

INVESTIGATION AND CALCULATION OF DOSE COEFFICIENTS FOR
RADIONUCLIDES PRODUCED IN A SPALLATION NEUTRON SOURCE USING
THE ENSDF AND NUBASE NUCLEAR DATABASES

by

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ABSTRACT

Investigation and Calculation of Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source Using the ENSDF and NUBASE Nuclear Databases

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Dose coefficients are useful for risk assessment during the design and siting of accelerator-driven nuclear facilities including the Spallation Neutron Source. There are seventy-two radionuclides with half-lives equal to or greater than one minute that will be produced by the spallation of a mercury target for which no published dose coefficients exist. Out of these seventy-two, twenty-four currently have conflicting published nuclear data in the Evaluated Nuclear Structure Data Files (ENSDF) and the NUBASE data files. In this research these twenty-four radionuclides have been studied. Because of missing ENSDF records, internal and external dose coefficients were determined for only six radionuclides using both ENSDF and NUBASE values, which had conflicting data. The methodology used was adopted from Federal Guidance Report 13. Information from this study will augment radiological data and be used to quantify the radiological risks to workers and members of public.

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CHAPTER 1

INTRODUCTION

The U.S. Department of Energy (DOE) was authorized by Congress in 2001 to establish the Advanced Accelerator Applications (AAA) program to address the many urgent nuclear issues facing the nation. The AAA Program is a national program for development of nuclear technology through advanced accelerator applications. The University of Nevada, Las Vegas (UNLV) Transmutation Research Program was built in March 2001 as part of the national AAA program to develop the technologies required for the ecological and economical management of spent nuclear fuel. These research programs span the range of technology areas in transmutation, from uranium separation from spent nuclear fuel to accelerator-related issues (Hechanova et al. 2001).

The UNLV Transmutation Research Program has been assigned to support DOE efforts to assess the health risks associated with the operation of each of their accelerator-driven nuclear facilities. It is important to assess radiological risk in order to limit radiation exposures to workers during the design and siting of accelerator-driven nuclear systems. Because of the developments of high-energy accelerators, radiation protection for high-energy radiation has become important. It is necessary to guarantee the safety of workers at accelerator-driven nuclear systems before these systems can be deployed for nuclear transmutation or other missions. Spallation is a nuclear reaction in which a heavy atomic nucleus is bombarded by a fast incident particle of high energy resulting in freeing

some neutrons. The energy that is deposited in the target nucleus causes the nucleus to disintegrate into a number of smaller nuclear components. Some neutrons are ejected and the target nucleus is left in an excited state. It is deexcited by emitting particles, preferentially neutrons which may still have enough energy to induce new reactions of spallation with neighboring nuclei.

The spallation neutron source (SNS) at Oak Ridge National Laboratory can produce the most intense pulsed neutron beams for scientific research and industrial development. Powerful neutron beams are produced in the SNS facility by striking a mercury target with high-energy protons from a large accelerator complex. The protons excite the mercury nuclei and release neutrons to form an intense beam. Around accelerator systems, a number of secondary radiations are produced due to interactions of charged particles with shielding materials.

At the SNS facility, the components of the proton accelerator complex, the target and the shielding materials, are exposed to primary and secondary high-energy particles causing generation of different spallation byproducts in the surrounding area. To limit exposure to workers and ensure their safety, their radiation risk should be evaluated in these environments. The effective dose recommended by publication 60 of the International Commission on Radiological Protection (ICRP) is the primary radiological protection quantity to assess radiation risks (ICRP 1991). Dose coefficients defined as effective doses and organ doses per unit intake are used by radiation safety personnel to calculate effective dose and organ dose via a measured intake from a given exposure.

During the spallation process, many rare radionuclides are created which may be a potential radiation exposure to workers. More than 520 radionuclides have been

identified as spallation byproduct materials. It is the focus of this work to investigate the possibility of calculating internal and external dose coefficients (DCs) for these radionuclides. DC results from this study will augment radiological data in Federal Guidance Reports No. 11 and 12 (EPA 1988; EPA 1993) and in Publications 68 and 72 of the International Commission on Radiological Protection (ICRP 1995; ICRP 1996). Information obtained will contribute to the health physics community and to the development and safe operation of accelerator-driven nuclear facilities.

1.1 Internal Dosimetry

Due to the complicated nature of an inhalation or ingestion, simple compartmental models are used to describe the movement of radionuclides through the body. The body is comprised of a number of separate compartments with any organ or tissue consisting of one or more compartments. Elimination of radionuclides from any compartment is assumed by first order kinetics and retention is expressed as exponential terms. After inhalation or ingestion, a radionuclide will be transferred to the body fluids at a rate determined by rate constants for different components of the respiratory and gastrointestinal (GI) models and by the radioactive decay of the radionuclide.

In internal dosimetry, two sets of anatomical regions in the body are concerned. Source regions specify the location of radionuclide in the body. Target regions involve the organs and tissues for which the radiation dose is of interest. For a specific radionuclide, source regions comprise anatomical regions along the intake route (respiratory and gastrointestinal tract), regions associated with the systemic behavior of the radionuclide, and regions along the elimination routes. A region may involve multiple

compartments. A radionuclide is assumed to be distributed uniformly either by volume or surface area in each region.

In order to compute radiation dose, it is necessary to relate compartments of the biokinetic model to anatomical regions of the body referred to as source regions. The mean absorbed dose to the target region is the basic quantity. Age-specific internal dose coefficients for ingestion and inhalation could be calculated for both occupational and public exposure. Dose coefficients for effective dose equivalent and effective dose can be calculated according to corresponding ICRP 26 and ICRP 60 recommendations (ICRP 1977; ICRP 1991). The metabolic models and data in ICRP 30 and ICRP 66 (ICRP 1979; ICRP 1994) are used to determine dose coefficients for target organs of interest and total committed effective dose. The committed effective dose to an individual is determined by numerous physical, chemical, and biological factors.

Biokinetic models describe tissue distribution, retention, and excretion of systemic activity. Some biokinetic models used now are more complicated than those traditionally used because of a recent movement toward biological realism in dealing with radionuclides in the body. For example, the new age-specific respiratory tract model (ICRP 1994) reflects significantly greater anatomical and physiological realism and incorporates many factors affecting dose assessment of inhaled radioactivity. It depends on two factors to calculate the committed dose equivalents $H_{50,i}$ for each type of radiation i in target organ T due to radionuclide j in source organ S .

- Total number of transformations of radionuclide j in the source organ over 50 years following the intake.

- Energy deposited per unit mass in the target organ weighted by the quality factor from radiation type of i per transformation of radionuclide j in source organ.

The committed dose equivalent in Sv in an organ T due to radiation type i from radionuclide j in source organ S is given by the expression (ICRP 1979):

$$H_{50}(T \leftarrow S)_i = 1.6 \times 10^{-10} \times U_s \times SEE(T \leftarrow S)_i, \quad \text{Eq. 1.1}$$

where U_s represents the total number of transformations of radionuclide j in the 50-year period after intake. It is the time integral of activity of the radionuclide in source organ over the stated period of time. $SEE(T \leftarrow S)_i$ is the Specific Effective Energy for radiation type i . Accordingly for all types of radiation from radionuclide j , the committed dose equivalent in Sv is (ICRP 1979):

$$H_{50}(T \leftarrow S)_j = 1.6 \times 10^{-10} [U_s \sum_i SEE(T \leftarrow S)_i]_j. \quad \text{Eq. 1.2}$$

In general, the total committed dose equivalent $H_{50,T}$ in Sv in target tissue or organ T from activities in various source organs could be expressed as (ICRP 1979):

$$H_{50,T} = 1.6 \times 10^{-10} \sum_S \sum_j [U_s \sum_i SEE(T \leftarrow S)_i]_j, \quad \text{Eq. 1.3}$$

where the summation is over all radiations resulting from all radionuclides in all source organs involved. The committed effective dose equivalent $H_{50,E}$ in Sv could then be calculated by:

$$H_{50,E} = \sum_T w_T H_{50,T}, \quad \text{Eq. 1.4}$$

where w_T is the tissue weighting factor (ICRP 1991).

1.2 External Dosimetry

The three external exposure modes considered in our study are: submersion in a semi-infinite contaminated air cloud, exposure from ground surface contamination, and exposure from soil contaminated to an infinite depth. The external dose quantities involve the dose equivalent in various target organs and the effective dose equivalent (ICRP 1977). The corresponding new quantities are equivalent dose to tissues and effective dose (ICRP 1991). Skin is included in target organs for the dose equivalents since it is the most frequently irradiated tissue from external exposure. For an external source, only penetrating radiations are concerned because they could pass the overlying tissues from outside to deposit energy in radiosensitive tissues of the body. Penetrating radiations are restricted to photons and electrons, for which the quality factor or radiation weighting factor is unity. Therefore the dose equivalent is numerically equal to the absorbed dose in a tissue.

The dose-rate coefficients for external exposures are based on calculating the energy and angular distribution of the radiation incident on the body and the transport of these radiations within the body. The methods of calculation include two independent steps: (1) the calculation of the radiation field incident on a closed surface surrounding the human phantom model, and (2) the calculation of organ doses due to a surface source equivalent to the angular flow rate entering the boundary surrounding the phantom. The radiation dose depends strongly on the temporal and spatial distribution of the radionuclide.

It is difficult to compute the dose to tissues from radiations emitted by a random distribution of a radionuclide in the environment. Hence it is common to consider simplified and idealized exposure geometries. It is assumed that the contaminated

medium has a radionuclide concentration that is uniform and effectively infinite or semi-infinite in extent. Usually a semi-infinite source region is assumed for air submersion and an infinite source region is assumed for exposures to contaminated soil. Consequently, in an assumed infinite or semi-infinite source region with a uniform concentration $C(t)$ of a radionuclide at time t , the dose equivalent H_T in Sv in tissue T could be calculated from:

$$H_T = h_T \int C(t) dt, \quad \text{Eq. 1.5}$$

where h_T is defined as the dose to tissue T of the body per unit time-integrated exposure, which is expressed in terms of the time-integrated concentration of the radionuclide. The time-independent dose coefficient for external exposure h_T in Sv per Bq s m⁻³ is thus given by:

$$h_T = \frac{H_T}{\int C(t) dt}. \quad \text{Eq. 1.6}$$

An alternative definition of h_T is the instantaneous dose rate for organ T per unit activity concentration of the radionuclide in the environmental medium.

In most cases, the dose H_T is the quantity of interest. The limits of integration are not specified here because they depend on the nature of the dose quantity H_T . The dose coefficient h_T for a specific radionuclide is determined by the type, intensity, and energy of the radiations emitted, the mode of exposure, and the anatomical variables that affect the energy absorption in organ T . The dose coefficient combines the transport of radiations in the environmental medium, their subsequent transport within the body, and determination of the energy deposition in the tissues.

There are three major steps to calculate the dose coefficients: (1) calculate the energy and angular distributions of the radiations incident on the body for a range of initial

energies of monoenergetic sources distributed in environment; (2) determine the transport and energy deposition in organs and tissues of the incident radiations; and (3) compute organ dose for specific radionuclides, taking into account the energies and intensities of the radiations emitted by the radionuclides (Eckerman et al. 1993).

1.2.1 Organ Doses from Photon Sources

Organ doses are calculated for monoenergetic environmental photon emitters at twelve energies from 0.01 to 5.0 MeV for 25 target tissues. The results are then used to determine the dose coefficients, considering the detailed photon spectrum of each radionuclide. The dose to the body is very sensitive to the direction of incident radiation, hence the methods used to compute organ dose coefficients for monoenergetic sources give a precise description of the energy and angular dependence of radiation field incident on the body. The source of the submersion dose calculations is a semi-infinite cloud containing a uniformly-distributed monoenergetic photon emitter of unit strength (1 Bq m^{-3}) surrounding a human phantom standing on the soil at the air-ground interface. The source for the contaminated soil calculations is an infinite isotropic plane source of monoenergetic photons of unit strength (1 Bq m^{-2}) at the air-ground interface or a specified depth in the soil. The organ dose due to a source in the soil that is uniformly distributed from the surface to a specified depth could be calculated from the doses due to a series of plane sources at different depths. The organ dose coefficients for isotropic plane sources at the six source depths are integrated over source depth to calculate organ dose coefficients for uniformly distributed volume sources having thicknesses of 1, 5, and 15 cm, and for an effectively infinitely thick source (Eckerman et al. 1993).

1.2.2 Skin Doses from Electrons

The skin dose due to monoenergetic environmental electron sources should be concerned for air submersion, and ground-surface and volume exposures. Because of the short range of electrons in the tissue, it is not necessary to take into account the contribution of electrons to doses to tissues other than the skin. However, bremsstrahlung radiation caused by electrons slowing down in a medium is sufficient to penetrate the skin and contribute to the dose to underlying tissues.

1.2.3 Dose Coefficient Formulation

The energy spectra of radiations emitted are either discrete for gamma emissions or continuous for beta particles and bremsstrahlung. The beta spectra are used to estimate the contribution of the beta particles to skin dose and to evaluate the bremsstrahlung yield. The dose coefficient h_T^S for tissue T for any exposure mode S in *Sv per Bq s m⁻³* is given by:

$$h_T^S = \sum_{j=e,\gamma} \left[\sum_i y_j(E_i) \hat{h}_{Tj}^S(E_i) + \int_0^\infty y_j(E) \hat{h}_{Tj}^S(E) dE \right], \quad \text{Eq. 1.7}$$

where $y_j(E_i)$ is the yield of discrete radiations of type j and energy E_i , and $y_j(E)$ is the yield of continuous radiations per nuclear transformation with energy between E and $E + dE$. The summation is over all electron and photon radiations. Each radiation has two compositions: the discrete energy emissions and the continuous emissions. The contribution of radiations to dose in organ T is represented by $\hat{h}_T^S(E)$ which is a function of energy for organ T for each exposure mode (Eckerman et al. 1993).

CHAPTER 2

METHODOLOGY

2.1 Background

A methodology to calculate dose coefficients for radionuclides produced in spallation neutron sources was developed by Omar Wooten of the Georgia Institute of Technology and Tony Andrade of Los Alamos National Laboratory. Mr. Wooten computed dose coefficients for 3-4 radionuclides using this method although several problems existed in this prototype code. Therefore, the methodology has been refined and optimized with the help of the research consortium consisting of representatives from several universities and national laboratories. Finally, the Dose Coefficient Working Group (DC Working Group) developed a reproducible methodology which was adopted from that used to develop Federal Guidance Report 13 and is shown in Fig. 2.1. In this methodology, radiological data is acquired from the Evaluated Nuclear Structure Data File (ENSDF) library maintained by the National Nuclear Data Center at Brookhaven National Laboratory and used as input data for the computer code EDISTR. The input data comprise decay modes, decay energy levels, and radiation energies and intensities, which are processed by the EDISTR code to create a radioactive decay data file. The processed data file contains not only the average or unique energies and intensities of the radiations, but also the beta spectra. The Decay Data (DECDAT) file arranges the output file of EDISTR into the proper format to incorporate into the nuclear decay data libraries within

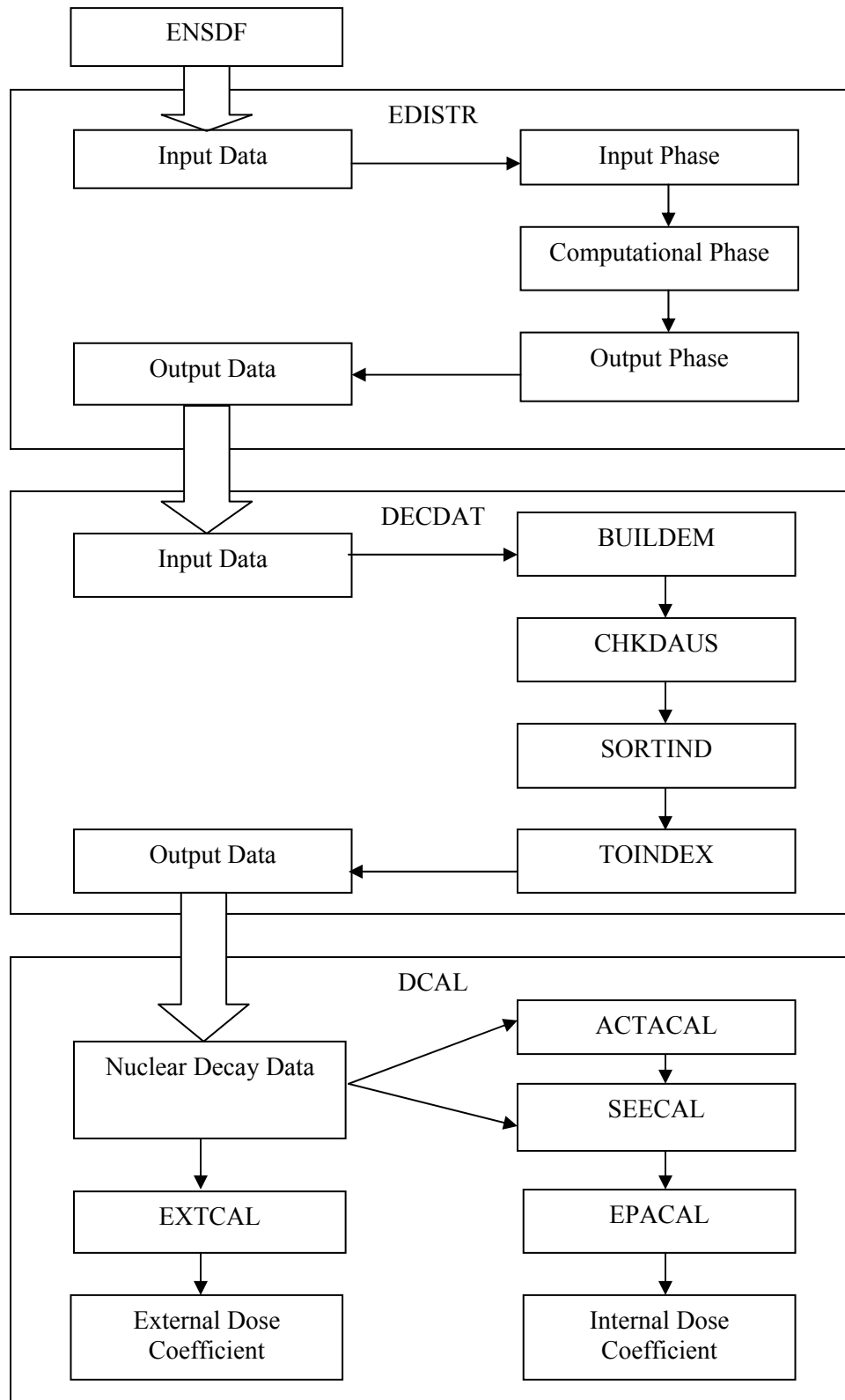


Figure 2.1 Flow chart of computer program for dose coefficient calculations

the Dose and Risk Calculation (DCAL) program developed by the Oak Ridge National Laboratory. DCAL software comprises a series of computational modules and extensive libraries of biokinetic, anatomical, and dosimetric data required for the dose coefficient computation.

2.2 Evaluated Nuclear Structure Data File (ENSDF)

The Evaluated Nuclear Structure Data File (ENSDF) is used to store nuclear structure properties of nuclides and the results from a variety of experiments to derive those properties. ENSDF is composed of a group of data sets, which have one of the following kinds of information: 1) the summary information for a mass chain, 2) the references used in the data sets for the given mass number, 3) the adopted level and gamma-ray properties for each nuclide, 4) the evaluated results of a single type of experiment, e.g., a radioactive decay or a nuclear reaction for a given nuclide, and 5) the combined evaluated results of a number of experiments of the same kind for a given nuclide.

A data set consisting of 80-column records has at least two records: the beginning and the end record. A data set must begin with an IDENTIFICATION record and end with an END record. The IDENTIFICATION record denotes the kind of data in the data set. The END record is blank and just tells the computer the end of the data set has been reached. Between these two records, there could be as many additional records as are required to depict fully the experimental or the evaluated information.

A DECAY data set is the data set that provides the radioactive decay information of a radionuclide. A DECAY data set IDENTIFICATION record includes three parts: the decaying nucleus, the decay type, and the word DECAY. Following the

IDENTIFICATION record is a NORMALIZATION record, and a PARENT record. Following the NORMALIZATION and PARENT records is a group of records, consisting of LEVEL, ALPHA, B-, EC, B+, and GAMMA records, which describe the measured or deduced properties of levels, gamma rays, alpha particles, and etc. Each LEVEL record is followed by a group of B-, EC, B+, or ALPHA records describing charged particle decay into the level and gamma ray out of the level (Tuli 1987).

2.3 EDISTR Computer Program

The computer program EDISTR extracts basic nuclear decay information from the ENSDF and prepares a nuclear decay database for radiation dosimetry computations. As we know for medical and health physics dosimetry calculations, the mean energies and absolute intensities of all the radiations associated with the radioactive decay process should be known. The average energy of the β^+ or β^- particles and the energies and intensities of radiations associated with spontaneous fission must also be known in dosimetry calculations. In addition bremsstrahlung yield and spectral shape information are also needed in a few cases which bremsstrahlung radiation may contribute to dose. The computer program EDISTR calculates intensities and mean energies of alpha, beta, gamma, internal-conversion-electron, x-ray, Auger-electron, and bremsstrahlung radiations resulting from nuclear decay.

The yields of radiations associated with spontaneous fission are computed in a few spontaneous fission cases. Moreover, spectral shapes of the continuous beta distributions are also calculated. The program gives a physical database for radiation dosimetry calculations or other calculations requiring detailed decay data. The principal calculations

performed by EDISTR are the determination of 1) the average energy of beta particles in a beta transition, 2) the beta spectra as a function of energy, 3) the energies and intensities of X rays and Auger electrons generated by radioactive decay processes, 4) the bremsstrahlung spectra accompanying beta decay and monoenergetic Auger and internal conversion electrons, and (5) the radiations accompanying spontaneous fission.

EDISTR program has three phases: 1) the input phase, in which the input data are prepared and put into a suitable format for computational purposes, 2) the computational phase, in which the computations required to implement the theory and empirical methods are completed, and 3) an output phase. For alpha decay, the kinetic energies of the alpha particles and the recoil nuclei are calculated via conservation of energy and momentum principles. The input data involve the ground-state Q value which is the total energy associated with a ground-state to ground-state transition, the different excitation energies of the daughter at which the alpha transitions end, and the corresponding alpha intensities. Beta decay results in a continuous spectrum of beta kinetic energies even when a transition to a single-daughter level is involved. Therefore, beta decay needs more computations. In the EDISTR program, the average energies of the beta particles in the emitted continuous spectrum can be calculated for dosimetry work. Electron capture leads to vacancies in the atomic subshells of the daughter nuclide. The distribution of primary vacancies generated in the different atomic shells and subshells due to the electron-capture process should be calculated using the K/L/M capture ratios because this distribution influences the relative intensities of the X-rays and Auger-electrons from the initial vacancies. Internal conversion of gamma rays is a process by which the transition energy between two states of a nucleus is transferred to an orbital electron. The orbital

electron is released from the atom with energy equal to the transition energy minus the binding energy of the shell from which the electron is released. The distribution of the primary vacancies and the energies and intensities of conversion electrons are calculated. Intensities of X-rays and Auger-electrons are acquired using the numbers of primary vacancies for electron capture or internal conversion. X-ray and Auger-electron energies are also calculated. Spontaneous fission results in various radiations. The fission decay fraction, the number of neutrons per fission, and the mass number and atomic number of the parent nuclide are used to compute intensities and energies for spontaneous fission fragments, neutrons, beta particles, prompt gamma rays, and delayed gamma rays. Bremsstrahlung radiation is the continuous spectrum of electromagnetic radiation produced when charged particles are slowed down by medium. Bremsstrahlung spectra associated with beta particles and monoenergetic conversion and auger electrons are calculated. The energy of the electromagnetic quantum may range from zero up to the kinetic energy of the charged particle. Bremsstrahlung radiation is important in dosimetry for high-end-point-energy beta decay or situation in which the bremsstrahlung radiation accounts for a significant fraction of penetrating radiations. There are two basic types of bremsstrahlung radiation: internal and external. Internal bremsstrahlung takes place when the beta particle is released from a nucleus and is regarded as an inherent radiation from decay process. External bremsstrahlung is due to the interaction of a beta particle with external atoms after its emission from the nucleus. The spectrum of external bremsstrahlung relies on the atomic composition of the external matter. The important external media for dosimetry are air, muscle, fat, and bone. The external bremsstrahlung radiations produced in these four media are calculated (Dillman 1980).

2.4 Decay Data (DEC DAT)

The Decay Data (DEC DAT) directory comprises the codes written to format the EDISTR output file properly and produce DCAL-type nuclear decay data files for inclusion into nuclear decay data library of DCAL system. Four programs (BUILDEM, CHKDAUS, SORTINDEX and TOINDEX) in the DEC DAT are run in a predefined order. The Data File INPUTDAT is used as a template and read by the BUILDEM program. This file tells the code the name and location of the EDISTR output file for the specified radionuclide. The BUILDEM code reads the INPUTDAT template file and picks the EDISTR output file to generate three decay data files (DEC DATA.IDX, DEC DATA.RAD, DEC DATA.BET) in a format usable by DCAL. The CHKDAUS program checks for the inclusion in the INDEX file of all daughter products in the decay chain of interest. This code creates a separate output file named MISSDAUS.DAT in order to indicate missing daughter information. The SORTINDEX code sorts the INDEX file alphabetically by radionuclide name. The TOINDEX program changes the original INDEX file into one that is usable by DCAL and renamed DEC DATA.NDX. The change is to substitute the record number of daughter products for their name in the INDEX file (Eckerman 2001).

2.5 The Dose and Risk Calculation (DCAL) System

The DCAL System is an integrative dosimetric software and numerical database developed at Oak Ridge National Laboratory for the U.S. Environmental Protection Agency (EPA). The DCAL software is used to compute internal and external dose and risk coefficients especially in the computation of age-specific dose coefficients for public.

The software includes a series of computational modules written in FORTRAN and driven either in an interactive or batch mode. The software comprises extensive libraries of biokinetic and dosimetric data so that it can make biokinetic and dosimetric calculations for the acute intake of a radionuclide via inhalation or ingestion at a specified age. DCAL depends on libraries of biokinetic and dosimetric models, nuclear decay data, and anatomic data for internal dosimetric computations which consist of three main steps:

1. Activity module (ACTACAL module): the module computes time-dependent activity for a parent radionuclide and any radioactive progeny in source regions of the body given a unit intake at a specific age. The time-dependent distribution of activity within the body is predicted by a biokinetic model.
2. SEE Calculations (SEECAL module): For the parent radionuclide and every decay chain member included in the ACTACAL calculation, the SEECAL module computes age-specific SEE values for each source and target tissue pair shown in the SEECAL request file created by ACTACAL. These SEE calculations are based on: a) nuclear decay data in the ICRP38.RAD file; b) a library of specific absorbed fractions which are now used by ICRP for non-penetrating radiations and photons; and c) age-dependent organ masses which are based on data from ICRP Publication 23.
3. Dose rate module (EPACAL module): The main job of this module is to combine the time-specific activity calculated by ACTACAL with SEE values for the source-target pairs generated by SEECAL to compute absorbed or equivalent dose rates in target tissues. Choosing equivalent dose option in ACTACAL could generate age-specific dose coefficients which are committed equivalent doses to

organs and committed effective doses per unit intake. Dose coefficients are determined by integrating the equivalent dose rates from the third step.

The systemic biokinetic models and GI absorption fractions (f_1 values) in the DCAL library represent primarily those recommended in recent publications of the ICRP such as ICRP Publication 68 for workers and ICRP Publication 72 for the public. GI absorption fractions (f_1 values) are the fraction of a stable element reaching the body fluids after ingestion. The biokinetic libraries also involve: for respiratory tract kinetics the latest ICRP model of ICRP Publication 66 and the older model of ICRP Publication 30; the ICRP's current gastrointestinal tract model recommended in ICRP Publication 30; and the urinary bladder voiding model in ICRP Publication 67. The biokinetic files are read by the ACTACAL module, which calculates time-dependent activity in the compartments specified in the biokinetic data files.

The nuclear decay library includes nuclear decay data in a file called ICRP38.RAD which represents information for the energies and intensities of radiations from natural nuclear transformation of radionuclides. The file ICRP38.NDX contains indicators to the original record of each radionuclide in the RAD file and other data including the half-life and fraction of the nuclear transformation to generate a radioactive daughter. The file ICRP38.BET comprises the beta spectra for beta emitters.

The batch mode can process a user-defined set of radionuclides for dose coefficient calculations. The input data are controlled by a user-supplied input file and a DOS batch file. A batch input file with extension INP should be generated. A batch input file includes the global data block and the case-specific data block. The global data block shows: 1) the dose type to be calculated; 2) the route of intake; 3) the exposure type for

inhalation; 4) the default particle size for inhalation; 5) the number of ages; and 6) a list of ages at intake. The case input data includes: 1) name of the radionuclide; 2) the length of the decay chain; 3) the assumption concerning the biokinetics of decay chain members; 4) the name of the biokinetic file; 5) the name of the f_1 file; 6) the intake age of the adult if different from the global value; 7) the behavior of the aerosol for inhalation; 8) the exposure type and activity median aerodynamic diameter (AMAD) for inhalation if different from the global values, and 9), the name of the lung file for inhalation if special assumptions are made regarding the respiratory kinetics of a radionuclide. The output dose file with extension GDB or HDB represents ingestion or inhalation respectively. DCAL also calculates dose rate from exposure to external sources distributed in environment. The external computational module EXTCAL calculates the dose coefficients for external exposure to photons and electrons for submersion in contaminated air, exposure to contaminated ground surfaces and volumes (Eckerman et al. 2001).

2.6 Quality Assurance

In order to provide a quality check on the adopted methodology used in our study, some radioisotopes with published dose coefficients were selected and given to various members at the DC working group. Individuals of the group calculated committed effective dose per unit intake for internal dose coefficient and external dose coefficient for air submersion for the following six radionuclides: ^{144}Nd , ^{50}V , ^{38}S , ^{201}Au , ^{41}Ar , and ^{61}Co . The DC results generated by different members were then compared with each other and compared with the published values. The values showed good agreements. The

outcome proves that the methodology was successfully adopted and could be used for dose coefficient calculations.

CHAPTER 2

METHODOLOGY

2.1 Background

A methodology to calculate dose coefficients for radionuclides produced in spallation neutron sources was developed by Omar Wooten of the Georgia Institute of Technology and Tony Andrade of Los Alamos National Laboratory. Mr. Wooten computed dose coefficients for 3-4 radionuclides using this method although several problems existed in this prototype code. Therefore, the methodology has been refined and optimized with the help of the research consortium consisting of representatives from several universities and national laboratories. Finally, the Dose Coefficient Working Group (DC Working Group) developed a reproducible methodology which was adopted from that used to develop Federal Guidance Report 13 and is shown in Fig. 2.1. In this methodology, radiological data is acquired from the Evaluated Nuclear Structure Data File (ENSDF) library maintained by the National Nuclear Data Center at Brookhaven National Laboratory and used as input data for the computer code EDISTR. The input data comprise decay modes, decay energy levels, and radiation energies and intensities, which are processed by the EDISTR code to create a radioactive decay data file. The processed data file contains not only the average or unique energies and intensities of the radiations, but also the beta spectra. The Decay Data (DECDAT) file arranges the output file of EDISTR into the proper format to incorporate into the nuclear decay data libraries within

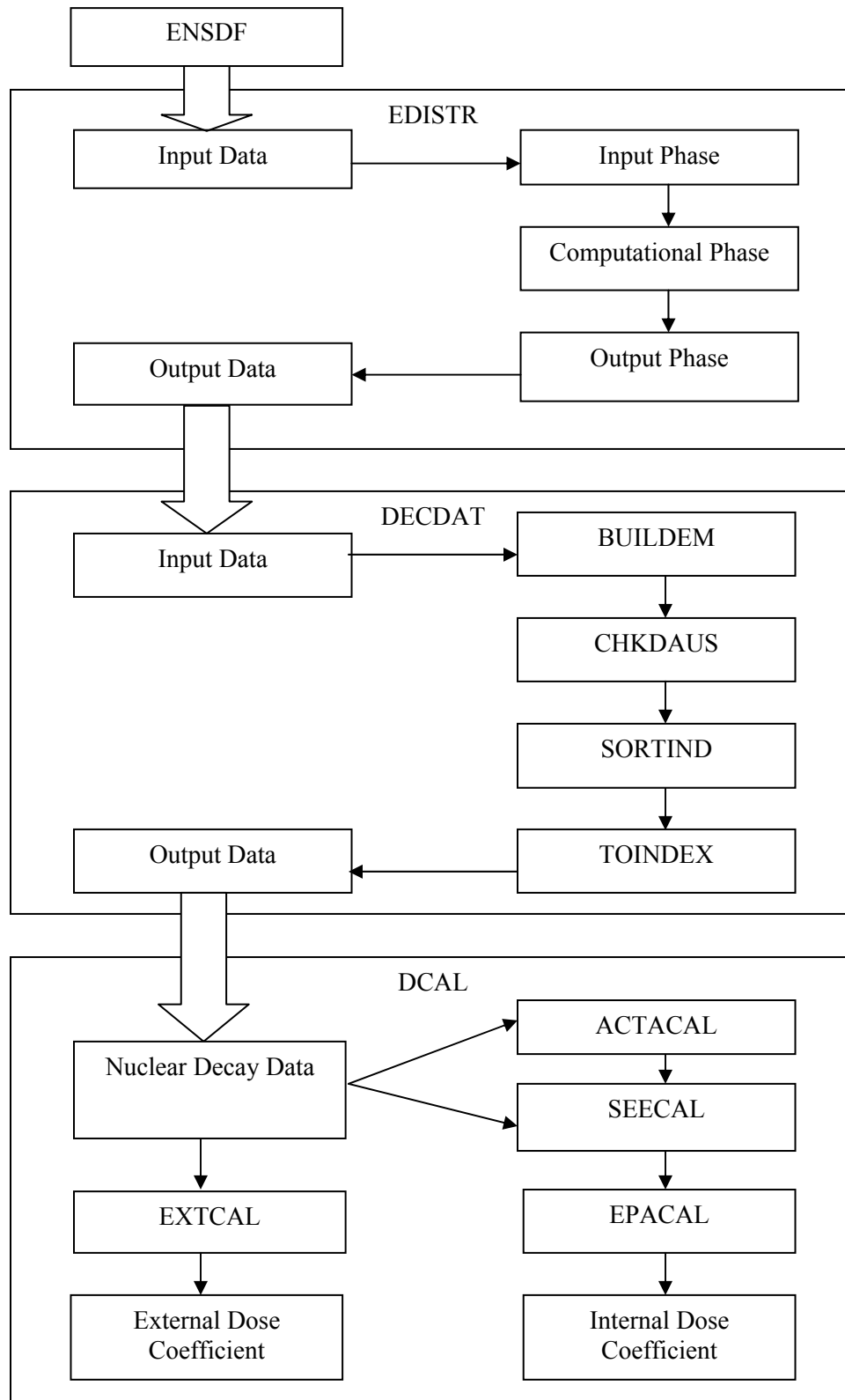


Figure 2.1 Flow chart of computer program for dose coefficient calculations

the Dose and Risk Calculation (DCAL) program developed by the Oak Ridge National Laboratory. DCAL software comprises a series of computational modules and extensive libraries of biokinetic, anatomical, and dosimetric data required for the dose coefficient computation.

2.2 Evaluated Nuclear Structure Data File (ENSDF)

The Evaluated Nuclear Structure Data File (ENSDF) is used to store nuclear structure properties of nuclides and the results from a variety of experiments to derive those properties. ENSDF is composed of a group of data sets, which have one of the following kinds of information: 1) the summary information for a mass chain, 2) the references used in the data sets for the given mass number, 3) the adopted level and gamma-ray properties for each nuclide, 4) the evaluated results of a single type of experiment, e.g., a radioactive decay or a nuclear reaction for a given nuclide, and 5) the combined evaluated results of a number of experiments of the same kind for a given nuclide.

A data set consisting of 80-column records has at least two records: the beginning and the end record. A data set must begin with an IDENTIFICATION record and end with an END record. The IDENTIFICATION record denotes the kind of data in the data set. The END record is blank and just tells the computer the end of the data set has been reached. Between these two records, there could be as many additional records as are required to depict fully the experimental or the evaluated information.

A DECAY data set is the data set that provides the radioactive decay information of a radionuclide. A DECAY data set IDENTIFICATION record includes three parts: the decaying nucleus, the decay type, and the word DECAY. Following the

IDENTIFICATION record is a NORMALIZATION record, and a PARENT record. Following the NORMALIZATION and PARENT records is a group of records, consisting of LEVEL, ALPHA, B-, EC, B+, and GAMMA records, which describe the measured or deduced properties of levels, gamma rays, alpha particles, and etc. Each LEVEL record is followed by a group of B-, EC, B+, or ALPHA records describing charged particle decay into the level and gamma ray out of the level (Tuli 1987).

2.3 EDISTR Computer Program

The computer program EDISTR extracts basic nuclear decay information from the ENSDF and prepares a nuclear decay database for radiation dosimetry computations. As we know for medical and health physics dosimetry calculations, the mean energies and absolute intensities of all the radiations associated with the radioactive decay process should be known. The average energy of the β^+ or β^- particles and the energies and intensities of radiations associated with spontaneous fission must also be known in dosimetry calculations. In addition bremsstrahlung yield and spectral shape information are also needed in a few cases which bremsstrahlung radiation may contribute to dose. The computer program EDISTR calculates intensities and mean energies of alpha, beta, gamma, internal-conversion-electron, x-ray, Auger-electron, and bremsstrahlung radiations resulting from nuclear decay.

The yields of radiations associated with spontaneous fission are computed in a few spontaneous fission cases. Moreover, spectral shapes of the continuous beta distributions are also calculated. The program gives a physical database for radiation dosimetry calculations or other calculations requiring detailed decay data. The principal calculations

performed by EDISTR are the determination of 1) the average energy of beta particles in a beta transition, 2) the beta spectra as a function of energy, 3) the energies and intensities of X rays and Auger electrons generated by radioactive decay processes, 4) the bremsstrahlung spectra accompanying beta decay and monoenergetic Auger and internal conversion electrons, and (5) the radiations accompanying spontaneous fission.

EDISTR program has three phases: 1) the input phase, in which the input data are prepared and put into a suitable format for computational purposes, 2) the computational phase, in which the computations required to implement the theory and empirical methods are completed, and 3) an output phase. For alpha decay, the kinetic energies of the alpha particles and the recoil nuclei are calculated via conservation of energy and momentum principles. The input data involve the ground-state Q value which is the total energy associated with a ground-state to ground-state transition, the different excitation energies of the daughter at which the alpha transitions end, and the corresponding alpha intensities. Beta decay results in a continuous spectrum of beta kinetic energies even when a transition to a single-daughter level is involved. Therefore, beta decay needs more computations. In the EDISTR program, the average energies of the beta particles in the emitted continuous spectrum can be calculated for dosimetry work. Electron capture leads to vacancies in the atomic subshells of the daughter nuclide. The distribution of primary vacancies generated in the different atomic shells and subshells due to the electron-capture process should be calculated using the K/L/M capture ratios because this distribution influences the relative intensities of the X-rays and Auger-electrons from the initial vacancies. Internal conversion of gamma rays is a process by which the transition energy between two states of a nucleus is transferred to an orbital electron. The orbital

electron is released from the atom with energy equal to the transition energy minus the binding energy of the shell from which the electron is released. The distribution of the primary vacancies and the energies and intensities of conversion electrons are calculated. Intensities of X-rays and Auger-electrons are acquired using the numbers of primary vacancies for electron capture or internal conversion. X-ray and Auger-electron energies are also calculated. Spontaneous fission results in various radiations. The fission decay fraction, the number of neutrons per fission, and the mass number and atomic number of the parent nuclide are used to compute intensities and energies for spontaneous fission fragments, neutrons, beta particles, prompt gamma rays, and delayed gamma rays. Bremsstrahlung radiation is the continuous spectrum of electromagnetic radiation produced when charged particles are slowed down by medium. Bremsstrahlung spectra associated with beta particles and monoenergetic conversion and auger electrons are calculated. The energy of the electromagnetic quantum may range from zero up to the kinetic energy of the charged particle. Bremsstrahlung radiation is important in dosimetry for high-end-point-energy beta decay or situation in which the bremsstrahlung radiation accounts for a significant fraction of penetrating radiations. There are two basic types of bremsstrahlung radiation: internal and external. Internal bremsstrahlung takes place when the beta particle is released from a nucleus and is regarded as an inherent radiation from decay process. External bremsstrahlung is due to the interaction of a beta particle with external atoms after its emission from the nucleus. The spectrum of external bremsstrahlung relies on the atomic composition of the external matter. The important external media for dosimetry are air, muscle, fat, and bone. The external bremsstrahlung radiations produced in these four media are calculated (Dillman 1980).

2.4 Decay Data (DEC DAT)

The Decay Data (DEC DAT) directory comprises the codes written to format the EDISTR output file properly and produce DCAL-type nuclear decay data files for inclusion into nuclear decay data library of DCAL system. Four programs (BUILDEM, CHKDAUS, SORTINDEX and TOINDEX) in the DEC DAT are run in a predefined order. The Data File INPUTDAT is used as a template and read by the BUILDEM program. This file tells the code the name and location of the EDISTR output file for the specified radionuclide. The BUILDEM code reads the INPUTDAT template file and picks the EDISTR output file to generate three decay data files (DEC DATA.IDX, DEC DATA.RAD, DEC DATA.BET) in a format usable by DCAL. The CHKDAUS program checks for the inclusion in the INDEX file of all daughter products in the decay chain of interest. This code creates a separate output file named MISSDAUS.DAT in order to indicate missing daughter information. The SORTINDEX code sorts the INDEX file alphabetically by radionuclide name. The TOINDEX program changes the original INDEX file into one that is usable by DCAL and renamed DEC DATA.NDX. The change is to substitute the record number of daughter products for their name in the INDEX file (Eckerman 2001).

2.5 The Dose and Risk Calculation (DCAL) System

The DCAL System is an integrative dosimetric software and numerical database developed at Oak Ridge National Laboratory for the U.S. Environmental Protection Agency (EPA). The DCAL software is used to compute internal and external dose and risk coefficients especially in the computation of age-specific dose coefficients for public.

The software includes a series of computational modules written in FORTRAN and driven either in an interactive or batch mode. The software comprises extensive libraries of biokinetic and dosimetric data so that it can make biokinetic and dosimetric calculations for the acute intake of a radionuclide via inhalation or ingestion at a specified age. DCAL depends on libraries of biokinetic and dosimetric models, nuclear decay data, and anatomic data for internal dosimetric computations which consist of three main steps:

1. Activity module (ACTACAL module): the module computes time-dependent activity for a parent radionuclide and any radioactive progeny in source regions of the body given a unit intake at a specific age. The time-dependent distribution of activity within the body is predicted by a biokinetic model.
2. SEE Calculations (SEECAL module): For the parent radionuclide and every decay chain member included in the ACTACAL calculation, the SEECAL module computes age-specific SEE values for each source and target tissue pair shown in the SEECAL request file created by ACTACAL. These SEE calculations are based on: a) nuclear decay data in the ICRP38.RAD file; b) a library of specific absorbed fractions which are now used by ICRP for non-penetrating radiations and photons; and c) age-dependent organ masses which are based on data from ICRP Publication 23.
3. Dose rate module (EPACAL module): The main job of this module is to combine the time-specific activity calculated by ACTACAL with SEE values for the source-target pairs generated by SEECAL to compute absorbed or equivalent dose rates in target tissues. Choosing equivalent dose option in ACTACAL could generate age-specific dose coefficients which are committed equivalent doses to

organs and committed effective doses per unit intake. Dose coefficients are determined by integrating the equivalent dose rates from the third step.

The systemic biokinetic models and GI absorption fractions (f_1 values) in the DCAL library represent primarily those recommended in recent publications of the ICRP such as ICRP Publication 68 for workers and ICRP Publication 72 for the public. GI absorption fractions (f_1 values) are the fraction of a stable element reaching the body fluids after ingestion. The biokinetic libraries also involve: for respiratory tract kinetics the latest ICRP model of ICRP Publication 66 and the older model of ICRP Publication 30; the ICRP's current gastrointestinal tract model recommended in ICRP Publication 30; and the urinary bladder voiding model in ICRP Publication 67. The biokinetic files are read by the ACTACAL module, which calculates time-dependent activity in the compartments specified in the biokinetic data files.

The nuclear decay library includes nuclear decay data in a file called ICRP38.RAD which represents information for the energies and intensities of radiations from natural nuclear transformation of radionuclides. The file ICRP38.NDX contains indicators to the original record of each radionuclide in the RAD file and other data including the half-life and fraction of the nuclear transformation to generate a radioactive daughter. The file ICRP38.BET comprises the beta spectra for beta emitters.

The batch mode can process a user-defined set of radionuclides for dose coefficient calculations. The input data are controlled by a user-supplied input file and a DOS batch file. A batch input file with extension INP should be generated. A batch input file includes the global data block and the case-specific data block. The global data block shows: 1) the dose type to be calculated; 2) the route of intake; 3) the exposure type for

inhalation; 4) the default particle size for inhalation; 5) the number of ages; and 6) a list of ages at intake. The case input data includes: 1) name of the radionuclide; 2) the length of the decay chain; 3) the assumption concerning the biokinetics of decay chain members; 4) the name of the biokinetic file; 5) the name of the f_1 file; 6) the intake age of the adult if different from the global value; 7) the behavior of the aerosol for inhalation; 8) the exposure type and activity median aerodynamic diameter (AMAD) for inhalation if different from the global values, and 9), the name of the lung file for inhalation if special assumptions are made regarding the respiratory kinetics of a radionuclide. The output dose file with extension GDB or HDB represents ingestion or inhalation respectively. DCAL also calculates dose rate from exposure to external sources distributed in environment. The external computational module EXTCAL calculates the dose coefficients for external exposure to photons and electrons for submersion in contaminated air, exposure to contaminated ground surfaces and volumes (Eckerman et al. 2001).

2.6 Quality Assurance

In order to provide a quality check on the adopted methodology used in our study, some radioisotopes with published dose coefficients were selected and given to various members at the DC working group. Individuals of the group calculated committed effective dose per unit intake for internal dose coefficient and external dose coefficient for air submersion for the following six radionuclides: ^{144}Nd , ^{50}V , ^{38}S , ^{201}Au , ^{41}Ar , and ^{61}Co . The DC results generated by different members were then compared with each other and compared with the published values. The values showed good agreements. The

outcome proves that the methodology was successfully adopted and could be used for dose coefficient calculations.

CHAPTER 3

RESULTS AND DISCUSSION

3.1 Radionuclide Classification

A list of radionuclides expected to be present in the mercury SNS target was provided by Dr. Keith Eckerman from Oak Ridge National Laboratory. Five hundred and twenty four radionuclides were identified as possible byproduct materials from the spallation of a liquid mercury target. According to the three radiation safety databases (ICRP 68, FGR 13 and JAERI), 158 radionuclides were found lacking a published dose coefficient. These radionuclides were prioritized according to half-life, with the highest priority given to those radionuclides with a half-life greater than or equal to one minute. This prioritization scheme was based on an assumed radiological risk associated with an exposure and the computational capabilities of the dosimetry codes.

It was found that 86 out of the 158 radionuclides had a half life less than one minute, 57 had a half life between one and ten minutes, and 15 had a half life greater than ten minutes. Therefore 72 radionuclides were identified in the first priority group. Further refinement of the prioritization scheme evolved from an effort to quantify the accuracy of the input data used in the dosimetry codes. First, whether or not an ENSDF reference exists for the assigned radionuclides and their decay chain members was checked. Second, the accuracy of the ENSDF data was evaluated by comparing its information to

another nuclear decay database NUBASE. Hence the final prioritization of the original list involved an interdatabase comparison to assure the accuracy of the input information.

Mr. John Shanahan of UNLV has developed a nuclear physics database comparison methodology (Shanahan 2003). Relevant nuclear structure and decay parameters compared between ENSDF and NUBASE for the 72 radionuclides and their decay chain members includes: decay mode, branching fraction, half-life, excitation energy of isomers, spin and parity, and Q value (total decay energy). A radionuclide can fall into one of the following three categories based on the comparison results. Category one indicates that each member of a radionuclide associated decay chain has a complete ENSDF data file and shows good agreement between databases for relevant nuclear structure and decay data. Category two indicates that each member of the decay chain has relevant ENSDF data sets but one or more members show poor agreement between the databases. Category three indicates that an ENSDF data file is missing for one or more members of the decay chain. The result of the 72 radionuclides shows: 30 radionuclides fell into category one, 24 radionuclides fell into category two, and 18 radionuclides fell into category three so that DC could not be calculated now.

3.2 Initial Research and Results for Category Two Radionuclides

The internal and external dose coefficients for the category one radionuclides have been calculated by Mr. John Shanahan (Shanahan 2003). The study then focused on category two radionuclides which need additional research to solve the inconsistencies between the databases. The 24 category two radionuclides are given in Table 3.2.1. Twelve of these twenty four radionuclides have a corresponding ENSDF reference with

complete information for all the parameters. These nuclides are: Cs-120, Er-158, Hf-168, Hg-186, Lu-162, Pt-182, Sm-139, Ta-168, Ta-169, Tm-158, Yb-159, and Yb-160. The parameters which require further investigation for each category two nuclide and its relative decay chain members were determined from the previous study. Some radionuclides or their related decay chain members are completely missing ENSDF information for one or more parameters.

Table 3.2.1 Category two radionuclides

Radionuclide	Radionuclide	Radionuclide	Radionuclide
Ba-125	Hg-191	Pt-182	Ta-169
Cs-120	Lu-162	Re-174	Tm-156
Dy-148	Os-175	Re-175	Tm-158
Er-158	Pb-190	Sm-138	W-175
Hf-168	Pb-193	Sm-139	Yb-159
Hg-186	Pr-132	Ta-168	Yb-160

The initial purpose of the further research for these 24 category two radionuclides is to determine the appropriate value for the conflicting or missing nuclear data so that DC calculation could be performed. With this goal all available nuclear databases were utilized to obtain as much information as possible for the inconsistent or missing parameters. The collected information was then compared in order to determine the appropriate or most uniformly accepted value for the parameters which could be used to calculate dose coefficients. The available databases include: Nubase Physics Database

(NUBASE), Evaluate Nuclear Structure Data File (ENSDF), Japan Atomic Energy Research Institute (JAERI) Nuclear Data Center, Korea Atomic Energy Research Institute (KAERI) Nuclear Data Evaluation Laboratory, International Atomic Energy Agency (IAEA) Nuclear Data Center, China Nuclear Data Center (CNDC), Ukrainian Nuclear Data Center (UKRNDC), Lund/LBNL Nuclear Data Search, and Berkeley Laboratory Isotopes Projects.

For example, the radionuclide Ta-168 shown in Table 3.2.2, the branching fraction for positron decay of decay chain member Lu-168m showed poor correlation between ENSDF and NUBASE. The branching fraction information for positron decay of Lu-168m obtained from all other databases is also presented in the table. From the information, the decision was made that 95% is the most uniformly accepted value.

Table 3.2.2. Branching fraction information for Lu-168m

Nuclide Chain:		Ta-168		
Branching Fraction:		Poor Correlation		
Category		2		
Symbol	Lu	Mass Number 168	Atomic Number 71	Isomer
Data File Date:		ENSDF 199407	NUBASE July 2002	percent difference
Branching Fraction:	B+	100	95	5.26
	IT	5	5	0
		Branching fraction	Other database	
	B+	95	JAERI	
	B+	95	KAERI	
	B+	95	UKRNDC	
	B+	95	IAEA	
	B+	95	CNDC	

After carefully investigating all twenty-four category two radionuclides from the available databases mentioned above, the information obtained was still not sufficient to determine the most popular value for all the conflicting or missing parameters required for DC calculations. All the collected data are included in Appendix A and the corresponding databases are documented. This information could be used as references and may be utilized in the future study.

3.3 Further Research and Results for Category Two Radionuclides

In order to make the DC calculations feasible for category two radionuclides with discrepancies between databases unsolved and determine the influence of conflicting nuclear data on the DC results, the Dose Coefficient Working Group finally decided to calculate dose coefficients using both the ENSDF value and NUBASE value for the parameters which have discrepancies. The corresponding two dose coefficient results will be compared. If the percent difference between these two DC results is less than or equal to 5%, the most conservative value will be recommended as the dose coefficient. If the percent difference is more than 5%, a range will be reported for the dose coefficients.

Before calculating the dose coefficients for these radionuclides, a more detailed evaluation of the ENSDF record and the energy balance data from EDISTR output file was performed.

3.3.1 Detailed Record Analysis for ENSDF Input File

Some of the ENSDF decay data sets are incomplete because of the lack of data from experiments. Before DC calculation, the accuracy of the input data from ENSDF used in the dosimetry codes was further evaluated. The ENSDF data files were evaluated to make sure the corresponding data sets had complete records. For each data set, we checked all

the contained records such as IDENTIFICATION, NORMALIZATION, PARENT, LEVEL, ALPHA, B-, EC, B+, GAMMA records, and the information included in each record.

Problems in the data sets must be corrected prior to processing with EDISTR. It was found that the daughter ground level record is a common problem in ENSDF. The half-life information of the ground state of daughter nuclides is missing in many decay data sets. EDISTR uses the half-life value to decide whether the daughter is radioactive or stable. If there is no half-life value at the ground state level of daughter nuclide, EDISTR treats the nuclide as stable. Since half-life is important to produce the decay chain data, it is required to add either the half-life value or the word “STABLE” to the daughter’s ground state level record. This correction insured all members of the whole decay chain would be considered during the computational process of the codes. The comments about these modifications are included in Appendix B. After carefully checking the ENSDF records for all the radionuclides in category two, as indicated in Appendix B, it was finally found that 19 out of the 24 radionuclides had one or more decay chain members missing their corresponding Electron Capture (EC) records. One radionuclide, Dy-148, had one of its decay chain members, Gd-148, missing related ENSDF data file. As a result, it should be placed into category three. The nineteen radionuclides with at least one decay chain member lacking an EC record are presented in Table 3.3.1.

Because of the missing experimental information, the dose coefficient calculations presented a large challenge. As has been stated before for external exposure, we only consider skin dose from electrons due to the short range of electrons in tissue. Therefore we are interested in the impact of lacking EC records on the external dose calculation

because of the short range of electrons. For this purpose, ten example radionuclides with published dose coefficients were selected to evaluate the influence of a missing EC record. These ten radionuclides include: I-125, La-132, Nd-139, Os-180, Pt-191, Ta-172, Tl-195, Xe-120, Xe-127 and Yb-169. The Nuclear Decay library was built using the adopted methodology for the selected radionuclides with all members of interest having or lacking EC records. As a result, two external dose coefficient values were generated for each selected radionuclide with EC record present and removed. External dose coefficients were calculated for air submersion and ground-surface and volume exposures. The two external DC values were compared and the percent error between these two values was calculated. The results are given in Appendix C. The ten example radionuclides could be classified according to the calculated percent error for each external exposure mode and the results are shown in Figure 3.3.1.

Table 3.3.1. Category two radionuclides with absent electron capture record

Radionuclide	Radionuclide	Radionuclide	Radionuclide
Cs-120	Os-175	Re-174	Tm-156
Hf-168	Pb-190	Re-175	W-175
Hg-186	Pb-193	Sm-138	Yb-159
Hg-191	Pr-132	Ta-168	Yb-160
Lu-162	Pt-182	Ta-169	

As indicated by Figure 3.3.1, for external dose coefficients related to air submersion and exposure to soil contaminated to an infinite depth, only two out of these ten

radionuclides have a percent error less than 5%. For ground plane dose coefficients, none of the ten has a percent error less than 5%. For air submersion, two out of the ten have a percent error between 5 to 10%; four radionuclides have a percent error between 10 to 50%; and two have a percent error greater than 50%. For ground surface exposure, two out of the ten radionuclides have a percent error between 5 to 10%; six have a percent error between 10 to 50%; and two have a percent error greater than 50%. For soil volume dose coefficients, three out of ten radionuclides have a percent error between 5 to 10%; three have a percent error between 10 to 50%; and two radionuclides have a percent error greater than 50%.

In summary, the majority of the ten example radionuclides have a percent error greater than 5% for each of the three external exposure modes. These results indicate that the missing electron capture records do have a significant impact on the external dose calculation and this impact can not be ignored. Thus the external dose coefficient can not be determined for those radionuclides with associated decay chain members lacking EC records. Hence nineteen category two radionuclides should be excluded from both internal and external DC calculations because of missing relative EC records in the ENSDF decay data sets. Excluding the nineteen radionuclides and Dy-148, which should be placed into category three, there are only four radionuclides left which are Ba-125, Er-158, Tm-158 and Sm-139.

Here, one thing should be mentioned: for radionuclides with several decay modes, radiations released from some decay modes do not significantly contribute to the dose due to their small branching fractions. In this study, the branching fraction on the order of 10^{-7} or less was neglected for dose calculation. Based on this reason, we have some

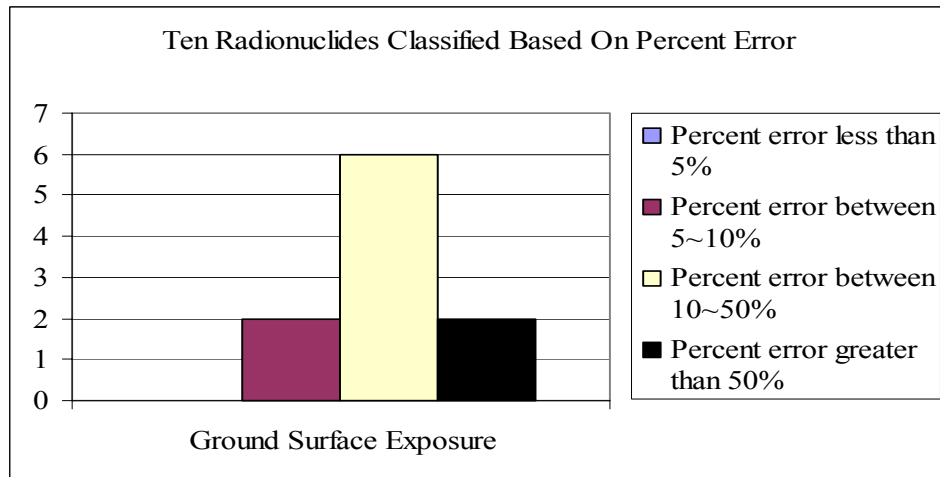
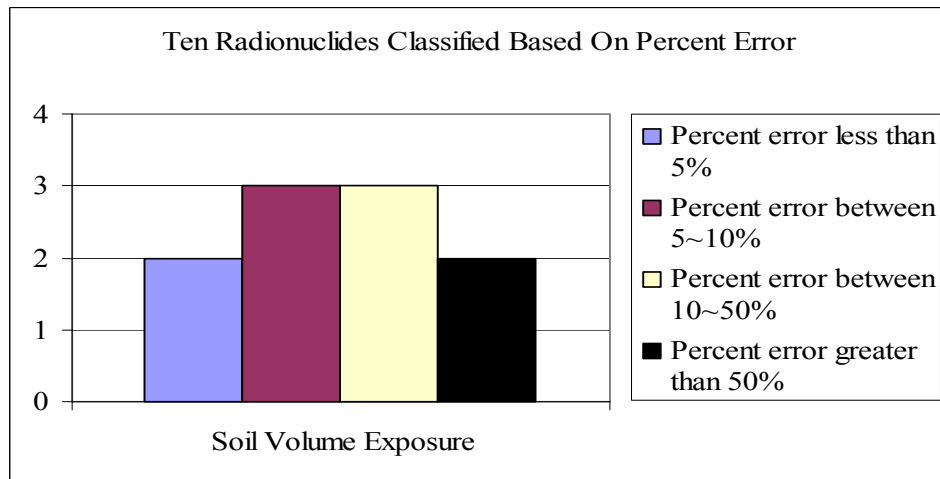
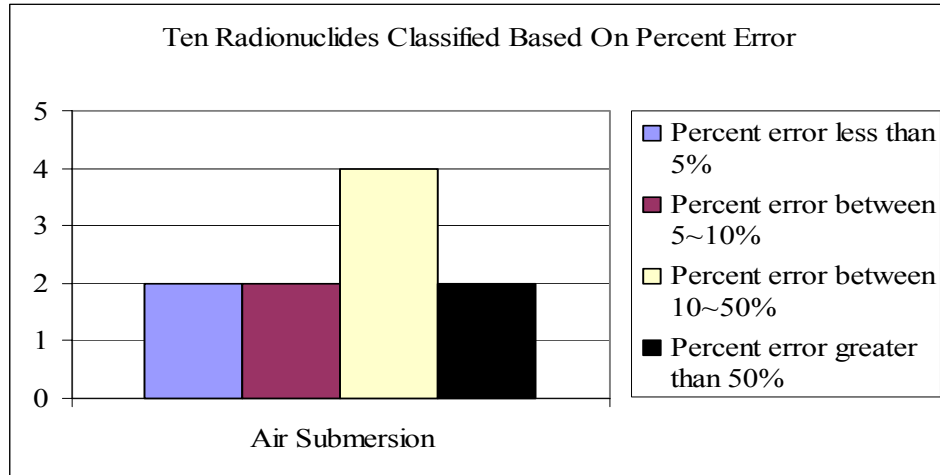


Figure 3.3.1. Ten example radionuclides classified based on percent error calculated for external dose coefficients with and without electron capture records.

special consideration for two radionuclides: Pb-190 and Hg-186. Three decay products (Pt-182, Ir-182 and Os-178) of these two radionuclides lack corresponding EC records in ENSDF data sets. But as indicated in Figure 3.3.2, all three decay products are included in the secondary decay mode (alpha decay) of radionuclide Hg-186, and Hg-186 is the daughter of radionuclide Pb-190 for its secondary decay (alpha decay) mode.

For Pb-190 the branching fraction of the alpha decay is 0.4%, while the branching fraction for its primary decay (electron capture) is 99.6%. Similarly for Hg-186 the branching fraction of its secondary decay (alpha decay) is only 0.016% in contrast to its primary decay (electron capture) with the branching fraction of almost 100%. The three decay chain members with electron capture records missing are all in the alpha decay chain of Hg-186 with the small branching fraction. Though for Hg-186 the value of the branching fraction for alpha decay is not less than the value for which the associated dose from that decay mode could be negligible, the associated dose from alpha decay is rather small compared to the dose from its primary decay with large branching fraction.

Obviously for Pb-190, the contribution to dose from the alpha decay members of Hg-186 is even less significant compared to Hg-186 itself. Without any question the primary decay chain has more dosimetry significance for both Pb-190 and Hg-186. For this reason, the dose coefficients were calculated including all the primary members for Pb-190 and Hg-186, removing the alpha decay chain of Hg-186, which contains three members without an EC record. Thus, even though some experimental data are missing, the relative radiological risk could still be estimated for Pb-190 and Hg-186 based on the internal and external dose coefficients determined for the primary decay mode. Now we have six category two radionuclides for which the DC can be calculated after record

examination for ENSDF input file. These six radionuclides are Ba-125, Er-158, Hg-186, Pb-190, Sm-139 and Tm-158.

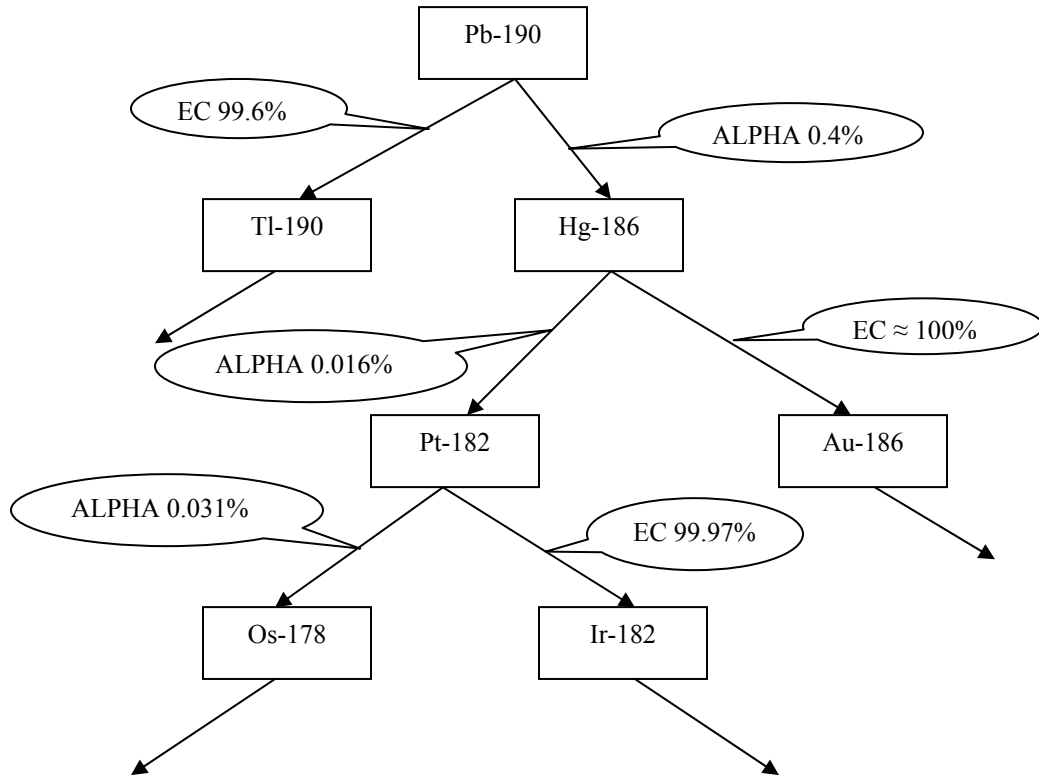


Figure 3.3.2. Decay chain of Pb-190 and Hg-186 with branching fraction of decay mode

3.3.2 Evaluation of EDISTR Output File

EDISTR computes the intensity and energy balance of the decay data sets in order to examine their self-consistency and provides warnings for the imbalance. These warnings include:

Intensity imbalance:

- Total intensity to ground and isomeric states of the daughter differs from 100% by more than 10%.
- Total intensity from the parent differs from 100% by more than 5%.

Energy imbalance:

- Total energy content of all radiations differs from the total decay energy by more than 5% (Endo 2001).

Therefore the energy balance data, radioactive decay data and intensity contained in the EDISTR output file should be evaluated before computing the dose coefficient. The energy balance data is particularly important. Theoretically the total energy available in a decay is equal to the total energy content of all radiations in that decay. So the percent error related to the total energy balance data should be zero. Actually we accept the percent error associated with the energy balance data if the value is less than or equal to 5%.

The EDISTR output file of the six radionuclides mentioned above and their associated decay chain members were checked for energy balance. Three radionuclides (Hg-186, Pb-190 and Sm-139) had percent errors associated with the energy balance data less than 5%. It should be noted for Pb-190 and Hg-186, the energy balance check results do not include all the alpha decay progenies of Hg-186. The other three radionuclides (Ba-125, Er-158 and Tm-158) had an energy imbalance problem. The energy imbalance results for the six radionuclides are given in Table 3.3.2.

Table 3.3.2. Energy imbalance results for six radionuclides

No.	Radionuclide	Decay chain member with energy imbalance	Energy Balance Data (Percent Error %)
1	Ba-125	Ba-125	100
2	Er-158	Ho-158	91.82
3	Hg-186	no	
4	Pb-190	no	
5	Sm-139	no	
6	Tm-158	Ho-158	91.82

3.3.3 Investigation for Energy Imbalance

In an effort to solve the imbalance problem before DC calculations, possible reasons for the imbalance were analyzed. Then proper revision of the input data sets was made based on the analysis. The ENSDF record has been updated by mass chains with a cycle of about six years and can represent a source of error. According to the previous research, the basic nuclear parameters that are especially important for computing energies and intensities of emission were double checked with the most recent version of NUBASE application module as reference (08 Jan 2004). The version of NUBASE used in interdatabase comparison was the 08 July 2002 version. Both ENSDF and NUBASE are based on the 1995 update to the atomic mass evaluation, but NUBASE has its own evaluation, which is updated more frequently. NUBASE is believed to more accurately reflect current scientific literature on nuclear structure and decay data as compared to the ENSDF library.

Decay mode, branching fraction, half-life, excitation energy of isomers, spin and parity, and Q value (total decay energy) were double checked. These parameters are important, because EDISTR uses the excitation energy and Q value to calculate the energies of alpha particles and beta end-point energies of beta particles and also uses the spin and parity to classify the shape of the beta particle spectrum. During the double check, it was found that NUBASE did update some parameter information in its new version. Consequently, the corresponding information was updated in the evaluation worksheet for a relative radionuclide. The goal is to solve the energy imbalance problems based on the fact that NUBASE could probably fill the experimental data gaps in the ENSDF library. Literature sources associated with the radionuclides of interest were also

searched by the Nuclear Science Reference (NSR) database for systematic evaluation of nuclear structure data.

The reason for the energy balance discrepancy was also investigated with the ENSDF analysis and utility programs, FMTCHK, GTOL and LOGFT, distributed from NNDC. These programs generally use ENSDF formatted files as input files. The FMTCHK program checked the format errors of an ENSDF data file to verify that it conforms to “Evaluated Nuclear Structure Data File”. Using the GTOL program, net feedings at the ground and excited levels were calculated from the input gamma ray intensities and internal conversion coefficients and were compared with the input values of the decay data sets. Gamma-ray energies were used to derive a set of least-squares adjusted level energies (Burrows 2003). The LOGFT program computed logft for beta decays. It also calculated the L/K ratio for electron capture decay and the E/B+ ratio for positron decay (NNDC 2001).

According to all these investigations and analyses, possible revisions of the input data sets were made for the three radionuclides with energy balance discrepancies. After the revisions, the decay data sets of the relative radionuclides were processed by the EDISTR program again. The updated EDISTR output data for corresponding radionuclides are shown in Table 3.3.3.

Table 3.3.3. The updated energy imbalance results for three radionuclides

No.	Radionuclide	Decay chain member with energy imbalance	Energy Balance Data (Percent Error %)
1	Ba-125	Ba-125	13.08
2	Er-158	Ho-158	91.82
3	Tm-158	Ho-158	91.82

After analyzing possible reasons of imbalance and making relevant revisions for Ba-125 and Ho-158, the percent error for Ba-125 is now 13.08%. The percent error for Ho-158 is still the same. After analysis, the possible imbalance reason for Ho-158 is the combination of Ho-158m (first isomeric state) and Ho-158 (ground state) for their electron capture decay in one ENSDF decay data set. It's hard to make any changes for this reason without an expert evaluator's suggestion. The problem for Ba-125 is lacking relative experimental data. Our effort is to make a best estimation based on the analysis. The energy balance discrepancy could be corrected if proper ENSDF updates and further experimental information becomes available in the future study. The final results of dose coefficients will definitely be impacted by energy balance discrepancies, but the importance of this inaccuracy is unknown unless additional information can be obtained in the future. For this reason we still calculated the dose coefficients for these three radionuclides: Ba-125, Er-158 and Tm-158.

3.4 Internal and External Dose Coefficient Results

3.4.1 Internal Dose Coefficients

Age-specific internal dose coefficients for ingestion and inhalation were computed by DCAL's batch mode facility for the adult worker and members of the public. The committed equivalent dose coefficients ($h_{T,50}$) and the committed effective dose coefficients (e_{50}) were calculated using both ENSDF and NUBASE databases for the six radionuclides: Ba-125, Er-158, Hg-186, Pb-190, Sm-139 and Tm-158. The systemic biokinetic models and f_1 values rely on models and assumptions specified in ICRP Publication 68 (ICRP 1994) for workers and in Federal Guidance Reports No. 13 (EPA

1999) for the public. With a few exceptional cases, the biokinetic models and f_1 values used in Federal Guidance Reports No. 13 are the same as those used in ICRP Publication 72. The radionuclides in our study are not included in those exceptional cases. As indicated before, f_1 values are the fraction of ingested material absorbed into body fluids (blood) in the absence of radioactive decay. For the public, exposure calculations for inhalation were based on an activity median aerodynamic diameter of $1\mu\text{m}$, but for workers both $1\mu\text{m}$ and $5\mu\text{m}$ were calculated. The activity median aerodynamic diameter (AMAD) is defined as the diameter of a unit density sphere with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol.

The dose coefficients for the public were calculated for six age groups (3 month old, 1-, 5-, 10-, 15- and 20-year olds,) based on the ICRP 56 age groups. For workers only one acute intake age of 20 years (7300 days) was used. For inhalation there are three absorption types according to the respiratory tract model of ICRP Publication 66 (ICRP 1994): Type F materials (fast dissolution and high level of absorption into blood from the respiratory tract), Type M materials (intermediate dissolution rates and level of absorption into blood from the respiratory tract), and Type S materials (slow dissolution and low level of absorption). The dose coefficients for inhalation were determined for each absorption type materials. The results are given in Appendix D.

The radionuclide Hg-186 is a special case. Inhalation dose coefficients were calculated for inorganic mercury, organic mercury and mercury vapour. For inorganic or organic mercury, the adopted lung file is the respiratory tract model of ICRP 66. The special biokinetic files and special f_1 data files are identified as Hg and Hg_org for

inorganic mercury and organic mercury respectively. The corresponding special biokinetic file and special f_1 data file are referred to as Hg for mercury vapour. Ingestion dose coefficients were calculated for inorganic mercury, methyl mercury and unspecified organic mercury compounds. The desired biokinetic files are specified as Hg for inorganic mercury and Hg_org for organic mercury. For organic mercury, the desired f_1 data files are Hg_meth and Hg_org for methyl mercury and unspecified organic compounds respectively.

Ingestion dose coefficients for environmental exposure and internal dose coefficients for occupational exposure using ENSDF databases were also determined by DCAL interactive mode for Ba-125, Er-158, Sm-139, Tm-158 and Pb-190. All the results generated by interactive mode showed perfect agreement with those calculated by batch mode. These results indicate there is no difference for dose coefficients outcome between the two DCAL modes.

After comparison, it is found that internal dose coefficients generated after building the relative Nuclear Decay library based on updating the ENSDF input file with NUBASE values for the six radionuclides showed good agreement with those determined by ENSDF values. The committed effective dose coefficients (e_{50}) generated using both databases were compared and the corresponding percent differences were calculated. The percent differences for e_{50} via inhalation or ingestion for occupational or environmental exposure were all less than 1% for the six radionuclides. All the outcomes are attached in Appendix D.

Table 3.4.1 shows the ingestion dose coefficients (e_{50}) for adults generated by the two database nuclear data. The ingestion dose coefficients ($h_{T,50}$ and e_{50}) of radionuclide Ba-

125 for adults calculated by two database values and the relative percent differences were given in Table 3.4.2 as an example. As indicated by Table 3.4.2, the percent differences related to committed equivalent dose coefficients ($h_{T,50}$) for all the organs of interest were less than 5% for radionuclide Ba-125.

Table 3.4.1. Comparison of ingestion dose coefficients (e_{50}) for adult based on two database nuclear decay data.

Nuclide	Age (day)	f_1	ENSDF $e_{50}(SvBq^{-1})$	NUBASE $e_{50}(SvBq^{-1})$	Percent difference (%)
Sm-139	7300	0.0005	2.04×10^{-11}	2.04×10^{-11}	0.000
Tm-158	7300	0.0005	3.56×10^{-11}	3.56×10^{-11}	0.000
Er-158	7300	0.0005	4.79×10^{-10}	4.79×10^{-10}	0.000
Ba-125	7300	0.1	2.08×10^{-11}	2.08×10^{-11}	0.0961
Pb-190	7300	0.2	8.94×10^{-12}	8.96×10^{-12}	0.223
Hg-186 (inorganic)	7300	0.02	6.74×10^{-12}	6.73×10^{-12}	0.134
Hg-186 (methyl)	7300	1	5.91×10^{-12}	5.90×10^{-12}	0.153
Hg-186 (organic)	7300	0.4	6.47×10^{-12}	6.46×10^{-12}	0.139

Table 3.4.2 Ingestion dose coefficients of radionuclide Ba-125 for adult based on ENSDF and NUBASE data.

$h_{T,50} (SvBq^{-1})$	ENSDF	NUBASE	Percent Difference (%)
Adrenals	1.01×10^{-12}	1.02×10^{-12}	0.197
Bladder Wall	3.33×10^{-13}	3.29×10^{-13}	1.06
Bone Surfaces	4.00×10^{-13}	3.97×10^{-13}	0.832
Brain	1.51×10^{-14}	1.47×10^{-14}	2.44
Breast	2.26×10^{-13}	2.26×10^{-13}	0.221
GI-Tract			
St Wall	1.64×10^{-10}	1.65×10^{-10}	0.243
SI Wall	1.77×10^{-11}	1.77×10^{-11}	0.340
ULI Wall	7.45×10^{-12}	7.25×10^{-12}	2.69
LLI Wall	1.53×10^{-12}	1.48×10^{-12}	3.30
Kidneys	1.05×10^{-12}	1.05×10^{-12}	0.0951
Liver	6.15×10^{-13}	6.15×10^{-13}	0.0163
Resp. Tract			
ET Region	2.99×10^{-14}	2.96×10^{-14}	0.979
Lung	4.02×10^{-13}	4.03×10^{-13}	0.248
Muscle	4.89×10^{-13}	4.88×10^{-13}	0.0615
Ovaries	1.18×10^{-12}	1.17×10^{-12}	0.769
Pancreas	4.49×10^{-12}	4.51×10^{-12}	0.311
Red Marrow	4.99×10^{-13}	4.96×10^{-13}	0.565
Skin	1.84×10^{-13}	1.84×10^{-13}	0.109
Spleen	2.69×10^{-12}	2.70×10^{-12}	0.297
Testes	6.80×10^{-14}	6.73×10^{-14}	1.04
Thymus	1.60×10^{-13}	1.60×10^{-13}	0.188
Thyroid	8.10×10^{-14}	8.10×10^{-14}	0.0741
Uterus	9.43×10^{-13}	9.37×10^{-13}	0.566
Remainder	8.58×10^{-13}	8.57×10^{-13}	0.152
$e_{50} (SvBq^{-1})$	2.08×10^{-11}	2.08×10^{-11}	0.144

3.4.2 External Dose Coefficients

The tissue equivalent dose coefficient $h_{T,ext}$ and the effective dose coefficient e_{ext} were computed for external exposure from submersion in air, radionuclides on ground surface, and radionuclides in the soil volume contaminated to an infinite depth. The external dose

coefficient results based on both ENSDF and NUBASE nuclear data are presented in Appendix E. In analogy to internal dose coefficients, the effective dose coefficients e_{ext} based on ENSDF or NUBASE data were compared and the percent difference were calculated. Accordingly, the percent differences for e_{ext} were equal to zero for submersion, ground plane, and soil volume for all these six radionuclides. The effective dose coefficient (e_{ext}) for submersion calculated from ENSDF or NUBASE nuclear data and the relative percent differences are shown in Table 3.4.3. The submersion dose coefficients ($h_{T,ext}$ and e_{ext}) for radionuclide Sm-139 according to ENSDF or NUBASE nuclear data were given in Table 3.4.4 as an example. As shown in Table 3.4.4, the associated percent differences were equal to zero.

From the comparison, it is indicated that there is no difference between the two external dose coefficient outcomes based on ENSDF and NUBASE respectively.

Table 3.4.3. The effective dose coefficients ($Sv\ per\ Bq\cdot s/m^3$) for air submersion

Nuclide	ENSDF e_{ext_sub}	NUBASE e_{ext_sub}	Percent difference (%)
Sm-139	1.66×10^{-13}	1.66×10^{-13}	0.00
Tm-158	2.01×10^{-13}	2.01×10^{-13}	0.00
Er-158	1.39×10^{-14}	1.39×10^{-14}	0.00
Ba-125	1.25×10^{-13}	1.25×10^{-13}	0.00
Pb-190	9.31×10^{-14}	9.31×10^{-14}	0.00
Hg-186	4.54×10^{-14}	4.54×10^{-14}	0.00

Table 3.4.4 Submersion dose coefficients (*Sv per Bq-s/m³*) for radionuclide Sm-139

$h_{T,ext}$	ENSDF	NUBASE	Percent difference (%)
Red Marrow	6.60×10^{-14}	6.60×10^{-14}	0.00
Adrenals	5.69×10^{-14}	5.69×10^{-14}	0.00
Bone Surface	1.20×10^{-13}	1.20×10^{-13}	0.00
Brain	7.26×10^{-14}	7.26×10^{-14}	0.00
Breasts	7.84×10^{-14}	7.84×10^{-14}	0.00
GB Wall	5.59×10^{-14}	5.59×10^{-14}	0.00
Esophagu	5.65×10^{-14}	5.65×10^{-14}	0.00
St Wall	6.08×10^{-14}	6.08×10^{-14}	0.00
SI Wall	5.44×10^{-14}	5.44×10^{-14}	0.00
ULI Wall	5.54×10^{-14}	5.54×10^{-14}	0.00
LLI Wall	5.66×10^{-14}	5.66×10^{-14}	0.00
Ht Wall	6.02×10^{-14}	6.02×10^{-14}	0.00
Kidneys	6.11×10^{-14}	6.11×10^{-14}	0.00
Liver	6.15×10^{-14}	6.15×10^{-14}	0.00
Lung	6.82×10^{-14}	6.82×10^{-14}	0.00
Ovaries	5.28×10^{-14}	5.28×10^{-14}	0.00
Pancreas	5.40×10^{-14}	5.40×10^{-14}	0.00
Skin	1.56×10^{-13}	1.56×10^{-13}	0.00
Spleen	6.18×10^{-14}	6.18×10^{-14}	0.00
Testes	6.88×10^{-14}	6.88×10^{-14}	0.00
Thymus	6.37×10^{-14}	6.37×10^{-14}	0.00
Thyroid	7.01×10^{-14}	7.01×10^{-14}	0.00
UB Wall	5.71×10^{-14}	5.71×10^{-14}	0.00
Uterus	5.31×10^{-14}	5.31×10^{-14}	0.00
Muscle	6.67×10^{-14}	6.67×10^{-14}	0.00
$h_{T,ext_remaind}$	2.07×10^{-12}	2.07×10^{-12}	0.00
e_{ext}	1.66×10^{-13}	1.66×10^{-13}	0.00

CHAPTER 4

CONCLUSION AND FUTURE RESEARCH

The twenty-four category two radionuclides have been studied for dose coefficient calculations. The initial research was to investigate all available databases to collect information. But the data obtained were still not adequate to determine the proper value for all the conflicting or missing parameters required for DC calculations. The collected data could be utilized as references in the future work. The internal and external dose coefficients were finally calculated for six radionuclides using both the ENSDF value and NUBASE value for the parameters which have discrepancies. The corresponding two dose coefficient results were compared.

In order to quantify the accuracy of the input data, an additional examination was performed for the ENSDF record and the EDISTR output file. Consequently, nineteen category two radionuclides were found to be missing relative electron capture records in their ENSDF data sets. One radionuclide lacked an ENSDF data file and was moved to category three. Based on the results from ten example radionuclides with published dose coefficients, the impact of missing electron capture records on the external dose calculation can not be neglected.

Considering the small branching fraction of the secondary decay chain which contains progenies lacking EC records for Pb-190 and Hg-186, only the primary decay members of these two radionuclides were taken into account, excluding the alpha decay

chain of Hg-186 for DC computations. Therefore dose coefficients were determined for six category two radionuclides: Ba-125, Er-158, Hg-186, Pb-190, Sm-139 and Tm-158. The DC results of three radionuclides (Ba-125, Er-158 and Tm-158) with energy balance problems are worth investigating in the future.

In review, internal and external dose coefficient calculations were performed for six category two radionuclides using both ENSDF and NUBASE values for the conflicting nuclear data. The results indicated the internal and external DC values determined by ENSDF nuclear data showed good agreement with those determined by NUBASE data. That means the conflicting data between the two databases for the six radionuclides do not have a significant impact on the dose coefficient outcomes. The most conservative value will be recommended as the dose coefficient. It is necessary to generate more experimental data in future research to fill the data gaps in the ENSDF library. It will be useful to have expert evaluators to evaluate and update the ENSDF data sets more frequently. Thus, the accuracy of the input data can be improved and the corresponding imbalance problem can be solved. The impact from energy balance discrepancy on the DC results can be evaluated. Based on more experimental information released in future studies, DC calculations can be performed for more radionuclides in categories two and three.

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