2001

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A. Arndt
Idaho State University

John P. Shanahan
University of Nevada, Las Vegas

C. Gold
Idaho State University

R. Brey
Idaho State University

T. Gesell
Idaho State University

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A. Arndt[1], J. Shanahan[2], C. Gold[1], R. Brey[1*], T. Gesell[1], P. Patton[2], M. Rudin[2], K. Eckerman[3], V. Rusetski[4], S. Pagava[4]

[1]Idaho State University, Pocatello, Idaho 83209 [2]University of Nevada Las Vegas, Las Vegas, Nevada, [3]Oak Ridge National Laboratory, Oak Ridge, Tennessee, 37831 [4] I. Javakhishvili Tiblisi State University, Georgia (Southern Caucasus) [*]Author to whom correspondence should be addressed

Abstract

The development of a spallation neutron source with a mercury target may lead to the production of rare radionuclides. The dose coefficients for many of these radionuclides have not yet been published. A collaboration of universities and national labs has taken on the task of calculating dose coefficients for the rare radionuclides using the software package: DCAL. The working group developed a procedure for calculating dose coefficients and a quality assurance (QA) program to verify the calculations completed. The first portion of this QA program was to verify that each participating group could independently reproduce the dose coefficients for a known set of radionuclides. The second effort was to divide the group of radionuclides among the independent participants in a manner that assured that each radionuclide would be redundantly and independently calculated. The final aspect of this program was to resolve any discrepancies arising among the participants as a group of the whole. The output of the various software programs for six QA radionuclides, ^{144}Nd, ^{201}Au, ^{50}V, ^{61}Co, ^{41}Ar, and ^{38}S were compared among all members of the working group. Initially, a few differences in outputs were identified. This exercise identified weaknesses in the procedure, which have since been revised. After the revisions, dose coefficients were calculated and compared to published dose coefficients with good agreement. The present efforts involve generating dose coefficients for the rare radionuclides anticipated to be produced from the spallation neutron source should a mercury target be employed.

Introduction

Dose coefficients; the committed effective dose per unit activity (internal) and effective dose equivalent per unit activity (external) (Sv Bq^{-1}), have been calculated for many radionuclides using Dose and Risk Calculation (DCAL) software developed in the Life Science Division at Oak Ridge National Laboratory (ORNL). DCAL software takes into account biokinetic models of organs and the parameters of reference man. The software has been used to calculate the dose coefficients found in such publications as ICRP 68, ICRP 72, FGR 12, and FGR 13 (Eckerman et al 2001, EPA 1999, ICRP 1996, ICRP 1995, EPA 1993).

The development of the Spallation Neutron Source with a mercury target has led to concerns about the production of rare radionuclides. Table 1 list the radionuclides with unpublished dose coefficients should a mercury target be employed in the proposed spallation neutron source. This list was developed by the Spallation Neutron Source (SNS) group at ORNL . The Dose Coefficient (DC)project/working group is a joint effort of the University of Nevada Las Vegas, Idaho State University, Tbilisi State University (Georgia Southern Caucasus), Georgia Institute of Technology, and Oak Ridge National Laboratory to calculate dose coefficients for these radionuclides. DC is a component of the larger Advanced Fuel Cycle Initiative (AFCI) effort.

The DC working group adopted the methodology employing the software package DCAL and the ancillary codes EDISTR, BUILDEM, CHKDAUS, SORTINDEX, and TOINDEX to calculate the dose coefficients for radionuclides found in Table 1. The procedure produces three database files that are incorporated into the DCAL software package to generate dose coefficient values. Each file requires substantial effort as the existing database files ICRP38.bet, ICRP38.rad, and ICRP38.ndx do not contain all the information on the rare radionuclides of interest. The working group was given the task of creating the three files necessary for DCAL.

Procedure

DCAL software uses three digital database files. The names of the files are: the Radiation file (.RAD), the beta spectrum file (.BET), and the index file (.NDX). The radiation file includes information on the different types of radiations potentially emitted from a particular radionuclide, the average or unique energy of each radiation emitted, and the yield of each radiation. The beta spectrum file provides information about the beta spectrum for each emitted beta particle. The index file directs DCAL to the appropriate areas of the radiation file and beta spectrum file for all members of a radionuclides decay chain. The database files are developed primarily based on information found in the Evaluated Nuclear Structure Data File (ENSDF).

ENSDF files are downloaded from the Brookhaven National Laboratory website (www.nndc.bnl.gov/nndc/ensdf/). ENSDF files contain information such as:

1. Summary of the mass chain information including the name and affiliation of the evaluator as well as their remarks and the references used in the data sets.
2. The adopted level and gamma-ray properties for each nuclide.
3. Single type experiment results such as radioactive decay or a nuclear reaction, or results for a number of experiments such as Coulomb excitation.
4. Modes of decay.
5. Half-life.
6. Q-values.
7. Spin, and parity (Tuli 1994).

ENSDF files do not exist for all radionuclides. In addition, many existing files have not been updated recently and thus are not very current. The information in the ENSDF library is updated by mass chains with a present cycle time of approximately six years, and can represent a significant source of error relative to current scientific literature. This deficit in quality nuclear structure data has put a hold on the calculation of many of the radionuclides of concern. The radionuclides which do not have existing ENSDF files are being researched to develop the fundamental nuclear physics needed for dosimetric calculations. The ENSDF files that have not been updated recently were compared to another nuclear database, NUBASE. If the comparisons between the two databases were in agreement, calculation of dose coefficients continued. If the comparisons were not in agreement, dose coefficients were calculated using both ENSDF data and NUBASE data. If an insignificant difference ($\leq$ 5% difference) exists between the dose coefficients, the more conservative of the two dose coefficients will be published. If a significant difference exists, the two dose coefficients will be published.

These data are manipulated by a computer code called EDISTR (Dillman 1980). EDISTR converts a Nuclear Structure and Decay Data file into a radioactive decay database for dosimetry purposes as described below.

EDISTR was developed at Oak Ridge National Laboratory for generating a nuclear-decay database for radiation dosimetry. EDISTR calculates:

1. The average energy of beta particles in the beta transitions
2. The beta spectrum as a function of energy
3. Energies and intensities of x-rays and auger electrons generated by radioactive decay
4. Bremsstrahlung spectra associated with beta decay and mono energetic Auger and internal conversion electrons
5. The radiations accompanying spontaneous fissions.

EDISTR performs the calculations and develops a decay file with the results of the calculations. This decay file is used in developing the three database files for DCAL.

The Decay Data (DECDAT) directory contains a series of MS-DOS executables, developed at Oak Ridge National Laboratory by Dr. Keith Eckerman, which are designed to take the output EDISTR file and suitably format it so that it can be incorporated into

the DCAL software package. The executables include BUILDEM.exe, CHKDAUS.exe, SORTINDEX.exe, and TOINDEX.exe. The BUILDEM executable takes an input file, the directory of the EDISTR output files, and creates the Radiation File (.RAD), the Beta Spectrum file (.BET), and the Index file (.NDX) database precursor, the IDX file, by extracting and sorting the data into the appropriate database files. The CHKDAUS executable file is then run for quality assurance. The CHKDAUS executable checks the database files created to ensure that all of the daughters of a decay chain are included. If there are missing daughters, a flag is put up to alert the user. After passing the CHKDAUS stage, SORTINDEX executable is run to organize the radionuclides alphabetically in the database files. TOINDEX converts the IDX file to an NDX file. Figure 1illustrates the flow of the software and files produced.

Quality Assurance Program

Proper identification of relevant decay chain members and the multiple codes used during the computational process all have the potential of introducing error into the final outcome.

The DCC working group's first quality assurance priority after extensive training efforts was to verify that each participant could successfully follow the procedure necessary for the generation of dose coefficients. To accomplish this each participant generated three database files for a collection of six radionuclides and their progeny. The six radionuclides were ^{144}Nd, ^{201}Au, ^{50}V, ^{61}Co, ^{41}Ar, and ^{38}S. The files generated by each independent participant were then compared so that inconsistencies could be identified.

These dose coefficients were also compared to other published dose coefficients such as ICRP 68 and the Japanese Atomic Energy Research Institute (JAERI)

Results

Few discrepancies were identified among the data files generated among members of the working group. All extractions from the ENSDF files matched for all radionuclides. The discrepancies identified did not exist due to software manipulations. The discrepancies identified were generated by either inclusion or exclusion of a given decay mode or progeny by those individuals generating files. Table 2 shows that all groups matched for the beta spectrum file for ^{201}Au but not for ^{50}V, because the beta decay mode for ^{50}V was excluded by group 3. Table 3 shows that the .RAD file for ^{61}Co was exact, but the ^{38m}Cl progeny of ^{38}S was not included by group 2 thus leading to inconsistency. These discrepancies demonstrated that a clear protocol was necessary for decisions regarding the inclusion or exclusion of bits of information. Currently the protocol is as follows:

1. Include all metastable states.
2. Level scheme data must be reviewed to determine which metastable states should be included in the decay chain of interest.
3. Decay modes that have a defined branching ratio less than 10^{-6}% can be excluded.

4. If one progeny ENSDF data is missing include the progeny that is further down the chain.

Employing these protocols the working group was able to successfully regenerate essentially identical values for the set of six test radionuclides.

The dose coefficients reported in Table 4 that were generated by members of the working group for the six test radionuclides were calculated after the QA issues had been addressed. After revisions to the group's procedure and development of inclusion protocols, the values calculated by members of the working group compared favorably. The DC working group was able to develop the same database files as used by ICRP and JAERI. Table 4 demonstrates only minor differences among our values and other published values for dose coefficients. Differences between the calculated and published dose coefficients exist due to input parameter selection in the DCAL software package and not from the database files generated. The effective dose for the QA radionuclides and the previously published doses coefficients are provided in Table 4.

Discussion

An initial evaluation of the DC working group's procedure for calculating dose coefficients values demonstrated few discrepancies. However, the discrepancies identified demonstrated that a stronger protocol for inclusion or exclusion of data was necessary.

The dose coefficients for the QA radionuclides were calculated after refining the procedure. These calculated dose coefficients were compared to published dose coefficients from such sources as ICRP 68 and JAERI. There exist insignificant differences between our calculated values and the published dose coefficients. The differences may exist due to more recent nuclear structure data and/or different biokinetic data and models. Clearly, the application of this refined protocol enhanced the reproducibility of independently calculated values. The DC working group is confident in the procedure and quality assurance program implemented for calculating dose coefficients using the DCAL and associated software.

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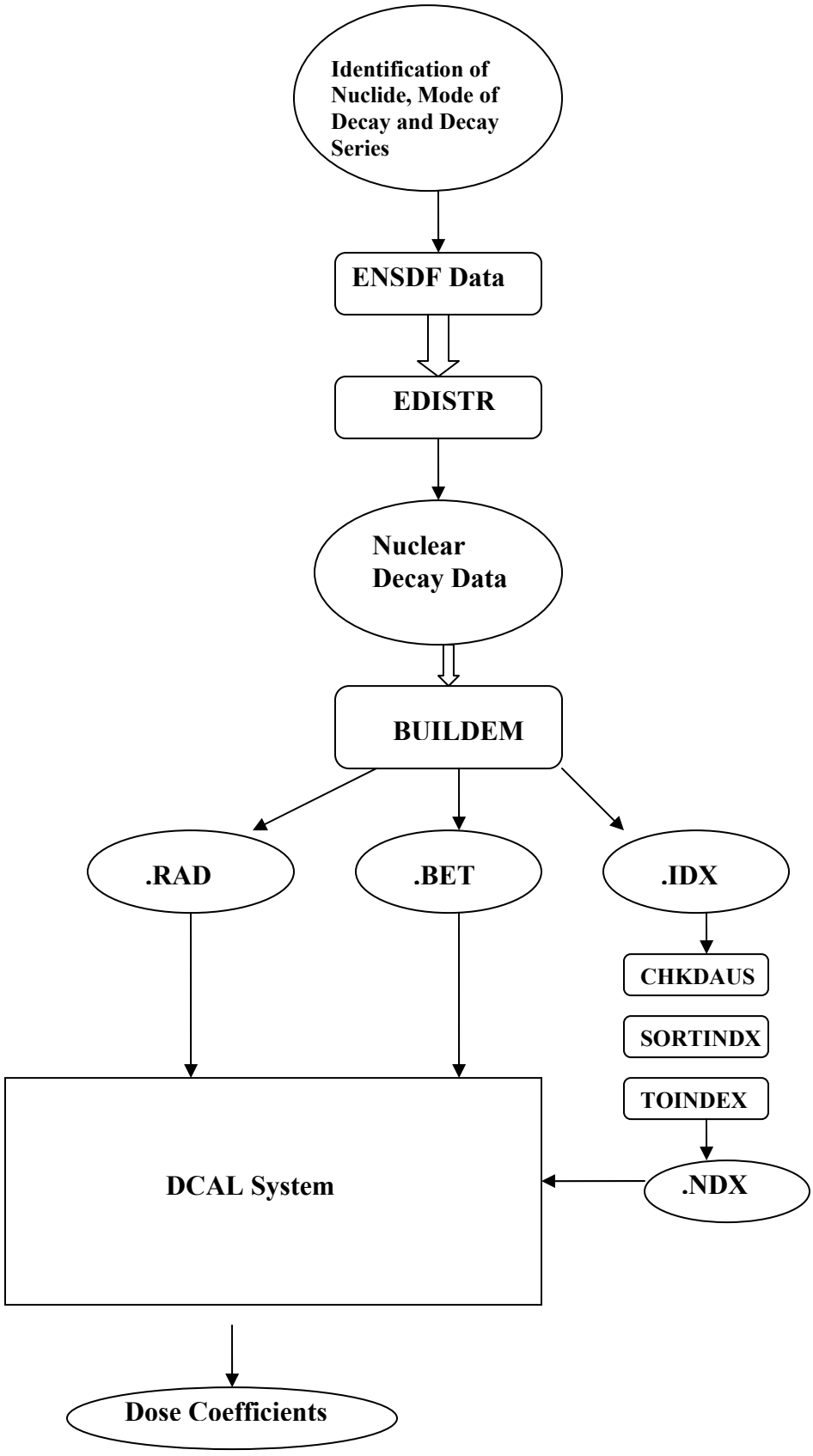
Identification of Nuclide, Mode of Decay and Decay Series
ENSDF Data
EDISTR
Nuclear Decay Data
BUILDEM
.RAD
.BET
.IDX
CHKDAUS
SORTINDX
TOINDEX
.NDX
DCAL System
Dose Coefficients

Figure1: Illustration of the flow of data and data files to produce dose coefficients

Table 1: List of unpublished radionuclides produced from a spallation neutron source upon a Hg target.

Z	A	Nuclide	T 1/2	Units	Z	A	Nuclide	T 1/2	Units
55	120	Cs-120	64	s	75	176	Re-176	5.3	m
81	188	Tl-188	71	s	71	168	Lu-168	5.5	m
82	190	Pb-190	71	s	54	119	Xe-119	5.8	m
67	150	Ho-150	72	s	77	197	Ir-197	5.8	m
82	191	Pb-191	1.33	m	75	175	Re-175	5.89	m
71	162	Lu-162	1.37	m	55	123	Cs-123	5.94	m
80	186	Hg-186	1.38	m	26	61	Fe-61	5.98	m
69	156	Tm-156	83.8	s	59	133	Pr-133	6.5	m
73	167	Ta-167	1.4	m	74	172	W-172	6.6	m
76	175	Os-175	1.4	m	51	113	Sb-113	6.67	m
70	158	Yb-158	1.49	m	72	166	Hf-166	6.77	m
70	159	Yb-159	1.58	m	79	185	Au-185m	6.8	m
59	132	Pr-132	1.6	m	74	173	W-173	7.5	m
73	168	Ta-168	2	m	80	189	Hg-189	7.6	m
82	193	Pb-193	2	m	69	160	Tm-160	9.4	m
81	189	Tl-189	2.3	m	81	192	Tl-192	9.6	m
74	171	W-171	2.38	m	82	195	Pb-195	15	m
61	137	Pm-137	2.4	m	68	157	Er-157	18.65	m
75	174	Re-174	2.4	m	81	193	Tl-193	21.6	m
74	170	W-170	2.42	m	73	171	Ta-171	23.3	m
75	180	Re-180	2.44	m	72	168	Hf-168	25.95	m
78	201	Pt-201	2.5	m	79	189	Au-189	28.7	m
62	139	Sm-139	2.57	m	74	174	W-174	31	m
56	123	Ba-123	2.7	m	69	161	Tm-161	33	m
78	182	Pt-182	3	m	78	185	Pt-185m	33	m
62	138	Sm-138	3.1	m	74	175	W-175	35.2	m
66	148	Dy-148	3.1	m	54	120	Xe-120	40	m
56	125	Ba-125	3.5	m	54	121	Xe-121	40.1	m
82	192	Pb-192	3.5	m	80	191	Hg-191	49	m
76	176	Os-176	3.6	m	78	185	Pt-185	70.9	m
69	157	Tm-157	3.63	m	18	41	Ar-41	109.34	m
68	154	Er-154	3.7	m	54	123	Xe-123	2.08	h
69	158	Tm-158	3.98	m	68	158	Er-158	2.29	h
70	161	Yb-161	4.2	m	72	171	Hf-171	12.1	h
79	185	Au-185	4.25	m	54	125	Xe-125	16.9	h
64	144	Gd-144	4.5	m	54	122	Xe-122	20.1	h
70	160	Yb-160	4.8	m	68	160	Er-160	28.58	h
73	169	Ta-169	4.9	m	18	37	Ar-37	35.04	d
57	128	La-128	5	m	54	127	Xe-127	36.4	d
76	178	Os-178	5	m	18	39	Ar-39	269	y
57	127	La-127	5.1	m	67	163	Ho-163	4570	y
61	153	Pm-153	5.25	m					

Table 2: Example of comparisons of .BET files where P(E) is the yield of the beta particle as a function of energy and the comparison are yes for exact matches and percent difference for inconsistencies among the results reported by members of the working group.

	Group 1	Group 2	Group 3			
Energy	P(E)	P(E)	P(E)	1 vs. 2	2 vs. 3	3 vs. 1
Au-201						
0.3	1.30	1.30	1.30	yes	yes	yes
0.32	1.29	1.29	1.29	yes	yes	yes
0.36	1.27	1.27	1.27	yes	yes	yes
0.4	1.23	1.23	1.23	yes	yes	yes
0.45	1.18	1.18	1.18	yes	yes	yes
0.5	1.12	1.12	1.12	yes	yes	yes
V-50						
0.01	1.40	1.40	0.00	yes	100%	100%
0.011	1.40	1.40	0.00	yes	100%	100%
0.012	1.39	1.39	0.00	yes	100%	100%
0.013	1.38	1.38	0.00	yes	100%	100%
0.014	1.38	1.38	0.00	yes	100%	100%
0.015	1.37	1.37	0.00	yes	100%	100%

Table 3: Examples of comparisons of .RAD files where Y(E) is the yield of the radiation at the given energy and the comparison are yes for exact matches and percent difference for inconsistencies among the results reported by members of the working group.

	Group 1		Group 2		Group 3		1 vs. 2		2 vs. 3		1 vs. 3	
Emitted radiation	Yield (%)	E (MeV)	Yield (%)	E (MeV)	Yield (%)	E (MeV)	Yield (%)	E (MeV)	Yield (%)	E (MeV)	Yield (%)	E (MeV)
Co-61												
x-ray	2.00×10^{-03}	7.43×10^{-04}	2.00×10^{-03}	7.43×10^{-04}	2.00×10^{-03}	7.43×10^{-04}	yes	yes	yes	yes	yes	yes
x-ray	8.12×10^{-04}	7.60×10^{-04}	8.12×10^{-04}	7.60×10^{-04}	8.12×10^{-04}	7.60×10^{-04}	yes	yes	yes	yes	yes	yes
gamma ray	2.01×10^{-06}	7.32×10^{-03}	2.01×10^{-06}	7.32×10^{-03}	2.01×10^{-06}	7.32×10^{-03}	yes	yes	yes	yes	yes	yes
gamma ray	1.28×10^{00}	7.46×10^{-03}	1.28×10^{00}	7.46×10^{-03}	1.28×10^{00}	7.46×10^{-03}	yes	yes	yes	yes	yes	yes
Beta	4.40×10^{00}	1.29×10^{-01}	4.40×10^{00}	1.29×10^{-01}	4.40×10^{00}	1.29×10^{-1}	yes	yes	yes	yes	yes	yes
Beta	9.56×10^{01}	4.74×10^{-01}	9.56×10^{01}	4.74×10^{-01}	9.56×10^{01}	4.74×10^{-1}	yes	yes	yes	yes	yes	yes
Cl-38m												
x-ray	1.91×10^{-05}	2.00×10^{-04}	0.00×10^{00}	0.00×10^{00}	1.91×10^{-05}	2.00×10^{-04}	100%	100%	100%	100%	yes	yes
x-ray	1.27×10^{-06}	2.63×10^{-04}	0.00×10^{00}	0.00×10^{00}	1.27×10^{-06}	2.63×10^{-04}	100%	100%	100%	100%	yes	yes
gamma ray	1.09×10^{-10}	2.55×10^{-03}	0.00×10^{00}	0.00×10^{00}	1.09×10^{-10}	2.55×10^{-03}	100%	100%	100%	100%	yes	yes
gamma ray	1.62×10^{-03}	2.62×10^{-03}	0.00×10^{00}	0.00×10^{00}	1.62×10^{-03}	2.62×10^{-03}	100%	100%	100%	100%	yes	yes
Internal conversion	2.51×10^{-03}	2.25×10^{-03}	0.00×10^{00}	0.00×10^{00}	2.51×10^{-03}	2.25×10^{-03}	100%	100%	100%	100%	yes	yes
Internal conversion	9.55×10^{-03}	2.31×10^{-03}	0.00×10^{00}	0.00×10^{00}	9.55×10^{-03}	2.31×10^{-03}	100%	100%	100%	100%	yes	yes

Table 4: Dose coefficients developed by members of the working group for inhalation and ingestion in terms committed effective dose (Sv Bq^{-1}), and air submersion in terms of effective dose per unit air concentration and time (Sv Bq^{-1} s^{-1}) compared to those published by JAERI and ICRP

		1AMAD		5AMAD		Ingestion		External	
Radionuclide	Condition	Effective dose	JAERI/ICRP	Effective dose	JAERI/ICRP	Effective dose	JAERI/ICRP	H sub E	FGR-13
Nd-144[a]						4.08×10^{-08}	4.1×10^{-08}		
Nd-144[a]	M	7.37×10^{-06}	7.4×10^{-06}	5.01×10^{-06}	5.0×10^{-06}				
Nd-144[a]	S	3.12×10^{-06}	3.2×10^{-06}	1.61×10^{-06}	1.6×10^{-06}				
V-50[a]						3.41×10^{-09}	4.2×10^{-09}		
V-50[a]	F	6.47×10^{-08}	8.4×10^{-08}	7.57×10^{-08}	9.9×10^{-08}				
V-50[a]	M	2.74×10^{-08}	3.5×10^{-08}	1.93×10^{-08}	2.5×10^{-08}				
S-38[a]	S_Org vapor (SO_2)	1.43×10^{-10}	2.0×10^{-10}						
S-38[a]	S_Org vapor (CS_2)	1.69×10^{-10}	2.0×10^{-10}						
S-38[a]	S_Org					2.66×10^{-10}	2.7×10^{-10}		
S-38[a]	S_Inorg F	1.44×10^{-10}	1.4×10^{-10}	2.44×10^{-10}	2.4×10^{-10}				
S-38[a]	S_Inorg M	2.51×10^{-10}	2.5×10^{-10}	3.64×10^{-10}	3.6×10^{-10}				
S-38[a]	S Elemental					6.09×10^{-10}	6.4×10^{-10}		
S-38[a]	S_Inorg					4.29×10^{-10}	4.3×10^{-10}		
Co-61[b]	Unspecified					7.51×10^{-11}	7.4×10^{-11}		
Co-61[b]	M	4.86×10^{-11}	4.8×10^{-11}	7.14×10^{-11}	7.1×10^{-11}				
Co-61[b]	S	5.18×10^{-11}	5.1×10^{-11}	7.55×10^{-11}	7.5×10^{-11}				
Au-201[b]						2.44×10^{-11}	2.4×10^{-11}		
Au-201[b]	F	9.31×10^{-12}	9.2×10^{-12}	1.58×10^{-11}	1.6×10^{-11}				
Au-201[b]	M	1.72×10^{-11}	1.7×10^{-11}	2.75×10^{-11}	2.8×10^{-11}				
Au-201[b]	S	1.81×10^{-11}	1.8×10^{-11}	2.88×10^{-11}	2.9×10^{-11}				
Au-201								4.08×10^{-15}	2.62×10^{-15}
Co-61								9.77×10^{-15}	3.75×10^{-15}
Ar-41								1.55×10^{-13}	6.15×10^{-14}
S-38								2.13×10^{-13}	Not Reported
Cl-38								1.82×10^{-13}	7.58×10^{-14}
V-50								1.74×10^{-13}	Not Reported
Nd-144								0.00×10^{00}	Not Reported