection limits, exposure scenario uncertainties and cost implications. These are the challenges faced by the current bioassay techniques [SC03]. The features of different bioassay techniques and their limitations including monitoring frequency and technique sensitivity are each dealt with in turn below.

3.3.1 Bioassay techniques

A survey done under the auspices of the OMINEX project found different preferences for routine monitoring of uranium [RA03]. Subsequently, the OMINEX project investigated various bioassay monitoring programmes [NR04] and recommended the programme given in Table 3.16. The programme

recommended by the DOE site at Hanford is given in Table 3.17 [CA09]. The various different techniques are detailed below.

Table 3.16: OMINEX monitoring programme for uranium [NR04].

Compound	**Type F**	**Type M**	**Type S**
Monitoring	Urine	Urine and Faeces	Lung and Faeces

Table 3.17: Hanford monitoring programme for uranium [CA09].

Compound	**Type F: Chronic exposure**	**Type F: Infrequent or acute exposure**	**Type M and S: Chronic exposures**
Monitoring	Biweekly or monthly urine samples	Quarterly monitoring or post task monitoring for short duration work.	Quarterly urine samples and annual or semi-annual lung counts.

3.3.1.1 Urine analysis

Urine monitoring is in most cases not interpreted quantitatively, but rather used as confirmation of satisfactory conditions [IC97]. This is also the case for Necsa and only intakes exceeding a prescribed action level are converted to a CED. Necsa currently use a technique called Delayed Neutron Counting (DNC) for monthly analysis of uranium in urine for chronic exposures to Types M and S. Quarterly confirmatory DNC analysis is done for infrequent and low exposures of Type M uranium. DNC is a neutron activation analysis technique that determines the ^{235}U activity based on delayed neutron activation after irradiation. Delayed neutrons are produced during fission by thermal neutrons. The number of delayed neutrons can be related back to the original ^{235}U activity. This technique is very sensitive (see Section 3.3.3) but requires an irradiation facility. The samples are irradiated at the Necsa SAFARI-1 research reactor. This implies turnaround times for results up to two weeks.

Type F uranium is monitored quarterly with fluorometry to ensure chemical toxicity levels are not exceeded. Fluorometry is based upon the fluorescence of the uranium compound when exposed to ultraviolet light. The intensity is proportional to the uranium concentration. Fluorometry is significantly faster than DNC but its sensitivity is at least an order of 100 less sensitive, which is inadequate for monitoring of Type M and S uranium. Exposed workers at the uranium plant are monitored monthly with DNC. All samples are singe voids and 24 hour samples are only given in instances of unexpected intakes.

Literature study, Ref. [HU03] and [SC07], indicated that alpha spectrometry is the technique mostly used for the determination of alpha emitters in urine (such as uranium), but the main disadvantage is the long counting time required (up to several days). Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) is not often used, but the technique has shown to be as sensitive as alpha spectrometry and modified newer ICP-MS techniques have even much lower sensitivities [OE07]. ICP-MS's main advantage is its short analysis time (several minutes), but the equipment is expensive. ICP-MS will be the preferred technique used in future according to Refs. [SC03] and [SC07].

Another technique (other than DNC) with better sensitivity is not necessary for Type M monitoring, but better sensitivity is necessary for Type S monitoring (as will be seen in Section 3.3.3). However, the required sensitivity levels for Type S are within the order of dietary intakes (see Section 3.3.2). Thus, it problematic to distinguish between dietary intake and occupational intakes at levels close to the required sensitivity levels. No immediate improvements thus are necessary for Necsa's uranium urine programme, but techniques with faster turnaround time and lower sensitivity would be advantageous.

3.3.1.2 Lung counting

Lung counting at Necsa is done annually for all workers exposed to all forms of uranium. The counting time is 30 minutes. Longer counting times (60 minutes) are used during unexpected intakes. The newly purchased Whole Body Lung

Counter (WBLC) from Canberra consists of 4 Broad Energy Germanium (BEGE) detectors for lung counting and a Sodium Iodide (NaI) detector for whole body scanning. It is also housed (shielded) in a low background steel chamber thus decreasing background levels significantly, thereby improving signal to noise ratio and subsequently also the detection capability. The previous counter set-up consisted of a single NaI(Tl) detector. The new WBLC was also supplied with the latest analysis software (GENIE 2000^{TM} and ABACOS^{TM}) allowing efficient analysis of spectra and data. The BEGE detectors provide better resolution than the NaI detector at lower energy ranges, particularly for the ^{235}U 185 keV peak. Sensitivity has also improved from the previous set-up.

Taking into consideration the improved sensitivity, lung counting on its own is still not adequate for routine uranium monitoring, due to poor sensitivity (see Section 3.3.3). It is only complementary to urine monitoring and useful for detecting significant intakes, especially Type S. Lung counting is an indication of accumulated exposures over longer periods (6 or 12 months), thus is useful to monitor for long term build-up, especially for highly insoluble compounds. Previous chronic (up to several months) intakes should thus be taken into account when evaluating lung counts. Carbaugh [CA03] has recommended lung monitoring for plutonium and is equally true for uranium [FI94]. Urine analysis is less reliable for detecting significant Type S intakes and lung counting provides more reliable results [TA03]. This is due to the biokinetic behaviour of Type S uranium, staying longer in lungs. Lung counting is more reliable but not necessary more sensitive as seen in Section 3.3.3.

To improve the sensitivity, it is suggested summing lung counts over a group of individuals in cases where chronic exposure to Type S is expected [TA03]. It entails counting a group of individuals and the summing of all of the spectra. This will lead to an improvement in the detection capability and an average CED can be calculated and allocated to each individual. However, this is limited to a group of individuals who have had similar exposure scenarios and who were exposed to the same single uranium compound i.e. same physiochemical properties and

chronic exposure. This type of exposure scenario is unlikely for a uranium plant at Necsa and would thus not be applicable for the present study. But it can be used in unexpected intakes where more than one individual was involved.

The current lung counting technique at Necsa is optimised and the best technology available is used. It compares well with sensitivities of other systems published and no improvements are thought to be necessary.

3.3.1.3 Faecal analysis

Faecal monitoring is invasive (inconvenient) and there are usually resistance against it from workers. Samples are also cumbersome to collect. Alpha spectrometry is the preferred method for analysis of faeces and minimum detected activity of 1 mBq/day is achievable [HU03]. Faecal monitoring is also recommended for monitoring of Type S uranium due to its lower sensitivity. A further advantage of faecal monitoring is its ability to aid in the differentiation between intakes of Type M and S (see later in this section), an indication of route of intake and can also be used as indication of the AMAD (see Section 3.1.1).

Another advantage of faecal monitoring is in the determination of the route of intake [DO08]. Ingested and inhaled uranium will both deposit in the upper respiratory tract and will clear through the faeces within a few days. Faecal results after 10 days will only be inhaled material. These data can then be extrapolated to the first few days after intake and so indicate the inhalation component in the faecal results during that time. Subsequently, the ingestion component of the faecal results for the first few days can be calculated. Necsa is not yet capable of analysing faeces and it is currently under development.

3.3.1.4 Technique combination

As shown in Section 2.1.8, the chemical and physical form (physiochemical nature) of the uranium compound will determine its behaviour on intake and its subsequent biokinetic behaviour in the human body, which will determine the stay time within the human body and consequently the rate of excretion. The

difference in biokinetic behaviour between respiratory tract (lungs) and gastrointestinal tract (faeces) and between kidneys (urine) and gastrointestinal tract (faeces) will result in different stay times and excretion rates. This can be used to differentiate between intakes of different solubility classes and intake pathways. Uranium can be placed into three categories [EI94], the first being those compounds (Type F) that dissolve rapidly in the lungs and are thus excreted almost instantaneously in the urine with no retention in the lungs and subsequently very low excretion in the faeces. The second category are compounds (Type M) with intermediate dissolution, such that both mechanical clearance via the gastrointestinal tract and dissolution are dominant with a certain retention in the lungs. Subsequently, uranium will be detected in the lungs, urine and faeces. The third category of compounds (Type S) dissolves slowly with significant retention in the lungs, resulting in predominantly mechanical clearance and subsequent high excretion in the faeces. Thus, these techniques can provide valuable information for dose reconstruction. The differences can be seen in Fig. 3.6, Fig. 3.7 and Fig. 3.8.

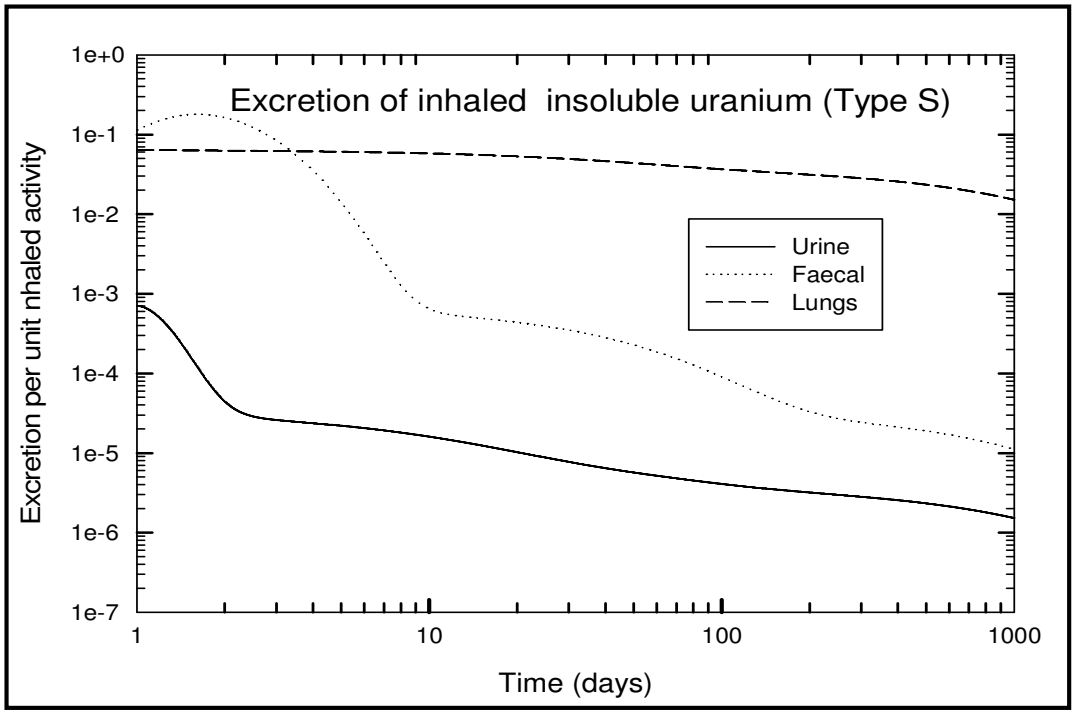

Figure 3.6: Difference in excretion rate of Type S uranium urine, faeces and lungs (taken from [IC97]).

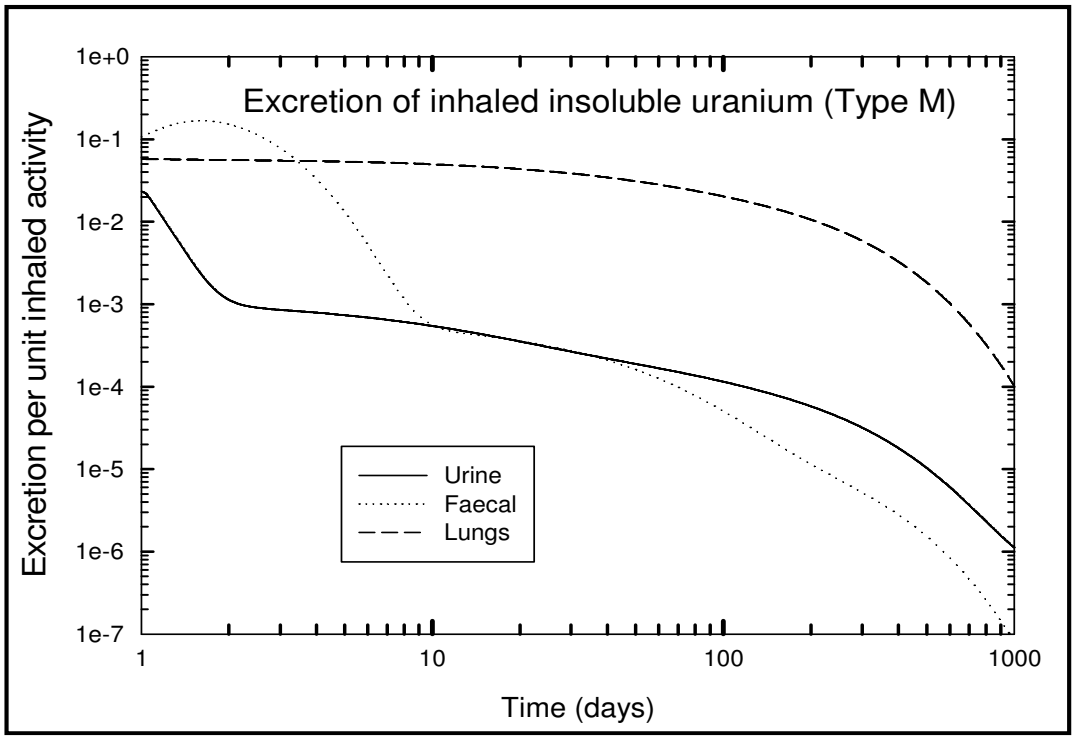

Figure 3.7: Difference in excretion rate of Type M uranium in urine, faeces and lungs (taken from [IC97]).

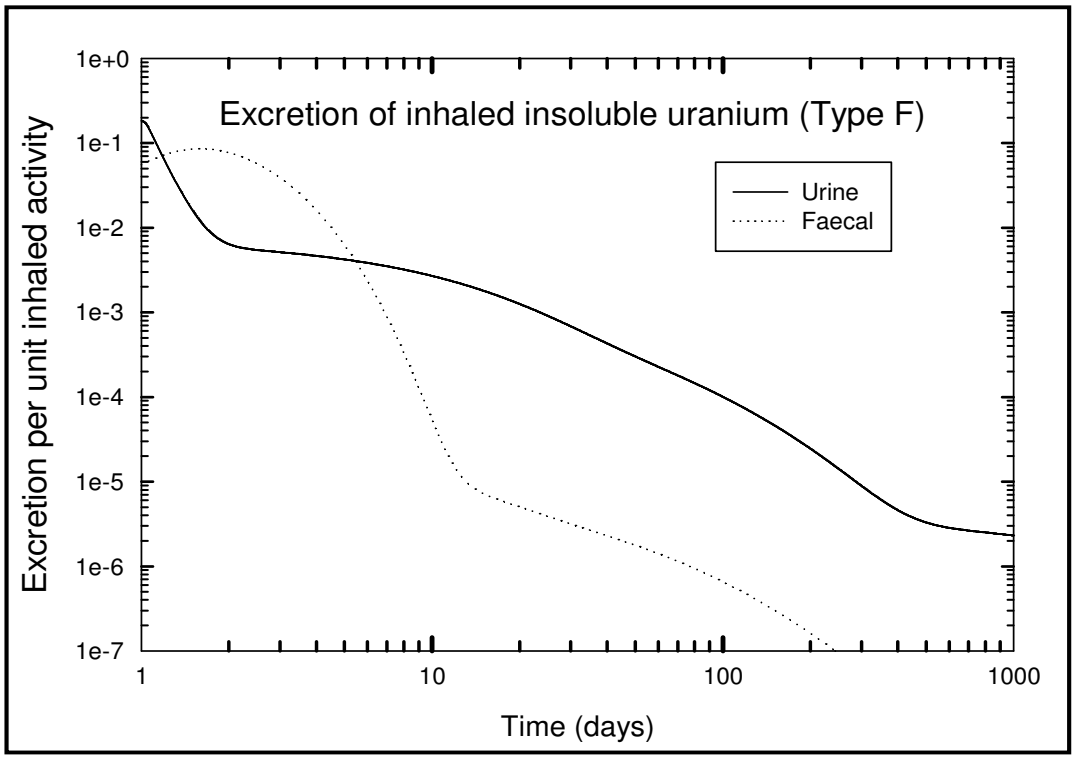

Figure 3.8: Difference in excretion rate of Type F uranium in urine and faeces (taken from [IC97]).

Type F uranium is highly soluble and it is almost instantaneously absorbed in the lungs and subsequently will not be detected. After the first few days (> 10 days) of intake (Type M), the excretion in the urine and faeces will be almost similar where after (> 40 days) the excretion rate in the faeces will be less than in the urine. This is in contrast to Type S uranium where the faeces excretion will always be more than the urine. The ratio between the lung counts and urine results will also be significantly higher for Type S uranium, compared to Type M. It is not within the scope of the present study to calculate urine *versus* faeces *versus* lung ratios but is recommended that it be calculated and used for unexpected intakes.

The above deliberation infers that the assessment of doses from intakes of uranium requires a combination of techniques in order to provide the best estimate. Workers can be exposed to a mixture of uranium compounds with different routes of intake. A combination of urine, lung and faecal analysis has shown to be the best methodology for assessing doses in such situations.

3.3.1.5 Other

Additional bioassay techniques such are hair and nail analysis are recommended [KA01] for better understanding and better estimation of exposures to uranium compounds. This is due to the uncertainties associated with single void urine samples (see Section 3.3.2) and is recommended for special monitoring and not routine monitoring. Hair and nails have the benefit of retaining the uranium for weeks or even months after an exposure and can also provide a time profile of the exposure. As the hair grows it will keep record of the exposure at a specific time.

Nasal swabs provide a quick and easy screening method to indicate unexpected intakes. Nose blows or nasal swabs should be taken as a screening test (see Refs. [DO99] and [DO04]). Nose blows are less time consuming, cost less and give an instant indication of a very recent intake (i.e. during the task). Nasal swabs should be done immediately after the task but swabs may be negative if a worker

is predominately a mouth breather [MA06]. Nasal swabs may revert to background levels within 30 min to 60 min after intake.

An individual monitoring programme, which includes personal air sampling and workplace air sampling, is recommended in order to ensure that intakes below the detection levels of the bioassay techniques (urine and lungs) are not missed. It is specifically recommended for Type S exposures. This type of monitoring is used to trigger additional bioassay monitoring in cases of abnormal levels of airborne contamination and also for dose calculations.

3.3.2 Specific limitations and/or uncertainties for urine analysis

Biokinetic models use urine excretion rates relative to a single day (24 hours). However, collection of such samples on a routine basis is often not convenient. A 24 hour sample implies that a person has to urinate in a collection bottle and this should be done for 24 hours (e.g. at work, at home, when they go shopping etc.). This can lead to missed voids or an incomplete 24 hour sample. Collection of a single void sample is easy and compliance is generally high [KA01]. Subsequently, most routine monitoring protocols use a single void which are normalised to a 24 hour sample. The first section below discusses the two sampling methods and makes recommendation on when to use which method. The second section will describe the different normalisation techniques. Dietary intake of uranium contributes to more uncertainty in the evaluation of urine and the effect thereof will also be discussed [MC99].

3.3.2.1 Singe void sample versus 24 hour sample

A 24 hour sample on two or three consecutive days is recommended to avoid large uncertainties in the quantification of daily urinary excretion values [OE07]. Excreted radionuclides may vary during the day and also varies from day to day due to variation in biological functioning of the human body. The volume excreted per day is also highly individual, being age and gender dependant [OE07]. A single void is acceptable for routine monitoring, but it is thus

recommended that 24 hour sample should be done in instances where more accurate and representative data are necessary.

Currently, Necsa's routine and task urine sampling programme consists of a single void sample. Only during special monitoring (investigations) are 24 hour samples taken. These samples are not necessarily taken in consecutive days. This author recommends continuing with a single void sample for routine and task monitoring. Continuous 24 hour sampling is also recommended for special monitoring, i.e. in instances where a predefined reference level has been exceeded or in case of an unexpected intake. This could be if a screening test such as a nasal swab indicates an unexpected intake or if workplace monitoring indicates abnormal levels of airborne contamination. A 24 hour sample should be taken for three consecutive days every week. This should be done until enough information is available for dose calculation or until excreted levels have declined below the detection limits. The above recommendations are in line with a "Graded Approach" where the effort (for gathering enough and accurate data) should commensurate with the risk.

Another option besides true 24 hour sampling is a simulated 24 hour sample. Simulated 24 hour sampling consists of collecting samples just before retiring at night and all samples until and including the first void after rising in the morning on two successive days [MC99]. This will approximate the total volume for 24 hour sampling. To ensure consistency and not to add any uncertainty regarding methodology, the author recommends using just one method for 24 hour collection, i.e. true 24 hour sampling.

3.3.2.2 Daily excretion rate normalisation

As mentioned before, routine urine samples are a single void sample. The activity concentration within this single void sample needs to be converted to total activity excreted per day in order to determine the CED. Normalisation includes use of a reference man excretion rates, creatinine content in the urine or specific gravity.

Necsa uses 1500 mℓ/day as the standard daily urinary excretion volume, irrespective of gender. The conversion is depicted by the following equation:

$$\frac{\text{activity}}{\text{day}}\left(\frac{\text{Bq}}{\text{day}}\right) = \text{activity concentration}\left(\frac{\text{Bq}}{\text{m}\ell}\right) \times 1.5\left(\frac{\text{m}\ell}{\text{Day}}\right) \quad . \tag{3.4}$$

ICRP recommends a conversion factor of 1600 mℓ/day for an adult male and 1200 mℓ/day for an adult female [IC02]. Oeh *et al.* [OE07] have found mean daily urinary excretion values of 1684 ± 719 mℓ/day for males and 1682 ± 823 mℓ/day for females. The value for males compares well with ICRP recommended values, but differs significantly for females. Other literature could not be found to confirm these values. This confirms the high variability in excretion rates and it supports the recommendation in the previous section for 24 hour sampling. A 24 hour sampling protocol will negate the need for normalisation. The relative deviation from the ICRP values (shown in brackets) is given in Table 3.18.

Table 3.18: Urine daily excretion rate comparison normalised to ICRP.

ICRP (mℓ/day)		**Necsa (mℓ/day)**	**Oeh *et al.* [OE07]**
Male:	1600	1500 (0.94)	1684 (1.05)
Female:	1200	1500 (1.25)	1682 (1.40)

The effect of the Necsa's values (see Table 3.18) is a 25% (1.25) overestimation in the calculated daily excretion for females and a 6% (0.94) underestimation for males with subsequently the same effect on the CEDs.

The current 1500 mℓ /day is still recommended for routine CED calculations below the predetermined reference level. This will simplify calculations for routine exposures and the underestimation for males and over estimation for females is still within acceptable criteria (see Section 2.3.3) and also provide a more accurate estimation of CED for higher doses. It is this author's recommendation to rather use the recommended conversion factors from ICRP in

the case where a predetermined reference level is exceeded or for unexpected intakes. It will thus ensure that Necsa's approach is "Harmonised and Accurate" and in line with the "Graded Approach" principle.

Normalisation to creatinine levels which are much more stable between sexes, ages etc is should also be considered [MC99]. Discussion with Necsa's radioanalytical laboratory pointed to some disadvantages of creatinine normalisation. Determining creatinine levels involves analysis by off-site laboratories and as such is cumbersome and expensive. Specific gravity analysis could be done by Necsa's analytical laboratories and this needs further study and investigation.

3.3.2.3 Dietary intake of uranium

Dietary intake of uranium contributes to more uncertainty in the evaluation of urine samples and the reliance on single void samples [MC99]. There could be significant levels of uranium in urine due to dietary intakes. Several studies have been done leading to significant variation in excretion rates which is dependent on intake foodstuffs and drinking water [MU07]. It should also be noted that these values can vary significantly between individuals and geographical locations. ICRP states values between 0.02 µg and 0.5 µgU/day. This is consistent with studies done at the DOE's Hanford site [MA06]. Average levels of 0.2 µgU/day and levels up to 0.6 µgU/day were reported. It is recommended that Necsa should not correct for dietary intake, i.e. subtract it from urine results but rather determine a dietary intake range for Necsa workers and take it into consideration during intake evaluations. It is recommended that urine values, below dietary level, are recorded, but not interpreted quantitatively.

3.3.3 Monitoring frequency and sensitivity

The two factors to consider are the recommended maximum underestimation and the maximum missed dose set by the ICRP (see Section 2.3.3). The ICRP [IC97] limits the underestimation of the calculated CED to a factor 3, compared to the default assumption of an intake in the middle of the monitoring period. By using

Eq. (2.11) the required sensitivity (decision level, L_C) can be calculated as based on the missed dose. This is shown in Table 3.19.

Table 3.19: Required sensitivity for various monitoring periods

Monitoring period (days)	L_C **(mSv)**
14	0.04
30	0.08
90	0.25
180	0.50
365	1.00

3.3.3.1 Urine analysis

Delayed Neutron Counting (DNC) is currently used by Necsa to determine uranium concentration in urine. The detection capability (MDA) is 0.72 ng ^{235}U/ℓ. The MDA as total uranium mass and as total uranium activity is calculated as follows:

$$Y \text{ (ng U/l)} = X \text{ (ng } ^{235}\text{U/l)} / \text{ ME}_{235_U} \quad , \tag{3.5}$$

where

Y = MDA (total uranium mass)

X = ^{235}U MDA

ME_{235_U} = mass enrichment for the ^{235}U nuclide.

$$Z \text{ (Bq U/l)} = \text{Y (ng U/l) x SA}_{\text{compound}} \tag{3.6}$$

where

Z = MDA (total uranium activity)

$\text{SA}_{\text{compound}}$ = specific activity of the mixture for the specific ^{235}U ME.

Substituting the specific activity of 5.1 x 10^5 Bq/g (ME = 30%, see Table 3.11), it equates to an MDA of 1.2 mBq/ℓ (100 ng total U/ℓ). Using the reference male excretion rate of 1.6 ℓ/day (Table 3.18), it is equal to 2 mBq/day. Mass enrichment of 30% was assumed as representative default enrichment (see Section 3.1.3.4). The MDA at different enrichments are also calculated and shown below. The MDA (stated as total activity) first decreases as the enrichment increases, but increase later on with increasing enrichment. This is due to the conversion from a ^{235}U mass to total activity (see section 3.1.3.3 and Table 3.12).

Table 3.20: MDA for urine analysis by DNC at different mass enrichments.

Mass enrichment: (^{235}U mass contribution)	**MDA (total U ng/ ℓ)**	**MDA (mBq/ℓ)**	**MDA (mBq/day)**
0.20%	360.0	5.3	9
0.50%	144.0	2.8	5
0.72%	100.0	2.6	4
1%	72.0	2.0	3
2%	36.0	1.6	2
5%	14.4	1.3	2
10%	7.2	1.2	2
20%	3.6	1.2	2
30%	2.4	1.2	2
40%	1.8	1.3	2
50%	1.4	1.4	2
60%	1.2	1.6	3
70%	1.0	1.9	3
80%	0.9	2.2	4
93%	0.774	2.8	4

The detection capabilities of ICP-MS are typically 1-10 ng U/ℓ [OE07], [SC07]. However, the detection capabilities can be bettered significantly (≈ 30 pg ^{235}U/ℓ) with new improved ICP-MS methods. For alpha spectrometry, detection values are in the order of 1 - 2 mBq [SC07]. The MDA of 2.4 ng U/ℓ (~ 2 mBq/ℓ) for the DNC technique compares well with values from other techniques. It was unclear from the literature whether Decision Level or MDA where quoted by Oeh *et al.* [OE07] and Schmitzer *et al.* [SC07]. The author assumed it to be MDA. It should be noted that the 2 mBq/ℓ MDA of the DNC technique is also in the same order as the 3.7 mBq/ℓ required in performance standards set by the American National Standard Institute (ANSI) [AN96]. The MDA (stated as CED) of the Necsa's DNC technique for different solubility classes and monitoring periods are given in Table 3.21.

Table 3.21: MDA of Necsa's uranium in urine analysis (mSv).

Monitoring period (days)	**Required L_C (mSv)**	**MDA (Type M) [a])** **(mSv)**	**MDA (Type S) [a])** **(mSv)**
30	0.08	0.009	1.0
90	0.25	0.015	1.7

[a]) Calculations were done with IMBA assuming:

- chronic intake over 28 and 88 days, respectively,
- ICRP default values,
- ME = ^{235}U (30%),
- MDA = 2 mBq total uranium per day and measured in urine sample on day 30 and 90.

For Type M uranium, the MDA is well below the L_C goals set in Table 3.19. It is also recommended to keep to a monitoring period of 30 days where exposure scenarios indicate a high possibility of intake. This will enable better gathering of information in case of an unexpected intake. However, for Type S, the MDAs are significantly higher than the L_C goals. Indeed, L_C values are roughly half of the MDA values (see Eq. (2.10)). Thus, the L_C for Type S will not meet the set L_C goals. As indicated in Section 3.2.1, chronic exposure to Type S uranium is unlikely. A routine monitoring programme is thus not necessary, but special and task monitoring is required for Type S exposures. Other urine analysis techniques

as indicated above with better sensitivities can also be investigated. The differentiation in excretion rates (biokinetic behaviour) between urine and faeces can be used to differentiate between intakes of different solubility classes. Thus, although urine analysis (Type S) does not have the required sensitivity, it will provide valuable information for dose reconstruction.

Assuming a worst case scenario where an acute intake occurred on the first day of a monitoring period, the underestimation for various monitoring periods for chronic intake is indicated in Table 3.22. For Type M and S, the underestimation complies with the ICRP recommended underestimation of not more than a factor of 3 (0.33) for both monitoring periods. It is thus recommended that no change is made in the monitoring periods of 90 and 30 days.

Table 3.22: Underestimation for Type M & S intakes, urine monitoring.

Monitoring period (days)	[a)]**Underestimation (Type M)**	[a)]**Underestimation (Type S)**
30	0.58	0.57
90	0.47	0.53

a) Calculations were done with IMBA assuming:

- chronic intake (over 28 and 88 days, respectively) *versus* acute intake on first day,
- measurement done on day 30 and 90 respectively
- ICRP default values and
- ME = IMBA low enrichment option (3.5%).

3.3.3.2 Lung counting

Lung counts are reported by the analysis software if it exceeds the critical level. The critical level is equivalent to the decision level (L_C). An MDA value is also calculated by the analysis software and is given in Table 3.23. The MDA is based on Eq. (2.10), corrected for counting time and attenuation by the chest wall. The MDA values are for ^{235}U activity and for counting times of 30 min and 60 min respectively. These MDAs are similar to values (LLD = 3 Bq) found in the literature [LO05] and is less than the 7.4 Bq MDA recommended by ANSI

[AN96]. A survey done by the EURADOS project reported MDA values of 3 - 5 Bq [LO07]. The corresponding CED values are given in Table 3.24.

Table 3.23: Sensitivity of Necsa's lung counter for ^{235}U.

Counting Time (min)	**MDA (^{235}U Bq)**
30	4.5
60	2.6

Table 3.24: Sensitivity of Necsa's lung counter.

Monitoring Period (Day)	**Required L_C (mSv)**	**MDA (Type M) [a])** **(mSv)**	**MDA (Type S) [a])** **(mSv)**
180 (30 min)	0.5	9.3	19.0
180 (60 min)	0.5	5.4	10.9
365 (30 min)	1.0	14.7	22.2
365 (60 min)	1.0	8.4	12.8

[a]) Calculations were done with IMBA assuming:

- chronic intake over 180 and 365 days,
- counting times 30 min and 60 min,
- MDA of 4.5 Bq and 2.6 Bq ^{235}U (30 min and 60 min, respectively),
- ICRP default values,
- ME = ^{235}U (50%) and
- measured in lungs on day 180 and day 365.

The CED values are significantly higher than the ICRP required L_C goals set in Table 3.19. A rule of thumb is that the L_C value is approximately half of the MDA value. As such, lung monitoring cannot on its own be used for routine monitoring for Type M and S uranium. It is complimentary to urine monitoring as described in Section 3.3.1.2. Increasing the monitoring period to 30 days resulted in a calculated measured MDA of ± 1.5 mSv for Type M which is still not in the same order as for urine monitoring. Such low levels required count times of at least 1 hour which is also impractical for monitoring large number of people.

A monitoring frequency of 6 months (180 days) and a count time of 30 min are recommended for routine Type M monitoring (L_C = 4.3 mSv). This is to confirm that annual CEDs are less than intervention levels which are typically set at 6 mSv to 8 mSv annual CED. Routine monitoring of Type S with only lung counting is impractical, but can still be used as an indicator of significant intakes (see Section 3.3.1.2). Improved detection capability can be attained by summing a number of spectra acquired over several consecutive days [TA03]. This is recommended in cases of unexpected intakes of Type S uranium. The differences in biokinetic behaviour between the retention in the lungs and excretion in the faeces can be used to differentiate between intakes of different solubility classes. Thus, although lung counting does not have the required sensitivity, it will provide valuable information for dose reconstruction.

Table 3.25: Underestimation for Types M and S intakes, lung counting.

Monitoring period (days)	**Underestimation** [a]) **(Type M)**	**Underestimation** [a]) **(Type S)**
90	0.64	0.78
180	0.47	0.76
365	0.25	0.76

[a]) Calculations were done with IMBA assuming:

- chronic intake (180 and 365 days) *versus* acute intake on first day,
- Measurement done on day 180 and 365,
- ICRP default values and
- ME = IMBA low enrichment option.

Assuming a worst case scenario where an acute intake occurred on the first day of a monitoring period, the underestimation for various monitoring periods assuming the intake was chronic is given in Table 3.25. For Type S, the underestimation is well within compliance of the ICRP recommended factor 3 (0.33) for all three monitoring periods. For Type M, the underestimation is acceptable for the 90 and 180 day monitoring periods, but below the criteria for the 1 year monitoring period. Since mostly exposure to Type M is expected, it is thus recommended to change the monitoring period from 365 days to 180 days. Lung counting is

complimentary to urine monitoring and a monitoring period of 90 days would not add significant value.

3.3.3.3 Faecal analysis

The detection capability for faecal monitoring is substantially lower than lung counting and urine analysis and is able to confirm Type S uranium exposures several days after intake. MDA levels of 1 mBq/day by means of alpha spectrometry are possible [HU03] and translates to a CED of 1 µSv for an acute intake 7 days prior (Type S uranium enriched to 50%).

3.3.4 Suitably qualified and experienced dosimetrist

Intercomparison studies have shown that discrepancies between results can be attributed amongst other factors to the level of expertise of the dosimetrist. Doses estimated shall be done by suitable qualified individuals using a person-specific approach. Due to assumptions made, dose estimated independently by two persons can lead to variations up to a factor of 3. This was not dealt with in the present study, but is an important factor. The USA Department of Energy recommends the following [DO07a]:

"The analysis of workplace and radiobioassay measurement data and the evaluation of internal dose involve complex evaluation and professional judgment. Personnel with responsibility for internal dose evaluation should have the necessary expertise and skill, based on appropriate education and training in conjunction with practical experience, to perform their assigned duties." "It is important that internal dosimetry specialists be capable of recognizing conditions warranting follow-up radiobioassay and dose evaluation. Personnel should be familiar with the relevant internal dosimetry literature and the recommendations of national and international scientific organizations with regard to internal dose evaluation."

The author thus recommends that a review and audit by another qualified individual is performed if the calculated dose exceeds a specific level. During the

review, the assumptions and models used to derive the radiation dose must be evaluated, with mathematical derivations and the associated uncertainty. This is in line with the requirements from project IDEAS.

3.4 Monitoring strategy and protocol

In summary, the following monitoring strategy and protocol is recommended for uranium monitoring at the uranium plan at Necsa. This is based on abovementioned analysis and literature research. Factors such as particle size, solubility, route of intake, time of intake, the required sensitivity and available bioassay techniques have been considered. In essence, only routine monitoring for Type M uranium needs to be considered. Due to the *ad-hoc* nature of Type S exposures, only task related monitoring is necessary. This is also possible since exposures to Type S uranium are unlikely due to administrative and engineering controls applicable where Type S uranium is processed. Therefore, based on a graded approach, the following monitoring programmes are recommended and detailed below.

3.4.1 Monitoring protocol (IDMP) for Type M uranium intakes

Monitoring for Type M uranium should be based on routine urine sampling. Complimentary to the urine monitoring, lung counting should also be done as confirmation of no significant intakes. However, faecal sampling is not recommended on a routine basis, but should be done in the instance of unexpected intakes. This recommendation is summarised in Table 3.26.

Table 3.26: Recommended monitoring programme for Type M uranium.

Bioassay	**Analysis technique**	**Monitoring period**
Urine analysis	DNC (Single void)	Monthly
Lung counting	Gamma spectroscopy	180 days, (30 min count time)
Faecal analysis	Alpha spectroscopy	Unexpected intakes

Based on a graded approach, the following different levels of CED calculation are recommended:

Level 0: Annual projected CED < 1 mSv or less than dietary level

- Record only urine analysis results.
- CED need not be calculated.

Level 1: Annual projected CED between 1 – 3 mSv

- Uncomplicated CED calculation based on ICRP default parameters for solubility, AMAD and chronic intake.
- Assume daily urine excretion rate of 1500 mℓ/day
- No additional bioassay monitoring needed.

Level 2: Annual projected CED between 3 – 6 mSv

- Perform additional bioassay monitoring including 60 min lung count and additional single-void urine analysis. The lung count is to confirm that the CED does not exceed 6 mSv and the additional urine analysis will confirm a positive intake.
- Perform realistic dose calculation. Typical parameters to be adjusted relates to the material (AMAD, solubility and enrichment) and exposure scenario (time of intake).
- Peer review of dose calculations.

Level 3: Annual projected CED > 6 mSv

- Comprehensive set of measurement data is needed, which includes 24 hour urine sampling protocol, 60 min lung counting and faecal analysis. The estimation of the CED will be based on the comprehensive set of measurement data from all of the available bioassay techniques. The 24 hour sampling will ensure normalisation to individual specific excretion rate. Lung and faecal results can also be used to assist in determining route of intake, solubility and AMAD. Typically, parameters to be adjusted relate

to the material (AMAD, solubility and enrichment), the exposure scenario (time and route of intake) and the individual (e.g. HRTM particle transport rates in case of inhalation). Systematically change default model parameters until the fit is acceptable to all of the data and provide valid justification for rejected data

- Consider alpha spectroscopy of urine in order to determine enrichment.
- Peer review of dose calculations.
- Evaluate results from previous monitoring periods and consider correcting for intakes during dose calculations. The reconstruction of an intake is usually performed on a basis of a single data point in a time series of measurements. If more than 10% of the actual measured quantity can be attributed to intakes in previous monitoring intervals, making a corresponding correction is recommended.
- Review the working conditions and circumstances of the exposure.

The annual projected dose is accumulated over a period of 12 months assuming similar intakes in each monitoring period (MPe) and is given by

Annual projected CED = Measured dose x Number of MPe per year. (3.7)

3.4.2 Monitoring protocol (IDMP) for Type S uranium intakes

Section 3.3.3 indicates that the sensitivities of current urine analysis and lung counting are inadequate to detect the slow absorbing Type S uranium to a satisfactory level. The detection capabilities can be reduced significantly (≈ 30 pg $^{235}U/\ell$) with new improved ICP-MS methods. This is well within the required sensitivity level. Routine monitoring for Type S uranium is not essential when based on the current exposure scenario (see Section 3.2.1), but improved techniques capable (including faecal analysis) of detecting small Type S intake is recommended.

CHAPTER 4

CONCLUSIONS

The primary objective of the present study was to benchmark Necsa' Internal Dosimetry and Monitoring programme (IDMP) for uranium against best international practices. This was done with an extensive literature survey, addressing most of the pertinent issues in an IDMP. A knowledge base was built up containing best practices and these practices were presented in the present study. The literature survey entailed studying and understanding exposure scenarios within a uranium plant, and specifically the physiochemical nature of the uranium compounds found in a uranium plant. A uranium monitoring programme, i.e. monitoring techniques and its capabilities, as recommend in literature, was described. Shortcomings of Necsa's programme were highlighted. The important parameters used during dose calculations and the effect it has on the calculated Committed Effective Dose (CED) were investigated. Specifically, the present study included calculations of and validation of limits of detection and performance criteria for the various monitoring techniques used by Necsa.

The objective of an IDMP is to obtain intake data and to assess the committed effective dose in order to contribute to the control of operations. This aim was achieved firstly by the optimization of the monitoring programme (based on workplace exposure and physiochemical characterisation) in order to ensure that monitoring results are representative of the real exposure. The second step is the optimising the evaluation of monitoring data (individual dosimetry) to ensure it is the best estimate (harmonised and accurate following a graded approach). Each of the contributing factors for an effective IDMP is summarised below highlighting the impact of the factor, its effect on Necsa's IDMP and recommendations.

4.1 Contributing factors to a internal dosimetry and monitoring programme

Activity Median Aerodynamic Diameter (AMAD)

CED calculations have shown a significant dependence on the AMAD value. Literature survey has shown that AMAD for a typical uranium plant such as Necsa are mostly between 4 μm and < 8 μm with a limited effect on CED (within ± 5%). Following a graded approach, the default ICRP AMAD value (5 μm) will be acceptable for routine Type M monitoring (lung counting and urine analysis). The recommended default ICRP value are representative of the inhaled aerosol and do not significantly under- or over-estimate dose for AMAD values within the range found at Necsa. However, more accurate AMAD values should be used for intakes close to or exceeding reference levels (in the case of lung counting).

Solubility

CED calculations indicated significant dependence on solubility. The solubility classes of compounds at Necsa's uranium plant can be classified as Type M, except in limited steps in the production process where it is classified as Type S uranium. However, these either occur in a glove box or are done with the necessary respiratory protective equipment. Literature confirmed ICRP recommended solubility classes but cautioned there could be large deviation from recommended default values especially during high temperate operations. High temperature treatment of UO_2 does occur, but the process is in an enclosed glove box, thus minimising the possibility of intake. Thus, one can recommend a routine monitoring programme assuming exposure to Type M. However, more representative solubility values should be used for intakes close to or exceeding reference levels.

Enrichment

CED calculations are not dependent on the enrichment since dose conversion factors of the three main isotopes of uranium are very similar. However, determination of the total uranium activity from a measured value is strongly dependant on enrichment. The total activity (specifically ^{234}U activity) is derived

from the measure ^{235}U activity. A default 30% mass enrichment is recommended based on the exposure scenarios at Necsa (between 5% and 50%). This will result in a limited effect on the resultant total activity and CED calculations (- 2% to + 17%). The $^{235/234}U$ mass ratio is main factor determining the total activity. This ratio is enrichment process specific and can differ significantly between enrichment processes. Purchased uranium compounds could in future form a significant part of the uranium mix at Necsa and a different $^{235/234}U$ mass ratio could have a significant impact on the CED calculations. Thus, one can recommend a routine monitoring programme assuming a mass enrichment of 30%. However, more representative enrichment values should be used for intakes close to or exceeding reference levels. It should be noted that Necsa has embarked on a programme to use low enriched uranium target plates for the production of medical isotopes. This could also significantly impact the assumptions regarding enrichment.

Route of intake

Literature survey recommended inhalation as the most probable rout of intake. However, even a small fraction of ingestion can have a significant impact on the CED, leading to large overestimations. Thus, it is recommended that the distribution between inhalation and ingestion should be investigated for intakes close to or exceeding reference levels. A combination of bioassay techniques can be used to assist in the determination of the different intake fractions.

Time of intake

Literature survey has indicated that most exposures are chronic and not acute. Newer versions of software for calculating CEDs are more capable of simulating exposure scenarios and thus are recommended that chronic option should be used for routine exposure calculations.

Urine analysis

Another technique (other than DNC) with better sensitivity is not necessary for Type M monitoring, but better sensitivity is necessary for Type S monitoring. It

should be noted that the required sensitivity levels for Type S are within the order of dietary intakes and the evaluation of low intake values should take dietary intake into consideration. It is recommended that Necsa should not correct for dietary intake, i.e. subtract it from urine results but rather determine a dietary intake range for Necsa workers and take it into consideration during intake evaluations.

The current DNC technique was found to be adequate and no immediate improvements thus are necessary. However, techniques with a faster turnaround time and lower sensitivity would be advantageous.

A singe void sample is recommended for routine urine monitoring. It is recommended that Necsa continue with normalisation of the singe void sample using 1500 mℓ/day. ICRP default excretion values or a 24 hour sampling protocol for intakes close to or exceeding reference levels are recommended.

Lung Counting

Lung counting on its own is not adequate for routine uranium monitoring, due to poor sensitivity. It is only complementary to urine monitoring and useful for detecting significant intakes, especially Type S. Lung counting is an indication of accumulated exposures over longer periods (6 or 12 months), thus is useful to monitor for long term build-up, especially for highly insoluble compounds. The current lung counting technique at Necsa is optimised and the best technology available is used. Although, sensitivities are not within the required levels, it still compares well with sensitivities of other systems published and no improvements are thought to be necessary. It is also recommended that the monitoring frequency for routine monitoring be changed from one year to six monthly.

Faecal analysis and technique combination

Faecal analysis is currently not part of the routine monitoring programme. The technique is not yet available at Necsa. Faecal analysis has better sensitivity than urine monitoring for exposures to Type S uranium. The difference in biokinetic

behaviour between respiratory tract (lungs) and gastrointestinal tract (faeces) and between kidneys (urine) and gastrointestinal tract (faeces) will result in different stay times and excretion rates. This can be used to differentiate between intakes of different solubility classes, AMAD and intake pathways. The assessment of doses from intakes of uranium requires a combination of techniques in order to provide the best estimate. Although faecal analysis is an invasive technique, it is recommended that Necsa implement this technique.

4.2 Summary

It was found that Necsa's bioassay techniques for uranium are adequate and compares very well with international best practices. DNC analysis of urine is adequate (acceptable sensitivity) for Type M uranium, however, turnaround time is still problematic. A more sensitive technique (specifically for Type S uranium) with a better turnaround time will be beneficial and is recommended. Urine analysis techniques with lower sensitivity would be an advantage for Type S monitoring but at such low levels it will be difficult to distinguish from dietary intakes. Faecal analysis has a much improved sensitivity and could also assist in entry route determination. The implementation of faecal monitoring is recommended. No immediate improvements are thus necessary for Necsa's uranium urine programme, but techniques with faster turnaround time and lower sensitivity would be advantages.

The current lung counting technique at Necsa is optimised and the best available technology is used. The sensitivity of the Necsa's lung counter compares well other published sensitivities and no improvements are necessary. Since exposure to Type M is mostly expected, it is thus recommended that the monitoring period be changed from 365 days to 180 days based. This is to ensure the underestimation is less than a factor 3 as recommended by ICRP. Lung counting is complementary to urine analysis. As such, good sensitivity is not essential, however detection capability can be improved with increasing the counting time from 30 min to 60 min. This is not recommended for routine counting, but only in cases of unexpected intakes.

Due to high variability in dose calculations, the aim is to have a consistent approach in determining the best estimate of the dose. Thus, two independent evaluations should get the same result and the result should be as close as possible to the true dose. Analysis done to quantify the effect of differences in physiochemical characteristic of the uranium compound (e.g. AMAD) and differences in human metabolic parameters (e.g. daily urine volume) has brought an understanding on how these parameters can influence CED calculation. Literature study and calculations support the defaults assumption to be used during CED calculations. The dose calculation steps as described in Section 3.4.1 ensures that Necsa's methodology is aligned (harmonised) with international recommendation. When using the appropriate parameters (depending on the estimated dose) and the recommended bioassay protocol, one is ensured that the calculated CED is also the best estimate (accurate). Using IMBA, by a relative expert user, will further ensure that results are the best estimate (harmonised and accurate).

It was shown that Necsa's IDMP is in line with best international practices and adequate to demonstrate that exposures are optimised and below regulatory limits. The IDMP is adequate and able to demonstrate compliance with the performance criteria for monitoring and dose calculation. Knowledge gained from the present study will further enhance the programme and assist in developing the necessary documentation, providing the technical justification for Necsa's uranium IDMP.

REFERENCES

[AH07] Ahmed, S.N., Physics and engineering of radiation detection, 1st Edition, Elsevier, 2007

[AN96] American National Standard, HPS N13.30-1996: Performance Criteria for Radiobioassay, 1996

[AN97] Ansoborlo, E., Boulaud, D., Leguen, B., Distribution granulométrique d'aérosols d'uranium dans l'industrie française de fabrication du combustible, Radioprotection, 32, 1997

[AN99] Ansoborlo, E., Hengé-Napoli, H., Chazel, V., Gilbert, R., Guilmette. A. Review and critical analysis of available in-vitro dissolution test, Helath Physics, 77(6), 1999

[AN01] American National Standard, HPS N13.39-2001: Design of Internal Dosimetry Programme, 2001

[AN02] Ansoborlo, E., Chazel, V., Hengé-Napoli, M., Pihet, P., Rannou, A., Bailey, M., Stradling, N., Determination of the physical and chemical properties, biokinetics and dose coefficients of uranium compounds handled during nuclear fuel fabrication in France, Health Physics, 82(3), 2002

[AN03] Ansoborlo, E., Berard, P., Eckerman, K., Berkovski, V., Birchall, A., Guilmette, R., Miller, G., Ishiure, N., Lipsztein, J., Nosske, D., Review of methods and computer codes for interpretation of bioassay data, Radiation Protection Dosimetry, 105(104), 2003

[AN07] Anderson, J.L., Spitz, H.B., Yin, J.H., Characterization of internal exposures to enriched uranium at a former gaseous diffusion plan, Health Physics, 93(6), 2007

[AR11] Argonne National Laboratory, Natural Decay Series: Uranium, Radium, and Thorium, Available from http://www.ead.anl.gov/pub/doc/natural-decay-series, 2011

[BI94] Birchall, A., James AC., A Rapid Method for Modelling the Kinetics of Radioactive Progeny Applied to Thorium in the Lungs, Health Physics. 67, 1994.

[BI06] Birchall, A., Puncher, M., Obtaining an unbiased estimate of intake in routine monitoring when time of intake is unknown, Draft Guidance Document on Interpretation of Bioassay Data, November 2005, Available from Alan.Birchall@hpa.org.uk

[BR11] Bruker Axs, Available from http://www.bruker-axs.de/fileadmin/user_upload/xrfintro/sec1_6.html, 2011

[CA03] Carbaugh, E.H., Minimum detectable dose as a measure of bioassay programme capability, Health Physics, 105(1-4), 2003

[CA04] Casper, S., Brookhaven Technical Basis Document for the Internal Dosimetry Program. Power Point Slideshow available from CASPER@BNL.GOV, 2004

[CA09] Carbaugh, E.H., Bihl, D., Maclellan, J., Hanford Internal Dosimetry Program Manual, PNL-MA-552, PNNL-15613 Rev.1, Pacific Northwest National Laboratory, 2009. Available from http://www.pnl.gov/publications/

[CE96] Cember, H., Introduction to Health Physics, 3rd Edition, McGraw Hill Companies Inc., 1996

[CU68] Currie, L.A., Limits for qualitative detection and quantitative determination, Analytical Chemistry, 40(3), 1968

[DO95] Dorrian, M.D., Bailey, M.R., Particle Size Distribution of Radioactive Aerosols measured in Workplaces, Radiation Protection Dosimetry 60(2); 1995

[DO99] Department of Energy USA, DOE-STD-1121-1998: Internal Dosimetry. December 1999

[DO04] Department of Energy USA, DOE-STD-1136-2004: Guide of Good Practices for Occupational Radiological Protection in uranium Facilities. December 2004

[DO07a] Department of Energy USA, DOE-G-441.1-3A: Radiation protection programs guide, March 2007

[DO07] Doerfel, H., Andrasi, A., Bailey, M., Blanchardon, E., Cruz-Suarez, R, Berkovski, V., Castellani, C.M., Hurtgen, C., Le Guen, B., Malatova, I., Marsh, J., Stather, J., Zeger, J., General Guidelines for Assessment of Internal Dose from Monitoring Data: Progress of the IDEAS Project, Radiation Protection Dosimetry 125(1-4); 2007

[DO08] Doerfel, H., Andrasi, A., Bailey, M., Berkovski, V., Blanchardon, E., Castellani, C.M., Cruz-Suarez, R, Etherington, G., Hurtgen, C., Le Guen, B., Malatova, I., Marsh, J., Stather, J., Refresher Course 6 - Internal dosimetry: The science and art of internal dose assessment, Presented at the 12th IRPA Congress, Buenos Aires, Argentina, 2008, Available from www.irpa.org

[EI94] Eidson, A.F., The effect of solubility on inhales uranium compound clearance: a review. Health Physics, 67(1), 1994

[ET03] Etherington, G., Stradling, G.N., Rahola, T., Le Guen, B., Hurtgen, C., Jourdain, J.R., Bérard, P., Design and implementation of monitoring programmes for internal exposure (Project OMINEX), Radiation Protection Dosimetry, 105 (1-4), 2003

[FE04] Fell, T.P., Phillips, A.W., Stradling, G.N., Assessment of internal doses to workers potentially exposed to enriched uranyl fluoride and uranium Tetrafluoride, NRPB Report NRPB-W56, 2004

[FI94] Fishwick, A.H., Finlayson, J.L., Dower, B., Internal dosimetry for uranium fuel manufacture at BNFL, Radiation Protection Dosimetry, 53 (1-4), 1994

[FU03] Fujita, M. Sato, M. Assessment of the internal effective dose of workers independent of inhaled particle size, Radiation Protection Dosimetry 105(1-4); 2003

[HE08] Hegyi, G., Richardson. R., Sizing particles of natural uranium and nuclear fuels using polyally-diglycol carbonate autography, Radiation Protection Dosimetry 130(4), 2008

[HU03] Hurtgen, C., Cossonnet, C., A survey of uncertainty in bioassay measurements carried out within the OMINEX project, Radiation protection Dosimetry, 105(104), 2003

[IA99a] International Atomic Energy Agency, IAEA Safety Guide No. RS-G-1.2, Assessment of occupational exposure due to intakes of radionuclides, 1999

[IA99b] International Atomic Energy Agency, IAEA Safety Guide No. RS-G-1.1, Occupation radiation protection, 1999

[IA04a] International Atomic Energy Agency, IAEA Practical radiation technical manual, IAEA-PRTM-1 Rev. 1, Workplace monitoring for radiation and contamination, 2004

[IA04b] International Atomic Energy Agency, IAEA Safety Reports Series No. 37, Methods for Assessing Occupational Radiation Doses from Intakes of Radionuclides, (2004)

[IC88] International Commission on Radiation Protection, ICRP 54: Individual monitoring for intakes of radionuclides by workers: Design and Interpretation, Annals of the ICRP, 19(1-3), 1988

[IC94] International Commission on Radiation Protection, ICRP 66: Human respiratory tract model for radiation protection, Annals of the ICRP, 24(1-4), 1994

[IC97] International Commission on Radiation Protection, ICRP 78: Individual Monitoring for Internal exposure of workers. Annals of the ICRP, 27(3-4), 1997

[IC02] International Commission on Radiation Protection, ICRP 89: Basic Anatomical and Physiological Data for Use in Radiological Protection Reference Values, Annals of the ICRP, 32(3-4), 2002

[IC06] International Commission on Radiation Protection, ICRP 100: Human Alimentary Tract Model for Radiological Protection, Annals of the ICRP, 36(1-2), 2006

[IC07] International Commission on Radiation Protection, ICRP 103: The 2007 Recommendations of the International Commission on Radiological Protection, Annals of the ICRP, 37(2–4), 2007

[ID06] Project IDEAS-EU, Report FZKA 7243: General Guidelines for Estimation of Committed Effective Dose from Incorporated Monitoring Data, June 2006

[IR11] IRP 2010 -2030, Integrated Resource Plan for Energy, March 2011 Available from www.energy.gov.za

[IS10] International Standards Organisation, ISO/DIS 27048: Radiation Protection – Dose Assessment for the monitoring of workers for internal radiation exposure, April 2010

[KA01] Karpas, Z., uranium Bioassay – Beyond Urinalysis, Health Physics, 81(4), 2001

[KI05] Kim, P.K., Wu, C., Birky, B.K., Bolch, W.E., Effective dose scaling factors for use with cascade impactor sampling data in TENORM inhalation exposures, Health Physics, 89(4), 2005

[KN99] Knoll, G.F., Radiation detection and measurement, 3rd Edition, John Wiley and Sons Inc., 1999

[KR05] Kramer, G.H., Hauck, B.M., The Human Monitoring Laboratory's new lung counter: Calibration and comparison with the previous system at the CAMECO Corporation's lung counter, Health Physics, 89(4), 2005

[LE89] Leggett, R.W., The behaviour and chemical toxicity of U in the kidney: a reassessment, Health Physics, 57, 1989

[LE10] Lee, J.K., Potential errors in committed effective dose due to the assumption of a single intake path in the interpretation of bioassay results. Presented at the HPS 52nd Annual Meeting, Utah, 2010

[LO05] Lopez-Fidalgo, J., Sanchez, G., Statistical criteria to establish bioassay programs, Health Physics, 89(4), 2005

[LO07] Lopez, M.A., Castellani, C.M., Currivan, L., van Dijk, J., Falk, R., Olko, P., Wernli, C., State of the art dosimetric methods for internal and external exposures: Conclusions of a EURADOS action, Radiation Protection Dosimetry, 125(1-4), 2007

[MA07] Marcus, C.S., Siegel, J.A., Sparks, R.B., Medical Management of Internally Radiocontaminated Patients, June, 2006. Report available from http://cdrp.net/ee/MMRSManual.pdf

[MC99] McDiarmid, M.A., Hooper, F. J., Squibb, K., McPhaul, K., The utility of spot collection for urinary uranium determinations in depleted uranium exposed gulf war veterans, Health Physics, 77(3), 1999

[MU07] Muikku, M., Heikkinen, T. Puhakainen, M., Rahola, T., Salonen, L., Assessment of occupational exposures to uranium by direct methods needs information on natural background variations, Radiation Protection Dosimetry, 125(1-4), 2007

[NR96] National Radiological Protection Board UK, NRPB-SR287: LUDEP 2.0 Personal computer programme for calculating internal doses using the ICRP Publication 66 respiratory tract model, 1996

[NR99] National Radiological Commission USA, NRC Document 10CFR835: Occupational Radiation Protection. 1999

[NR04] National Radiological Protection Board UK, NRPB-W60 OMINEX Report: Optimization of Monitoring of Internal exposure, Project funded by the European Commission, 2004

[OE07] Oeh, U., Andrasi, A., Bouvier-Capely, C., de Carlan, L., Fischer, H., Franck, D., Höllriegl, V., Li, W.B., Ritt, J., Roth, P., Schmitzer, C., Wahl, W., Zombori, P., Implementation of Bioassay Methods to Improve Assessment of Incorporated Radionuclides. Radiation Protection Dosimetry, 125(1-4); 2007

[PN03] Pacific Northwest National Laboratory, Hanford Manual PNNL-MA-860: Methods and Models of the Hanford Internal Dosimetry Program. 2003. Available URL: http://www.pnl.gov/

[PO04] Potter, A., Internal Dosimetry, A Review. Health Physics, 87(5), 2004

[RA94] Raabe (1994) Raabe, O. G., Internal Radiation Dosimetry, Medical Physics Publishing, 1994

[RA03] Rahola. T., Etherington, G., Bérard, P., Le Guen, B., Hurtge, C., OMINEX: Survey of internal dose monitoring programmes for radiation workers, Radiation Protection Dosimetry, 105 (1-4), 2003

[RU98] Rucker, T.L., Johnson, C.M., relationship between isotropic uranium activities and total uranium at various uranium enrichments, Journal of radioanalytical and nuclear chemistry, 235(1-2), 1998

[RU01] Rucker, T.L., Uranium lung solubility class selection at Bechtel Jacobs Company LLC-Operated Facilities, Presented at the 47th Annual Radiochemical Measurements Conference, Honolulu, Hawaii (November 2001). Available from www.lanl.gov/BAER-

Conference/BAERCon-47p-Rucker.pdf

[SC03] Schmitzer, C., Brandl, A., Wahl, W., Roth, P., Franck, D., de Carlan, L., Andrasi, A., Development in internal monitoring techniques, Radiation Protection Dosimetry, 105(1-4), 2003

[SC05] StudieCentrum voor Kernenergie, Report: CK•CEN-BLG-1018, IDEAS / IAEA Intercomparison Exercise on Internal Dose Assessment, October 2005

[SC07] Schmitzer, C., Fischer, H., Andrasi, A., Bouvier, C., de Carlan, L., Franck, D., Höllriegl, V., Li, W.B., Oeh, U., Ritt, J., Roth, P., Wahl, W., Zombori, P., Improvements in routine internal monitoring- An overview of the IDEA project, Radiation Protection Dosimetry, 125(1-4), 2007

[SH92] Shleien, B., The health physics and radiological health handbook, Revised edition, Scinta Inc., 1992

[ST03] Strom, D.J., Eliminating bias in routine bioassay when there is an unknown time of intake, Radiation Protection Dosimetry, 105(1-4), 2003

[ST08] Stabin, M., Radiation protection and dosimetry, 1st Edition, Springer, 2008

[TA03] Takala, J.M., Kramer, G.H., Use of group monitoring data in lung dose estimation for intakes of uranium, Health Physics, 105(1-4), 2003