

METHOD OF ESTIMATING MAXIMUM VOC CONCENTRATION IN VOID VOLUME OF VENTED WASTE DRUMS USING LIMITED SAMPLING DATA - APPLICATION IN TRANSURANIC WASTE DRUMS

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ABSTRACT: A test program has been conducted at the Idaho National Engineering Laboratory to demonstrate that the concentration of volatile organic compounds (VOCs) within the innermost layer of confinement in a vented waste drum can be estimated using a model incorporating diffusion and permeation transport principles as well as limited waste drum sampling data. The model consists of a series of material balance equations describing steady-state VOC transport from each distinct void volume in the drum. The primary model input is the measured drum headspace VOC concentration. Model parameters are determined or estimated based on available process knowledge. The model effectiveness in estimating VOC concentration in the headspace of the innermost layer of confinement was examined for vented waste drums containing different waste types and configurations. This paper summarizes the experimental measurements and model predictions in vented transuranic waste drums containing solidified sludges and solid waste.

INTRODUCTION

In the final conditional Waste Isolation Pilot Plant (WIPP) No-Migration Determination, the Environmental Protection Agency (EPA) imposed several waste characterization requirements on the U. S. Department of Energy (DOE) specific to sampling and analyzing the headspace gases in waste containers to be emplaced in the WIPP facility.¹ The objectives of these requirements are to demonstrate that flammable gas mixtures or mixtures of gases that could become flammable when mixed with air do not exist within a container; to demonstrate that the waste received at the WIPP is comparable to that described in the DOE No-Migration Variance Petition; and to demonstrate that hazardous constituents will not exceed concentrations above health-based limits established by the EPA at the WIPP repository unit boundary. In order to comply with these requirements, the DOE must sample and analyze all layers of confinement (drum headspace, large polymer bag headspace, and innermost layer of confinement headspace) to ensure that data are representative of the volatile organic compounds (VOCs) within the entire void space within the waste container. The EPA expects that all layers of confinement in a container will have to be sampled until the DOE can demonstrate to the EPA, based on the data collected, that sampling of all layers is either unnecessary or can be safely reduced.

A test program has been conducted at the Idaho National Engineering Laboratory (INEL) to demonstrate that the concentration of VOCs within the innermost layer of confinement in a vented waste drum can be estimated using a model incorporating diffusion and permeation transport principles as well as limited waste drum sampling data.

VOC TRANSPORT MODEL

A transport model was developed to estimate the VOC concentration in void volumes within a vented drum containing waste contaminated with or containing VOCs. Model parameters are defined from knowledge of the waste drum configuration. A waste drum consists of a vented drum with a rigid drum liner that contains the waste inside one or two large polymer bags. The waste may have been placed directly in the innermost

large bag (solidified sludges) or wrapped in one or more layers of smaller polymer bags, which were then placed inside the innermost large polymer bag (solid waste). A small opening in the drum liner lid allows gas and vapor transport between the drum liner and drum headspaces. The waste drum configuration includes the type of filter vent in the drum lid, the dimensions of the opening in the drum liner lid, and the thickness of polymer bags surrounding the waste. The model, consisting of a series of material balance equations describing steady-state VOC transport from each distinct void volume in the drum, is presented in this section.

The primary mechanisms for steady-state gas transport across a polymer boundary are permeation across the polymer and diffusion across an opening in a layer of confinement. The steady-state gas transport via permeation across a polymer film is defined as

$$r_p = \left[\frac{A_0 \rho A_p P}{x_p} \right] c \Delta y_p = K_p \Delta y_p \quad (1)$$

where

- r_p = gas permeation rate, mol s⁻¹
- A_0 = correction factor for temperature and pressure relative to standard temperature and pressure (STP)
- P = gas pressure, Pa
- ρ = gas permeability coefficient, m³ (STP) m⁻¹ Pa⁻¹ s⁻¹
- A_p = permeable surface area of polymer film, m²
- x_p = polymer boundary thickness, m
- c = total gas concentration, mol
- Δy_p = gas mole fraction difference across polymer film
- K_p = gas permeation characteristic, 44.6 $\rho A_p P x_p^{-1}$, mol s⁻¹.

The steady-state gas transport across an opening in a layer of confinement via gas diffusion is defined as

$$r_d = \left[\frac{D A_d}{x_d} \right] c \Delta y_p = K_d \Delta y_p \quad (2)$$

where

- r_d = gas diffusion rate, mol s⁻¹
- D = gas diffusivity in air, m² s⁻¹
- A_d = cross-sectional area of opening, m²
- x_d = diffusional length across opening, m
- K_d = gas diffusion characteristic, mol s⁻¹.

In the case of a sludge waste drum, the large polymer bag immediately surrounding the waste is the innermost layer of confinement and the headspace surrounding the waste is referred to as the first void volume. The drum liner headspace not included in the large bags and the drum headspace outside the drum liner are the second and third void volumes, respectively. The VOC transport rate from the innermost layer of confinement, r , is defined as

$$r = K_p (y_1 - y_2) \quad (3)$$

where y_i is the VOC mole fraction in the i^{th} void volume. The VOC transport rate from the drum liner is defined as

$$r = K_d (y_2 - y_3) \quad (4)$$

The VOC transport rate from the drum headspace across the drum filter vent is defined as

$$r = D_{VOC}^* (y_3 - y_\infty) \quad (5)$$

where

$$\begin{aligned} D_{VOC}^* &= \text{drum filter vent VOC diffusion characteristic, mol s}^{-1} \\ y_\infty &= \text{VOC mole fraction outside waste drum.} \end{aligned}$$

The values of K_p and K_d are calculated based on process knowledge. The VOC mole fraction in the drum headspace, y_3 , is determined from analysis of gas samples collected below the filter vent. Combining Equations (3) through (5), the VOC mole fraction within the innermost layer of confinement can be defined directly in terms of the measured VOC mole fraction in the drum headspace

$$y_1 = y_3 + D_{VOC}^* y_3 \left[\frac{1}{K_p} + \frac{1}{K_d} \right] \quad (6)$$

Most solid waste drums contain waste packaged in one or more layers of small polymer bags. These smaller bags are located inside a larger polymer bag. The polymer bag immediately surrounding the waste is the innermost layer of confinement and the headspace inside this layer of confinement is referred to as the first void volume. Large bag, drum liner, and drum headspaces are referred to as the second, third, and fourth void volumes, respectively. The VOC transport rate from the innermost layer of confinement of the i^{th} waste package wrapped in one or more polymer bags is defined by the equation

$$r_i = K_{p,1,i} (y_{1,i} - y_2) \quad (7)$$

where

$$\begin{aligned} K_{p,1,i} &= \text{VOC permeation characteristic of first layer of confinement surrounding the } i^{\text{th}} \text{ waste} \\ &\quad \text{package, mol s}^{-1} \\ y_{1,i} &= \text{VOC mole fraction in headspace of } i^{\text{th}} \text{ waste package.} \end{aligned}$$

The total VOC transport rate exiting the waste packages and entering the large bag headspace is equal to the sum of individual VOC transport rates from each waste package

$$r = \sum_{i=1}^N r_i \quad (8)$$

where N is the total number of small bags inside the drum. The maximum model estimate of the VOC mole fraction in the innermost layer of confinement results if all VOC-contaminated waste is assumed to exist in

one bag or $N = 1$. The VOC transport rate across the large polymer bags containing the waste packages is defined as

$$r = K_{p,2} (y_2 - y_3) \quad (9)$$

The VOC transport rate across the drum liner is defined as

$$r = K_d (y_3 - y_4) \quad (10)$$

The VOC transport rate from the drum headspace across the filter vent is defined as

$$r = D_{VOC}^* (y_4 - y_{\infty}) \quad (11)$$

Combining Equations (7) through (11), the VOC mole fraction within the innermost layer of confinement of a drum containing solid waste is defined in terms of the measured VOC mole fraction in the drum headspace

$$y_1 = y_4 + D_{VOC}^* y_4 \left[\frac{1}{K_{p,1}} + \frac{1}{K_{p,2}} + \frac{1}{K_d} \right] \quad (12)$$

MODEL ASSUMPTIONS

Some assumptions were based on an understanding of the thermodynamic and kinetic nature of VOC transport in the waste drum. Other assumptions were based on process knowledge of the system. In cases where process knowledge was lacking, conservative assumptions that would result in higher model estimates of VOC concentration were made. Some of the model assumptions are listed below.

1. An equilibrium exists between the VOC-contaminated waste and the vapor phase in the innermost layer of confinement.
2. Ideal gas behavior exists.
3. In waste drums containing solid waste, all VOC-contaminated waste is contained inside one waste package.
4. The VOC transport rates across all layers of confinement are equal and at steady state.
5. The primary mechanisms for VOC transport are permeation across the polymer bags and diffusion across the drum liner and drum filter vent.
6. A layer of confinement defined by multiple layers of polymer bags is considered a single polymer bag with a bag thickness equal to the sum of the bag thicknesses of the individual bags.
7. The VOC concentration throughout each void volume is uniform and is zero outside the waste drum.
8. All VOC properties and other model parameters remain constant.

MODEL PARAMETERS

The permeability coefficients for 8 VOCs across polyethylene were measured at the INEL using a mixed-component chromatographic detection method. The VOCs were carbon tetrachloride, dichloromethane, 1,1,1-trichloroethane (TCA), 1,1,2-trichloro-1,2,2-trifluoroethane (Freon-113), trichloroethylene (TCE), toluene, methanol, and p-xylene. When the VOC permeability across polyethylene was not known, it was calculated based on the estimated values of VOC diffusivity in polyethylene, D_{PE} , and VOC solubility in polyethylene, S .^{2,3}

$$\rho = D_{PE} S \quad (13)$$

The diffusivities of most VOCs in air at a given temperature and pressure were identified in the literature.⁴ In the case where diffusivity data were not identified, the VOC diffusivity in air was estimated.⁵ The diffusion characteristic for specific VOCs and hydrogen across different drum filter vents was measured in experiments performed at the INEL. The drum filter vent VOC diffusion characteristic, D_{VOC}^* , can be estimated using the measured or estimated VOC diffusivity in air and process knowledge of the hydrogen diffusion characteristic across the drum filter vent, $D_{H_2}^*$

$$D_{VOC}^* = \frac{D_{VOC-air}}{D_{H_2-air}} D_{H_2}^* \quad (14)$$

Waste drum model parameters were measured or estimated from available process knowledge. The large bag surface area in all drums was estimated to be 0.26 m^2 . This corresponds to the cross-sectional area of the drum liner lid. From process knowledge, two large polyethylene bags were assumed to be in each waste drum. Each bag was assumed to be $2.8 \times 10^{-4} \text{ m}$ thick.

In the sludge waste drums, the cross-sectional area of the opening in the drum liner lid is $5 \times 10^{-4} \text{ m}^2$. The diffusion length across the drum liner⁶ was estimated to be 0.014 m . The VOC diffusion characteristics across the drum filter vents ranged from 9×10^{-7} to $2 \times 10^{-6} \text{ mol s}^{-1}$. All waste drums were maintained at ambient room temperature and pressure. In the solid waste drums, waste was assumed to be wrapped inside two or three layers of polymer bags. Small polyethylene bags were assumed to be $1.3 \times 10^{-4} \text{ m}$ thick and PVC bags were assumed to be $2.8 \times 10^{-4} \text{ m}$ thick. All small bags were assumed to be polyethylene unless process knowledge indicated the possible use of a PVC bag. The cross-sectional area of the drum liner lid opening is $7.1 \times 10^{-5} \text{ m}^2$. The diffusion length across the drum liner lid was estimated to be 0.012 m . The measured VOC diffusion characteristics across the drum filter vents on these drums ranged from 2×10^{-7} to $6 \times 10^{-7} \text{ mol s}^{-1}$.

All model parameters, except the small bag surface area, can be estimated from process knowledge prior to characterization of waste drum contents. During waste characterization of solid waste drums, the operator estimated the bag shape (rectangular, triangular, elliptical, or cylindrical) of each bag gas sampled and measured the characteristic dimensions of the bag. Estimated bag surface areas ranged from 0.02 to 0.5 m^2 . Based on this information, a conservative assumption was made that the permeable surface area of all small polymer bags was 0.05 m^2 .

EXPERIMENTAL

The model effectiveness in estimating VOC concentration was examined for vented waste drums containing different waste types and configurations. Sludge waste drums contained a maximum of 2 layers of polymer

bags. The solid waste drums sampled had a maximum of 4 or 5 layers of polymer bags. Drum headspace samples were initially collected from waste drums that had been vented for a minimum of 8 weeks to identify drums with detectable VOC concentrations. Drums with low concentrations of total VOCs (less than 10 ppm) were not sampled further or used to test the accuracy of the VOC transport model.

Gas samples from 22 sludge waste drums were collected and analyzed at the Rocky Flats Environmental Technology Site. Drum headspace samples were collected by removing the top of the drum filter vent and inserting a sample needle into the carbon composite medium. After this sample was collected, the drum lid and drum liner lid were removed and a gas sample was collected from the innermost layer of confinement surrounding the waste sludge. The elapsed time for all samples to be collected from a drum was between 10 and 30 minutes.

Gas sampling and waste characterization of 42 waste drums containing solid waste were performed in the Argonne National Laboratory-West Hot Fuel Examination Facility Waste Characterization Chamber (WCC). The WCC gas sampling system is a semi-automated process. Operators were responsible for installing an evacuated canister on the sample manifold and inserting the sample needle in the drum or bag headspace. The operator activated the computer-controlled process which opened and then automatically closed the sample line upon sample collection. The complete gas sampling sequence took approximately 20 minutes. A drum headspace sample was collected by inserting the sample needle into the drum filter vent in the drum lid. After gas sample collection, the drum and drum liner lids were removed. A gas sample was collected from the headspace of the innermost layer of large polymer bags. The top of each large polymer liner bag was then removed to allow the removal of the smaller polymer bags. A gas sample from each separate bag was collected from the innermost layer of confinement that was not breached and contained at least 100 mL of void volume as decided by the operator. An effort was made to conduct the sampling of the drum headspace, the drum liner bag headspace, and the innermost layer of confinement of all small bags in one working day. In some cases, the drum and drum liner bag headspaces were sampled in one day and all small bags were sampled within the next 24 hours. In this case, the lids of the drum and drum liner were placed back on after gas sampling of the large bag and the drum liner bags were not opened until the second day. Visual characterization of waste consisted of estimating bag material and wall thickness, identifying bag shape, and measuring linear dimensions of each bag.

RESULTS

Model calculations were performed for two sets of VOCs. The first set consisted of VOCs in which most VOC-specific parameters, such as VOC permeability across the polyethylene bags and the drum filter vent VOC diffusion characteristic, were measured. Model calculations were performed for other VOCs using estimated values for VOC-specific parameters. In the case where duplicate measurements were made, the higher VOC concentration was used as model input. Model estimates of VOC concentration within the innermost bag headspace were compared to the maximum measured VOC concentration in the bag headspaces. The estimated or measured total VOC concentration was the sum of the individual estimated or measured VOC concentrations. In order to quantify the accuracy of the model results, the logarithm of the ratio of the predicted VOC concentration in the innermost layer of confinement, Y_{mod} , to the maximum VOC concentration measured in all bag headspaces, Y_{max} , was calculated

$$\psi = \log_{10} \left[\frac{Y_{mod}}{Y_{max}} \right] \quad (15)$$

In the case where the model estimate and measured VOC concentrations are identical, ψ equals zero. A statistical test was developed to test the null hypothesis, H_0 , that the mean of ψ , μ_ψ , is equal to zero and, therefore, the model is accurate. The alternative hypothesis, H_a , is that μ_ψ is not equal to zero

$$H_o: \mu_{\psi} = 0 \quad (16)$$

$$H_a: \mu_{\psi} \neq 0 \quad (17)$$

The VOC transport model is considered to be accurate if the confidence limits bound the case of $\psi = 0$. Otherwise, the model is said to have a positive or negative bias. This means that model estimates of the maximum VOC concentration in a specific population of waste drums are consistently greater or less than the maximum measured VOC concentration. The mean and standard deviation for ψ as well as the upper and lower 95% confidence limits for each waste drum configuration based on the maximum number of polymer bags and for each VOC in all waste drums are summarized in Table 1.

Table 1. Mean, standard deviation, and 95%-confidence limits of ψ for total VOC concentration in drums by waste configuration and single VOCs in all waste drums.

Variable	Number of drums	Mean	St. deviation	Lower limit	Upper limit
Two bag layers	21	0.078	0.073	0.040	0.111
Four bag layers	12	-0.014	0.176	-0.125	0.097
Five bag layers	24	0.087	0.199	0.003	0.171
CCl ₄ ^{a,b}	21	-0.014	0.371	-0.182	0.154
Methanol ^b	7	0.061	0.326	-0.241	0.363
Dichloromethane ^b	22	0.043	0.147	-0.022	0.108
TCA ^b	51	0.118	0.150	0.077	0.159
TCE ^b	24	-0.044	0.135	-0.101	0.013
Toluene ^b	40	-0.044	0.150	-0.090	0.003
Freon-113 ^b	16	0.335	0.076	0.295	0.375
p-xylene ^b	7	-0.092	0.093	-0.178	-0.006
Acetone ^c	15	-0.085	0.110	-0.146	-0.024
1,1-dichloroethane ^c	6	0.055	0.102	-0.047	0.157
Chloroform ^c	8	-0.097	0.157	-0.225	0.031
1,1-dichloroethene ^c	7	0.124	0.094	0.040	0.208
Tetrachloroethene ^c	5	-0.016	0.116	-0.149	0.117

a. Carbon tetrachloride.

b. VOC-specific model parameters were measured.

c. VOC-specific model parameters were estimated.

DISCUSSION

The model was accurate in estimating the total VOC concentration within waste drums with a maximum of four layers of polymer bags as well as the concentration of carbon tetrachloride, methanol, methylene chloride, TCE, toluene, 1,1-dichloroethane, chloroform, and tetrachloroethene. The model exhibited a positive bias for waste drums with a maximum of two layers of polymer bags, waste drums with a maximum of five layers of polymer bags, and in estimating the concentration within the innermost layer of confinement of TCA, Freon-113, and 1,1-dichloroethene. The model exhibited a negative bias in the case of p-xylene and acetone.

The assumption of equal VOC permeability across all polymers were made because most large and small polymer bags were made of polyethylene and there were limited VOC permeability data available for PVC. Calculation of the mean value of ψ and the confidence interval about the mean for 14 waste drums identified as containing one or PVC bags indicates that there is no bias in the current model that assumes VOC permeability across polyethylene and PVC are identical.

Conservative assumptions that were applicable to all or most waste drums, thus eliminating the need for detailed waste characterization, were made for a number of model parameters. Conservative assumptions inherently result in a higher estimated VOC concentration within the innermost layer of confinement. Assumptions concerning total bag thickness and the permeable surface area of large and small polymer bags are considered to be conservative. The maximum number of layers of polymer bags was assumed in each waste drum. The large bag surface area only gives credit to the top of the large bag. A survey of nearly 40 waste drums containing different waste types indicated that nearly all drums contained small polymer bags with a total permeable surface area greater than 500 cm². Other conservative assumptions included a VOC concentration of zero outside the waste drum, maximum bag thicknesses, and the placement of all VOC-contaminated waste inside one small bag.

Although most solid waste drums were completely sampled in one day, seven drums were sampled over two days. It was assumed that sample results would not be significantly different. The mean and standard deviation of ψ for total VOC concentration in these waste drums sampled over two days were 0.185 and 0.156, respectively. The 95%-confidence limits were both greater than $\psi = 0$ indicating a positive model bias. This suggests that longer sampling time may have resulted in lower measured VOC concentration in the small bags. Despite reasonable precautions, the VOC concentration within small bags may decrease before they are sampled. A gas sample can be collected from the large bag headspace more quickly after drum lid removal than can be collected from most waste packages after the large bags are opened. In this case, it is reasonable then to use the VOC concentration measured in the large bag headspace if it the highest reported concentration. Although a model bias was detected for waste drums with a maximum of five layers of polymer bags, the lower confidence limit was only 0.003. The 7 drums that were sampled over two days were listed as having a maximum of 5 layers. Excluding the waste drums sampled over two days, the mean and standard deviation of ψ for total VOC concentration in 17 waste drums containing a maximum of five layers of polymer bags were 0.057 and 0.198, respectively. The inclusion of $\psi = 0$ within a 95%-confidence interval indicates that the model is accurate for these waste drums that were completely sampled within one day.

Model bias in estimating the concentration of individual VOCs is attributed to the values of VOC-specific parameters used in model calculations, specifically VOC permeability. Most sludge waste drums contained TCA, Freon-113, and carbon tetrachloride. The positive bias in model results for sludge waste drums is attributed to the bias in estimating TCA and Freon-113 concentration, since these VOCs are primary constituents in the waste drums. Carbon tetrachloride plasticization of polyethylene results in an increase in amorphous fractional free volume.⁷ The VOC permeability will increase with an increasing amorphous polymer volume fraction. Thus, the VOC permeability measured in unplasticized polyethylene may have been less than VOC permeability in plasticized polyethylene. The negative model bias in predicting p-xylene and acetone concentrations suggests a smaller permeability value would result in more accurate model results. P-xylene and acetone had the largest permeability values used in model calculations.

CONCLUSIONS

The objective of this study was to demonstrate that the VOC concentration within the innermost layer of confinement of a vented waste drum can be estimated using a model incorporating diffusion and permeation transport principles and limited waste drum sampling data. The following conclusions can be made:

1. The VOC transport model was accurate in estimating the VOC concentration within the innermost layer of confinement for 8 VOCs. The VOC-specific model parameters for 5 compounds (carbon tetrachloride, TCE, toluene, methylene chloride, and methanol) were measured. The VOC-specific model parameters for 3 compounds (1,1-dichloroethane, chloroform, and tetrachloroethene) were estimated. The VOC transport model was accurate in estimating the total VOC concentration within the innermost layer of confinement for waste drums containing a maximum of 4 or 5 layers of polymer bags that were completely sampled within one day.
2. The VOC transport model exhibited a positive bias in estimating the VOC concentration within the innermost layer of confinement for 3 VOCs. The VOC-specific model parameters for 2 compounds (TCA and Freon-113) were measured and for 1,1-dichloroethene were estimated. The VOC transport model exhibited a positive bias in estimating the total VOC concentration within the innermost layer of confinement for sludge waste drums containing a maximum of 2 layers of polymer bags that were sampled within one day. It is believed that the bias is related to model bias in estimating TCA and Freon-113 concentrations which are primary constituents in sludge waste drum headspace.
3. The VOC transport model exhibited a negative bias in estimating the VOC concentration within the innermost layer of confinement for 2 VOCs. The VOC-specific model parameters for p-xylene were measured and for acetone were estimated.

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