8-2003

Dose Coefficients for Radionuclides Produced in a Spallation Neutron Source

John P. Shanahan
University of Nevada, Las Vegas

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

Graduate College
University of Nevada, Las Vegas
August 2003

ABSTRACT

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

by

John Patrick Shanahan

Dr. Phillip W. Patton, Examination Committee Chair
Assistant Professor of Health Physics
University of Nevada, Las Vegas

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_1 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_1 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.

TABLE OF CONTENTS

LIST OF FIGURES

ACKNOWLEDGEMENTS

There are several people that contributed a great deal to this work that I must thank. First, I would like to thank my advisor, Dr. Phillip W. Patton, for his support and guidance throughout this project. I sincerely appreciate the opportunity that he provided me to pursue my interest in health physics. I must also thank Dr. Keith F. Eckerman, Dr. Mark J. Rudin, Dr. William H. Johnson, and Dr. Richard D. Tandy for their suggestions and for being members of my committee.

I would also like to thank all the members of the UNLV Dose Coefficient Working Group. It was an honor and privilege to have been associated with such distinguished individuals and academic institutions.

Finally, I would like to thank some people who are very special to me. These include my children, Sara, Kyle, and Ryan, who patiently tolerated my endless hours of studying, my parents, John and Patricia Shanahan, who have always loved and supported me, and my wife, Marguerite, who I continue to fall more in love with each day.

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):

$$D = \frac{d\bar{\varepsilon}}{dm} \qquad \text{(Gy)}, \tag{1.1a}$$

where $d\bar{\varepsilon}$ 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):

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

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):

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

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 workers 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 ($h_{T,ext}$) from external exposure, or the effective dose per unit time-integrated exposure to a radionuclide (e_{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):

$$H_{T,50} = K \sum_S U_S SEE(T \leftarrow S) \qquad \text{(Sv)}. \qquad (1.2.1a)$$

The constant, K, depends on the units specified for $H_{T,50}$, $SEE(T \leftarrow S)$, and U_S. In the ICRP methodology, K is equal to 1.6×10^{-10} Sv g MeV^{-1} when $SEE(T \leftarrow S)$ is expressed in megaelectron volts (MeV) per gram (g) per nuclear transformation, and U_S in nuclear transformations. The specific effective energy, $SEE(T \leftarrow S)$, 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:

$$E_{50} = \sum_T w_T H_{T,50} \qquad \text{(Sv)}, \qquad (1.2.1b)$$

where w_T 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_T 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

Organ or Tissue	w_T
Gonads	0.20
Breast	0.05
Colon	0.12
Red bone marrow	0.12
Lung	0.12
Stomach	0.12
Urinary Bladder	0.05
Liver	0.05
Esophagus	0.05
Thyroid	0.05
Bone Surfaces	0.01
Skin	0.01
Remainder	0.05

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

$$h_{T,ext} = \frac{H_T}{\int C(t)dt} \qquad \text{(Sv per Bq s m}^{-3}\text{)}. \qquad (1.2.2a)$$

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_T^S, for any tissue *T* for any exposure mode *S* can be expressed as (Eckerman et al. 1993):

$$h_T^S = \sum_{j=e,\gamma}\left[\sum_i y_j(E_i)\hat{h}_T^S(E_i) + \int_0^\infty y_j(E)\hat{h}_{Tj}^S(E)dE\right] \qquad \text{(Sv per Bq s m}^{-3}\text{)}, \qquad (1.2.2b)$$

where $y_j(E_i)$ is the yield of discrete photon radiations of type *j* and energy E_i, and $y_j(E)$ 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 $\hat{h}_T^S(E)$ 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 process 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

minute. 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.

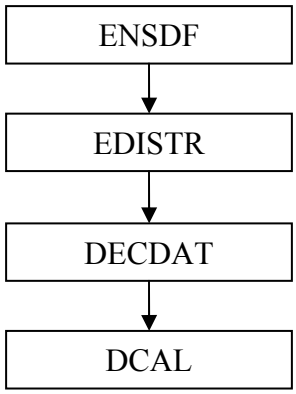

Figure 2.1. Schematic showing input data, dosimetry codes, and data flow in the dosimetric system applied in this work.

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

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}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.

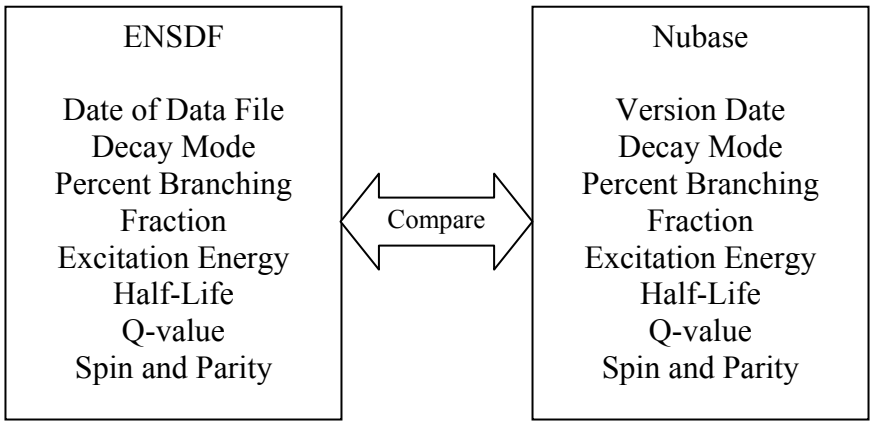

Figure 2.2. Nuclear structure and decay parameters crossed referenced between ENSDF and Nubase. ENSDF data files showing good agreement between the databases will be utilized in a dose coefficient calculation.

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

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}H) for a beta minus decay would require Helium-3 (^{3}He) as the input parameter since the progeny of the ^{3}H beta minus decay is ^{3}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

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).

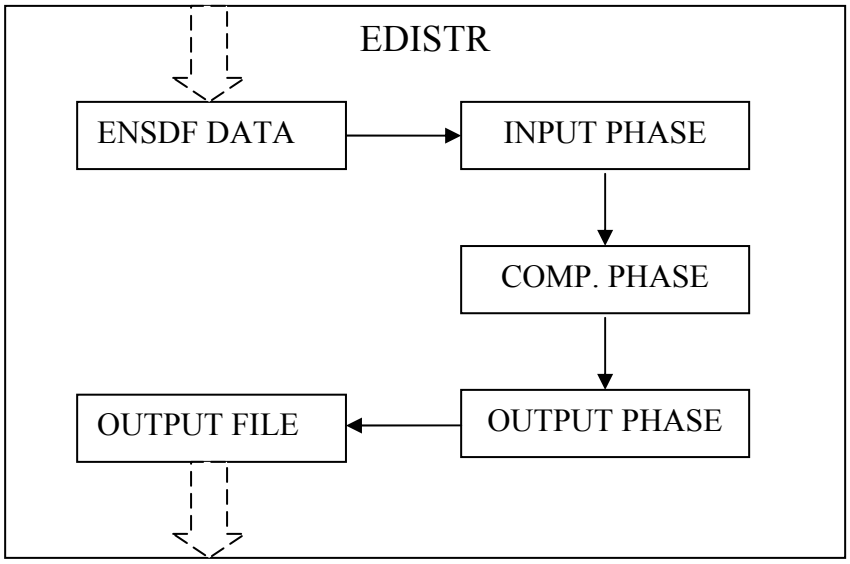

Figure 2.3.1. The three functional phases of the EDISTR code as described by the ORNL/TM-6689 (Dillman 1980).

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.

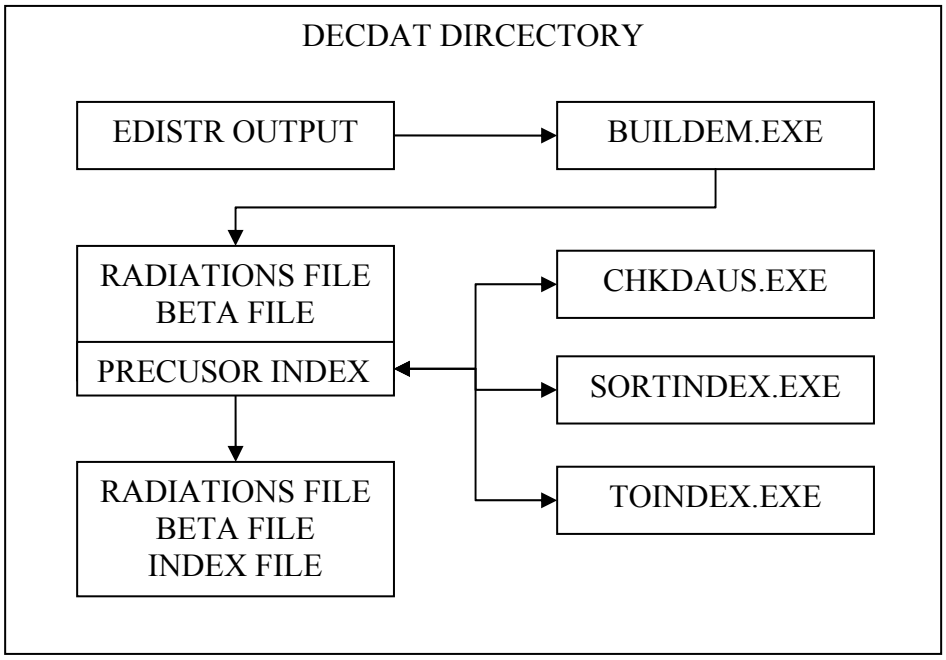

Figure 2.3.2. The Decay Data (DECDAT) directory as described by Eckerman 2001.

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

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)

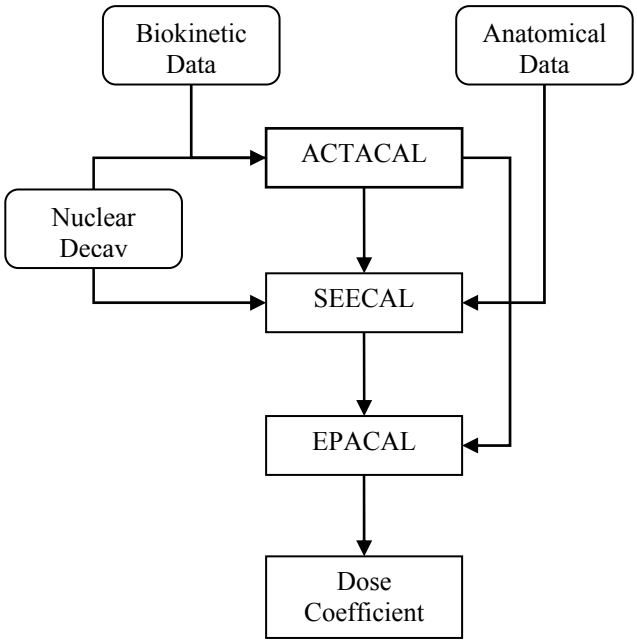

Figure 2.3.4.1. Schematic of DCAL system for internal radiation dosimetry calculations– adopted from ORNL/TM-2001/190 (Eckerman et al. 2001)

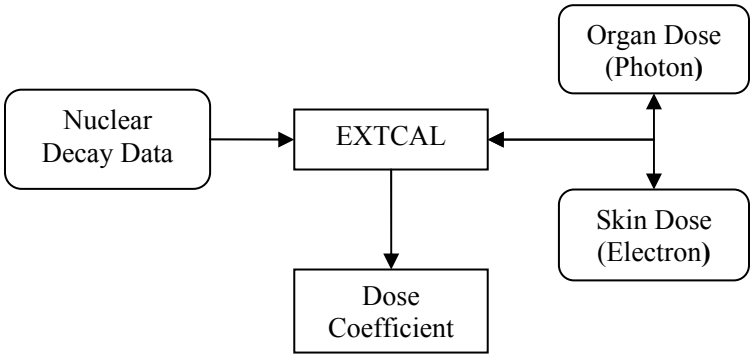

Figure 2.3.4.2. Schematic of DCAL's dosimetric system for the computation of an external dose coefficient calculation.

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.

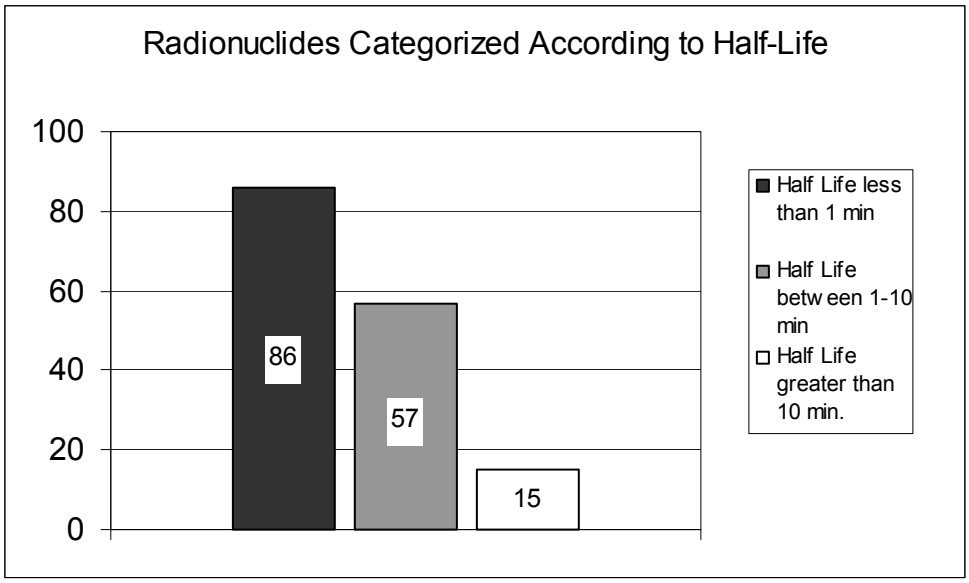

Figure 3.1. Radionuclides identified as lacking a published dose coefficient according to a query of existing radiation safety dose coefficient databases. Seventy two radionuclides with a half-life of greater than or equal to one minute were selected for a dose coefficient evaluation as part of this study.

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.

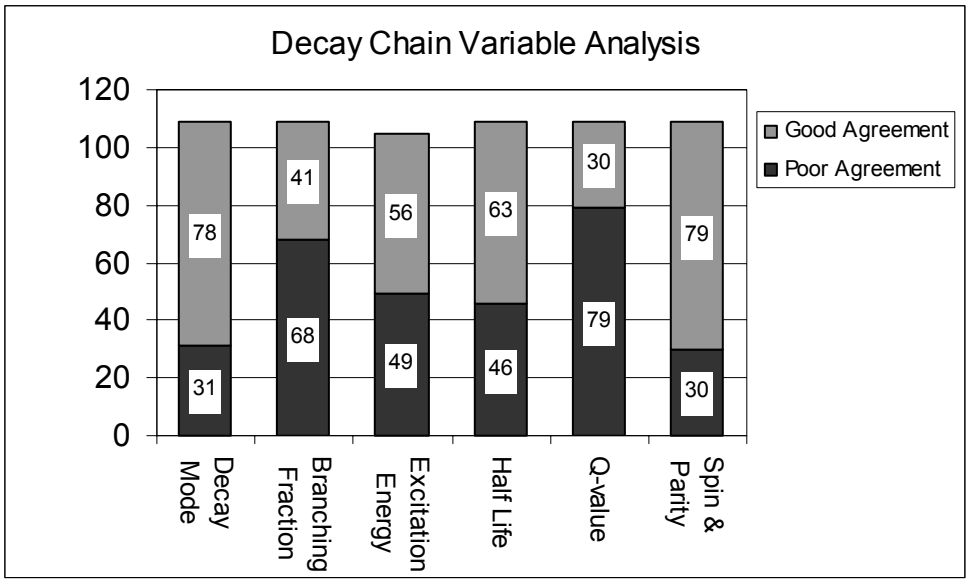

Figure 3.2. Tabulated radionuclidic results for each of the variables evaluated in this work after being crossed-referenced and scored.

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:

$$Q = (\Delta M)c^2 \qquad \text{(MeV)}, \tag{3.2}$$

where ΔM 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 (Δ) 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 ± 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

STATISTICS	QCALC vs. Ame'95	NUBASE vs. Ame'95
Mean	0.0499	0.9196
Standard Error	0.0098	0.6395
Median	0.0029	0.0041
Mode	0.0	0.0
Number equal to Mode	65	93
Range	19.005	1820.20
Minimum	0.0	0.0
Maximum	19.005	1820.20
Count	2862	2862

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.

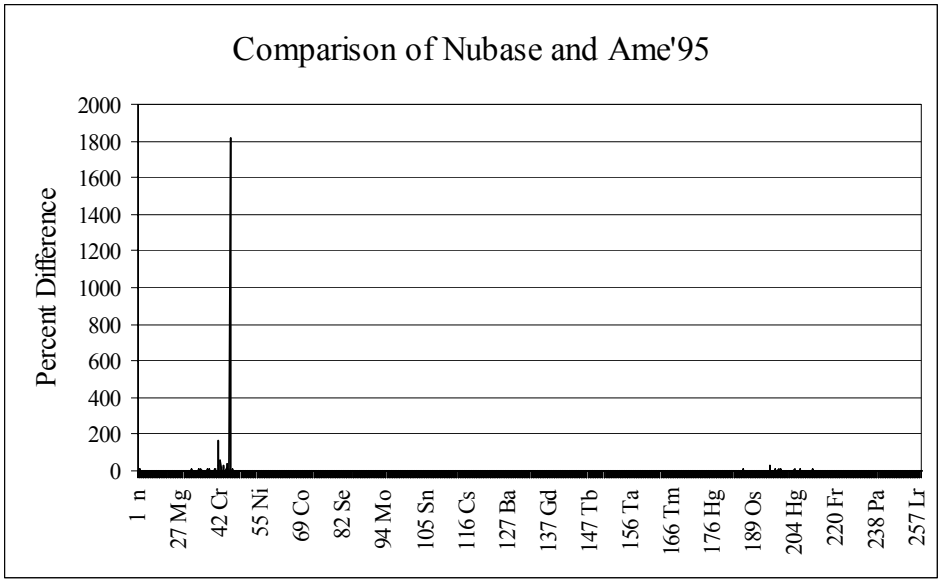

Figure 3.2.1. Comparison of Nubase to Ame'95 showing recent updates to Nubase related to mass excess data.

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

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.

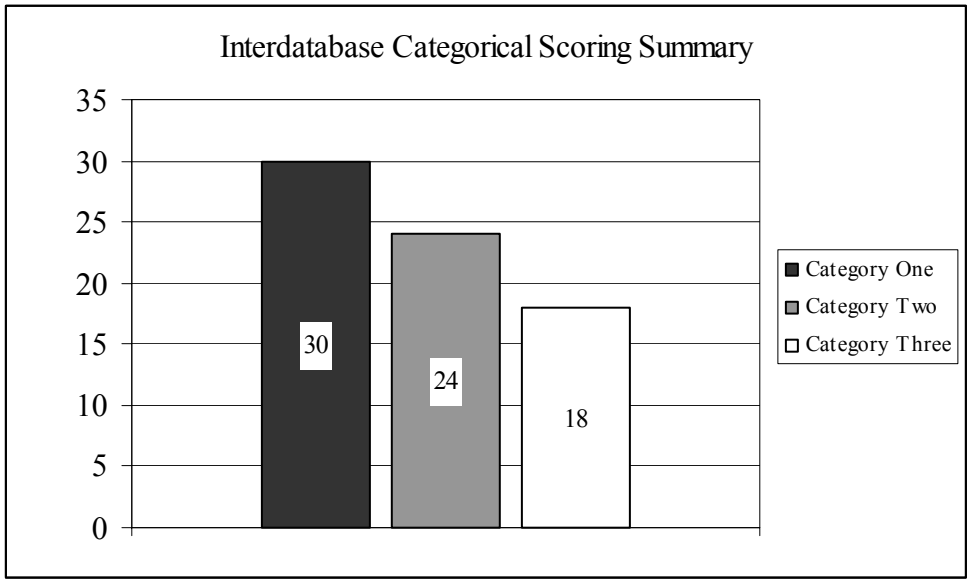

Figure 3.2.2. Categorical scoring summary for the 72 radionuclides identified as lacking a published dose coefficient.

Table 3.2.1 Category One Radionuclides

Radionuclide	Radionuclide	Radionuclide
^{157}Er	^{178}Os	^{193}Tl
^{160}Er	^{195}Pb	^{157}Tm
^{61}Fe	^{153}Pm	^{160}Tm
^{144}Gd	^{133}Pr	^{161}Tm
^{171}Hf	^{201}Pt	^{171}W
^{197}Ir	^{176}Re	^{172}W
^{127}La	^{113}Sb	^{173}W
^{128}La	^{167}Ta	^{174}W
^{168}Lu	^{171}Ta	^{119}Xe
^{176}Os	^{192}Tl	^{161}Yb

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 by 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_1 values and absorption types for the adult worker. Values of f_1 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_1 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

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 1.6×10^3 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 μm and 5 μm particulates along with the f_1 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_1 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.

APPENDIX C

ENSDF Records

The first record in each data set is always the IDENTIFICATION record and indicates the type of data to be found in the data set. In this work, ENSDF data files are selected based on the radioactive decay of a given radionuclide. Therefore, the types of data sets used in this work are known as decay data sets. A decay data set IDENTIFICATION record contains a field consisting of three parts; the decaying nucleus, the type of decay, and the word "DECAY" separated by one or more blanks. The decaying nucleus is specified by the mass number followed by the chemical symbol of the parent. The EDISTR code uses the data in this three-part field to assign a mass number, atomic number, and decay type to the parent nuclide. The only other item read from the IDENTIFICATION record is the date the data set was entered into the ENSDF library. Immediately following the IDENTIFICATION record is a group of records which contain information about the entire data set. These records include: (1) the PARENT record, designated by the letter P, (2) the general COMMENT record, designated by the letter C, (3) the NORMALIZATION record, designated by the letter N, (4) the Q-VALUE record, designated by the letter Q, and (5) the CROSS-REFERENCE record, designated by the letter X. It should be noted that not all these records are contained in every data set for each radionuclide evaluated for a dose coefficient calculation. Following the group of records containing information about the entire data set is the main body of the data set, composed of LEVEL, ALPHA, B-, EC, B+, and GAMMA

records, which describe measured or deduced nuclear properties. These records are associated with the level that decays (for GAMMA records) or the level that is populated (for B-, EC, B+, or ALPHA records). Thus, each LEVEL record is followed by a group of radiation records (B-, EC, B+, or ALPHA) describing charged-particle decay into the level and GAMMA records describing gamma-ray decay out of the level. The LEVEL records, and the corresponding radiations records, are placed in the data set in order of increasing energy, and are the primary records used by the EDISTR code as it generates radioactive decay data file. The last record in a data set is always the END record (a blank card) and is used to indicate the end of an ENSDF data file (Tuli 1987).

APPENDIX D

EDISTR Methods of Solutions Outline

Methods of Solutions Outline (PSR-191/EDISTR Code Package)

1. Alpha decay.
    a. Kinetic energies of the alpha particles and the associated recoil nuclei are computed using conservation of energy and momentum principles. The input data consist of the ground state Q value, the various excitation energies of the levels in the daughter nuclide at which the alpha transitions end, and the corresponding alpha intensities.
2. Beta decay.
    a. The average energies of the beta particles and the emitted continuous spectra are calculated using the Fermi theory of beta decay with the input of additional data to determine the forbiddenness of the beta spectra.
3. Electron-Capture decay.
    a. The distribution of primary vacancies created in the various atomic shells and sub shells as a result of the electron-capture process are calculated using the K/L/M capture ratios.
4. Internal conversion of gamma rays.
    a. This is a process by which the energy of a transition between two states of a nucleus is transferred to an orbital electron. The distribution of the primary vacancies in the various atomic shells and the energies and intensities of conversion electrons are calculated.
5. X-ray and Auger-electron intensities and energies.
    a. Intensities of X-rays and Auger electrons are obtained using the numbers of primary vacancies in the various sub shells for electron capture or for internal conversion of electrons.
6. Spontaneous fission.
    a. The fission decay fraction, the number of neutrons emitted per fission, the mass number of the parent nuclide, and the 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.
7. Bremsstrahlung radiation.
    a. Bremsstrahlung spectra associated with beta particles and monoenergetic conversion and Auger electrons are calculated.

APPENDIX E

Decay Data (DECDAT)

There are four applications located in the DECDAT directory and they are executed in a predefined sequence. To initiate the reformatting process a template file is first built to indicate the name(s) and location of the EDISTR output file. Once the template file is built the first application, BUILDEM.EXE, is executed to read the template file and create the three decay data files in a format that can be read by DCAL. It should be noted that a precursor to the INDEX file is generated at this time and will later be converted into the actual file used in DCAL by the TOINDEX application. A CHKDAUS application is initiated immediately following the BUILDEM application to check for the inclusion of all daughter products in the INDEX file. This application generates a separate file call MISSDAUS.DAT to flag users of missing daughter products. Following the CHKDAUS application the SORTINDEX application is used to sort the precursor INDEX file alphabetically by radionuclide name. After this has been completed the TOINDEX application converts the precursor INDEX file into the one used by DCAL. This conversion is a matter of replacing the name of daughter products by their record number in the INDEX file (Eckerman 2001).

APPENDIX F

Internal Dose Coefficient Calculations

Prior to initiating a dose coefficient calculation the user must define the systemic biokinetic files that will be utilized by the ACTACAL module during the computational process. In this work the systemic biokinetic models and f_1 values are based on models and assumptions given in ICRP Publication 68 (ICRP 1994b).

Invoking the ACTACAL module initiates an interactive session which calculates activity as a function of time in the compartments specified in the biokinetic data files for a given radionuclide. ACTACAL performs the following functions: (1) prompts you to describe the intake scenario, such as the parent radionuclide, exposure mode, and, for the inhalation case, size absorption type or solubility classification of inhaled particles; (2) uses the biokinetic files to determine model compartments, transfer rates between compartments, and source organs to be used in dose calculations; and (3) calculates activity of the parent and radioactive progeny in each source organ as a function of time. There were two groups of individuals evaluated in this study and they included the adult workers and members of the public. Specific responses to interactive prompts in the ACTACAL module varied according to the group of individuals being evaluated. For the occupational worker, specific responses included: (1) one acute intake age of 7300 days (20 years), (2) the selection of the equivalent dose option, (3) the selection of "no" for a compartment and source region activity computation, (4) acute intake routes of inhalation (h) and ingestion (g) for each radionuclide, (5) the selection of the ICRP Publication 66

(ICRP 1994a) lung model for inhalation cases, (6) absorption types of fast (f), medium (m), and slow (s) as required, and (7) input Activity Median Aerodynamic Diameters (AMAD) of 1.0 and 5.0 microns based on an occupational exposure scenario. For members of the public, specific responses include: (1) the use of the ICRP 56 age groups (100 d, 1 y, 5 y, 10 y, 15 y, and adult), (2) the selection of the equivalent dose option, (3) the selection of "no" for a compartment and source region activity computation, (4) acute intake routes of inhalation (h) and ingestion (g) for each radionuclide, (5) the selection of the ICRP 66 (ICRP 1994a) lung model for inhalation cases, (6) absorption types of fast (f), medium (m), and slow (s) as required, and (7) an input AMAD of 1.0 micron based on an environmental exposure scenario. In cases were radioactive progeny exist, the ICRP 30 shared kinetics approach was assumed for both groups of individuals.

Pressing "enter" initiates the SEE calculations by the module SEECAL. Once this module has been initiated no additional information is required from the user. As SEE calculations start, SEECAL writes brief credits to the screen, such as the version of the code and the names of the authors. Next, a "major program loop" is entered where the photon Specific Absorbed Fractions (SAFs) and the electron and alpha Absorbed Fractions (AFs) are read for the radiation types associated with the parent radionuclide and decay chain members. These values are read for the ages specified by the user in the ACTACAL module. Once these values are read, the module proceeds to calculate the SEE for each radionuclide with the results being written to an output file. The program ends with a message that SEECAL has ended normally, and pressing any key will return the user to the DCAL main menu. The third menu item, Compute Dose (EPCAL), will then be highlighted.

Pressing “enter” initiates the Compute Dose calculations of the EPCAL module. The principle task of the EPCAL module is to combine the time and age specific activities calculated by ACTACAL with the age specific SEE values calculated by SEECAL to calculate dose rates to target organs. In the case of this research the output is in the form of equivalent dose rate calculations since the equivalent dose option was selected in ACTACAL. As with the SEECAL module, once the EPCAL is initiated no additional information is required from the user. As EPCAL calculations start, EPCAL writes brief credits to the screen, such as the version of the code and the names of the authors. Next, information on the radiation types associated with the parent radionuclide and decay chain members appears on the screen and is updated as progress is made by EPCAL. The results are written to an output file followed by a message indicating that the program has ended normally. Pressing any key will return the user to the DCAL main menu.

The Tabulate Dose Coefficient (HTAB) utility is used to generate a concise table of dose coefficients, similar in form to the tables provided in the ICRP’s series documents on doses to the public from intake of radionuclides (ICRP 1989, 1993, 1995a, 1995b, 1996). Highlighting this utility on the main menu and pressing “enter” initiates the utility. Next, a screen will appear indicating that HTAB is working on the file that was produced as a result of the calculations performed by the EPCAL module. After HTAB has finished working on the file the results are written to an output file. Pressing any key will return the user to the DCAL main menu. These results, as well as any files generated during the computational process, can be viewed using the DCAL Work File (LIST) utility.

APPENDIX G

External Dose Coefficient Calculations

Selecting EXTCAL from the DCAL main menu will initiate an external dose calculation. Following the credit display, a prompt appears requiring the user to indicate how radionuclides will be entered into the system. A template file can be created or the user can input individual radionuclides into the system. In this work, radionuclides were input individually. Once the radionuclides have been entered into the system, a source media must be chosen. Air and soil source media options were selected for each of the radionuclides evaluated in this study. If the air media option was selected then no additional information was required to complete the computational process. The soil option, however, required additional information regarding the distribution of the radionuclide in the media. Distributional options included surface or volume and both were investigated. If the surface option was selected then no additional information was required to complete the computational process. The volume option, however, required additional information regarding the depth to which the radionuclide was distributed in the media. In this work that depth was assumed to be infinite and the corresponding option of infinite thickness was thus selected. Finally, once all the appropriate selections have been made for a given media option calculations proceed resulting in an output file being generated. Pressing any key returns the user to the DCAL main menu. As previously noted, results can be viewed using the DCAL Work File (LIST) utility.

REFERENCES

Audi G, Bersillon O, Blachhot J, Wapstra AH. The Nubase evaluation of nuclear and decay properties. Nuclear Physics. A624: 1; 1997.

Audi G, Wapstra AH. The 1995 update to the atomic mass evaluation. Nuclear Physics A595: 409; 1995.

Burrows TW, Evaluated Nuclear Structure Data File. Nucl. Instrum. Meth. 286: 595; 1990.

Dillman LT, EDISTR-A Computer Program to Obtain a Nuclear Data Base for Radiation Dosimetry. Oak Ridge National Laboratory, Oak Ridge, TN: ORNL/TM-6689; 1980.

Eckerman KF, DECDAT: Readme.1st file, Oak Ridge National Laboratory, Oak Ridge, TN; 2001.

Eckerman KF, Leggett RW, Cristy M, Nelson CB, Ryman JC, Sjoreen AL, Ward RC. DCAL: User's Guide to the DCAL System. Oak Ridge National Laboratory, Oak Ridge, TN: ORNL/TM-2001/190; 2001.

Eckerman KF, Ryman JC. External Exposure to Radionuclides in Air, Water, and Soil. Federal Guidance Report No. 12. Oak Ridge, TN: Oak Ridge National Laboratory; prepared for U.S. Environmental Protection Agency; EPA 402-R-93-081; 1993.

Eckerman KF, Wolbarst AB, Richardson CB. Limiting values of radionuclide intake and air concentration and dose conversion factors for inhalation, submersion, and ingestion. Federal Guidance Report No. 11. Oak Ridge, TN: Oak Ridge National Laboratory; prepared for U.S. Environmental Protection Agency; EPA 520/1-88-020; 1988.

Endo A, Yamaguchi Y. Compilation of new nuclear decay data files used for dose calculation. J. Nucl. Sci. Technol. 38: 689-696; 2001.

Federal Guidance Report No. 13. Cancer Risk Coefficients for Environmental Exposure to Radionuclides: CD supplement, EPA-402-C-99-001, Rev 1, U.S. Environmental Protection Agency, Washington, DC; 2002.

Hechanova A, Johnson E, Cerefice G. University of Nevada, Las Vegas transmutation research program annual report 2001. Harry Reid Center for Environmental Studies, Las Vegas, NV; 2001.

International Commission on Radiation Units and Measurements. Quantities and units in radiation protection dosimetry. Bethesda, MD: ICRU; ICRU Report 51; 1993.

International Commission on Radiological Protection. Report of the Task Group on Reference Man. Oxford: Pergamon Press; ICRP Publication 23; 1975.

International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publications 30, Part 1; Ann ICRP 2(3/4); 1979.

International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publication 30, Part 2; Ann ICRP 4(3/4); 1980.

International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publications 30, Part 3; Ann ICRP 6(2/3); 1981.

International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publications 30, Part 4; Ann ICRP 19(4); 1988.

International Commission on Radiological Protection. Human respiratory tract model for radiological protection. Oxford: Pergamon Press; ICRP Publication 66; Ann ICRP 24(1-3); 1994.

International Commission on Radiological Protection. Dose coefficients for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publication 68; Ann ICRP 24(4); 1995.

International Commission on Radiological Protection. Age-dependent doses to members of the public from intake of radionuclides: Compilation of ingestion and inhalation dose coefficient. Oxford: Pergamon Press; ICRP Publication 72; Ann ICRP 26(1); 1996.

International Commission on Radiological Protection. The ICRP database of dose coefficients: workers and members of the Public, Version 2.01. CD-ROM Prepared by the Task Group on Dose Calculations of Committee 2 of the International Commission on Radiological Protection. Oxford: Pergamon Press; distributed by Elsevier Science Ltd; 2001.

Kawai K, Endo A, Noguchi H. Dose coefficients for radionuclides produced in high energy proton accelerators facilities: coefficients for radionuclides not listed in ICRP

publications. Japan Atomic Energy Research Institute: JAERI-Data/Code 2002-013; 2002.

Tuli JK. Evaluated Nuclear Structure Data File: a manual for the preparation of data sets. Brookhaven National Laboratory, Upton, Long Island, New York: BNL-NCS-51655; 1987.

VITA

Graduate College
University of Nevada, Las Vegas

John P. Shanahan

Home Address:
1013 Snow Bunting Court
Henderson, Nevada 89015

Degrees:
Bachelor of Science, Nuclear Medicine Technology, 1993
State University of New York at Buffalo

Special Honors and Awards
H. Wade Patterson Award

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

Thesis Examination Committee:
Chairperson, Dr. Phillip W. Patton, Ph. D.
Committee Member, Dr. Keith F. Eckerman, Ph. D.
Committee Member, Dr. Mark J. Rudin, Ph. D.
Graduate Faculty Representative, Dr. Richard D. Tandy, Ph. D.

I. Database Comparison Worksheet Analysis

A. Scoring System Notes

1. The scoring system was developed to:

a) Carry out a quantitative comparison of two nuclear decay databases (ENSDF and NUBASE) for selected radionuclides and their associated decay chains.

b) Determine if the information in the ENSDF library was accurate and up to date compared to the NUBASE database found in the NUCLEUS software package.

c) Insure that each member of the decay chain was referenced by or had a corresponding ENSDF decay data file before a dose coefficient calculation was performed.

d) Serve as a reference tool for users of the EDISTR code to update INPUT.ENS files before performing a dose coefficient calculation.

2. The data comparison worksheets serve as a platform to carry out a quantitative comparison analysis of specific nuclear decay data from the ENSDF library and NUBASE database for individual members of a nuclear decay chain. The outcome of this analysis is shown under the parameters respective initial results column. Individual parameter results are then tabulated and summarized for the entire decay chain in the classification worksheet. Concurrently, a global analysis of all the parameters is also occurring in the classification worksheet and is used to place the decay chain into one of three categories.

3. Updates allow the user to:

a) Enter any missing decay data or clarify any observed discrepancies during the initial evaluation of a parameter as a result of Nuclear Science Reference (NSR) search or after utilizing a Brookhaven National laboratory (BNL) data analysis software package.

b) Eliminate the comparison methodology in evaluating a nuclear decay parameter.

4. Updated results are based only on the information entered for updates and no comparison is made with any other information on the data comparison worksheet. It is assumed that updated information is correct and will override the initial results when they are summarized in the classification worksheet.

B. Database Comparison Worksheet

1. Date of data file

a) ENSDF is formatted as year/month e.g. 200210.

b) NUCLEUS/NUBASE:

(1) NUCLEUS is the software package.

(2) NUBASE is the database within the software package.

(3) Software version 08, July 2002

2. Decay Mode(s)

a) Method of Analysis

(1) Initial Results: A binary score is generated after the appropriate decay mode(s) are selected from the drop down menu. A score of zero indicates that information was entered for both databases and that these values are in agreement. A score of one indicates that information is missing, unknown, double beta decay, or that the values entered are not in agreement.

(2) Updated Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that the information was within acceptable limits. A score of one indicates

that the information was not within acceptable limits.

3. Branching Fraction(s)

    a) Method of Analysis

        (1) Initial Results: A score of one is generated if information is missing or unknown from one of the databases; otherwise a percent difference is calculated.

        (2) Updated Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that the information was within acceptable limits. A score of one indicates that the information was not within acceptable limits.

4. Excitation Energy

    a) Method of Analysis

        (1) Initial Results: A score of one is generated if information is missing or unknown from one of the databases; otherwise a percent difference is calculated.

        (2) Updated Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that the information was within acceptable limits. A score of one indicates that the information was not within acceptable limits.

5. Half Life

    a) Method of Analysis

        (1) Initial Results: A score of one is generated if information is missing or unknown from one of the databases; otherwise a percent difference is calculated.

(2) Updated Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that the information was within acceptable limits. A score of one indicates that the information was not within acceptable limits.

6. Q-value(s)

a) Method of Analysis

(1) Initial Results: A score of one is generated if information is missing or unknown from one of the databases; otherwise a percent difference is calculated.

(2) Updated Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that the information was within acceptable limits. A score of one indicates that the information was not within acceptable limits.

7. Spin and Parity

a) Method of Analysis

(1) Initial Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that information was entered for both databases and that these values are in agreement. A score of one indicates that information is missing, unknown, or that the values entered are not in agreement.

(2) Updated Results: A binary score is generated after the appropriate information is entered. A score of zero indicates that the information was within acceptable limits. A score of one indicates that the information was not within acceptable limits.

II. Classification Worksheet Analysis

A. Classification Worksheet

1. Primary Decay Mode

a) A good correlation indicates that each member of the decay chain has a corresponding reference in the ENSDF library for this value, and the sum of the binary scores is equal to zero after the entire chain has been evaluated.

b) A poor correlation indicates either one or more members of the decay chain do not have a corresponding reference in the ENSDF library for this value, or the sum of the binary scores is not equal to zero after the entire chain has been evaluated.

2. Secondary Decay Mode

a) A good correlation indicates that each member of the decay chain has a corresponding reference in the ENSDF library for this value, and the sum of the binary scores is equal to zero after the entire chain has been evaluated.

b) A poor correlation indicates either one or more members of the decay chain do not have a corresponding reference in the ENSDF library for this value, or the sum of the binary scores is not equal to zero after the entire chain has been evaluated.

3. Branching Fraction

a) A good correlation indicates that each member of the decay chain has a corresponding reference in the ENSDF library for this value, and less than one percent difference exists between the databases for any member of the decay chain after the entire chain has been evaluated.

b) A poor correlation indicates either one or more members of the decay chain do not have a

corresponding reference in the ENSDF library for this value, or one or more members of the decay chain show a percent difference greater than one percent after the entire chain has been evaluated.

4. Excitation Energy

   a) A good correlation indicates that each isomeric member of the decay chain has a corresponding reference in the ENSDF library for this value, and less than one percent difference exists between the databases for any isomeric member of the decay chain after the entire chain has been evaluated.

   b) A poor correlation indicates either one or more isomeric members of the decay chain do not have a corresponding reference in the ENSDF library for this value, or one or more isomeric members of the decay chain show a percent difference greater than one percent after the entire chain has been evaluated.

5. Half Life

   a) A good correlation indicates that each member of the decay chain has a corresponding reference in the ENSDF library for this value, and less than one percent difference exists between the databases for any member of the decay chain after the entire chain has been evaluated.

   b) A poor correlation indicates either one or more members of the decay chain do not have a corresponding reference in the ENSDF library for this value, or one or more members of the decay chain show a percent difference greater than one percent after the entire chain has been evaluated.

6. Q value(s)

   a) A good correlation indicates that each member of the decay chain has a corresponding reference in the ENSDF library for this value, and less than one percent difference exists between the databases for any

member of the decay chain after the entire chain has been evaluated.

b) A poor correlation indicates either one or more members of the decay chain do not have a corresponding reference in the ENSDF library for this value, or one or more members of the decay chain show a percent difference greater than one percent after the entire chain has been evaluated.

7. Spin and Parity

a) A good correlation indicates that each member of the decay chain has a corresponding reference in the ENSDF library for this value, and the sum of the binary scores is equal to zero after the entire chain has been evaluated.

b) A poor correlation indicates either one or more members of the decay chain do not have a corresponding reference in the ENSDF library for this value, or the sum of the binary scores is not equal to zero after the entire chain has been evaluated.

B. Categories

1. Category Notes:

a) The objective of this scoring system is to generate a comprehensive global score for the entire decay chain so that it can be easily identified, categorized, and referenced.

b) Decay chains are placed into one of three categories depending on the results of individual parameter scores tabulated and summarized on the classification worksheet. These categories are briefly summarized below.

2. Category 0

a) Category zero is the default score and is used as a reference flag for empty workbooks.

3. Category 1

    a) Category one indicates that all parameters summarized on the classification worksheet showed good correlation and that the decay chain can be worked up for a dose coefficient calculation.

4. Category 2

    a) Category two indicates that one or more of the following parameters showed poor correlation after being summarized on the classification worksheet: branching fraction, excitation energy, half life, or spin and parity. This category is used to identify decay chains that have complete data sets but require further research to clarify discrepancies between the databases.

5. Category 3

    a) Category three indicates that ENSDF does recognize or reference a primary or secondary mode of decay for one or more members of the decay chain. This category is used to identify decay chains that can not be worked up for a dose coefficient calculation.

Chain	Radionuclide	ENSDF Record Comments
Er-157	Er-157	Added T1/2 (12.6 M 2) to Level 0.
	Ho-157	Added T1/2 (8.14 H 4) to Level 0.
		Updated Level (199.34->199.5):
		added T1/2 (21.6 MS 16).
	Dy-157m	Added T1/2 (8.14 H 4) to Level 0.
	Dy-157	Added T1/2 (71 Y 7) to Level 0.
	TB-157	Added "STABLE" to Level 0.
Er-160	Er-160	
	Ho-160m(IT)	
	Ho-160(25.6 M+5.02 H)	Added "STABLE" to Level 0.
	Ho-160	Added "STABLE" to Level 0.
Fe-61	Fe-61	Added T1/2 (1.650 H 5) to Level 0.
	Co-61	Added "STABLE" to Level 0.
Gd-144	Gd-144	
	Eu-144	Added "STABLE" to Level 0.
Hf-171	Hf-171	Missing E records.
	Lu-171m	
	Lu-171	
Ir-197	Ir-197	Missing B records.
	Pt-197m(IT)	
	Pt-197m(B-)	
	Pt-197	
	Au-197m	
La-127	La-127	Missing E records.
	Ba-127m	
	Ba-127	
	Cs-127	
	Xe-127m	
	Xe-127	
La-128	La-128	
	Ba-128	
	Cs-128	Modified T1/2(3.62 M -> 3.66 M):
		Header and P record.
Lu-168	Lu-168	Missing E records.
Os-176	Os-176	Updated Q-value (3100->2960).
		Missing E records.
		Added T1/2 (5.3 M 3)
		and Spin and Parity (3+) to Level 0.
	Re-176	
	W-176	Updated Q-value (820->790).
		Missing E records.
	Ta-176	Added "STABLE" to Level 0.

Os-178	Os-178	Updated Q-value (2330->2210).
		Missing L and E records.
	Re-178	Added T1/2 (21.6 D 3) to Level 0.
	W-178	
	Ta-178	Added "STABLE" to Level 0.
Pb-195	Pb-195	N record missing data.
		Added T1/2 (1.16 H 5) to Level 0.
	Tl-195	
	Hg-195m(IT)	
	Hg-195m(EC)	Updated Q-value (1510->1571).
	Hg-195	Updated Q-value (1510->1571).
	Au-195m	
	Au-195	
Pm-153	Pm-153	
	Sm-153	Updated T1/2 (46.50 H 21->46.27 H 1).
Pr-133	Pr-133	Updated Q-value (4330->4488).
		Added T1/2 (97 M 4) to Level 0.
		Missing E records.
	Ce-133	Updated Q-value (2.94E+3->2900).
	La-133	
	Ba-133m(IT)	
	Ba-133m(EC)	
	Ba-133	
Pt-201	Pt-201	Added T1/2 (26 M 1) to Level 0.
	Au-201	Added "STABLE" to Level 0.
Re-176	Re-176	
	W-176	Updated Q-value (820->790).
		Missing E records.
	Ta-176	Added "STABLE" to Level 0.
Sb-113	Sb-113	
	Sn-113m(IT)	Updated P record Level (77->77.39 2).
		Updated Level (77->77.39 2).
	Sn-113m(EC)	
	Sn-113	
	In-113m(IT)	
Ta-167	Ta-167	Missing E records.
	Hf-167	
	Lu-167	
	Yb-167	
	Tm-167	
	Er-167m	

Ta-171	Ta-171	Updated Q-value (3740->3700).
		Missing E records.
	Hf-171m	
	Hf-171	Missing E records.
	Lu-171m	
	Lu-171	
Tl-192	Tl-192	Missing E records.
	Hg-192	Updated Q-value (700->744).
	Au-192	
Tl-193	Tl-193	Updated Q-value (3640->3560).
		Missing E records.
	Hg-193	Updated Q-value (2534->2340).
	Au-193m(IT)	
	Au-193m(EC)	Added T1/2 (50 Y 9) to Level 0.
	Au-193	
	Pt-193m(IT)	
	Pt-193	
Tm-157	Tm-157	Missing E records.
		Added T1/2 (18.65 M 10) to Level 0.
	Er-157	Added T1/2 (12.6 M 2) to Level 0.
	Ho-157	Added T1/2 (8.14 H 4) to Level 0.
		Updated Level (199.34->199.5)
		added T1/2 (21.6 MS 16).
	Dy-157m	Added T1/2 (8.14 H 4) to Level 0.
	Dy-157	Added T1/2 (71 Y 7) to Level 0.
	TB-157	Added "STABLE" to Level 0.
Tm-160	Tm-160	Updated Q-value (5890->5600).
		Missing E records.
	Er-160	
	Ho-160m(IT)	
	Ho-160(25.6 M+5.02 H)	Added "STABLE" to Level 0.
	Ho-160	Added "STABLE" to Level 0.
Tm-161	Tm-161	Updated T1/2 (8 US->7.5 US 7),
		396.44 Level.
	Er-161m(IT)	Updated P record Level:
		(397->396.44 4)
	Er-161	Added T1/2 (6.76 S 7) to 211.16 Level
	Ho-161m(IT)	
	Ho-161	

W-171	W-171	Updated Q-value (4560->4660).
	Ta-171	Updated Q-value (3740->3700).
		Missing E records.
	Hf-171m	
	Hf-171	Missing E records.
	Lu-171m	
	Lu-171	
W-172	W-172	Added T1/2 (36.8 M 3) to Level 0.
		Missing E records.
	Ta-172	Added T1/2 (1.87 Y 3) to Level 0.
	Hf-172	Added T1/2 (6.70 D 4) to Level 0.
		Added T1/2 (3.7 M 5) to Level 41.86.
	Lu-172m(IT)	Added T1/2 (1.87 Y 3) to Level 0.
	Lu-172n(IT)	Added T1/2 (1.87 Y 3) to Level 0.
		Added T1/2 (3.7 M 5) to Level 41.86.
	Lu-172	Added "STABLE" to Level 0.
W-173	W-173	Added T1/2 (3.14 H 13) to Level 0.
	Ta-173	Updated Q-value (2790 SY->2690 200)
	Hf-173	
	Lu-173	
W-174	W-174	Missing L and E records.
	Ta-174	Added T1/2 (2.0E+15 Y 4) to Level 0.
	Hf-174	Added "STABLE" to Level 0.
Xe-119	Xe-119	Updated Q-value (5000->4880).
		Data missing in N record.
		Missing E records.
	I-119	
	Te-119m(EC)	
	Te-119	
	Sb-119	
Yb-161	Yb-161	Updated Q-value (4150->4200).
		Changed T1/2 (38 M 4->30.2 M 8),
		Level 0.
	Tm-161	Updated T1/2 (8 US->7.5 US 7),
		396.44 Level
	Er-161m(IT)	Updated P record Level:
		(397->396.44 4).
	Er-161	Added T1/2 (6.76 S 7) to 211.16 Level
	Ho-161m(IT)	
	Ho-161	

Chain	Radionuclide	Energy Balance Data (Percent Error)
Er-157	Er-157	2.36
	Ho-157	1.51
	Dy-157m	2.67
	Dy-157	1.42
	TB-157	0.00
Er-160	Er-160	6.82
	Ho-160m	0.33
	Ho-160	0.16
Fe-61	Fe-61	3.07
	Co-61	0.00
Gd-144	Gd-144	0.11
	Eu-144	0.00
La-128	La-128	3.30
	Ba-128	0.01
	Cs-128	0.00
Pm-153	Pm-153	0.99
	Sm-153	0.13
Pt-201	Pt-201	44.64
	Au-201	0.19
Sb-113	Sb-113	0.77
	Sn-113m	0.21
	Sn-113	0.43
	In-113m	0.00
Tm-161	Tm-161	22.77
	Er-161m	69.48
	Er-161	3.48
	Ho-161m	0.00
	Ho-161	0.08
W-173	W-173	8.54
	Ta-173	0.09
	Hf-173	0.15
	Lu-173	0.33
Yb-161	Yb-161	0.51
	Tm-161	22.77
	Er-161m	69.48
	Er-161	3.48
	Ho-161m	0.00
	Ho-161	0.08

Radionuclides with Unpublished Dose Coefficients (half-life less than one minutes).

Z	A	Nuclide	T 1/2	Units
4	8	Be-8	7.70E-16	s
5	13	B-13	17.36	ms
5	12	B-12	20.2	ms
56	136	Ba-136m	0.3084	s
2	6	He-6	806.7	ms
3	8	Li-8	838	ms
79	191	Au-191m	0.92	s
70	155	Yb-155	1.8	s
63	142	Eu-142	2.4	s
78	175	Pt-175	2.54	s
80	180	Hg-180	2.56	s
79	178	Au-178	2.6	s
69	154	Tm-154	3.3	s
76	169	Os-169	3.4	s
80	181	Hg-181	3.6	s
81	195	Tl-195m	3.6	s
7	17	N-17	4.173	s
77	172	Ir-172	4.4	s
74	165	W-165	5.1	s
74	183	W-183m	5.2	s
72	159	Hf-159	5.6	s
78	176	Pt-176	6.3	s
79	179	Au-179	7.1	s
7	16	N-16	7.13	s
76	170	Os-170	7.3	s
79	197	Au-197m	7.73	s
76	171	Os-171	8	s
77	176	Ir-176	8	s
77	198	Ir-198	8	s
77	173	Ir-173	9	s
77	174	Ir-174	9	s
77	175	Ir-175	9	s
61	140	Pm-140	9.2	s
75	170	Re-170	9.2	s
63	144	Eu-144	10.2	s
68	152	Er-152	10.3	s
80	182	Hg-182	10.83	s
78	177	Pt-177	11	s
81	184	Tl-184	11	s

79	181	Au-181	11.4 s
77	178	Ir-178	12 s
72	160	Hf-160	13.6 s
4	11	Be-11	13.81 s
49	116	In-116	14.1 s
75	172	Re-172	15 s
79	182	Au-182	15.6 s
75	192	Re-192	16 s
76	173	Os-173	16 s
72	161	Hf-161	18.2 s
74	166	W-166	18.8 s
76	172	Os-172	19.2 s
6	10	C-10	19.255 s
78	178	Pt-178	21.1 s
78	179	Pt-179	21.2 s
69	155	Tm-155	21.6 s
65	146	Tb-146	23 s
68	151	Er-151	23.5 s
82	188	Pb-188	24 s
70	156	Yb-156	26 s
8	19	O-19	26.96 s
81	186	Tl-186	27.5 s
79	202	Au-202	28.8 s
77	177	Ir-177	30 s
79	195	Au-195m	30.5 s
80	184	Hg-184	30.6 s
55	124	Cs-124	30.8 s
73	166	Ta-166	34.4 s
67	151	Ho-151	35.2 s
68	153	Er-153	37.1 s
70	157	Yb-157	38.6 s
64	143	Gd-143	39 s
47	109	Ag-109m	39.6 s
79	204	Au-204	39.8 s
63	141	Eu-141	40 s
61	142	Pm-142	40.5 s
79	183	Au-183	42 s
76	174	Os-174	44 s
61	136	Pm-136	47 s
80	185	Hg-185	49 s
78	181	Pt-181	51 s
82	189	Pb-189	51 s

77	196	Ir-196	52 s
78	180	Pt-180	52 s
79	184	Au-184	53 s
79	203	Au-203	53 s
57	126	La-126	54 s

Committed Effective Dose Coefficients (Sv/Bq): Ingestion.

Radionuclide	Calculated	JAERI_02	Percent Error
Au-201	2.44E-11	2.44E-11	0.00
Co-61	7.51E-11	7.51E-11	0.00
Nd-144	4.08E-08	4.08E-08	0.00
S-38(Organic)	2.66E-10	2.66E-10	0.00
S-38(Inorganic)	6.09E-10	6.09E-10	0.00
V-50	3.41E-09	3.41E-09	0.00