RESPIRATION OF GASES FROM

NEAR-SURFACE RADIOACTIVE WASTE BURIAL TRENCHES

John M. Matuszek and Lawrence W. Robinson[a]
Radiological Sciences Institute
Center for Laboratories and Research
New York State Department of Health
Albany, New York 12201

ABSTRACT

Experience at the low-level radioactive waste burial site at West Valley, New York, indicates that the NRC, in its proposed regulation 10CFR61, has incompletely evaluated the major exposure pathways at this wet site - and perhaps for all other sites, whether in a wet or dry environment. Offsite exposures at West Valley via the surface-water and groundwater pathways are so slight as to be virtually unmeasurable. Radioactive gases emanating from the burial trenches, on the other hand, may exceed, not only the 25-mrem annual limit of 10CFR61, but the 500-mrem annual limit of 10CFR20. Tritium, ^{222}Rn, ^{14}C, and ^{85}Kr are the principal contributors to offsite and onsite exposures at this site.

The West Valley data indicate that to minimize exposures, wastes should perhaps be segregated beyond the requirements of 10CFR61, or greater use should be made of incineration of biodegradable wastes.

Extension of these findings to other sites seems reasonable, but no studies of similar scope have been performed at any other site. Even the work at West Valley is as yet incomplete. However, laboratory studies of trench-water composition performed at Brookhaven National Laboratory and analysis of a few trench-gas samples collected at the Sheffield, Illinois, burial site indicate that operators at all wet sites in the United States must be concerned with the gaseous pathway. For arid sites the NRC acknowledges that gas respiration is likely to be the most significant dose pathway.

INTRODUCTION

Burial sites for low-level radioactive wastes were once expected to completely retain these contaminants. However, total isolation is simply not possible even in the short term so small amounts of the buried radionuclides have been mobilized via surface-water and groundwater, resulting in public and regulatory concern about the adequacy of present burial practices. Reflecting this concern, regulators of sites used for shallow-land burial of radioactive wastes have concentrated in recent years on controlling the risks of potential migration of radioactivity via the groundwater pathway. It appears that in enforcement of its newly proposed regulation, 10CRF61, the U.S. Nuclear Regulatory Commission (NRC) will continue to focus on groundwater as "the primary long-term pathway of release of radioactivity from near-surface disposal,"[1,2] at least at wet sites.

To reinforce this emphasis the NRC asserts - without experimental support - that at wet sites "gaseous releases do not have a large impact and can be reduced by assuring stable site conditions."[2] Yet the NRC acknowledges that at arid sites gas and/or vapor may be the most significant dose pathway[1,2]; and at one wet site the NRC assumption is demonstrably wrong. Our radiochemical studies at the low-level waste burial site in West Valley, New York, indicate that the predominant uncontrolled release pathway is respiration of gases ($^{3}HCH_3$, $^{14}CH_4$, ^{85}Kr, ^{222}Rn, and perhaps HTO) from the waste trenches [3-9], whereas the surface-water and groundwater pathways have little impact[3,4,6,10,11].

A study of the chemical and radiochemical character of the West Valley low-level trenches by our institute showed that the trenches are chemically similar to sanitary landfills.[12] As with a sanitary landfill, the large biomass buried in the trenches undergoes aerobic and anaerobic biodegradation to produce water-soluble and -insoluble products and gases, such as CO_2, CH_4, H_2O, and other hydrocarbons.[9] At West Valley the predominant modes for waste mobilization appear to be leaching, erosion, and gas generation, while the common transport pathways are surface-water runoff and atmospheric diffusion. Venting of radioactive gases through the trench cover and dispersion into the atmosphere is the transport and dose pathway likely to exceed the limits in 10CFR61, while leachate in groundwater or surface water has been shown to be inconsequential as a dose pathway.[13] Eroded waste could present a significant surface water pathway if appropriate precautions are not taken to stabilize the trenches against this process.[3]

Unfortunately the federal agencies which regulate radioactive waste burial have made little use of the available data. The U.S. Environmental Protection Agency (EPA), an early cosponsor of studies at West Valley, dismissed the first reports of gas respiration as inconsequential and terminated further work on this pathway. The NRC continued partial funding of the studies at West Valley but only for hydrologic and geologic work. Estimates of biogenic gas production rates were a serendipitous result of Lu's work on the GASFLO model[10], which was intended primarily to evaluate cap transmissivity as a measure of precipitation infiltration into the trenches. Robinson's work on the ADIFF model[8] earned him a master's degree, but time restrictions limited his efforts to simulation of of microscale diffusion from a single trench. Despite these limitations, we are able to use the models to

[a] Present address: Cornell University Medical College, New York, NY 10021

obtain order-of-magnitude estimates of downwind concentrations of respired trench gas for comparison to regulatory standards.

EXPERIMENTAL APPROACH

Experimental details have been published in the several references cited here; for additional experimental information see the references cited in a recent review.[13] This report will focus on the implications of concentration estimates reported in the works referenced here.

Transport of trench gas through a fracture network within the cover has been modeled.[4,5] Rising and falling atmospheric pressure acts as a pump to respire air into or gas out of the trench voids over short time periods, moving a few thousand cubic meters of gas into and out of each trench annually.[5,6] However, the net amount of waste-generated gases transported to the atmosphere is governed by the biodegradation processes in the trenches, which constantly produce a slight net positive pressure against the atmosphere to move as little as a few cubic meters annually from a "young" trench to as much as a few hundred cubic meters annually from an "old" trench.[5,6]

Atmospheric dispersion, both transport and diffusion, of a radioactive gas can be described up to a few meters above the trench-cap surface. The diffusion equation has been solved as a microscale dispersion model to estimate the concentration of radioactive gases in air over low-level waste trenches and to distances of 50 to 100 m downwind.[8]

A conventional mesoscale diffusion model can be used to estimate concentrations of radioactive gases in air at greater distances from the trenches, but this work remains to be completed. Adaptation of a mesoscale model will require consideration of the fact that outward gas respiration is associated with decreasing atmospheric pressure created by passage of a low front. Since winds at West Valley usually flow from the south as each low front moves in, transport of respired gases tends to be dispersed to the north.

For the West Valley study measurements of gas concentrations beneath trench caps[12] were used in the GASFLO model to predict gas concentrations at the fractured surface of the caps.[4-6,8] Mixing into surface air was then projected via the ADIFF model[8] to obtain radionuclide concentrations in air downwind from a single trench to distances of 50 to 100 m.

The validity of the several model calculations have not been tested by concentration measurements in surface air. The ADIFF calculations suffer particularly from our inability to continue the field studies necessary to properly describe input data for the model, so only upper and lower bounds can be set for surface-air concentrations. Since the upper and lower bounds differ a millionfold, their relevance to 10CFR61 and/or 10CFR20 limits must be inferred as described below.

DISCUSSION

Concentrations of Respired Gases

If we assume that the maximum concentrations developed from the ADIFF model apply, radioactive gases emanating from the burial trenches may exceed, not only the long-term annual limit of 25 mrem to the public set by 10CFR61, but the 500-mrem annual limit of 10CFR20 as well.[14] Tritium (as hydrogen, methane, and hydrocarbons) would require 200,000-fold dilution to meet the 25-mrem limit. Respiration of tritiated water vapor would increase the concentration of tritium in air and require a correspondingly greater dilution, but transport of water vapor through the trench caps has not yet been evaluated sufficiently for meaningful calculation.

Although somewhat less dilution of ^{14}C, ^{85}Kr, and ^{222}Rn would be needed (20,000-, 30,000-, and 60,000-fold respectively), other factors must be considered in evaluating the long-term dose commitment. The long physical half-life of ^{14}C means that, unless biogenic processes decrease significantly with time, respiration of this radionuclide from the trench surface may continue to exceed 10CFR61 limits long after site closure and perhaps even past the period of institutional control. Since concentrations of ^{222}Rn increase as a function of trench age (measured as a function of time since closure)[9] and since its ^{226}Ra parent is long-lived, emanation of this gas may place restrictions on site operations, e.g., limits on the amount of ^{226}Ra accepted for burial or on the amount of biogenically degradable material which may be included with radium.

If transport and mixing occur more nearly as approximated by the minimum values obtained with ADIFF[8] all gases will meet 10CFR61 limits (by approximately 5-, 15-, 30-, and 50-fold respectively for tritium, ^{222}Rn, ^{85}Kr, and ^{14}C), although the margin by which the regulated limit is achieved for tritium will be narrowed somewhat by the contribution of tritiated water vapor. However, the boundary conditions used to develop these lowest concentrations do not appear representative of actual conditions as observed from wind-tunnel tests.[15]

If more representative values are used for the ADIFF model parameters, how far will the upper-bound concentrations be reduced? The most significant reduction is likely to develop from recalculation of the crack area relative to the total trench-surface area. Such an assumption is obviously incorrect, but no better assumption is available for insertion into the model at this time, Actually the crack area varies seasonally[5], so a variety of field measurements are required to obtain satisfactory estimates of the crack area presented to the atmosphere.

A more realistic estimate of gas concentrations downwind can nevertheless be obtained. The crack area can be roughly estimated from observations made during studies at West Valley that opening and closing of the sampling pipe on each trench altered the pressure differential between the atmosphere and the gases trapped beneath the cap.[5,10] Thus the crack area and pipe area must be similar if both are to affect the pressure differential at about the same rate. Constriction in the sampling pipes and in the perforations which open the pipes to the gas beneath the caps will reduce the cross-sectional pipe area to less than that observed at the land surface, so the estimated areas must be considered very poorly defined. Pipes used for gas measurements in the field varied from 50 to 400 cm in diameter. The trench surface area varies from 1500 to 2100 m^2, but here also constrictions beneath the caps may reduce the cap area in contact with the sampling pipes. Thus the ratio of crack area to trench area might range from about 10^{-6} to 10^{-3}, with a $\pm$ 10-fold error for each value.

Concentrations will not be reduced as severaly as the ratios would indicate, however. Combining field measurements with model calculations [4,13], we see that the mass transfer (volume of gas vented) changes only two- or threefold for a 100-fold change in the cap transmissivity coefficient (which is in turn proportional to the crack area in the Darcy flow calcula-

tion). Similar comparisons of mass flow to transmissivity for values below those normally observed in the field indicate only a 15-fold decrease in mass flow for a 1000-fold decrease in the transmissivity coefficient (crack area). Thus, the maximum concentrations obtained by Robinson using resistance-free flow from the trenches appear to be reduced at most about 15-fold when flow resistance is taken into account.

Robinson assumed[8] that the cracks extend smoothly and with uniform diameter throughout the cap (Darcy flow assumption) in order to obtain surface concentration values by the GASFLO model, which are three times the measured values at standard conditions. In fact, the fractures are much wider at the surface of the caps than at depth, and the trench gas which is compressed in flowing through the orifice at the bottom of the crack is likely to expand to the atmospheric conditions by the time it reaches the cap surface. While Robinson's assumption has little influence on the mass-transfer calculations performed with GASFLO[4], it appears to overestimate by threefold the surface concentrations.

Thus corrections for crack area, mass transfer, and departure from Darcy behavior suggest that realistic estimates of concentrations of radionuclides vented from a single trench would be in the range of 1 to 10% of the upper-bound values calculated by Robinson. These corrected concentrations still exceed 10CFR61 limits.

The chance of exceeding 10CFR61 and/or 10CFR20 limits is further enhanced if we consider that Robinson performed calculations only for air movement across a single trench. Air movement across a series of trenches would markedly increase the air concentration of each radionuclide but not necessarily in direct proportion to the number of trenches. (Upward mixing of respired gases from upwind trenches continues even as new material is introduced from the downwind trenches, and the trench-gas concentrations vary from trench to trench.) The same is true for airflow along the length of a trench when several tranches are in place. As a result, when an entire site is properly modeled, 10CFR61 limits are even more likely to be exceeded than is indicated by the corrected concentration estimates given above.

Finally, respiration of gases out of the trenches is associated primarily with atmospheric lows, which in turn are associated with a relatively fixed wind direction. The maximally exposed individual will likely experience a much greater dose than would be estimated from average annual winds.

Comparison to Water Pathways

Despite NRC's announced concern we have found conclusively that at West Valley the groundwater pathway is not significant.[3,4,6,10,11,13] Furthermore, radionuclide concentrations measured in water samples collected from the West Valley burial trenches themselves would generally not give a dose equivalent to the annual limit of 25 mrem established as a long-term performance objective in 10CFR61, even if the water were consumed directly.[12] Only ^{63}Ni, ^{3}HHO, ^{90}Sr, and ^{137}Cs would then require dilution - by 10,000-, 5,000-, 200- and 10-fold respectively. Sufficient dilution clearly does occur in the surface-water pathway at West Valley, as offsite samples of water pumped from the trenches and collected downstream would produce exposures <0.1 mrem/yr[11] - at least 250-fold less than the 10CFR61 limit.

Other Sites

The West Valley experience can likely be extrapolated to other sites. Studies conducted at Brookhaven National Laboratory of trench-water composition and biogenic character[7] have shown the close relationship among burial sites in wet climates. (Although only a single report is cited here, a voluminous literature is available from the Brookhaven authors). Gas production at sites with unsaturated trenches may be at least as great as at West Valley.[16]

Preliminary measurements were performed on trench gas samples collected by the U.S. Geological Survey from two of 21 trenches at Sheffield, Illinois. The two Sheffield trenches appear biogenically "younger" than those at West Valley which were closed at about the same time (i.e., gas production is more aerobic). Although tritium and ^{14}C concentrations are less than at West Valley, ^{222}Rn concentrations beneath the caps are at least as great. Since the controlling mechanism for transport of ^{222}Rn through the cap is biogenic gas production, ^{222}Rn concentrations at the surface of the Sheffield trenches may not be at a corresponding level. The degree to which ^{222}Rn venting at Sheffield exceeds the 10CFR61 limit should be evaluated now and again later, as the biogenic gas production rate increases from increasingly anaerobic decomposition of the buried wastes (See reference 9 for a discussion of gas production mechanisms.)

Finally, the annual intake values (in pCi/year) of ^{210}Pb (from decay of ^{222}Rn), ^{3}H, and ^{14}C via gas pathways at West Valley exceed those for the same radionuclides via groundwater and resuspension pathways at Barnwell, South Carolina[17], by factors of 10^{25} (!), 10^{7}, and 10^{4} respectively. For the Barnwell site, at least, studies of the gas pathway may be more cost-effective than would a continued effort to define the groundwater pathways.

Incineration as an Alternative to Burial

Respiration of gaseous ^{14}C from low-level waste trenches means that most of the buried ^{14}C will eventually reach the atmosphere, as will a lesser but significant fraction of the buried tritium. Thus the waste classification and handling required by 10CFR61 will do little to reduce the collective population dose from these radionuclides. Burial of ^{14}C wastes, in particular, serves only to shift the point of release from the generator to the burial site. If ^{14}C must be buried, a more stable waste form should be considered for the biodegradable wastes.

Biogenic gases also appear to carry ^{222}Rn through the trench caps at a sufficient rate to survive physical decay. Segregation of ^{226}Ra from the biodegradable wastes may therefore be necessary, if the biodegradable wastes are not incinerated.

Although the ^{85}Kr respiration rate also appears to be controlled by biogenic gas production, its physical decay (unlike ^{222}Rn) will not significantly reduce the total amount released. The time over which the release occurs can be extended, however, by reducing the respiration rate of biogenic gases, either by segregation or incineration. Individual exposures at the burial site will be correspondingly decreased.

An alternative to simple burial of biodegradable wastes may be incineration at *the generator* site or even at the burial site. Incineration offers an opportunity to release the ^{14}C and tritium to the atmosphere at a controlled rate and with sufficient buoyancy to

reduce individual exposures well below the limits of 10CFR61. Such a step would also reduce ^{222}Rn and ^{85}Kr exposures around the burial site.

Incineration of biodegradable wastes appears to offer a plausible approach for reducing individual exposures from water pathways as well, whether or not the exposures exceed 10CFR61 limits. Compaction of wastes by incineration would greatly reduce the void space within the trenches and the gas pressure against the caps, thus helping to stabilize the trenches against water infiltration.

These advantages of incineration suggest that a recent modification[18] to 10CFR20, which permits greater use of local incineration of biomedical wastes containing small quantities of tritium and ^{14}C, should perhaps be extended to wastes produced in even greater quantities by other generators as well. Since unrestricted release of ^{14}C and tritium will not add significantly to the worldwide inventory of either radionuclide[19], there appears little reason to continue restrictions on incineration beyond those necessary to maintain safe levels of exposure at the sites of incineration.

CONCLUSIONS

Estimates of breathing-zone concentrations of gases respired from the low-level waste burial trenches at West Valley indicate that the gas pathway is significantly more important than the water pathways, although neither presents an imminent health hazard. This conclusion can apparently be applied to other sites as well. Regardless of whether sites are wet or dry, doses from respired gases appear likely to exceed the exposure limits set by 10CFR61.

Reduction of exposure levels may be better accomplished by more extensive use of incineration, which will also aid greatly to stabilize the trench caps. If incineration is not feasible, segregation of radium and ^{85}Kr sources from biogenically degradable wastes may be desirable.

Further study is required of the implications of gas production and transport, as much for improvement of burial procedures and site maintenance as for dose reduction.

ACKNOWLEDGMENT

This discussion has drawn extensively from earlier publications by staff of the Radiological Sciences Institute, especially those of Drs. An-Hua Lu and Charles O. Kunz.

REFERENCES

1. U.S. Nuclear Regulatory Commission, "Licensing Requirements for Land Disposal of Radioactive Waste," Fed. Reg. 46, 38081 (1981).

2. U.S. Nuclear Regulatory Commission, "Draft Environmental Impact Statement on 10CFR61, Licensing Requirements for Land Disposal of Radioactive Waste," NUREG-0782, Vol. 1 (September 1981).

3. J. M. Matuszek, L. Husain, A. H. Lu, J. F. Davis, R. H. Fakundiny, and J. W. Pferd, "Application of Radionuclide Pathway Studies to Management of Shallow, Low-Level Radioactive Waste Burial Facilities," Symposium on the Management of Low-Level Radioactive Waste, Atlanta, Georgia (1977), Pergamon Press, New York (1979).

4. A. H. Lu and J. M. Matuszek, "Solute and Gas Transport from Radioactive Waste," Trans. Amer. Nucl. Soc. 30, 94 (1978).

5. A. H. Lu and J. M. Matuszek, "Transport Through a Trench Cover of Gaseous Tritiated Compounds from Buried Nuclear Wastes," International Atomic Energy Agency Symposium on the Behavior of Tritium in the Environment, San Francisco, California (October 1978), IAEA, Vienna, 1979.

6. J. M. Matuszek, "Biochemical and Chemical Processes Leading to Radionuclide Transport from Low-Level Waste Burial Sites," Trans. Amer. Nucl. Soc. 34, 155 (1980).

7. A. J. Francis, S. Dobbs, and R. F. Doering, "Biogenesis of Tritiated and Carbon-14 Methane from Low-Level Radioactive Waste," Nucl. Chem. Waste Manage. 1, 153 (1980).

8. L. W. Robinson, Jr., "Modeling Atmospheric boundary Layer Transport and Diffusion of Radioactive Gases from a Low-Level Waste Burial Area," Rensselaer Polytechnic Institute Thesis (August 1980).

9. C. O. Kunz, "Radioactive Gas Production and Venting at a Low-Level Radioactive Waste Burial Site," Nucl. Chem. Waste Manage. 3, 185 (1982).

10. A. H. Lu, "Modeling of Radionuclide Migration from a Low-Level Radioactive Waste Burial Site," Health Physics, 34, 39, 1978).

11. J. M. Matuszek, F. V. Strnisa, and C. F. Baxter, "Radionuclide Dynamics and Health Implications for the New York Nuclear Services Center's Radioactive Waste Burial Site," International Atomic Energy Agency Symposium on the Management of Radioactive Wastes from the Nuclear Fuel Cycle, Vienna, Austria (March 1976).

12. L. Husain, J. M. Matuszek, J. Hutchinson, and M. Wahlen, "Chemical and Radiochemical Character of a Low-Level Radioactive Waste Burial Site," Symposium on the Management of Low-Level Radioactive Waste, Atlanta, Georgia (1977), Pergamon Press, New York (1979).

13. J. M. Matuszek, "Radiochemical Measurements for Evaluating Air Quality in the Vicinity of Low-Level Waste Burial Sites - The West Valley Experience," Symposium on Low-Level Waste Disposal: Site Characterization and Monitoring, Arlington, Va. (June 1982), U.S. Nuclear Regulatory Commission, NUREG/CP-0028, CONF-820674, Vol. 2, December 1982.

14. U.S. Nuclear Regulatory Commission, "10CFR20 - Standards for Protection Against Radiation," Code of Federal Regulations, U.S. Government Printing Office, Washington, D.C. (1982), page 214.

15. O. G. Sutton, Micrometeorology, McGraw-Hill, New York (1953), pages 105-156, 229-241 and 273-283.

16. P. Colombo, Brookhaven National Laboratory, private communication (1982).

17. D. E. Fields, C. A. Little, C. J. Emerson and G. Hiromoto, "Shallow-Land Burial of Low-Level Radioactive Wastes: Preliminary Simulation of Long-Term Health Risks," Waste Management '82 (March 1982), University of Arizona Press, in press.

18. U.S. Nuclear Regulatory Commission, "10CFR20-Biomedical Waste Disposal," Fed. Reg. 46, 16230 (1981).

19. M. Eisenbud, "The Status of Radioactive Waste Management: Need for Reassessment," Health Physics, 40, 429 (1981).