

2002

Development of Dose Coefficients for Radionulides Produced in Spallation Neutron Sources: Annual Report

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Patton, P. W., Rudin, M. (2002). Development of Dose Coefficients for Radionulides Produced in Spallation Neutron Sources: Annual Report.

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Annual Report

Development of Dose Coefficients for Radionuclides Produced in Spallation Neutron Sources

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Graduate Students: John P. Shanahan and Yayun Song

DOE Collaborator: Keith F. Eckerman, Oak Ridge National Laboratory

Other Collaborators: Faculty and students from Idaho State University, Georgia Institute of Technology, and Tbilisi State University, and faculty from the University of Florida.

Goals and Background

The University of Nevada, Las Vegas (UNLV) Transmutation Research Program has been tasked to support U.S. Department of Energy (DOE) efforts to assess the health risks associated with the operation of each of their accelerator-driven nuclear facilities for both NEPA and PSAR development. Quantifying the radiological risks to workers will have to be addressed during the design and siting of each of these facilities. U.S. 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 0.05 Sv/yr for stochastic effects, or a committed dose equivalent to an individual organ or tissue of 0.5 Sv/yr for deterministic 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 (EPA 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 dose equivalent to an organ or tissue per unit intake, the committed effective dose equivalent per unit intake, the dose equivalent rate per unit air concentration of radionuclide, or the effective dose equivalent rate per unit air concentration of radionuclide. These dose coefficients (DCs) allow simple determination of radiation dose associated with various exposure scenarios, and ultimately, assess the health risks to workers in a nuclear facility.

Even though the ALIs, DACs, and DCCs calculated in Federal Guidance Report No. 11 adhere to the derived limits in Publication 30 (ICRP 1979), which incorporate current knowledge of radionuclide dosimetry and biological transport in humans, the report is not

exhaustive in reference to anthropogenic radionuclides. Unfortunately, many of the rare radionuclides produced during the spallation process are not addressed in current radiation protection standards either. There may be as many as several hundred radionuclides that would be produced in either the target or blanket of proposed accelerator facilities for which no data exists in Federal Guide Report No. 11 or in Publications 68 and 72 of the ICRP.

It is the intent of the current research to develop a methodology and generate internal and external dose coefficients for radionuclides produced in spallation neutron sources. Results from this study will expand the ALI and DAC data of Federal Guidance Report No. 11 in order to include radionuclides produced by current technology, such as that used in the AAA and SNS programs.

Project Objectives

There were four research objectives for Year 1 of this project:

- to establish a research consortium comprised of representatives from several Universities and National Laboratories
- to develop a prioritized list of radionuclides produced during the spallation process that will be considered as part of this study
- to further Georgia Institute of Technology's work on developing a reproducible methodology to determine internal and external DCs
- to generate internal and external DC values for selected radionuclides

Research Accomplishments

Each of the above project objectives were accomplished through the completion of specific tasks. Completed tasks associated with each objective are identified below:

Objective 1 – Establish a research consortium comprised of representatives from several Universities and National Laboratories

Work performed under this project has continued to draw upon the experience and expertise residing at a number of respected health physics academic programs across the United States and representatives from DOE national laboratories. Faculty, and students from the following academic institutions are currently participants in the consortium: Georgia Institute of Technology, Idaho State University, University of Florida, and the University of Nevada, Las Vegas (UNLV). Oak Ridge National Laboratory (ORNL) also has representation on the Consortium.

Efforts were initiated in Year 1 to add representatives from other countries to serve as members on the TRP DC Consortium. For example, faculty and students from Tbilisi State University in Tbilisi, Georgia have participated in a number of project activities and have been invited to formally become members of the Consortium. Project personnel will continue to work to expand the number of participants and role of the existing TRP DC Consortium in a reasonable manner.

A TRP DC Working Group was established in Year 1 to direct and oversee consortium activities. The following individuals served as members of the Working Group in Year 1:

Phillip Patton ,UNLV, Project Coordinator
Wesley Bolch, University of Florida
Richard Brey, Idaho State University
Adam Arndt, Idaho State University
Keith Eckerman, ORNL
Tom Gesell, Idaho State University
Nolan Hertel, Georgia Tech
Omar Wooten, Georgia Tech
Samson Pagava, Tbilisi State University
Mark Rudin, UNLV

The Project Coordinator was responsible for scheduling and hosting the two DC Working Group meetings in Las Vegas, NV (January and May 2002). It should be emphasized that Working Group members and all participating members of the consortium worked collaboratively to complete all tasks associated with the project. UNLV personnel will host future working group meetings periodically to continually encourage collaboration and ensure project activities are completed in a timely manner.

Objective 2 – Develop a prioritized list of radionuclides produced during the spallation process that will be considered as part of this study

The DC Working Group decided initially to concentrate efforts to on generating DCs for those radionuclides that could be produced from the spallation of a liquid mercury target for which no DCs currently exist. Over 520 radionuclides were identified as possible by product materials, all of which have the potential to be involved in various radiation exposure scenarios involving workers. Prioritization of the 520 radionuclides included tabulating the list according to half-life and obtaining further physical information on each radionuclide (e.g., mode of decay, etc.), and determining which radionuclides currently have no DCs published in the literature. The Working Group recommended focusing on those radionuclides with half-lives between one and ten minutes. Radionuclides from the list with half-lives less than one minute and greater than ten minutes may be considered at a later date. The prioritization effort identified 86 radionuclides from the original list of 520 that will be considered for this phase of the project.

Objective 3 - Further Georgia Institute of Technology's work on developing a reproducible methodology to determine internal and external DCs

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 (Oak Ridge National Laboratory). A methodology to take the output of EDISTR and generate

nuclear decay data files suitably formatted for use by DCAL (Dose and Risk Calculation software) was developed by personnel from the Georgia Institute of Technology. This methodology was later replaced by a series of MS-DOS executables found in the DECDAT (Decay Data) directory and introduced by Keith Eckerman at the student workshop.

At the present time the group has adopted the methodology used to develop Federal Guidance Report 13 from the U.S. Environmental Protection Agency (EPA). In this methodology, relevant nuclear decay data is extracted from ENSDF (Evaluated Nuclear Structure Data File) and used by the computer code EDISTR to calculate mean energies and absolute intensities of all principal radiations associated with the radioactive decay of a radionuclide. The executables of DECDAT arrange the output of EDISTR into the proper format for inclusion into the nuclear data libraries within DCAL. DCAL consists of a series of computational modules for the calculation of dose and risk coefficients. The DCAL system also includes extensive libraries of biokinetic, anatomical, and dosimetric data representing the current state-of-the-art. Specific information regarding each step of the methodology is presented in the *Dose Coefficient (DC) Methodology Report* that was developed as part of this project.

Objective 4 - Generate internal and external DC values for selected radionuclides

Internal dose coefficients for inhalation and ingestion scenarios were determined using the methodology described above for 5 of the 86 radionuclides identified by the DC Working Group. The five radionuclides selected for this report are presented in Table 1 as well as the source of the nuclear decay data sets. Source information can be found and is documented in the journal *Nuclear Data Sheets*.

Table 1. Radionuclides included for evaluation in current study.
Ingestion and inhalation of particulates

Atomic Number Data Set	Nuclide	Physical Half-Life	Source of the Nuclear Decay
26	Fe-61	5.98m	ENSDF
51	Sb-113	6.67m	ENSDF
55	Cs-123	5.94m	ENSDF
56	Ba-125	3.5m	ENSDF
62	Sm-139	2.57m	ENSDF

The NUBASE database was utilized for these selected radionuclides to note primary and secondary decay chains, as well as, the status of an ENSDF for a particular radionuclide. This essentially allowed project personnel to identify “exotic” decay chains with complete decay data sets available for dose calculation purposes. It also allowed personnel to compare the information in the ENSDF database to another database to identify missing or inaccurate information in the decay data sets. Previous investigators (Endo and Yamaguchi 2001) effectively utilized this database to reexamine and update the decay data sets for the 817 radionuclides that are listed in ICRP Publication 38, 6 additional isomers, and 162 additional radionuclides with half-lives ≥ 10 min. not listed in ICRP #38.

The effective dose coefficients for workers for the five radionuclides are presented in Table 2. The results are presented in the same format as ICRP Publications 68 and 72. It should be noted that the ICRP will report results to two significant figures and Table 2 contains the output results from DCAL which are given in three significant figures. As shown in Table 2, dose coefficients for inhalation of 1 μm and 5 μm particulates and ingestion are presented along with the f_1 values and absorption types.

Calculating external dose coefficients for environmental exposure scenarios for the 5 selected radionuclides were not performed at this time. The software version of DCAL did allow for these calculations to occur within the framework of the EXTCAL module but only for those radionuclides identified in FGR 13. In other words, these calculations could only be performed on those radionuclides included in the nuclear decay data libraries that came with the software package. The user’s manual did provide instructions on how to calculate an external dose coefficient for radionuclides not listed in the nuclear decay data libraries once the appropriate data files were built (e.g. ICRP38.NDX, ICRP38.BET, and ICRP38.RAD). Not included in this version of the software package was the example file EXTLIST.INP, which would allow the user to setup a template in order to carry out these calculations.

Table 3 is a biokinetic comparison of current ICRP recommendations and the output form DCAL utilizing the ICRP 68 biokinetic subdirectory default parameters. Inhalation and ingestion f_1 values and absorption types are presented for the selected elements presented in this report. As seen from the table several f_1 values do not correspond to current ICRP recommendations. It is noted that DCAL does provide the user with the flexibility to define or update existing biokinetic subdirectories and utilize these parameters during calculations.

Table 2. Effective dose coefficients for Workers - Ingestion and inhalation of particulates

Nuclide	t _{1/2}	Type	Effective dose coefficient (Sv Bq ⁻¹)				
			Inhalation, $e_{inh}(50)$			Ingestion	
			f_1	1 μ mAMAD	5 μ mAMAD	f_1	$e_{ing}(50)$
Iron							
Fe-61	5.98m	F	0.1	6.77E-12	1.14E-11	0.1	2.26E-11
		M	0.1	8.74E-12	1.44E-11		
		S	0.1	8.96E-12	1.48E-11		
Antimony							
Sb-113	6.67m	F	0.1	5.62E-12	9.54E-12	0.1	1.68E-11
		M	0.01	7.41E-12	1.22E-11		
		S	0.1	7.65E-12	1.26E-11		
Cesium							
Cs-123	5.94m	F	1.0	5.62E-12	9.32E-12	1.0	1.96E-11
		M	1.0	7.29E-12	1.19E-11		
		S	1.0	7.47E-12	1.21E-11		
Barium							
Ba-125	3.5m	F	0.1	4.91E-12	7.67E-12	0.1	7.41E-12
		M	0.1	6.33E-12	9.68E-12		
		S	0.1	6.49E-12	9.90E-12		
Samarium							
Sm-139	2.57m	F	5E-4	3.00E-12	4.98E-12	5E-4	1.01E-11
		M	5E-4	3.44E-12	5.69E-12		
		S	5E-4	3.49E-12	5.77E-12		

Table 3. Biokinetic comparison of f_1 values: ICRP versus DCAL output (bio\i68).

Nuclide	Publication	Type	Inhalation		Ingestion	
			ICRP	DCAL	ICRP	DCAL
Iron	69	F	0.1	0.1	0.1	0.1
		M	0.1	0.1		
		S	0.01	0.1		
Antimony	69	F	0.1	0.1	0.1	0.1
		M	0.01	0.01		
		S	0.01	0.1		
Cesium	56	F	1.0	1.0	1.0	1.0
		M	0.1	1.0		
		S	0.01	1.0		
Barium	67	F	0.2	0.1	0.2	0.1
		M	0.1	0.1		
		S	0.01	0.1		
Samarium	30	All	5.0E-4	5.0E-4	5.0E-4	5.04E-4

Data for ^{123}Cs from the ENSDF and NUBASE databases are presented in Figures 1 and 2, respectively. They are included in this report to illustrate the need to implement quality control measures to ensure the accuracy and credibility of the data. It is noted from the figures that both ENSDF and NUBASE report different half-life and Q values for the same radionuclide. These values are particularly important because EDISTR uses the Q-value to compute the energy of an alpha particle and the end point energy of a beta particle. It uses the half-life value to produce decay chain data as well as a reference to judge whether a daughter product is radioactive. Previous work by Endo and Yamaguchi, 2001 updated an ENSDF decay data sets when the values of half-life, branching fraction, excitation energy, and total decay energy differed by more than 1% from those of NUBASE.

Published: 1993 Nuclear Data Sheets.

 ^{123}Cs Parent: $E_x=0.0$; $J^\pi=1/2^+$; $T_{1/2}=5.94$ min 4; $Q_{g.s.\rightarrow g.s.}=4210$ 50; $\% \epsilon=100$ **History**

Type	Author	Citation	Cutoff Date
Full evaluation	S. Ohya and T. Tamura	Nuclear Data Sheets 70,531 (1993)	1-Jan-1993

Figure 1. ENSDF heading for Cs-123 EC decay data set. Note $t_{1/2}$ value of 5.94 min and a $Q_{g.s.\rightarrow g.s.}$ value of 4210 KeV.

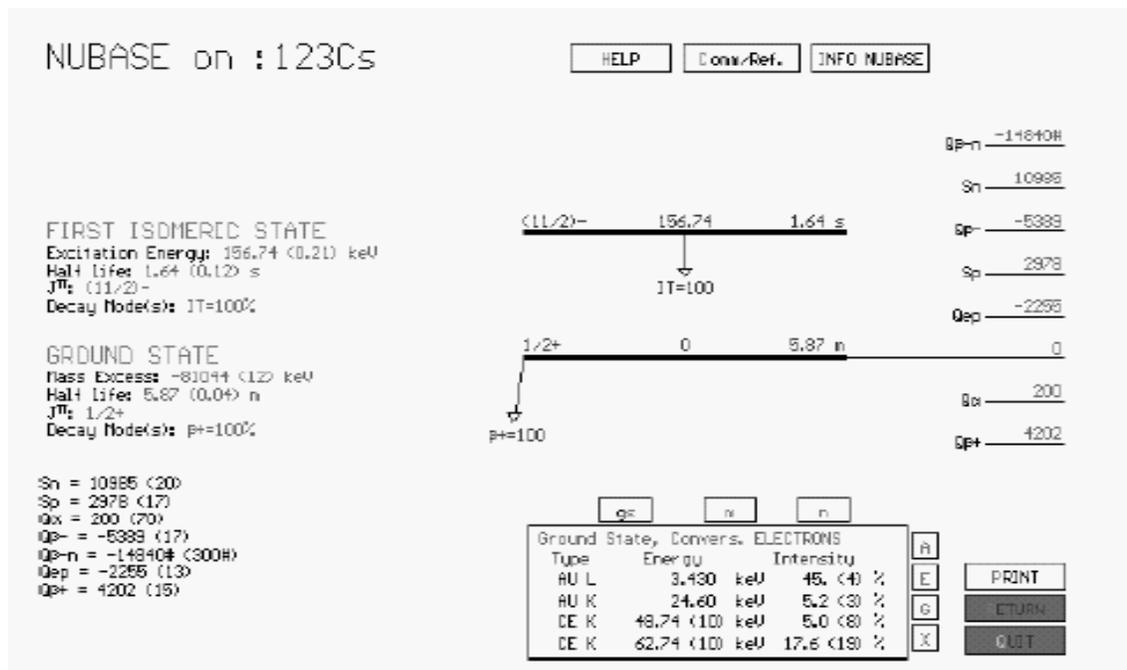


Figure 2. Radionuclide information sheet for Cs-123 from NUBASE. Note $t_{1/2}$ value of 5.87 min and a $Q_{g.s.\rightarrow g.s.}$ value of 4202 KeV. The reference (under Comm/Ref.) given for this radionuclide was the 1994 ENSDF.

It is interesting to note that the NUBASE information sheet for this particular radionuclide sites ENSDF as the reference source and yet reports a different half-life and Q value. In this particular case a more thorough evaluation of the literature would be required before a dose coefficient could be reported.

Deliverables

The following deliverables were completed during Year 1 of the project:

DC Methodology Report (August 2002)

Annual Report (September 2002)

Professional Meeting Presentations/Publications (September 2002)

Project Highlights

- The research consortium, including Georgia Institute of Technology, Idaho State University, University of Florida, UNLV, Los Alamos National Laboratory (LANL), Oak Ridge National Laboratory (ORNL), and Tbilisi State University in Tbilisi, Republic of Georgia, was established.
- A student workshop was hosted at UNLV to train graduate students from participating universities on the methodology of generating dose coefficients.

- Researchers presented the “Development of Dose Conversion Coefficients for Radionuclides Produced in Spallation Neutron Sources” at the 47th Annual Bioassay, Analytical, and Environmental Radiochemistry Conference, Las Vegas in November 2001 and at the American Nuclear Society Winter Meeting, Student Mini-Conference in Reno, NV November 2001.
- “Development of Dose Conversion Coefficients for Radionuclides Produced in Spallation Neutron Sources” was presented as a poster at the Annual Meeting of the Health Physics Society, Tampa, FL June 2002.
- Project personnel completed a report titled *Dose Coefficient (DC) Methodology Report*.
- Project personnel used the DC methodology to generate internal DCs for 6 radionuclides for which no DCs currently exist. The results are presented in the *Dose Coefficient (DC) Methodology Report*.