

SANDIA REPORT

SAND2021-15855

Printed 12/15/2021



Sandia
National
Laboratories

Environmental Protection Agency Drinking Water Protective Action Guides Implementation Recommendations

Kevin G. Hart
Lainy D. Cochran

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico
87185 and Livermore,
California 94550

Issued by Sandia National Laboratories, operated for the United States Department of Energy by National Technology & Engineering Solutions of Sandia, LLC.

NOTICE: This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government, nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, make any warranty, express or implied, or assume any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represent that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government, any agency thereof, or any of their contractors or subcontractors. The views and opinions expressed herein do not necessarily state or reflect those of the United States Government, any agency thereof, or any of their contractors.

Printed in the United States of America. This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831

Telephone: (865) 576-8401
Facsimile: (865) 576-5728
E-Mail: reports@osti.gov
Online ordering: <http://www.osti.gov/scitech>

Available to the public from

U.S. Department of Commerce
National Technical Information Service
5301 Shawnee Rd
Alexandria, VA 22312

Telephone: (800) 553-6847
Facsimile: (703) 605-6900
E-Mail: orders@ntis.gov
Online order: <https://classic.ntis.gov/help/order-methods/>



ABSTRACT

This report documents the analysis conducted by Sandia National Laboratories on the effect of various alterations to published methodologies to calculate Derived Response Levels for Environmental Protection Agency Drinking Water Protective Action Guides. Specifically, this study sought to assess and provide recommendations on calculation of the Derived Response Level accounting for decay during the consumption period, assess the impact of decay on laboratory Minimal Detectable Concentration in water samples as compared to the Derived Response Level, make a recommendation on the calculation of Derived Response Level consistent with existing Public Protection Methods, and make a recommendation on the use of six age groups versus eight age groups based on available dose coefficients for calculation of the Derived Response Level. The authors analyzed these various factors using nominal radionuclide mixes from four scenarios and compared calculation of the Derived Response Level accounting for decay and no decay and then compared those results to the laboratory Minimal Detectable Concentrations. The authors concluded that decay should be included in the calculation of the Derived Response Level, existing Public Protection Methods should be employed to calculate the Derived Response Level, six age groups should be used versus eight, and the use of both decay and Public Protection Methods result in little to no concern for water samples meeting the Minimum Detectable Concentration requirements. The results of this study may be used in further developing and implementing a method for the Environmental Protection Agency Water Derived Response Level calculation in the Federal Radiological Monitoring and Assessment Center Assessment Manual.

ACKNOWLEDGEMENTS

Sean Fournier

Brian Hunt

Tom Laiche

Autumn Kalinowski

Terry Kraus

Robert Litman

Robert Shannon

CONTENTS

1.1. Background	8
1.2. Purpose.....	8
2. Study Methods	9
2.1. Scenarios Evaluated	9
2.1.1. Nuclear Power Plant.....	9
2.1.2. Plutonium Improvised Nuclear Device.....	10
2.1.3. Plutonium Radioisotope Thermoelectric Generator.....	11
2.1.4. Radiological Dispersal Device.....	12
2.2. Dose Coefficients	13
2.3. Derived Response Level Calculation	14
2.4. Ingestion Dose Parameter Calculation	15
2.4.1. Ingestion Dose Parameter – No Decay.....	15
2.4.2. Ingestion Dose Parameter – Decay.....	16
3. Analysis	17
3.1. Sum of Fractions Methods	17
3.2. DRL Calculations Considering Decay versus No Decay	17
3.3. DRL versus MDC Comparison.....	18
4. Conclusions and Recommendations	20
4.1. Six Age Groups versus Eight Age Groups.....	20
4.2. Sum of Fractions Methods.....	20
4.3. DRL Calculations Considering Decay versus No Decay	20
4.4. DRL versus MDC Comparison.....	20
Appendix A. Scenario DRL and MDC Comparison Tables.....	22

LIST OF TABLES

Table 1. Nuclear Power Plant Radionuclides and Water Concentration	9
Table 2. Plutonium Improvised Nuclear Device Radionuclides and Water Concentration	10
Table 3. Plutonium Radioisotope Thermoelectric Generator Radionuclides and Water Concentration	12
Table 4. Radiological Dispersal Device Radionuclides and Water Concentration	13
Table 5. Number of Radionuclides without a Fetus or Breastfed Infant DC.....	13
Table 6. Intake Rates (EPA PAG Manual)	15
Table 7. DRL Increase Factor	18
Table 8. Summary of DRL versus MDC Comparison	19

This page left blank

ACRONYMS AND DEFINITIONS

Abbreviation	Definition
AWG	Assessment Working Group
DC	dose coefficient
DCFPAK	Dose Coefficient File Package
DRL	Derived Response Level
EPA	Environmental Protection Agency
FRMAC	Federal Radiological Monitoring and Assessment Center
ICRP	International Commission on Radiological Protection
IND	improvised nuclear device
IngDP	Ingestion Dose Parameter
MDC	Minimal Detectable Concentration
NAREL	National Analytical Radiation Environmental Laboratory
PAG	Protection Action Guide
NPP	nuclear power plant
NRC	Nuclear Regulatory Commission
NUREG	Nuclear Regulatory Commission Regulatory Guide
RDD	radiological dispersal device
RTG	radioisotope thermoelectric generator
SNL	Sandia National Laboratories
SDWA	Safe Drinking Water Act
WGpu	weapons grade plutonium

INTRODUCTION

In 2017, the Environmental Protection Agency (EPA) published, *PAG Manual: Protective Action Guides and Planning Guidance for Radiological Incidents* which included new Protective Action Guides (PAGs) for Drinking Water [5]. This report documents the analysis conducted by Sandia National Laboratories (SNL) on the effect of various alterations to published methodologies to calculate Derived Response Levels (DRLs) for EPA Drinking Water PAGs.

1.1. Background

Water DRLs represent the concentration of radioactive material in drinking water that, in the absence of any intervention, could lead to an individual receiving a dose equal to the appropriate PAG if consumed over one year. Protective actions should be considered when radionuclide activity concentrations in water reach the DRL. In 2017, the Assessment Working Group (AWG) of the Federal Radiological Monitoring and Assessment Center (FRMAC) noted an issue with the method specified to calculate the Water DRL in the PAG Manual [5]. The issue noted was that the DRL values were so low that a radiochemistry laboratory would have difficulty meeting the minimum detectable concentration (MDC) limits necessary to “see” the DRL values. The AWG commissioned the report, *Use of Drinking Water Methods for Analysis of Radionuclides in Water Following a Radiological Event* [7] (referred to later in this report as “the Griggs report”) to assess whether MDC limits could be met using several case studies and their associated DRLs calculated using the method described in the PAG Manual [5]. This report concluded that EPA approved drinking water methods for compliance with the Safe Drinking Water Act (SDWA) could detect many but not all radionuclides at the required MDC [7]. The report also cautioned that the DRL values used were scenario-specific and the results would not apply universally to other scenarios.

1.2. Purpose

The purpose of this report is to make recommendations on how to implement an improved method for calculating DRLs for the EPA Drinking Water PAGs in Turbo FRMAC[®] [8].

Specifically, the analysis in this report sought to assess the impact of considering decay during the consumption period on DRL values, use of the Public Protection Method DRL calculation methodology presented in the FRMAC Assessment Manual Vol. 1, Method 1.1 [6], and assess the fraction of DRL values for each scenario that meet both SDWA approved methods as well as common radiochemical analytical method MDCs for the similar radionuclide mixes assessed in *Use of Drinking Water Methods for Analysis of Radionuclides in Water Following a Radiological Event* [7].

2. STUDY METHODS

2.1. Scenarios Evaluated

Several accident scenarios were considered in the analysis. The radionuclide mixture for each scenario is based on a default mixture to be used by FRMAC Assessment until event-specific information is known. These default mixtures are not yet published but form the basis of a revision to FRMAC Assessment Manual, Vol. 2 – Pre-Assessed Scenarios, currently in draft. The water concentration values provided in the tables in this section are generally inferred based on ground deposition and were not actually modeled using a water model. The concentration values were based on the default mixture at the end of release.

2.1.1. Nuclear Power Plant

The potential radionuclides for the nuclear power plant (NPP) accident in Table 1 were obtained from a scenario described in Nuclear Regulatory Commission (NRC) Regulatory Guide (NUREG)/CR-7110 Vol. 1 Rev 1 [12] and further down selected to the top dose contributors for offsite consequence. This scenario is conservative and generally representative of other severe accident scenarios; it may not reflect other NPP accidents that are likely to occur. Because the selected scenario is for a boiling water reactor, this analysis does not address other radionuclides that could be preferentially released from other reactor types and potentially classified as “hard-to-detect” that could also challenge the capabilities of EPA-approved drinking water methods. It is also noted that the presence of additional radionuclides contributing dose would likely result in lower DRLs and more restrictive detection capability requirements than were evaluated [7]. For the analysis, equilibrium was not assumed due to inclusion of progeny as parents in the original core inventories.

Table 1. Nuclear Power Plant Radionuclides and Water Concentration

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)
Ba-140	9.31E-04	Np-239	5.58E-04
Ce-141	6.40E-05	Pu-238	7.56E-08
Ce-144	4.60E-05	Pu-239	1.51E-08
Cs-134	9.89E-06	Pu-240	1.22E-08
Cs-136	3.72E-06	Pu-241	4.25E-06
Cs-137	9.89E-06	Sr-89	5.64E-04

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)
I-129	2.34E-15	Sr-90	4.42E-05
I-131	6.87E-04	Sr-91	3.96E-04
I-132	8.94E-04	Te-127m	6.98E-06
I-133	1.17E-03	Te-129m	2.50E-05
I-134	3.09E-06	Te-131m	8.14E-05
I-135	6.26E-04	Te-132	7.56E-04
La-140	1.22E-04	Zr-95	6.98E-05
Nb-95	1.92E-05	Zr-97	4.83E-05

2.1.2. Plutonium Improvised Nuclear Device

The plutonium improvised nuclear device (IND) scenario mixture shown in Table 2 includes the top dose producing radionuclides at a distance of 10 km downwind at 1 hour post-denotation assuming no fractionation and a weapons grade plutonium (WGPu) implosion assembled weapon with stainless steel cladding [11]. Detonation of different device designs in different detonation and weather conditions will result in different fallout mixtures. Similar considerations about “hard-to-detect” radionuclides and impacts on analyte lists, DRLs and detection capability requirements apply here. For the analysis, equilibrium was not assumed due to inclusion of progeny as parents in the mixture.

Table 2. Plutonium Improvised Nuclear Device Radionuclides and Water Concentration

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)
Ba-140	4.08E-04	Mn-54	9.10E-07	Tc-99m	2.00E-04
Ba-141	4.02E-02	Mn-56	6.23E-02	Tc-101	1.05E-01
Ba-142	1.18E-02	Mo-99	2.11E-03	Te-131	5.47E-02

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)
Ce-143	2.88E-03	Mo-101	3.72E-02	Te-131m	5.66E-04
Ce-144	1.27E-05	Pu-239	1.11E-07	Te-132	1.54E-03
Co-58	5.23E-06	Rb-89	1.26E-02	Te-133m	3.76E-02
Co-58m	8.43E-04	Ru-103	1.70E-04	Te-134	5.95E-02
Cs-134m	4.66E-05	Ru-105	2.51E-02	Y-93	8.35E-03
Cs-137	5.88E-07	Ru-106	1.17E-05	Y-94	3.38E-02
Cs-138	9.65E-02	Sb-128	1.12E-03	Y-95	1.07E-02
I-131	2.12E-04	Sb-129	6.81E-03	Zr-95	6.79E-05
I-132	1.47E-03	Sb-130	1.29E-02	Zr-97	7.10E-03
I-133	5.84E-03	Sb-131	2.90E-02		
I-134	8.19E-02	Sn-128	9.42E-03		
I-135	1.95E-02	Sr-90	1.93E-07		
La-141	2.48E-02	Sr-91	5.80E-03		
La-142	5.08E-02	Sr-92	2.02E-02		

2.1.3. Plutonium Radioisotope Thermoelectric Generator

The radioisotope thermoelectric generator (RTG) mixture in Table 3 is based on the inventory for the Mars Science Laboratory RTG launch in 2011 [1]. The SDWA does not regulate any specific alpha-emitting transuranic (TRU) radionuclides and, as a result, there are no methods approved by the EPA for their determination [7]. SDWA-approved methods, however, do include techniques for screening samples for gross alpha and gross beta radioactivity which could be used to screen samples containing simple source terms such as those in the RTG and radiological dispersal device (RDD) scenarios where the source terms consist of single alpha or beta-emitting radionuclides. For the analysis, equilibrium was not assumed due to the inventory including plutonium progeny such as Am-241, U-234, and U-237 as parents.

Table 3. Plutonium Radioisotope Thermoelectric Generator Radionuclides and Water Concentration

Radionuclide	Concentration ($\mu\text{Ci/l}$)
Am-241	1.89E-08
Pu-238	9.73E-05
Pu-239	5.23E-08
Pu-240	3.85E-08
Pu-241	7.92E-07
Pu-242	1.64E-11
U-234	1.57E-09
U-237	1.90E-11

2.1.4. Radiological Dispersal Device

The radionuclides listed under the RDD scenario in Table 4 are considered potential radionuclides of concern for use in an RDD and were collated from two sources: a joint Department of Energy and NRC study [2] on the most likely sources available for potential terrorist use and a study [13] by SNL on source prioritization for use in an RDD of national security significance. Although the event is assumed to be a single-radionuclide-event, multiple nuclide events could be encountered. If more than one radionuclide is associated with a source term, the list of analytes, and DRLs and detection capability requirements would vary significantly from the scenarios evaluated and the required detection capability for each radionuclide would be determined considering the dose from all of the radionuclides in the mixture. As such, detailed evaluation of potential multi-nuclide RDD scenarios was considered as beyond the scope of this analysis. For this study each radionuclide DRL was calculated assuming the release contained a single radionuclide not in a mixture. The analysis for each RDD radionuclide assumed progeny present in equilibrium, if applicable (e.g., Y-90 present in equilibrium with Sr-90).

Table 4. Radiological Dispersal Device Radionuclides and Water Concentration

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Radionuclide	Concentration ($\mu\text{Ci/l}$)
Am-241	1.00E+00	Pu-239	1.00E+00
Cf-252	1.00E+00	Ra-226	1.00E+00
Cm-244	1.00E+00	Se-75	1.00E+00
Co-60	1.00E+00	Sr-90	1.00E+00
Cs-137	1.00E+00	Tm-170	1.00E+00
Ir-192	1.00E+00	U-235	1.00E+00
Po-210	1.00E+00	Yb-169	1.00E+00
Pu-238	1.00E+00		

2.2. Dose Coefficients

Dose Coefficients (DCs) from Dose Coefficient File Package (DCFPAK) were used to calculate Ingestion Dose Parameters (IngDPs) and subsequent calculation of DRLs for the six age groups: Infant, 1 year old, 5 year old, 10 year old, 15 year old, and Adult [3] [4]. EPA's Water DRL methodology considers eight age groups, adding the fetus and breastfed infant to the standard six [5]. However, ingestion DCs for the fetus and breastfed infant are only available for a limited set of radionuclides in International Commission on Radiological Protection (ICRP) Publication 88 [9] and ICRP Publication 95 [10], respectively. Therefore, the DRLs for eight age groups were not calculated in this analysis.

Implementation of the additional two age groups (fetus and breastfed infant) is not possible with the available ingestion DC information. Table 5 shows the number of radionuclides in the scenarios considered in this report that do not have DCs for both the fetus and breastfed infant.

Table 5. Number of Radionuclides without a Fetus or Breastfed Infant DC

Scenario	Radionuclides Without DC
NPP	3 of 27
Pu IND	28 of 46

Scenario	Radionuclides Without DC
RTG	2 of 8
RDD	4 of 15

2.3. Derived Response Level Calculation

The PAG Manual specifies an individual radionuclide DRL calculation method (Eq. 1).

$$DRL_{water,age,i} = \frac{PAG_{water,age}}{DWIR_{age} * t_c * IngDC_{E,age,i}} \quad (\text{Eq. 1})$$

where:

- $DRL_{water,age,i}$ = Water Derived Response Level, the concentration of radionuclide i in drinking water at which the ingestion dose to the most sensitive population (age group) from all radionuclides in a release has the potential to equal the applicable ingestion PAG, $\mu\text{Ci/l.}$;
- $PAG_{water,age}$ = Age Specific Water PAG (100 or 500, dependent on Age Group), mrem;
- $DWIR_{age}$ = Daily Water Intake Rate for a specific age group. (See Table 6), $1/\text{d.}$
- t_c = Consumption Time, the length of the consumption period (default 365 days), d.
- $IngDC_{E,age,i}$ = Ingestion Dose Coefficient, the ingestion pathway dose coefficient for the whole body (E) for a specific age group for radionuclide i, $\text{mrem}/\mu\text{Ci.}$

FRMAC calculates mixture based DRLs that account for dose contributions from multiple radionuclides representing the hazard of the entire mixture. A description of this approach is included in FRMAC Assessment Manual Vol. 1, Method 1.1. Applying this same approach to calculate the mixture Water DRL yields the following equation:

$$DRL_{water,age,i} = \frac{c_{water,i} * PAG_{water,age,i}}{DWIR_{age} * FWC * \sum_i (IngDP_{E,age,i})} \quad (\text{Eq. 2})$$

where:

- $DRL_{water,age,i}$ = Water Derived Response Level, the concentration of radionuclide i in drinking water at which the ingestion dose to the most sensitive population (age group) from all radionuclides in a release has the potential to equal the applicable ingestion PAG, $\mu\text{Ci/l.}$;

$C_{\text{water},i}$	= Water Contamination, the level of contamination of radionuclide i in a drinking water, $\mu\text{Ci/l}$; At beginning of the consumption period;
$\text{PAG}_{\text{water},\text{age}}$	= Age Specific Water PAG (100 or 500, dependent on Age Group), mrem ;
DWIR_{age}	= Daily Water Intake Rate for a specific age group. (See Table 6), l/d ;
FWC	= Fraction of Water Contaminated, unitless; (Default of 1)
$\text{IngDP}_{E,\text{age},i}$	= Ingestion Dose Parameter, the committed effective dose received from ingestion of radionuclide i in water by a specific age group, $\text{mrem}\cdot\text{d/l}$.

Table 6. Intake Rates (EPA PAG Manual)

Age	Water Intake Rates (l/day)
0	0.191
1	0.223
5	0.542
10	0.725
15	0.9
Adult	1.643

2.4. Ingestion Dose Parameter Calculation

The PAG Manual assumes no decay during the consumption period. This is a conservative assumption based on the unlikely scenario where radionuclides are continuously replenished [5]. This study includes cases with and without decay during the consumption period. This is handled in the IngDP term, described in the following sections.

The PAG Manual assumes that there is no decay during the consumption period as a conservative assumption [5]. The current EPA methodology has a great deal of conservatism through assuming 100% of the water consumed during the consumption period is contaminated, no measures are implemented to mitigate the magnitude of contamination during the consumption period, and the sampling point is representative of the water intake point. Decay is a physical process that would be applicable in the case of a single short duration release.

2.4.1. Ingestion Dose Parameter – No Decay

Equation 3 shows the calculation of IngDP. For the no decay case, there is no integrated decay term in the equation. All radionuclides were assumed to remain at their initial concentration for the duration of the ingestion period. It should be noted that Equation 2 simplifies to Equation 1 for the case of a single radionuclide. The decay term in Equation 3 can be used in the denominator of Equation 1 to account for the decay occurring between the end of the release also referred to a time of deposition and the beginning of the consumption period or the sampling time. For this study t_s

was assumed to be zero as the concentration values used in the calculation were assumed to have already been decay corrected.

$$IngDP_{E,age,i} = C_{water,i} * IngDC_{E,age,i} * e^{-\lambda_i t_s} * t_c \quad (\text{Eq. 3})$$

where,

IngDP _{E,age,i}	= Ingestion Dose Parameter, the committed effective dose received from ingestion of radionuclide i in water by a specific age group, mrem·d/l;
C _{water,i}	= Water Contamination, the level of contamination of radionuclide i in a drinking water, $\mu\text{Ci/l}$; At beginning of the consumption period;;
IngDC _{E,age,i}	= Ingestion Dose Coefficient, the ingestion pathway dose coefficient for the whole body (E) for a specific age group for radionuclide i, mrem/ μCi ;
λ_i	= Decay constant for radionuclide i, d^{-1} ;
t_s	= Time relative to end of the release when the water sample was collected. Also corresponds to the start of the consumption period, d;
t_c	= Consumption Time, the length of the consumption period (default 365 days), d.

2.4.2. Ingestion Dose Parameter – Decay

For cases that consider decay during the consumption period, the integrated decay term for each radionuclide appears at the end of the equation.

$$IngDP_{E,age,i} = C_{water,i} * IngDC_{E,age,i} * e^{-\lambda_i t_s} * \frac{1 - e^{-\lambda_i t_c}}{\lambda_i} \quad (\text{Eq. 4})$$

where,

IngDP _{E,age,i}	= Ingestion Dose Parameter, the committed effective dose received from ingestion of radionuclide i in water by a specific age group, mrem·d/l;
C _{water,i}	= Water Contamination, the level of contamination of radionuclide i in a drinking water, $\mu\text{Ci/l}$; At beginning of the consumption period;;
IngDC _{E,age,i}	= Ingestion Dose Coefficient, the ingestion pathway dose coefficient for the whole body (E) for a specific age group for radionuclide i, mrem/ μCi ;
λ_i	= Decay constant for radionuclide i, d^{-1} ;
t_s	= Time relative to end of the release when the water sample was collected. Also corresponds to the start of the consumption period, d;
t_c	= Consumption Time, the length of the consumption period (default 365 days), d.

3. ANALYSIS

3.1. Sum of Fractions Methods

The PAG Manual specifies the use of the sum of fractions method (Eq. 5) when determining whether total dose from the radionuclide mixture exceeds the PAG.

$$F = \sum_i^n \left(\frac{C_i}{DRL_i} \right) \quad (\text{Eq. 5})$$

where:

F = sum of fractions;

C_i = the measured concentration of radionuclide i in the water supply, $\mu\text{Ci/l}$;

DRL_i = derived response level for the i^{th} radionuclide from Equation 2, $\mu\text{Ci/l}$.

This methodology presupposes that concentrations of radionuclides for all radionuclides in the mix are known which is a challenging consideration that might be difficult to implement operationally during a response. Additionally, this method does not allow for an *a priori* determination of DRLs to be used to set laboratory Analytical Action Levels or Critical Levels since no initial concentration values are available to calculate individual DRLs. This methodology only lends itself to an *a posteriori* determination if the PAG has been exceeded for the mixture and age group.

The mixture-based DRL approach used in the FRMAC Assessment Manual, Vol 1. [6] provides the added benefit of not having a requirement to know the concentrations of all radionuclides in the mixture provided the mixture concentration ratio is known or assumed. The $DRL_{\text{water,age},i}$ (Eq. 1) represents the concentration at the sampling time of radionuclide i at which the total dose from *all radionuclides* in a release from the ingestion pathway included in the assessment would equal the PAG over the time phase under consideration. Once the relative amount of each radionuclide present is known or assumed, $DRL_{\text{water,age},i}$ can be calculated for any radionuclide in the mixture to represent the hazard of the entire mixture. The DRLs must be recalculated for water sources with differing ratios.

Initial concentration values for the mixture to calculate $DRL_{\text{water,age},i}$ (Eq. 1) can come from a knowledge of the released or deposited mixture. Initially the modelled or assumed released inventory mixture can be assumed to be deposited on the ground without regard the mixing that would occur in the water column assuming the mixture and its radionuclide ratios represent the same radionuclide ratios appearing in the water column. This approach was taken for the analysis documented in this report. Alternatively, the concentration values can be obtained by modelling the dispersion into water source of concern.

3.2. DRL Calculations Considering Decay versus No Decay

Table 7 summarizes percent increase of the $DRL_{\text{water},i}$ for the five scenarios and their mixtures in this study (Appendix A, Tables A-1 thru A-4). The large increases shown for the NPP and Pu IND scenarios demonstrate the considerable amount of decay that occurs for these fission product mixtures over the one-year consumption period.

Table 7. DRL Increase Factor

Scenario	DRL _{water,i} Percent Increase (%)
NPP ^a	1400
Pu IND ^a	28200
RTG ^a	4
RDD ^b	0-3600

^aMixture DRL calculation using most restrictive DRL from 6 age groups.

^bIndividual radionuclide DRL calculation using the most restrictive DRL from 6 age groups.

3.3. DRL versus MDC Comparison

The Griggs report outlined the variety of methods used to compare the scenario radionuclide mix DRL values to the MDC [7]. Table 8 summarizes the comparison of the mixture DRL values (Appendix A, Tables A-1 thru A-4) in this study with the MDC values from the Griggs report as well as from other sources. Table 8 notes how many radionuclides would be eliminated from the analysis due to their short half-life relative to a supposed sampling time after release. For this study radionuclides with a half-life of less than 9 hours were counted as not being available in the water sample and therefore there would be no need to evaluate whether they could be detected by laboratory analysis. The Pu IND has the largest number of short half-lived radionuclides (25) in the mix that would be neglected in laboratory analysis.

The Number of Radionuclides not Meeting MDC for the No Decay Case column of Table 8 identifies the number of radionuclides that would not meet MDC from the original mix excluding short half lived radionuclides. The RDD scenario has zero radionuclides not meeting MDC for the no decay case which would be expected due to the relative ease at which these radionuclides are detected. When decay is included in the DRL calculation the Number of Radionuclides not meeting MDC for the Decay Case column of Table 8 shows the number of radionuclides not meeting MDC after consideration of decay in the DRL calculation. Both the Pu IND and the RDD have no DRL values that would not meet the MDC. The last column Number of Radionuclides with no Identified Laboratory Method is the number of radionuclides which have no method identified to determine concentration. Both the NPP and the Pu RTG have a small number of radionuclide DRLs that would not meet MDC requirements.

Ultimately, Table 8 shows that when decay and removal of short-lived radionuclides are considered in calculation of the DRL, there are only a small number of radionuclides (if any) for which a laboratory would not be able to determine concentrations.

Table 8. Summary of DRL versus MDC Comparison

Scenario	Total Number of Radionuclides in Mix	Short Half Life	Number of Radionuclides not Meeting MDC ^{cd} for <u>No Decay Case</u>	Number of Radionuclides not Meeting MDC ^{cd} for <u>Decay Case</u>	Number of Radionuclides with no Identified Laboratory Method
NPP	27	3	4	1	2
Pu IND	46	25	5	0	0
RTG	8	0	4	4	1
RDD ^b	15	0	0	0	0

^a Individual radionuclide DRL calculation using the most restrictive DRL from 6 age groups.

^b John Griggs, *Use of Drinking Water Methods for Analysis of Radionuclides in Water Following a Radiological Event*, National Analytical Radiation Environmental Laboratory (NAREL), U.S. EPA, December 2019.

^c Included MDC values for other common methods not included in the Griggs report for gamma and alpha spec.

4. CONCLUSIONS AND RECOMMENDATIONS

4.1. Six Age Groups versus Eight Age Groups

Given that DCs are not available for many of the radionuclides (Table 5) included in the nominal mixtures for the scenarios in this study, it is recommended that implementation of the Water DRL be restricted to the standard six age groups (Infant, 1 year old, 5 year old, 10 year old, 15 year old, and Adult) until such time as DCs are available for all radionuclides of interest. Another option such as using the next conservative age group in place of the fetus or breastfed infant was rejected as the difference between the fetus and breastfed infant and the next most conservative age group can vary by an order of magnitude or more. The use of a single age group was again rejected as the difference among the DCs among the six age groups can vary by several orders of magnitude.

Further research is needed to develop DCs for the fetus and breastfed infant for a more comprehensive list of radionuclides.

4.2. Sum of Fractions Methods

As stated previously, the sum of fractions method specified in the PAG Manual [5] has multiple limitations. The authors recommend the implementation of the DRL calculation method in Equation 1 which allows for *a priori* analysis of AALs and restricting laboratory analysis to a smaller set of radionuclides in the mix to make a determination if the PAG could be exceeded.

4.3. DRL Calculations Considering Decay versus No Decay

The assumption of continuous discharge is not valid for the bulk of nuclear/radiological incident scenarios. While the PAG Manual acknowledges that this assumption is conservative, there are other assumptions in the Water DRL method [5] that make it sufficiently conservative even if decay is included. These include assuming contaminated water would be consumed for a year and no mitigation of radionuclides in water would occur. Decay is a natural process exhibited by radionuclides. Decay during the consumption period is well understood and accounted for in Equation 4. It is therefore recommended that implementation of the method account for decay both prior to sampling and during the consumption period.

4.4. DRL versus MDC Comparison

Only a small number of radionuclides in the mixtures considered in this report lack an identified laboratory method (Table 8) to achieve an MDC that would allow comparison to the DRL once short lived radionuclides are removed, SDWA methods are considered, and other standard laboratory methods are considered. With the implementation of Equation 1 for the calculation of the DRL over the sum of fractions methods specified in the PAG Manual [5], the number of radionuclides that would be analyzed in a laboratory is small once the radionuclide ratios in the mix are known. It is recommended that no additional effort is needed to identify laboratory methods to support comparison of samples to the DRL.

REFERENCES

- [1] Department of Energy “MSL MMRTG Fuel Inventory at Assay at Launch (November 25, 2011),” Communication to U.S. DOE, March 2010.
- [2] Department of Energy/Nuclear Regulatory Commission, *Radiological Dispersal Devices: An Initial Study to Identify Radioactive Materials of Greatest Concern and Approaches to their Tracking, Tagging, and Disposition*, prepared by the DOE/NRC Interagency Working Group on Radiological Dispersal Devices, report to the NRC and Secretary of Energy, May 2003.
- [3] Eckerman, K., et al., *Dose Coefficient Factor Package*, Oak Ridge National Laboratory, Oak Ridge, TN, 1996.
- [4] Eckerman, K. and R.W. Leggett, *User Guide to DCFPAK 3.0*, Oak Ridge National Laboratory, Oak Ridge, TN, 2011.
- [5] Environmental Protection Agency, *PAG Manual: Protective Action Guides and Planning Guidance for Radiological Incidents*, EPA-400/R-17/001, January 2017.
- [6] Federal Radiological Monitoring and Assessment Center, *FRMAC Assessment Manual Volume 1, Overview and Methods*, SAND2020-10446 R, Sandia National Laboratories, July 2020.
- [7] Griggs, J., *Use of Drinking Water Methods for Analysis of Radionuclides in Water Following a Radiological Event*, National Analytical Radiation Environmental Laboratory, December 2019.
- [8] Fulton, J., et al., Turbo FRMAC© Assessment Software Package, Sandia National Laboratories, Albuquerque, NM, 2021.
- [9] International Council on Radiation Protection, *Doses to the Embryo and Fetus from Intakes of Radionuclides by the Mother*, ICRP Publication 88, Ottawa, Ontario, Canada, 2002.
- [10] International Council on Radiation Protection, *Doses to Infants from Ingestion of Radionuclides in Mother’s Milk*, ICRP Publication 95, Ottawa, Ontario, Canada, 2004.
- [11] Kraus, T., Foster, K. *FINAL: Analysis of Top Dose Producing Radionuclides from Uranium- and Plutonium-Fueled Improvised Nuclear Devices*, FY12 Final Report (SNL-CM-082J), SAND2012-8533, October 2012.
- [12] Nuclear Regulatory Commission, *State-of-the-Art Reactor Consequence Analyses Project Volume 1: Peach Bottom Integrated Analysis*, NUREG/CR 7110, May 2013.
- [13] Rhodes, W., *Radioactive Material Downselection and Source Prioritization Methodology*, SAND2009-1076P, Sandia National Laboratories, Albuquerque, NM, September 2008.

APPENDIX A. SCENARIO DRL AND MDC COMPARISON TABLES

Table A-1. NPP Scenario Six Age Group Water DRL to MDC Comparison for No Decay and Decay Cases

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - No Decay ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - Decay Included ($\mu\text{Ci/l}$)	MDC (Griggs Report) (pCi/l)	Meet MDC -No Decay ($\Delta\text{pCi/l}$)	Meet MDC - Decay Included ($\Delta\text{pCi/l}$)	Comments
Ba-140	9.31E-04	9.31E-04	1.40E-02	1.32E+01	9.18E+02	1.40E+04	
Ce-141	6.40E-05	6.40E-05	9.60E-04	5.40E+00	5.86E+01	9.55E+02	
Ce-144	4.60E-05	4.60E-05	6.90E-04	1.67E+01	2.93E+01	6.73E+02	
Cs-134	9.89E-06	9.89E-06	1.48E-04	3.40E+00	6.49E+00	1.45E+02	
Cs-136	3.72E-06	3.72E-06	5.59E-05	2.50E+00	1.22E+00	5.34E+01	
Cs-137	9.89E-06	9.89E-06	1.48E-04	4.00E+00	5.89E+00	1.44E+02	
I-129	2.34E-15	2.34E-15	3.51E-14	Not Included			Very low energy 30kev and 40kev lines, special method like neutron activation analysis is required to get a reasonable detection limit
I-131	6.87E-04	6.87E-04	1.03E-02	3.60E+00	6.84E+02	1.03E+04	
I-132	8.94E-04	8.94E-04	1.34E-02	3.50E+00	8.90E+02	1.34E+04	
I-133	1.17E-03	1.17E-03	1.75E-02	3.70E+00	1.16E+03	1.75E+04	
I-134	3.09E-06	3.09E-06	4.64E-05	Not Included			52 min half-life, gamma spec (847, 884, and 1075kev lines)
I-135	6.26E-04	6.26E-04	9.39E-03	Not Included			6 hr half-life, gamma spec (250, 1260, and 1131kev)
La-140	1.22E-04	1.22E-04	1.83E-03	4.10E+00	1.18E+02	1.83E+03	
Nb-95	1.92E-05	1.92E-05	2.88E-04	3.50E+00	1.57E+01	2.85E+02	
Np-239	5.58E-04	5.58E-04	8.38E-03	0.00E+00	5.58E+02	8.38E+03	
Pu-238	7.56E-08	7.56E-08	1.13E-06	0.18	-1.04E-01	9.55E-01	MDC estimate based on a 100 min count time
Pu-239	1.51E-08	1.51E-08	2.27E-07	0.18	-1.65E-01	4.70E-02	MDC estimate based on a 100

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - No Decay ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - Decay Included ($\mu\text{Ci/l}$)	MDC (Griggs Report) (pCi/l)	Meet MDC -No Decay ($\Delta\text{pCi/l}$)	Meet MDC - Decay Included ($\Delta\text{pCi/l}$)	Comments
							min count time
Pu-240	1.22E-08	1.22E-08	1.83E-07	0.18	-1.68E-01	3.32E-03	Note: Pu-239+Pu-240 from alpha spec
Pu-241	4.25E-06	4.25E-06	6.37E-05	3.60E+00	6.47E-01	6.01E+01	
Sr-89	5.64E-04	5.64E-04	8.47E-03	8.00E-01	5.63E+02	8.47E+03	
Sr-90	4.42E-05	4.42E-05	6.63E-04	5.00E-01	4.37E+01	6.63E+02	
Sr-91	3.96E-04	3.96E-04	5.94E-03	Not Included			9hr half-life, decent gamma yield
Te-127m	6.98E-06	6.98E-06	1.05E-04	Not Included			Very low gamma yield, hard to detect, could not find a good method for it
Te-129m	2.50E-05	2.50E-05	3.75E-04	1.44E+03	-1.42E+03	-1.07E+03	
Te-131m	8.14E-05	8.14E-05	1.22E-03	6.60E+00	7.48E+01	1.22E+03	
Te-132	7.56E-04	7.56E-04	1.13E-02	2.80E+00	7.53E+02	1.13E+04	
Zr-95	6.98E-05	6.98E-05	1.05E-03	6.50E+00	6.33E+01	1.04E+03	
Zr-97	4.83E-05	4.83E-05	7.25E-04	Not Included			16hr half-life, good gamma signature

Table A-2. Pu IND Scenario Six Age Group Water DRL to MDC Comparison for No Decay and Decay Cases

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - No Decay ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - Decay Included ($\mu\text{Ci/l}$)	MDC (NIRT Report) (pCi/l)	Meet MDC -No Decay ($\Delta\text{pCi/l}$)	Meet MDC - Decay Included ($\Delta\text{pCi/l}$)	Comments
Ba-140	4.081E-04	7.10E-05	2.01E-02	13.2	5.78E+01	2.01E+04	
Ba-141	4.021E-02	7.00E-03	1.98E+00	Not Included			18min half-life
Ba-142	1.180E-02	2.05E-03	5.81E-01	Not Included			10min half-life
Ce-143	2.881E-03	5.01E-04	1.42E-01	6.3	4.95E+02	1.42E+05	
Ce-144	1.270E-05	2.21E-06	6.25E-04	Not Included			Gamma spec at 133kev
Co-58	5.231E-06	9.10E-07	2.58E-04	2.5	-1.59E+00	2.55E+02	
Co-58m	8.432E-04	1.47E-04	4.15E-02	Not Included			9hr half-life, 810 kev gamma 100% yield
Cs-134m	4.661E-05	8.11E-06	2.30E-03	Not Included			3hr half life
Cs-137	5.882E-07	1.02E-07	2.90E-05	4	-3.90E+00	2.50E+01	
Cs-138	9.653E-02	1.68E-02	4.75E+00	Not Included			33min half-life
I-131	2.121E-04	3.69E-05	1.04E-02	3.6	3.33E+01	1.04E+04	
I-132	1.470E-03	2.56E-04	7.24E-02	3.5	2.52E+02	7.24E+04	
I-133	5.842E-03	1.02E-03	2.88E-01	3.7	1.01E+03	2.88E+05	
I-134	8.192E-02	1.43E-02	4.03E+00	Not Included			52min-half life but can be detected by gamma spec with relative ease (847, 884, and 1075kev lines)
I-135	1.951E-02	3.39E-03	9.60E-01	Not Included			6hr half-life, good gamma signature at 250, 1260, and 1131kev
La-141	2.481E-02	4.32E-03	1.22E+00	Not Included			4hr half-life
La-142	5.081E-02	8.84E-03	2.50E+00	Not Included			91min half-life
Mn-54	9.102E-07	1.58E-07	4.48E-05	Not Included			Gamma spec at 834kev
Mn-56	6.232E-02	1.08E-02	3.07E+00	Not Included			2.6hr half-life
Mo-99	2.111E-03	3.67E-04	1.04E-01	28.9	3.38E+02	1.04E+05	

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - No Decay ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - Decay Included ($\mu\text{Ci/l}$)	MDC (NIRT Report) (pCi/l)	Meet MDC -No Decay ($\Delta\text{pCi/l}$)	Meet MDC - Decay Included ($\Delta\text{pCi/l}$)	Comments
Mo-101	3.721E-02	6.47E-03	1.83E+00	Not Included			14min half-life
Pu-239	1.110E-07	1.93E-08	5.47E-06	0.18	-1.61E-01	5.29E+00	Alpha spec, 100 min count time
Rb-89	1.260E-02	2.19E-03	6.21E-01	Not Included			15min half-life
Ru-103	1.700E-04	2.96E-05	8.37E-03	3.5	2.61E+01	8.37E+03	
Ru-105	2.511E-02	4.37E-03	1.24E+00	33.6	4.33E+03	1.24E+06	
Ru-106	1.170E-05	2.04E-06	5.76E-04	33.6	-3.16E+01	5.43E+02	
Sb-128	1.120E-03	1.95E-04	5.52E-02	Not Included			9hr half-life
Sb-129	6.812E-03	1.19E-03	3.35E-01	Not Included			4hr half-life
Sb-130	1.290E-02	2.24E-03	6.35E-01	Not Included			40min half-life
Sb-131	2.901E-02	5.05E-03	1.43E+00	Not Included			23min half-life
Sn-128	9.422E-03	1.64E-03	4.64E-01	Not Included			60min half-life
Sr-90	1.931E-07	3.36E-08	9.51E-06	0.5	-4.66E-01	9.01E+00	
Sr-91	5.802E-03	1.01E-03	2.86E-01	Not Included			9hr half-life
Sr-92	2.021E-02	3.52E-03	9.95E-01	Not Included			2.7hr half-life
Tc-99m	2.001E-04	3.48E-05	9.85E-03	3	3.18E+01	9.85E+03	
Tc-101	1.050E-01	1.83E-02	5.17E+00	Not Included			14 min half-life
Te-131	5.471E-02	9.52E-03	2.69E+00	2.6	9.52E+03	2.69E+06	
Te-131m	5.661E-04	9.85E-05	2.79E-02	6.6	9.19E+01	2.79E+04	
Te-132	1.540E-03	2.68E-04	7.58E-02	2.8	2.65E+02	7.58E+04	
Te-133m	3.761E-02	6.54E-03	1.85E+00	Not Included			55 min half-life
Te-134	5.952E-02	1.04E-02	2.93E+00	Not Included			41 min half-life
Y-93	8.352E-03	1.45E-03	4.11E-01	Not Included			10 hr half-life
Y-94	3.381E-02	5.88E-03	1.66E+00	Not Included			18 min half-life
Y-95	1.070E-02	1.86E-03	5.27E-01	Not Included			10 min half-life
Zr-95	6.792E-05	1.18E-05	3.34E-03	6.5	5.32E+00	3.34E+03	
Zr-97	7.102E-03	1.24E-03	3.50E-01	2.6	1.23E+03	3.50E+05	

Table A-3. Pu RTG Scenario Six Age Group Water DRL to MDC Comparison for No Decay and Decay Cases

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - No Decay ($\mu\text{Ci/l}$)	Most Restrictive Mixture 365d DRL - Decay Included ($\mu\text{Ci/l}$)	MDC (NIRT Report) (pCi/l)	Meet MDC -No Decay ($\Delta\text{pCi/l}$)	Meet MDC - Decay Included ($\Delta\text{pCi/l}$)	Comments
Am-241	1.89E-08	1.88552E-08	1.89297E-08	0.022	-3.14E-03	-3.07E-03	
Pu-238	9.73E-05	9.7282E-05	9.76665E-05	0.07	9.72E+01	9.76E+01	
Pu-239	5.23E-08	5.23028E-08	5.25095E-08	0.034	1.83E-02	1.85E-02	
Pu-240	3.85E-08	3.85302E-08	3.86825E-08	0.034	4.53E-03	4.68E-03	
Pu-241	7.92E-07	7.92466E-07	7.95598E-07	4.478	-3.69E+00	-3.68E+00	
Pu-242	1.64E-11	1.63959E-11	1.64607E-11	0.07	-7.00E-02	-7.00E-02	
U-234	1.57E-09	1.56854E-09	1.57474E-09	0.059	-5.74E-02	-5.74E-02	
U-237	1.90E-11	1.90192E-11	1.90944E-11	Not Included			Gamma spec if there was enough

Table A-4. RDD Scenario Six Age Group Water DRL to MDC Comparison for No Decay and Decay Cases

Radionuclide	Concentration ($\mu\text{Ci/l}$)	Most Restrictive Individual 365d DRL - No Decay ($\mu\text{Ci/l}$)	Most Restrictive Individual 365d DRL - Decay Included ($\mu\text{Ci/l}$)	MDC (NIRT Report) ($\mu\text{Ci/l}$)	Meet MDC - No Decay ($\Delta\mu\text{Ci/l}$)	Meet MDC - Decay Included ($\Delta\mu\text{Ci/l}$)
Am-241	1.00E+00	1.04E-04	3.90E-03	24.996	7.89E+01	3.87E+03
Cf-252	1.00E+00	7.78E-05	8.85E-05	0.074	7.78E+01	8.84E+01
Cm-244	1.00E+00	1.32E-04	1.35E-04	0.07	1.32E+02	1.35E+02
Co-60	1.00E+00	7.15E-03	7.63E-03	3.716	7.15E+03	7.63E+03
Cs-137	1.00E+00	6.14E-03	6.21E-03	3.998	6.14E+03	6.21E+03
Ir-192	1.00E+00	2.89E-02	1.02E-01	3.356	2.89E+04	1.02E+05
Po-210	1.00E+00	1.49E-05	3.25E-05	0.07	1.48E+01	3.24E+01
Pu-238	1.00E+00	9.74E-05	9.78E-05	0.07	9.73E+01	9.77E+01
Pu-239	1.00E+00	9.25E-05	9.25E-05	0.07	9.25E+01	9.25E+01
Ra-226	1.00E+00	5.41E-05	5.41E-05	0.489	5.36E+01	5.37E+01
Se-75	1.00E+00	1.63E-02	3.92E-02	4.386	1.63E+04	3.92E+04
Sr-90	1.00E+00	1.04E-03	1.06E-03	0.575	1.04E+03	1.05E+03
Tm-170	1.00E+00	2.42E-02	5.54E-02	145.166	2.41E+04	5.53E+04
U-235	1.00E+00	1.10E-03	1.10E-03	4.794	1.10E+03	1.10E+03
Yb-169	1.00E+00	5.48E-02	4.33E-01	6.559	5.48E+04	4.33E+05

This page left blank

DISTRIBUTION

Email—Internal

Name	Org.	Sandia Email Address
Lainy Cochran	06631	bhunt@sandia.gov
Kevin Hart	06634	kghart@sandia.gov
Brian Hunt	06631	bhunt@sandia.gov
Technical Library	01977	sanddocs@sandia.gov

Email—External [REDACTED]

Name	Company Email Address	Company Name
Holly Arrigoni	Arrigoni.Holly@epa.gov	US EPA
Sara DeCair	DeCair.Sara@epa.gov	US EPA
Wagnus Prioleau	prioleau.wagnus@epa.gov	US EPA

This page left blank

This page left blank



**Sandia
National
Laboratories**

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.