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Dose coefficients for radionuclides produced in a spallation neutron source

John Patrick Shanahan
University of Nevada, Las Vegas

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UMI®
DOSE COEFFICIENTS FOR RADIONUCLIDES PRODUCED IN A
SPALLATION NEUTRON SOURCE

by

John Patrick Shanahan

Bachelor of Science
State University of New York at Buffalo
1993

A thesis submitted in partial fulfillment
of the requirements for the

Master of Science Degree in Health Physics
Department of Health Physics
College of Health Sciences

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Master of Science

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ABSTRACT

Evaluation of Nuclear Decay Chains and Calculation of Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source

by

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Internal and external dose coefficient values have been calculated for 14 anthropogenic radionuclides which are not currently presented in Federal Guidance Reports No. 11, 12, and 13 or Publications 68 and 72 of the International Commission on Radiological Protection. Internal dose coefficient values are reported for inhalation and ingestion of 1 μm and 5 μm particulates along with the $f_i$ values and absorption types for the adult worker. Internal dose coefficient values are also reported for inhalation and ingestion of 1 μm particulates as well as the $f_i$ values and absorption types for members of the public. Additionally, external dose coefficient values for air submersion, exposure to contaminated ground surface, and exposure to soil contaminated to an infinite depth are also presented.

Information obtained from this study will be used to support the siting and licensing of future accelerator-driven nuclear initiatives within the U.S. Department of Energy complex, including the Spallation Neutron Source (SNS) and Accelerator Production of Tritium (APT) Projects.
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CHAPTER 1

INTRODUCTION

1.1 Background

High intensity proton accelerators have been developed for the production of intense and pulsed neutron beams for basic scientific research and for the development of transmutation technology of long-lived transuranic nuclides. At these facilities, components of the accelerator system such as the target or blanket are subjected to high energy particle bombardment resulting in the production of various radionuclides by a variety of nuclear reactions. These radionuclides have the potential to be involved in both internal and external exposure scenarios involving workers. Quantifying the radiological health risks to workers and the general public from the production of these radionuclides will be essential for radiation safety and protection.

In general, the term dose, as it applies to radiation dosimetry, denotes the quantity of energy absorbed in a medium per unit mass. For radiation protection purposes it must be appropriately qualified. If unqualified, it refers to absorbed dose, $D$, and can be calculated from (ICRU 1993):

$$D = \frac{d\bar{E}}{dm} \quad \text{(Gy)},$$  \hspace{1cm} (1.1a)
where $dE$ is the mean energy imparted by ionizing radiation to mass, $dm$, of the medium. Because the biological effects of ionizing radiation depend not only on the amount of energy absorbed but the type of radiation involved, the dosimetric quantity of the equivalent dose is employed for radiation protection purposes. The equivalent dose, $H_T$, in a tissue or organ is given by (ICRP 1991):

$$H_T = \sum_R w_R D_{T,R} \text{ (Sv)}, \quad (1.1b)$$

where $D_{T,R}$ is the mean absorbed dose in the tissue or organ, $T$, due to radiation $R$, and $w_R$ is the corresponding radiation weighting factor. The equivalent dose is therefore a measure of the risk associated with a given exposure to a specific type of ionizing radiation. Risks due to exposures to different types of radiations can be directly compared in terms of equivalent dose. If the exposure to ionizing radiation for organ or tissue, $T$, is not uniform, as is the case when radionuclides are preferentially deposited in different body tissues, one uses the effective dose. The effective dose expresses the risk from an exposure of a single organ or tissue in terms of the equivalent risk from an exposure of the whole body being uniformly irradiated. The effective dose, $E$, is calculated from (ICRP 1991):

$$E = \sum_T w_T H_T \text{ (Sv)}, \quad (1.1c)$$

where $H_T$ is the equivalent dose in organ or tissue, $T$, and $w_T$ is the corresponding tissue weighting factor obtained from ICRP report 60 (1991). The tissue weighting factor is a dimensionless number that expresses the relative sensitivity of the given organ or tissue to radiation.

From the standpoint of radiation protection, radiation doses from ionizing radiation are classified as either internal or external, however, both contribute to an
individual's total dose. Internal radiation dosimetry deals with radionuclides that are accidentally ingested or inhaled and deposit ionizing energy into organs or tissues of the body. External radiation dosimetry deals with radiation originating outside the body with sufficient penetrating power to traverse overlying tissues and deposit ionizing energy within the body. By law, employers are required to initiate a radiation protection monitoring program if an employee can receive a radiation dose in excess of 10% of the established limits. There are a number of strategies that an employer may adopt to meet this requirement and they depend largely on the type of work their employees are performing. Monitoring programs provide radiation safety personal numerical data from which to ascertain an individual's radiation dose after an exposure has occurred. However, in planning work with radioactive materials it is often necessary to have an idea of the radiation dose a worker may receive in order to provide adequate radiation protection. It is also desirable, in situations involving an accidental exposure, to be able to immediately estimate a worker's radiation dose. Applying dose coefficients in these situations assist radiation safety personal in calculating the dose an individual may receive and the potential health risks associated with that exposure.

In this work several dosimetry codes are used to compile internal and external dose coefficients for radionuclides produced in a spallation neutron source (SNS) that are not published in current radiation safety standards. The theory used to calculate dose coefficients for occupational exposures follow recommendations set forth by the ICRP as well as previously developed dosimetric methodologies found in Federal Guidance Reports No. 11 and 12. The dosimetry codes adopted for this work were also used in the development of Federal Guidance Reports No. 11 and 12 and will be briefly described.
1.2 Calculating Dose

In the United States radiation protection programs are administered by the Environmental Protection Agency (EPA) through federal guidance approved by the President. Current guidance is consistent with recommendations set forth by the ICRP and was adopted by the EPA in 1987. This guidance provides Federal agencies with the necessary tools to develop and implement their own regulatory standards. Under this guidance, the EPA states that a system of dose limitations is provided which is based on the following principles: (1) Justification – There should not be any occupational exposure of workers to ionizing radiation without the expectation of an overall benefit from the activity causing the exposure; (2) Optimization – A sustained effort should be made to ensure that collective doses, as well as annual, committed, and cumulative lifetime individual doses, are maintained as low as reasonably achievable (ALARA), economic and social factors being taken into account; and (3) Limitation – Radiation doses received as a result of occupational exposure should not exceed specified limiting values (Eckerman et al. 1988). This guidance provides for two types of limits. These limits are: (1) the primary guides which are expressed in terms of limiting doses, and whose primary objective is to minimize the risk of stochastic effects (effects that occur randomly) and to prevent the occurrence of non-stochastic, or deterministic (effects with thresholds whose magnitude increases with the size of the dose) effects from ionizing radiation; and (2) the derived guides which are expressed in terms of quantities or concentrations of radionuclides and are chosen to insure radiation doses do not exceed the primary guide. Implementing the primary guides is largely accomplished through the use of regulations based on derived guides (Eckerman et al. 1988).
Environmental Protection Agency (EPA) Federal Guidance Report No. 11 "Limiting Values of Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion, and Ingestion", developed two derived guides, Annual Limit on Intake (ALI) and the Derived Air Concentration (DAC), to be used to control radiation exposure in the workplace. The ALI is the annual intake of a radionuclide which would result in a committed effective dose equivalent of 50 mSv yr\(^{-1}\) for stochastic effects, or a committed equivalent dose to an individual organ or tissue of 500 mSv yr\(^{-1}\) for non-stochastic effects, to Reference Man (ICRP 1975). A DAC is that concentration of a radionuclide in air which, if breathed by Reference Man for a work-year, would result in an intake corresponding to its ALI (Eckerman et al. 1988). Therefore, ALIs and DACs can be used for assessing radiation doses due to accidental ingestion and inhalation of radionuclides and are used for limiting radionuclide intake through breathing of, or submersion in, contaminated air.

In addition to determining ALIs and DACs, in many situations it is useful to know the committed equivalent dose to an organ or tissue per unit intake \((h_{T,50})\), the committed effective dose per unit intake \((e_{50})\), the tissue dose equivalent per unit time-integrated exposure to a radionuclide \((h_{T,\text{ext}})\) from external exposure, or the effective dose per unit time-integrated exposure to a radionuclide \((e_{\text{ext}})\) from external exposure. These are collectively referred to as dose coefficients, and give either the equivalent dose to a tissue or the effective dose to an individual that is characterized adequately by reference man (ICRP 1975). Tabulated dose coefficients for the 825 radionuclides listed in Publication 38 of the ICRP (1983) are found in both Federal Guidance Reports No. 11 and 12. Federal Guidance Report No. 11 reports dose coefficients (dose conversion factors) for
inhalation, ingestion, and submersion in contaminated air scenarios. While Federal Guidance Report No. 12 reports dose coefficients for immersion in contaminated water, exposure to contaminated soil, and updates Federal Guidance Report No. 11 with respect to dose coefficients for submersion in contaminated air.

1.2.1 Internal Dose Coefficients Methodology

In internal radiation dosimetry programs the risk of a given biological effect is assumed to relate linearly to the equivalent dose. The risk of an effect is determined by the total equivalent dose averaged throughout the organ or tissue, and is independent of the time in which the equivalent dose is delivered. The intake of certain long-lived radionuclides may result in the continuous deposition of dose to tissues far into the future. To account for this fact in planning work with radioactive materials, the ICRP recommends that the appropriate period for integration of equivalent dose is a working life time of 50 years. The committed equivalent dose, \( H_{T,50} \), to a given organ or tissue from a single intake of radioactive material into the body is defined as the integrated equivalent dose accumulated over the next 50 years from that intake, and can be calculated from (Eckerman et al. 1988):

\[
H_{T,50} = K \sum_{S} U_{S} \text{SEE}(T \leftarrow S) \quad \text{(Sv).} \tag{1.2.1a}
\]

The constant, \( K \), depends on the units specified for \( H_{T,50} \), \( \text{SEE}(T \leftarrow S) \), and \( U_{S} \). In the ICRP methodology, \( K \) is equal to \( 1.6 \times 10^{-10} \text{ Sv g MeV}^{-1} \) when \( \text{SEE}(T \leftarrow S) \) is expressed in megaelectron volts (MeV) per gram (g) per nuclear transformation, and \( U_{S} \) in nuclear transformations. The specific effective energy, \( \text{SEE}(T \leftarrow S) \), depends on the details of the nuclear transformations of the radionuclide, including the weighting factors of the emitted radiations, and the distribution of absorbed energy among body tissues.
Computation of $U_S$ reflects the metabolic activity of a radionuclide in the body. Models such as the “Dosimetric Model for the Gastrointestinal Tract” (ICRP 1979) and the “Human Respiratory Tract Model for Radiation Protection” (ICRP 1994) are used to facilitate these calculations and are based on the assumption that the body consists of a number of separate compartments (ICRP 1979). Details of the uptake, distribution, and retention of a particular radionuclide into the body or body tissues are given in the metabolic data for a given element, while models are used to describe its translocation and clearance (biokinetics) from the body.

The committed effective dose, $E_{50}$, reflects both the distribution of dose among various tissues of the body and the relative sensitivity of those tissues to the stochastic effects of ionizing radiation (Eckerman et al. 1988). The committed effective dose is calculated from:

$$E_{50} = \sum_r w_r H_{r,50} \text{ (Sv)}, \quad (1.2.1b)$$

where $w_r$ is the tissue weighting factor and equates the risk of cancer induction in a single irradiated tissue or organ to the risk of cancer induction if the whole body were uniformly irradiated. For occupational exposures the ICRP recommends the values of $w_r$ shown in Table 1.2.1.

Dose coefficient calculations, for internal dosimetry applications, require unit activity of a given radionuclide be used in Eq. 1.2.1a to calculate the committed equivalent dose per unit intake for a specific organ or tissue. Accordingly, when individual organ and tissue committed equivalent doses are summed after applying the appropriate tissue weighting factors the result is the committed effective dose per unit intake.
Table 1.2.1. ICPR 60 Tissue Weighting Factors for Stochastic Effects

<table>
<thead>
<tr>
<th>Organ or Tissue</th>
<th>$w_T$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gonads</td>
<td>0.20</td>
</tr>
<tr>
<td>Breast</td>
<td>0.05</td>
</tr>
<tr>
<td>Colon</td>
<td>0.12</td>
</tr>
<tr>
<td>Red bone marrow</td>
<td>0.12</td>
</tr>
<tr>
<td>Lung</td>
<td>0.12</td>
</tr>
<tr>
<td>Stomach</td>
<td>0.12</td>
</tr>
<tr>
<td>Urinary Bladder</td>
<td>0.05</td>
</tr>
<tr>
<td>Liver</td>
<td>0.05</td>
</tr>
<tr>
<td>Esophagus</td>
<td>0.05</td>
</tr>
<tr>
<td>Thyroid</td>
<td>0.05</td>
</tr>
<tr>
<td>Bone Surfaces</td>
<td>0.01</td>
</tr>
<tr>
<td>Skin</td>
<td>0.01</td>
</tr>
<tr>
<td>Remainder</td>
<td>0.05</td>
</tr>
</tbody>
</table>

1.2.2 External Dose Coefficients Methodology

In contrast to internal radiation dosimetry, external radiation dosimetry deals with photons and electrons emitted by radionuclides outside the body with sufficient energy to penetrate overlying tissues and deposit their energy internally. As a result, the need to account for the continuous deposition of energy far into the future from the intake of a radionuclide into the body (and the required 50 year period of integration of the equivalent dose) is no longer applicable. Also, from the definition of the external dose coefficient, the time integrated concentration of a radionuclide is used in the denominator instead of unit activity, thus

$$h_{T, ext} = \frac{H_T}{\int C(t) dt} \quad \text{(Sv per Bq s m}^{-3})$$  \quad (1.2.2a)
Therefore, the tissue equivalent dose, $H_T$, and the effective dose, $E$, now become the quantities of interest in evaluating an external radiation dose to a worker for a specific control period.

According to Federal Guidance Report No. 12 calculations of external dose coefficients involve three major steps: (1) computation of the energy and angular distributions of the radiation incident on the body for a range of initial energies of monoenergetic sources distributed in environmental media, (2) evaluation of the transport and energy deposition in organs and tissues of the body of the incident radiations, characterized above in terms of their energy and angular distributions, for each of the initial energies considered, and (3) calculation of the organ or tissue dose for specific radionuclides, considering the energies and intensities of the radiations emitted during nuclear transformations of those nuclides. The result of the first two steps is a set of dose coefficients for monoenergetic sources of photon or electron radiations. The last step simply scales these coefficients to the emissions of the radionuclide of interest (Eckerman et al. 1993).

With respect to steps one and two, Federal Guidance Report No. 12 reports that the estimation of dose to tissues of the body from radiation emitted by an arbitrary distribution of a radionuclide in an environmental medium is an extremely difficult computational task and requires solution to a complex radiation transport problem involving radiations incident on and through the body. As a result, it becomes impractical to solve this problem for the precise spectrum of photons emitted by each radionuclide of interest. Therefore, organ doses for 25 organs in an adult hermaphrodite phantom were computed using various codes for monoenergetic photon sources at 12 energies ranging
from 0.1 to 5.0 MeV. The results are tabulated in various look up tables found in Federal Guidance Report No. 12 for each source, $S$, and are utilized by interpolating photon energy data specific to the radionuclide of interest to obtain the equivalent dose for the organ or tissue of interest. Additionally, the skin dose from environmental electron sources represents a complex radiation transport problem. Skin dose coefficients were calculated for a series of monoenergetic electron emissions that were convoluted to the spectra of the various radionuclides found in ICRP Publication 38 (1983) using the energy and intensity data of the beta and electron emissions. It should be noted that the electron dose to organs and tissues of the body other than the skin are negligible, due to the short range of electrons. These results were also tabulated for each source, $S$, and are presented graphically in various look up charts found in Federal Guidance Report No. 12. Obtaining the skin dose coefficient for the radionuclide of interest then becomes a matter of integrating energy, $E$, between $E$ and $E + dE$ for the continuous spectrum (Eckerman et al. 1993).

Finally, an external dose coefficient, $h^x_T$, for any tissue $T$ for any exposure mode $S$ can be expressed as (Eckerman et al. 1993):

$$ h^x_T = \sum_{j=0}^{\infty} \sum_{i} y_{ij}(E_i) \hat{Y}^x_{ij}(E_i) + \int_{0}^{\infty} y_{ij}(E) \hat{Y}^x_{ij}(E) dE $$  \hspace{1cm} (Sv per Bq s m$^{-3}$), \hspace{1cm} (1.2.2b)

where $y_{ij}(E)$ is the yield of discrete photon radiations of type $j$ and energy $E_i$, and $y_{ij}(E)$ denotes the yield of continuous electron radiations per nuclear transformation with energy between $E$ and $E + dE$. The summation is performed over all photon and electron radiations. Note that each radiation potentially has two components: (1) the discrete energy emission, and (2) the continuous emissions. The continuous component is only
accounted for when calculating the tissue dose equivalent for the skin and can be effectively ignored in all other tissue dose coefficient calculations. The contribution of the radiations to the dose in tissue $T$ is defined by the quantity $\hat{h}^2(E)$ which is tabulated as a function of energy for tissue $T$ for each exposure mode $S$ and obtained from the various look up tables previously described. The modes of exposure described here are for: (1) submersion in a contaminated atmospheric cloud, (2) immersion in contaminated water, and (3) exposure to contamination on or in the ground (Eckerman et al. 1993).

1.3 Transmutation Research Program

The 2001 annual report of the University of Nevada, Las Vegas (UNLV) Transmutation Research Program (TRP) states that over 20% of the electricity generated in the United States is provided by nuclear power reactors. It is also estimated that the amount of used nuclear fuel in the United States will reach 140,000 tons by the end of the operational period of current reactors (Hechanova et al. 2001). The United States is currently pursuing a waste management strategy of placing spent nuclear fuel in deep geologic repositories. This waste management strategy separates the nuclear waste from the biosphere and allows long-lived radionuclides to decay to more manageable daughter products. Transmutation is an alternative waste management strategy undergoing research and development in the United States in partnership with other countries. Transmutation changes one radionuclide into another with more favorable characteristic properties by altering its nuclear structure. Nuclear structure is defined by the number of protons and neutrons in the nucleus. Transmutation can be accomplished with two different processes: (1) neutron induced fission, or (2) neutron capture. Both processes
start with the target nucleus absorbing neutrons. Depending on the energy of the incident neutron the target nucleus will either undergo nuclear fission (splitting) or radioactive decay. Both processes lead to the same final result: the altering of the original isotopes nuclear structure and the transmutation of waste.

Transmutation as an alternative waste management strategy was authorized by Congress in the Fiscal Year 2001. The goals of the national program are to: (1) develop and demonstrate transmutation of civilian used nuclear fuel, (2) provide a test bed to conduct nuclear fuel science and material engineering research and development, (3) provide capability of producing tritium for the nation's nuclear stockpile, and (4) provide capability of producing other isotopes for civilian and defense needs. The UNLV Transmutation Research Program was established in March 2001 as part of the national Advanced Accelerator Application (AAA) program to develop the technologies necessary for the ecological and economical treatment of spent nuclear fuel. The goal of this current research project is to compliment the advancement of transmutation technology as it relates to the use of radioactive materials in the workplace.
CHAPTER 2

METHODS AND MATERIALS

2.1 Introduction

The Department of Health Physics has been tasked by the UNLV Transmutation Research Program to develop the methodology necessary for the calculation of dose coefficients for radionuclides produced in spallation neutron sources. In the first year of this multi-year study, a research consortium consisting of members from participating universities and national laboratories was established. This research consortium, the UNLV Dose Coefficient Working Group, was formed to implement the goals and objectives underlining the consortium efforts. The first year goals and objectives included: (1) the development of a methodology to calculate dose coefficients, (2) developing a methodology to identify and prioritize the radionuclides given to the Dose Coefficient Working Group for a dose coefficient evaluation, and (3) instituting a quality assurance (QA) program to begin to assess the effectiveness of the adopted dose coefficient calculation methodology.

Working closely with one of the principal authors of Federal Guidance Reports No. 11 and 12, Dr. Keith Eckerman from Oak Ridge National Laboratory (ORNL), the Working Group adopted the methodology used to calculate dose coefficients in these Federal Guidance Reports. Implementing this methodology required the use of several
dosimetry codes and much of the first year of the study was spent with Working Group members familiarizing themselves with the use of these codes.

Five hundred and twenty four radionuclides, based on a mercury SNS target, were given to the Working Group for a dose coefficient evaluation. The initial list was provided by the SNS Group at ORNL. The identification of radionuclides lacking a published dose coefficient was accomplished by comparing the initial list to three existing radiation safety dose coefficient databases. The databases utilized in this analysis included: (1) International Commission on Radiological Protection (ICRP) Database of Dose Coefficients: Workers and Members of the Public, CD Supplement, Version 2.0.1 (ICRP 2001), (2) Federal Guidance Report 13: Cancer and Risk Coefficients for Environmental Exposure to Radionuclides, CD Supplement, EPA-402-C-99-001, Rev 1 (EPA 2002), and (3) Dose Coefficients for Radionuclides Produced in High Energy Proton Accelerator Facilities: Coefficients for Radionuclides not Listed in ICRP Publications (JAERI-Data/Code 2002-013). Although the Japan Atomic Energy Research Institute (JAERI) database was included in this work it is not generally recognized internationally as an established radiation safety database. ORNL has a collaborative effort underway under the auspices of the U.S. EPA with the JAERI for the processing of nuclear decay data using the Energy Distribution (EDISTR) code. The EDISTR code is also used in this work to generate a radioactive decay database; therefore, the JAERI database was included in our research so that work would not be duplicated.

The list of radionuclides was initially prioritized according to half-life, with the highest priority given to those radionuclides with a half-life greater than or equal to one
This prioritization scheme was based on an assumed radiological risk associated with an exposure and the computational capabilities of the dosimetry codes. Further refinement of the prioritization scheme evolved from an effort to quantify the accuracy of the input data used in the dosimetry codes. In this work, nuclear decay and structure data files in the Evaluated Nuclear Structure Data File (ENSDF) library maintained by the National Nuclear Data Center (NNDC) at Brookhaven National Laboratory (BNL) serve as input files for the EDISTR code. The information in the ENSDF library is updated by mass chains with a present cycle time of approximately six years, and can represent a source of error relative to current scientific literature. Therefore, the final prioritization of the initial list included a nuclear physics database comparison of relevant nuclear structure and decay data for both the radionuclide of interest, and associated decay chain members, to determine the accuracy of this information before it was utilized in a dose coefficient computation. It should be noted that a similar approach was employed by the JAERI as they prepared radionuclides for possible dose coefficient computation and this methodology served as a template for this work. In the JAERI methodology, ENSDF values of half-life, branching fraction, excitation energy, and total reaction energy (Q-value) were updated when these values differed by more that 1% from those found in Nubase. Nubase is a nuclear physics application module found in the Nucleus software package which gives experimentally known and extrapolated nuclear properties for some 3000 nuclides in the ground-state, and when applicable isomeric-states (Audi 2002). Additionally, ENSDF spin and parity values were also revised and updated according to Nubase if any differences were found between the databases. In the JAERI study a
branching fraction on the order of $10^{-7}$ or less was assumed to be insignificant to the dose and thus was ignored for computational purposes (Endo 2001).

The general scheme of the input data, dosimetry codes, and data flow through the dosimetric system applied in this work is given in Fig. 2.1. The computation of a dose coefficient begins with an ENSDF data file as the input file. Once the appropriate ENSDF data file has been selected, the file is downloaded into an input directory within the EDISTR code. The primary objective of the EDISTR code is to extract relevant nuclear structure and decay information from the ENSDF file for the purpose of generating a radioactive decay data file. The EDISTR output file contains the necessary dosimetric data needed to perform a dose coefficient calculation, and is ultimately used by the computational modules within the Dose and Risk Calculation (DCAL) software package. Before the EDISTR output file can be utilized in the computation of a dose coefficient it must be properly formatted for use by DCAL. To facilitate this formatting requirement, a series of MS-DOS executables were developed and can be found in the Decay Data (DECDAT) directory. Files that have been appropriately formatted are then incorporated into DCAL's Nuclear Decay library for a dose coefficient computation. The DCAL software package contains a series of modules or subroutines necessary for the computation of a dose coefficient calculation.

The purpose of this work is to: (1) identify which of the 524 radionuclides given to the UNLV Dose Coefficient Working Group do not have published dose coefficients, (2) determine whether or not an ENSDF reference exists for each of the radionuclides identified as not having a published dose coefficient as well as for any associated decay chain members, (3) determine the accuracy of the data contained within the ENSDF
library by comparing this information to another nuclear physics database, (4) identify those decay chains with complete data sets so that a dose coefficient can be calculated, and (5) calculate dose coefficients for those radionuclides that have a corresponding ENSDF reference with a complete and precise set of data files.

Results from this study will be used to expand current radiation safety dose coefficient databases to assist radiation safety personal in evaluating the risk to a worker from a planned or accidental exposure to radioactive materials not currently defined in existing standards. Also, ensuring the protection of workers at proposed nuclear accelerator facilities is paramount before these facilities can be implemented. Therefore, quantifying the health risks to workers at these facilities from the production of radionuclides generated during operations directly supports ongoing national efforts to promote transmutation technology and basic scientific research. Finally, the 524 radionuclides were based on a spallation neutron source of mercury. Information

![Diagram](image-url)

Figure 2.1. Schematic showing input data, dosimetry codes, and data flow in the dosimetric system applied in this work.
obtained in this study can be used and applied to other spallation neutron sources as they become available.

2.2 Interdatabase Comparison

In order to insure the accuracy of the input data and to facilitate a means in which to prioritize the radionuclides identified in this study, a comparison methodology was developed to compare the information in the ENSDF file to another nuclear physics database. The Nubase application module was used in this research to carry out a direct comparison of relevant nuclear structure and decay data found in an ENSDF data file for the radionuclides identified in this study. The Nubase application module was chosen because it is believed to more accurately reflect current scientific literature on nuclear structure and decay data as compared to ENSDF. The Nubase database includes “The 1995 Update to the Atomic Mass Evaluation” (Audi 1995), “The Nubase Evaluation of Nuclear and Decay Properties” (Audi 1997), and the “Evaluated Nuclear Structure Data File” (Burrows 1990) as references for published data. Although ENSDF and Nubase both rely on the 1995 Update to the Atomic Mass Evaluation (Ame’95), Nubase includes its own evaluation, “The Nubase Evaluation of Nuclear and Decay Properties”, which is updated more frequently. For example, the version of Nucleus used in this work was 08 July 2002. At the time of this writing, a more recent version of Nucleus had just been released (09 May 2003).

The process developed to cross reference the databases utilizes Microsoft Excel workbooks, one for each of the radionuclides requiring an evaluation, with a series of worksheets formatted to carry out the evaluation. There are two types of worksheets
found in the workbooks. These include: (1) the data comparison worksheet, used to cross reference the databases for each member of the decay chain, and (2) the classification worksheet, used to tabulate the results from each data comparison worksheet so that a decay chain categorization score can be generated. An example workbook showing the decay chain analysis for $^{61}\text{Fe}$ is given in Appendix A. Specific variables analyzed in the data comparison worksheets correspond to principal input parameters utilized by the EDISTR code in compiling a radioactive decay data file. These parameters are: (1) decay mode(s), (2) excitation energy, (3) half-life, (4) Q-value, and (5) spin and parity and are given in Fig. 2.2. In this work, the excitation energy parameter is used to quantify the energy released, in kiloelectron volts (KeV), for the isomeric transition of a particular radionuclide. After the appropriate information has been transcribed into the data comparison worksheets, the results are analyzed for either a percent difference or a binary score. A percent difference was generated for the excitation energy value, half-life value, and Q-value, while a binary score was used to evaluate the decay modes and the spin and parity values. Note that a binary score of one or a percent difference greater than or equal to one in the data comparison worksheets indicates poor agreement between the databases for the parameter in question. Results from the data comparison worksheets are tabulated and logically tested in a classification worksheet so that the decay chain can be categorized. Logic testing is used to generate a binary score for each parameter after the entire decay chain has been evaluated and these results are then weighted and summed so that a final categorization score is generated. A decay chain can fall into one of three categories based on the results of the logic testing. These categories include: (1) each member of the decay chain has a corresponding ENSDF data file and shows good
agreement between the databases, (2) each member of the decay chain has a corresponding ENSDF data file and one or more members of the decay chain shows poor agreement between the databases, and (3) an ENSDF data file is missing for one or more members of the decay chain. With respect to the category scores, good agreement is defined as having less than one percent difference and the sum of the binary scores equal to zero after the entire decay chain has been evaluated and cross referenced.

![Comparison of ENSDF and Nubase parameters](image)

Figure 2.2. Nuclear structure and decay parameters crossed referenced between ENSDF and Nubase. ENSDF data files showing good agreement between the databases will be utilized in a dose coefficient calculation.

### 2.3 Dose Coefficient Methodology

A more detailed explanation of the dose coefficient methodology used in this work is developed so that the dosimetric system can be adequately described. As noted in the previous section, a comparison methodology was developed to analyze the information in an ENDSF data file. Radionuclides identified as lacking a published dose coefficient were analyzed and categorized using this system prior to performing a dose coefficient calculation. In this dosimetric system the information in an ENSDF data file

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is used to create a radioactive decay database or dosimetry file. This file is ultimately used by the computational modules of DCAL to calculate a dose coefficient according to Equations (1.2.1a), (1.2.1b), and (1.2.2b).

2.3.1 Evaluated Nuclear Structure Data File (ENSDF)

The ENSDF library contains evaluated nuclear structure and decay data information for selected radionuclides with mass numbers ($A$) less than 263. For radionuclides with $A \geq 45$, this information is documented in the journal “Nuclear Data Sheets”. For radionuclides with $A < 45$, the data in ENSDF is based on compilations published in the “Nuclear Physics” journal. ENSDF data files for a radionuclide are located within the library according to progeny of the parent and the decay mode of interest. For example, to locate the ENSDF data file for tritium ($^3\text{H}$) for a beta minus decay would require Helium-3 ($^3\text{He}$) as the input parameter since the progeny of the $^3\text{H}$ beta minus decay is $^3\text{He}$. Files downloaded from the library are formatted in FORTRAN and consists of a collection of data sets. A data set can represent one of the following kinds of information: (1) the evaluated results from a single experiment, (2) the combined evaluated results of a number of experiments of the same kind, (3) the adopted properties of the nucleus, (4) the references (key numbers) used in all the data sets for a given mass number, and (5) the summary information for a mass chain giving information (e.g. Nuclear Data Sheet publication details). A data set is composed of records, with each record being made up of one or more 80-column images (Tuli 1987).

The records of the ENSDF data file contain specific information which describes measured or deduced nuclear properties for the various levels of the decaying nucleus. This information is used by the EDISTR code to generate a corresponding radioactive

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decay database for the radionuclide of interest. Missing or incomplete ENSDF records will affect the output results from EDISTR in the form of intensity and energy balance discrepancies. A more detailed explanation of the ENSDF records and the information they contain can be found in Appendix C.

2.3.2 Energy Distribution (EDISTR) Code

The EDISTR computer code was initially developed to compile a nuclear decay database for internal radiation dosimetry calculations by the Biomedical Effects and Instrumentation Section of the Health and Safety Research Division of ORNL. The EDISTR code uses an ENSDF data file (a basic radioactive decay data set) to calculate the mean energies and absolute intensities of all principal radiations associated with the radioactive decay of a nuclide. 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-ray spectra accompanying beta decay and monoenergetic Auger and internal conversion electrons, and (5) the radiations accompanying spontaneous fission (Dillman 1980).

The EDISTR code is essentially divided into three functional phases. These phases are shown in Fig. 2.3.1 and include: (1) the input phase, in which the ENSDF data file is read 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, in which the results of the computational phase are prepared for print (Dillman 1980). An outline of the methods and solutions used by the EDISTR code during the computational process of generating a radioactive decay data file are presented in Appendix D. A more detailed discussion of
the theory and empirical methods used during the computational process is given in a text by Dillman (1980).

---

Figure 2.3.1. The three functional phases of the EDISTR code as described by the ORNL/TM-6689 (Dillman 1980).

---

2.3.3 Decay Data (DECDAT)

The Decay Data directory contains a series of MS-DOS executables designed to take the output EDISTR file and suitably format it so that it can be incorporated into the Nuclear Decay Data Directory of DCAL. Executing the codes found in this directory results in three output files being generated. These output files include: (1) the index file, a pointer file used to indicate the first records in the beta and radiations file for a radionuclide, (2) the beta file, a tabulated beta spectrum for those radionuclides that exhibit beta decay, and (3) the radiations file, a file that contains information on the energies and intensities of the radiations associated with spontaneous nuclear
transformation of the radionuclide. The flow of data and the executables found in the DECDAT directory is given in Fig. 2.3.2. A more detailed explanation of the use of these executables as it relates to this research is given in Appendix E.

![DECDAT Directory Diagram](image)

Figure 2.3.2. The Decay Data (DECDAT) directory as described by Eckerman 2001.

2.3.4 Dose and Risk Calculation Software Package (DCAL)

The DCAL system consists of a series of computational modules (written in FORTRAN) driven by an interactive interface written in Professional BASIC. There are two modes of operation of DCAL: an interactive mode designed for evaluation of a given exposure case; and a batch mode that allows non-interactive, multiple-case calculations. DCAL has been used in the development of two Federal Guidance Reports 12 and 13 (EPA 1993; EPA 1999) and several publications of the ICRP, specifically in the

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computation of age-specific dose coefficients for members of the public (ICRP 1989, 1993, 1995a, 1995b, 1996). The Dosimetry Research Group (now the Biosystems Modeling Team within the Advanced Biomedical Science and Technology Group) at ORNL developed DCAL under the sponsorship of the U.S. Environmental Protection Agency.

DCAL contains two Nuclear Decay Data libraries that were initially documented in ORNL/TM-12350 (Eckerman et al. 1993). The "ICRP 38" collection consists of data for 825 radionuclides that appeared in Publication 38 (ICRP 1983), plus an additional 13 radionuclides evaluated during the preparation of a monograph for the Medical Internal Radiation Dose (MIRD) Committee of the Society of Nuclear Medicine (Weber et al. 1989). Additionally, the "JAERI 02" library was provided to the Working Group as part of the DCAL software package and consists of data for 1023 radionuclides from both the ICRP 38 collection and the JAERI-Data/Code 2002-013. It is important to note that additional libraries can be incorporated into the DCAL software package to include radionuclides that have not been previously documented.

DCAL performs biokinetic and dosimetric calculations for the case of acute intake of a radionuclide by inhalation, ingestion, or injection into blood at a user-specified age. For the intake of the radionuclide, the user may compute either equivalent or absorbed (low and high LET) dose rates as a function of time following intake. Selection of the equivalent dose option allows the generation of a table of age-specific dose coefficients. In addition, DCAL also includes a computational module for the evaluation of dose rate resulting from exposure to radionuclides distributed in an environmental media. This computational module utilizes the photon and electron...
dosimetric data tabulated in Federal Guidance Report 12 (EPA 1993) to generate radionuclide specific dose coefficients. Besides the Nuclear Decay Data library, DCAL also contains libraries of Biokinetic Models and Anatomical Data which are utilized during the computational process. These libraries contain current state-of-the-art data and allow the best available estimates of radiation dose and risk from internally deposited radionuclides, with minimal input by the user.

2.3.4.1 Internal Dose Coefficient Calculations

Dosimetric calculations for the intake of a radionuclide proceed in three main steps and are schematically presented in Fig. 2.3.4.1. The three main steps include: (1) the calculation of time-dependent activity of the parent radionuclide and any radioactive progeny present in anatomical regions (source regions) of the body, (2) the calculation of specific effective energies (SEE values) for specified source and target organs, and (3) the calculation of dose rates or equivalent dose rates, based on output generated in the first two steps. Dose coefficients may be computed after the third step has been completed using the Tabulate Dose Coefficient (HTAB) utility. A more detailed discussion of the computational modules and the specific parameters used in this work to calculate internal dose coefficients for the adult worker and members of the public is given in Appendix F.

2.3.4.2 External Dose Coefficients Calculations

Dosimetric calculations for the external exposure of radionuclides in the environment proceed in two main steps and are shown schematically in Fig. 2.3.4.2. These steps include: (1) the identification of photons and electrons with sufficient energy as a result of spontaneous nuclear transformation of a given radionuclide, and (2)
Figure 2.3.4.1. Schematic of DCAL system for internal radiation dosimetry calculations—adopted from ORNL/TM-2001/190 (Eckerman et al. 2001)

Figure 2.3.4.2. Schematic of DCAL’s dosimetric system for the computation of an external dose coefficient calculation.
comparing the photon and electron energy data to tabulated look up tables of external
dose coefficients, based on a hermaphrodite phantom, to extrapolate the contribution of
dose for a given exposure scenario involving a person adequately described by reference man. Specific parameters used in this work to calculate external dose coefficients are
given in Appendix G.

2.4 Quality Assurance

Six radionuclides with published dose coefficient values were selected to evaluate the adopted methodology used in this work. The radionuclides selected for this study included three from the ICRP Publication 68 database ($^{201}$Au, $^{41}$Ar, and $^{61}$Co), and three from the more recently released JAERI database ($^{144}$Nd, $^{50}$V, and $^{38}$S). The appropriate ENSDF data files were selected for each of these radionuclides and any associated decay chain members according to the Nubase decay chain. These files were then processed by the EDISTR code and the executables of Decay Data to build a Nuclear Decay library for inclusion into the DCAL software package. A committed effective dose coefficient value and a dose coefficient for air submersion value were generated for each radionuclide using the Nuclear Decay library that was built as part of this study. These results were compared to the corresponding dose coefficient values generated using the “JAERI 02” Nuclear Decay library contained within the DCAL software package. The “JAERI 02” library was selected because it contains results from both the ICRP 38 database and JAERI-Data/Code 2002-013. The results were also collectively compared among the various student members of the Working Group so that problem areas could be identified related to the use of the dosimetry codes.
CHAPTER 3

RESULTS AND DISCUSSION

3.1 Radionuclide Identification

One hundred fifty eight of the 524 radionuclides given to the UNLV Transmutation Research Program have been identified as lacking an appropriate reference for a published dose coefficient according to existing radiation safety dose coefficient databases queried as part of this study. A list of these radionuclides is provided in Appendix B. The 158 radionuclides identified in this study were categorized according to half-life and the results are presented in Fig. 3.1. As seen in Fig. 3.1 the majority of radionuclides, 86, had a half-life less than one minute, 57 had a half-life between one and ten minutes and, 15 had a half-life equal to or greater than ten minutes. The half-life categorization of these radionuclides served as an initial means to prioritize the list and is based on an intake scenario involving workers at a nuclear facility. Radionuclides identified with a half-life greater than or equal to one minute present the greatest radiological risk to workers and were therefore given the highest priority in this study. As a result, 72 radionuclides were identified and included in the interdatabase comparison study as outlined in Section 2.2 for a possible dose coefficient calculation. Those radionuclides identified with a half-life of less than one minute were set aside because of concerns regarding the computational capabilities of the dosimetry codes used in this work. These radionuclides will be addressed at a later date.
3.2 Interdatabase Comparison Study

Nuclear decay data for the 72 radionuclides identified as lacking a published dose coefficient and their associated decay chains members were established using Nubase and cross referenced with the ENSDF library. In all, a total of 109 decay chains were evaluated as part of this study after secondary and tertiary decay chains were included. The 109 decay chains included 699 radionuclides in their ground and isomeric states with each decay chain having approximately six decay chain members. A quantitative comparison was made of relevant nuclear structure and decay data utilized by the EDISTR code between the two databases for the 699 radionuclides. Radionuclidic results were tabulated for each parameter relative to its associated decay chain so that systemic trends could be identified. The results of this analysis are given in Fig. 3.2.
As shown in Fig. 3.2 the largest observed discrepancy occurred between reported Q-values with 79 out of the 109 decay chains showing poor agreement. Only 105 decay chains were evaluated for an excitation energy value, indicating four decay chains without at least one member having an associated isomeric state of transition.

The energy associated with exothermic nuclear reactions comes from the conversion of mass into energy. The energy released, $Q$, is given by Einstein's relation:

$$Q = (\Delta M)c^2 \quad \text{(MeV)},$$

where $\Delta M$ is the mass loss associated with the reaction and $c$ is the velocity of light. In this work we are only interested in the energy differences in nuclear decay reactions and can obtain the energy released, Q-value, directly from the atomic mass excess ($\Delta$) data. Thus the observed Q-value discrepancies between ENSDF and Nubase can be
investigated as they relate to documented atomic mass excess data for a given radionuclide. ENSDF cites both the “1993 Atomic Mass Evaluation” (Ame’93) and “The 1995 update to the atomic mass evaluation” (Ame’95) as references for atomic mass excess data, whereas Nubase relies on the Ame’95 plus additional updates provided by the authors. Additionally, the NNDC website provides a utility, QCALC, for calculating Q-values to update ENSDF data files and is based on Ame’95. Therefore, in theory, QCALC and Nubase should both provide the same Q-value results since they both utilize the same atomic mass excess data excluding the updates. In an effort to update ENSDF Q-value records showing greater than 1% difference in the interdatabase comparison study mass excess data for QCALC and Nubase was compared to Ame’95. The objective of this comparison was to determine which reference contained more accurate data, relative to current scientific literature, so that it could be used to update ENSDF Q-value records. Mass excess data for 2862 nuclides were compared and a percent difference was calculated for QCALC versus Ame’95 and Nubase versus Ame’95. A statistical analysis was performed on the percent difference results and is given in Table 3.2.

Based on the statistical results given in Table 3.2, QCALC more closely reflected the values found in Ame’95 with a mean and standard error of 0.0499 and 0.0098 respectively. The mean represents the average percent error value of the data set and the standard error represents the variability about the mean. The fact that the QCALC mean is not equal to zero is the result of rounding errors associated with the reported data. For example, Ame’95 reports the mass excess data for $^{41}$V as $-242 \pm 252$ KeV while QCALC reports the value as $-2.00 \times 10^2 \pm 3.00 \times 10^2$ KeV giving a percent difference of 19.005.
Table 3.2 Statistical Results of Mass Excess Comparison Study

<table>
<thead>
<tr>
<th>STATISTICS</th>
<th>QCALC vs. Ame'95</th>
<th>NUBASE vs. Ame'95</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean</td>
<td>0.0499</td>
<td>0.9196</td>
</tr>
<tr>
<td>Standard Error</td>
<td>0.0098</td>
<td>0.6395</td>
</tr>
<tr>
<td>Median</td>
<td>0.0029</td>
<td>0.0041</td>
</tr>
<tr>
<td>Mode</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Number equal to Mode</td>
<td>65</td>
<td>93</td>
</tr>
<tr>
<td>Range</td>
<td>19.005</td>
<td>1820.20</td>
</tr>
<tr>
<td>Minimum</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>Maximum</td>
<td>19.005</td>
<td>1820.20</td>
</tr>
<tr>
<td>Count</td>
<td>2862</td>
<td>2862</td>
</tr>
</tbody>
</table>

Also shown in Table 3.2 is the maximum percent difference value for each data set. A large discrepancy is noted for the maximum percent difference value for the Nubase versus Ame'95 data set (1820.20) and is the result of the reported mass excess data for $^{46}$S from Nubase. This value was confirmed after corresponding with the author's of Nubase and was the result of a recent update. This large discrepancy also accounts for the order of magnitude difference in the calculated mean values between the data sets and explains why the Nubase mean value is not closer to zero. Several other updates were also noted during this analysis and are shown in Fig. 3.2.1. Therefore, although QCALC showed better statistical results when compared to Ame'95 the value of Nubase is shown in its ability to provide information relative to current scientific literature. As a result, Nubase was used to update ENSDF Q-value records when greater than 1% difference was noted between the databases.
Figure 3.2.1. Comparison of Nubase to Ame’95 showing recent updates to Nubase related to mass excess data.

The 79 decay chains identified as having a Q-value discrepancy were updated according to the values found in Nubase so that a categorical score for the decay chain could be performed. Categorical scores for the 72 radionuclides identified as lacking a published dose coefficient are presented in Fig 3.2.2. As shown in Fig. 3.2.2, 30 radionuclides had category one scores, 24 had category two scores, and 18 had category three scores. In the final analysis only 42% (30 out of 72) of the radionuclides identified as lacking a published dose coefficient showed good agreement between the databases for relevant nuclear structure and decay data so that a dose coefficient calculation could be performed. Thirty three percent (24 out of 72) of the radionuclides require further research to resolve observed discrepancies between the databases before a dose coefficient calculation can be performed. While 25% (18 out of 72) of the radionuclides had missing ENSDF data files for one or more members of its decay chain and can not be

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evaluated for a dose coefficient calculation at this time. The 30 category one radionuclides identified as lacking a published dose are presented in Table 3.2.1.

3.3 Quality Assurance

Six radionuclides with published dose coefficient values (three from the ICRP database and three from the more recently released JAERI database) were selected to evaluate the adopted methodology used in this work. These results were compared to the corresponding dose coefficient values generated using the “JAERI 02” Nuclear Decay data library contained within the DCAL software package and are given in Appendix H.

As indicated by the percent error results in Appendix H, dose coefficient values generated after building the appropriate Nuclear Decay library for the six radionuclides evaluated as part of this study showed excellent agreement with those values generated using the “JAERI 02” library as indicated by the percent error results. These results show that the methodology was successfully adopted and implemented to perform a dose coefficient computation and will be utilized for the category one radionuclides identified in this study.
Figure 3.2.2. Categorical scoring summary for the 72 radionuclides identified as lacking a published dose coefficient.

Table 3.2.1 Category One Radionuclides

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Radionuclide</th>
<th>Radionuclide</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{157}$Er</td>
<td>$^{178}$Os</td>
<td>$^{193}$Tl</td>
</tr>
<tr>
<td>$^{160}$Er</td>
<td>$^{195}$Pb</td>
<td>$^{157}$Tm</td>
</tr>
<tr>
<td>$^{61}$Fe</td>
<td>$^{153}$Pm</td>
<td>$^{160}$Tm</td>
</tr>
<tr>
<td>$^{144}$Gd</td>
<td>$^{133}$Pr</td>
<td>$^{161}$Tm</td>
</tr>
<tr>
<td>$^{171}$Hf</td>
<td>$^{201}$Pt</td>
<td>$^{171}$W</td>
</tr>
<tr>
<td>$^{197}$Ir</td>
<td>$^{176}$Re</td>
<td>$^{172}$W</td>
</tr>
<tr>
<td>$^{127}$La</td>
<td>$^{113}$Sb</td>
<td>$^{174}$W</td>
</tr>
<tr>
<td>$^{128}$La</td>
<td>$^{167}$Ta</td>
<td>$^{119}$Xe</td>
</tr>
<tr>
<td>$^{168}$Lu</td>
<td>$^{171}$Ta</td>
<td></td>
</tr>
<tr>
<td>$^{176}$Os</td>
<td>$^{192}$Tl</td>
<td>$^{161}$Yb</td>
</tr>
</tbody>
</table>
3.4 Dose Coefficient Calculations

Prior to performing a dose coefficient calculation for the 30 category one radionuclides a more detailed analysis was made of the ENSDF data files, the energy balance data output from EDISTR, and the missing daughter products identified by the Check Daughters executable of the Decay Data directory. The results from this analysis serve to document any observed discrepancies or editing manipulations associated with the data used to compute a dose coefficient value.

3.4.1 ENSDF Record Analysis

Analysis of the ENSDF data files for each category one radionuclide decay chain was performed to insure that the corresponding data sets had complete records prior to performing a dose coefficient computation. The results of this analysis are given in Appendix I. As shown in Appendix I, several ENSDF data files required the addition of either a half-life value or the word "STABLE" to the ground state level (level 0). This editing manipulation preserved the integrity of the decay chain by insuring that all members of a given decay chain would be recognized by the codes used in this work. For example, if the half-life value, 12.6 minutes, were not added to the ground state level of $^{157}$Er, the $^{157}$Er decay chain would effectively end with the decay of $^{157}$Er and no other decay chain members would be considered during the computational process of calculating a dose coefficient for $^{157}$Er. Nineteen out of the 30 radionuclides had one or more decay chain members missing ENSDF records, most notably, electron capture and beta minus records. These results indicate a lack of experimental data and effectively eliminate a given radionuclide for a dose coefficient computation. Appendix J also documents any Q-value updates that were performed as part of the interdatabase
comparison study for the 30 category one radionuclides and their associated decay chain members.

### 3.4.2 EDISTR Energy Balance Data

The output file generated by the EDISTR code contains, among other radioactive decay information, intensity and energy balance data. This data can be used to evaluate a given radionuclide’s decay level scheme with respect to the total energy associated with the decay. In other words, the total energy available for a given decay should equal the total energy content of all the radiations associated with that decay. In theory, the percent error associated with the energy balance data should be equal to zero. In this work, a percent error of less than or equal to 5% was considered acceptable. The percent error related to the total energy balance data for the 11 radionuclides and their associated decay chain members included in this study is given in Appendix J as well as an example EDISTR output file for $^{61}$Fe. A total of 38 radionuclides were analyzed for a percent error related to their total energy balance data. Five radionuclides had deficiencies greater than 5% effecting 5 out of the 11 radionuclides being worked up for a dose coefficient calculation. Those radionuclides showing a percent error greater the 5% included: $^{160}$Er (6.82%), $^{201}$Pt (44.6%), $^{161}$Tm (22.8%), $^{161m}$Er (69.5%), and $^{173}$W (8.54%). However, even though these radionuclides showed a percent error greater than 5% they were not excluded from this study. Although the final outcome of a dose coefficient computation involving these radionuclides will undoubtedly be affected, the magnitude of this error will remain unknown until further information becomes available.
3.4.3 Inclusion of Daughter Products

The output file generated by the Check Daughters (Chkdaus) executable found in the Decay Data (DECDAT) directory was unremarkable for missing daughter products related to the 11 radionuclides and their associated decay chains being evaluated as part of this study. Also, no warning flags were noted in the Index file after it was produced. These results indicate that DCAL-type nuclear decay data files were successfully generated which can be incorporated into the DCAL software package and utilized for dose coefficient calculation purposes.

3.4.4 Dose Coefficient Results

The calculated committed equivalent dose coefficients, $h_{T,50}$, and the calculated committed effective dose coefficients, $e_{50}$, are presented in Appendix K for the adult worker and members or the public. The dose coefficients for inhalation of 1 μm and 5 μm particulates and ingestion are presented along with the $f_i$ values and absorption types for the adult worker. Values of $f_i$ represent the fraction of a stable element reaching the body fluids following ingestion. Absorption types describe the rate of absorption of a particular radionuclide into the various tissues and compartments of the Human Respiratory Tract Model (ICRP 1994). Absorption types are denoted as: (1) type F (fast) for materials that are readily absorbed into the blood, (2) type M (moderate) for materials with intermediate rates of absorption, and (3) type S (Slow) for relatively insoluble materials. These dose coefficients results are followed by those for members of the public and include inhalation of 1 μm particulates and ingestion as well as the $f_i$ values and absorption types. Dose coefficients for air submersion, exposure to contaminated ground surface, and exposure to soil contaminated to an infinite depth are given in

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Appendix L. For each radionuclide the organ equivalent dose coefficient $h_{T,ext}$, and the effective dose coefficient, $e_{ext}$, are presented. The coefficients are for a soil at a density of $1.6 \times 10^3$ kg m$^{-3}$.

Three additional radionuclides were identified as lacking a published dose coefficient value and were included in the results presented in Appendices K and L. These radionuclides were not included on the original list for a dose coefficient evaluation; however, these radionuclides were included as members of the decay chains being evaluated as part of this study. The additional radionuclides include $^{144}$Eu, $^{161m}$Er, and $^{160m}$Ho.
CHAPTER 4

CONCLUSIONS AND FUTURE WORK

The 72 radionuclides identified as lacking a published dose coefficient value with a half-life value greater than or equal to one minute were successfully evaluated utilizing the interdatabase comparison methodology developed as part of this study. This methodology emphasized the need to quantify the accuracy of the input data relative to another nuclear physics database prior to performing a dose coefficient computation given the frequency in which ENSDF data files are updated. Several radionuclides evaluated as part of this study had their most recent ENSDF evaluations performed prior to 1995 suggesting an evaluation cycle time significantly longer than the stated six years.

Although the interdatabase comparison study served its purpose of prioritizing the list given to the Working Group it can be improved upon by incorporating the ENSDF records check into the system. It was noted that 19 out of the 30 category one radionuclides lacked appropriate ENSDF records in their data sets resulting in their removal from consideration of a dose coefficient computation. These results effectively placed these radionuclides into category two of the current scoring scheme. They will essentially remain category two radionuclides until further experimental data becomes available. Incorporating this step into the current system could dramatically improve the accuracy of the final categorical results.
The Nubase database proved to be invaluable in terms of identifying radioactive decay chains and providing basic nuclear property information relative to current scientific literature for comparative purposes. Although Nubase Q-values were only used to update ENSDF Q-value records in this research, the JAERI study demonstrated its true potential in updating an ENSDF data file in preparation for dose coefficient computation. As noted previously both ENSDF and Nubase rely on information found in Ame’95 for basic nuclear structure and decay property data. Nubase, however, is maintained by the same authors of Ame’95 and is used as a platform to release new experimental data to the scientific community more frequently. Given the rare nature of the anthropogenic radionuclides included in this study the use of alternative sources, such as Nubase, to confirm or fill experimental data gaps in the ENSDF library may prove to be useful in further research.

The adopted dosimetric methodology used in this work was successfully implemented to perform a dose coefficient computation. Dose coefficient results from the QA radionuclide analysis study showed good agreement with those values obtained using the “JAERI 02” library found within the DCAL software package. For the purpose of this analysis good agreement is defined by the results of the percent error computations given in Appendix H. As indicated by the results each radionuclide evaluated as part of this study had a percent error equal to zero for inhalation, ingestion, and air submersion dose coefficients relative to the “JAERI 02” database.

Dose coefficient values were calculated for 11 radionuclides from the initial list. Three additional dose coefficient values are reported for radionuclides not included on this list. Although dose coefficient values are reported for 14 different radionuclides in
this work six have results that require further investigation due to EDISTR total energy balance discrepancies. Those radionuclides include: (1) $^{160}$Er, (2) $^{161m}$Er, (3) $^{201}$Pt, (4) $^{161}$Tm, (5) $^{173}$W, and (6) $^{161}$Yb. There are several utilities available on the NNDC website to assist evaluators in assessing an ENSDF data file for continuity. For example, the GABS utility calculates absolute gamma-ray intensities and a decay scheme normalizing factor for converting relative intensities to absolute values per 100 decays of the parent nucleus. Utilities such as these maybe employed in the future to correct for deficiencies noted in the EDISTR output file given the appropriate training. It should also be noted that both internal and external dose coefficient values are reported for three radionuclides whose half-life values are less than one minute. Those radionuclides include: (1) $^{144}$Eu, (2) $^{161m}$Er, and (3) $^{161m}$Ho. Given the computational limitations of the DCAL dosimetric system the internal dose coefficient values associated with these radionuclides require further investigation; however, the external dose coefficient values can be directly utilized in evaluating a dose to an individual.

In closing, internal and external dose coefficient values have been calculated for 14 anthropogenic radionuclides which are not currently presented in Federal Guidance Reports No. 11, 12, and 13 or Publications 68 and 72 of the International Commission on Radiological Protection. Internal dose coefficient values are reported for inhalation and ingestion of 1 $\mu$m and 5 $\mu$m particulates along with the $f_i$ values and absorption types for the adult worker. Internal dose coefficient values are also reported for inhalation and ingestion of 1 $\mu$m particulates as well as the $f_i$ values and absorption types for members of the public. Additionally, external dose coefficient values for air submersion, exposure
to contaminated ground surface, and exposure to soil contaminated to an infinite depth are also presented.
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