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MEASUREMENTS OF THE PASSIVE VENTILATION RATES OF HIGH-LEVEL RADIOACTIVE WASTE TANKS USING TRACER GASES

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Abstract

Passive ventilation rates of high-level radioactive waste tanks at the Hanford Site in Richland, Washington have been determined using a tracer gas method. Helium and/or sulfur hexafluoride (SF₆) were injected into the headspaces of 14 tanks, and their depletion with time was observed. Helium was found to be a more reliable tracer under the highly radioactive/high-alkaline conditions found in Hanford tanks. Ventilation rates ranged from 1.7 to 114 m³/h. The major factor in predicting ventilation rates appears to be the availability of airflow pathways. In this study, ventilation was shown to occur via connecting lines between tanks. Barometric pressure fluctuations appear to account for only a minor portion of the ventilation rates. These measurements give useful information about rates of hydrogen release from the tanks and drying rates of organic salt-nitrate salt mixtures.

I. Introduction

Most single-shell, high-level radioactive waste tanks at the Hanford Site in Richland, Washington are passively ventilated with the atmosphere to prevent accumulation of gases released by the waste and pressurization of the tanks. Passive ventilation rates are needed to calculate parameters involved in two key safety issues associated with the rates of flammable gas production and accumulation and the rates at which organic salt-nitrate salt mixtures dry out.

In the past, simple models have been suggested to bound the ventilation rate^(1,2) because direct measurement of passive ventilation rates using mass flow meters is not feasible. Ventilation occurs via multiple pathways to the atmosphere (filtered breather riser and unsealed tank risers and pits) and through underground connections to other tanks, junction boxes, and inactive ventilation systems. More complex numerical models have been developed to estimate passive ventilation rates, but these depend on physical parameters that have not been measured. Ventilation rates for a small number of flammable gas-generating tanks have been measured indirectly by monitoring the decrease in hydrogen concentration after a significant amount of trapped hydrogen was released by the waste.^(3,4) Although this method provides credible results, it cannot be applied to tanks that do not trap and release significant amounts of hydrogen or tanks without hydrogen monitoring instrumentation. Thus a different method was needed for determining ventilation rates. One possibility was suggested by the extensively documented use of tracer gases to monitor rates of gaseous emissions from different types of sources.

In this paper, the tracer gas method is applied to Hanford tanks to provide a measurement of the rate at which gases are removed from the tanks by ventilation, which, in turn, can be used to calculate the ventilation rate. The unique conditions in the Hanford Site tanks are discussed in light of the potential problems associated with using tracer gases in the tanks. Actual ventilation rate measurements are presented for 15 tanks.

II. Experimental

The tracer gas method for measuring tank ventilation rates requires injecting a tracer gas into the tank headspace and periodically collecting and analyzing headspace samples to determine the tracer gas concentration as a function of time. If key assumptions are satisfied, the tracer gas concentration will decrease logarithmically with time, and the ventilation rate of the tank can be calculated from the headspace volume and the rate at which the tracer gas concentration decreases.

Selection of Tracer Gases

Both helium (He) and sulphur hexafluoride (SF₆) were used as tracer gases in this study. These gases are commonly used as tracers because of their chemical stability, low solubility in water,^(5,6) and lack of toxicity. Both are very difficult to ionize, and He is not subject to radiolysis. Both are also easily sampled and measured. An advantage of SF₆ is that it is purely anthropogenic, so the natural background levels are essentially zero,⁽⁷⁾ in contrast to He, which is found at levels of ~5 ppmv in air.

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Tracer Gas Injection and Sampling

Tracer gases were introduced into the tank headspaces via tubing that passed through a tank riser to a location approximately halfway between the waste surface and the bottom of the riser. Helium was supplied from a commercial high-pressure gas cylinder. The SF₆ was supplied by the laboratory in flow-through cylinders that were purged with the He being injected. After the tracer gas was introduced, the tank headspaces were allowed to stabilize for one day before baseline (time zero) samples were collected.

Headspace samples were collected in evacuated SUMMA™ passivated stainless steel canisters from the mid-elevation of the headspace via stainless steel or PFA Teflon tubing that had been previously purged with headspace air. Samples were collected in duplicate or triplicate to examine sampling precision and provide spare samples if needed. Headspace samples collected before tracer gas was injected gas established background levels of tracer gas in each tank headspace.

Samples were collected one day and seven days after tracer gas injection to establish an approximate ventilation rate for each tank. If this initial ventilation rate was high, the sampling schedule was accelerated to complete sampling before the tracer gas concentrations dropped below measurable levels. If the ventilation rate of a tank was expected to be very high (as for Tanks AX-101 and C-104), the sampling schedule was accelerated to obtain one-day, two-day, etc., samples.

Helium Analysis

Helium was analyzed by a micro gas chromatograph (GC) equipped with an integral thermal conductivity detector (TCD) (P200H Micro GC from MTI Analytical Instruments, Inc.). A gas sampling loop was used to introduce the gas onto a 4 m x 0.32 mm ID 5Å molecular sieve column. The oven was held isothermal at 35°C for the one-minute analysis time. Under those conditions, He and hydrogen eluted as well-separated peaks in the 11-to 17-second time range. A small correction was included in all calculations for an unresolved neon component found in ambient air.

Calibration standards had nominal concentrations of 2000, 1000, 500, 200, 50, 10, and 2 ppmv. The instrument exhibited a very high degree of linearity for components over that range. Sensitivity requirements were demonstrated daily through repeat analysis of the low level standard (2 ppmv). Each daily calibration was also verified using an independently prepared standard at the 100 ppmv level. Precision of the method was found to be 1–3% in most cases. Overall accuracy requirements were set at 10%; however, most available data showed significantly better performance. The instrument detection limit was estimated to be 1 ppmv based on the sensitivity of the integration algorithm, and the estimated quantitation limit was 2 ppmv based on the use of a lower-level standard.

Several samples were analyzed for He by a high-sensitivity mass spectrometer (MS) equipped with both a Faraday cup detector (operated in the current mode and linear to 1 part in 100,000) and a secondary electron multiplier (SEM) that was used for ion counting. The MS system used has a replicate precision of about 2% relative standard deviation (RSD) for He. The instrument detection limit is typically less than 1 ppmv.

SF₆ Analysis

SF₆ was analyzed by a GC with an electron capture detector (ECD). SUMMA™ canister samples were first pressurized with ultra-high-purity nitrogen to exactly double the initial filling pressure. A 2 mL gas sample loop was used to introduce the gas onto a 2.4 m molecular sieve (HP5Å 45/60 mesh) GC column. The oven temperature was held isothermal at 40°C for the eight-minute analysis.

Commercially purchased calibration standards with concentrations of SF₆ certified to 5% accuracy were used to calibrate the GC-ECD. Three calibration ranges were needed to minimize the effects of the nonlinear response of the ECD to this strongly electron-capturing compound.⁽⁸⁾ The three calibration curves ranged from 0.030 to 1.0 ppbv (linear regression based on 3 points), 1 to 20 ppbv (quadratic fit based on 4 points), and 20 to 160 ppbv (cubic fit based on 5 points). The GC-ECD system used for SF₆ had an RSD of less than 8% for each of the three calibration ranges. The estimated instrument detection limit was 0.006 ppbv, and the estimated quantitation limit was 0.032 ppbv.

Ventilation Rate Calculations

The tracer gas is assumed to be inert and insoluble in the waste, so that its only depletion mechanism is ventilation. The tracer is also assumed to be absent from, or at a relatively low constant level in, the ambient air introduced to the headspace.

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Assuming also that the tracer is uniformly distributed in the headspace, the decrease in tracer concentration with time is proportional to its concentration:

$$\frac{dC}{dt} = -\frac{v}{V} C \quad (1)$$

where C is the concentration of the tracer, v is the volumetric ventilation rate, V is the headspace volume, and t is time. This equation can be solved for the ventilation rate, v , between any two sample events:

$$v = \frac{V}{(t_i - t_j)} \ln \left(\frac{C_j}{C_i} \right) \quad (2)$$

where C_i and C_j are the concentrations of the tracer gas at any two different times t_i and t_j , respectively.

When concentration is plotted against time on a log-linear scale, the slope of the resulting line is proportional to the estimated ventilation rate. Figure 1 illustrates this with data from Tank U-111. Nonlinearity of more than two points may be caused by sampling or analytical measurement errors, or by real differences in the average ventilation rate.

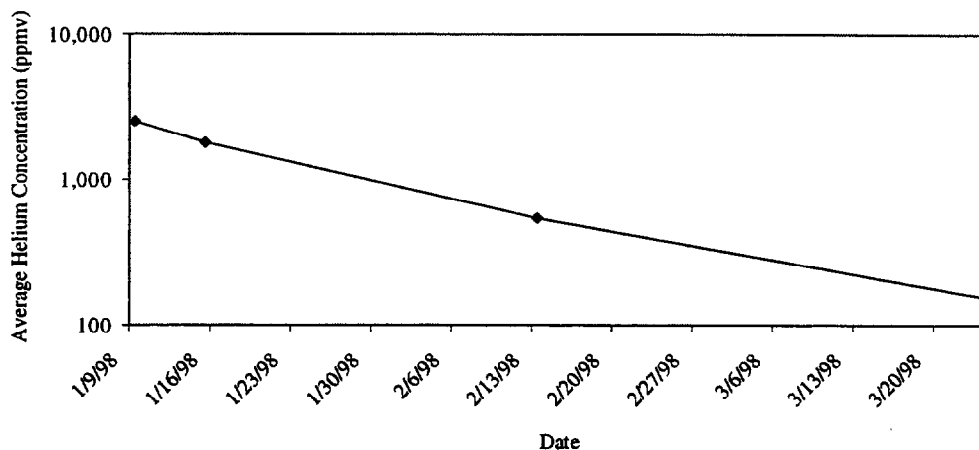


Figure 1 Logarithmic plot of helium tracer concentration in Tank U-111.

III. Results and Discussion

A key assumption in using the tracer gas method to calculate ventilation rates (as in equation 2) is that the tracer gas is essentially uniformly distributed within the tank headspace. This condition is also required to ensure that representative samples can be collected via any available tank riser. Two aspects of this assumption are that the injected tracer gas be well mixed within the headspace before its baseline concentration is measured, and that fresh air brought into the headspace via the ventilation system be mixed within the headspace in a relatively short time period (i.e., the concentration of tracer in the air leaving the tank must be approximately the average concentration in the headspace). These aspects of mixing were considered by Huckaby et al.⁽⁹⁾ and found to be satisfied for the eight passively ventilated tanks they studied; tracer gases were essentially at their final concentrations at the sampling point within one hour after injection was completed.

Another key assumption in the method is that the only significant change in tracer gas concentration is loss via ventilation. The tracer gas must therefore be chemically and radiolytically inert and insoluble in the wastes. The validity of this assumption for the two tracer gases, He and SF₆, was investigated.⁽⁹⁾ It was found that the SF₆ loss rate was higher than that for He in several, but not all, of the waste tanks (Table 1), even when He and SF₆ were injected and sampled on the same dates. They attributed the higher loss of SF₆ to radiolytic decomposition caused by beta radiation from fission product-derived beta emitters such as H³, Sr⁹⁰, Tc⁹⁹, and Cs¹³⁷. This is consistent with literature reports of the use of SF₆ in radiation studies as an electron scavenger and its reactivity under these conditions.^(10,11,12) Thus, in the current study, He has been the primary tracer gas for quantitative vent rate measurements in each tank, and SF₆ has been employed only when He data were not available and for qualitative assessment of intertank air exchange in Tank C-104.

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Tank Ventilation Rates

Ventilation rates were measured in 14 tanks at different times and for different durations. Averaged tracer gas concentrations are available for each sampling event and ventilation rates between each consecutive pair of sampling events.^(9,13) The average ventilation rate for each tank, presented in Table 2, was calculated from the initial and final tracer concentrations and is not an average of the consecutive ventilation rates. Ventilation rate tolerances given in Table 2 represent the 95% confidence limits. Passive ventilation rates were found to be significantly different for individual tanks, ranging from 1.7 m³/h during a 28-day period in Tank C-107 to 114 m³/h during a four-day period in Tank C-104. Ventilation rates were also observed to exhibit short- and long-term variations.

The initial hypothesis, that barometric pressure fluctuations caused by diurnal variations or weather system phenomena were the cause of passive ventilation of tanks, would not account for such large differences among the tanks. In addition, calculations using the barometric pressure changes predicted ventilation rates of 0.2 to 0.9 m³/h, much lower than the measured rates. Thus, alternative reasons were explored for these large tank-to-tank variations by examining individual tanks more closely. Specifically, variations in available airflow pathways were thought to be a major factor and tank diagrams were examined to determine these pathways. In addition, temporal variations were examined in Tank U-103 to determine the magnitude of variability within a tank. More detailed information about selected tanks follows.

Tanks C-107, S-102, TX-104, U-105, U-106, and U-111

These tanks are each part of a series, connected via a 7.6-cm diameter, 7.6-m long underground pipe to one or two adjacent tanks. Their headspaces are allowed to breathe with the atmosphere via a high-efficiency particulate air (HEPA) filter. No other open connections to tanks or the atmosphere were identified. Visual inspection of the pit covers and the above-ground portions of risers in January 1998 indicated no significant pathways for air leakage. The relatively low ventilation rates are consistent with having only one or two connections to other tanks and one outlet to the atmosphere.

Tanks A-101, AX-102, AX-103, and BY-105

The tanks in AX-farm are connected via an underground ventilation system that was originally part of the AX-farm exhaust system. The ventilation system has been isolated from the exhausters, but the 51-cm and 61-cm-diameter air ducts connecting the headspaces of the tanks in the farm remain open. A similar ventilation system exists in A-farm. It is thought that the high rates observed for these tanks were associated with these large air ducts. The high ventilation rate of Tank BY-105 is thought to be associated with air leakage around its large risers; it has five 107-cm-diameter risers in covered concrete pits above the tank, and it is known that the blind flange covers of such risers are often not bolted down.

Tank C-104

Tank C-104 is the first tank in a three-tank cascade. Like most of the other tanks studied, it is connected via a 7.6-cm diameter, 7.6-m long underground pipe to an adjacent tank. However, in this case, the adjacent tank is the actively ventilated Tank C-105. A similar cascade line connects Tank C-105 to Tank C-106, which is also actively ventilated. Vapor samples obtained in 1994 from the headspace of Tank C-104 and the exhaust riser of Tank C-105 indicated that Tank C-104 was being actively ventilated via the cascade line,⁽¹⁴⁾ and this has been confirmed by the tracer study.

Table 1 Comparison of ventilation rates using He and SF₆ as tracers.

Tank	Ventilation rate based on He	Ventilation rate based on SF ₆
AX-103	32 ± 2	100 ± 6
C-107	1.7 ± 0.4	15 ± 1
S-102	3.4 ± 0.8	3.8 ± 0.3
U-103	3.9 ± 0.2	3.0 ± 0.2

Table 2 Summary of average ventilation rates by tank.

Tank	Number of days	Ventilation rate (m ³ /h)
A-101	6	18 ± 1
AX-101	4	Not determined
AX-102	11	28 ± 3
AX-103	6	32 ± 2
BY-105	6	35 ± 5
C-104	4	114 ± 9
C-107	28	1.7 ± 0.4
S-102	140	3.4 ± 0.8
TX-104	30	5.9 ± 0.5
U-102	75	3.5 ± 0.2
U-103	145	4.0 ± 0.2
U-105	28	8.5 ± 0.5
U-106	75	2.2 ± 0.1
U-111	75	3.2 ± 0.2

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Approximately 5.7-m³ of He and 1.3-L of SF₆ were injected into the headspace of Tank C-104 on April 13, 1998. Samples were collected from the headspace on April 14, 16, and 17, 1998. Average ventilation rates, calculated from measured He concentrations and equation (2), ranged from 113 to 115 m³/h. The ventilation rates for the two periods considered are essentially identical and clearly indicate that the ventilation rate of Tank C-104 at that time was much higher than those of other passively ventilated tanks.

Samples were also collected from the ventilation risers of Tanks C-105 and C-106 on April 14, and the ventilation rates of both tanks were measured by another method on the same day. Samples from Tank C-105 ventilation riser had an average He concentration of 112 ppmv, well above background He levels, and qualitatively consistent with the measured airflows of Tanks C-104 and C-105 and the premise that Tank C-104 is venting via the cascade line. Samples from the ventilation riser of Tank C-106 did not have elevated He concentrations but did have detectable levels of SF₆, indicating that the cascade line between Tanks C-105 and C-106 is not plugged.

Figure 2 depicts the Tank C-104, C-105, and C-106 cascade and labels the airflows of the system. The ventilation rates of Tanks C-105 and C-106 were reported to be $v_4 = 15 \text{ m}^3/\text{min}$ (900 m³/h) and $v_7 = 83 \text{ m}^3/\text{min}$ (4980 m³/h), respectively, on April 14. To estimate the ventilation rate of Tank C-104 when the ventilation rate of Tank C-105 is different, it may be assumed that the ratio of the Tank C-104 and C-105 ventilation rates is constant:

$$\frac{v_2}{v_4} = \frac{1.9 \text{ m}^3/\text{h}}{15 \text{ m}^3/\text{h}} = 0.13 \quad (3)$$

This assumes that both v_2 and v_4 are turbulent flows and behave similarly to changes in system pressure differentials. Given that SF₆ was detected in Tank C-106 exhaust, air was apparently flowing from Tank C-105 to C-106 (i.e., $v_5 > 0$), but since the magnitude of this airflow was not determined, it is assumed to not appreciably affect the relationship of equation (3).

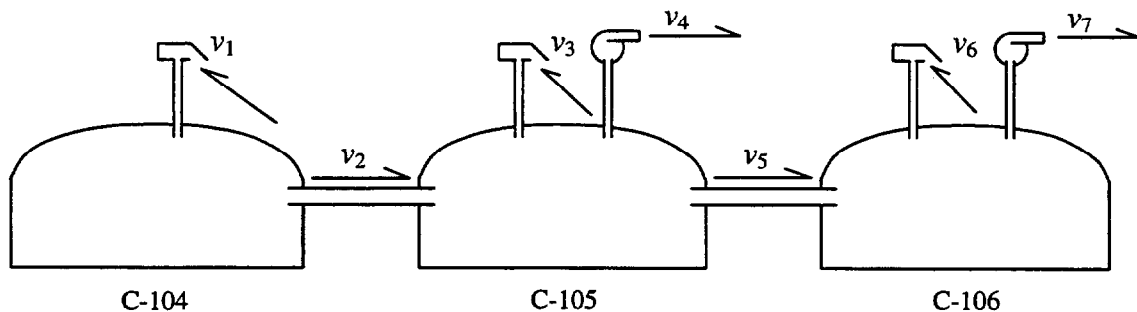


Figure 2 Schematic of airflow in the Tank C-104, C-105, and C-106 cascade.

Tanks U-102 and U-103

Tanks U-102 and U-103 have similar airflow pathways to the other U-farm tanks discussed above. However, these are the only connected tanks that have been studied (i.e., sampled for tracer gases) simultaneously. The use of equation (2) assumes incoming ventilation air has a constant, known concentration of tracer. That assumption is valid if the incoming air comes directly from the atmosphere, but it may not be valid if the ventilation of the tank involves a second tank, with a tracer gas concentration different from that in ambient air.

Helium tracer was introduced first into Tank U-103 in February, July, and November of 1997. No He was injected into U-102 until January 8, 1998. Table 3 lists the average He concentrations in samples from these two tanks from January 8 through March 24, 1998. Note that the January 8, 1998, He concentration in Tank U-102 was significantly higher than the 5 ppmv background level, despite the fact that He had not been injected directly into this tank previously. This observation clearly indicates that air from Tank U-103 had flowed into Tank U-102 and that the cascade line between the two tanks was not plugged. Airflow in the other direction (from Tank U-102 to Tank U-103) was also shown to occur after the January 8 injection of He into Tank U-102 by the temporary rise in He concentrations in Tank U-103. This demonstrates that ventilation can and does occur via the cascade lines between tanks.

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Figure 2 depicts the connections and tracer-containing airflows of Tanks U-102 and U-103. The airflows indicated are non-negative (either zero or positive). Though the airflow between Tanks U-102 and U-103 can evidently change direction and may do so frequently, the tracer method as applied to these tanks provides only enough data for an examination of average flow rates and limiting cases. Assuming the tracer is uniformly distributed (well-mixed) in each tank headspace, the concentrations of tracer in U-102, C_{102} , and in U-103, C_{103} , are described by the following coupled differential equations:

Table 3 Helium concentrations measured in Tanks U-102 and U-103.

Date	Average helium concentration (ppmv)	
	Tank U-102	Tank U-103
11/17/97	No tracer injected	5.8 m ³ of He injected
01/08/98	195	119
01/08/98	5.7 m ³ of He injected	No tracer injected
01/09/98	2,085	140
01/15/98	1,637	215
01/13/98	265	135
01/24/98	71	38

$$\frac{dC_{102}}{dt} = -\frac{v_1 + v_3}{V_{102}} C_{102} + \frac{v_4}{V_{102}} C_{103} \quad (4)$$

$$\frac{dC_{103}}{dt} = -\frac{v_2 + v_4}{V_{103}} C_{103} + \frac{v_3}{V_{103}} C_{102} \quad (5)$$

where the average volumetric flow rates v_i are as indicated in Figure 3, and V_{102} and V_{103} are the headspace volumes of Tanks U-102 and U-103, respectively. Solution of this equation system yields

$$C_{102} = C_{102}^0 \exp\left(-\frac{v_1 + v_3}{V_{102}} t\right) + C_{103}^0 \frac{\frac{v_4}{V_{102}} \left(\exp\left(-\frac{v_2 + v_4}{V_{103}} t\right) - \exp\left(-\frac{v_1 + v_3}{V_{102}} t\right) \right)}{\frac{v_1 + v_3}{V_{102}} - \frac{v_2 + v_4}{V_{103}}} \quad (6)$$

$$C_{103} = C_{103}^0 \exp\left(-\frac{v_2 + v_4}{V_{103}} t\right) + C_{102}^0 \frac{\frac{v_3}{V_{103}} \left(\exp\left(-\frac{v_1 + v_3}{V_{102}} t\right) - \exp\left(-\frac{v_2 + v_4}{V_{103}} t\right) \right)}{\frac{v_2 + v_4}{V_{103}} - \frac{v_1 + v_3}{V_{102}}} \quad (7)$$

where C_{102}^0 and C_{103}^0 are the concentrations of tracer in Tanks U-102 and U-103, respectively, at $t = 0$. In this formulation, either v_3 or v_4 is zero. Note that there are only two equations and three unknown flow rates. If there is airflow from Tank U-103 into U-102, then v_1 , v_2 , and v_4 are unknown (and v_3 is set to zero); and if there is airflow from Tank U-102 into Tank U-103, then v_1 , v_2 , and v_3 are unknown (and v_4 is set to zero). The system is consequently indeterminate, and an additional constraint must be imposed to obtain a solution. Two constraints were considered, one that maximizes the ventilation rate of a given tank (either $v_1 + v_3 = \text{maximum}$ or $v_2 + v_4 = \text{maximum}$), and one that minimizes the ventilation

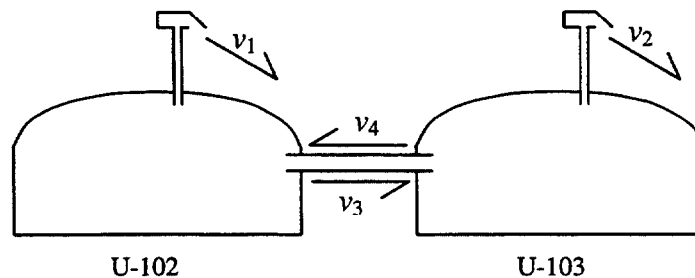


Figure 3 Schematic of airflow in the Tank U-102 and U-103 cascade.

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rate of a given tank (either $v_1 + v_3 = \text{minimum}$ or $v_2 + v_4 = \text{minimum}$). The system of equations (5), (6), and a constraint were solved numerically using Microsoft Excel and based on the assumption that the He concentration in Tank U-102 on November 18, 1997 was equal to that in the ambient air. The calculated minimum and maximum ventilation rates ranged from 0.4 to 6.0 m³/h for U-102 and from 0.5 to 24.7 m³/h for U-103. Though useful as bounding estimates, these maximum and minimum ventilation rates generally require improbable conditions. For example, the maximum ventilation rate of Tank U-103 occurs when all the exhaust air from Tank U-102 goes through the cascade line into Tank U-103.

Tank AX-101

Tank AX-101 is one of four tanks in AX-farm. As noted above, the AX tanks are connected with a system of large air ducts that remain open. Thus, a high ventilation rate was expected for this tank. Approximately 5.4-m³ of He was injected into Tank AX-101 headspace on March 5, 1998, to provide a calculated initial tracer concentration of about 1,100 ppmv. Samples were collected one and four days after tracer injection, with unexpected results, and the study was aborted.

Table 4 lists sample collection dates and times, and average analytical results for He and hydrogen measured in the Tank AX-101 samples. As shown in Table 4, the three samples collected on March 6 contained significantly different concentrations of He, even though they were collected only minutes apart. No sampling problems had been noted, and the consistency of hydrogen concentrations measured in the same samples suggests that no accidental dilution with air (which would have affected the hydrogen concentration) or partial filling of the samples occurred. Analytical results from replicate analyses were highly reproducible, and there were no indications of any analytical

Table 4 Measured helium and hydrogen concentrations in Tank AX-101.

Sample date and time	Helium (ppmv)	Hydrogen (ppmv)
03/05/98 09:53 AM	6.7	6.4
03/05/98 09:59 AM	6.5	6.3
03/05/98 10:05 AM	6.7	6.4
03/05/98 10:16 AM	He injection started	--
03/05/98 10:46 AM	He injection stopped	--
03/06/98 09:48 AM	218.8	7.1
03/06/98 09:54 AM	112.9	7.1
03/06/98 10:00 AM	83.3	7.4
03/09/98 10:08 AM	7.7	5.6
03/09/98 10:15 AM	6.0	6.7
03/09/98 10:21 AM	7.2	6.5

problems. A possible explanation for the inconsistent He results is that, because the samples were collected from a headspace location near the opening of the underground ventilation system, they may have been affected by a transient inflow of air from the other AX-farm tanks.

Samples collected four days after sample injection, on March 9, were found to have essentially ambient air levels of He. These samples may also have been affected by an inflow of air from the AX-farm ventilation system, but it is also possible that the He level in the headspace had actually been reduced to this low level via a moderate ventilation rate combined with poor mixing of the tracer. If, for example, the ventilation air introduced to Tank AX-101 was cooler than the bulk headspace, it would tend to displace the tracer-containing air in the headspace without proper mixing. Ventilation air leaving the headspace would be correspondingly concentrated in the tracer and cause the ventilation rate calculated from equation (2) to appear higher than it is. The tracer method was deemed to have failed on Tank AX-101, due possibly to an inappropriate sampling location and/or to a nonuniform distribution of the tracer in the headspace.

Temporal Variations in Ventilation Rates

Though local meteorology is thought to have a strong influence on variations in the ventilation rates of the passively ventilated tanks, no definitive correlations were established between the average ventilation rates measured with tracer studies and data from the Hanford Meteorological Station. Furthermore, a year-long study of Tank U-103 indicated only a moderate correlation between measured ventilation rates and seasonal effects.^(9,13) The correlation suggests that temperature differences between the headspace and atmosphere do play a role in passive ventilation (higher rates are correlated with larger temperature differences), but apparently their role in this tank is weak.

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Using Ventilation Rates to Estimate Hydrogen Release Rates

Ventilation rates can be used to estimate the rate at which hydrogen is released by the waste when hydrogen concentrations are measured in the tracer samples. This has been done using the following equation:

$$R_{ij} = v_{ij} \left(\frac{C_i + C_j}{2} \right) + \frac{V(C_j - C_i)}{\Delta t} \quad (8)$$

where R_{ij} is the release rate, v_{ij} is the calculated ventilation rate for the period between the i th and j th samplings, C_i and C_j are the concentrations of hydrogen associated with the i th and j th samplings, V is the headspace volume, and Δt is the time between samplings. Conversion of the hydrogen concentration, which is reported in ppmv, to a mass concentration, is done using the ideal gas law and average headspace temperatures. Average headspace temperatures were calculated from readings of thermocouples in the headspaces.

Average hydrogen concentrations and release rates for several tanks are found in Table 5. The average hydrogen release rate for the entire study is a weighted average of the individual periods during which hydrogen concentrations were measured. For all the tanks tabulated, the consistency of headspace hydrogen concentrations measured on different dates supports the observed consistency in the ventilation rates for the time periods studied.

Table 5 Calculated hydrogen release rates.

Tank	Time period	Headspace temp (°C)	H ₂ concentration (ppmv)	H ₂ release rate (g/day)
C-104	04/14/98 – 04/17/98	27.7	36 ± 5	7.5
TX-104	01/14/98 – 02/12/98	18.1	17.0 ± 1.2	0.17
U-102	01/09/98 – 03/24/98	21.8	511 ± 65	3.7
U-103	11/18/97 – 01/08/98	22.4	611 ± 59	51
U-106	01/09/98-3/24/98	21.6	444 ± 31	2.2
U-111	01/09/98 – 03/24/98	20.7	389 ± 39	2.4

For all the tanks tabulated, the consistency of headspace hydrogen concentrations measured on different dates supports the observed consistency in the ventilation rates for the time periods studied.

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