START

0028455

RHO-RE-SA-216 P

Characterization of Airborne Radionuclide Particulates in Ventilated Liquid Waste Tanks

June 1987

R. T. Kimura
D. W. Lindsey
Process Engineering
Research and Engineering

Presented at the American Nuclear Society
Annual Meeting in Dallas, Texas June 7-11, 1987

Prepared for the U.S. Department of Energy
under Contract DE-AC06-77RL01030

9313022.1743

Rockwell International
Rockwell Hanford Operations
P.O. Box 800
Richland, Washington 99352

JUN 1993
RECEIVED
EDMC

DISCLAIMER

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

Printed in the United States of America

DE83022744

ABSTRACT

Tank vapor spaces in nine selected underground double-wall tanks at the U.S. Department of Energy Hanford Site have been sampled to characterize airborne radionuclides present in the form of particulates or aerosols. These tanks are actively ventilated and are kept under negative pressure. The vent gases containing the particulates are drawn through high-efficiency particulate air (HEPA) filters prior to sampling and discharging to the environment. Quantification of airborne radioactive particulates in these operating tanks is important in the development of credible accident scenarios for safety analysis, and these data are also useful for estimating HEPA filter effectiveness.

The sample system pulled gas samples isokinetically from the ventilation duct near the tank at a rate of 62.3 L/min for durations ranging between 30 min and 2 h. The filters were subjected to tests to determine the types of radioactivity present and to identify and quantify all significant radionuclides present. Gross alpha and beta analyses were initially performed followed by a gamma energy analysis (GEA). Analysis for strontium-90 was also performed when beta-emitting radionuclides identified from the GEA could not account for a majority of the beta activity on a filter.

Results indicate that the highest airborne activity is present in a tank that uses airlift circulators for agitation and mixing of waste. Tanks

9313022.1745

regularly receiving waste have higher concentrations in a vapor space than do stagnant tanks. Comparison of concentrations of Cesium-137, strontium-90, ruthenium-106, and other radionuclide concentrations in the vapor space to those in the tank liquid at the time of sampling showed partition factors between 10^{-9} and 10^{-13}. A partition factor is the ratio of vapor to liquid concentration for a given radionuclide. The highest partition factors for ^{137}Cs was observed in the tank having airlift circulators. Higher radionuclide particulate concentrations could result in more frequent HEPA filter replacements and a higher source term for use in evaluating potential accident scenarios. The lowest partition factors for ^{137}Cs and other radionuclides were observed in tanks not actively receiving wastes or not having airlift circulators. This behavior was expected because both of these latter operations can product higher concentrations of airborne particulates and aerosols.

9913022.1745

CONTENTS

Introduction 1

Description of Equipment 1
Ventilation System 1
Sampler 1
Filters and Filter Handling 4
Miscellaneous Equipment 5

Results and Discussions 5
Sample Results During Mixing Operations 5
Sample Results During Transfer Operations 6
Sample Results Under Stagnant Conditions 10
Isokinetic Sample Data 10

Conclusion 10

References 14

Figures:
1. Underground Double-Wall Storage Tank 2
2. Sampler Schematic 3
3. Cesium-137 Partition Factors 13

Tables:
1. Mixing Tank 102-AW Results 7
2. Partition Factors 8
3. Waste Receiving Tanks 9
4 Stagnant Tanks 11
5 Isokinetic Ratios 12

9313022.1747

THIS PAGE INTENTIONALLY
LEFT BLANK

INTRODUCTION

The vapor spaces of selected Hanford Site underground double-wall tanks were sampled to estimate the type and quantity of airborne radionuclide particulates present. The purpose of the characterization effort was to obtain data on radioactive aerosols and particulates under a variety of operating conditions. Concentrations were obtained under three distinct operating conditions. These conditions included sampling above stagnant liquids, sampling during liquid waste transfer activities, and sampling during agitation and mixing operations involving airlift circulation. The goal of the study was to determine a partition factor (PF), which is the ratio of air to liquid concentration. Radionuclides were detected by Gamma Energy Analysis (GEA). These data are useful for safety analyses of the double-walled tanks and for estimating high-efficiency particulate air (HEPA) filter decontamination factors (DF). Cesium-137 and cesium-134 are prevalent in nearly all double-wall tank wastes; therefore, they were used for comparison purposes. Process control of these tank farm facilities will be improved when the specific operating variables that are the major contributors to higher partition factors are determined. Controlling these parameters will help reduce airborne activity inside the tanks and will reduce loading of the HEPA filters.

DESCRIPTION OF EQUIPMENT

9313022.1761

VENTILATION SYSTEM

Double-wall tanks have two ventilation systems. The first system insures that a slight vacuum is maintained on the primary tank, and the second system maintains a vacuum for the annular space between the primary and secondary tank. The primary tank system is of concern (fig. 1). The underground ventilation line from the primary tank passes through an underground containment structure or pit. Sampling is performed in these pits because they provide a barrier between the sampler and the environment. The ventilation air passes through the pit and then enters a HEPA filter system. Air samples are pulled from a 0.0254-m diameter access port located on the ventilation line. The transportable sampler unit is lowered into the pit and connected to an access port (fig. 2).

SAMPLER

Air is drawn into the sampler through a nozzle attached to a short segment of tubing. The inlet nozzle points against the direction of flow and is centered inside the ventilation pipe. The length of the inlet tubing is kept as short as possible and only one 90-degree, 6-in. radius bend is allowed. This minimizes the amount of line loss upstream of the filter papers. Heat tape and insulation are wrapped around the inlet tubing.

9313022.1760

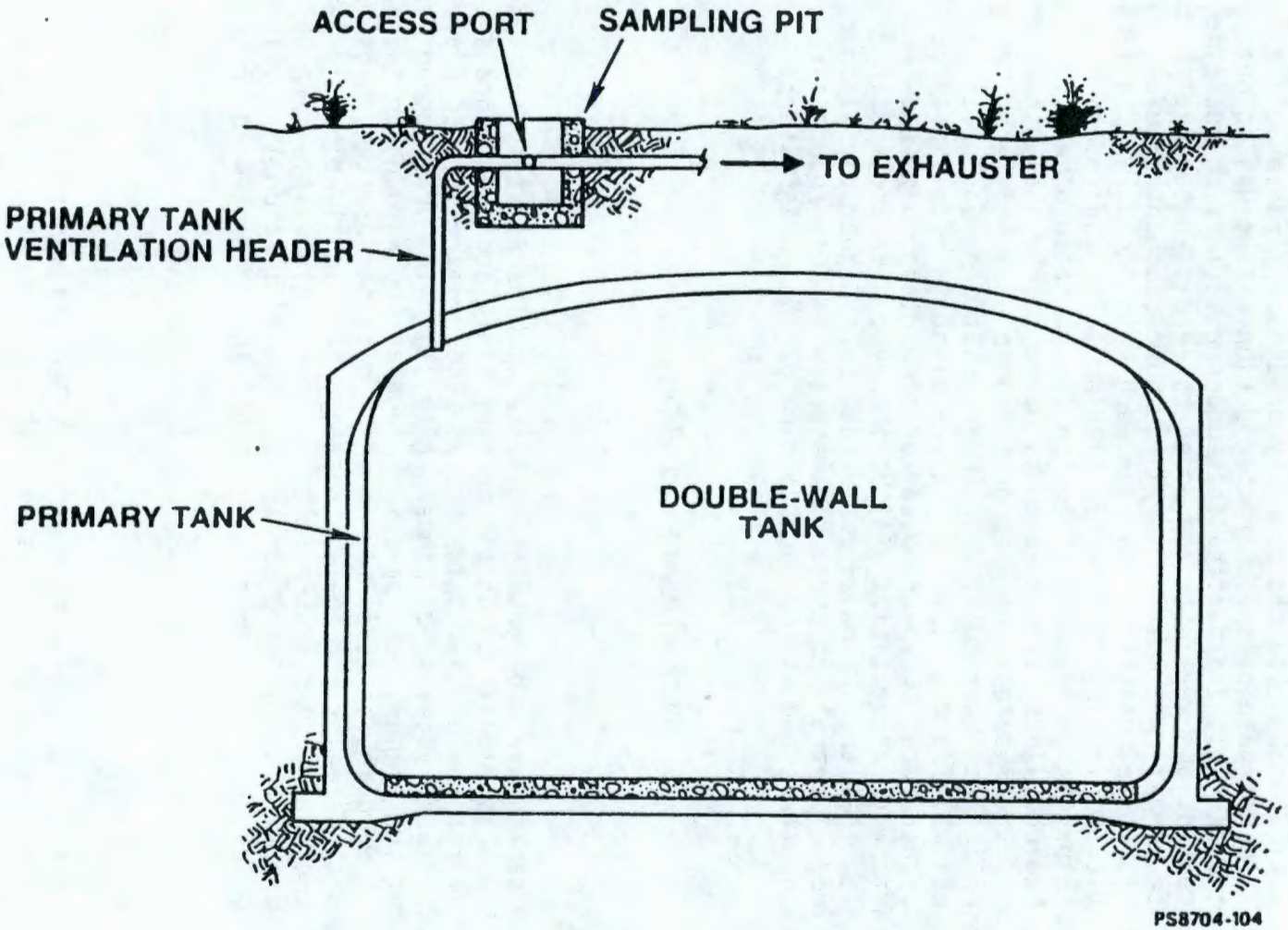

Figure 1. Underground Double-Wall Storage Tank.

RHO-RE-SA-216

9313022.1759

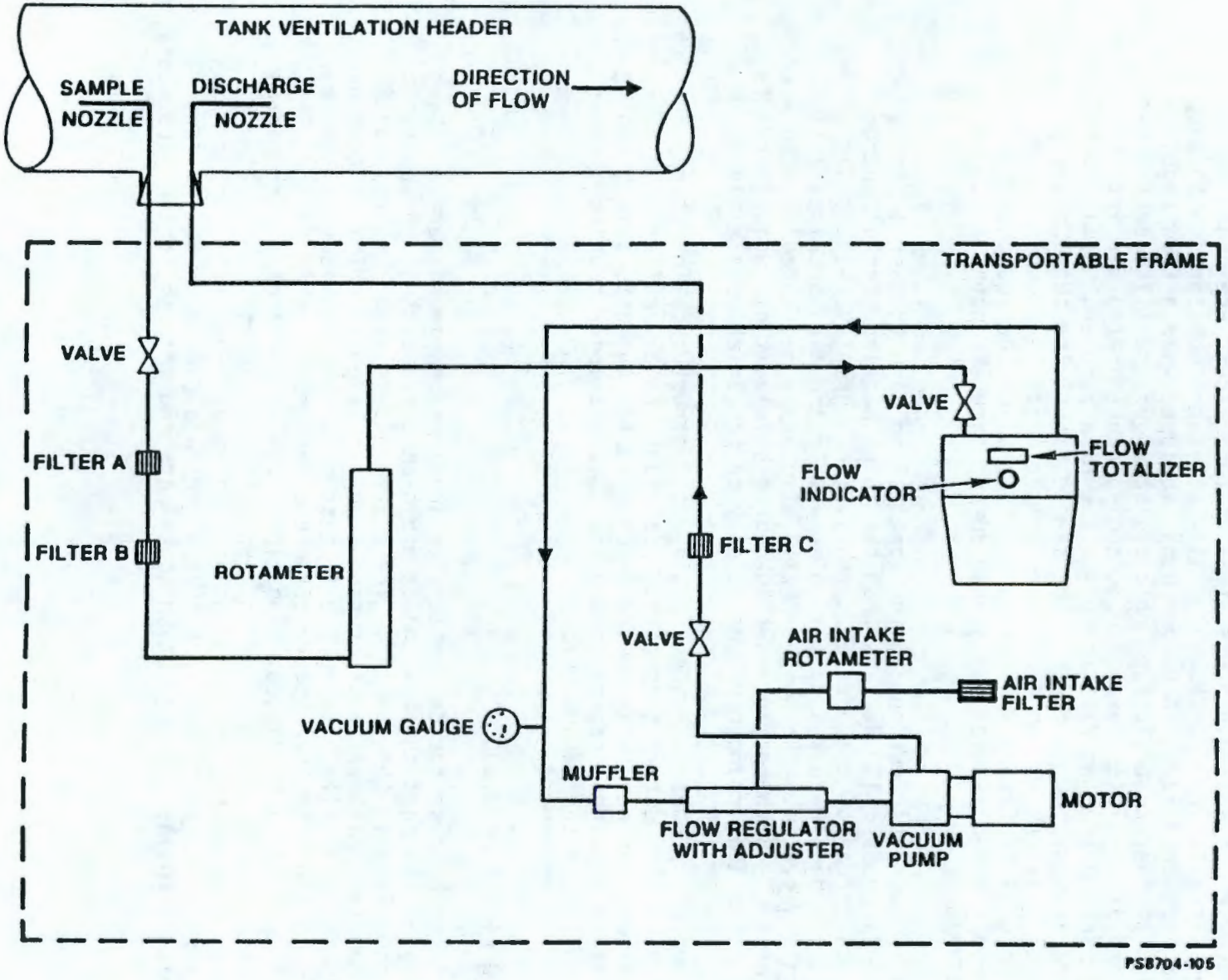

Figure 2. Sampler Schematic.

This prevents moisture from condensing on the tubing or wetting the filter paper. The sample line is replaced if any smearable activity is detected.

Inside the sampler, the air is filtered through two 3-μm pore size filters arranged in series. Stainless steel filter holders are designed for easy filter installation and removal. Flow rates are monitored with a rotameter and a flow totalizer. A vacuum pump, equipped with a flow regulator to maintain isokinetic sample flow rates, discharges the filtered air back into the vent line. This prevents discharging contaminated air into the environment should the two filters fail. The flow regulator admits outside air into the vacuum pump to control the sample flow. The outside air passes through an air filter, and an air rotameter measures the amount of air bypassing the filter papers. Three ball valves allow the equipment to be isolated from the ventilation air stream during sampler installation and filter replacement.

The discharged air returns to the ventilation line through a discharge nozzle that is opposed 180 degrees from the inlet nozzle. The discharged air flows downstream of the nozzle assembly. This greatly reduces the potential for recycling air back into the inlet nozzle and also provides a safe, closed sampling system. The outlet nozzle also has a 6-in. radius bend like the inlet nozzle. Both nozzles have 30-degree beveled tips, which serve to minimize particulate impingement, especially on the inlet nozzle tip.

The sampler is housed inside a rigid metal structure. The sturdy housing protects the sampler during transport. Each tank has a ventilation pit into which the sampler is carefully lowered. Once inside the pit, sample tubing is connected to fittings located on the top of the sampler housing. Power is then supplied to the vacuum pump.

9313022.1758

FILTERS AND FILTER HANDLING

Two filter holders inside the sampler are arranged in series to remove the majority of the particulates and to minimize the amount of contamination that may accumulate in the sampler. The filter papers used are Versapor 3000 Supported Membrane Filters manufactured by Gelman Sciences. The filters had a 3-μm diameter pore size. Using two filters in series provides data which can be used to estimate the capture efficiency of the first filter for individual radionuclide particulates. Two clean 0.047-m diameter filter papers are inserted into the holders. After a specified volume of sample air is drawn through the two filters, the vacuum pump is shut off, the sampler is isolated by closing the valves, and the filters are removed. The filters are then placed inside transportation containers and shipped to the laboratory for immediate analysis.

MISCELLANEOUS EQUIPMENT

Plastic bags and steel drums are used to safely dispose of contaminated equipment. Used tubing, gloves, fittings, nozzle assemblies, heat tape, and insulation are wrapped and taped in plastic and stored in drums for disposal. Different types of radiation detection equipment are used to survey the waste, the sampler, and housing prior to transport. The sampler is also wrapped in plastic as a precautionary measure because it is potentially contaminated.

RESULTS AND DISCUSSION

Results from a GEA of each filter paper provided an estimate of the total activity of each gamma-emitting radionuclide present above its minimum detectable level. Filters A and B were analyzed within 8 h after removal from the filter holders. Concentrations were then calculated using sample volume data provided by flow totalizer. These concentrations were then corrected using an estimated filtration efficiency. The efficiency was calculated from the activity present on both filters. Concentrations were not corrected for line losses because the line length, configuration of inlet tubing, and the total volume of each sample served to minimize the error. In general, surveys of the sample tubing and holders showed below-background radiation levels although there were a few samples that did deposit minor contamination on the tubing and filter holders. Sample volumes were kept as large as possible to reduce the line loss error and to draw a representative volume of air from the tank vapor space. A constraint was also imposed on the volume the sample taken to minimize the exposure to workers handling and analyzing the filter papers.

9313022.1757

SAMPLE RESULTS DURING MIXING OPERATIONS

The highest cesium PFs were seen in Tank 102-AW. This tank contains two airlift circulators that agitate and mix a variety of radioactive liquid wastes. An airlift circulator mixes liquid wastes by injecting air into a large vertical submerged pipe (see fig. 1). The air-liquid mixture inside the pipe rises because it is less dense than the surrounding liquid. This induces flow into the bottom of the pipe and causes the tank liquids to circulate. The air is released at the liquid surface and is removed by the ventilation system. Airborne particulates are generated at the turbulent air-liquid interface. Those particulates that do not fall back into the liquid serve to increase the concentration of airborne particulates that are carried into the ventilation system. Other factors affecting the concentration, which are recommended for future study, are liquid level, liquid temperature, and airlift circulator air rate. The effect of these parameters on PF or particulate concentration may be as significant as the operational parameters studied here. Higher liquid concentrations may also affect both the airborne activity and the PF for other radionuclides.

Partition factor values were calculated using both the corrected concentration and the liquid concentrations of a given radionuclide inside the 102-AW Tank. Data for the three samples are shown in table 1, and PF values are listed in table 2. The liquid concentrations shown in table 2 were obtained at the time of sampling. Both cesium-137 and cesium-134 were detected in all three samples. In addition, ruthenium-rhodium-106 and niobium-95 were also detected.

Cesium-137 concentrations ranged between 3,106 Bq/m^3 and 11,489 Bq/m^3. Cesium-137 PF values ranged between 1.02×10^{-9} and 2.49×10^{-9} while cesium-134 PF values ranged between 3.35×10^{-11} and 8.84×10^{-8}.

For future characterization work, more detailed studies are recommended to be performed on this tank due to its high airborne activity. Characterization work on radioactive gases is also recommended. Radioactive gas sampling is commonly performed on gaseous effluent streams. Iodine-129 and antimony-125 can be captured with filter media specific to these nuclides. The sampler can be modified to include a charcoal and/or silver zeolite cartridge for collection of iodine and antimony (McManus and Fernandez 1986). Work can even be performed on individual iodine species if they are present (Mandler et.al. 1986)

In addition, alpha energy analyses are recommended to be performed in future studies on Tank 102-AW. Trace quantities of transuranic elements as well as certain uranium isotopes exist in some waste tanks.

9313022.1756

SAMPLE RESULTS DURING TRANSFER OPERATIONS

In general, waste-receiving tanks had higher PF values than did stagnant tanks, but they had lower PF values than Tank 102-AW. Table 3 lists five samples that were taken from three tanks. Filter activity on both A and B filters, the estimated efficiency, and the corrected concentration based on the GEA were calculated for each radionuclide detected. Cesium-137 concentrations ranged between 5 and 649 Bq/m^3 and cesium-134 concentrations ranged between 0.06 and 44 Bq/m^3.

Partition factors for these samples are shown in table 2. The highest PF for cesium-137 was seen in Tank 105-AW, sample number 105-AW-3, at 5.25×10^{-10}. Higher airborne activity can be attributed to surface agitation and spraying of waste as it falls into the tank. Temperature and composition of the pumped liquid and the tank liquid level are all contributing factors to airborne activity. The PF values obtained here also closely represent typical vapor to liquid concentrations. The PF values are typical because the liquid composition in each of these three tanks are relatively constant due to segregation of waste streams. The in-tank liquid composition may change slightly if settling occurs or if surface evaporation takes place.

Table 1. Mixing Tank 102-AW Results.

Sample number	Nuclide	Filter A (Bq)	Filter B (Bq)	Efficiency (%)	Corrected concentration (Bq/m^3)
102-AW-1	^{137}Cs	40,700	0[a]	100	3,107
	^{134}Cs	81,400	0[a]	100	6,214
	^{106}RuRh	5,550	25	99 +	424
102-AW-2	^{137}Cs	9,990	14	99 +	5,876
	^{134}Cs	237	0[a]	100	139
	^{106}RuRh	1,739	0[a]	100	1,023
	^{95}Nb	13	0[a]	100	8
102-AW-3	^{137}Cs	21,830	30	99 +	11,489
	^{134}Cs	8	0[a]	100	4

[a]0 = below minimum detectable levels.

PST87-3225-1

9313022.1755

Table 2. Partition Factors.

Sample number	Nuclide	Vapor (Bq/m3)	Liquid (Bq/m3)	PF (vapor/liquid)
102-AW-1	^{137}Cs	3,107	3.05 E + 12	1.02 E – 09
	^{134}Cs	6,214	7.03 E + 10	8.84 E – 08
	^{106}RuRh	424	6.33 E + 11	6.70 E – 10
102-AW-2	^{137}Cs	5,876	3.05 E + 12	1.93 E – 09
	^{134}Cs	139	7.03 E + 10	1.98 E – 09
	^{106}RuRh	1,023	6.33 E + 11	1.62 E – 09
	^{95}Nb	8		
102-AW-3	^{137}Cs	11,500	4.63 E + 12	2.49 E – 09
	^{134}Cs	4	1.22 E + 11	3.35 E – 11
105-AW-1	^{137}Cs	5	1.24 E + 12	4.14 E – 12
	^{134}Cs	0	1.01 E + 11	5.95 E – 13
105-AW-2	^{137}Cs	3	1.24 E + 12	2.45 E – 12
105-AW-3	^{137}Cs	649	1.24 E + 12	5.25 E – 10
	^{134}Cs	45	1.01 E + 11	4.43 E – 10
	^{106}RuRh	292	2.23 E + 11	1.31 E – 09
	^{95}Nb	238		
	^{95}Zr	199		
	^{113}Sn	12		
101-AW-1	^{137}Cs	43	1.65 E + 11	2.63 E – 10
	^{134}Cs	2	1.40 E + 10	1.65 E – 10
	^{106}RuRh	44	2.84 E + 11	1.56 E – 10
	^{95}Nb	9		
	^{95}Zr	1		
	^{144}CePr	52		
106-AW-1	^{137}Cs	7	7.10 E + 12	1.02 E – 12
	^{113}Sn	1		
101-SY-1	^{137}Cs	1,808	2.62 E + 13	6.91 E – 11
101-SY-2	^{137}Cs	172	2.62 E + 13	6.58 E – 12
105-AN-1	^{137}Cs	1	2.52 E + 13	4.23 E – 14
106-AN-1	^{137}Cs	3	1.28 E + 13	1.97 E – 13
	^{95}Nb	151		
107-AN-1	^{137}Cs	160	1.67 E + 13	9.62 E – 12
	^{134}Cs	8		
	^{106}RuRh	46		
	^{95}Nb	468		
	^{95}Zr	2		
	^{103}Ru	4		
104-AW-1	^{137}Cs	19	1.87 E + 14	1.00 E – 13
	^{106}RuRh	6		
	^{90}Sr	3	8.55 E + 10	3.45 E – 11

NOTE: Blank indicates no data available.

PST87-3225-2

9313022.1754

Table 3. Waste Receiving Tanks.

Sample number	Nuclide	Filter A (Bq)	Filter B (Bq)	Efficiency (%)	Corrected concentration (Bq/m^3)
105-AW-1	^{137}Cs	35	0[a]	100	5
	^{134}Cs	0.6	0[a]	100	<1
105-AW-2	^{137}Cs	38	2	94	3
105-AW-3	^{137}Cs	2,013	10	99 +	649
	^{134}Cs	138	0[a]	100	45
	$^{106}RuRh$	907	0[a]	100	292
	^{95}Nb	740	0[a]	100	239
	^{95}Zr	618	0[a]	100	199
	^{113}Sn	36	0[a]	100	12
101-AW-1	^{137}Cs	481	241	50	43
	^{134}Cs	19	1	94	2
	$^{106}RuRh$	370	21	94	44
	^{95}Nb	104	52	50	9
	^{95}Zr	11	0[a]	100	1
	$^{144}CePr$	814	555	32	52
106-AW-1	^{137}Cs	56	19	67	7
	^{113}Sn	4	0[a]	100	1

[a]0 = below minimum detectable levels.

PS187-3225-3

9313022.1753

SAMPLE RESULTS UNDER STAGNANT CONDITIONS

In general, the lowest airborne activities were found in the stagnant tanks in which no agitation or waste transfers were occurring (table 4). Only minor air movement was present in the vapor spaces of these tanks due to the tank ventilation systems. Ventilation was present in all tanks sampled. The largest number of data points were obtained from stagnant tanks, which comprise the majority of double-wall tanks on the Hanford Site.

Cesium-137 concentrations varied between 1 and 1,808 Bq/m^3, and the PFs varied between 4.23×10^{-14} and 6.91×10^{-11}. No cesium-134 PFs could be calculated because cesium-134 was only found in 107-AN-1, and no representative liquid data was available for this tank. Although liquid concentrations were as high or higher than the more active tanks, PFs were much lower. This was expected due to the lack of a motive force to generate aerosols or particulates.

ISOKINETIC SAMPLE DATA

All samples were taken as close to isokinetic as possible (table 5). Ventilation line flow velocities varied for each tank, and sample flow rates were adjusted to meet the line velocity. All samples were very close to isokinetic except for samples taken from Tanks 105-AN, 106-AN, and 107-AN. This was due to exceptionally low line velocities. Sampling time would have been excessive at the lower flow rates in this particular farm. These three samples (105-1, 106-1, and 107-1) should be taken again at a later date when time permits sampling at lower flow velocities.

9313022.1752

CONCLUSION

Figure 3 contains all cesium-137 PFs as calculated from all 13 samples representing nine different double-wall tanks. The highest PFs occurred in Tank 102-AW, where mixing operations took place. Figure 3 shows that there is indeed a correlation between airborne cesium-137 activity and the operating condition or status of a tank.

Data points 1, 2, and 3 represent PF values calculated while airlift circulators were in operation. The higher PFs represent higher air to liquid concentrations. These PF values were used in a safety analysis report and provided an order of magnitude estimate for future airborne activity at higher liquid concentrations.

Data points 4, 5, 9, and 10 represent partition factors that were obtained during transfer operations. The wide range of PF values were attributed to significantly different and segregated transfer operations that were occurring. This is most likely a function of temperature and composition of the waste entering a tank and the tank liquid level.

Table 4. Stagnant Tanks.

Sample number	Nuclide	Filter A (Bq)	Filter B (Bq)	Efficiency (%)	Corrected concentration (Bq/m^3)
101-SY-1	^{137}Cs	6,882	275	96	1,808
101-SY-2	^{137}Cs	370	56	85	172
105-AN-1	^{137}Cs	8	0[a]	100	1
106-AN-1	^{137}Cs	18	2	90	3
	^{95}Nb	11	0[a]	100	2
	^{90}Sr	17	0[a]	99	2
107-AN-1	^{137}Cs	1,554	740	52	160
	^{134}Cs	70	33	53	8
	^{106}RuRh	444	244	45	46
	^{95}Nb	3,182	0[a]	100	468
	^{95}Zr	11	0[a]	100	2
	^{103}Ru	28	0[a]	100	4
104-AW-1	^{137}Cs	148	3	98	19
	^{106}RuRh	48	0[a]	100	6
	^{90}Sr	23	0[a]	99	3

a = below minimum detectable limits

PSTB7-3225-4

9313022.1751

Table 5. Isokinetic Ratios.

Sample number	Vent line (m^3/s)	Vent line velocity (m/s)	Sampler (m^3/s)	Sampler velocity (m/s)	Isokinetic ratio
106-AW-1	0.41390	5.67	0.00105	5.73	1.01
105-AW-1	0.41390	5.67	0.00104	5.69	1.00
105-AW-2	0.41390	5.67	0.00102	5.60	0.99
105-AW-3	0.41390	5.67	0.00102	5.60	0.99
102-AW-1	0.41390	5.67	0.00110	6.04	1.06
102-AW-2	0.41390	5.67	0.00105	5.78	1.02
102-AW-3	0.42570	5.83	0.00098	5.39	0.92
104-AW-1	0.43750	6.00	0.00110	6.04	1.01
101-AW-1	0.46110	6.32	0.00113	6.21	0.98
107-AN-1	0.28317	3.88	0.00106	5.82	1.50
106-AN-1	0.28317	3.88	0.00105	5.73	1.48
105-AN-1	0.28317	3.88	0.00105	5.78	1.49
101-SY-1	0.42570	5.83	0.00102	5.60	0.96
101-SY-2	0.42570	5.83	0.00104	5.69	0.98

PST87-3225-S

9313022.1750

9313022.1749

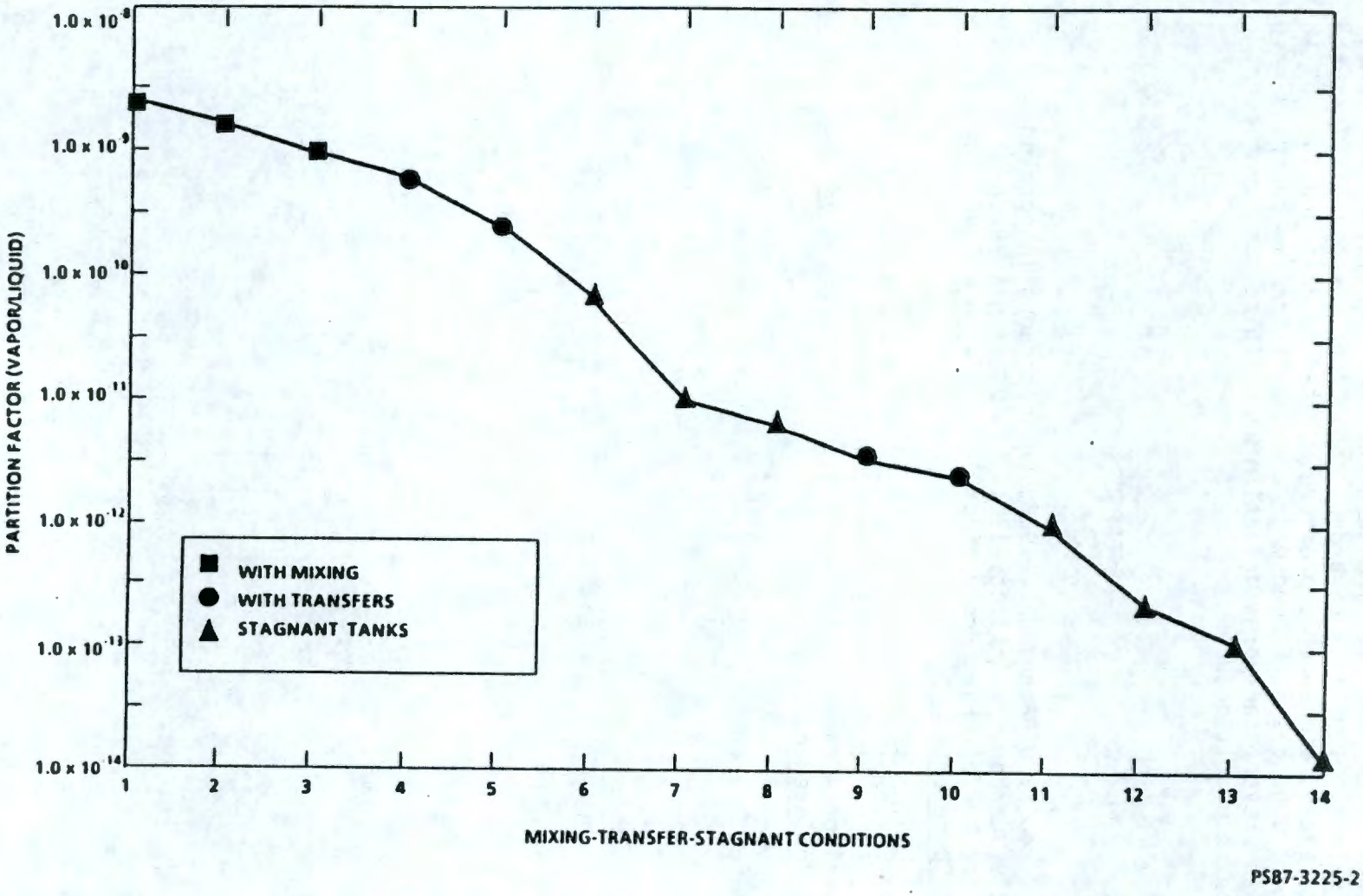

Figure 3. Cesium-137 Partition Factors.

Stagnant tanks, as represented by data points 6, 7, 8, 11, 12, 13, and 14, are generally the lowest PFs. These lower PFs are the most desirable during tank operation. Safety analyses indicate that failure of the tank ventilation system may create a potentially hazardous condition if vacuum is lost and tank pressures exceed atmospheric pressure. Based on data provided by this study, equipment upgrades have been made and administrative controls have been revised to prevent tank pressurization or minimize the duration of tank pressurization should it occur.

REFERENCES

McManus, G. J. and S. J. Fernandez, 1986, "A Continuous Integrated Iodine-129, Carbon-14, Antimony-125, Tritium, and Krypton-85 Off-Gas Monitor for Nuclear Fuel Processing Plants," 19th DOE/NRC Nuclear Air Cleaning Conference.

Mandler, J. W., J. W. Tkachyk, J. T. Case, J. H. McDaniel, and H. K. Peterson, 1986, "In-situ Measurement of Efficiency of Silver Zeolite for Removal of Airborne Radioiodine Species," 19th DOE/NRC Nuclear Air Cleaning Conference.

9311027 220193
EAT