

Calculation of Radionuclide Inventories for Use in NUTS  
in the Performance Assessment Baseline Calculation

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## TABLE OF CONTENTS

1.	INTRODUCTION.....	3
1.1	ACRONYMS.....	4
1.2	SYMBOLS.....	4
2.	PROBLEM DESCRIPTION.....	5
3.	COMPUTATIONAL METHODOLOGY .....	6
3.1	THEORY .....	6
3.2	IMPLEMENTATION.....	7
3.3	EQUATIONS.....	7
4.	RESULTS .....	9
5.	RELEVANT PROCEDURES AND REFERENCES.....	9
5.1	PROCEDURES .....	9
5.2	REFERENCES .....	10

## TABLES

Table 1: Percent Contribution at 2033 to EPA Units for Isotopes Modeled in NUTS .....	5
Table 2: Radionuclide Half-life Values Used in Calculation .....	6
Table 3: Lumped Radionuclide Inventory Values As of 12/31/2033 .....	9

## 1. INTRODUCTION

In 1996 the Department of Energy (DOE) completed a performance assessment (PA) calculation for the Waste Isolation Pilot Plant (WIPP). The PA was part of the Compliance Certification Application (CCA) submitted to the Environmental Protection Agency (EPA) to demonstrate compliance with the radiation protection regulations of 40 CFR 191 and 40 CFR 194 (DOE 1996). As required by the WIPP Land Withdrawal Act (Public Law 102-579), DOE is required to submit documentation to EPA for the recertification of the WIPP every five years in order to continue operating the site. This required that a Compliance Recertification Application (CRA) be prepared, and CRA-2004 was submitted to the EPA in March 2004 (DOE 2004).

EPA is currently performing a completeness review of CRA-2004 and will soon start a final evaluation to determine if CRA-2004 demonstrates continued compliance with the disposal regulations governing WIPP. It is expected that as part of their final evaluation, the EPA will require that another PA, the Performance Assessment Baseline Calculation (PABC), is performed.

During the final preparation of CRA-2004, Mr. P.E. Shoemaker (Shoemaker 2003) of Sandia National Laboratories (SNL) requested a review of the waste stream profiles that form the basis of the inventory estimates for CRA-2004 (DOE 2004). The review was performed by Mr. J.P. Harvill (2004) of Washington TRU Solutions LLC (WTS). Mr. Harvill (2004) found a number of inconsistencies and possible omissions in the reporting of transuranic (TRU) waste inventories for CRA-2004. In response to Mr. Harvill's review, LANL-CO and SNL investigated the noted inconsistencies and possible omissions. The results of the laboratories investigations are documented in Leigh and Crawford (2004).

In addition, the EPA as part of its completeness review for CRA-2004 thoroughly examined the CRA-2004 inventory and visited several of the TRU waste sites to review information about the TRU waste inventory. The EPA review highlighted the need for inventory updates that should be incorporated in the PABC.

An update to the CRA-2004 inventory, governed by AP-113, *Analysis Plan For Inventory Reconciliation: Compliance Recertification Application*, has been completed. This inventory update is the basis for the PA inventory for the PABC.

This analysis is governed by AP-119, *Analysis Plan For Deriving Radionuclide Inventory Information for Performance Assessment Calculations: Post CRA Performance Assessment Baseline Calculation* which discusses the methodology that will be used by Sandia National Laboratories (SNL) to determine the WIPP repository radionuclide inventory information for use in the PA calculation for the PABC. In particular, the radionuclide inventories for the NUTS code are needed.

This analysis was performed in accordance with the SNL Quality Assurance Program and was prepared as prescribed by the SNL NWMP Procedure, NP 9-1, *Analyses*.

## 1.1 ACRONYMS

AP	Analysis Plan
CCA	Compliance Certification Application
CFR	Code of Federal Regulations
CH	Contact Handled
CRA	Compliance Recertification Application
DOE	Department of Energy
EPA	Environmental Protection Agency
ERMS	Electronic Records Management System
LANL-CO	Los Alamos National Laboratories – Carlsbad Operations
NP	NWMP Procedure
NWMP	Nuclear Waste Management Program
ORIGEN	Oak Ridge Isotope Generation and Depletion Code
PA	Performance Assessment
PABC	Performance Assessment Baseline Calculation
RH	Remote Handled
SNL	Sandia National Laboratories
TRU	Transuranic
WIPP	Waste Isolation Pilot Plant
WTS	Washington TRU Solutions LLC

## 1.2 SYMBOLS

$A_L(\text{Am}241)_{\text{CH}}$	Activity of the parameter INVCHD for the material AM241L
$A_L(\text{Th}230)_{\text{CH}}$	Activity of the parameter INVCHD for the material TH230L
$A_L(\text{Pu}238)_{\text{CH}}$	Activity of the parameter INVCHD for the material PU238L
$A_L(\text{U}234)_{\text{CH}}$	Activity of the parameter INVCHD for the material U234L
$A_L(\text{Pu}239)_{\text{CH}}$	Activity of the parameter INVCHD for the material PU239L
$A_L(\text{Am}241)_{\text{RH}}$	Activity of the parameter INVRHD for the material AM241L
$A_L(\text{Th}230)_{\text{RH}}$	Activity of the parameter INVRHD for the material TH230L
$A_L(\text{Pu}238)_{\text{RH}}$	Activity of the parameter INVRHD for the material PU238L
$A_L(\text{U}234)_{\text{RH}}$	Activity of the parameter INVRHD for the material U234L
$A_L(\text{Pu}239)_{\text{RH}}$	Activity of the parameter INVRHD for the material PU239L
$A(\text{Am}241)_{\text{CH}}$	Activity of $^{241}\text{Am}$ from Table 1 for CH-TRU
$A(\text{Th}230)_{\text{CH}}$	Activity of $^{230}\text{Th}$ from Table 1 for CH-TRU
$A(\text{Th}229)_{\text{CH}}$	Activity of $^{229}\text{Th}$ from Table 1 for CH-TRU
$A(\text{Pu}238)_{\text{CH}}$	Activity of $^{238}\text{Pu}$ from Table 1 for CH-TRU
$A(\text{U}234)_{\text{CH}}$	Activity of $^{234}\text{U}$ from Table 1 for CH-TRU
$A(\text{U}233)_{\text{CH}}$	Activity of $^{233}\text{U}$ from Table 1 for CH-TRU
$A(\text{Pu}239)_{\text{CH}}$	Activity of $^{239}\text{Pu}$ from Table 1 for CH-TRU
$A(\text{Pu}240)_{\text{CH}}$	Activity of $^{240}\text{Pu}$ from Table 1 for CH-TRU
$A(\text{Pu}241)_{\text{CH}}$	Activity of $^{241}\text{Pu}$ from Table 1 for CH-TRU
$A(\text{Pu}242)_{\text{CH}}$	Activity of $^{242}\text{Pu}$ from Table 1 for CH-TRU
$A(\text{Am}241)_{\text{RH}}$	Activity of $^{241}\text{Am}$ from Table 1 for RH-TRU
$A(\text{Th}230)_{\text{RH}}$	Activity of $^{230}\text{Th}$ from Table 1 for RH-TRU

A(Th229) <sub>RH</sub>	Activity of <sup>229</sup> Th from Table 1 for RH-TRU
A(Pu238) <sub>RH</sub>	Activity of <sup>238</sup> Pu from Table 1 for RH-TRU
A(U234) <sub>RH</sub>	Activity of <sup>234</sup> U from Table 1 for RH-TRU
A(U233) <sub>RH</sub>	Activity of <sup>233</sup> U from Table 1 for RH-TRU
A(Pu239) <sub>RH</sub>	Activity of <sup>239</sup> Pu from Table 1 for RH-TRU
A(Pu240) <sub>RH</sub>	Activity of <sup>240</sup> Pu from Table 1 for RH-TRU
A(Pu241) <sub>RH</sub>	Activity of <sup>241</sup> Pu from Table 1 for RH-TRU
A(Pu242) <sub>RH</sub>	Activity of <sup>242</sup> Pu from Table 1 for RH-TRU
τ <sub>1/2</sub> (RN)	Half-life of a radionuclide

## 2. PROBLEM DESCRIPTION

LANL (2005) reports that the DOE waste generator sites are expecting to dispose of TRU waste containing 138 different radionuclides in the WIPP. Not all of these radionuclides have significant inventories for disposal and not all of these radionuclides are expected to contribute to potential releases from the WIPP repository over the 10,000 year regulatory period. The analysis, *Radionuclides Expected to Dominate Potential Releases in the Performance Assessment Baseline Calculation Revision 0* (Leigh and Fox 2005a), identifies which radionuclides are expected to contribute to potential releases from the repository for the individual transport pathways modeled in performance assessment. For transport through the Salado to the accessible boundary and transport through the Salado to the Culebra which is modeled using the NUTS code in performance assessment, the important radionuclides are shown in Table 1.

Table 1: Percent Contribution at 2033 to EPA Units for Isotopes Modeled in NUTS

ID	Total Inventory [Curies] <sup>a</sup>			Source EPA Unit <sup>b</sup>			
	CH	RH	Total	CH	RH	Total	Cum %
<b>Pu-238</b>	1.13E+06	2.96E+03	1.13E+06	4.85E+03	1.28E+01	4.86E+03	47.72
<b>Pu-239</b>	5.77E+05	5.24E+03	5.82E+05	2.49E+03	2.26E+01	2.51E+03	72.37
<b>Am-241</b>	5.01E+05	1.65E+04	5.17E+05	2.16E+03	7.09E+01	2.23E+03	94.25
<b>Pu-240</b>	9.38E+04	1.58E+03	9.54E+04	4.04E+02	6.81E+00	4.11E+02	98.29
<b>U-233</b>	1.10E+03	1.27E+02	1.23E+03	4.74E+00	5.47E-01	5.29E+00	98.34
<b>U-234</b>	3.13E+02	3.08E+01	3.44E+02	1.35E+00	1.33E-01	1.48E+00	98.35
<b>Pu-242</b>	1.22E+01	4.80E-01	1.27E+01	5.25E-02	2.07E-03	5.46E-02	98.35
<b>Th-229</b>	4.65E+00	5.64E-01	5.21E+00	2.00E-02	2.43E-03	2.25E-02	98.35
<b>Th-230</b>	1.69E-01	1.07E-02	1.80E-01	7.30E-04	4.61E-05	7.76E-03	98.35

<sup>a</sup>Decayed radionuclide data taken from Leigh and Fox 2005b; <sup>b</sup>Source EPA Units taken from Leigh and Fox 2005a.

The radionuclides in Table 1 are important contributors to the unit of waste at the time of repository closure and they have half-lives that are long enough to contribute to potential releases via the Salado. In addition, Leigh and Fox 2005a identifies <sup>241</sup>Pu as being important because it is the parent isotope for <sup>241</sup>Am and it has a significant initial inventory (Leigh and Fox 2005a). The inventory for <sup>241</sup>Pu at the time of repository closure (2033) is 4.20E+05 curies in contact handled (CH) TRU waste and 2.80E+04 curies in remote handled (RH) TRU waste (Leigh and Fox 2005b).

The radionuclides in Table 1 are members of the following decay chains. The half-lives used in equations 10 – 13 are given in Table 2.

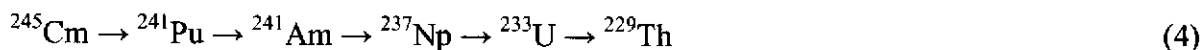
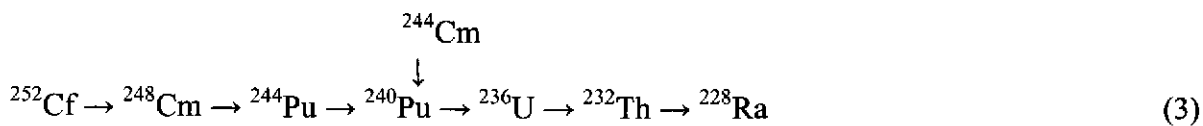
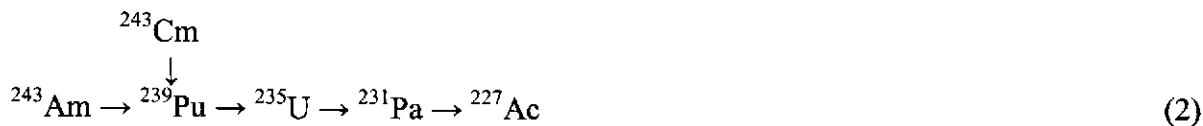
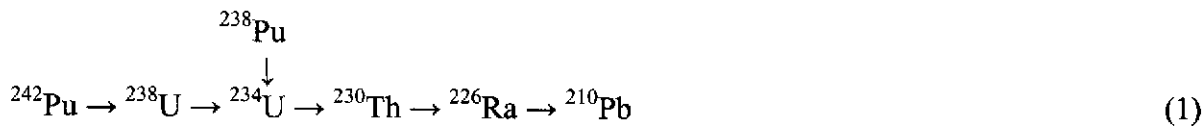


Table 2: Radionuclide Half-life Values Used in Calculation<sup>1</sup>

Radionuclide	Half-life (seconds)
<sup>241</sup> Am	1.364E+10
<sup>239</sup> Pu	7.594E+11
<sup>241</sup> Pu	4.544E+8
<sup>242</sup> Pu	1.221E+13

<sup>1</sup> PAPDB, 2002

Because NUTS is a computationally intensive code, minimizing the number of radionuclides that NUTS must track in a calculation is *beneficial*. Therefore, inventories for the uranium, plutonium, americium and thorium isotopes need to be combined into “lumped” inventories to facilitate the NUTS calculations for performance assessment. Technical justification for lumping is provided below.

### 3. COMPUTATIONAL METHODOLOGY

#### 3.1 THEORY

The theory behind combining radionuclide inventories into “lumped” inventories for transport calculations is that radionuclides of the same elemental form will transport at the same rate. Therefore, the inventories for isotopes of uranium can be combined. Inventories for isotopes of

plutonium can be combined, and inventories for isotopes of thorium can be combined. Using the isotopes identified in Table 1 as important: (1) the activity of  $^{234}\text{U}$  and  $^{233}\text{U}$  will be combined to produce values for the material U234L (the lumped uranium inventory) for the NUTS calculation, (2) the activity of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{242}\text{Pu}$  will be combined to produce values for the material PU239L for the NUTS calculation, and (3) the activity of  $^{230}\text{Th}$  and  $^{229}\text{Th}$  will be combined to produce values for the material TH230L in the NUTS calculation.

$^{238}\text{Pu}$  is also listed as an important radionuclide for the NUTS calculation in Table 1. The inventory of  $^{238}\text{Pu}$  is not combined with the other plutonium isotopes because of its relatively short half-life.

Finally, the inventory of  $^{241}\text{Pu}$  is combined with the inventory of  $^{241}\text{Am}$  because  $^{241}\text{Pu}$  is a parent isotope to  $^{241}\text{Am}$  and  $^{241}\text{Pu}$  has a relatively short half-life when compared to  $^{241}\text{Am}$ . Combining the inventories for  $^{241}\text{Am}$  and  $^{241}\text{Pu}$  produces the inventory value for the material AM241L in the NUTS calculation.

The resulting decay chains for the NUTS calculations are:



### 3.2 IMPLEMENTATION

One can either add the curies of two isotopes to get a combined inventory or one can add the moles. Depending on the half-lives of the isotopes that are being combined, one method will result in a larger combined activity than the other. If for example, isotope A has a longer half-life than isotope B and the inventory of isotope B is to be combined with the inventory of isotope A, the method that results in the largest combined activity for isotope A is to add the curies of isotope B to the curies of isotope A. If on the other hand, the half-life of isotope A is shorter than the half-life of isotope B, the method that results in the largest combined activity for isotope A is to add the moles of isotope B to the moles of isotope A. Therefore, when combining the plutonium isotopes,  $^{242}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{239}\text{Pu}$ , the curies of  $^{240}\text{Pu}$  are added to the curies of  $^{239}\text{Pu}$  but the moles of  $^{242}\text{Pu}$  are added to the moles of  $^{239}\text{Pu}$ . When combining the thorium isotopes, the curies of  $^{229}\text{Th}$  are added to the curies of  $^{230}\text{Th}$ . When combining the uranium isotopes, the curies of  $^{233}\text{U}$  are added to the curies of  $^{234}\text{U}$ .

### 3.3 EQUATIONS

Equations 8 through 17 show the computational methodology for combining radionuclide activities into lumped activity values.

$$A_L(\text{Pu238})_{\text{CH}} = A(\text{Pu238})_{\text{CH}} \tag{8}$$

$$A_L(\text{Pu238})_{\text{RH}} = A(\text{Pu238})_{\text{RH}} \quad (9)$$

where:

$A_L(\text{Pu238})_{\text{CH}}$  activity value for the property INVCHD for the material PU238L  
 $A(\text{Pu238})_{\text{CH}}$  activity value for  $^{238}\text{Pu}$  from Table 1 for CH-TRU  
 $A_L(\text{Pu238})_{\text{RH}}$  activity value for the property INVRHD for the material PU238L  
 $A(\text{Pu238})_{\text{RH}}$  activity value for  $^{238}\text{Pu}$  from Table 1 for RH-TRU

$$A_L(\text{Am241})_{\text{CH}} = A(\text{Am241})_{\text{CH}} + A(\text{Pu241})_{\text{CH}} * \tau_{1/2}(\text{Pu241}) / \tau_{1/2}(\text{Am241}) \quad (10)$$

$$A_L(\text{Am241})_{\text{RH}} = A(\text{Am241})_{\text{RH}} + A(\text{Pu241})_{\text{RH}} * \tau_{1/2}(\text{Pu241}) / \tau_{1/2}(\text{Am241}) \quad (11)$$

where:

$A_L(\text{Am241})_{\text{CH}}$  activity value for the property INVCHD for the material AM241L  
 $A(\text{Am241})_{\text{CH}}$  activity value for  $^{241}\text{Am}$  from Table 1 for CH-TRU  
 $A(\text{Pu241})_{\text{CH}}$  activity value for  $^{241}\text{Pu}$  from Table 1 for CH-TRU  
 $A_L(\text{Am241})_{\text{RH}}$  activity value for the property INVRHD for the material AM241L  
 $A(\text{Am241})_{\text{RH}}$  activity value for  $^{241}\text{Am}$  from Table 1 for RH-TRU  
 $A(\text{Pu241})_{\text{RH}}$  activity value for  $^{241}\text{Pu}$  from Table 1 for RH-TRU  
 $\tau_{1/2}(\text{Am241})$  half-life of  $^{241}\text{Am}$  from Table 2  
 $\tau_{1/2}(\text{Pu241})$  half-life of  $^{241}\text{Pu}$  from Table 2

$$A_L(\text{Pu239})_{\text{CH}} = A(\text{Pu239})_{\text{CH}} + A(\text{Pu240})_{\text{CH}} + A(\text{Pu242})_{\text{CH}} * \tau_{1/2}(\text{Pu242}) / \tau_{1/2}(\text{Pu239}) \quad (12)$$

$$A_L(\text{Pu239})_{\text{RH}} = A(\text{Pu239})_{\text{RH}} + A(\text{Pu240})_{\text{RH}} + A(\text{Pu242})_{\text{RH}} * \tau_{1/2}(\text{Pu242}) / \tau_{1/2}(\text{Pu239}) \quad (13)$$

Where:

$A_L(\text{Pu239})_{\text{CH}}$  activity value for the property INVCHD for the material PU239L  
 $A(\text{Pu239})_{\text{CH}}$  activity value for  $^{239}\text{Pu}$  from Table 1 for CH-TRU  
 $A(\text{Pu240})_{\text{CH}}$  activity value for  $^{240}\text{Pu}$  from Table 1 for CH-TRU  
 $A(\text{Pu242})_{\text{CH}}$  activity value for  $^{242}\text{Pu}$  from Table 1 for CH-TRU  
 $A_L(\text{Pu239})_{\text{RH}}$  activity value for the property INVRHD for the material PU239L  
 $A(\text{Pu239})_{\text{RH}}$  activity value for  $^{239}\text{Pu}$  from Table 1 for RH-TRU  
 $A(\text{Pu240})_{\text{RH}}$  activity value for  $^{240}\text{Pu}$  from Table 1 for RH-TRU  
 $A(\text{Pu242})_{\text{RH}}$  activity value for  $^{242}\text{Pu}$  from Table 1 for RH-TRU  
 $\tau_{1/2}(\text{Pu239})$  half-life of  $^{239}\text{Pu}$  from Table 2  
 $\tau_{1/2}(\text{Pu242})$  half-life of  $^{242}\text{Pu}$  from Table 2

$$A_L(\text{U234})_{\text{CH}} = A(\text{U234})_{\text{CH}} + A(\text{U233})_{\text{CH}} \quad (14)$$

$$A_L(\text{U234})_{\text{RH}} = A(\text{U234})_{\text{RH}} + A(\text{U233})_{\text{RH}} \quad (15)$$



Where:

$A_L(U234)_{CH}$  activity value for the property INVCHD for the material U234L  
 $A(U234)_{CH}$  activity value for  $^{234}U$  from Table 1 for CH-TRU  
 $A(U233)_{CH}$  activity value for  $^{233}U$  Table 1 for CH-TRU  
 $A_L(U234)_{RH}$  activity value for the property INVRHD for the material U234L  
 $A(U234)_{RH}$  activity value for  $^{234}U$  from Table 1 for RH-TRU  
 $A(U233)_{RH}$  activity value for  $^{233}U$  Table 1 for RH-TRU

$$A_L(Th230)_{CH} = A(Th230)_{CH} + A(Th229)_{CH} \quad (16)$$

$$A_L(Th230)_{RH} = A(Th230)_{RH} + A(Th229)_{RH} \quad (17)$$

Where:

$A_L(Th230)_{CH}$  activity value for the property INVCHD for the material TH230L  
 $A(Th230)_{CH}$  activity value for  $^{230}Th$  from Table 1 for CH-TRU  
 $A(Th229)_{CH}$  activity value for  $^{229}Th$  Table 1 for CH-TRU  
 $A_L(Th230)_{RH}$  activity value for the property INVRHD for the material TH230L  
 $A(Th230)_{RH}$  activity value for  $^{230}Th$  from Table 1 for RH-TRU  
 $A(Th229)_{RH}$  activity value for  $^{229}Th$  Table 1 for RH-TRU

## 4. RESULTS

Using the radionuclide activity values from Table 1 and the equations from Section 3.3 gives the values in Table 3 for the lumped radionuclide inventories at the end of 2033.

Table 3: Lumped Radionuclide Inventory Values As of 12/31/2033

Material	INVCHD (Total Curies)	INVRHD (Total Curies)
AM241L	5.15E+05	1.74E+04
PU238L	1.13E+06	2.96E+03
PU239L	6.71E+05	6.83E+03
TH230L	4.82E+00	5.75E-01
U234L	1.41E+03	1.58E+02

## 5. RELEVANT PROCEDURES AND REFERENCES

### 5.1 PROCEDURES

AP-113, *Analysis Plan for Inventory Reconciliation: Compliance Recertification Application*, Sandia National Laboratory Nuclear Waste Management Analysis Plan, February 3, 2005.

AP-119, *Analysis Plan For Deriving Radionuclide Inventory Information for Performance Assessment Calculations: Post CRA Performance Assessment Baseline Calculation*. Sandia National Laboratory Nuclear Waste Management Program Analysis Plan, April 4, 2005.

NP 9-1, *Analyses*. Sandia National Laboratory Nuclear Waste Management Program Procedure, August 29, 2001.

## 5.2 REFERENCES

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