

# EFFECTS OF ASSUMPTIONS ABOUT GAS BEHAVIOR ON MODELED RELEASES OF VOLATILE ORGANIC COMPOUNDS FROM THE WIPP

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## ABSTRACT

The Waste Isolation Pilot Project (WIPP) is authorized under Section 213 of Public Law 96-164 to demonstrate the safe disposal of radioactive waste from the defense activities and programs of the United States that are exempt from Nuclear Regulatory Commission (NRC) regulation. Both CH-TRU and RH-TRU from ten defense sites are destined for the WIPP, but only CH-TRU is discussed. The TRU waste will contain volatile organic compound (VOC) residues absorbed on metal, paper, cloth or other absorbent surfaces. WIPP is regulated by the Resource Conservation and Recovery Act (RCRA, 42 USC 6905 *et seq*) as a miscellaneous land disposal unit under 40 CFR Part 264 Subpart X, and must demonstrate under 40 CFR 268.6 that VOCs will not migrate from the site. This study examines the effects of several different assumptions about the physico-chemical behavior of the VOCs in question on the VOC concentration that may leak through the unit boundary. The CH-TRU waste described in the Part B Permit Application (2) contains the following organic solvents in quantities greater than 1 ppm: carbon tetrachloride (CCl<sub>4</sub>), methylene chloride (CH<sub>2</sub>CCl<sub>2</sub>), 1,1,1-trichloroethane and 1,1,2-trichloroethylene (TCE).

## INTRODUCTION

The Waste Isolation Pilot Project (WIPP) is authorized under Section 213 of Public Law 96-164 to demonstrate the safe disposal of radioactive waste from the defense activities and programs of the United States that are exempt from Nuclear Regulatory Commission (NRC) regulation. Both CH-TRU and RH-TRU from ten defense sites are destined for the WIPP, but only CH-TRU is discussed; the term "TRU waste" in this paper means only CH-TRU waste. No free liquid will be brought to the WIPP for disposal. The TRU waste will contain volatile organic compound (VOC) residues absorbed on metal, paper, cloth or other absorbent surfaces (1). WIPP is regulated by the Resource Conservation and Recovery Act (RCRA, 42 USC 6905 *et seq*) as a miscellaneous land disposal unit under 40 CFR Part 264 Subpart X, and must demonstrate under 40 CFR 268.6 that VOCs will not migrate from the site. WIPP is also subject to the EPA Standards for Safe Disposal of High-Level and Transuranic Radioactive Wastes under 40 CFR Part 191.

Potential migration of VOCs from the WIPP cannot be determined directly, but must be modeled. The CH-TRU waste described in the Part B Permit Application (2) contains the following organic solvents in quantities greater than 1 ppm: carbon tetrachloride (CCl<sub>4</sub>), methylene chloride (CH<sub>2</sub>CCl<sub>2</sub>), 1,1,1-trichloroethane (CCl<sub>3</sub>CH<sub>3</sub>), and 1,1,2-trichloroethylene (CCl<sub>2</sub>HCl, or TCE). The solvent concentrations in the application were estimated by DOE from process knowledge rather than by direct measurement. The VOC concentrations calculated in this paper based on two data sets:

1. Experimentally determined headspace concentrations obtained from the DOE Bin Case Data Report Summary (Ref. 2 addendum). Between three and five replicate measurements, generally within a range of +/- 5%, were reported for each drum. In order to be conservative, the datum used in this analysis was the largest of the replicate measurements. Detection limits was reported in lieu of a measurement.
2. Limits allowed by the Environmental Protection Agency (EPA) in the No-Migration Determination (NMD) (2,3). These allowed NMD (1) limits were

based on data from headspace measurements made on about 220 drums (4) that were categorized as Type I, Type II, Type III and Type IV wastes (5). The average VOC headspace vapor concentrations for each waste type (including a nominal value for drums where the VOC concentration was below detection limits) was then multiplied by 10, and the maximum for each waste type, by two, to give the limits allowed by EPA in the NMD.

In the subsequent discussion, these two parameters are referred to as "experimental concentrations" and "NMD limits," respectively.

## METHODS

The evaporation model discussed in this paper is based on solvent in equilibrium with its vapor, under one atmosphere pressure and at 293°K (20°C). The resulting total vapor concentration is essentially the same as that obtained by Kehrman using the Clausius-Clapeyron Eq. (6). The Clausius-Clapeyron equation

$$\frac{dP_{vp}}{dT} = \frac{\Delta H}{T\Delta H} \quad (1)$$

In which  $\Delta H$  and  $\Delta V$  are the differences in enthalpy and molar volume, respectively, between the saturated vapor and saturated liquid, and  $P_{vp}$  is the vapor pressure. Integrating Eq. 1 yields an expression first developed by Antoine (7) of the form

$$\ln P_{vp} = A - \frac{B}{T + C} \quad (2)$$

in which A, B, and C are constants characteristic of the particular substance. Although the solvents in the waste are not present as free liquid but are absorbed on a variety of surfaces, neither the evaporation rate nor the various conditions affecting that rate, like evaporation through a capillary, are considered. The evaporation model thus overestimates the amount of VOC vapor being produced. Chemical interactions and formation of new VOCs are not addressed because the time frame under consideration, five years, is too short.

This model has no relationship at all to the gas generation model of Brush (8).

## RESULTS

Table I shows the headspace gas concentration of VOCs in the 32 drums currently characterized for the test phase, as taken from the Bin Case Data Reports. Table II shows the values allowed by EPA in the No-Migration Determination (NMD).

TABLE I  
Experimentally Determined Headspace Gas  
Concentrations in  $\mu\text{g}/\text{m}^3 \times 10^6$

	Drum		Bin	
	Average	Maximum	Average	Maximum
CH <sub>2</sub> Cl <sub>2</sub>	0.0751	0.191	0.0632	0.161
Cl <sub>3</sub> CCH <sub>3</sub>	1.36	5.45	1.15	4.59
Cl <sub>2</sub> CCHCl	0.0392	0.296	0.0331	0.249
CCl <sub>4</sub>	0.00465	0.0128	0.00391	0.0108

EPA has listed a proposed rule for solid waste management units (Proposed Subpart S of 40 CFR Part 264) that includes air concentrations that meet EPA action levels for the four contaminants in question (9). These air concentrations are

- CH<sub>2</sub>Cl<sub>2</sub>: 0.3  $\mu\text{g}/\text{m}^3$
- CH<sub>3</sub>CCl<sub>3</sub>: 1000  $\mu\text{g}/\text{m}^3$
- CHClCCl<sub>2</sub>: 0.6  $\mu\text{g}/\text{m}^3$
- CCl<sub>4</sub>: 0.03  $\mu\text{g}/\text{m}^3$

Drum headspace concentrations allowed by EPA in the NMD can be shown not to exceed these air concentrations under the following conditions:

- No more than 1% of the ultimate inventory of CH<sub>2</sub>TRU waste is placed underground in WIPP,
- All the VOCs in the drum headspaces are released into the WIPP,
- The VOCs are diluted by the 17 million m<sup>3</sup> of air ventilating the WIPP each day,
- The VOCs are also trapped further by a 95% efficient carbon adsorption control device (10).

These conditions will not apply during the disposal phase, and potential emissions from the WIPP during the disposal phase must therefore be recalculated for disposal phase conditions.

EPA is now requiring that environmental standards for soil concentrations (given in the proposed 40 CFR 264.521, USEPA 1990b) be met, rather than those for air concentra-

tions (11). Appendix A of the proposed 40 CFR 264.521 shows that, for the four compounds under consideration, the action level soil concentration is two to three orders of magnitude larger than the action level air concentration, and thus much easier to comply with. Lappin (12) has shown that, under a variety of assumptions, the VOC concentration in the soil of the accessible environment would be well below the soil action level. However, consideration of the air pathway is necessary in the event of inadvertent human intrusion into the waste, when a direct pathway to the ground surface and the air would be established. Human intrusion has not yet been considered with respect to the disposal phase of WIPP. After WIPP closure, VOCs in the vapor phase could be released to the ambient air if a pathway directly to the accessible environment were opened.

VOC vapor concentrations may be calculated under various conditions. The following calculations of VOC concentration, that are analogous to those performed by Lappin, are shown in detail for CCl<sub>4</sub>, and summarized in Table IV for the four VOCs in question. These calculations assume that the VOC vapor behaves like an ideal gas -- assumption of non-ideality would yield smaller concentrations. Ambient concentrations of VOC vapors in the drums were calculated for a 147 liter void volume.

If there is no human intrusion, and the WIPP gradually closes around the waste, the volume available to any gas, including VOC vapor, is the pore space in the anhydrite layer of the WIPP. Lappin (1993) has assumed that the total accessible void volume, assuming gas-accessible room porosity of 0.40, gas-accessible anhydrite porosity of 0.30, 2 m effective flow thickness and a cylindrical anhydrite volume 716 m. deep and with a radius of 150 m, is 1.2 million m<sup>3</sup>.

The drum headspace concentrations allowed by EPA in the NMD are given in Table II, above; the average CCl<sub>4</sub> concentration for Type I waste is 5.13 g/m<sup>3</sup>. If we assume that the full complement of 850,000 drums in the WIPP contain only Type I waste, and that the average void volume per drum is 147 liters (Lappin 1993), the total grams of CCl<sub>4</sub> in the WIPP would be

$$(5.13 \frac{\text{g}}{\text{m}^3})(0.147 \text{ m}^3/\text{drum})(850,000 \text{ drums}) = 6.41 \times 10^5 \text{ g CCl}_4 \quad (3)$$

If the VOC vapor is assumed to be at equilibrium in the accessible void volume, at lithostatic pressure (attained about 150 years after closure),

$$\frac{6.41 \times 10^5 \text{ g}}{1.20 \times 10^6 \text{ m}^3} = 0.534 \frac{\text{g}}{\text{m}^3} \text{ of CCl}_4 \quad (4)$$

This calculation assumes that no additional gas has been generated by radiolysis, corrosion or biochemical action (or by any other mechanism). If a borehole were drilled into the

TABLE II  
Drum Concentrations Allowed by the EPA No-migration Determination, in  $\mu\text{g} \times 10^6$

	Maximum				Average			
	I	II	III	IV	I	II	III	IV
CH <sub>2</sub> Cl <sub>2</sub>	15.6	29.7	17.7	50.2	13.8	14.9	11.7	32.9
Cl <sub>3</sub> CCH <sub>3</sub>	104	316	118	831				
Cl <sub>2</sub> CCHCl	13.7	18.6	15.8	20.8	4.38	15.3	15.3	15.3
CCl <sub>4</sub>	15.5	16.7	37.2	524	5.13	11.5	19.2	442

WIPP, then at the surface, the VOC vapor would be diluted further in going from lithostatic pressure to atmospheric pressure. Again assuming equilibrium, volume diluting the VOC vapor may be obtained using Boyle's Law:

$$p_{\text{lith}} V_{\text{lith}} = p_{\text{atm}} V_{\text{atm}}$$

$$V_{\text{atm}} = \frac{(148 \text{ atm})(1.2 \times 10^6 \text{ m}^3)}{1 \text{ atm}} = 1.77 \times 10^8 \text{ m}^3 \quad (5)$$

The VOC vapor is then diluted by this volume to

$$\frac{(0.534 \text{ g/m}^3)(10^6 \text{ } \mu\text{g/g})(1.2 \times 10^6 \text{ m}^3)}{1.77 \times 10^8 \text{ m}^3} = 3620 \text{ } \mu\text{g/m}^3 \text{ CCl}_4 \quad (6)$$

An ambient air concentration of  $3620 \text{ } \mu\text{g/m}^3$  is about six orders of magnitude greater than the proposed action level of  $0.03 \text{ } \mu\text{g/m}^3$  for  $\text{CCl}_4$ . The results of these calculations are given in Tables III through IX. As may be seen from the table titles, a successive series of assumptions was made, generally with each assumption building on, and adding to, the preceding assumption. These progression from higher to lower concentrations in these tables has only relative significance; a different set of assumptions would clearly yield different numbers. The exercise demonstrates the sensitivity of these calculations to the assumptions that go into them. Numbers have been rounded off to three significant figures, although probably no more than two are warranted. The "NMD weighted average" takes into account the estimated fraction of each type of waste

(Type I = 0.2338, Type II = 0.2403, Type III = 0.4339, Type IV = 0.0320) as used in the no-migration petition to EPA (6,9).

The effects of several of these modeling assumptions on the ambient air concentration are highlighted below.

#### Effect of Porosity

The VOC vapor is considered to occupy the entire sub-surface void space. The VOC vapor concentration is thus heavily dependent on assumptions about porosity: to wit, a tenfold decrease in porosity would result in a tenfold increase in concentration.

#### Effect of Escape of VOC Vapor Directly to the Surface

When gas escapes for the underground, as through a borehole, the decrease from lithostatic to atmospheric pressure results obviously in expansion and, therefore, dilution of the VOC vapor. This may be seen by comparing Table IV with Table V.

#### Effect of Gas Generation

Kosiewicz (13) has established that, under appropriate conditions, radiolysis, corrosion and biochemical action will produce a combination of hydrogen,  $\text{CO}$  and  $\text{CO}_2$  in drums of TRU waste. This phenomenon has been modeled by Brush, *et al* (8). A small amount of VOCs, presently thought to be negligible compared to the quantities emplaced and therefore neglected in the calculations, may also be produced by radiolysis (8). The generated gas may be treated as an inert

TABLE III  
Total Gas in WIPP: Headspace Concentration x  $147/\text{m}^3$  x 850,000 Drums, in Grams

	$\text{CH}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	401	3,410	9,380	1.70E+5
Experimental max	1055	25,700	23,900	6.80E+5
NMD Type I av	1.72E+6		5.47E+5	6.41E+5
NMD Type II av	1.86E+6		1.91E+6	1.44E+6
NMD Type III av	1.46E+6		1.91E+6	2.40E+6
NMD Type IV av	4.11E+6		1.91E+6	5.53E+7
NMD Type I max	1.95E+6	1.31E+7	1.71E+6	1.92E+6
NMD Type II max	3.71E+6	3.94E+7	2.33E+6	2.08E+6
NMD Type III max	2.21E+6	1.47E+7	1.98E+6	4.65E+6
NMD Type IV max	6.28E+6	1.04E+8	2.60E+6	6.55E+7

TABLE IV  
Assume No Gas is Generated; Pressure is Lithostatic; Gas Concentration In WIPP Per Year, in  $\mu\text{g/m}^3$

	$\text{CH}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	0.0003	0.0028	0.0078	0.142
Experimental max	0.0009	0.0215	0.0199	0.567
NMD Type I av	1.44		0.46	0.53
NMD Type II av	1.55		1.60	1.20
NMD Type III av	1.22		1.60	2.00
NMD Type IV av	3.43		1.60	46.1
NMD Type I max	1.62	10.9	1.42	1.60
NMD Type II max	3.09	32.9	1.94	1.74
NMD Type III max	1.84	12.3	1.65	2.17
NMD Type IV max	5.23	86.6	2.17	54.6
NMD weighted av	1.42	1.33	2.88	
NMD weighted max	2.40	0.35	1.59	23.6

TABLE V

Assume No Gas is Generated; Pressure is Atmospheric; Gas Concentration in WIPP Per Year, in  $\mu\text{g}/\text{m}^3$ \*

	$\text{H}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	2.26	19.2	52.8	959
Experimental max	5.93	145	134	3830
NMD Type I av	9700		3080	3610
NMD Type II av	10,400		10,800	8110
NMD Type III av	8210		10,800	13,500
NMD Type IV av	23,100		10,800	3.11E + 5
NMD Type I max	10,900	73,400	96,200	10,800
NMD Type II max	20,900	2.22E + 5	13,100	11,700
NMD Type III max	12,400	82,800	11,200	26,100
NMD Type IV max	35,300	5.84E + 5	14,400	3.69E + 5
NMD weighted av	9750		8980	19,400
NMD weighted max	16,200	2350	10,700	1.59E + 5

\*Please note the change in units: Table IV is in  $\mu\text{g}/\text{m}^3$  but Table V is in  $\mu\text{g}/\text{m}^3$

TABLE VI

500 Moles of Gas Generated/drum; Atmospheric Pressure; Concentration of Gas Per Year in  $\mu\text{g}/\text{m}^3$ 

	$\text{CH}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	2.13	18.1	49.8	905
Experimental max	5.60	137	127	3614
NMD Type I av	9160		3080	3610
NMD Type II av	9860		10,800	8110
NMD Type III av	7750		10,800	13,500
NMD Type IV av	21,800		10,800	3.11E + 5
NMD Type I max	10,300	73,400	96,200	10,800
NMD Type II max	19,700	2.22E + 5	13,100	11,700
NMD Type III max	11,700	82,800	11,200	26,100
NMD Type IV max	33,300	5.84E + 5	14,400	3.69E + 5
NMD weighted av	9036		8980	19,400
NMD weighted max	15,300	2350	10,700	1.59E + 5

TABLE VII

500 Moles of Gas Generated/drum; Atmospheric Pressure; Concentration of Gas After 1000 Years in  $\mu\text{g}/\text{m}^3$ 

	$\text{CH}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	0.04	0.32	0.87	15.9
Experimental max	0.10	2.40	2.22	63.4
NMD Type I av	161		51.0	59.7
NMD Type II av	173		178	134
NMD Type III av	136		178	224
NMD Type IV av	383		178	5150
NMD Type I max	181	1220	159	179
NMD Type II max	346	3670	217	194
NMD Type III max	206	1370	185	433
NMD Type IV max	585	9670	242	6100
NMD weighted av	158		149	321
NMD weighted max	268	38.9	178	2630

TABLE VIII

500 Moles of Gas Generated/drum; Atmospheric Pressure; NMD Amounts Normalized to the Fraction of Each Type of Waste; NMD Averages Divided by 10; NMD Maxima Divided by 2; Concentration of Gas After 1000 Years in  $\mu\text{g}/\text{m}^3$

	$\text{CH}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	0.04	0.32	0.87	15.9
Experimental max	0.098	2.40	2.22	63.4
NMD Type I av	3.75		1.19	1.40
NMD Type II av	4.16		4.29	3.23
NMD Type III av	6.71		8.81	11.1
NMD Type IV av	1.23		0.57	16.5
NMD Type I max	21.2	142	18.6	20.9
NMD Type II max	41.6	441	26.0	23.3
NMD Type III max	50.8	339	45.6	107
NMD Type IV max	9.35	155	3.87	97.6
NMD weighted av	15.8		14.9	32.1
NMD weighted max	33.3	142	32.3	51.7

TABLE IX

500 Moles of Gas Generated/drum; Atmospheric Pressure; NMD Amounts Normalized to the Fraction of Each Type of Waste; NMD Averages Divided by 10; NMD Maxima Divided by 2; Only 40% of Waste Contains VOCs; Concentration of Gas After 1000 Years in  $\mu\text{g}/\text{m}^3$

	$\text{CH}_2\text{Cl}_2$	$\text{CCl}_3\text{CH}_3$	$\text{CCl}_2\text{CHCl}$	$\text{CCl}_4$
Experimental av	0.01	0.13	0.35	6.40
Experimental max	0.040	0.96	0.890	25.4
NMD Type I av	1.50		0.48	0.56
NMD Type II av	1.66		1.71	1.29
NMD Type III av	2.68		3.52	4.42
NMD Type IV av	0.49		0.23	6.59
NMD Type I max	8.47	56.8	7.45	8.37
NMD Type II max	16.6	177	10.4	9.32
NMD Type III max	20.3	135	18.3	42.7
NMD Type IV max	3.74	61.9	1.55	39.1
NMD weighted av	6.34		5.94	12.9
NMD weighted max	13.3	56.8	12.9	20.7

ideal gas, and will dilute the VOC vapor, as may be seen by comparing Table V with Table VI. It may be noted, however, that a 50,000% increase in gas generated results in less than 10% increase in dilution. Expansion from lithostatic to atmospheric pressure has a much more significant effect on VOC concentrations in air.

#### Effect of Limiting VOC Content of Emplaced Waste

It is estimated that approximately 70% of the CH-TRU waste destined for WIPP is not stored and has yet to be generated (3). The waste being generated need not contain VOCs or other gas phase hazardous constituents, though much of it may be classified as mixed waste because of the lead content (leaded gloves, aprons, etc.). The waste currently being generated at the Plutonium Finishing Plant at Hanford, for example, is not even mixed waste. One may then assume that compliance with RCRA can logically require removal of detectable amounts of VOCs before packaging. Thus, the WIPP need not contain 850,000 drums of VOC-containing waste, but about 260,000.

#### CONCLUSIONS

Unless a pathway is made directly into the WIPP repository, as by drilling, VOCs from the WIPP will not be released into the ambient air. According to the model used in this particular study, if inadvertent human intrusion were to create such a pathway 1000 years or more after closure, the concentration of VOC vapor released into the ambient air would be below or about the same order of magnitude as the proposed EPA Air Action Level, except for  $\text{CCl}_4$ . The ambient concentration of  $\text{CCl}_4$  would still exceed the proposed EPA Air Action Level by about two orders of magnitude. This conclusion should be considered in view of the constraints posed by the model:

- Assumption of gaseous equilibrium is conservative; any assumptions about VOC transport would decrease the modeled ambient air concentration. VOC transport within the drums containing the waste was not considered.
- The model assumes that all vapor phase VOCs are released from the waste containers.

- The model is heavily dependent on assumptions about porosity of the geologic medium.
- Hydrogen, CO<sub>2</sub>, and CH<sub>4</sub> generated by corrosion, microbial action, and radiolysis could impede the flow of VOCs to an unknown extent, although the diluting effect of generated gas is relatively small.
- The EPA Air Action Levels are not a promulgated rule, but are still in the draft stage.
- No consideration is given here to emitters of these same VOCs that are exempt from RCRA regulation under 40 CFR 261.4.

Even in view of these constraints, however, the risk from VOC emissions into the ambient air can be reduced considerably by some straightforward expedients. First, TRU waste now being generated, and that still to be generated, need not contain any VOCs. Second, VOCs are readily chemisorbed -- this is how they came to contaminate the TRU waste initially. Possible engineered barriers to the movement of radionuclides from the WIPP repository would also chemically or physically adsorb VOC vapors. In sum, this study does not show either that the WIPP will violate RCRA or that the VOCs in the TRU waste destined for the WIPP pose a significant health risk.

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