Conceptual Model for Hanford 242-Z Area Soil Disposal Sites and the Hanford Soil Inventory Model (SIM) - 11565

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ABSTRACT

Previous work has shown that the soil disposal sites at Hanford's Z-Area comprise a significant challenge for future cleanup. Prior to 1973, both High Salt Waste and Low Salt Waste from Z-plant operations were disposed to the soil column at Hanford. The current effort updates and corrects information already published in the Hanford Soil Inventory Model (SIM) with new information including for the first time the historical context of the various soil disposal sites for Hanford's plutonium processing plant.

Although past SIM reports showed inventories, the current work assembles for the first time information from many more disparate sources into a more comprehensive conceptual model for Z area that outlines not only disposals to cribs, but also to French drains, ditches, and the U-10 pond. Also there were errors in previous SIM results that have been corrected.

There remain many uncertainties in the overall distribution of waste constituents among these soil sites. Those uncertainties are captured and propagated within SIM using Oracle's Crystal Ball Excel add-in. This work is not yet complete but when complete, it will lay the groundwork for more detailed assessments in the future of the amount and distribution of important contaminants of concern such as carbon tetrachloride, nitrate, plutonium, and americium. This will the next step in this project.

INTRODUCTION

Previous work has shown that the soil disposal sites at Hanford's Z-Area comprise a significant challenge for future cleanup. [1,2] This paper represents a portion of a much larger ongoing effort to evaluate new information and update soil inventories at Hanford including the Z area Hanford Soil Inventory Model [4, 5]. This work includes not only updates to waste definition and distribution among soil sites but also corrections to previous SIM results due to errors.

The Z area comprises 9 different SIMwastes distributed among 25 disposal sites (SIMsites) as settling tanks, cribs, French drains, trenches, and the U-10 Pond shown in Fig. 1. Two of the Z area process wastes, Low Salt and High Salt Wastes (LSW and HSW), were routed to waste tanks after 1973 and are not included in SIM.

The Hanford Soil Inventory Model predicts chemical and radionuclide inventories and uncertainties for a set of ~400 of Hanford's soil disposal sites, SIMsites. Using archived data, SIM sends to each SIMsite volumes from various plants and processes and defines waste streams based on plant flowsheets and other information about those streams. Waste stream volume and inventory uncertainties depend on a set of derivations and SIM propagates those resultant inventory uncertainties for each of eight Hanford campaigns for the ~1200 nuclear fuel batches

of each of those eight campaigns. For chemical species SIM uses HDW species uncertainty wherever available and otherwise the radionuclide uncertainty defines the chemical uncertainty.

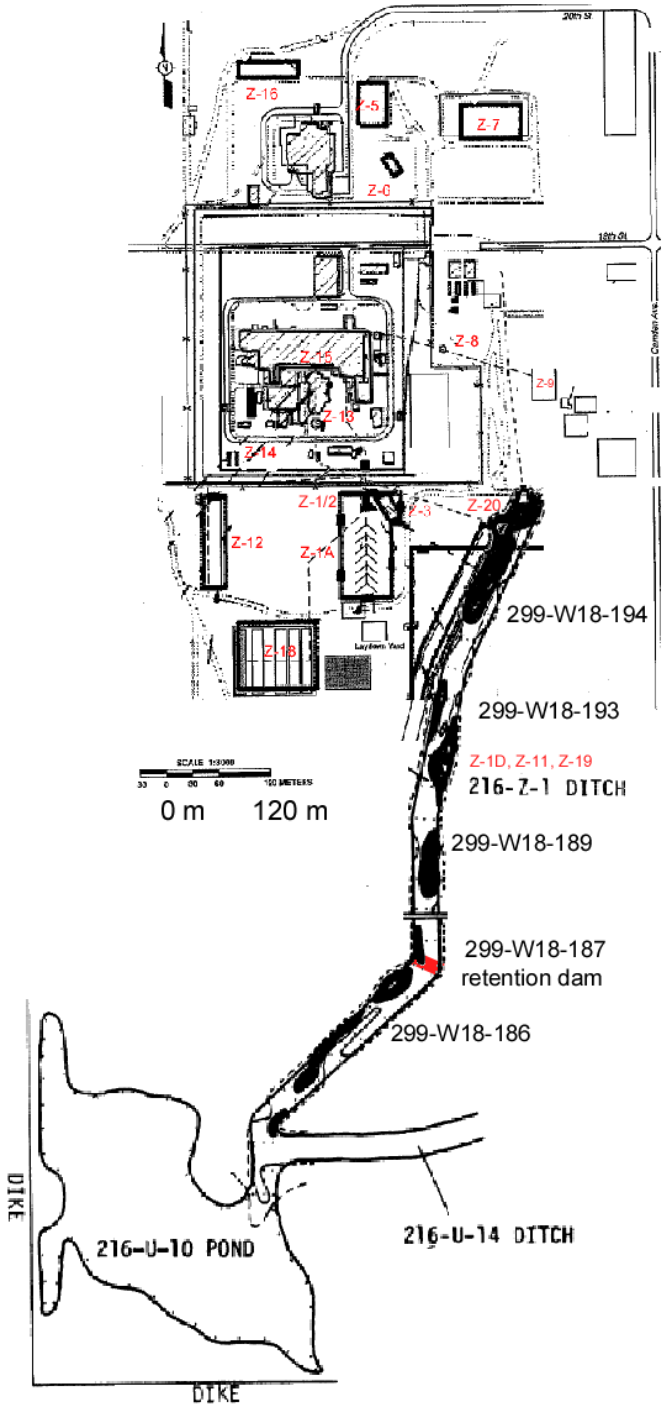

Fig. 1. Diagram of Z area facilities, cribs, French drains, trenches, and U-10 pond.

The SIM inventory has 75 species comprising 46 radionuclides, 29 chemicals discharged to the SIMsites, and one property, density. These SIMwaste discharges between 1944 and 2001 were to 377 disposal waste sites located on the central plateau portion of the Hanford site. The SIM2005

documents its calculations and contains SIMsite inventories and uncertainties for 377 SIMsites, including unplanned releases and tank leaks, by distributing some 196 waste streams, SIMwastes, among the 377 SIMsites.

METHODOLOGY

The calculation for SIMsites inventories simply multiplies a SIMsite volume and a SIMwaste composition, summing all SIMwastes at each SIMsite as

$$simS_i = \sum_j simVol_{ij} \mathbf{simW}_j \qquad \text{(Eq. 1)}$$

where

- simSi = composition vector for SIMsite i in kg or Ci
- simWj = composition vector for SIMwaste j in ppm or μCi/g x density (kg/L)
- simVolij = ML (1e6 L) volume of SIMwaste j for SIMsite i

to calculate a SIMsite inventory vector.

Each SIMwaste uncertainty is one of three types of probability distribution functions for each of the SIMwaste species and SIMsite volumes: triangular, lognormal, and beta.

The schematic in Fig. 2 shows the logic and information flow for SIM. A variety of information records provide the radionuclide and chemical concentrations for each SIMwaste as well as the SIMwaste volumes released to each SIMsite. Variabilities and uncertainties in these data then provide uncertainty bases as probability distribution functions (pdf's) for each species and volume. Sampling those pdf's with Monte-Carlo trials results in a large number of realizations (~450,000) that forecast the uncertainty distribution for each inventory species.

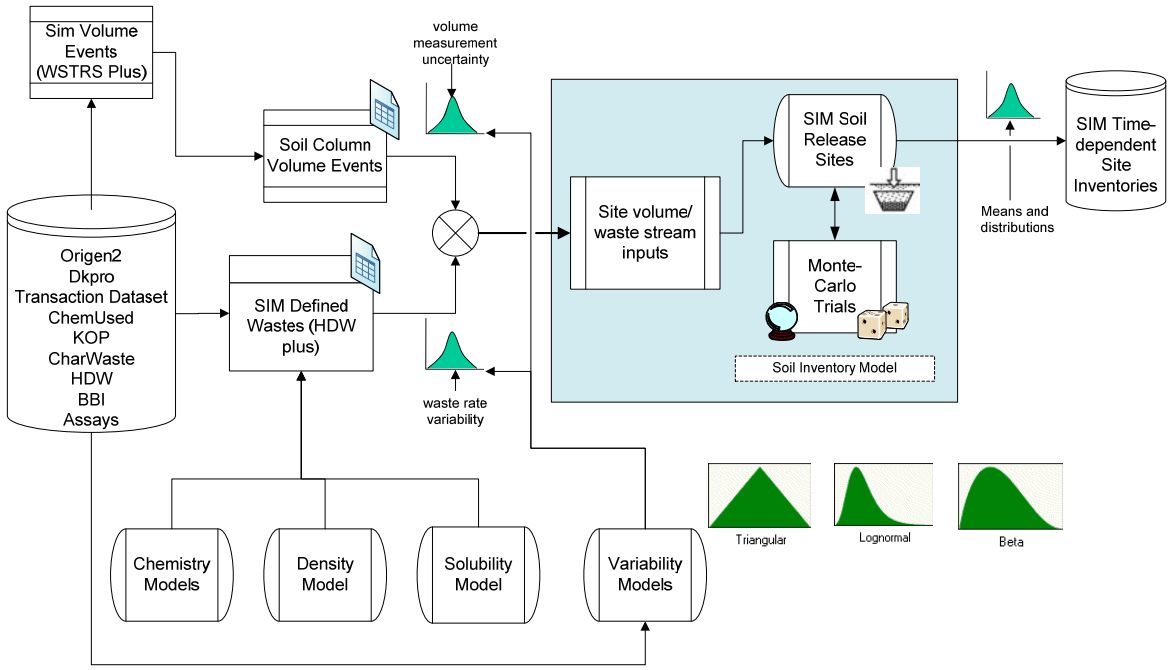

Fig. 2. Schematic of SIM logic and information flow.

There is no solubility model in SIM and thus there is no rigorous solids partitioning in SIM. However SIM does have very limited *ad hoc* retention and propagation assumptions. Therefore SIM does not account very well for insoluble solids that sedimented in the settling tanks associated with SIMsite cribs [9,10]. Furthermore, the retention and propagation of liquids and solids from open trenches that flowed into the U-10 pond are subject to very simple *ad hoc* SIM partitioning assumptions, not to a sedimentation and transport model.

The SIM also does not account for the various past and ongoing campaigns to remove and/or encapsulate contaminants from selected Z-area SIMsites. Among these are a Z-9 soil mining operation in 1976, a 450 MT soil vitrification of Z-12 in 1987, and a CCl_4 vapor stripping operation ongoing since 1996. These operations all have significant effects on the SIMsite inventories but those inventory changes are not completely accounted for by the SIM.

RESULTS AND DISCUSSION

Z Area SIMwastes and SIMsites

Operations over the history of the Z Area [2] produced plutonium metal “buttons” from a variety of sources. Primary plutonium sources were solutions from each extraction facility. Secondary plutonium was recycled or recovered from scrap and other wastes from the metal button processes as shown in Fig. 3. Tertiary plutonium was recovered along with americium from the CAW stream during a limited campaign with ion exchange resins (PuAmRec). There were nine unique SIMwastes for Z Area and three different campaigns radionuclide probability distributions {BT1, Z1, Z2} for most of these nine SIMwastes resulting in a total of 23 SIMwaste variations.

The Z Area SIMwastes were nearly all disposed to the soil column among some 25 Z Area SIMsites as cribs, French drains, closed trenches, open trenches, settling tanks, and the U-10 open pond or swamp.

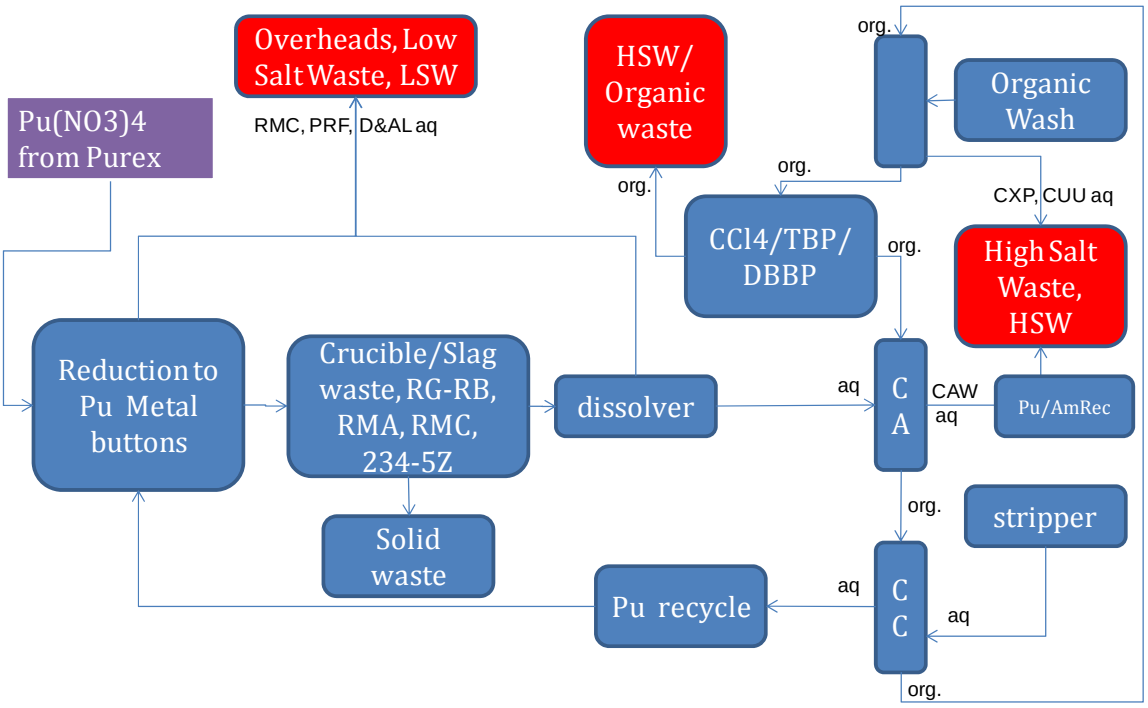

Fig. 3. Diagram of Z plant processes that resulted in three wastes shown: HSW-aq, HSW-org, and LSW.

Figure 4 shows Z Area production scaled as the total fuel burn-up expressed as GWd's/year (gigawatt-days per year). These Z area operations produced wastes that were proportional to this production curve. In addition, Fig. 4 shows variability as a vertical bar for each production year per batch of fuel processed. About 1,200 batches of fuel were processed over the history of Hanford operations and the batch-to-batch variability burn-up represents a fundamental uncertainty or variance in the partitioning of radionuclide inventories among SIMwastes.

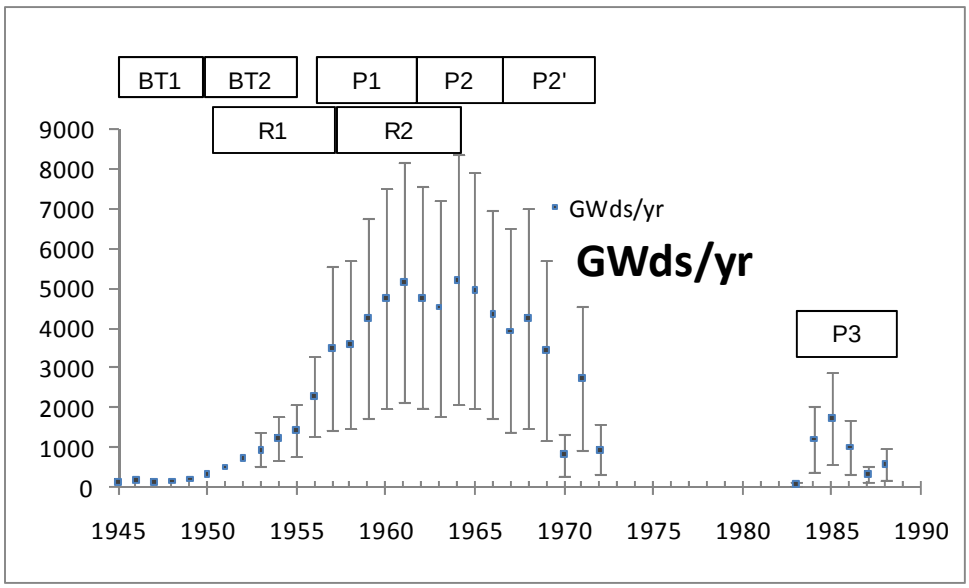

Fig. 4. Reactor production per year for Z Plant feed Pu with bars showing campaigns and error bars showing variation of GWds/batch for those campaigns.

Conceptual Model for Z Plant Wastes

Figure 5 shows Z-Plant waste streams broadly categorized into three primary groups: (1) Process wastes that originated from both metal production lines (Low-Salt Waste, LSW) and solvent extraction processes used to recover plutonium (High-Salt Waste, HSW-aq and HSW-org) (2) stack drainage from the main stack and HVAC systems with trace contaminants; and (3) cooling water and chemical sewer waste streams with much lower amounts of contaminants compared to the process streams.

Waste streams were discharged to separate waste-disposal sites, as Fig. 5 shows in a simplified manner according to these designations. The HSW and LSW process wastes are responsible for the vast majority of chemical and radiological contaminants discharged to the soil column while the CoolingWtr/ChemSewer/Lab wastes have by far the most volume.

The presentation of waste streams generated and disposed in Fig. 5 is simplified compared to the actual handling and disposal. Several different types of HSW and LSW streams were generated as reported by Barrington [7] and disposed at various waste sites, sometimes with both HSW and LSW routing to the same crib leading to a more complicated representation of actual disposed amounts.

Understanding these complex histories and developing a conceptual model is part of the current on-going work that will result in updating SIM inputs. Figure 6 shows a timeline of HSW and LSW SIMwastes resulting from six Z-Plant processes along with the various SIMsites that were used for disposal. The upper half shows the SIMsites that received the HSW stream while the lower half shows the SIMsites that received very high volumes but only mildly contaminated waste streams. Also shown are timelines from 1983-92 for wastes from periods when some wastes were routed to the tank farms instead of being sent to the SIMsites.

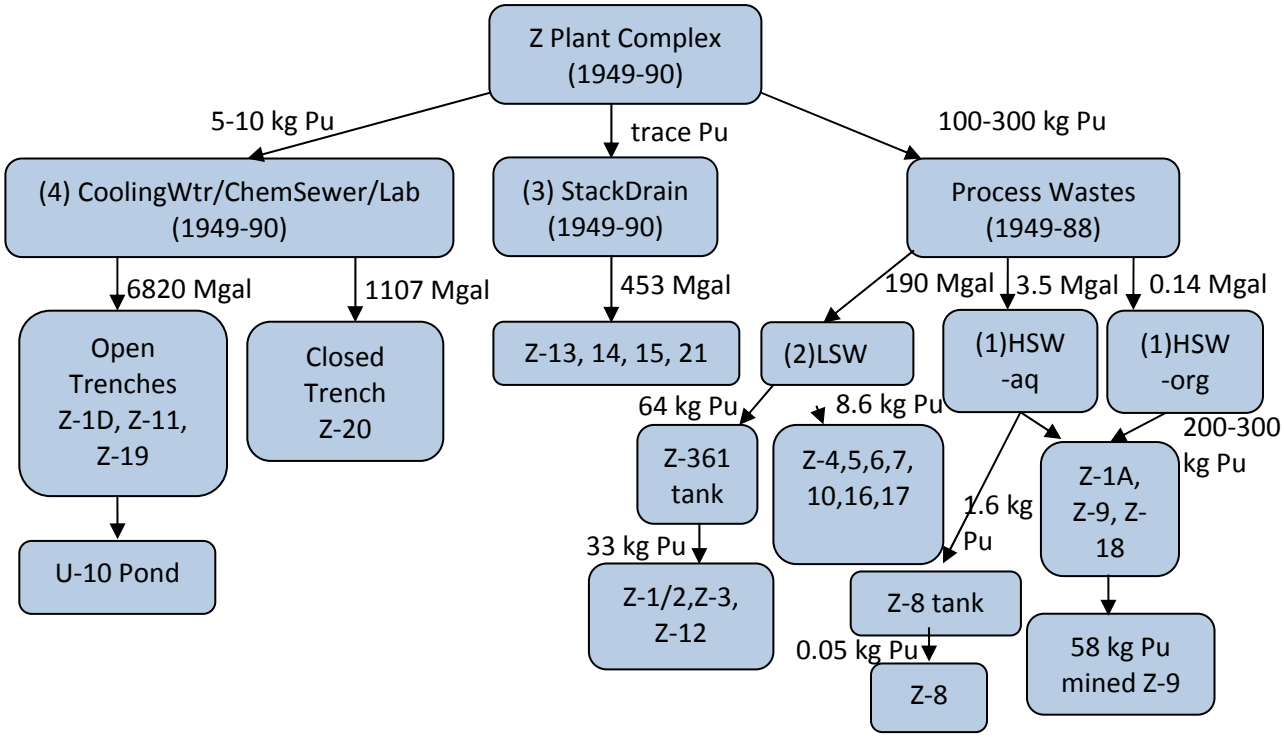

Fig. 5. Logic and Pu mass balance proposed for various Z-Plant Area soil sites.

Note that the SIMsite Z-1/2 was used mainly for LSW but also had some HSW sent to it through the Z-361 settling tank, but no spent organic waste, HSW-org.

The lower portion of Fig. 6 shows a timeline for disposal of large volumes of CoolingWtr / ChemSewer / Lab wastes among four trenches: Z-1D, Z-11, Z-19, and Z-20. Each of the three trenches Z-1D, Z-11, and Z-19 were open and flowed directly into U-10 pond (see Fig. 1).

There were no Z wastes discharged directly to U-10 pond but disposed liquids did flow through unlined trenches into U-10 pond. There are ad hoc assumptions in SIM that 20% of the liquids discharged to Z-1D, Z-11, and Z-19 were absorbed by the trenches and a much larger fraction, 99%, of Pu was absorbed in those trenches prior to the disposed liquid flowing into U-10 pond.

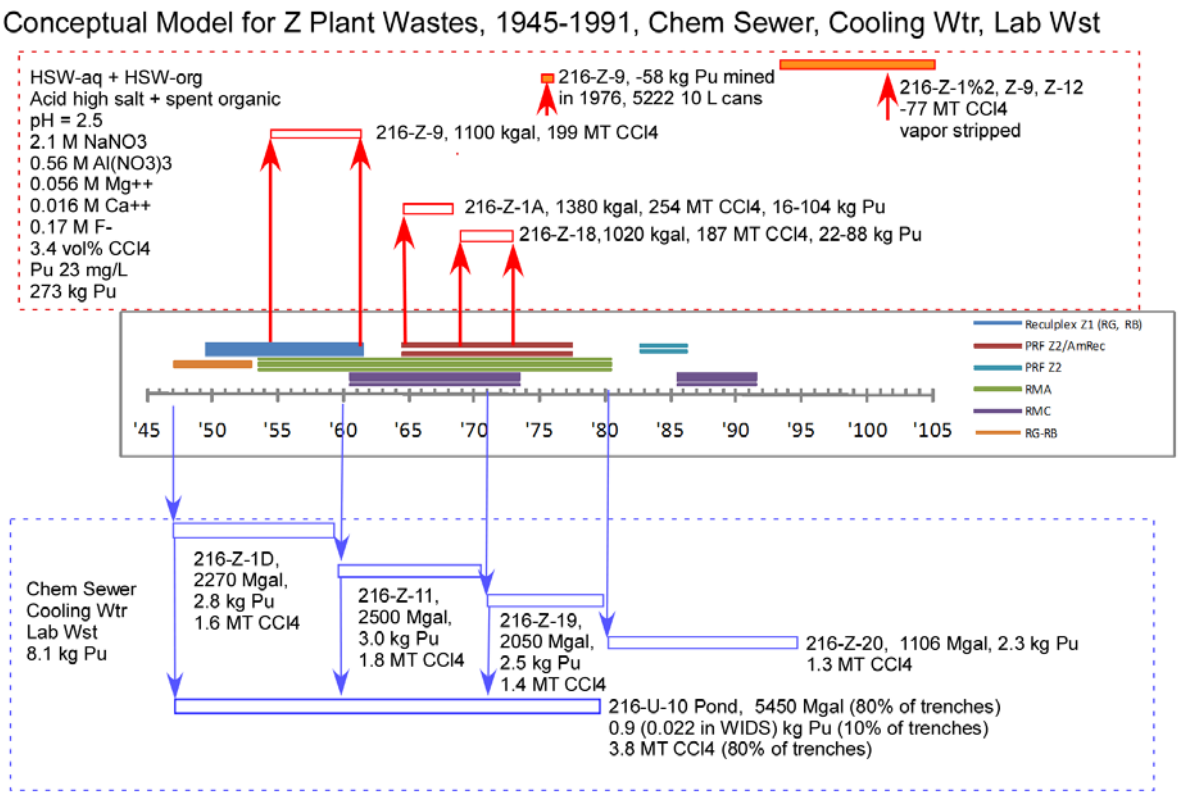

Fig. 6. Conceptual model for Z-Plant operations for HSW, ChemSewer, CoolingWtr, and LabWst for waste crib additions and mining along with timeline. The wastes sent to the lower cribs represented very large volumes much of which ended up in the U-10 pond before 1980.

High and Low Salt Aqueous Waste (HSW-aq and LSW)

The operations in Z plant were fairly complex and varied over time and there were, for example, a number of Recuplex waste stream descriptions. The most complete set are those of Barrington [7]. At the time of this description in 1989, though, HSW and LSW Z plant wastes were no longer disposed to the soil column. They were rather neutralized, combined, and disposed to the tank farms (tanks TX-118 and SY-102). Nevertheless, these flowsheet compositions apply to the previous campaigns before 1973 as well.

The amounts of Pu that were discharged to each waste stream were measured and accounted by assay. For reasons that are not entirely clear, this accounting method understated the actual amounts of Pu that were discharged to Z-9. For example, the Pu concentration varies from 14.8 to 8.6 mg Pu/L for HSW between Barrington and Upington, but the amount of Pu actually sent to Z-9 has been determined quite well [3] and is consistent with an HSW Pu concentration of 23 mg Pu/L, which is 2 to 3 times as much as the flowsheets note. This totals 90 kg Pu sent to Z-9 some 58 kg of which was mined in 1976.

Since the Pu accounting to Z-9 is the only definitive Pu concentration for HSW, all HSW is set to this same Pu concentration, 23 mg Pu/L. This increases the amount for Z-1A and Z-18 by as much as 40-50 kg each relative to WIDS as shown in Table II.

High Salt Waste Organic (HSW-org)

The organic solvent used during PRF operations was a mixture of carbon tetrachloride, tributylphosphate, and dibutylbutylphosphonate [6,7,8]. The amounts of these species that were disposed to the HSW organic crib sites has been described elsewhere [1] and Table I shows those reported amounts. Although a range of inventories were reported for Z-9, SIM fixes Z-9 CCl_4 to an average 3.1 vol% CCl_4 that is consistent with amounts reported to have been disposed to Z-1A and Z-18.

The SIM2005 HSW-org organic waste streams contained a large fraction of Pu and Am sent to HSW cribs. While it is certain that these organic streams were contaminated with Pu, the amounts likely varied and were not reported separately from the aqueous high salt waste, HSW-aq. Since both HSW-aq and HSW-org were disposed to the same soil crib sites, Z-1A, Z-9, and Z-18, we propose associating all Pu/Am residues with HSW-aq. Any further analysis of transport of these COPC's would need to better account for organic versus aqueous partitioning of contaminants of concern.

Table I. Carbon tetrachloride distribution among Z soil disposal sites

Crib	Waste Type	Total aq+org kgal	kgal ORG	CCl4 MT	References
Z-1A	HSW-org	1,380	184	254	WIDS, 3.1 vol%
Z-9	HSW-org	1,080	125	199	Proportional to Z-1A, Z-18
Z-18	HSW-org	1,020	115	187	WIDS, 3.1 vol%
Z-1D	ChemSewer	2,270,000		1.6	Scaled to Z-20
Z-11	ChemSewer	2,500,000		1.8	Scaled to Z-20
Z-19	ChemSewer	2,050,000		1.4	Scaled to Z-20
U-10 pond	Z-1D + Z-11 + Z-19	5,450,000		(3.8)	80% of Z-1D + Z-11 + Z-19
Z-20	ChemSewer	1,106,000		1.3	71 ppmv, k_H = 0.0305 atm-m^3/mol
total				633	
Total Vapor Stripped				77	Vapor stripping campaign, Z-9, Z-1/2, Z-18, Z-12, 1992-2006.

Table II shows the proposals for both plutonium and carbon tetrachloride inventories along with their variances from previous work based on the Monte-Carlo propagation [4]. Although these species distributions are not normal, the variances shown can be interpreted as roughly comparable to a relative standard deviation (RSD) of a normal distribution function. In the final SIM update, these distribution functions will be propagated as realizations and forecasts with a Monte-Carlo tool.

Table II. SIMsites for Z Area

index	SIMsite	Type	ML (1e6 L)	Lower Bound Pu (kg)	Upper Bound Pu (kg)	Mean CCl_4 (kg)	CCL_4 Variance†
1	216-Z-1/2	LSW crib	34	3.1	12	31	12.8%
2	216-Z-1A	HSW crib	6.2	16	104	254,000	21.1%
3	216-Z-3	LSW crib	178	4.2	7.0	65	12.5%
4	216-Z-4	LSW crib	0.011	0.0060	0.016		
5	216-Z-5	LSW crib	31	0.39	0.61	10	21.3%
6	216-Z-6	LSW crib	0.098	0.013	0.035		
7	216-Z-7	LSW crib	80	6.6	9.2	24	13.2%
8	216-Z-8	HSW French drain	0.067**	0.63	2.8		
9	216-Z-9	HSW crib	4.1	61	119	199,000	12.3%
	216-Z-9	mined in 1976		-58	-58		
10	216-Z-10	LSW crib	1.0	0.13	0.35	0.3	31.8%
11	216-Z-11	Open trench to U-10	9,460	1.9	4.1	1,750	
12	216-Z-12	LSW crib	257	12	26	60	10.6%
	216-Z-12	450 MT soil vitrified '87					
13	216-Z-13	French drain	50				
14	216-Z-14	French drain	52				
15	216-Z-15	French drain	48				
16	216-Z-16	LSW crib	102	0.030	0.110		
17	216-Z-17	LSW crib	37				
18	216-Z-18	HSW crib	3.9	22	88	187,000	15.2%
19	216-Z-19	Open trench to U-10	7,740	0.09	4.9	1,440	
20	216-Z-20	Closed trench	4,190	0.083	4.5	1,310	
21	216-Z-21	Seepage Basin	1,570				
22	216-Z1D	Open trench to U-10	8,600	0.10	5.5	1,600	
		Totals	32,400	90	310	646,000	
		vapor stripping				-77,000	
			~same as Rev. 1	was 174 kg		was 912,000 kg	
23	216-U-10	Open pond fed by Z-1D, Z-11, Z-19	25,800***	8.3***		3,800***	
24	241-Z-361	LSW settling tank for Z-1/2, Z-3, Z-12	469*	32*			
25	241-Z-8	HSW settling tank	0.067**	1.7**			

*Total volume routed through Z-361 settling tank but only 48% of Pu kg is amount now in tank sludge.

**Actual amounts are split between Z-8 settling tank and Z-8 French drain.

***Actual amounts in U-10 Pond are a fraction of listed sums of Z-1D, Z-11, Z-19.

†Variance equivalent to relative standard deviation (rsd) for a normal distribution.

SUMMARY

These results for Z Area SIMsites are a portion of a larger project to update the SIM and only represent proposed changes at this time. In particular, we propose standardizing amounts and variances of two important COPC's, carbon tetrachloride and plutonium, among the Z area soil disposal sites along with an large number of other species.

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