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SUBJECT: DISPERSION OF SOLUBLE RADIOACTIVE MATERIAL
IN WATER

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FROM: Fred S. Dunning¹ and John C. LeDoux¹ABSTRACT

Theories of diffusion processes in water are discussed and the dispersion of radioactivity is calculated from a solution to the molecular diffusion equation with modifications which tend to fit the equation to experimental data. From this the dose rate resulting from fission products in solution are made for a number of situations of interest. Parametric solutions are obtained and plotted on graphs which may be used to obtain water concentrations and thence dose levels.

- 1 U. S. Navy personnel. The material in this report was prepared as a part of their work at the ORNL Reactor School, 1956-1957

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DISPERSION OF SOLUBLE RADIOACTIVE MATERIAL IN WATER

If a soluble substance is placed in water at a point, the concentration at the instant of deposition will be infinite at that point and zero at all others in the medium. As time increases, the concentration at the initial point becomes finite and decreases, while the concentration at other points in the medium increases to a maximum, then decreases. This behavior is the result of diffusion; and, in turbulent water, the dominant diffusion process is turbulent eddy diffusion, where the size and strength of the eddies effectively transport dissolved materials.

The equation describing molecular diffusion of a soluble material is:²

$$\frac{DQ}{Dt} = \nabla A (\nabla Q) - \lambda Q - R \quad (1)$$

where: Q = concentration, number/centimeter cubed
 t = time, seconds
 D' = total derivative operator (includes convection)
 ∇ = Laplacian operator
 A = Diffusion coefficient, cm^2/sec
 λ = decay constant, sec^{-1}
 R = Removal function, such as for settling out of suspended or dissolved material.

The solution to Equation (1) for an instantaneous point source, implying that the material introduced is immediately completely soluble and the diffusion coefficient is constant,

$$Q = S_0 \frac{e^{-\frac{(x-vt)^2}{4A_x t}}}{(4\pi A_x t)^{1/2}} \frac{e^{-\frac{y^2}{4A_y t}}}{(4\pi A_y t)^{1/2}} \frac{e^{-\frac{z^2}{4A_z t}}}{(4\pi A_z t)^{1/2}} e^{-\lambda t} \quad (2)$$

where S_0 = quantity of material introduced
 The function describing removal by settling out is omitted, and a velocity in the "x" direction only is assumed.

Difficulties are encountered in the application of the molecular diffusion equation to turbulent eddy diffusion phenomena for two major reasons.

One, the turbulent eddy diffusion coefficient, A , is not constant, nor isotropic,¹ but depends on currents, winds, temperatures and surface irregularities in a very complex manner. In any particular application, a spectrum of eddies exists; thus making the choice of a single coefficient of diffusion difficult if not impossible. Further, density currents can radically change a diffusion pattern.²

Second, according to Richardson³ as explained by Stommel,⁴ the "distance-neighbor" theory is shown to be the process describing diffusion by turbulent eddies. The "distance-neighbor" theory states that the probability of two particles being ℓ distance apart is a function of their initial separation distance, not on their separate original distance from some other fixed point, i.e., an origin. Diffusion theory uses distance from an origin as a basis for the probability that a particle will be that distance from the origin at a given later time.

Richardson's equation takes the following form:

$$\frac{\partial Q}{\partial t} = \frac{\partial Q}{\partial \ell} K(\ell) \frac{\partial Q}{\partial \ell} \quad (3)$$

where ℓ = distance between two particles considered
 $K(\ell)$ = a diffusion coefficient which is a function of distance between particles.

Experiment shows;

$$K(\ell) = k \ell^{4/3} \quad (4)$$

(where K is an empirical constant),
 and that there is some correlation between $K(\ell)$ and $A(x)$.⁴

The solution of Richardson's equation as a function of ℓ , of course, has no direct applicability; but, is of interest since it shows equation (1) may not be applied in the strictest sense.

One approach to the practical problem is to present a solution to the molecular diffusion equation with modifications which tend to fit the equation to experimental data. Sutton⁵ has done this, in atmospheric

diffusion theory, with a stability parameter, Sverdrup¹ assumes the turbulent eddy diffusion coefficient proportional to the mean radius of the diffusion substance, implying that the diffusion coefficient is a function of the scale of observation. Glover,² in studying diffusion in streams, suggests that the diffusion coefficient is proportional to the stream velocity with a value one tenth as great in the cross stream direction.

In this report it is proposed to use characteristic values of turbulent eddy diffusion coefficients in the molecular diffusion equation in order that estimates of dose rates resulting from fission products in solution may be made. Accordingly, typical values of turbulent eddy diffusion coefficients are presented. Table 1, Turbulent Eddy Diffusion Coefficients, will be found at the end of this text.

These coefficients are used in the diffusion equation to estimate concentration of soluble materials as a function of distance and time. The diffusion of soluble fission products is believed by the authors to be similar to the diffusion of dye marker, or fluorescein dye. Using typical values of diffusion coefficients derived from experiments with dye marker is extremely pessimistic and should result in overestimating dose rates. However, the AEC used values of the diffusion rate, typical for dye marker, in calculations for the spread of contaminant in Operations Crossroads.⁶ Using small values of diffusion coefficients also implies a relatively stagnant water such as might be found in a large harbor.

1. Gamma Ray Dose Rates Analyses

It is assumed that all the fission products released in water go into solution immediately. The dose rate received is calculated assuming a slowly varying concentration of gamma ray sources in an infinite medium. In such a system the energy absorbed must equal the energy emitted and the dose rate in the medium is determined by the application of a conversion factor.¹¹

$$D^0(\text{Rads/hr}) = Q(\text{Mev/cm}^3\text{-sec}) 5.767 \times 10^{-5} \quad (5)$$

The rad is a unit of dose equivalent to the absorption of 100 ergs per gram of tissue.

At one meter above a water medium, the dose rate may be approximated by taking one-half of equation (5).

Concentration distribution may be assumed by one of three processes: one by assuming a fixed geometry and a uniform distribution of sources; then

$$Q = S_o/V \quad \text{where } V = \text{volume assumed;} \quad (6a)$$

next, by point source geometry in an infinite medium such as

$$Q = \frac{e^{-\frac{(x-vt)^2}{4A_x t}} e^{-\frac{y^2}{4A_y t}} e^{-\frac{z^2}{4A_z t}}}{(4\pi A_x t)^{1/2} (4\pi A_y t)^{1/2} (4\pi A_z t)^{1/2}} \quad (6b)$$

and lastly, by assuming vertical line source geometry (cylindrical) as

$$Q = \frac{S_o}{D} \frac{e^{-\frac{(x-vt)^2}{4A_x t}} e^{-\frac{y^2}{4A_y t}}}{(4\pi A_x t)^{1/2} (4\pi A_y t)^{1/2}} \quad (6c)$$

where D is the depth of the water (i.e., length of line source).

If isotropic turbulence exists and it is desired to use the relation $A/r = \text{constant}$ (1), then for spherical geometry and velocity equal to zero

$$Q = \frac{S_o}{r^{3/2}} \frac{e^{-\frac{r^2}{4(A/r)t}}}{[4\pi (A/r) t]^{3/2}} \quad (6d)$$

1.1 Isotropic Turbulence

1.1.1 Dose Rate from an Instantaneous Source: The equation describing isotropic diffusion as derived above is,

$$Q = \frac{S_o}{(4\pi A t)^{3/2}} e^{-\frac{r^2}{4A t} - 0.2} \quad (7)$$

where $t^{-0.2}$ is included as the decay factor appropriate to reactor fission products. Equation (7) may be rewritten as

$$\frac{Q}{S_o} = e^{-X} t^{-0.2} \quad (8)$$

where X is a function of (r, A, t) .

Figures 1, 2, 3 and 4 show X as a function of time and distance from the point of deposition for diffusion coefficients of 3, 30, 300 and $10^5 \text{ cm}^2/\text{sec}$. The dose rate is

$$\begin{aligned} D^o(r, A, t) &= 5.767 \times 10^{-5} Q \\ &= 5.767 \times 10^{-5} S_o e^{-X} t^{-0.2} \text{ rads/hr} \end{aligned} \quad (9)$$

As an example enter Figure 3 for $A = 300 \text{ cm}^2/\text{sec}$ and assume $r = 1000 \text{ cm}$ and $t = 1000$ seconds. Assume an initial source of 1.2×10^{19} Mev per second

$$x \approx 25$$

$$\begin{aligned} D^0(1000, 1000) &= (5.767 \times 10^{-5}) (1.2 \times 10^{19}) e^{-25} 1000^{-0.2} \\ &= 6.9 \times 10^3 \text{ Rads/hr} \end{aligned} \quad (10)$$

If $r = 5000 \text{ cm}$

$$\begin{aligned} D^0(5000, 1000) &= (5.767 \times 10^{-5}) (1.2 \times 10^{19}) e^{-46} 1000^{-0.2} \\ &= 1.7 \times 10^{-6} \text{ Rads/hr} \end{aligned} \quad (11)$$

The presence of boundaries such as the ocean floor, harbor bottom or water surfaces acts as a bar to the diffusion process. However, Figures 1, 2, 3, and 4 may be used for cases of isotropic turbulence with a source under water for dose rates on a boundary provided the time curves do not continue horizontally past the distance (boundary) in question. (Otherwise see non-isotropic solution following this discussion.)

Since the relaxation length of one Mev gamma ray is about 100 meters in air, the dose to a person one meter above the water surface is essentially one half that of a person in the water. With a point source on a boundary such as the ocean floor or harbor bottom the semi-infinite geometry essentially multiplies the source by two. Therefore, the two previous examples apply, but are multiplied by two in the semi-infinite case for a person in the water. The dose rate is the same as shown one meter above the water in the semi-infinite geometry.

Figure 5 shows the minimum allowable distance from point of deposition based on one Rad per hour, as a function of time for deposition of 1.2×10^{19} mev per second of fission products.

1.1.2 Dose Rate at the Center of Deposition: The dose rate at the center of

deposition of a soluble radioactive material, assuming a buildup factor of unity in an infinite homogeneous water system is,¹¹

$$D^0(r,t) \text{ rad/hr} = 5.767 \times 10^{-5} \left(\frac{\mu_a}{\rho}\right) E_0 \Gamma^0(r,t) \quad (12)$$

where

E_0 = photon energy

$\frac{\mu_a}{\rho}$ = energy absorption mass attenuation coefficient for the energy and material (tissue) considered

$\Gamma^0(r,t)$ = current density of unscattered photons summed or integrated over all sources

Isotopic decay may be included in equation (12) by multiplying by a decay function, such as $t^{-0.2}$ or $e^{-\lambda t}$. Furthermore,¹¹

$$\Gamma^0(r,t) = \int \frac{Q e^{-\mu r}}{4\pi r^2} dV \quad (13)$$

where

μ = linear absorption coefficient for water

Q = defined by equation (7)

$dV = 2\pi r dr$

Now, let

$$\alpha = {}^2_{At}$$

and

$$w = \left(\mu r/2\alpha^{1/2}\right)^2 + \mu r + \alpha = \left(\mu r/2\alpha^{1/2}\right) + \alpha^{1/2}^2$$

Substitute α and w in equation (12) and get,

$$D^0(r,t) = \frac{5.767 \times 10^{-5}}{(4\pi)^{3/2}} S_0 \left(\frac{\mu_a}{\rho}\right) \mu^2 \left\{ \frac{e^\alpha}{\alpha} \int_\alpha^\infty e^{-w} w^{1/2} dw \right\} \quad (14)$$

Noting that

$$\begin{aligned} \int_\alpha^\infty e^{-w} w^{1/2} dw &= \int_0^\infty e^{-w} w^{-1/2} dw - \int_0^\alpha e^{-w} w^{-1/2} dw \\ &= \Gamma\left(\frac{1}{2}\right) - \int_0^\alpha e^{-w} w^{-1/2} dw \end{aligned} \quad (15)$$

Then:

$$D^0(r,t) = \frac{5.767 \times 10^{-5}}{(4\pi)^{3/2}} S_0 \left(\frac{\mu_a}{\rho} \right) \mu^2 \frac{e^{-a}}{a} \left\{ \sqrt{\pi} - 2a^{1/2} \left[1 - \frac{a}{3} + \frac{a^2}{5 \cdot 2} - \frac{a^3}{7 \cdot 3} \dots \right] \right\} \quad (16)$$

For evaluation purposes, Figure 6 shows a plot of a versus

$$\frac{e^{-a}}{a} \int_a^\infty e^{-w} w^{-1/2} dw, \quad \text{for } a = .01 \text{ to } 1000. \quad (17)$$

Further, for $a \gtrsim 2$

$$\int_a^\infty e^{-w} w^{-1/2} dw \approx \frac{0.78 e^{-a}}{a^{1/2}} \quad (18)$$

Using buildup factors as a function of relaxation length makes the integral extremely difficult to evaluate analytically. A method used at ORNL (original source not known to the authors) is to divide the mass attenuation coefficient by 1.2 as an approximation for the buildup.

As an example, suppose soluble radioactivity is placed instantaneously on the surface of water with isotropic turbulence. To approximate a dose rate, assume:

$$S_0 = 1.2 \times 10^{19} \text{ mev/sec}$$

$$A = 300 \text{ cm}^2/\text{sec}$$

$$t = 100 \text{ sec}$$

$$\mu = .07 \text{ for one mev gamma rays}$$

$$\therefore a = 147, \text{ or } 122 \text{ if the factor } 1.2 \text{ is used.}$$

Enter Figure 6 at $\alpha = 147$ and get 0.00045 for the function of α .

$$D^0(0,100) = \frac{2(5.767 \times 10^{-5})(1.2 \times 10^{19})(0.03)(0.07)^2(4.5 \times 10^{-4})}{(4\pi)^{3/2}} \approx 2.0 \times 10^6 \text{ Rads/hr} \quad (19)$$

Using $\alpha = 122$,

$$D^0(0,100) = 2.8 \times 10^6 \text{ Rads/hr}$$

Compare this result to that obtained from Figure 3 as follows,

$$D^0(0,100) \approx e^{(34.1 - X)} = e^{34.1 - 20} = e^{14.1} = 3.6 \times 10^6 \text{ Rads/hr}$$

The comparison is good considering the extreme extrapolation and approximations involved.

1.1.3 Dose Rate from a Continuous Source: In the case of continuous isotropic point source in an infinite isotropic medium, the concentration at a point is defined by a modified form of equation (7),

$$Q = \int_{\xi=0}^t \frac{e^{-\frac{r^2}{4A(t-\xi)}}}{[4\pi A(t-\xi)]^{3/2}} s_o d\xi \quad (20)$$

where s_o = source emission rate, no./sec
 ξ = infinitesimal time at which a particular $s_o d\xi$ is introduced

$$S_o = \int_{\xi=0}^t s_o d\xi$$

Equation (20) is the product of the sum of the material introduced and the distribution function for the material in the dispersing media.

To obtain a solution, let

$$B = \frac{r^2}{4A} \text{ and } W = \frac{r^2}{4A(t-\xi)}; \text{ therefore,}$$

$$\frac{B}{W} = (t - \xi)$$

Substitute into equation (20) and get,

$$Q = \frac{s_o}{(4\pi A)^{3/2}} \int_{B/t}^{\infty} e^{-W} W^{-1/2} dW \quad (21)$$

Now let

$$\frac{B}{t} = \alpha' = \frac{r^2}{4At}$$

substitute in equation (21) and get,

$$Q = \frac{s_o e^{-\alpha'} \alpha'}{(4\pi A)^{3/2}} \left\{ \frac{e^{\alpha'}}{\alpha'} \int_{\alpha'}^{\infty} e^{-W} W^{-1/2} dW \right\} \quad (22)$$

The term in brackets may be evaluated from Figure 6 or the integral may be approximated by $(0.78 e^{-\alpha'^{1/2}})/\alpha'^{1/2}$, giving,

$$Q = \frac{0.78}{(4\pi A)^{3/2}} s_o \frac{e^{-\alpha'}}{\alpha'^{1/2}} \text{ for } \alpha' \gtrsim 2 \quad (23)$$

Dose rates may be calculated as shown in the example following equation (18).

Another method approximation of concentration is:

$$Q = \sum_i s_o \frac{\Delta \xi_i e^{-\frac{r^2}{4At_i}}}{(4\pi A t_i)} \quad (24)$$

where

$\Delta \xi_i$ = the i^{th} increment of time

t_i = time from start of i^{th} increment of source

As an example, assume

$$s_o = 1.2 \times 10^{19} / 1,000 = 1.2 \times 10^{16} \text{ mev/sec}^2$$

$$\Delta \xi_i = 100 \text{ sec}$$

What is the dose rate at $r = 1,000 \text{ cm}$ and $t = 1,000 \text{ seconds}$: for $A = 300 \text{ cm}^2\text{-sec}$?

<u>t_i</u>	<u>Increment</u>	<u>Q/s_o</u>
1000	1	.050 x 10 ⁻⁹
900	2	.055
800	3	.058
700	4	.061
600	5	.062
500	6	.064
400	7	.0575
300	8	.045
200	9	.029
100	10	.001
	$\sum_i$	<u>.500 x 10⁻⁹</u>

$$D^0(1000,1000) \approx (5.767 \times 10^{-5})(1.2 \times 10^{16})(.5 \times 10^{-9})$$

$$\approx 3.5 \times 10^2 \text{ Rads/hr}$$

This method may be applied to a varying source.

1.2 Non-Isotropic Turbulence

The equation for non-isotropic diffusion is:

$$Q = \frac{S_o e^{-\frac{(x-vt)^2}{4A_x t}}}{(4 \pi A_x t)^{1/2}} \cdot \frac{e^{-\frac{y^2}{4A_y t}}}{(4 \pi A_y t)^{1/2}} \cdot \frac{e^{-\frac{z^2}{4A_z t}}}{(4 \pi A_z t)^{1/2}} \quad (25)$$

If the source is at a boundary multiply by two, if at the intersection of two boundaries multiply by four, and if at the intersection of three boundaries (diffusing in only one quadrant) multiply by eight.

Suppose the source lies at the bottom of a channel. (The typical depth assumed in this report is ten meters, 36 feet.) The existence of the water surface acts as a bar to diffusion in the "Z" direction. However, the solution is applicable if²,

$$\frac{D^2}{4 \pi A t} > 4$$

where D = depth

Otherwise, solve for t_2 in, $D^2 = 16\pi A_z t_2$, and substitute t_2 in the following series,

$$\frac{e^{-\frac{z^2}{4A_z t}}}{(4\pi A_z t)^{1/2}} \approx \frac{0.4820}{D} e^{-\frac{A_z \pi^2 (t-t_2)}{D^2}} \cos \frac{\pi z}{D} + 0.0722 e^{-\frac{4 A_z \pi^2 (t-t_2)}{D^2}} \cos \frac{2\pi z}{D} + \frac{0.433}{D} \quad (26)$$

In general, the interest is in the maximum dose rate. This occurs at $y = 0$, $x = 0$, $v = 0$, or $y = 0$, $x = vt$ if the velocity is constant with depth. If the velocity is not constant with z , then the maximum concentration is approximately the same but varies with x as shown in the diagram below.

Diagram: The Effect of Variable Velocity on Concentration

Note that both curves have approximately the same maximum.

Figures 7 and 8 show X as a function of time, and depth of 1000 centimeters and 1500 centimeters (36 and 45 feet) respectively for three combinations of diffusion coefficients; $A_x = A_y = A_z = 300 \text{ cm}^2/\text{sec}$, $A_x = 300$, $A_y = A_z = 30 \text{ cm}^2/\text{sec}$, and $A_x = A_y = 300$, $A_z = 30 \text{ cm}^2/\text{sec}$. Surface dose rate be calculated from:

$$D^0(1000, t) = (5.767 \times 10^{-5}) S_0 e^{-X} \quad (27)$$

For example, with $1.2 \times 10^{19} \text{ mev/sec}$ source and $A_x = A_y = A_z = 300 \text{ cm}^2/\text{sec}$.

$$D^0(1000, 1000) = (5.767 \times 10^{-5})(1.2 \times 10^{19})(e^{-30}) = 69 \text{ Rads/hr}$$

1.2.1 Instantaneous Point Source at the Surface

Effectively at $x = y = z = 0$, the concentration varies as

$$Q_{\max} = \frac{2 S_0}{(4\pi)^{3/2} A_x^{1/2} A_y^{1/2} A_z^{1/2} t^{3/2}} \quad (28)$$

1.2.2 Vertical Line Sources

The equation applicable to a vertical line source is:

$$Q = \frac{S_0 e^{-\frac{(x-vt)^2}{4A_x t}} e^{-\frac{y^2}{4A_y t}}}{D(4\pi A_x t)^{1/2} (4\pi A_y t)^{1/2}} \quad (29)$$

where D is the depth of the channel

Effectively only the first 100 cm in depth contributes to the dose rate on the surface. This is diffusion in two dimensions and similar to the point isotropic source on the surface.

$$Q_{\max} = \frac{S_0}{D 4\pi A_x^{1/2} A_y^{1/2} t} \quad (30)$$

1.2.3 Continuous Point Sources

The case consisting of a continuous point source under water may be solved in the following manner.

$$Q = \int_0^t \frac{s_o e^{-\frac{(x-v[t-\xi])^2}{4A_x(t-\xi)}} e^{-\frac{y^2}{4A_y(t-\xi)}} e^{-\frac{z^2}{4A_z(t-\xi)}}}{(4\pi)^{3/2} A_x^{1/2} A_y^{1/2} A_z^{1/2} (t-\xi)^{3/2}} d\xi \quad (31)$$

Neglecting diffusion blocking due to surface boundary and taking the case for the maximum dose rate on the surface; i.e., $x = y = v = 0$, and substituting,

$$W = \frac{z^2}{4A_z(t-\xi)}, \quad \alpha = \frac{z^2}{4A_z t}$$

get:

$$Q = \frac{s_o e^{-\frac{z^2}{4A_z t}} + \frac{z^2}{4A_z t} \left(\frac{z^2}{4A_z t}\right)^{1/2}}{(4\pi)^{3/2} A_x^{1/2} A_y^{1/2} A_z^{1/2} t^{1/2} \left(\frac{z^2}{4A_z t}\right)^{1/2}} \int_0^{\frac{z^2}{4A_z t}} e^{-W} W^{-1/2} dW \quad (32)$$

which is put into the form:

$$Q = \frac{s_o e^{-\alpha} \alpha^{1/2}}{(4\pi)^{3/2} A_x^{1/2} A_y^{1/2} A_z^{1/2}} \left\{ \frac{e^{\alpha}}{\alpha} \int_{\alpha}^{\infty} e^{-W} W^{-1/2} dW \right\} \quad (33)$$

The term in brackets has been plotted in Figure 6 as a function of α . The graph may be used to calculate dose rates. For large α , ($\alpha \geq 2$), the integral term behaves approximately like $0.78e^{-\alpha/\alpha^{1/2}}$ and:

$$Q = \frac{s_o (0.78) e^{-\alpha}}{(4\pi)^{3/2} A_x^{1/2} A_y^{1/2} A_z^{1/2} \alpha} \quad (34)$$

1.2.4 Alternate Method for Continuous Point Source

The continuous source problem is approximated in a manner similar to the point isotropic case.

$$Q = \frac{s_o \Delta \left\{ e^{-\frac{(x-vt_i)^2}{4A_x t_i}} e^{-\frac{y^2}{4A_y t_i}} e^{-\frac{z^2}{4A_z t_i}} \right\}}{(4\pi A_x t_i)^{1/2} (4\pi A_y t_i)^{1/2} (4\pi A_z t_i)^{1/2}} \quad (35)$$

It is believed that if diffusion coefficients in the 30 to 300-cm²/sec range are chosen that reasonable values of dose rate will be determined if the scale of observation is less than 100 meters (100 yards).

The formulas may be modified to include A/r for larger scales of diffusion phenomena. Then the concentration varies as $r^{-1/2}$, r^{-1} or $r^{-3/2}$, depending on the number of degrees of freedom.

Since the time element enters in as the product (At), it is instructive to compare the different methods of assuming the distribution of fission products.

Suppose a source of $\left(\frac{1.2 \times 10^{19}}{50}\right)$ mev/sec of which 10% were fission products which were soluble or small enough to be suspended. Postulate that this activity is released below the surface of the water. Some time later assume the soluble fission products occupy a space 10 x 100 x 100 meters. The dose rate above the surface would be:

$$D^o = \frac{S_o (\% \text{ dissolved}) (5.767 \times 10^{-5})}{2 (\text{volume of water})} \quad (36)$$

$$D^o = \frac{(5.767 \times 10^{-5}) (1.2 \times 10^{19}) (0.1)}{(2) (50) (1 \times 10^{11})} \quad (37)$$

This may be compared to a calculation using line source diffusion theory and substituting the following assumptions into a modification of equation (6c) times the conversion factor.

$$\begin{aligned} D &= 1,000 \text{ cm} \\ S_o &= 1.2 \times 10^{19} \text{ mev/sec} \\ A &= 1,000 \text{ cm}^2/\text{sec} \\ D^o &= \frac{1}{2} \left(\frac{5.767 \times 10^{-5}}{4\pi t A} \right) \left(\frac{1.2 \times 10^{19}}{D} \right) \left(\frac{0.1}{50} \right) e^{-\frac{r^2}{4(1000t)}} \quad (38) \end{aligned}$$

$$D^o = \frac{5.6 \times 10^4}{t} e^{-\frac{r^2}{4000t}} \approx \frac{5.6 \times 10^4}{t} \quad (39)$$

for $r^2 < 4000 t$, and in about five hours, this drops to about 6 rads/hr.

In the ocean taking $A_H = 10^5 \text{ cm}^2/\text{sec}$,

$$D^0 = \frac{5.6 \times 10^2}{t} e^{-\frac{r^2}{4 \times 10^5 t}} \quad (40)$$

and in 100 seconds this activity would be attenuated to 6 rads/hr. Note the dose rate drops off rapidly beyond 60 meters from the source.

2. The Thermal Column Problem

It is assumed that fission products from a reactor which had been operated at 100 Mw are dumped into water. Furthermore it is assumed that all the fission products are dissolved in the water and that they have associated with them from the preceding reactor operation a latent heat, H_L , of 3.4×10^6 Btu. These latter assumptions are both pessimistic and in any real situation could be less by orders of magnitude. The resulting calculated doses from the release would then be proportionally lower than those calculated as shown here. In particular if the fuel is in canned elements of one kind or another only a small fraction of the total fission products would be released, whereas with a liquid fuel all but a small fraction would be released.

The initial afterheat (fission product decay heat) is estimated at 6% of reactor power. Since reactor power is assumed to be 100 Mw, the initial decay heat is,

$$\begin{aligned} H_a &= (0.6) P \text{ (C.F.)} = \frac{(0.6)(100 \times 10^6)(3.4)}{3.6 \times 10^3} \quad (41) \\ &= 5670 \text{ Btu/sec} \end{aligned}$$

Approximately 50% of H_a is gamma ray energy and 50% beta particle energy, therefore,

$$H_{a\gamma} = 3000 \text{ Btu/sec}$$

$$H_{a\beta} = 3000 \text{ Btu/sec}$$

It is seen that the afterheat is small compared to the latent heat and is subsequently neglected. (Various authorities agree that afterheat contributes to dilution and dissipates quickly.)⁷ It is then assumed that the entire latent heat is transferred to water while the fuel descends to the bottom of a 36-ft channel.

The fuel, then, upon exposure to the water, will transfer its heat as it sinks and will leave a stream of fuel particles behind. (If introduced under water near the bottom, will form a "cloud" of fuel particles near the point of introduction.) In both cases, instantaneous sources of radioactive fission products are available.

For a certain period of time the turbulent eddy diffusion coefficients would be large and localized; estimated greater than 2000 cm²/sec.⁸ Also, vertical currents generated, if sufficient in magnitude, may carry particles and dissolved radioactive fission products to the surface in a thermal column.

A preliminary estimate of the thermal column possibility may be made by considering the temperature change possible in water, assuming cylindrical geometry, a nominal channel depth of 36 ft and a latent heat (H_L) equal to 3.4×10^6 Btu.

The volume of the water heated would be

$$V = \pi R^2 D = \frac{H_L}{C_p \Delta T \rho_{H_2O}} \quad (42)$$

and the associated temperature rise would be

$$\Delta T = \frac{H_L}{C_p \rho \pi D R^2} = \frac{3.4 \times 10^6}{(1)(62.4)(\pi)(36)R^2}$$

The table below shows assumed radii and the corresponding temperature rise for this equation.

Temperature Rise in a Column of Water
($H_L = 3.4 \times 10^6$ Btu), (Channel depth = 36 ft)

<u>R (feet)</u>	<u>T (degrees F)</u>
1	482
2	120
3	53.3
4	30.0
5	19.3
6	13.4
7	9.8
8	7.5
9	6.0
10	4.8

It is seen that a realistic temperature rise might be on the order of 10° .

The vertical acceleration of a column of water would be due to the difference in density of the water in the column and the ambient water, the density being a function of temperature.

Representative water densities are:⁹

<u>Temperature, $^\circ\text{F}$</u>	<u>Density, slugs/ft³</u>
30	1.940
40	1.940
50	1.940
60	1.938
70	1.936
80	1.934
90	1.931
100	1.927
110	1.923
120	1.918
130	1.913

If the assumed maximum ΔT were to be as high as 50°F and a nominal ambient water temperature of 50°F is assumed, then the acceleration is determined from

$$a = \frac{\Delta \rho}{\rho} g$$

$$= \left(\frac{1.940 - 1.927}{1.927} \right) g = 0.218 \text{ ft/sec}^2 \quad (43)$$

The velocity of this column at the end of one second would be, from

$$v = at$$

$$= (0.218)(1) = 0.218 \text{ ft/sec} \quad (44)$$

Assuming a more probable or reasonable ΔT of 10°F in 50°F ambient water, the acceleration would be

$$a = \left(\frac{1.940 - 1.938}{1.938} \right) g = 0.033 \text{ ft/sec}$$

and, at the end of one second,

$$v = 0.033 \text{ ft/sec}$$

The authors estimate the settling velocities of the fuel particles at about 0.16 ft/sec .¹ Comparing this to the results of the above analysis, it is seen that a significant ΔT is required before the fuel particles will be convected to the surface. It must be kept in mind that the ΔT will continually and rapidly drop as a result of the turbulent eddy diffusion. On the other hand, some of the fission products will actually dissolve; xenon, for instance, and that the comparison of settling velocities to rising velocities in this case will have no meaning.

If the diffusion of heat energy has the same behavior as the diffusion of dissolved particles, then equation (6c) is applicable except for the substitution of the heat source in place of the radiation source.

$$h = \frac{H_L e^{-\left(\frac{x-vt^2}{4A_x t}\right)} e^{-\frac{y^2}{4A_y t}}}{D 4\pi A_x^{1/2} A_y^{1/2} t} \quad (45)$$

where $h = \text{Btu/ft}^3$

Recalling that

$$h = \rho C_p \Delta T \quad (46)$$

then, for the column case with stream velocity equal to zero and for cylindrical geometry,

$$\Delta T = \frac{H_L e^{-\frac{r^2}{4At}}}{D \rho C_p 4\pi A t} \quad (47)$$

Assume the nominal channel depth of 36 ft and $A = 2.15 \text{ ft}^2/\text{sec},^8$

$$\Delta T = \frac{3.4 \times 10^6 e^{-\frac{r^2}{8.6 t}}}{(36)(62.4)(1)(4\pi)(2.15)t}$$

$$= \frac{56}{t} e^{-\frac{r^2}{8.6 t}}$$

The table below shows the calculated temperature rise for different radii and times.

Temperature Rise in Thermal Column for Various Radii and Times

<u>Time (seconds)</u>	<u>Radius (feet)</u>				
	0	2	5	10	20
1	56.0°F	35.0°F	3 °F	- °F	- °F
2	28.0	22.0	6.5	-	-
3	18.7	16.0	7.3	-	-
4	14.0	12.5	6.8	0.8	-
5	11.2	10.2	6.2	1.1	-
10	5.6	5.4	4.2	1.8	-
25	2.2	2.2	2.0	1.8	1.1
50	1.1	-	-	-	-
75	0.8	-	-	-	-
100	0.6	-	-	-	-

The above table shows that the diffusion of the heat energy is quite rapid and should be complete in a maximum time of 100 seconds. A thermal column, if formed, would, therefore, be dissipated within 100 seconds.

Some vertical thermal motion is expected but not long-lasting nor to any significant magnitude. Further, if a current is flowing, the inherent character of the velocity distribution (profile) and the associated shear stresses will tend to dissipate the effective thermal convection, since rising columns of warmer water would be required to push colder water aside rather than follow an accelerating column.

If 1.2×10^{19} mev/sec of fission product activity were to be completely dissolved, the dose rate at the assumed deposition line with the previously assumed column geometry and depth and a diffusion coefficient of $2.15 \text{ ft}^2/\text{sec}$ would be from equation (9),

$$D^0(0,100) = (Q)(5.767 \times 10^{-5})$$

Obtain "Q" from equation (6c) and get

$$\begin{aligned} D^0(0,100) &= \frac{(5.767 \times 10^{-5})(1.2 \times 10^{19}) e^{-\frac{0}{8.6 t}}}{(28,500 \text{ cm}^3/\text{ft}^3)(4\pi)(36)(2.15)(100)} \quad (48) \\ &= 2.5 \times 10^5 \text{ rads/hr} \end{aligned}$$

The dose at various distances from the source may be obtained by substituting the desired value of "r" in the above equation. The dose at the surface will persist if thermal convection brings the warmer water to the surface where it will remain and spread horizontally with time.

It is probable that the actual dose rates from an accident of the type under consideration would be much lower if the fission products were deposited at least 10 ft below the water surface, and hence, the radiation will be attenuated. Further, the fission products, except for iodine and bromine, tend to attach to sediment and settle out.

3. Solubility of the Fission Product Gases

The question arises as to whether the gaseous fission products will dissolve in the water or escape as a cloud.

For one week's operation at 100 Mw with holdup of all the fission product gases, about 30 gram-moles of gas will be present, about 80% of which is xenon.

Assume water at 10°C (46°F); Henry's Law constant,⁹ $c = 0.555 \times 10^7$ and 30 gram-moles gas released at a depth of 36 ft,

$$c = \frac{p}{n} \quad (54)$$

where p = partial pressure of the xenon

n = mole fraction in solution

$$n = \frac{p}{c} = \frac{14.7 + \left(\frac{62.4}{144} \times 36\right)}{0.555 \times 10^7} = 5.0 \times 10^{-6} \frac{\text{moles Xe}}{\text{moles H}_2\text{O}} \quad (55)$$

$$\begin{aligned} 1 \text{ ft}^3 \text{ H}_2\text{O} &= 62.4 \left(\frac{\text{lb}}{\text{ft}^3} \right) \left(\frac{1000 \text{ gm}}{2.2 \text{ lb}} \right) \left(\frac{\text{mole}}{18 \text{ gm}} \right) \\ &= 1,550 \text{ moles H}_2\text{O/ft}^3 \end{aligned} \quad (56)$$

$$\begin{aligned} 30 \text{ gm moles Xe requires } &\frac{30}{(1550)(5.0 \times 10^{-6})} \\ &= 3,870 \text{ ft}^3 \text{ of H}_2\text{O} \end{aligned} \quad (57)$$

Assuming 10 meters (36 ft³) depth,

$$V = 36 \pi R^2 \quad (58)$$

$$R = \left(\frac{3870}{36 \pi} \right)^{1/2} = (34)^{1/2} = 6 \text{ ft}$$

With this required volume and in view of the low column velocities, it is quite possible that this amount of gas will dissolve before reaching the surface. Furthermore, if extremely high diffusion coefficients can be applied or the gas is spread over a large area during release, assuming all the gas was released, it will almost certainly be dissolved.

TABLE 1. TURBULENT EDDY DIFFUSION COEFFICIENTS

Horizontal	
<u>A_h, cm²/sec</u>	<u>Source</u>
1.2 - 1.8 x 10 ⁵	Measurement, Bikini area ⁸
20-1000	Measurement, dye marker ¹⁰
3.23 x 10 ⁵	Measurement, Mohawk River ²
10 ⁵	Calculated, ideal reservoir ⁶
Vertical	
<u>A_v, cm²/sec</u>	<u>Source</u>
300-360	Measurement, Bikini area ⁸
2000	Measurement, Bikini area (special case of diurnal heating and advection over a reef)
1-300	Measurement, oceans ¹ (a function of stability and currents)
Ratio A/r	
<u>A/r, cm/sec</u>	<u>Source</u>
0.5	Measurement, Bikini area ⁸
0.3 - 0.5	Suggested range, oceans ¹
0.2	Measurement, Framingham Reservoir ⁶

References

1. Sverdrup, Johnson, Fleming, "The Oceans", Prentice-Hall, Inc., 1942; Reprint 1949.
2. Glover, Robert E., "The Dispersion of Dissolved or Suspended Materials in Flowing Streams", Preliminary Draft, United States Department of the Interior, June, 1956.
3. Richardson, L. F., "Atmospheric Diffusion on a Distance-Neighbor Graph", Proc. Royal Society of London, 107A, 709, 1926.
4. Stommel, H., "Hydrography of the Western Atlantic, Diffusion Due to Oceanic Turbulence", Woods Hole Oceanographic Institution, NP-926.
5. Sutton, O. G., "Atmospheric Turbulence", Methuen and Company, Ltd., London, 2nd ed., 1955.
6. Parker, F. L., "Mechanisms of Dilution of Radiocontaminants in Streams", Thesis, Harvard University, 1955.
7. Conference on the Disposal of Radioactive Waste; Woods Hole Oceanographic Institute, August 5-6, 1954, Unpublished.
8. Munk, Ewing, Revelle, "Diffusion in the Bikini Lagoon", Trans. American Geophysical Union, Vol. 30, No. 1, February, 1949.
9. Hodgman, C. D., "Handbook of Chemistry and Physics", Chemical Rubber Publishing Co., 30th Edition, 1946.
10. Burke, C. J., "Horizontal Diffusion of Dyes in the Ocean", Scripps Institute of Oceanography, Oceanographics Report No. 2, Unpublished, 1946.
11. Blizzard, E. P., "Nuclear Radiation Shielding", (prepared for the McGraw-Hill Nuclear Reactor Handbook), September 17, 1956.

List of Symbols

A	diffusion coefficient, cm^2/sec or ft^2/sec
a	acceleration, ft/sec^2
α	$\mu^2 At$
α'	$r^2/4At$
B	$r^2/4A$
c	Henry's Law Constant, $\text{psi}/\text{mole fraction dissolved}$
C_p	specific heat at constant pressure, $\text{Btu}/\text{lb-}^\circ\text{F}$
D'	total derivative operator
D°	dose rate, rad/hr
D	depth of water, ft or cm
∇	Laplacian operator
g	acceleration of gravity, $32.2 \text{ ft}/\text{sec}^2$
h	volume heat, Btu/ft^3
H_a	heat rate, Btu/sec
H_o	initial heat, Btu
H_L	latent heat, Btu
k	experimental constant
$K(\ell)$	diffusion coefficient based on distance-neighbor theory
ℓ	distance between two particles
λ	decay constant, sec^{-1}
μ	linear absorption coefficient, cm^{-1}
μ_a/ρ	mass absorption coefficient, cm^2/g
n	mole fraction in solution
p	partial pressure, psi
Q	concentration, $\text{no.}/\text{cm}^3$ or $\text{no.}/\text{ft}^3$
R	removal function, $\text{n}/\text{cm}^3\text{-sec}$
r	radial distance from source, cylindrical or spherical geometry
ρ	density, g/cm^3 or lb/ft^3
S_o	quantity of material
s_o	quantity of material/sec
s	distance
T	temperature
T_o	initial temperature

List of Symbols (continued)

t	time, sec
V	volume
v	velocity, cm/sec
w	$(\mu_r/2\alpha^{1/2})^2 + \mu_r + \alpha$
W	$r^2/4A (t - \xi)$
ξ	time, sec
β	beta particles
γ	gamma rays
X	F (r,A,t), see section 1.1.1
x	distance in x direction, cm or ft
y	distance in y direction, cm or ft
z	distance in z direction, cm or ft

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FIGURE 1 CONCENTRATION FOR A = 3 cm²/sec

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FIGURE 2. CONCENTRATION FOR A = 30 CM²/SEC

FIG.3 CONCENTRATION FOR A = 300 cm²/sec

FIGURE 4 - CONCENTRATION FOR $A = 10^5$ CM²/SEC

FIGURE 7. DISTANCE AT TIME AFTER RELEASE OF 1.2×10^{19} MEV/SEC
FISSION PRODUCTS IN WATER

FIGURE 6. - CONCENTRATION IN A CHANNEL

FIGURE 7 - CONCENTRATION IN A CHANNEL

FIGURE 8 - CONCENTRATION IN A CHANNEL

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