REVIEW BY WDOH OF US ECOLOGY'S RADIOACTIVE WASTE SHIPMENT RECORDS

(June 1965 – December 1981)

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F. A. Palmer and A.A. Owen, "Review of Radioactive Shipment Records for the US Ecology LLRW Disposal Site" at Richland, WA; December 9, 1994

SUMMARY

Review of the US Ecology's radioactive waste shipment records **(1965 through 1981)** by the Washington Department of Health (WDOH) for waste disposed at its LLRW facility located in Richland, Washington, is documented in Appendices A, B and C. Results of this review are provided in **Table 4.** The purpose of this study is to establish authenticity of the shipping records and formalize information (**mass and activity**) related to **U-235, U-238, natural uranium, depleted uranium; Tc-99, Cl-36, and I-129** are reviewed for their **activities** only. These nuclides are considered to be important contributors for the long-term doses in support of the Environmental Impact Statement (EIS) of the LLRW facility. **Tc-99m** is also included in this review, even though its long- term dose contribution is not considered to be significant because of its short half-life (6 hours) compared with **Tc-99**, which has a half-life of 212,000 years. Based on information from US Ecology, it was determined that much of the Tc-99m was mistakenly documented as Tc-99 by original shippers. **U-234** is not included in the list of nuclides reviewed by WDOH, since it was not identified in the US Ecology's shipping records. (However, because of its high specific activity compared to the rest of uranium nuclides, its contribution to the overall dose may be significant). **Plutonium nuclides** are also not included in this review because of the large degree of uncertainty in identifying its various isotopes as listed in the original manifests. A more comprehensive study of plutonium was performed by US Ecology (Ref. 5), and is included as Appendix F. It includes actual isotopic percentage for plutonium provided by the generators. Plutonium data from this study is believed to be more definitive, and hence is given preference over other data.

Since the start of its operation in 1965, the facility has received low-level radioactive wastes from various generators and waste processors through waste collectors. Records of shipment prior to February 26, 1982 existed only on paper copies of Radioactive Shipping Records (RSR's) and manifests. Review of these records, as they appear on the manifests, shows a large degree of inconsistency in the way the data was documented in the original manifests. This review identified several major concerns: (1) inconsistency in the activities (millicuries) of uranium nuclides with respect to their mass (kg); (2) Tc-99m listed as Tc-99; (3) plutonium nuclides lacking isotopic nomenclature; (4) one activity listed for multiple nuclides; (5) mathematical errors made by shippers when adding the mass and/or activity for nuclides of interest; (6) too many nuclides crowded in a small section of the manifest making it difficult to identify the nuclide as well as their mass and activity; (7) inconsistency in the shipping records and in the data base; (8) inaccurate data input by outside contractors not fully knowledgeable with the data base system and radioactivity. Some of these records were not standardized documents and, in many cases, did not provide the same level of information as present day records. Hence a need to rectify and formalize these records so that a more accurate data could be established for the US Ecology's EIS and the Closure Plan.

WDOH reviewed a total of **9,915 manifests,** covering a period from **June 1965** through the end of **1981.** These shipping records include information for about **a million packages.** The results of this review are summarized in Table 4. Mass and activity for **uranium nuclides** are documented in Appendices A and B; **Tc-99, Cl-36 and I-129** data is presented in Appendix C.

The **mass** (gm,kg) of uranium nuclides (U-235, U-238, U-depleted, U-natural) listed in Table 4, is obtained directly from the manifests. Since the activities of these nuclides as listed in the original manifests were determined to be inaccurate (shown in Table 2), the **activities** (mCi) for uranium

nuclides (shown in parenthesis in Table 4) are the **calculated values** based on **specific activity data** provided in **WAC 246-231-200**.

Since a large number of packages containing **Tc-99m** nuclides were mistakenly documented as **Tc-99** in the original manifests, a concentrated effort was made to modify these data based on knowledge of generator's waste stream. However, this effort was limited due to time constrain and a lack of definitive information concerning the source of their origin. The **Tc-99 activities** as presented in Table 4 are therefore considered to be **overly conservative.**

In many instances **Cl-36** activities were listed in the manifests along with other nuclides, making it difficult to get an accurate estimate of its activities.

Although **I-129** entries were very limited, the hand written data input for I-129 was poorly documented on the manifest making it difficult to identify. Most of the listed activity for **I-129** is based on the lower level of detection rather than actual detector activity. Studies have shown that this results in overestimate of I-129 (Ref.4).

An **internal audit** of the shipping records (reviewed by the WDOH reviewer) was independently performed by two staff members of the Radioactive Waste Management Section (WDOH). About **40%** of the manifests (**3,952** manifests out of a total of **9,915**) were internally audited. Results of their audit (data obtained by the two internal auditors compared with those of the original reviewer) showed less than **0.5 %** discrepancy in the mass (gm, kg) of U-235, natural and depleted uranium; U-238 showed a discrepancy of 1.71%. For the remaining nuclides such as Tc-99, Cl-36 and I-129, the discrepancies were 5.9%, 8.9% and 12 % respectively. The somewhat higher degree of discrepancy with Tc-99, Cl-36 and I-129 is related to, among other things, the lack of consistency in the data input by the shippers. Results of this audit are presented in Table 3.

BACKGROUND

This study was undertaken by Washington Department of Health (WDOH) to review the US Ecology's **radioactive shipment records** (referred to as **manifests**) for radioactive waste disposed at its Low-Level Radioactive Waste Facility (LLRWF), located in Richland, Washington.

Since the start of its **operation in 1965**, the facility has received low-level radioactive wastes from various generators and waste processors through waste collectors. Records of shipment prior to February 26, 1982 existed only on paper copies of Radioactive Shipping Records (RSR's) and manifests. These records were later entered into a computerize database. Review of these records (1965-1981) as they appear on the manifests show a large degree of inconsistency for uranium isotopes when compared with the US Ecology's computerized database for the same period. This problem in the data base system has been traced back (by US Ecology) to inconsistent data input by outside contractors not fully knowledgeable with radioactivity and low-level radioactive waste system. Some of these records were not standardized documents and, in many cases, did not provide the same level of information regarding isotopic activities as present day records. Therefore, there existed a need to rectify and formalize these records.

On January 21, 1987 the state of Washington issued an amendment to the US Ecology radioactive Material license, WN-I019-2 **(Ref. 1),** which required a complete record of the type, activity and location of all radioactive waste disposed at the site. This license condition, known as "**Condition**

58", required a complete inventory of all radioactive material previously buried at the LLRW facility. In order to fulfill the requirements of condition 58, US Ecology completed a review of its waste inventory and issued the "Condition 58" report to the department dated June 29, 1990. Modified data of the "Condition 58" report for the period 1965 through early 1982 (developed by US Ecology in support of the closure plan for the Richland LLRW facility) was completed and provided to the department dated December 9,1994 (Ref.5). This report (included as Appendix F) did not evaluate uranium and its isotopes. However, data contained in this report may be useful in comparing results obtained by the department from its review of the US Ecology radioactive waste shipment records.

State regulatory requirements for transfers of radioactive waste (LLRW) to a licensed low-level waste disposal facility is delineated in **WAC-246-249-090** (Ref.2)**.** The requirements of this regulation are designed to **control transfers** of low-level radioactive waste, **establish a manifest tracking system** and **supplement existing requirements** concerning transfer and record keeping for those wastes. This regulation further requires that the waste generator, collector, or processor who transports, or offers for transportation, low-level radioactive waste intended for ultimate disposal at a licensed LLRW facility, **prepare a manifest** reflecting information requested on applicable NRC Forms titled "**Uniform Low-Level Radioactive Waste Manifest";** NRC Form 540**, "Shipping Papers"** (Attachment 1); NRC Form 541**, "Container and Waste Description" (**Attachment 2)**;** NRC Form 542 **"Manifest Index and Compact Tabulation" (**Attachment 3). These NRC Forms must be completed and must physically accompany the pertinent low-level waste shipment. Additionally, it is required that the shipper of the radioactive waste provide, among other things, the following information regarding the waste shipment on the uniform manifest**: (1) the activity of each of the radio-nuclides H-3, C-14, Tc-99, and I-129 contained in the shipment (2) the total masses of U-233, U-235 and plutonium** in special nuclear material, and the total mass of uranium and thorium in source material.

Previous review of the radioactive shipment records by US Ecology (Ref.5) separated the inventory into three periods, **1965-1981, 1982-1987 and 1988-1994**. The last period is now modified to include the inventory from **1988 to 2003** (Ref. 4). Data entered during the first period (**1965-1981**) represents the data that was entered into the US Ecology's data base system retroactively. The period, **1981-1987,** represents data that was entered on a "real-time basis (i.e. as the shipments were received) but which has since been archived. The third period, **1988 to 2003**, represents data that has been accumulated on a "real-time" basis and still resides on the data base system.

This review is restricted to the following nuclides: **U-235, U-238, Depleted-U, Natural-U, Tc-99, Tc-99m, Cl-36 and I-129.** These nuclides are considered to be important contributors for the long-term doses in support of the Environmental Impact Statement (EIS) for the LLRW facility.). **Plutonium nuclides** are not included in this review because of the large degree of uncertainty in identifying its various isotopes as listed in the original manifests. A more comprehensive study of plutonium was performed by US Ecology (Ref. 5), and is included as Appendix F. It includes actual isotopic percentage for plutonium provided by the generators. Plutonium data from this study is believed to be more definitive, and hence is given preference over other data. **U-234** is also not included in this review since it was not identified in the US Ecology's shipping records. However, because of its high specific activity compared to the rest of uranium nuclides, its contribution to the overall dose may be significant.

PURPOSE

The **purpose** of this study is to **review each shipping record** (manifest) received by US Ecology's LLRW facility **starting June 1965 through the end of 1981**, and (formally) document the mass and activity of radionuclides related to **U-235, U-238, Depleted-U, Natural-U; Tc-99, Tc-99m, Cl-36 and I-129** are reviewed for their activities only**.** This documentation is needed to verify and establish a more accurate qualitative and quantitative data for the radionuclides disposed at the site.

MANIFEST REVIEW

A total of **9,915** radioactive waste **manifests (**covering June 1965 through December 1981) with approximately **one million packages, were reviewed by WDOH** for this study. The general finding of this review is documented in Appendix A. Appendices B and C provide more detailed information related to these nuclides. Each manifest ranged from a single page to over fifty pages containing radioactive waste information related to the type of nuclide, its mass (grams, kilograms and pounds), activity (mCi and curies), chemical and physical forms, and the weight and the volume (cubic feet) of packages containing the waste. The waste information provided in these shipping records (with few exception) is hand written, and in many cases include data that is poorly documented, lacking in clarity and uniformity (Attachments 4 and 5). Some waste shipping records appeared on non-standardized forms (work sheet) with incomplete data (Attachment 6). Attachment 7 is an example of a manifest that presents Cl-36 activity as combined with a host of other nuclides.

Although the manifests include a wide variety of radionuclides including tritium, Co-60, Sr-90 and Cs-137, (which make up more than 90% of the low-level waste), this review is limited to the following radionuclides: **U-235, U-238, depleted-U, natural-U, Tc-99, Tc-99m, Cl-36 and I-129 (Table 1).** Selection of these nuclides is based on their long half-life, along with their potential to contribute to the long-term individual and population dose resulting from ground water pathways and food chain factors. U-234 is not included in this review since the original manifests do not include data on U-234. A separate study on plutonium was conducted by US Ecology (Ref.5).

TABLE 1

Nuclides	**Half-life (yrs)**	**Sp. Activity** (WAC 246-231-200)
U-234	**2.46 E+5**	**6.2 E- 3 Ci/g = (6.2E+3 mCi/kg)**
U-235	**7.10 E +8**	**2.2 E - 6 Ci/g = (2.2 mCi/kg)**
U-238	**4.51 E +9**	**3.4 E - 7 Ci/g = (0.34 mCi/kg)**
U-Dep.	**-**	**5.0 E -7 Ci/g = (0.50 mCi/kg)**
U-Nat.	**-**	**7.1 E -7 Ci/g = (0.71 mCi/kg)**
Tc-99	**212,000**	
Tc-99m	**6.85 E- 4 (= 6 hrs)**	
Cl-36	**310,000**	
I-129	**16.0 E + 6**	

(Note: *All the nuclides included in the above table are long-lived except forTc-99m (half-life = 6 hours), which is included along with Tc-99 (half-life = 212,000 years) because of the difficulty encountered in differentiating between the two nuclides as listed in the manifests).*

The initial finding of this review, documented in **Appendix A,** shows data (mass and activity) related primarily to uranium nuclides, although limited data for other nuclides are also included. (A typical example of WDOH review material presented in Appendix A is shown in Attachment **9)**. A more comprehensive data related to **Tc-99, Tc-99m, Cl-36 and I-129** are included in **Appendix C**. The set of data in Appendix C documents only **the activity** (mCi) for Tc-99, Tc-99m, Cl-36 and I-129 **(**Attachment 11) since the mass for these nuclides are not documented in the original shipping records. Identity of companies and institutions generating Tc-99 and Tc-99m are included on these review data sheets whenever available.

Information related to uranium and its isotopes as documented in **Appendix A** is further **summarized on a yearly basis in Appendix B.** (An example of data presented in Appendix B is shown in Attachment 10)**.** Appendix B documents the mass (kilograms/pounds) and activity (mCi) of the uranium nuclides as they are listed in the US Ecology manifests even though activities related to the mass of uranium nuclides are considered to be inconsisten*t* and in disagreement with each other. These are shown in Attachment 8.

The following set of data derived from the US Ecology's manifests and presented in Table 2, highlight some of these inconsistencies when compared with the specific activity values given in WAC 246-231-200.

TABLE 2

Mass to Activity Conversion *(using WAC 246-231-200)*

Manifest	***Nuclide***	***Mass*** *(Manifest)*	***Activity*** *(Manifest)*	***Sp. Act.(WAC)***	***Calc. Activity (based on WAC)***
# 04068	***U-235***	*26.0 gm*	***1.0 mCi***	*2.2 mCi/kg*	*26 gm (2.2 mCi/kg)=* ***0.057 mCi***
# 04110	***U-235***	*360.0 gm*	***36.0 mCi***	*2.2 mCi/kg*	*360 gm (2.2 mCi/kg)=****0.792 mCi***
# 03781	***U-238***	*642.0 kg*	***7.2 mCi***	*0. 34 mCi/kg*	*642 kg (0.34 mCi/kg)=* ***218.3 mCi***
#08878	***U-238***	*540.0 kg*	***19.0 mCi***	*0.34 mCi/kg*	*540 kg (0.34 mCi/kg)=* ***183.6 mCi***
#09790	***U-depl***	*14.0 kg*	***0.0008 mCi***	*0.50 mCi/kg*	*14 kg (0.5 mCi/kg) =* ***7.0 mCi***
#09800	***U-depl***	*3,714.9 kg*	***1,340.0 mCi***	*0.50 mCi/kg*	*3,714kg (0.5 mCi/kg) =* ***1,857.5mCi***
#05435	***U-nat***	*5.77 kg*	***170.19 mCi***	*0.71 mCi/kg*	*5.77 kg (0.71 mCi/kg) =* ***4.10 mCi***
#09483	***U-nat***	*27.31kg*	***18.46 mCi***	*0.71 mCi/kg*	*27.31kg(0.71 mCi/kg) =* ***19.39 mCi***

A large degree of inconsistency relating to **technetium-99** (Tc-99) and **technetium-99m** (Tc-99m) was also observed in the data presented on the shipping records. In most cases Tc-99m was reported as Tc-99, partly due to the lack of knowledge and proper training among the shippers to differentiate between the two technetium isotopes; in many instances the manifests failed to identify the source (**generators)** for Tc-99m and Tc-99, making it difficult (for the reviewer) to conclude about the identity of the two nuclides. In many cases the activities of Tc-99 nuclides were reported as a combined activity with other nuclides and mixed fission products.

Cl-36 and **I-129** appear to form a very small percentage of the radionuclides identified in the waste shipping records. Their identification, like other nuclides, was at times, difficult to decipher, partly because of the poor quality of documentation in the original manifests. (Examples presented in Attachments 4 and 7).

Other observations related to the review of the radioactive waste shipment records are noted below:

1. Nuclides identified with chemical symbol (on the manifest), but missing the isotopic nomenclature e.g. U and Tc instead of U-238 and Tc-99 respectively.
2. Unreadable isotopes and isotope quantities.
3. Incomplete data entries e.g. missing the mass or the activity for a particular nuclide; also in many cases the date of shipment on the manifest was found missing.
4. Mass and activity of a nuclide often reported with other nuclides, e.g. U-238 activity often combined with mixed fission products.
5. In few instances, U-238 mass was reported combined with its container weight.
6. Plutonium nuclides (Pu-238, Pu239/240 and Pu-241) in many cases did not include their isotopic number.
7. Mathematical errors made by shippers when adding the mass and/or activity for nuclide of interest
8. Too many nuclides crowded in a small section of the manifest, making it difficult to identify the isotope and its mass and/or activity
9. Duplicate data provided with different control number
10. Name of waste generator(s) not identified on the manifest in many cases
11. The data input lacked uniformity and consistency, since most of the entries in the manifests were hand recorded by a large number of people, over a long period of time.
12. One activity listed for multiple nuclides in a package.

Uranium

One of the major problems associated with uranium and its isotope (U-235, U-238, U-depleted, U-natural) was the lack of consistency in the shipping records in relating the given mass (grams, kilograms) of uranium (isotope) with its activity (millcuries). An example to illustrate this point is shown earlier in Table 2.

Some manifests recorded the uranium isotopes including the special nuclear material (SNM) in grams, and their activities in curies, which did not relate and agree with each other, partly because the activities were reported as a total activity combined with activities of other nuclides and fission products. After comparing a number of uranium mass data with its activity (listed in the manifests, as well as those available with US Ecology's data base for the years 1965- 1981), it was determined that data related to the **activity** of uranium isotope (in most cases) is considered to be incorrect.

Appendix B documents the mass (kilograms/pounds) and activity (mCi) of the uranium nuclides as they are listed in the US Ecology manifests even though activities related to the mass of uranium nuclides are considered to be inconsistent and in disagreement with each other.

Occasionally data on the manifest did not include the isotopic number for uranium, even though it included the values for mass and activity. In such cases, when the quantity was given in **grams**, this

review assumed it to be **U-235**. Likewise, when the mass was given in **kilograms**, it was assumed to be the mass of **U-238.**

On a few occasions, the mass of uranium-235 is recorded on the shipping papers in kilograms instead of grams; based on experience gained from reviews of previous shipping records, this is considered to be a typographical error, and hence assumed to be part of U-238. Likewise, there are few instances when uranium is listed as U (without any isotopic nomenclature) and its mass listed in grams; this review assumed it to be part of U-235 inventory.

A number of manifests (# 02426, 2362, 02404 and more) listed mass values for **U-238** and included the weight of shipping containers, making it difficult to identify the actual amount of U-238. In such cases, the total mass (including the containers weight) as shown on the manifest, was included for inventory, resulting in an overestimate of U-238.

Plutonium

One of the major difficulties associated with plutonium and its nuclides was related to its isotopic nomenclature. Most of the plutonium listed on the manifests did not include sufficient information for activities to be correctly identified. For these reasons, data for plutonium are not included in this report. A more exhaustive study related to plutonium and its nuclides disposed at the LLRW facility, was performed by US Ecology's staff (Ref. 5) and is included as Appendix F. This study includes actual isotopic percentage for plutonium provided by the generators. Data available from this study are believed to be more accurate, and hence should be given preference over other data.

Technetium –99

Tc-99 (half-life = 212,000 years) and **Tc-99m** (half-life = 6 hours) appear extensively on the original shipping records submitted by US Ecology for this study. It was determined that much of the Tc-99m nuclides were **mistakenly** documented as Tc-99. This mishap resulted partly because the data input in the original shipping records was believed to be documented (over a long period of time) by a wide variety of people lacking proper training and education. During the course of this review, a number of discussions were held with the US Ecology staff to resolve this issue and determine the true identity of these two nuclides, so that activities associated with these nuclides could be properly quantified.

The following criteria were adopted to quantify Tc-99:

- Tc-99 shipments originating from hospitals was considered to be Tc-99m
- Tc-99 associated with Mo-99 in equilibrium quantities was assumed to be Tc-99m
- All other Tc-99 was assumed correct

In many cases, Tc-99 activities (mCi) were reported on the manifests as combined with other nuclides. In such situations, the total activity was divided by the number of nuclides to obtain the required activity for Tc-99 (example: total activity of 12 mCi includes three other nuclides; the value for Tc-99 would then be 12 mCi/4 = 3 mCi).

Chlorine – 36

Cl-36 is not widely found in the shipping records. However at times it was presented along with a number of other nuclides in a manner that was difficult to identify (Attachments 4 and 7).

Iodine-129

I-129 entries were very limited compared to other isotopes. A major problem associated with I-129 was the difficulty in differentiating it with I-125 due to hand written data on the manifest.

INTERNAL AUDIT

An **internal audit** on the previously reviewed material (by the WDOH reviewer) was performed to rectify any mistakes in the shipping records and to provide the necessary quality assurance to the project. For reasons stated earlier, this audit focused on the following nuclides: U-235, U-238, depleted and natural uranium, Tc-99, Tc-99m, Cl-36 and I-129.

The internal audit was performed by two staff members of the Waste Management Section. Both of these auditors, because of their long- term association with the section, required very little training for this assignment. Documentations of their audit are presented in Appendices D and E.

Auditor #1 reviewed a total of **3,816 manifests**, which included two sets of data; the first set included manifests **# 00001 (June 1965) through #03472 (June 27,1978);** the second set covered manifests **# 9577 (Nov. 26, 1981) through # 9920 (Jan. 6, 1982).**

Auditor #2 (thoroughly familiar with the history as well as the day to day operation of the waste disposal facility) conducted his assignment by auditing a selected group of manifests, a total of **136,** among the following manifests**: #08584 (6/10/81) through #08699 (6/23/81) and #08841 (7/20/81) through #09200 (9/21/81).**

Each (internal) auditor compared the radioactive waste data which was reviewed and documented by the original (WDOH) reviewer **(Appendix A, B and C)** with those recorded on the US Ecology's manifests. The audit involved verifying the mass and activity associated with each of the nuclides identified in Table 1. Examples of their audit are shown in Attachments 12 and 13 respectively.

Findings (Internal-Audit):

The findings of the internal audit are **summarized** in two different ways. In the **first case**, mass (kilograms) and activity (curies) associated with the nuclides obtained by the internal auditors are compared with the mass and activity data documented by the original WDOH reviewer. The **differences** (mass and activity as the case may be) in the two sets of data are used to estimate the percentage difference related to the mass and activity of each of the nuclides. These are summarized below:

TABLE 3

Nuclides	Total (WDOH)	Differences	% Difference
U-235	90.372 kg	(+) 0.323 kg	0.35 %
U-238	716,670.0 kg	(+) 12,272 kg	1.71 %
U-Dep.	98,369.7 kg	(+) 26.27 kg	0.03 %
U-Nat.	3,908.5 kg	(+) 11.03 kg	0.28 %
Tc-99	802.44 mCi	(+) 47.87 mCi	5.97 %
Cl-36	974.93 mCi	(+) 87.04 mCi	8.92 %
I-129	7.07 mCi	(+) 1.15 mCi	16.3 %

NOTE:

1) Numbers under the column "Differences" show the differences in values between the materials reviewed initially by the original reviewer and the auditors.
2) Tc-99m activities as listed in the manifests were mistakenly noted (by the original shippers) in many instances to be Tc-99. The audit was performed before correction was applied to Tc-99. The present data is still considered to be overly conservative.
3) The difference of 16.3% for **I-129** is related to the difficulties encountered in identifying I-129 nuclide as listed in the original manifests. Most of these errors are associated with manifests during the early years of waste disposal (1965 – 1977). The audit of shipping records for 1981 shows the percentage differences to be much smaller.

In the **second case**, the percentage of error is determined by comparing the number of entries reviewed with the number of entries requiring correction. These are shown below:

Auditor # 1

Manifests selected for audit: ***(1) #00001- #03472; (2) # 09577 - # 09920***
Total number of manifests audited = ***3816*** *manifests*
Total number of entries QA reviewed = ***45,792*** *entries*
*Number of entries requiring correction =****157***
Percentage requiring correction *= (157/45,792) 100 =* ***0.34 %***

Auditor # 2

Manifests selected for audit: **(1) # 08584 - 08699; (2) #08841 - #09200**
Total number of manifests selected for audit = **136 manifests**
Total number of entries QA reviewed = 1632 entries
Number of entries requiring correction = 13
Percentage requiring correction = (13/1632) 100 = **0.79 %**

Out of a total of **9,915 manifests** reviewed by WDOH staff, a total of **3,952 manifests** were internally audited **(40 %).** Most of the error related to **Cl-36** that had escaped documentation during the initial stages of our review. It should be pointed out that **Cl-36** along with **I-129,** was not part of the list of radionuclides originally ascribed for this study; these radionuclides were added to the list after the WDOH review had been in progress for number of months.

TABLE 4

RESULTS OF US ECOLOGY'S RADIOACTIVE WASTE SHIPMENTS (1965-1981)

YEAR	U-235 (kg)	U-238 (kg)	DEP. U (kg)	NAT. U (kg)	TC-99 (mCi)	TC-99m (mCi)	CL-36 (mCi)	I-129 (mCi)
YEARS 1965-1981								
1965	0.002 (0.0044 mCi)	0.80 (0.272 mCi)	--	--	3.25	--	--	2.50
1966	0.243 (0.535 mCi)	187.1 (63.614 mCi)	0.72 (0.360 mCi)	--	--	0.100	--	--
1967	0.0	1.02 (0.35 mCi)	--	--	5.00	--	0.050	2.50
1968	0.00024 (0.00053 mCi)	--	0.127 (0.064 mCi)	--	0.110	--	5.50	--
1969	0.0167 (0.0368 mCi)	17.895 (6.08 mCi)	0.0045 (0.0023 mCi)	--	--	--	1.00	--
1970	0.1504 (0.331 mCi)	0.192 (0.065 mCi)	26.27 (13.14 mCi)	--	1.70	--	0.786	--
1971	0.0025 (0.006 mCi)	171.182 (58.20 mCi)	133.68 (66.84 mCi)	38.77 (27.53 mCi)	--	--	101.99	--
1972	0.5576 (1.227 mCi)	2,464.090 (837.7 mCi)	<0.045 <(0.023 mCi)	--	44.60	213.69	6.82	--
1973	8.316 (18.3 mCi)	1,429.26 (485.95 mCi)	778.6 (389.2 mCi)	1.982 (1.41 mCi)	11.52	--	33.41	--
1974	4.9258 (10.837 mCi)	20.960 (7.126 mCi)	0.035 (0.0175 mCi)	1,174.33 (833.75 mCi)	0.60	--	778.38	--
1975	7.472 (16.44 mCi)	6,790.2 (2,308.7 mCi)	11.295 (5.649 mCi)	--	2.00	--	0.967	--
1976	25.90 (56.98 mCi)	137,169.1 (46,637.50 mCi)	167.450 (83.725 mCi)	0.0002 (0.00014 mCi)	7.50	15.00	0.030	--

YEAR	U-235 (kg)	U-238 (kg)	DEP. U (kg)	NAT. U (kg)	TC-99 (mCi)	TC-99m (mCi)	CL-36 (mCi)	I-129 (mCi)
1977	24.746 (54.44 mCi)	95,635.0 (32,516.00 mCi)	782.24 (391.0 mCi)	4.05 (2.88 mCi)	173.50	1,215.90	0.008	0.22
1978	18.52 (40.744 mCi)	34,712.3 (11,802.18 mCi)	6,911.4 (3,455.7 mCi)	13.06 (9.28 mCi)	305.13	3,697.87	2.443	0.033
1979	18.09 (39.798 mCi)	3,992.3 (1,357.38 mCi)	12,950.5 (6,475.25 mCi)	1,356.8 (963.4 mCi)	2,470.64	10,799.21	45.12	--
1980	0.002 (0.0044 mCi)	114,230.0 (38,838.20 mCi)	6,008.2 (3,004.1 mCi)	897.85 (637.47 mCi)	9,277.16	93,009.94	96.23	31.42
1981	0.0022 (0.0048 mCi)	1,018,393.8 (346,250 mCi)	93,733.96 (46,867.0 mCi)	8,654.86 (6,145.0 mCi)	455.09	13,989.13	83.935	4.82
TOTAL (1965-81)	**108.947 kg** (0.240 Ci)	1,425,215.20 kg (484.57 Ci)	**121,504.48 kg** (60.75 Ci)	**12,141.70 kg** (8.62 Ci)	**12,757.80** (12.76 Ci)	**122,940.74** (122.94 Ci)	**1,156.69** (1.157 Ci)	**41.49 mCi** (0.042 Ci)

1. An audit of US Ecology's waste shipments (1965-1981) was performed by DOH. It includes results for U-235, U-238, depleted and natural uranium, TC-99, TC-99m, CL-36, and I-129.

2. Mass (gm, kg) of uranium isotopes is obtained directly from the US Ecology shipping records (manifests). Activities (millicuries) for each of these isotopes shown in parentheses are obtained by converting mass into activity, using the specific-activity relationship provided in WAC 246-231-200. Activities of uranium nuclides as documented on the manifests are not considered to be correct, because of the inconsistency in the way they were recorded.

3. Activities of Tc-99 as shown in the above table are considered to be overly conservative, since a large number of data entries for Tc-99m in the original shipping records are believed to be incorrectly assigned to Tc-99.

4. In many instances, Cl-36 activities include activities from mixed fission products (MFP); hence the reported activities for Cl-36 are considered conservative.

5. Mass of U-isotopes (kg) is converted to millicuries using the specific-activity relationship given in WAC 246-231-200. The conversion factors are shown below:

U-235 = 2.2 E-6 Ci/gm U-238 = 3.4 E-7 Ci/gm Nat. U = 7.1 E-7 Ci/gm Dep. U = 5.0 E-7 Ci/gm

RESULTS

Table 1 identifies the nuclides considered for this review. **Table 2** provides data that compares the uranium mass and activity as listed in the manifests, with the uranium mass-activity relationship estimated using the specific activity based on WAC 246-231-200. This comparison reflects the inconsistency in the uranium mass to activity data documented in the manifests.

Results of the US Ecology's radioactive waste shipment records (**June 1965 through the end of 1981)** reviewed by WDOH is presented in **Table 4**. This table shows the **mass** (kg) of uranium nuclides (U-235, U-238, U-depleted, U-natural) obtained from the US Ecology's manifests for each year, as well as the total for these years. Since the activities of uranium nuclides as listed in these manifests (also documented in Appendices A and B) were determined to be inaccurate, the **activities** (mCi) of uranium nuclides (shown in parenthesis in **Table 4**) related to their respective mass are the **calculated values** based on **specific activity data** provided in **WAC 246-231-200**.

The activities of **Tc-99** presented in **Table 4**, are obtained directly from the manifests. The **accuracy** of these numbers has been the subject of much discussion with the US Ecology staff, since a large number of **Tc-99m** nuclides were mistakenly documented as **Tc-99** in the original manifests. Based on experience, **appropriate corrections** were applied to a limited number of **Tc-99** nuclides. However, due to time constrain, and a lack of definitive information concerning the source of their origin, a decision was reached to limit further effort in this regard. The Tc-99 activities as presented in Table 4 are therefore considered to be **overly conservative.**

Cl-36 and I-129 activities as shown in Table 4 are obtained directly from those documented in the manifests. Most of the manifested **I-129 activity** is based on the lower level of detection rather than actual detector activity. Studies have shown that this results in overestimate of I-129. However, the information required to further evaluate I-129 is not available, and any additional analysis is beyond the scope of this study.

CONCLUSION

Results of the US Ecology radioactive waste shipment records for the years **1965 through 1981** reviewed by WDOH is presented in **Table 4**. It includes data for **U-235, U-238, depleted U, natural U, Tc-99, Tc-99m, Cl-36 and I-129**. Selection of these nuclides is based on their long half-life, along with their potential to contribute to the long-term individual and population dose resulting from ground water pathways and food chain factors.

The **mass** (kg) of uranium nuclides listed in Table 4, is obtained directly from the manifests and has been determined to be more accurate than their corresponding activities documented in the original manifests. The **activities** (mCi) of uranium nuclides (shown in parenthesis in Table 4) are estimated from the mass (kg) which are converted to their corresponding activities using **specific activity data** provided in **WAC 246-231-200** (Ref. 2)

The **Tc-99 activities** as presented in Table 4 are considered to be **overly conservative,** since a large number of packages containing **Tc-99m** nuclides were mistakenly documented as **Tc-99** in the original manifests. Efforts to modify these data based on knowledge of generator's waste stream was

limited due to time constrain, and a lack of definitive information concerning the source of their origin.

Cl-36 and I-129 activities as shown in Table 4 are obtained directly from those documented in the manifests. Most of the manifested **I-129 activity** is based on the lower level of detection rather than actual detector activity. Studies have shown that this results in overestimate of I-129. However, the information required to further evaluate I-129 is not available, and any additional analysis is beyond the scope of this study (Ref.4).

Data for **plutonium nuclides** is not included in Table 4 because of the large degree of uncertainty involved in identifying its various isotopes in the original manifests. A more comprehensive study of plutonium nuclides by US Ecology (Ref. 5) that includes actual isotopic percentage for plutonium provided by the generators, is given preference over other data.

The internal audit by the Waste Management staff (WDOH), which covered approximately **40%** of the reviewed material, concluded that data related to **uranium nuclides** was in close proximity with those reviewed by the original reviewer (Table 3). **Cl-36** (8.92%) and **I-129** (16.3%) had somewhat higher percentage of differences, largely due to the difficulties associated with their documentation. Percentage difference, based on the number of entries reviewed, showed less than 0.8%.

REFERENCES

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