

MICROBIAL EFFECTS ON RADIOACTIVE WASTES AT SLB SITES*

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INTRODUCTION

A significant fraction of DOE and commercially generated low-level radioactive waste consists of carbonaceous compounds including paper, wood, cloth, animal carcasses, scintillation mixtures and chelating agents from decontamination solutions. These materials are subject to degradation by microorganisms present in the shallow land burial environment and may contribute to enhanced migration of radionuclides through the formation of gases, mobile complexes and bioaccumulation. Microorganisms may directly attack the waste package promoting container corrosion and waste form degradation. Problems relevant to subsidence and trench deterioration have also been attributed to microbial degradation of organic wastes.

While several mechanisms for enhanced radionuclide migration as a result of microbial degradation have been proposed, there is no consensus concerning which mechanisms predominate or whether they are even significant.

The objectives of this study are to determine the significance of microbial degradation of organic wastes on radionuclide migration in shallow land burial for humid and arid sites, establish which mechanisms predominate and ascertain the conditions under which these mechanisms operate.

A literature study was conducted in FY 82 to determine the present state of knowledge dealing with radionuclide movement attributed to microbial activities at shallow land burial sites.

The results of this study are given in detail in reports entitled Assessment of Microbial Processes on Gas Production at Radioactive Low-Level Waste Disposal Sites, BNL-51557, May 1982 and Assessment of Microbial Processes on Radionuclide Mobility in Shallow Land Burial, BNL-51574, July 1982.

This work was identified as a major data need in a report entitled, Unresolved Technical Issues in Land Burial of Low-Level Radioactive Wastes, ORNL/NFW-79/62 (October 1979). The program is in direct support of DOE/LLW Management Program Alpha Milestone B, "Develop technology for waste treatment, handling, and packaging for shallow land burial site disposal," Milestone C, "Develop technology and documentation to support a shallow land burial site," and Milestone D, "Develop remedial action technology for shallow land burial sites."

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MICROBIAL GENERATION OF GASES

Factors controlling gaseous emanations from low-level radioactive waste disposal sites are assessed. Importance of gaseous fluxes of methane, carbon dioxide and possibly hydrogen from the site stems from the inclusion of tritium and/or ^{14}C into the elemental composition of these compounds. In that the primary source of these gases is the biodegradation of organic components of the waste materials, primary emphasis of the study involved an examination of the biochemical pathways producing methane, carbon dioxide and hydrogen, and the environmental parameters controlling the activity of the microbial community involved. Initial examination of the data indicates that the ecosystem is anaerobic. Hence, reaction sequences of importance are fermentation of carbo-naceous substrates to fatty acids, alcohols, ammonia, carbon dioxide and hydrogen. The fatty acids and alcohols are further metabolized to acetate plus carbon dioxide. With a sufficiently reducing environment, acetate, carbon dioxide and hydrogen are converted to methane by the methanogenic bacteria. As the result of the complexity of the pathway leading to methane production, factors such as substrate availability, which limit the initial reaction in the sequence, greatly affect the overall rate of methane evolution.

Although the methane and carbon dioxide production rate indicates the degradation rate of the organic substances in the waste, it does not predict the methane evolution rate from the trench site. Methane fluxes from the soil surface are equivalent to the net synthesis minus the quantity oxidized by the microbial community as the gas passes through the soil profile. Methane metabolized within the soil profile will be converted to carbon dioxide, water and cell components. The ^{14}C in the methane converted to carbon dioxide would leave the ecosystem with carbon dioxide produced within the confines of the trench. The ^{14}C entering cell components would remain within the soil for a period of time dependent upon the turnover rate of the cell carbon.

FIELD STUDIES OF GASEOUS RELEASES

Gas studies were performed at three of the six commercial low-level radioactive waste disposal sites during the period 1976-1978. The first was an exploratory study performed at West Valley, New York, in June 1976 by personnel from USGS, NYGS, and the N.Y. State Health Department (1-3). Additional gas samples were collected at West Valley during periods in 1977 and 1978. In August 1978, a limited study was conducted at the Beatty, Nevada, disposal site (4,5) and in December, 1978, a similar study was made at Maxey Flats, Kentucky (6). The results of these studies are summarized in Tables 1-3.

Table 1

Radionuclide Concentrations in Field Air and Soil Gas Samples
Above Trench Soil Covers at West Valley, New York [3]

($\mu\text{Ci}/\text{cm}^3$ \pm error in parentheses)

Over-Trench	Date	$^3\text{H}_2$	$^{14}\text{C}_4$	$^3\text{HCH}_3$	$^{14}\text{C}(\text{HC})^a$	$^3\text{H}(\text{HC})^a$	$^{14}\text{CO}_2$	$^3\text{H}_2\text{O}$
14 (1 in.)	6/15/77	-	<8 E-8	<2 E-7	-	-	<1.6 E-7	-
11 (1 in.)	6/15/77	-	<1.1 E-7	<1.4 E-7	-	-	<1.3 E-7	-
9 (1 in.)	9/21/77	-	<3 E-8	<1.0 E-7	-	-	-	-
5 (1 in.)	9/21/77	-	<5 E-8	<1.7 E-7	-	-	-	-
9 (1 in.)	4/26/78	<6 E-8	<1.4 E-8	7 E-8(83)	-	-	<1.3 E-7	-
5 (5 ft.)	4/26/78	-	<1.3 E-12	8 E-13(51)	-	-	<4 E-11	2.5 E-12(17)
14 (5 ft.)	4/26/78	-	<9 E-13	4 E-12(34)	-	-	<1.3 E-11	< 4E-13
3 (Over-fracture)	6/04/76	<1.3 E-9	7 E-7(11)	1.1 E-7(10)	-	-	6.7 E-9(12)	-
			b					
5 (F) Flux ^c Box	9/21/77	4.1 E-9(40)	3.3 E-7(5)	9.0 E-6(5)	1.1 E-8(21)	1.1 E-6(5)	4.1 E-7(7)	<1.6 E-9
5(UF) Flux ^d Box	9/21/77	<3 E-10	9.1 E-8(5)	2.3 E-6(5)	<2 E-9	4.8 E-7(5)	9.3 E-7(5)	1.2 E-8

^aAs hydrocarbons above methane (C_2H_6 , C_3H_8 , and C_4H_{10})

^bTotal methane in gas sample < 0.02%

^cFractured

^dUnfractured

Table 2
 Radionuclide Concentrations and Gas Compositions
 of Trench and Soil Gases at Beatty, Nevada [4]

Location	H ₂ (% Vol.)	O ₂ (% Vol.)	CO ₂ (ppm)	CH ₄ (ppm)	Trace H ₂ O (mg/L)	Gaseous Tritium ($\mu\text{Ci}/\text{cm}^3$)	³ HHO ($\mu\text{Ci}/\text{cm}^3$)	Gaseous C-14 ($\mu\text{Ci}/\text{cm}^3$)
<u>Background</u>	78	22	300	10	1.95	<1.4 E-10	5.52 E-8	<9.01 E-11
<u>Trench Gas</u>								
Observation Well R-1	78	20	1860	7.7	2.81	2.68 E-7	8.22 E-8	5.90 E-8
Observation Well R-2	78	17	8100	19	2.74	5.68 E-9	3.08 E-8	3.60 E-8
Observation Well R-3	74	15	1270	41	2.55	4.73 E-8	2.93 E-8	3.04 E-8
Observation Well R-6	78	22	600	7	--	2.11 E-7	--	1.46 E-9
Sump Well Tr-14	76	21	530	16	2.88	9.86 E-8	5.52 E-6	2.59 E-8
Sump Well Tr-16	64	17	2200	9.8	3.59	9.82 E-9	9.35 E-8	8.56 E-9
<u>Soil Gas</u>								
Canister Tr-2	73	21	300	5.1	1.77	<1.4 E-10	5.18 E-8	<9.0 E-11
Canister Tr-3	77	22	300	7.1	2.11	1.99 E-7	1.90 E-8	2.21 E-10
Canister Tr-4	76	21	300	6.5	2.08	1.17 E-7	7.55 E-8	1.35 E-10
Canister Tr-5	73	21	300	6.5	2.02	7.75 E-8	1.27 E-8	5.27 E-9
Canister Tr-14	79	21	300	6.5	2.02	1.53 E-7	3.72 E-8	5.41 E-10
Canister Tr-16	78	22	300	7.3	2.58	1.98 E-7	1.62 E-8	8.11 E-10

Table 3
Radionuclide Concentrations and Gas Compositions
of Trench and Soil Gases at Maxey Flats, Kentucky [6]

Location	N ₂ (% Vol.)	O ₂ (% Vol.)	CO ₂ (ppm)	CH ₄ (ppm)	Trace H ₂ O (mg/L)	Gaseous Tritium (μCi/cm ³)	³ HHO (μCi/cm ³)	Gaseous C-14 (μCi/cm ³)
<u>Background</u>								
North Fence	77	22	98	2	0.95	4.18 E-8	2.48 E-9	N.D. ^a
South Fence	77	22	168	2	2.11	1.28 E-8	8.33 E-9	N.D.
<u>Trench Gas</u>								
Tr-7 Sump	77	22	148	2	1.48	1.29 E-8	6.31 E-9	N.D.
Tr-11S Sump	77	22	470	114	2.60	1.58 E-8	1.48 E-8	4.95 E-11
Tr-18 Sump	78	20	12000	6720	2.56	1.12 E-7	2.25 E-8	7.88 E-8
Tr-26 Sump	77	22	29	2	2.22	1.58 E-7	7.77 E-9	2.57 E-10
Tr-35 Sump	77	22	186	8	1.38	1.61 E-8	2.25 E-9	N.D.
<u>Soil Gas</u>								
Tr-11S	77	22	255	1	1.04	2.84 E-9	8.67 E-10	N.D.
Tr-18	77	21	2580	12	1.33	9.01 E-10	6.42 E-10	4.50 E-11
Tr-20	78	22	881	1.4	1.47	1.13 E-8	2.59 E-9	N.D.
Tr-26	77	22	819	2	2.12	5.81 E-9	4.95 E-10	N.D.
Tr-35	77	22	1410	4	2.83	9.50 E-9	1.80 E-9	N.D.

^a N.D. - Not detected

The concentrations in methane and carbon dioxide are orders of magnitude higher at West Valley, while the levels at Maxey Flats are comparable to Beatty. It would be expected that microbial activity at the humid West Valley site would produce more methane and carbon dioxide than at the arid Beatty site, but the observation that the levels of carbon-14 in soil gas are approximately the same at Beatty and Maxey Flats (humid site) is noteworthy. The observation that gaseous tritium and tritiated water vapor in soil gas samples at Beatty are higher than at Maxey Flats may be caused by (1) the higher evapotranspiration at Beatty, (2) the higher permeability of the trench covers at Beatty, or (3) the differences in atmospheric pressure at the time the samples were obtained.

The concentrations of tritium and carbon-14 measured in soil gas compared to maximum permissible concentrations (MPC) for release to unrestricted areas listed in Table II of 10 CFR Part 20 are a factor of 200 below MPC for tritium and a factor of 190 below MPC for carbon-14 (7). These calculations were based on the maximum tritium and carbon-14 concentration observed at the Beatty facility, viz., 2×10^7 $\mu\text{Ci}/\text{cm}^3$ gaseous tritium and 5.27×10^3 $\mu\text{Ci}/\text{cm}^3$ carbon-14. MPC values of 4×10^{-5} $\mu\text{Ci}/\text{cm}^3$ and 1×10^{-6} $\mu\text{Ci}/\text{cm}^3$ for submersion doses were used for tritium and carbon-14, respectively. The same comparisons for the highest tritium and carbon-14 soil gas emanations measured at Maxey Flats (1.1×10^8 $\mu\text{Ci}/\text{cm}^3$ [Tritium] and 9×10^7 $\mu\text{Ci}/\text{cm}^3$ [carbon-14]) indicate less than MPC releases for each radionuclide. The highest emanations measured at West Valley, in the flux boxes above Trench 5, were a factor of 4.4 below MPC for tritium and a factor of 1.1 below MPC for carbon-14. The actual concentrations in unrestricted areas would be much less due to atmospheric dispersion. Emanation rates at the facility boundary are expected to be considerably less than 25 mrem/yr whole body criteria proposed (7).

Additional field studies should be performed to verify these preliminary investigations, taking into consideration the effects of differences in site climate, soil conditions, and measurement techniques.

BIOACCUMULATION OF RADIONUCLIDES

The questions which must be considered to determine the impact of bioaccumulation on radionuclide mobility are: a) Does bioaccumulation occur in the trench ecosystem? b) Would bioaccumulation enhance or retard radionuclide mobility? c) What would the effect of radionuclide decay be on microbial growth and development? d) Upon death of the microbial cell would mineralization of the cell constituents and subsequent mobility of the radionuclides be greater or less than if bioaccumulation had not occurred? Due to the lack of data concerning occurrence of this phenomenon in the disposal trench, conclusions must be based on extrapolation of experimental results collected during study of alternate ecosystems (8-10).

The major physical manifestation of bioaccumulation is that soluble radionuclide molecules are converted to particulate substances of sizes approximating those of growing microbial cells. Thus, factors controlling mobility of the nuclide will essentially be those relating to microbial mobility in general. Within the trench confines, the microbes may be suspended in the trench waters or immobilized on surfaces of particulate substances. The latter microbes are of little consequence in radionuclide loss from the trench. The question, thus, becomes: Will the bacteria suspended in trench waters be able to penetrate the soil matrix and migrate from the trench? Considerable research has been conducted to evaluate movement of fecal coliforms contained in sewage effluent applied to soil surfaces or entering soils through septic systems. These data are useful in evaluating migration of microbes from the waste disposal trench. Both mineral and organic soils are effective filters of microbial cells from sewage and septic system effluents (11-17). Few difficulties are encountered, except in the cases of channeling and with saturated water flow. Thus, it can be concluded that construction of a compacted trench base and placement of the trench in regions of low soil water tables would minimize problems associated with bioaccumulation. Placement of trenches on sandy soils may create some difficulties in that microbial movement is greater in that soil type than in less permeable soils.

Material accumulated on growing microbial cells would be expected to remain attached to the cell until the death and subsequent decay of the cell constituents. Longevity of the microbes surrounded with the radionuclides may be anticipated to be shorter than that of the microbe adsorbing a nonradioactive metal, such as iron. Wildung and Garland (18) observed a significant decline in aerobic spore forming and anaerobic bacteria in soil amended in hydrolyzable plutonium ($^{239}\text{Pu}(\text{NO}_3)_4$) at concentrations as low as 1 $\mu\text{g/g}$ soil. Thus, stability of the radionuclide-microbe complex could be threatened by the radiation induced death of the microbe. Unfortunately, the effect of the attached radionuclide molecules on the mineralization of the dead cell debris is unknown. It could be hypothesized that sufficient radionuclide concentrations could be bound to the cell debris to inhibit or prevent its subsequent biodegradation. Thus, should this be the case, retention of the radionuclides in non-mobile forms would be extended. Research to elucidate this possibility is suggested.

CHELATION EFFECTS ON RADIONUCLIDES

Problems with chelators in trench waters are divisible into two major areas: 1) chelators originating with the waste materials, and 2) chelators synthesized by the microbial community indigenous to the trench. Although augmented radionuclide mobility is the major problem associated with each group, the importance and impact of each source is unique. Thus, the two types of chelators will be discussed separately.

CHELATORS ORIGINATING WITH THE WASTE MATERIALS

Chelators contained within the waste materials are primarily EDTA (ethylenediamine tetraacetic acid), DTPA (diethylenetriaminepentaacetic acid), and NTA (nitrilotriacetic acid). These compounds convert insoluble radionuclides into water soluble structures, which may migrate from the trench confines with trench waters (19). EDTA, DTPA and NTA are all biodegradable under aerobic conditions. Tiedje (20) reported the degradation of EDTA in soils and sediments under aerobic conditions and by microorganisms in mixed liquid cultures. Equivalent degradation rates were found for the copper, cadmium, zinc, manganese, calcium and iron salts in soil, whereas the nickel salt was degraded more slowly. No radionuclide salts were examined. Similar results were found by Belly et al. (21) with EDTA decomposition in an aerated lagoon. No degradation was found under anaerobic conditions.

Reports of DTPA degradation are less common. Wildung and Garland (18) demonstrated, however, that DTPA complexed with ^{238}Pu and ^{239}Pu were degraded by about 20% over a 95-day period when incubated aerobically.

Studies of NTA decomposition are more abundant. Tiedje and Mason (22) found NTA decomposition in a variety of agricultural soils receiving sewage effluent. Degradation was observed with NTA concentrations of 10 to 600 ppm and temperatures of 2 to 24°C, but not under anaerobic conditions. Similarly, Barnhart et al. (23) found no NTA decomposition under anaerobic conditions in the absence of nutrient supplements. Enfors and Molin (24,25) have reported anaerobic degradation of this chelator; nitrate was used as terminal electron acceptor. A wide variety of bacteria have been isolated that are capable of converting NTA to carbon dioxide and ammonia in the presence of oxygen (26-31).

These data demonstrate that DTPA, NTA and EDTA degradation occurs slowly under aerobic conditions and that little decomposition can be expected under anaerobic conditions. The sole case of significant anaerobic decomposition indicated a requirement for nitrate as a terminal electron acceptor. Nitrate would not be expected to occur in significant concentrations in low level radioactive waste disposal trenches. Thus, it is reasonable to conclude that reduction of solubility of nuclide chelates via microbial action would be minimal in the low level radioactive waste disposal trench. A more probable reduction of radionuclides complexing with these chelates would result from exchange of radionuclides with metals such as iron, which have a higher affinity for binding to the chelator.

CHELATORS SYNTHESIZED BY MICROORGANISMS

A wide variety of compounds that may solubilize radionuclides are produced by the microbial community. These include hydroxamates (produced by bacteria for solubilizing iron), fatty acids and metabolic products, as well as humic and fulvic acids, and metabolic intermediates

such as 2-ketogluconic acid. As indicated above, fatty acids are produced during anaerobic decomposition of the carbonaceous waste substituents in the disposal trenches. Fulvic and humic acids are soil organic matter components, which result from microbial and chemical transformations of the more biodegradation resistant carbon residues (32). These residues include the lignin common in plant debris and products from plants such as paper. Because of the abundance of acidic groupings on these molecules, solubilization of cationic nuclide residues will occur. Complexing with humic acid would be of little consequence in that these acids are not water soluble. Significance of the other complexes will depend upon the capability of iron, and other metals known to be present in trench waters, to displace the radionuclides, and the biodegradability of radionuclide-acid complexes. Further research is suggested to elucidate this problem.