Assessment of Fission Product Content of High-level Liquid Waste Supernate on E-Area Vault Package Criteria (U)

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INTRODUCTION

This report assesses the tank farm's high level waste supernate to determine any potential impacts on waste certification for the E-Area vaults (EAV). The Waste Acceptance Criteria procedure (i.e., WAC 3.10 of the 1S manual)³ imposes administrative controls on radioactive material in waste packages sent to the EAV, specifically on six fission products. Waste tank supernates contain various fission products, so any waste package containing material contaminated with supernate will contain these radioactive isotopes.

This report develops the process knowledge basis for characterizing the supernate composition for these isotopes, so that appropriate controls can be implemented to ensure that the EAV WAC is met.

SUMMARY

The characterization of fission products in High Level Waste supernate is based on process knowledge of reactor fission yield, the subsequent impacts of process chemistry, and the available sample data.

Both the fission yield calculations and the available sample data confirm that Se-79, Sr-90, and Sn-126 in supernate contaminated waste will be quite minor relative to the Package Administrative Criteria, so these isotopes will not need to be listed on the waste package manifests. The waste package manifests will have to list Tc-99, I-129 and Cs-137. The "supernate contaminated" waste packages will be limited by their I-129 content, so the package's Cs-137 content (i.e., their allowable dose rate) will have to be reduced ~20%.

The composition of waste streams related to In-Tank Processing will be documented separately. Both the In-Tank Precipitation (ITP) and Extended Sludge Processing (ESP) operations result in process streams where the Cs-137 has been concentrated or removed. Additionally, batches of spent washwater from the ESP operation will have a declining Cs-137 content, while other radionuclides could potentially maintain their original concentrations (i.e., the Cs-137 will be diluted as the sludge washing proceeds, while other isotopes which were partially insoluble could re-dissolve from the sludge). Thus these process streams, and potentially their associated solid waste streams, will have different isotopic ratios than the tank farm's historical supernate (i.e., which results from waste receipt and concentration/evaporation operations).

DISCUSSION

Fission Products of Interest

In a nuclear reactor, a wide distribution of "fission product" (FP) isotopes results from fission of the uranium and plutonium atoms. Each FP atom is a fragment of the original U/Pu atom, and is either a non-radioactive "stable" isotope, or else it is the radioactive "parent" of a "daughter" atom. Then, the daughter atom can either be stable, or it can be the parent of yet another generation of daughter. For each FP isotope, its atoms can be present from both mechanisms -- some of its atoms may have been formed directly, and while others may be the decay daughters which originated as some other FP isotope.

Six fission products are listed in the SRS "1S Manual" Waste Acceptance Criteria 3.10 (WAC)³:

- selenium, Se-79, which decays to bromine¹:
As-79 (half life of 9 minutes) ---> Se-79 (65,000 years) ---> Br-79
- strontium, Sr-90, which decays to niobium¹:
Kr-90 (half life of 32.3 seconds) ---> Rb-90^m (258 seconds) & Rb-90 (157 seconds) --->
Sr-90 (28.6 years) ---> Y-90 (64.1 hours) ---> Zr-90 (14.6 hours) ---> Nb-90
- technetium, Tc-99, which decays to ruthenium¹:
Mo-99 (half life of 66.0 hours) ---> Tc-99^m (6.0 hours) & Tc-99 (213,000 years) ---> Ru-99

- tin, Sn-126, which decays to tellurium¹:
 Sn-126 (half life of 100,000 years) → Sb-126^m (19 minutes) & Sb-126 (12.4 days) → Te-126
- iodine, I-129, which decays to xenon¹:
 Sb-129 (half life of 4.4 hours) → Te-129^m (33.6 days) & Te-129 (69.6 minutes) →
 I-129 (15,700,000 years) → Xe-129
- cesium, Cs-137, which decays to barium¹:
 Xe-137 (half life of 3.8 minutes) → Cs-137 (30.17 years) → Ba-137^m (2.52 min) → Ba-137

Characterizing these FP's in HLW supernate involves process knowledge about their production (in the nuclear reactors) and their subsequent behavior (as impacted by process chemistry).

Fission Products Formed in SRS Reactor Assemblies

In a nuclear reactor, the amount of each FP isotope produced depends primarily on two factors. First is the fissionable atom isotope distribution (e.g., the % of U-233, -235, -238; Np-237; Pu-238, -239, etc). This is largely synonymous with various fuel/target assemblies (e.g., Mark-16, -22, -31A, -31B, -53A, offsite fuel). The second factor that determines FP production is the reactor operating conditions (e.g., flux rate, megawatt-days, etc). Table 1 shows the amount of each of the six FPs generated in each type of assembly (i.e., Ci/assembly), as determined by reactor production-depletion calculations for the DWPF design basis². The different assemblies contain either enriched U-235 (the Mark-16's and a part of the Mark-22 assembly, which is the reactor fuel), or depleted U-238 (the Mark-31A's and -31B's, which produce Pu-239), or Np-237 (the Mark-53A's, which produce Pu-238). The conclusions drawn from Table 1 are:

- 1) For each isotope, the range between the lowest and highest amount of curies varies widely between the various assemblies (i.e., by factors of 19x-145x). However, this is largely due to differences in the total amount of FP isotopes in various assemblies. Thus the apparent variability is greatly reduced by tracking the isotope ratios, e.g., the "(Ci of isotope FP_x) / (Ci of Cs-137)" -- these vary over a relatively narrow range (i.e., <3x). For each isotope, a wide variability is seen in the "Ci/assembly" rows, compared to the lesser variations in the "Ci/Ci ratio" rows.

(Note: In this document, the term "Ci/Ci ratio" will always mean the ratio of that FP to Cs-137.)

The consistent Ci/Ci ratios mean that the exact origin of a given waste is not highly significant. That is, a waste container contaminated with HM supernate (from Mark-16's, -22's, and -53A's processed in H-area) will be similar to a container contaminated with PUREX supernate (from Mark-31A's and -31B's processed in F-area). Therefore, the "supernate contaminated" waste stream does not need to be subdivided and tracked individually. The next two sections of this report deal with the "average" waste composition, and then the variability will be addressed in the "maximum" waste composition section.

- 2) The ~60 year span of SRS's operation (from 1953 until DWPF completes operation ~2015) is about twice the ~30 year half live of Sr-90 and Cs-137. Therefore (holding other variables constant) the age of a particular waste can vary the isotope Ci/Ci ratios for the long lived FPs by a factor of 2-4. Comparing the "5 year" rows (in Table 1) with the "15 year" rows shows this over a shorter time.

Reactor Assembly Fission Yield Isotope Distribution

The relative quantity² of each type of assembly processed through the Separations canyons and the subsequent waste volumes generated (shown in Table 1) can be used to calculate the average composition of HLW waste (shown below) that results from processing these assemblies. The IS manual's Waste Acceptance Criteria³ (WAC) specifies the amount of each isotope allowed in a waste package -- these

values are called the Package Administrative Criteria (PAC). For comparison with the PAC, assume that a package contains enough "average assembly" waste to have reached the PAC for Cs-137 (i.e., 0.266 gal):

	15 year aged "avg assembly" waste composition <u>Ci / gal</u>	hypothetical package containing 0.266 gal of "avg assembly" waste <u>Ci / package</u>	WAC 3.10 PAC values <u>Ci / package</u>	ratio of package / PAC	waste volume req'd for each FP to equal its PAC <u>gal</u>
Se-79	0.000 0161	0.000 004 29	0.000 52	0.008 24	32.3
Sr-90	2.68	0.713	1.1	0.649	0.411
Tc-99	0.000 553	0.000 147	0.000 17	0.867	0.307
Sn-126	0.000 001 71	0.000 000 456	0.000 29	0.001 57	169.
I-129	0.000 000 942	0.000 000 251	0.000 000 083	3.02	0.088
Cs-137	2.85	0.76	0.76	1.	0.266

These supernate compositions and volumes are for fresh waste (i.e., as received from Separations). After the subsequent volume reduction in the Tank Farm evaporators, the waste concentrations are several fold higher, so the volume required to equal the PAC would be proportionately lower. For example, Cs-137 in the tank farm's average supernate is 5.05 Ci/gal (126.9 million Ci¹⁷, in 25.1 million gallons^{17a}), with a maximum of 44 Ci/gal¹⁵.

A useful way to compare the waste stream's composition to the PAC values involves the isotope ratios, i.e., Ci of FP_x per Ci of Cs-137. If the waste stream's Ci/Ci ratios are all less than the PAC's Ci/Ci ratios, then Cs-137 is the limiting isotope. For any isotope, if the waste stream's Ci/Ci ratio is higher than the PAC's Ci/Ci ratio, then the allowable Cs-137 content would be reduced proportionately, so the package's content of each isotope remains within its PAC value. Comparing the "average assembly" Ci/Ci ratios with the PAC Ci/Ci ratios:

	"average assembly" curie ratios <u>Ci / Ci Cs-137</u>	PAC curie ratios <u>Ci / Ci Cs-137</u>	ratio of "avg assembly" ratio to the PAC ratio	available margin factor
Se-79 / Cs-137	0.000 005 64	0.000 684	0.008 24	121.
Sr-90 / Cs-137	0.939	1.45	0.649	1.54
Tc-99 / Cs-137	0.000 194	0.000 224	0.867	1.15
Sn-126 / Cs-137	0.000 000 600	0.000 382	0.001 57	636.
I-129 / Cs-137	0.000 000 330	0.000 000 109	3.02	0.331
Cs-137 / Cs-137	1.	1.	1.	1.

These comparisons lead to these conclusions for packages contaminated with "average assembly" waste:

- 1) When the package reaches the PAC for Cs-137, it will only contain trace amounts of Se-79 and Sn-126 (only 0.82% and 0.16% of their PAC values respectively). A waste package would have to contain a large volume of supernate to exceed the PAC values (i.e., 32 and 169 gal respectively), and by that time the package would have already exceeded the Cs-137 PAC value by 121x-636x. This shows that these two isotopes are of no concern.
- 2) When the package reaches the PAC for Cs-137, then Sr-90 and Tc-99 will still meet their PAC, but not by large margins.
- 3) When the package reaches the PAC for Cs-137, the I-129 will already have exceeded its PAC by 200%. In order for the waste package to meet I-129's PAC value, it's Cs-137 content would have to be limited to 0.331 PAC.

The Ci/Ci ratio concept also parallels the tank farm's waste compliance program: i.e., to measure a waste package's external radiation dose rate, determine the Cs-137 content, and calculate the remaining isotope's contents by their predetermined "scaling factors" (i.e., the Ci/Ci composition ratios). If a waste stream's isotope ratios exceed the PAC ratios (i.e., when the "ratio of ratios" exceeds 1), then the allowable

radiation rate must be reduced until the Cs-137 content is low enough that the package's calculated inventory for all the other FPs will each meet their PAC values.

Alternative Fission Yield Isotope Distribution

Table 1 is based on Chandler's 1980 reactor fission product yield calculations² (done for the DWPF design basis), and this subject has recently been revisited by Webb^{2a} (for the SRS waste certification program). Comparing these calculations shows that both obtained essentially the same results (they used the same computer codes). Webb added one old style assembly (i.e., Mark-5E) and omitted two others (i.e., Mark-31B, -53A). Webb showed several decay times, where his case of 20 years is most comparable to Chandler's 15 years. During those additional 5 years, a noticeable portion of the Sr-90 and Cs-137 would decay (they each have half lives of ~30 years), so Webb's Ci/Ci ratios are generally slightly higher than Chandler's. Webb's calculation supports the conclusion that fission yield does not produce sufficient Se-79 and Sn-126 to limit the contents of waste packages. As stated in Webb's report^{2a}:

"These (Ci/Ci) relationships are true only if, subsequent to production, there have been no physical or chemical processes which preferentially separate one element relative to another. It is valid however, to combine data on chemical/physical separation processes with production data in order to justify a ratio different from those reported here, which are based solely on production data".

The next section develops the chemical/physical justification for reducing the Sr-90, Tc-99, and I-129 activity levels estimated for actual waste tank supernate.

Average Waste Tank Supernate

If all the FP atoms in the fuel/target assemblies went to the waste tanks and stayed in the supernate, then "average assembly" Ci/Ci isotope ratios would actually apply to "supernate contaminated" waste. In reality however, there are two "partition factors" which impact the supernate's Ci/Ci ratios: not all the FPs go the waste tanks, and not all the FPs stay in the supernate:

- 1) When Separations dissolves the assemblies in nitric acid, part of the iodine is volatilized into the dissolver off-gas system. Part of that volatile I-129 is collected as silver iodide (on silver coated Beryl saddles). Originally, the silver iodide coating was dissolved and flushed to the waste tanks; later the coated rings either went to Solid Waste or remain in Separations¹⁰. In the PUREX process (i.e., in F-area, and in H-area before 1960), an estimated 15-20% of the I-129 went to the waste tanks; compared to the 95-98% estimated for the HM process (i.e., for H-area since 1960)¹⁰.

Thus there are three routes whereby the waste tanks do not contain all the I-129 that was originally produced in the assemblies: venting to atmosphere, disposal, and storage. Of the 39 Ci of I-129 produced through 1974, 60% is estimated to have not gone to the waste tanks¹⁰. These "losses" reduce the I-129/Cs-137 ratio for "supernate contaminated" waste to 40% of the average assembly Ci/Ci ratio value (which was shown in Table 1).

In the waste tank's caustic environment, the I-129 will be present as non-volatile soluble salts (i.e., sodium iodide, and sodium iodate)^{10,12}. Chemically, the behavior of iodine will be analogous to chlorine. In acid solutions, chlorine is the semi-volatile hydrochloric acid (HCl); but when neutralized, it becomes sodium chloride (i.e., table salt, NaCl). Likewise, in the acidic Separations process, iodine is a semi-volatile molecule (HI); and then it becomes nonvolatile (NaI) in the waste tanks.

If iodine were volatile in caustic solutions, then the tank farm evaporators would have either sent the iodine to the Effluent Treatment Facility (if it condensed in the overheads stream), or it would have been vented to atmosphere (if it didn't condense). The available sample data (discussed later) indicates

that iodine "concentrates" in the evaporators (like cesium). This confirms that the iodine species present in the waste tanks is not volatile.

Aside from iodine, essentially 100% of the other FP isotopes in the assemblies do go to the waste tanks. Only trace amounts are "lost" to the environment or to Separation's (or Reactor's) solid waste streams.

- Many other isotopes are only slightly soluble in the waste tank's highly caustic environment, so a large fraction of those curies ends up in the sludge layer. Depending on the FP isotope's chemical behavior, the supernate may only contain part of the original activity. The portion can range from essentially everything (e.g., I-129, Cs-137 = 100%), to a moderate amount (e.g., Tc-99 = 57%, Se-79 = 10%, Sn-126 = 4%), or a very small fraction (e.g., Sr-90 = 0.025%).

The supernate portions (i.e., "partition factors") were determined empirically, from sample analyses of supernate samples taken during development of the DWPF^{5,8}. Thus the "supernate fraction" includes both the true chemical solubility and any "physical" effects such as suspended colloidal particulate.

Accounting for these two factors, the Ci/Ci ratios of "average supernate" waste are different than those of "average assembly" waste:

	"average assembly" Ci/Ci Cs-137	partition fractions to waste&supernate	"average supernate" Ci/Ci Cs-137	PAC ratios Ci/Ci Cs-137	ratio of the "avg supern" ratio to the PAC ratio	available margin factor
Se-79 / Cs-137	5.64E-06	1. & 0.1	5.64E-07	6.84E-04	0.000 824	1,510.
Sr-90 / Cs-137	9.39E-01	1. & 0.00025	2.35E-04	1.45	0.000 162	6,170.
Tc-99 / Cs-137	1.94E-04	1. & 0.57	1.10E-04	2.24E-04	0.493	2.02
Sn-126 / Cs-137	6.00E-07	1. & 0.04	2.40E-08	3.82E-04	0.000 0629	15,900.
I-129 / Cs-137	3.30E-07	0.4 & 1.	1.32E-07	1.09E-07	1.21	0.827
Cs-137 / Cs-137	1.	1. & 1.	1.	1.	1.	1.

Accounting for these two factors leads to several conclusions:

- The two isotopes which were un-important (Se-79 and Sn-126) become even less significant.
- One of the previously marginal isotopes (Sr-90) becomes totally un-important.
- The other previously marginal isotope (Tc-99) remains marginal.
- The isotope which previously appeared to be the actual limit (I-129) remains so, although to a somewhat lesser extent. To keep the package I-129 content within its PAC criteria, the Cs-137 content would have to be limited to 82% of its PAC.

Maximum Waste Tank Supernate

As was noted previously, for a given FP isotope, variations in the Ci/Ci ratios between types of assemblies is relatively small, and those variations do not materially change the above conclusions. This is demonstrated by hypothesizing a "maximum" supernate which contains each FP at the highest Ci/Ci ratio produced in any type of assembly (i.e., the "max" column shown in Table 1):

	"maximum assembly" Ci/Ci Cs-137	partition fractions to waste&supernate	"maximum supernate" Ci/Ci Cs-137	PAC ratios Ci/Ci Cs-137	ratio of the "max supern" ratio to the PAC ratio	available margin factor
Se-79 / Cs-137	5.81E-06	1. & 0.1	5.81E-07	6.84E-04	0.000 849	1,180.
Sr-90 / Cs-137	9.74E-01	1. & 0.00025	2.43E-04	1.45	0.000 168	5,940.
Tc-99 / Cs-137	2.00E-04	1. & 0.57	1.14E-04	2.24E-04	0.510	1.96
Sn-126 / Cs-137	1.30E-06	1. & 0.04	5.21E-08	3.82E-04	0.000 136	7,330.
I-129 / Cs-137	5.86E-07	0.4 & 1.	2.35E-07	1.09E-07	2.15	0.466
Cs-137 / Cs-137	1.	1. & 1.	1.	1.	1.	1.

Comparing these "maximum supernate" Ci/Ci ratios with the PAC ratios shows that:

- 1) Se-79, Sr-90, and Tc-99 are essentially un-affected (their "maximum assembly" Ci/Ci ratios are only ~3% higher than the "average assembly" Ci/Ci ratios).
- 2) Sn-126 could increase by 121% ($\text{max}=2.21 \times \text{avg}$), so the available margin before Sn-126 becomes the limiting isotope would "only" be 7,330x instead of 15,900x.
- 3) I-129 could increase by 78% ($\text{max}=1.78 \times \text{avg}$), which would raise the "supernate/PAC" ratio from 1.21 to 2.15. Covering this possibility would require limiting the waste packages to 46% of the Cs-137 criteria, instead of 82%.

The WAC says that "on average, each package from a given waste stream will meet the (PAC)". The interpretation of the term "on average" will determine whether the 1.78 factor needs to be applied. Over a long enough time period, the "average waste package" could be presumed to contain "average supernate". But over a sufficiently short time span, there is a possibility that disproportionate amounts of waste (e.g., resulting from non-uniform work loads) could be generated from tanks with high ratios of I-129/Cs-137. The PAC could be exceeded over that particular time span.

Inventory, Partition Factor, and Sample Data

The previous section developed the isotope Ci/Ci ratios expected for supernate contaminated waste, as based on the reactor's calculated production rates for the various fission products, and the subsequent impacts of process chemistry. This section compares those expectations to available sample data for these isotopes in actual waste tank supernate:

Se-79

Tank 50 contains decontaminated salt solution which was produced during the 1983 demonstration of the ITP process. One sample (taken 6/1/89) was analyzed⁴ for Se-79. The result (0.185 nCi/gm of salt) corresponds to $8.4\text{E-}7$ Ci/gal (for a solution having a 1.2 density), so the waste package would have to contain 615 gallons before it could exceed the $5.2\text{E-}4$ Ci PAC value. Note that the ratio of Se-79/Cs-137 in tank 50 is not indicative of supernate contaminated waste, since the ITP process had chemically removed (i.e., precipitated) almost all of the Cs-137 activity.

Table 4 shows several analyses obtained during previous waste characterization efforts (mostly related to developing the DWPF flowsheet) and during preparation of feed solutions for the 1982 demo of the ITP process (the tank 24 samples).

These analyses do not contradict the previous conclusion that Se-79 will not be a limiting isotope.

Sr-90

The total waste tank inventory of Sr-90 is estimated¹⁷ to be 122,000,000 Ci. Only a small fraction (0.00025) of the Sr-90 is expected⁵ to partition into the supernate (30,500 Ci), and the remainder will be in the sludge. This implies that the 25.1 million gallons of supernate^{17a} will average 0.00121 Ci/gal, so a waste package would have to contain 905 gal to exceed the 1.1 Ci PAC value.

Special supernate samples⁶ were taken from 9 waste tanks in July and November 1992. As shown in Table 2, the Sr-90 concentration averaged $1.76\text{E}6$ d/m/ml (0.0030 Ci/gal). At this concentration, a waste package would have to contain 363 gallons of supernate before it exceeds the 1.1 Ci PAC. Of the 9 samples, the average Sr-90/Cs-137 ratio is 0.00057 Ci/Ci. This is slightly higher (2.5x) than the "fission yield and partition factor" ratio of 0.00023 Ci/Ci, but is still well below the 1.45 Ci/Ci ratio where Cs-137 would cease to be the limiting isotope.

Waste tank supernate samples are taken routinely as part of the tank corrosion control program, and 33 of these have been analyzed¹⁵ for both Sr-90 and Cs-137 (see Table 3). The min/avg/max values shown in Table 3 do not include one sample (i.e., Tank 23 in 10/71), where the unusually low Cs-137 content is obviously suspect. The remaining 32 samples averaged 0.00228 Ci/gal for Sr-90, so a waste package would have to contain 482 gal of this supernate before it would exceed the 1.1 Ci PAC. The average Sr-90/Cs-137 ratio for these samples (0.00103 Ci/Ci) is 4.4x higher than the "fission yield and partition factor" ratio (0.000235 Ci/Ci), but is still well below the ratio where Cs-137 would cease to be the limiting isotope (1.44 Ci/Ci).

Table 4 shows several analyses obtained during previous waste characterization efforts (mostly related to developing the DWPF flowsheet) and during preparation of feed solutions for the 1982 demo of the ITP process (the tank 24 samples). These results indicate that Sr-90 is well below the PAC.

These analyses do not contradict the previous conclusion that Sr-90 will not be a limiting isotope.

Tc-99

As shown in Table 4, twelve supernate samples⁸ ranged from 0.00015 to 0.0010 Ci/gallon and averaged 0.00036 Ci/gal (normalized to a supernate containing 400 gm salt/liter), so a waste package would have to contain 0.47 gal to exceed the 0.00017 Ci PAC value.

Supernate samples⁶ were taken from 9 waste tanks in July and November 1992. As shown in Table 2, the Tc-99 concentration averaged $4.9\text{E}5$ d/m/ml (0.00084 Ci/gal). At this concentration, a waste package would have to contain 0.20 gallons of supernate before it would exceed the WAC value of 0.00017 Ci. Of these 9 samples, the average Tc-99/Cs-137 ratio is 0.00016 Ci/Ci. This agrees well with the "fission yield and partition factor" ratio of 0.00011 Ci/Ci, and also meets the 0.000224 Ci/Ci ratio which allows Cs-137 to be the limiting isotope.

Saltcake samples¹⁶ taken from two locations in tank 41 were found to contain $4.0\text{E}-4$ & $1.75\text{E}-4$ Ci of Cs-137/gm of salt, and 1.2 & $3.8\text{E}-8$ Ci of Tc-99/gm of salt. This 0.000263 Ci/Ci ratio slightly exceeds the 0.000224 ratio that would allow Cs-137 to be the limiting isotope.

Waste tank supernate samples are taken routinely as part of the tank corrosion control program, and 21 of these have been analyzed¹⁵ for both Tc-99 and Cs-137 (see Table 3). Note that the min/avg/max values shown in Table 3 do not include the 36 samples of tank 50H -- because 1) its Cs-137 was chemically removed by the ITP demo in 1983 (thus changing its Ci/Ci ratios) and 2) the contents of tank 50 have been dormant, so the repeated samples would skew the average. The min/avg/max values in Table 3 also exclude one sample (i.e., tank 20 on 8/82), where the unusually low Cs-137 content is obviously suspect (note that a sample taken 6 days later had a normal Cs-137 content, although that sample wasn't analyzed for Tc-99). The remaining 20 analyses average 0.00049 Ci/gal for Tc-99, so a waste package would have to contain 0.346 gal of this supernate before it would exceed the 0.00017 Ci PAC value. The average Tc-99/Cs-137 ratio of 0.00019 Ci/Ci agrees well with the 0.000194 ratio determined by the "average assembly" fission yield (it is 1.64x higher than the 0.000116 ratio determined by accounting for the expected partition factor), and it also meets the 0.000224 Ci/Ci ratio that allows Cs-137 to be the limiting isotope. The Tc-99 abundance shown in Table 3 could be related to analytical difficulties, or it may reflect the impact of other types of fuel/target assemblies (or their operating history) which are not included in the reactor fission yield calculations^{2, 2a}.

These analyses confirm the previous conclusion that Tc-99 is expected to be present in supernate contaminated waste packages, and they agree reasonably well with the "fission yield and partition factor" isotope ratio of 0.00011 Ci/Ci.

Note: Ref 9 discusses Tc-99 from a site-wide perspective, and ref 8 details the tank farm inventory and technetium's behavior/chemistry.

Sn-126

Supernate samples⁶ were taken from 9 waste tanks in July and November 1992. As shown in Table 2, the Sn-126 concentration averaged $4.93\text{E}3$ d/m/ml ($8.5\text{E}-6$ Ci/gal), so a waste package would have to contain 34.1 gal before it would exceed the $2.9\text{E}-4$ Ci PAC. The average Sn-126/Cs-137 ratio of $1.6\text{E}-6$ Ci/Ci:

- is 2.7x higher than the $6.0\text{E}-7$ ratio determined by the "average assembly" fission yield (and is 67x higher than the $2.4\text{E}-8$ ratio determined by accounting for the expected partition factor).
- is still well within the $3.81\text{E}-4$ Ci/Ci ratio which allows Cs-137 to be the limiting isotope.

The Sn-126 abundance shown in Table 2 could be related to analytical difficulties, or it may reflect the impact of other types of fuel/target assemblies (or their operating history) which are not included in the reactor fission yield calculations^{2,2a}.

These analyses do not contradict the previous conclusion that Sn-126 will not be a limiting isotope.

I-129

Dissolved salt and supernate from tank 24H was used in the full-scale demonstration of the In-Tank Precipitation process in 1983. That supernate was used to develop an analytical method (which involves Cs-137 precipitation, and ion exchange of the iodine) which found that the I-129 was $<1.7\text{E}-9$ Ci/gal (i.e., less than the 0.45 pCi/ml detection capability)¹¹.

Table 4 shows several waste tank analyses¹² for I-129. Compared to fresh supernate, the 3x higher I-129 content found in the concentrated supernate is directly related to the volume reduction (boildown) achieved by the tank farm evaporators. The low values found in the salt samples are also indicative of the high solubility of iodine (i.e., its reluctance to crystallize into the salt phase). A waste package would only have to contain 0.075 gal of the $1.1\text{E}-6$ Ci/gal supernate to exceed the $8.3\text{E}-8$ Ci PAC value. As expected based on a 3x concentration factor in the evaporators, this is $\sim 1/3$ of the 0.22 gal value determined by "fission yield and partition factor".

The total waste tank inventory of I-129 has been estimated¹² to be 9.1 Ci. This is comparable to the ~ 10 Ci introduced to the atmosphere by nuclear weapons testing, or to the environment's ~ 40 Ci natural inventory¹³. Note: Ref 13 discusses I-129 from a site-wide perspective, and ref 12 details the tank farm inventory and iodine's behavior/chemistry.

These sample results confirm the previous conclusion that I-129 is expected to be present in supernate contaminated waste packages. Relative to its PAC value, I-129 will limit the packages to an extent similar to Cs-137.

Cs-137

The total waste tank inventory of Cs-137 is estimated¹⁷ to be $1.269\text{E}8$ Ci. Part of the Cs-137 is associated with interstitial supernate in the layers of sludge and saltcake, but most⁵ will be in the 25.1 million gallons of supernate^{17a}, which would thus average 5.05 Ci/gal.

Supernate samples⁶ were taken from 9 waste tanks in July and November 1992. As shown in Table 2, the Cs-137 concentration averaged $3.08\text{E}9$ d/m/ml (5.3 Ci/gal). At this concentration, a waste package would have to contain 0.14 gallons of supernate before it would exceed the WAC value of 0.76 Ci.

Waste tank supernate samples are taken routinely as part of the tank corrosion control program. Chart 1 shows the 470 analyses¹⁵ which average 2.66 Ci/gal (they range from 0.000 000 007 to 44. Ci/gal). At the average concentration, a waste package would have to contain 0.29 gal to exceed the 0.76 Ci PAC value.

Note: Ref 14 discussed Cs-137 from a site-wide perspective.

Future Conditions

The DWPF melter is expected to volatilize Cs-137 more than the other isotopes, so the DWPF recycle stream (which returns to the tank farm) will contain more Cs-137 compared to the other isotopes (i.e., the other isotope Ci/Ci ratios will be lower). Thus future supernates, which will consist partially of DWPF recycle, should have (slightly) lower Ci/Ci ratios than the values discussed in this document. Therefore it would be conservative to keep using the same dose rate scaling factors derived for the current supernate, as this will (slightly) over-estimate the amounts of other FP isotopes in the waste packages.

Over a sufficiently long time span, the amount of Cs-137 present will eventually decline (due to its 30 year half life) enough that the Ci/Ci ratios used for the other FP isotopes will have to be adjusted.

CONCLUSIONS

Both the fission yield calculations and the available sample data confirm that Se-79, Sr-90, and Sn-126 in supernate contaminated waste will be quite minor relative to the Package Administrative Criteria. When a waste package contains enough supernate to equal the Cs-137 PAC, it will contain less than 1/1000th of the PACs for these three isotopes, thus they will not need to be reported in the package manifest. This applies to both an "average" mix of fuel/target assemblies, and to a hypothetical "maximum" assembly (which contains the worst concentrations of each fission product relative to the amounts of Cs-137).

The Tc-99, I-129, and Cs-137 are all relatively soluble and thus tend to stay in the evaporator's concentrated supernate instead of settling into the sludge layer or crystallizing into the saltcake. Compared to the PAC values, I-129 will be slightly more limiting than Cs-137. This means that the allowable package dose rate should be reduced 20%, such that the package's I-129 content also meets the PAC. When a waste package contains enough supernate to equal the Cs-137 PAC, it will contain ~50% and 120% of the PAC for Tc-99 and I-129 (respectively). These three isotopes will have to be reported in the package manifest.

REFERENCES

1. The Se-79 decay chain is from "Chart of the Nuclides", G.E./Knolls Lab, 4/88. The others are from appendix B of "External Dose-Rate Conversion Factors for Calculation of Dose to the Public", July 1988, DOE/EH-0070 DE88 014691.
2. "Preliminary Technical Data Summary - DWPF - Stage 2", 12/80, DPSTD-80-39 (specifically, section 13.6 "Radionuclide Compositions", by J.R.Chandler). Note that later versions of this document omit this material (i.e., DPSTD-80-39-2).
- 2a. "Production Limits for Fission Product Ratios", R.L.Webb, 6/15/94, EPD-CTG-94-0006.
3. "SRS Waste Acceptance Criteria Manual", WSRC-1S (specifically the LAWV column in table 2 of procedure WAC 3.10, rev-1, 5/21/94).
4. Analytical results from TMA/Norcal, dated 7/15/89; attached to "Tank 50 Sample Results", J.E.Batton, 8/3/89, OPS-DPF-89-0036.
5. "Update of Chemical and Radiochemical Composition of Decontaminated Soluble Waste From the Precipitation Process", J.R.Fowler, 8/6/82, DPST-82-759.
6. "Composition of Tank Farm Supernate Samples", D.D.Walker/et.al., 7/16/93, WSRC-RP-93-1009.
7. "Assessment of Strontium in the S.R.S. Environment", W.H.Carlton/et.al., (no date), WSRC-RP-92-984.
8. "Technetium-99 in SRP High-Level Waste and Saltstone", J.R.Fowler/et.al., 2/24/84, DPST-84-334.
9. "Assessment of Technetium in the S.R.S. Environment", W.H.Carlton/et.al., 7/93, WSRC-TR-93-217.
10. "Estimation of ^{129}I Distribution on SRP Separations Processes", W.R.Cornman, 11/21/74, DPST-74-533.
11. "Analysis of Decontaminated Defense Waste Salt Supernate for I-129", J.P.Ryan, 2/23/93, DPST-83-325.
12. "I-129 in SRP High-Level Waste and Saltstone", J.R.Fowler and J.R.Cook, 2/29/84, DPST-84-343.
- 12a. Personal communications with J.R.Fowler 6/17/94: ref 12 incorrectly reports a value 10x lower for I-129 in tank 15H. The original (correct) data is in notebook DPSTN-3302 pages 69 and 82-83.
13. "Radioiodine in the S.R.S. Environment", M.V.Kantelo/et.al., 1/15/93, WSRC-RP-90-424-2.
14. "Cesium in the S.R.S. Environment", W.H.Carlton/et.al., 3/92, WSRC-RP-92-250.
15. "Radionuclide Sample Analysis of Waste Tanks 1-51", J.A.Pike, 1/24/94, HLW-HLE-94-0328.
16. "Final Report: Analysis of Tank 41H Saltcake Sample #1 and Comparison to Sample #1", D.T.Hobbs and C.J.Coleman, 1/26/94, WSRC-TR-94-057.
17. "High Level Waste Engineering Monthly Data Record - October 1993", R.A.Scaggs, 12/13/94, WSRC-RP-93-78-10B.
- 17a. The tank farm inventory¹⁷ is 34.01E6 gal total, consisting of 3.67E6 gal sludge, 14.09E6 gal saltcake, and 16.25E6 gal supernate (ranging from ballast water to highly concentrated). Allowing for 60% of the sludge volume to be interstitial supernate (and 10% of the saltcake), the total equivalent supernate volume is 25.1E6 gal.

Table 1
Fission Product Yields in Various Types of Assemblies

		<u>Mark-16B</u>	<u>Mark-22</u>	<u>Mark-31A</u>	<u>Mark-31B</u>	<u>Mark-53A</u>	<u>offsite</u> <u>fuel</u>	<u>scrap Pu</u> <u>-238 -239</u>		<u>min</u>	<u>max</u>	<u>ratio of</u> <u>max / min</u>	<u>weighted</u> <u>average</u>
assembly fraction		0.1443	0.1461	0.4241	0.1409	0.0136	0.1035	0.0029	0.0246	--	--	--	--
waste prod., gal/assembly		564.	376.	258.	175.	588.	808.	1500.	475.	--	--	--	--
waste fraction		0.2153	0.1453	0.2894	0.0652	0.0212	0.2213	0.0114	0.0309	--	--	--	--
	<u>years</u> <u>out of</u> <u>reactor</u>	<u>Ci /</u> <u>assembly</u>	<u>Ci /</u> <u>assembly</u>	<u>Ci /</u> <u>assembly</u>	<u>Ci /</u> <u>assembly</u>	<u>Ci /</u> <u>assembly</u>	<u>Ci /</u> <u>kg U-235</u>	<u>Ci /</u> <u>kg Pu</u>	<u>Ci /</u> <u>kg Pu</u>	<u>Ci /</u> <u>assembly</u>	<u>Ci /</u> <u>assembly</u>	<u>ratio</u>	<u>Ci /</u> <u>gal waste</u>
Se-79	5	1.48E-02	1.31E-02	1.75E-03	1.28E-03	2.42E-04	1.06E-02	0	0	2.42E-04	1.48E-02	61.2	1.61E-05
Sr-90	5	3.28E+03	2.89E+03	3.06E+02	2.31E+02	2.28E+01	2.28E+03	0	0	2.28E+01	3.28E+03	143.7	3.43E+00
Tc-99	5	5.08E-01	4.48E-01	6.09E-02	4.50E-02	9.64E-03	3.66E-01	0	0	9.64E-03	5.08E-01	52.7	5.53E-04
Sn-126	5	1.34E-03	1.21E-03	3.41E-04	2.27E-04	6.87E-05	9.64E-04	0	0	6.87E-05	1.34E-03	19.5	1.71E-06
I-129	5	8.12E-04	7.22E-04	1.37E-04	9.78E-05	3.10E-05	5.89E-04	0	0	3.10E-05	8.12E-04	26.2	9.42E-07
Cs-137	5	3.32E+03	2.93E+03	3.98E+02	2.95E+02	6.65E+01	2.30E+03	0	0	6.65E+01	3.32E+03	50.0	3.59E+00
Se-79	15	1.48E-02	1.31E-02	1.75E-03	1.28E-03	2.42E-04	1.06E-02	0	0	2.42E-04	1.48E-02	61.2	1.61E-05
Sr-90	15	2.57E+03	2.26E+03	2.39E+02	1.81E+02	1.79E+01	1.78E+03	0	0	1.79E+01	2.57E+03	143.7	2.68E+00
Tc-99	15	5.08E-01	4.48E-01	6.09E-02	4.50E-02	9.64E-03	3.66E-01	0	0	9.64E-03	5.08E-01	52.7	5.53E-04
Sn-126	15	1.34E-03	1.21E-03	3.41E-04	2.27E-04	6.87E-05	9.64E-04	0	0	6.87E-05	1.34E-03	19.6	1.71E-06
I-129	15	8.12E-04	7.22E-04	1.37E-04	9.78E-05	3.10E-05	5.89E-04	0	0	3.10E-05	8.12E-04	26.2	9.42E-07
Cs-137	15	2.64E+03	2.33E+03	3.16E+02	2.34E+02	5.28E+01	1.83E+03	0	0	5.28E+01	2.64E+03	50.0	2.85E+00
	<u>years</u> <u>out of</u> <u>reactor</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>Ci/Ci</u>	<u>ratio</u>	<u>Ci/Ci</u>
Se-79 / Cs-137	5	4.45E-06	4.48E-06	4.40E-06	4.34E-06	3.64E-06	4.61E-06	0	0	3.64E-06	4.61E-06	1.266	4.48E-06
Sr-90 / Cs-137	5	9.88E-01	9.87E-01	7.69E-01	7.84E-01	3.44E-01	9.90E-01	0	0	3.44E-01	9.90E-01	2.879	9.54E-01
Tc-99 / Cs-137	5	1.53E-04	1.53E-04	1.53E-04	1.53E-04	1.45E-04	1.59E-04	0	0	1.45E-04	1.59E-04	1.096	1.54E-04
Sn-126 / Cs-137	5	4.03E-07	4.12E-07	8.55E-07	7.70E-07	1.03E-06	4.18E-07	0	0	4.03E-07	1.03E-06	2.563	4.77E-07
I-129 / Cs-137	5	2.44E-07	2.47E-07	3.44E-07	3.32E-07	4.66E-07	2.56E-07	0	0	2.44E-07	4.66E-07	1.907	2.62E-07
Cs-137 / Cs-137	5	1.	1.	1.	1.	1.	1.	0	0	1.	1.	1.	1.
Se-79 / Cs-137	15	5.61E-06	5.64E-06	5.54E-06	5.46E-06	4.59E-06	5.81E-06	0	0	4.59E-06	5.81E-06	1.266	5.64E-06
Sr-90 / Cs-137	15	9.72E-01	9.72E-01	7.56E-01	7.71E-01	3.38E-01	9.74E-01	0	0	3.38E-01	9.74E-01	2.879	9.39E-01
Tc-99 / Cs-137	15	1.92E-04	1.92E-04	1.93E-04	1.92E-04	1.83E-04	2.00E-04	0	0	1.83E-04	2.00E-04	1.096	1.94E-04
Sn-126 / Cs-137	15	5.09E-07	5.19E-07	1.08E-06	9.70E-07	1.30E-06	5.26E-07	0	0	5.09E-07	1.30E-06	2.559	6.00E-07
I-129 / Cs-137	15	3.08E-07	3.10E-07	4.33E-07	4.17E-07	5.86E-07	3.22E-07	0	0	3.08E-07	5.86E-07	1.907	3.30E-07
Cs-137 / Cs-137	15	1.	1.	1.	1.	1.	1.	0	0	1.	1.	1.	1.

Note: data is from chapter 13.6 of "Preliminary Technical Data
Summary - DWPF - Stage 2", 12/80, DPSTD-80-39.

Table 2
Recent Supernate Characterization Samples

		<u>average</u>	<u>25F</u>	<u>26F</u>	<u>27F</u>	<u>28F</u>	<u>29H</u>	<u>30H</u>	<u>32H</u>	<u>38H</u>	<u>43H</u>
sample analysis:											
Sr-89/90	d/m/ml	1.76E+06	6.90E+04	9.10E+04	7.50E+04	7.20E+04	8.50E+05	1.30E+07	1.30E+06	2.30E+05	1.70E+05
Tc-99	d/m/ml	4.89E+05	3.70E+05	4.00E+05	3.10E+05	4.00E+05	1.15E+06	5.80E+05	4.30E+05	4.40E+05	3.20E+05
Sb-125	d/m/ml	1.23E+05	7.00E+02	7.00E+02	7.00E+02	4.00E+02	4.90E+04	1.20E+05	2.50E+04	5.00E+05	4.10E+05
Sn-126	d/m/ml	4.93E+03	5.70E+03	6.00E+03	4.90E+03	6.20E+03	1.00E+04	4.40E+03	3.90E+03	1.70E+03	1.60E+03
Cs-134	d/m/ml	5.54E+07	not detect.	1.00E+08	not detect.	7.40E+07	2.00E+07	1.20E+08	6.60E+07	4.90E+06	3.20E+06
Cs-137	d/m/ml	3.08E+09	2.50E+09	3.20E+09	2.20E+09	2.60E+09	6.50E+09	4.80E+09	2.20E+09	2.20E+09	1.50E+09
Sr-89/90 / avg	ratio	1.	0.04	0.05	0.04	0.04	0.48	7.38	0.74	0.13	0.10
Tc-99 / avg	ratio	1.	0.76	0.82	0.63	0.82	2.35	1.19	0.88	0.90	0.65
Sb-125 / avg	ratio	1.	0.01	0.01	0.01	0.00	0.40	0.98	0.20	4.07	3.33
Sn-126 / avg	ratio	1.	1.16	1.22	0.99	1.26	2.03	0.89	0.79	0.34	0.32
Cs-134 / avg	ratio	1.	not detect.	1.80	not detect.	1.33	0.36	2.16	1.19	0.09	0.06
Cs-137 / avg	ratio	1.	0.81	1.04	0.71	0.84	2.11	1.56	0.71	0.71	0.49
Sr-89/90 / Cs-137	Ci/Ci	5.72E-04	2.76E-05	2.84E-05	3.41E-05	2.77E-05	1.31E-04	2.71E-03	5.91E-04	1.05E-04	1.13E-04
Tc-99 / Cs-137	Ci/Ci	1.59E-04	1.48E-04	1.25E-04	1.41E-04	1.54E-04	1.77E-04	1.21E-04	1.95E-04	2.00E-04	2.13E-04
Sb-125 / Cs-137	Ci/Ci	3.99E-05	2.80E-07	2.19E-07	3.18E-07	1.54E-07	7.54E-06	2.50E-05	1.14E-05	2.27E-04	2.73E-04
Sn-126 / Cs-137	Ci/Ci	1.60E-06	2.28E-06	1.88E-06	2.23E-06	2.38E-06	1.54E-06	9.17E-07	1.77E-06	7.73E-07	1.07E-06
Cs-134 / Cs-137	Ci/Ci	1.80E-02	not detect.	3.13E-02	not detect.	2.85E-02	3.08E-03	2.50E-02	3.00E-02	2.23E-03	2.13E-03
Cs-137 / Cs-137	Ci/Ci	1.	1.	1.	1.	1.	1.	1.	1.	1.	1.

Note: data is from "Composition of Waste Tank Supernate Samples", D.D.Walker/et.al., 7/16/93, WSRC-RP-93-1009.

Table 3
Routine Supernate Samples

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Tank	Se-79	Sr-90	Tc-99	I-129	Cs-137	Se-79 / Cs-137	Sr-90 / Cs-137	Tc-99 / Cs-137	I-129 Cs-137	notes & references ...
mCi/L of solution						isotope ratios, Ci/Ci				per J.R.Fowler's DPSTN-3302:
13	4.5E-04	9.1E-03	4.0E-04	-	2.5E+00	1.8E-04	3.6E-03	1.6E-04	-	supernate sampled 6/30/81, p 17
47	6.4E-05	1.1E-02	2.9E-04	-	8.6E-02	7.4E-04	1.3E-01	3.4E-03	-	concentrated supernate sampled 7/13/81, p 17
19	1.6E-04	1.4E-01	1.2E-02	-	-	-	-	-	-	dissolved salt soln sampled 8/19/81 (in Tank 18), p 21
20	1.7E-03	-	3.2E-02	-	-	-	-	-	-	dissolved salt solution sampled 8/19/81, p 22-23
24	1.1E-02	not detect.	7.7E-02	-	4.4E+00	2.5E-03	not detect.	1.8E-02	-	dissolved salt solution sampled 8/26/81, p 24-25
24	3.2E-03	6.0E-03	4.3E-02	-	3.5E+02	9.2E-06	1.7E-05	1.2E-04	-	dissolved salt solution sampled 10/21/81, p 35
24	-	-	9.6E-02	-	3.5E-01	-	-	2.7E-01	-	dissolved salt solution sampled 11/25/81, p 47
24	2.0E-03	not detect.	5.9E-02	-	1.9E+02	1.1E-05	not detect.	3.2E-04	-	dissolved salt solution sampled 6/17/82, p 63
mCi/L of salt cake										per J.R.Fowler's DPSTN-3302:
1	3.2E-02	4.3E+00	2.5E-02	-	1.0E+03	3.1E-05	4.2E-03	2.4E-05	-	1st analysis of dry saltcake sampled 5/21/81, p 7
1	2.9E-02	3.3E+00	1.8E-01	-	1.0E+03	2.8E-05	3.2E-03	1.7E-04	-	dupl analysis of dry saltcake sampled 5/21/81, p 11
Ci/gal of supernate (based on 400 gm salt / L)										per DPST-84-334:
4	-	-	1.0E-03	-	-	-	-	-	-	PUREX HAW supernate
8	-	-	2.4E-04	-	-	-	-	-	-	PUREX LAW supernate
8	-	-	3.0E-04	-	-	-	-	-	-	PUREX LAW supernate
8	-	-	2.7E-04	-	-	-	-	-	-	PUREX LAW supernate
8	-	-	1.7E-04	-	-	-	-	-	-	PUREX LAW supernate
8	-	-	1.5E-04	-	-	-	-	-	-	PUREX LAW supernate
8	-	-	1.7E-04	-	-	-	-	-	-	PUREX LAW supernate
8	-	-	1.7E-04	-	-	-	-	-	-	PUREX LAW supernate
47	-	-	6.5E-04	-	-	-	-	-	-	PUREX concentrated supernate
32	-	-	9.4E-04	-	-	-	-	-	-	HM HAW supernate
13	-	-	1.0E-03	-	-	-	-	-	-	HM LAW supernate
30	-	-	5.5E-04	-	-	-	-	-	-	HM concentrated supernate
1	-	-	3.0E-05	-	-	-	-	-	-	PUREX saltcake
1	-	-	2.4E-04	-	-	-	-	-	-	PUREX saltcake
19	-	-	3.0E-05	-	-	-	-	-	-	PUREX dissolved saltcake
20	-	-	9.1E-05	-	-	-	-	-	-	PUREX dissolved saltcake
9	-	-	6.1E-06	-	-	-	-	-	-	HM saltcake
10	-	-	9.1E-06	-	-	-	-	-	-	HM saltcake
Ci/gal of supernate										per DPST-84-343:
11	-	-	-	2.9E-07	-	-	-	-	-	fresh supernate (not yet conc by an evapr)
32	-	-	-	3.6E-07	-	-	-	-	-	fresh supernate (not yet conc by an evapr)
12	-	-	-	1.1E-06	-	-	-	-	-	concentrated supernate
15	-	-	-	1.1E-06	-	-	-	-	-	concentrated supernate (as corrected per ref 12a)
30	-	-	-	1.1E-06	-	-	-	-	-	concentrated supernate
9,10	-	-	-	<7.6E-09	-	-	-	-	-	saltcake (equiv supern at 400 gm salt/liter)

Table 4
Miscellaneous Supernate Samples

Chart 1
Cs-137 per Routine Supernate Samples