TRANSPORT OF LOW-LEVEL RADIOACTIVE SOIL AT DEEP-OCEAN DISPOSAL SITE

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ABSTRACT: Transport studies were conducted to assess ocean disposal of soil contaminated with low-level natural radioisotopes. The experimental approach involved characterization of the soil for parameters affecting transport and fate of radionuclides. Radioactivity was associated with discrete soil particles up to 6,000 μm. Gross and most individual isotope and element activities were more concentrated on smaller particles. The median size based on gross radio-activity was 125 μm, while the median size based on dry mass was 350 μm. Particle-settling velocities measured in a 1 m settling column ranged up to 8.2 cm/s (median 2.1 cm/s). Large-scale mixed water column experiments (5 m) confirmed that settling would be the dominant verticle transport mechanism for the soil particles. Experimental results were used to calibrate a convective-diffusive transport model, which was applied to a hypothetical ocean disposal site in 4,000 m of water. The model indicated that 95% of the soil and associated radionuclides would impact the bottom sediment within five days and up to 40 km from the disposal point, along the mean current flow.

INTRODUCTION

The ocean contains natural radioactivity, both long-lived radionuclides and their daughter products, and relatively short-lived radionuclides formed by the interaction of cosmic radiation. A liter of seawater contains approximately 350 pCi, and a kilogram of deep-sea sediment contains approximately 1×10^4 pCi (Park et al. 1983). Comparing these data with the ocean-volume-averaged anthropogenic radioactive loading indicates that there has been a background activity increase in about 0.1% since 1944 (Knauss 1984).

In 1981, Sandia National Laboratories initiated an evaluation of potential disposal sites, including the oceans, for soils contaminated with low, but significant, levels of natural radionuclides under the Department of Energy's Formerly Utilized Sites Remedial Action Program (FUSRAP). This program was designed to evaluate and clean up numerous inactive industrial plant sites remaining from the Manhattan Project (Kupferman et al. 1984). Sandia identified one of these sites, the sampling plant material at Middlesex, New Jersey, as a candidate for possible ocean disposal. Efforts were begun to document the feasibility and advantages of ocean disposal of the soils and rubble from this site and to provide information useful for evaluating potential permit applications to the U.S. Environmental Protection Agency (EPA) (Kupferman et al. 1984).

Approximately 95,000 m^3 (about 200,000 metric tons) of contaminated soil are stockpiled at the Middlesex site, with an associated maintenance

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cost of $200,000/year. The spatially averaged radioactivity has been estimated at between 40–80 pCi/g (as ^{226}Ra) by Dickson et al. (1977) and Leggett et al. (1980), and at 108 pCi/g (as ^{226}Ra) by Kupferman et al. (1984). These investigators also indicate that the isotopes of the ^{238}U and ^{232}Th decay series are in isotopic equilibrium, meaning that parent activity is equal to all of the individual daughter activities. Projected 1981 disposal cost estimates for the Middlesex soil ranged from $1,000,000 to $40,000,000, depending on assumptions concerning transportation, containment, and whether land or ocean disposal options were chosen. In general, ocean disposal of noncontained waste is the least-expensive option. Contained waste would tend to concentrate radionuclides on the ocean floor but would reduce water-column contamination during descent, and it would reduce potential surface contamination due to floatable material. This last consideration may not be important, as Hunt (1986) indicates that less than 1% of the total radioactivity in Middlesex soil is contained in floatable material. This study addressed the issue of radionuclide mobilization for short equilibrium times and found that most activity would be associated with settleable particulates.

The results of the Sandia studies were useful but not sufficient to determine the exposure fields that would result from disposal of the soil in the marine environment. A parametric study conducted by Klett and Klett (1984) was submitted as scoping calculations in the Kupferman et al. (1984) report to bracket velocities and settling times of FUSRAP soil dumped at sea in 4,000 m of water. Extreme-condition calculations indicated that the soil would arrive at the sediment some time between 4.5 min and 174 years. These numbers reflect assumptions concerning the dominance of the vertical transport by plume dynamics after release from a barge and gravitational settling of very small particles. Clearly, refined estimates of bottom-arrival time are needed in the examination of marine disposal as a management option.

The primary purpose of the present study was to provide better upper-bound estimates for the maximum water-column exposure time of the FUSRAP soil from Middlesex. This involved estimation of the settling time and horizontal travel distance of the contaminated soil for a given ocean depth and current. To accomplish this, process-oriented experiments were conducted to calibrate a transport model. Development of the model required estimates of soil-settling rates and physical and chemical properties that would modify the contaminants in the soil on contact and passage through seawater. The specific study objectives were to: (1) Characterize the Middlesex soil for parameters affecting transport; (2) develop and calibrate a vertical-transport model using data from the first objective; and (3) apply this model to estimate the transport and fate of the radionuclides associated with FUSRAP soils at a hypothetical deep-ocean disposal site.

The experimental efforts necessary for this development included the following: (1) Measurement of particle specific gravity, size and activity distributions, and radionuclides dissolution for representative soils from the Middlesex site; (2) measurement of dynamic soil-settling velocity distributions under quiescent hydrodynamic conditions; (3) simulation of deep-ocean vertical dispersion conditions over a range of ocean values (0–26 cm^2/s) by modification of large-scale water columns at the Marine Ecosystems Research Laboratory (MERL), at the University of Rhode Island; and (4) measurements of vertical transport of Middlesex soil in the mesocosms under turbulent conditions typical of the deep ocean.

The model used to describe transport of this soil in the ocean depended on the characteristics of the soil and behavior of associated contaminants. Tightly bound contaminants required a description of particle transport alone to determine the exposure-concentration fields of the contaminant. Our model only considered particle-bound radionuclides, although preliminary data indicated that approximately 10% of the gross activity could be solubilized during descent. This fraction of activity would be lost directly to the water column.

Our efforts indicated that a particulate convective-diffusive model can be used to describe the transport of natural radionuclides associated with soil from the FUSRAP site. When applied scaled to deep ocean conditions, this model predicted that the vast majority of soils (>95%) would impact the bottom sediments at a 4,000 m disposal site within 4.5 days and up to 40 km away, along the direction of mean current flow.

Experimental Methods

Samples of soil were excavated from the Middlesex FUSRAP site in 1982, sieved through a 1/4 in. (about 6,350 μm) mesh, characterized on site for total radioactivity, homogenized, and divided into 50 kg replicate batches. The results in this paper are based on the sample sets that had the highest activity (about 500 pCi ^{226}Ra/g dry weight). Specific activity determinations were carried out in 1982 by Sandia and in 1984 by the Eastern Environmental Radiation Facilities (EERF) of EPA, in Montgomery, Alabama.

The specific gravity of the Middlesex soil sample was determined by using standard pycnometric procedures at 20°C. The average-particle specific gravity was 2.31. Throughout this study this value was assumed for all size categories.

Particles obtained from a dry sieving size classification were analyzed for total radioactivity by two particles. A gross measure of total activity was determined on each size class by counting total gamma plus beta and total alpha particles emanating from a known mass of sample with a Nuclear Measurements, Inc., PC-5 proportional counting system. Results were calculated in terms of disintegration per minute per gram dry sample (DPM/g) and corrected for incident background radiation. These same samples were then sent to EERF for quantification of the radioisotopes ^{226}Ra, ^{210}Pb, ^{210}Po, ^{234}U, ^{235}U, ^{238}U, ^{227}Th, ^{228}Th, ^{230}Th, and ^{232}Th. In addition, autoradiography of the soil particles in several size classes was conducted.

The size distributions of the soil were measured by two methods. Method one involved oven drying 100 g of soil at 70°C to determine percent moisture. The dried sample was then sieved for 20 minutes with a Ro-Tap particle sieve shaking system (Carver 1971) using 0.5 ϕ-size increments between 4.0 and 1.0 ϕ units (63 μm and 2,000 μm). The soil retained on each sieve was quantified in terms of mass and the mass percent, and a cumulative size distribution was determined. The mass of particles passing through 63 μm sieves (0–< 63 μm) was collected and quantified.

The second technique utilized a HIAC-SSTA automatic particle counter calibrated to count particles in five sizes between 20 μm and 600 μm. The counting sensor was positioned vertically at the bottom of a 1 m bench-scale experimental settling column (Fig. 1), filled with 0.4 μm filtered lower Narragansett Bay seawater [salinity about 30 parts per thousand (ppt)]. The column was constructed using 20 cm diameter polyvinyl chloride (PVC) pipe. A known mass of soil was added at the top of the column and allowed

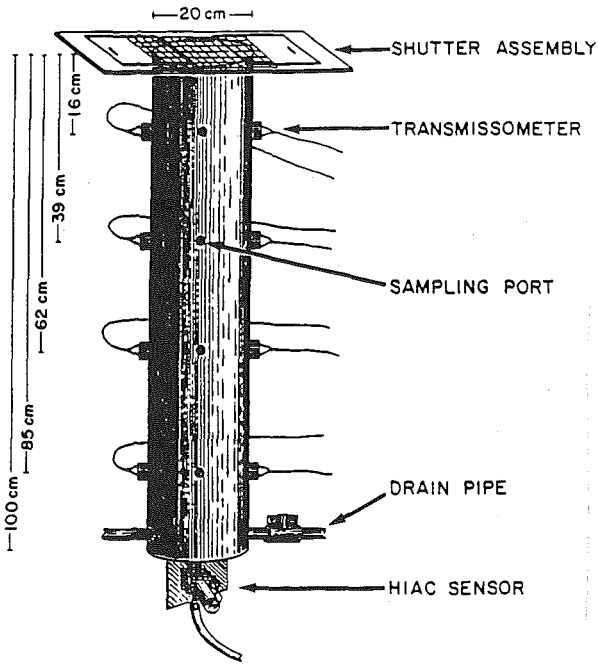


FIG. 1. Laboratory Settling Column

to settle through it. Total particle counts were maintained at less than 100/ml to avoid coincidence counts by the sensor. The hydrostatic head from the column was sufficient to force the fluid through the sensor at 380 ml/min. Bias resulting from differential particle arrival time was eliminated by continuing the experiment until background count rates were obtained. This method provided a measure of the dynamic behavior of the wetted particles. Particle counts were transformed to mass by multiplying average particle volume (assuming spherical particles) for a given size range by the measured average soil density.

Comparison of size classes between the two techniques was accomplished by adjusting and normalizing the dry size results (Bonner et al. 1985) to correspond with the HIAC automatic particle counter results. Numerical interpolation and integration techniques were used to reduce the dry cumulative size distribution (due to higher resolution data) from a range of 0–2,000 μm to 20–600 μm. Polynomial interpolation was used to determine absolute mass in size categories 20–40, 40–100, 100–200, 200–300, and 300–600 that are consistent with the HIAC wet-sizing technique. These data were then numerically integrated to generate a cumulative dry size distribution that was consistent with the HIAC cumulative wet size distribution.

Soil-settling velocities were also determined with the column in Fig. 1. Transmissometers interfaced to a microcomputer were mounted externally on the side of the column (with glass windows) at four depths (16, 39, 62, and 85 cm) to measure the attenuation of infrared light caused by the passage of particles as a function of time. Soil samples were added instantaneously across the entire surface area of the column by means of gridded shutter,

which was located 3 mm above the water surface. The shutter was interfaced with a microcomputer and triggered the transmissometer sampling when opened. Initially, the intensity of each transmissometer was matched by equalizing electronic outputs to filtered seawater.

As the descending particles settled past the transmissometers, the attenuation of the light beams increased in direct proportion to the particle-mass concentration ($C_k = \alpha_k R_k$) at each depth k (Bonner et al. 1985). For all analyses it was assumed that this proportionality (α_i) was a constant for each particle size and depth. The resulting time-dependent analog signals from each transmissometer (R_k) were digitized and the values stored. The time-dependent particle-mass flux at each depth was assumed to be advection (settling) dominated and equal to concentration multiplied by particle-settling velocity:

$$\text{flux}_k = vC_k \quad \text{(1)}$$

where v = Stoke's particle-settling velocity corresponding to a mass fraction at a specific time and is equal to depth$_k$/time assuming fluid velocity equal to zero. The flux at each depth was integrated over time and normalized, resulting in the fraction of the total mass passing a given depth as a function of time. The time-dependent cumulative mass fraction at each depth was represented as

$$f_k = \frac{h_k \int_0^t \frac{C_k}{t}\,\mathrm{d}t}{h_k \int_0^\infty \frac{C_k}{t}\,\mathrm{d}t} = \frac{h_k\alpha_k \int_0^t \frac{R_k}{t}\,\mathrm{d}t}{h_k\alpha_k \int_0^\infty \frac{R_k}{t}\,\mathrm{d}t} = \frac{\int_0^t \frac{R_k}{t}\,\mathrm{d}t}{\int_0^\infty \frac{R_k}{t}\,\mathrm{d}t} \quad \text{(2)}$$

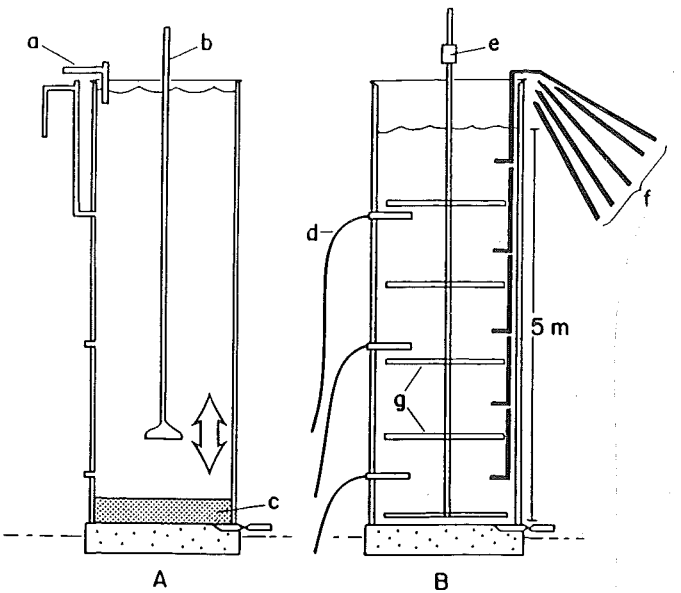
where h_k = depth of each transmissometer (Ducharme 1989). This ratio represents the mass passing location k at time = t divided by the total mass passing location k over all time. The time for all particle mass to pass location k ($t = \infty$) was determined by measuring R_k until it dropped to background levels (i.e. response for filtered seawater). The cumulative velocity distribution at each depth was determined by relating cumulative percent mass at each time to the depth (h_k) divided by time. The settling velocity distribution was, therefore, determined by measuring the time of travel for each fraction of total mass through a known distance (velocity = distance/time).

Mixed Water Column Experiments

A series of experiments in Marine Ecosystems Research Laboratory (MERL) mesocosms of the University of Rhode Island were designed to quantify the effects of turbulent mixing on settling rates to provide more realistic mass-transport estimates for the model predictions. These experiments were accomplished by modifying the normal up/down plunger mixing used by MERL (Nixon et al. 1980) with axially driven impellers, which rotated 360° in a clockwise/counterclockwise mode (Fig. 2). This design provided uniform mixing throughout the water column, as quantified by dye studies (Bonner et al. 1985).

The vertical dispersion in the mesocosm tanks was quantified at three levels of mixing. This was accomplished by first allowing the impeller to operate at a set rate sufficiently long for a dynamic equilibrium to be established in the tank. Rhodamine-WT dye was then sprayed onto the water

J. Environ. Eng. 1992.118:101-119.



a. Inflow to mesocosm
b. Plunger
c. Sediments
d. Sampling ports for suspended solids measurements
e. Axially driven impeller
f. Sampling ports for dye measurements
g. Impeller

FIG. 2. MERL Mesocosm: (*a*) Normal Up/Down Plunger Mixing; (*b*) Axially Driven Impeller Mixing

surface, and the concentration was measured at depths (0.5, 1.5, 2.5, 3.5, 4.5 m) as a function of time. These data were used to calibrate a simple dispersion model.

The effects of water-column mixing on the transport of the soil were tested by adding a known mass (about 4 kg) of soil, uniformly and instantaneously, to the surface of the water column for a fixed dispersion. Particle mass was intermittently sampled from three 2.5 cm diameter siphon tubes, 1.2, 2.2, and 4.4 m below the water surface. Samples (4.0 l at the rate of 1 l/s) were taken on a log time scale for up to 4 hr after soil addition.

Particles were separated from suspension by filtration. Particles greater than 63 μm were separated with a sieve, rinsed onto a filter, and total mass for the known volume calculated. The mass of <63 μm particles was determined by collecting a 500 ml aliquot of water after passing the sieve, then quantifying the mass retained on passage through a 1 μm filter. The mass per unit volume was calculated and added to the >63 μm fraction to give the concentration of total particulates.

Experimental Results

Size Distributions

Ninety-five percent of the Middlesex soil particle mass was less than 6,350 μm, and 90% was measured between 63 μm and 6,350 μm. Cumulative mass fraction versus particle size showed that the soil was dominated by sand size particles smaller than 1 mm (Fig. 3) with a median grain size of approximately 350 microns.

Comparison of bulk-sample size classifications between 20 μm and 600 μm (this range represents 62% of the total mass) by the wet and dry methods indicated that larger sizes were determined with the wet sizing technique (Fig. 4). This may have resulted from mechanisms such as particle agglomeration, surface wetting, and swelling from hydration, as well as from differences in the measuring techniques. Comparison of dry and wet size-distribution data for particles from single dry sieving size classes (63–90, 90–106, 106–180, and 180–355 microns) supported the apparent size shifts (Bonner et al. 1985). This tendency was less pronounced for the larger size classes than for the smaller size classes.

Activity Distributions

Significant radioactivity was found associated with each particle size class using the total beta plus gamma counts. The activity found in individual size classes was not the same as observed for whole samples, indicating non-uniform distribution of the radioactivity within the sample. The contribution to total sample activity by each size class was estimated by normalizing the activity in each size class with the mass fraction for that size class (i.e., DPM/g fraction $i \times g$ fraction i/g total sample). Cumulative distributions for mass and activity shown in Fig. 3 indicates median activity to occur at

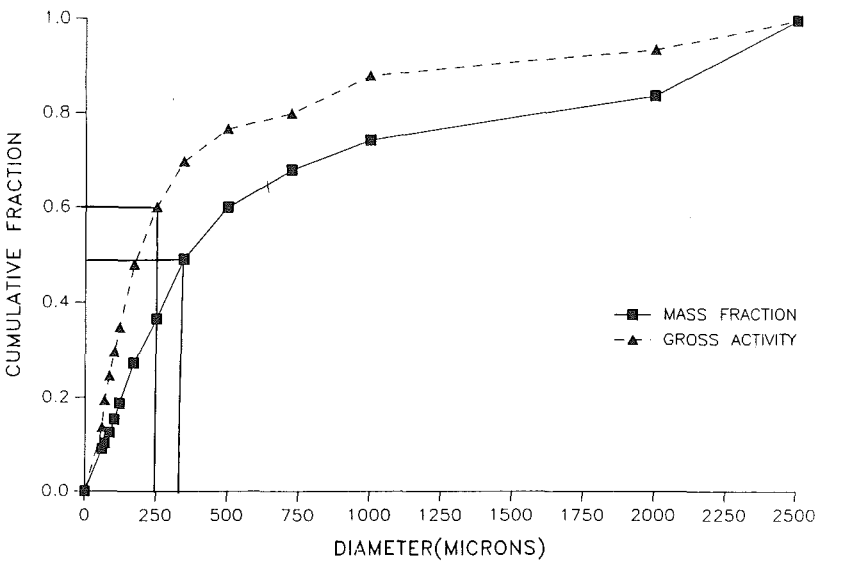


FIG. 3. Cumulative Mass Fraction and Gross Activity as Function of Particle Size (Dry)

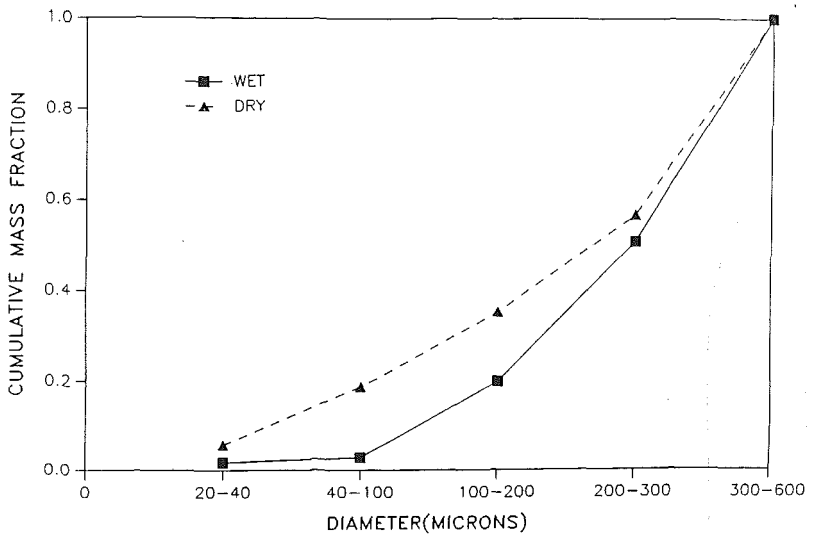


FIG. 4. Size Distribution between 20 and 600 μm as Determined by Wet and Dry Methods

the 125 μm size class. Measurement of individual isotope activity showed significant activity in each fraction when normalized to the mass fraction (Table 1). This table of results indicate that less than 10% of the activity was found associated with the silt-clay fraction (<63 μm) of the sample.

Autoradiography of these soils (Fig. 5) demonstrated the activity (large black dots) was associated with discrete, individual particles (small black dots). The discrete nature of the activity indicates the isotopes may be associated with residual ore particles originally at the plant.

The mass-weighted cumulative activity distributions for individual isotopes were different from the cumulative mass and the cumulative gross activity distributions (Fig. 6). The individual isotopes that were measured exhibited higher median sizes for activity than did the measure of gross activity, with ^{226}Ra, ^{234}U, ^{238}U, and ^{230}Th having similar median values (250 μm); ^{210}Pb and ^{210}Po had higher median distributions, namely 300 μm and 600 μm, respectively. The Pb distribution was biased by an abnormally large activity in the 1,000–2,000 μm size class relative to other isotopes in the ^{238}U series. Several isotopes—^{227}Th, ^{228}Th, ^{232}Th, and ^{235}U—did not show consistent cumulative activity distributions due to large counting errors resulting from low activity and small sample sizes.

Preliminary short-term studies (20 hr) (Bonner et al. 1985) and long-term studies (90 days) Hunt (1986) indicate that up to 25% of ^{226}Ra and the U isotopes would be mobilized to the dissolved phase after initial contact with seawater. This mobilized fraction represented less than 10% of the initial total gross activity. Therefore, with the exception of the fraction of the isotopes mobilized to the dissolved phase, transport of the isotopes should be described by processes controlling the transport of the particulate material.

TABLE 1. Radionuclide Activity for Middlesex Soil

Particle diameter (μm) (1)	^{226}Ra 2–3% (2)	^{210}Pb 14–41% (3)	^{210}Po 7–12% (4)	^{234}U 12–16% (5)	^{235}U 17–79% (6)	^{238}U 12–16% (7)	^{227}Th 26–62% (8)	^{228}Th 10–218% (9)	^{230}Th 6–15% (10)	^{232}Th 8–69% (11)
<63	50.3	56.0	57.2	49.2	4.6	49.1	4.3	7.0	54.9	8.2
63	8.6	8.4	8.4	9.7	3.8	9.7	0.7	1.1	10.2	1.2
75	14.2	19.1	13.4	11.4	2.3	11.8	0.9	1.3	11.2	1.5
90	15.1	16.4	13.7	17.3	0.8	16.2	1.0	1.6	15.2	1.3
106	13.6	14.9	15.9	14.8	1.8	14.0	1.1	1.5	13.5	1.7
125	59.3	61.5	52.9	65.4	2.4	63.9	3.9	4.2	57.9	47.9
180	28.5	27.7	37.7	26.6	1.1	28.1	2.3	1.9	31.6	2.0
250	24.9	29.9	27.2	30.1	1.8	28.5	1.7	0.9	21.7	1.3
355	20.8	19.9	19.3	14.2	0.7	15.2	1.2	0.5	16.6	1.3
500	18.8	29.2	20.0	24.8	1.1	24.7	2.2	<0.1	23.5	1.3
710	17.3	32.5	26.3	35.1	1.9	33.5	22.1	0.3	31.6	0.3
1,000	58.3	149.3	64.7	53.7	2.2	56.2	9.7	52.9	84.4	76.0
>2,000	29.1	49.6	31.8	24.1	0.9	22.2	3.2	<0.1	31.9	1.3
Total	358.8	514.5	388.3	376.5	25.5	373.2	54.2	73.1	404.1	145.2

[a]Percent counting error specified as two standard deviations.

Note: Activity expressed in pCi/g times mass fraction in that sieve size (g/g).

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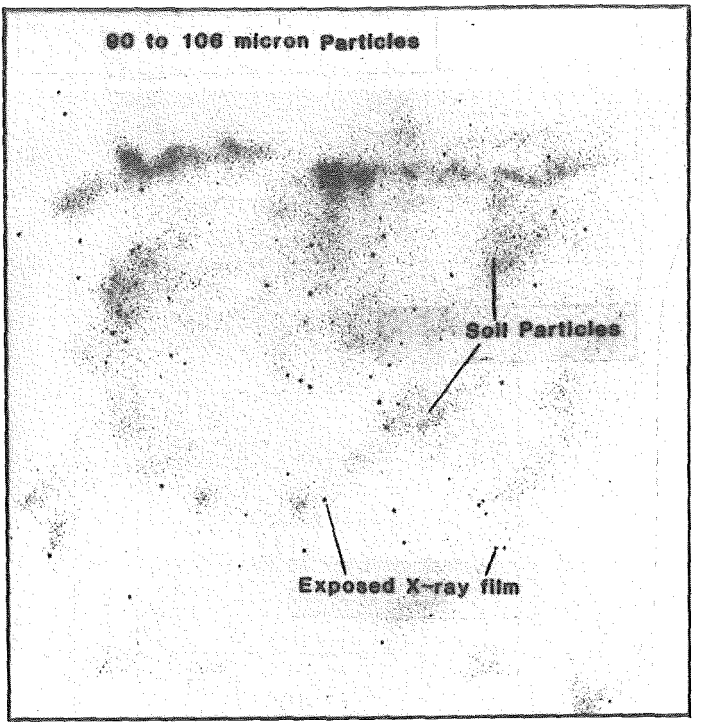

FIG. 5. Autoradiograph of 90–106 μm Middlesex (FUSRAP) Soil Particles

Settling Velocity Distributions

Soil settling velocities were determined for a series of dried composite Middlesex samples and a series of selected size classes that had been dried. Fig. 7 shows the observed settling velocity distributions for the dried composite Middlesex soil; 20% of the composite sample settled at greater than 5 cm/s in the upper 16 cm of the column. However, the fraction of particles descending past 39, 62, and 85 cm at velocities greater than 5 cm/s decreased to 10%. The momentum from the actual soil addition could have resulted in the observed decrease or other causes such as wetting or hydration of particles and disaggregation of soil clumps may be theorized. Regardless of the cause, the shift to a slower velocity was complete at 39 cm.

Minimum settling velocities for the composite sample increased from 0.2 cm/s at 16 cm to 1.3 cm/s at 85 cm. The median settling velocity followed a similar trend as did velocity distributions for individual size classes of dried soils (Bonner et al. 1985), particularly in the smaller size classes. The observed shift in velocity distributions suggests the particles had not reached a complete equilibrium state by 85 cm, possibly due to wetting or hydration and agglomeration of soil particles.

The settling-column results indicated that the Middlesex soil will fall rapidly through the water column, and that changes in settling velocity distribution would occur upon introduction into seawater. Whatever mechanism is responsible for the observed change, the increased velocities act

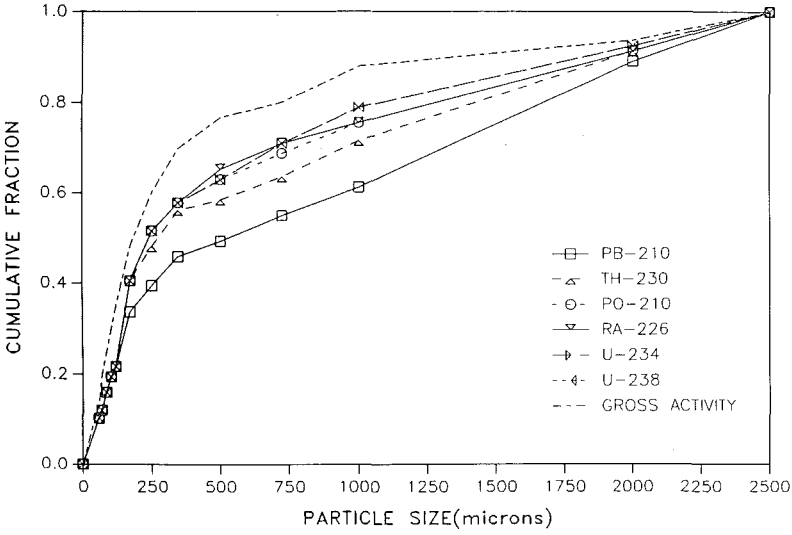

FIG. 6. Cumulative Individual Isotope Activity as Function of Particle Size

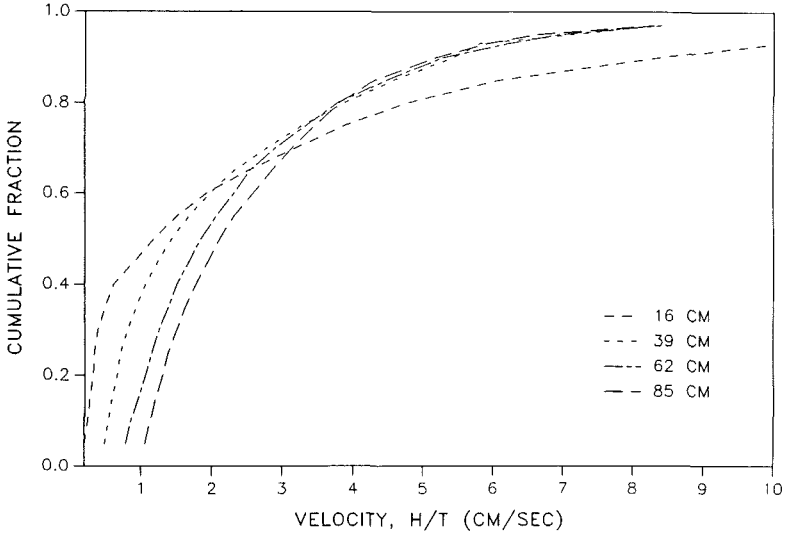

FIG. 7. Settling-Velocity Distribution for Middlesex Soil Determined from Laboratory Column Experiment

to reduce the water-column residence time for the finer particles. These particles begin to approach an equilibrium state in terms of size and velocity during the descent in the 1.0 m experimental settling column (Bonner et al. 1985). At a 4,000 m deep-ocean disposal site it can be safely assumed that the distance and time of travel to reach equilibrium would be relatively

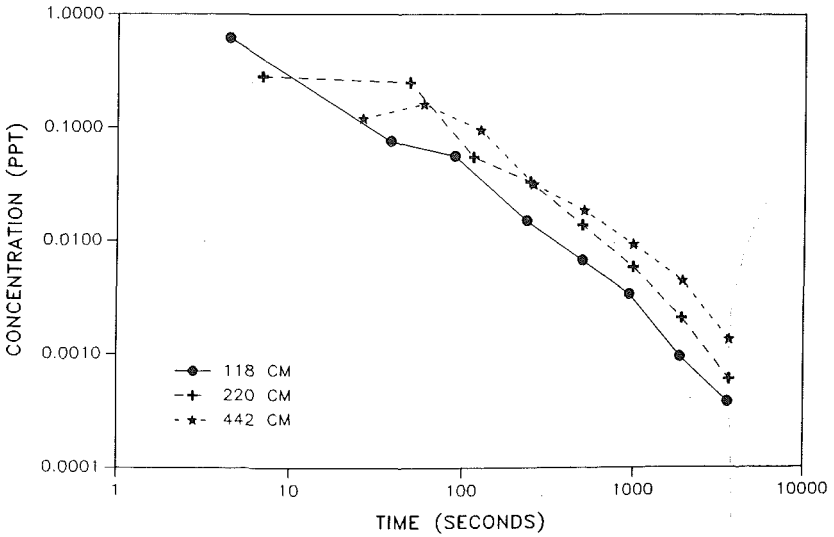

FIG. 8. Mass-Removal Experimental Results for Particles Greater than 63 microns and with 11 cm²/s Dispersion in MERL Mesocosm

small compared to total water-column depth and time to reach the bottom sediments.

Mesocosm-Scale Experiments

Vertical transport of particles under mixed-turbulent conditions is significantly different than under quiescent conditions (Lawler et al. 1983) and (Friedlander 1977). This is not only due to enhanced transport due to dispersion, but also the particle-particle interaction (breakup and flocculation). These processes affect the particle-size distribution and therefore the advective portion of transport formula. The effects of mixing and quantified vertical dispersion on the transport of the soil in the mesocosm tank was difficult to determine. The rapid descent of the particles in the MERL mesocosm limited the quality of data. Approximately 4 sec were required to obtain the 4.0 l used to determine the solids concentration. Thus, it was difficult to obtain synoptic samples for a given time. These sampling constraints did not permit detection of a peak in mass descending through the tank during the experiments (see Fig. 8). In spite of this, we were able to estimate the total mass of particles (1–6,350 μm) remaining in the mesocosm at various depths as a function of time and thus determine the mass removal from the water during the experiments. These results (Table 2) suggest that higher mixing and vertical dispersion increased the residence time of particles in the 5 m MERL water column. Differences in mass removal at different dispersions could be accounted for by an analytical expression relating gravitational settling (V) and vertical diffusion (D) (Table 2). In Table 2 V is estimated from mass-removal time by the following expression [derived from results in Csanady (1973)]:

$$X = \frac{1}{2}\left[\text{erf}\left(\frac{b - Vt}{2\sqrt{Dt}}\right) + \text{erf}\left(\frac{b + Vt}{2\sqrt{Dt}}\right)\right] \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots (3)$$

TABLE 2. Effects of Dispersion on Mass Removal of Middlesex from MERL Mesocosm Experiments

Mass removed (%) (1)	DISPERSION			
	11 cm²/s Mixing		26 cm²/s Mixing	
	t^a (sec) (2)	V^b (cm/s) (3)	t^a (sec) (4)	V^b (cm/s) (5)
50	94	5.3	78	6.4
84	229	2.5	363	1.7
90	385	1.6	611	1.2
95	1,322	0.6	2,543	0.4

[a]Estimated by volume-weighting concentrations for a sampling period, and linearly extrapolating between sampling periods.

[b]Estimated from mass-removal time according to (3).

where X = fraction remaining in water column; erf ( ) = error function; b = depth of water column; V = settling velocity; t = time for removal; and D = vertical diffusion. Extrapolating this analytical expression to a 4,000 m water column suggests that dispersion levels comparable to those used in the MERL tank would modify mass removal times by less than 5%. The vertical dispersion levels in the MERL tank experiments are within the range observed in the open ocean (Sverdrup et al. 1942).

Modeling Approach

Model Development

Particulate matter is vertically transported through the water column by a combination of convection, gravitational settling, and diffusion (turbulent). The model used here assumes that vertical convection of the fluid, including convective descent upon barge release, was not significant in estimating descent through the entire water column. It has been shown that vertical fluid convection is only significant in large water bodies where strong upwelling and downwelling occur, e.g., in nearshore coastal regions; and that settling and diffusion are ubiquitous phenomena (Lick 1982). The vertical distance over which convective descent and plume dynamics are important is just a small fraction of the water column in the deep ocean. The modeling approach further assumed the particles can be treated as a system of independent (or noninteracting) size classes, each with its own characteristic settling velocity.

The one-dimensional, time-dependent, convective-diffusive transport model presented here involved two vertical transport terms, settling and diffusion. The settling term for noninteracting, single-sized particles was the mean settling velocity of the material relative to the fluid V (the velocity of the fluid V_f minus the particle fall velocity V_p) multiplied by the concentration. The particle-fall velocity, V_p, was determined from laboratory settling-column experiments. Alternatively, V_p could have been expressed as a function of particle diameter for at least three different Reynolds-number regimes (Weber 1972). Vertical diffusion was characterized by the vertical eddy diffusivity coefficient, D_v.

The equation describing the vertical transport of single-sized particles is

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial z}(D_v)\frac{\partial C}{\partial z} - \frac{\partial VC}{\partial z} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (4)$$

where C = particle concentration; z = depth; and t = time (Csanady 1973). The boundary conditions used with (4) were that: (1) The upper boundary was treated as a surface with no net mass flux across it; and (2) the bottom boundary was treated as an adsorptive boundary, i.e., particles that reach this boundary "stick" to it. The initial condition was specified as having the entire mass of soil particles in the surface-water layer, i.e., the particles were considered to be added instantaneously to the surface at time $t = 0$.

There are several assumptions that govern our use of (4). These are: (1) The horizontal variation in concentration was negligible; (2) mixing was accounted for by a constant eddy diffusivity; (3) there was no hindered or nondiscrete settling of particles due to hydrodynamic influences; (4) vertical convective fluid transport was insignificant, i.e., $V_f = 0$; (5) the particle-size distribution was treated as a single-size category characterized by an average particle size with an average settling velocity; (6) the particles formed a stable suspension, i.e., no coagulation occurred. Most particulate systems, including the Middlesex soil, are not monodispersed suspensions. Under the condition in which the fifth assumption is not valid, a system of equations must be employed, each similar to (4). Each equation would correspond to a single-size category. This modeling approach assumes that several state variables, one for each representative size or velocity category, could be used to describe the vertical convective-diffusive transport of the polydispersed suspension. The sixth assumption means that the particles neither coagulate and form larger particles nor break up and form smaller particles. This assumption is only valid for stable, noncoagulating systems that are approaching or have reached an equilibrium particle-size distribution, i.e., the particle formation and breakup balance.

The system of particle differential equations used to describe the vertical transport of the polydispersed particles is

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial z}(D_v)\frac{\partial C_i}{\partial z} - \frac{\partial V_i C_i}{\partial z}; \qquad i = 1,2, \ldots, N \ldots\ldots\ldots\ldots\ldots\ldots (5)$$

where C_i = particle concentration in a size category i; V_i = mean settling velocity for size category i; and N = number of size categories. The equations within the foregoing system are not coupled as they would be for a coagulating suspension of particles, i.e., for a dynamic particle-size distribution. Therefore, the equation for each size category was solved independently.

Model Calibration and Data Synthesis

Calibration of the dispersion component of the model was conducted to quantify the vertical dispersion in a MERL tank. This calibration was based on the data obtained during the dye studies by iteratively adjusting the dispersion coefficient to minimize the difference between observed and predicted concentration over space and time. Fig. 9 shows an example of predicted model output superimposed onto a plot of the observed data. The model output, with its vertical dispersion at 11 cm^2/s, agrees well with observed data from the dye study conducted using an impeller mixing speed of 0.52 rad/s (Bonner et al. 1985). Similar agreement was obtained for the mixing levels of 8 and 26 cm^2/s, corresponding to impeller rotation speeds of 0.37 and 0.85 rad/s, respectively.

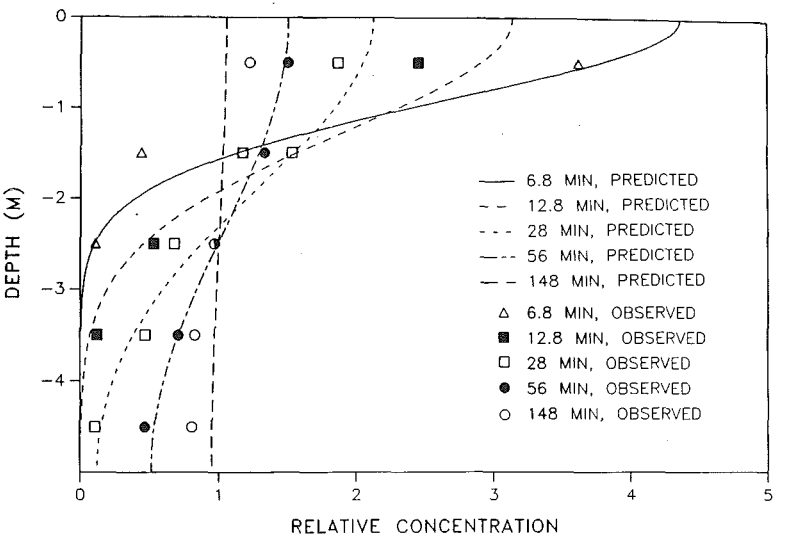

FIG. 9. Dye Experiment Results in MERL Mesocosm with 11 cm²/s Dispersion

The convective terms for (5) were determined empirically from the data obtained from the settling-column experiments. These data represent settling-velocity distributions for the polydispersed suspension of soil particles. The velocity distributions used in the model were those measured at the greatest depth in the experimental settling column (85 cm). The size and velocity distributions measured at this depth would most closely approach an equilibrium condition. Using these data to calibrate the transport model is reasonable for the projections to the deep-ocean environment. Settling particles in this environment would reach an equilibrium condition before descending through a significant fraction of the total depth (4,000 m). If for no other reason, equilibrium would occur due to reduced particle-particle interaction caused by simple particle-number dilution as a result of the turbulent dispersion and differential gravitational settling.

Model Application

The multiparticle convective-diffusive model was applied assuming conditions that would exist at a hypothetical deep-ocean disposal site in 4,000 m of water. The model was used to estimate the soil-particle residence time in the water column and the distance along the direction of mean flow to deposition on the bottom sediments. The empirically determined settling-velocity distribution and assumed vertical dispersion, ranging from 0 to 100 cm²/s, were used. Fig. 10 shows time of arrival at the bottom sediments for the Middlesex soil assuming vertical dispersions of 0, 50, and 100 cm²/s. The model predicts that at least 95% of the soil mass would arrive at the sediment surface in less than five days. Variations in vertical dispersion over the foregoing range, characteristic of deep-ocean sites, have negligible effect on the time of arrival of the Middlesex soil.

Information on the horizontal current structure off the northeast coast of the United States (Ingham et al. 1977) was used to calculate the horizontal distance particles would travel before they reached the bottom. The down-

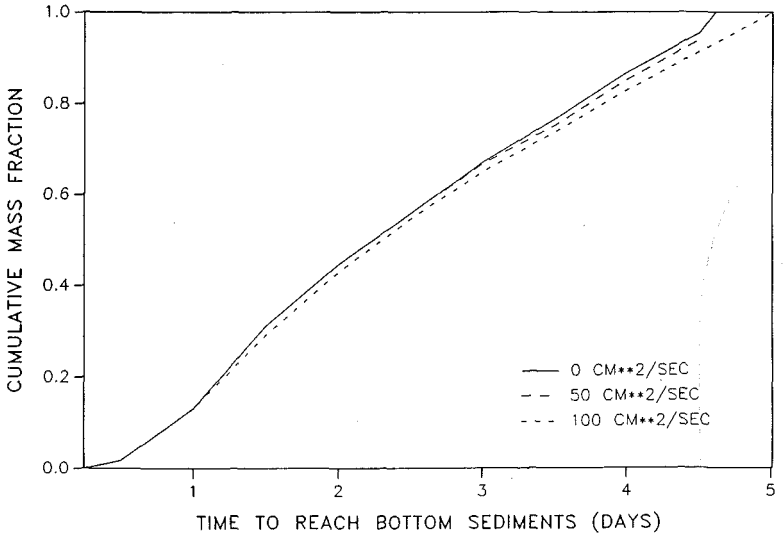

FIG. 10. Time of Arrival for Middlesex Soil at Bottom of 4,000 m Water Column

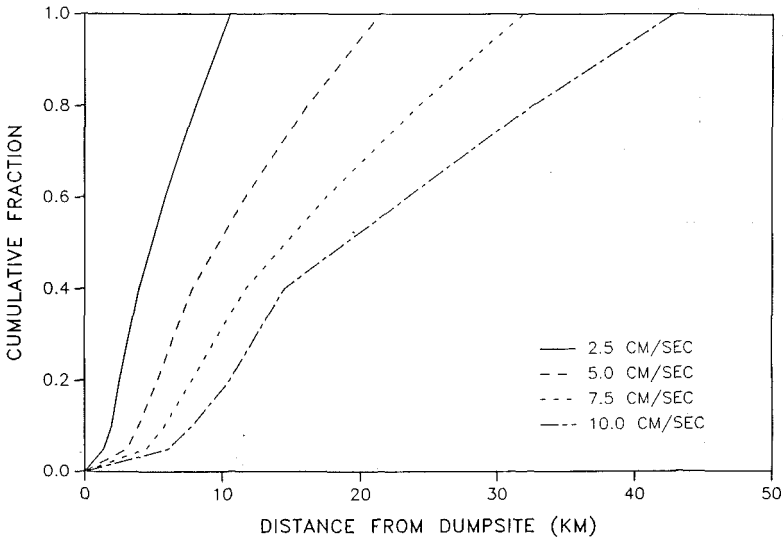

FIG. 11. Bottom Impact Distance along Mean Current Flow Direction for Middlesex Soil Disposed in 4,000 m of Water (D_v = 25 cm²/s)

stream bottom-impact distance from the disposal site is shown in Fig. 11 for time averaged horizontal current velocities of 2.5, 5.0, 7.5 and 10.0 cm/s assuming no episodic events (e.g., cold or warm core eddies, storms etc.). This figure indicates that the distance these particles would travel, for the assumed site condition (D_v = 25 cm²/s), ranges from 0.5 to 40.0 km. O'Con-

nor et al. (1985) reported a vertically weighted mean velocity of 2.0 cm/s, based on the deep-ocean current structure off the northeast coast of the United States. With this velocity, the impact distance along the direction of the mean flow for 95% of the mass would be approximately 6 km. For practical purposes, the soil would deposit within the confines of a typical disposal site based on the assumed time-averaged currents.

Our projections for a deep-ocean disposal site were based upon experimental data for the Middlesex soil that had been excavated, sieved, and homogenized. If an actual disposal operation were to commence, the physical characteristics of the soil disposed would be expected to be different than those of the homogenized soil we used. The density of uranium ore is several times that of soil. Certainly, if the radioactivity is confined to pitchblende particles, then significantly faster transport rates would occur and a more restricted impacted zone would result. Smaller impact zones would mean that the sediments would have higher concentrations of radioactive contaminants derived from waste-soil disposal. Our results, however, provide upper-bound estimates on the time of arrival at the bottom sediments and on the bottom-impact distance along the direction of mean flow.

Conclusions

A combined experimental and modeling approach was conducted to assess the vertical transport of low-level radioactive soil at a deep-ocean disposal site. Experimentation was conducted to characterize Middlesex soil in terms of particle-size distribution, specific gravity, and radioactivity. In addition, settling-velocity distributions were measured. The data derived from the experimental work were synthesized using a modeling approach that emphasized the vertical convective-diffusive transport of a polydispersed suspension of particles. The following conclusions may be drawn from our study.

The Middlesex soil in this study had 90% of the mass associated with particles between 63 μm and 6,350 μm, with a median size of 350 μm, and an average particle specific gravity of 2.31.

Gross activity (total gamma plus beta and alpha) was associated with particles up to 6,350 μm, with a median activity at 125 μm.

The specific element or isotope activities were also associated with particles up to 6,350 μm, with median activities ranging from 250 μm and 600 μm. The activity distributions of individual isotopes were not coincident with either the size or gross activity distributions.

The soil-settling-velocity distributions were initially dynamic, but tended to stabilize with increasing depth in a 1 m settling column. This indicated that the soil suspensions were approaching an equilibrium state in terms of size distribution and terminal velocity.

The measured soil-settling velocity ranged up to 8.2 cm/s, with a median of 2.1 cm/s.

The gravitational settling of the Middlesex soil would dominate the vertical transport of a deep-ocean disposal site; water-column mixing had a limited effect, with vertical dispersion having less than a 5% effect on the water-column residence time.

The Middlesex soil and the radioactivity associated with it would impact the bottom sediment in 4,000 m of water at distances from 0.5 to 40 km along the direction of the flow, for mean horizontal velocities in the range of 2.5 to 10 cm/s.

Ninety-five percent of the Middlesex soil disposed in 4,000 m of water would arrive at the bottom in less than five days. The particles with the largest settling velocities may reach the bottom in approximately 12 hr.

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APPENDIX I. REFERENCES

Bonner, J. S., Hunt, C. D., Paul, J. F., and Bierman, V. J., Jr. (1985). "Prediction of vertical transport of low-level radioactive Middlesex soil at deep-ocean disposal site." *Tech. Report,* Environmental Research Laboratory, U.S. Environmental Protection Agency, Narragansett, R.I.

Csanady, G. T. (1973). *Turbulent diffusion in the environment.* D. Reidel Publishing Co., Boston, Mass.

Dickson, H. W., Haywood, F. F., Cottrell, W. D., Leggett, R. W., Goldsmith, W. A., and Fox, W. F. (1977). "Formerly utilized MED/AEC sites remedial action program." *DOE/EV-0005: Radiological Survey of the Middlesex Sampling Plant,* Department of Energy, Washington, D.C.

Ducharme, S. L. (1989). "Design and validation of a settling column for particle transport studies," thesis presented to Texas A&M University, at College Station, Tex., in partial fulfillment of the requirements for the degree of Master of Science.

Friedlander, S. K. (1977). *Smoke, dust, and haze.* John Wiley & Sons, New York, N.Y.

Hunt, C. (1986). "Fate and bioaccumulation of soil-associated low-level naturally occurring radioactivity following disposal into a marine ecosystem." *Tech. Report: Prepared for Analysis and Support Division ORP-EPA EPA-520/1-86-017,* Office of Radiation Programs, Environmental Protection Agency, Washington, D.C.

Ingham, M. C., Bisagni, J. J., and Mizenko, D. (1977). "The general physical oceanography of Deepwater Dumpsite 106." *NOAA Dumpsite Evaluation Rept. 77-1,* U.S. Dept. of Comm. Pub., Washington, D.C., Vol. 1, 29–54.

Klett, R. D., and Klett, K. A. (1984). "Particle velocity systems study for ocean dumping." *FODOCS Annual Report, March 1–September 30, 1981: SAND82-0292,* Sandia National Laboratories, Albuquerque, N.M., 65–68.

Knauss, J. A. (1984). "Nuclear waste management and the use of the sea." *A Special Report to the President and the Congress.* National Advisory Committee on Oceans and Atmosphere, Washington, D.C.

Kupferman, S. L., Anderson, D. R., Brush, L. H., Gomez, L. S., and Shephard, L. E. (1984). *FODOCS Annual Report March 1–September 30, 1981: SAND82-0292,* Sandia National Laboratories, Albuquerque, N.M.

Lawler, D. F., Iswieta, E., and Koa, C. J. (1983). "Changes in particle size distribution in batch flocculation." *J. American Water Works Assoc.,* 75(12), 604–612.

Leggett, R. W., Christian, D. J., Crawford, D. J., Cottreal, W. D., Haywood, F. F., Doane, R. W., Goldsmith, W. A., Wagner, E. B., and Shinpaugh, W. H. (1980). "Formerly utilized MED/AEC sites remedial action program, radiological

survey of the Middlesex Municipal Landfill." *DOE/EV0005/20,* Department of Energy, Washington, D.C.
Lick, W. (1982). "Entrainment, deposition, and transport of fine-grain sediments in lakes." *Hydrobiologia,* 91(1), 31–40.
Nixon, S. W., Alonso, D., Pilson, M. E. Q., and Buckley, B. A. (1980). "Turbulent mixing in aquatic microcosms." *Microcosms in ecological research: DOE Symp. Series; CONF-781101,* G. P. Gisey, ed., Department of Energy, Washington, D.C., 818–849.
O'Connor, T. P., Walker, H. A., Paul, J. F., and Bierman, V. J., Jr. (1985). "A strategy for monitoring of contaminant distribution resulting from proposed sewage sludge disposal at the 106-mile ocean disposal site." *Marine Envir. Res.,* 16, 127–150.
Sverdrup, H. U., Johnson, M. W., and Fleming, R. H. (1942). *The oceans, their physics, chemistry, and general biology.* Prentice-Hall, Inc., New York, N.Y.
Wastes in the ocean; Volume 3: Radioactive wastes and the ocean. (1983). P. K. Park, D. R. Kester, I. W. Duedall, and B. H. Ketchum, eds., John Wiley, New York, N.Y.
Weber, W. J., Jr. (1972). *Physicochemical processes for water quality control.* Wiley-Interscience, New York, N.Y.

APPENDIX II. NOTATION

The following symbols are used in this paper:

b = depth of water column;
C = mass concentration of particles;
C_i = mass concentration of particles of type i where there are N particle types;
D_v = vertical dispersion;
N = total number of particle types;
t = time;
V = velocity;
V_f = fluid velocity;
V_i = settling velocity of particle type i;
V_p = particle settling velocity;
X = mass fraction; and
z = distance.