VOLATILE FISSION PRODUCTS

SOME ASPECTS OF VOLATILE FISSION PRODUCT BEHAVIOUR AND ASSOCIATED POPULATION DOSE IN POSTULATED ACCIDENTS

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ABSTRACT

In this report, models, equations, and methodology useful in the analyses of removal of radioactive iodine in the form of elemental iodine and methyl iodide from the reactor containment by sprays and deposition after a postulated Loss of Coolant and Loss of Emergency Core Coolant (LOC/LOECC) accident are discussed.

Also discussed are applicable methodology useful in the estimation of population dose as a result of the escape of radioactive iodine and noble gases out of the containment after such a postulated accident. A computer program POPDOSE was written to estimate population dose based on this methodology.

Results of analyses and details of POPDOSE are not important for the purposes of this report and are not included here.

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CONTENTS

Part I

Removal of Elemental Iodine and Methyl Iodide From the Containment Atmosphere by Sprays and Deposition

Part II

POPULATION DOSAGE AS A RESULT OF RELEASE OF RELEASE OF RADIOACTIVE IODINE AND NOBLE GASES FROM THE REACTOR CONTAINMENT AFTER A SINGLE FAILURE

LIST OF FIGURES

LIST OF TABLES

Part I

Removal of Elemental Iodine and Methyl Iodide from the Containment Atmosphere by Sprays and Deposition

1 Introduction

In the safety analysis of reactors, accidents are postulated some of which may lead to fuel failures and release of radioactive fission products to the atmosphere within the containment.* Among the various radioactive fission products released the iodine isotopes are the most biologically significant due to their abundance, volatility and ability to concentrate in a single organ (the thyroid).

Radioactive iodine may be released in different chemical forms: 99 % is expected to be a mixture of elemental iodine and hypo-iodous acid with the remaining 1 % being organic iodide. After release from the core the fission products will enter the containment building where some of the iodine will be removed from the atmosphere by water sprays and some will deposit on surfaces.

This part of the report looks at models, equations, and methodology useful in analysing the removal of elemental iodine and organic methyl iodide by doped water sprays and deposition.

2 Removal of Elemental Iodine

Elemental iodine can be removed at an appreciable rate

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* It should be noted that this does not imply that the fission products would then be released to the external atmosphere.

by sprays and the natural process of deposition onto surfaces inside the containment.

2.1 Removal by Spray

2.1.1 Model of Spray Removal

Elemental iodine reacts with water and therefore may be rapidly absorbed by sprays. The removal rate may be predicted from a model in which the spray is considered to be an assemblage of noninteracting single drops (Reference 1). The overall drop absorption process includes the following steps:

(a) mass transfer of elemental iodine across the gas film,

(b) equilibrium dissolution at the gas-liquid interface,

(c) diffusion into the drop,

(d) reaction within the liquid phase.

2.1.2 Removal Rate

The rate of change of iodine concentration in the reactor building atmosphere due to spray removal is,

$$\frac{dq}{dt} = -\lambda_s t \qquad (1)$$

Simplifying,

$$\frac{q}{q_o} = e^{-\lambda_s t} \qquad (2)$$

where t = time (s)

q_o = quantity of iodine initially released (g)

q = quantity of iodine (g)

The spray removal rate constant, λ_s, is related to the spray flow rate and drop absorption efficiency E by (Reference 2) :

$$\lambda_s = \frac{FHE}{V} \qquad (3)$$

where H = equilibrium partition coefficient

E = drop efficiency

V = volume of contained gas (cm^3)

F = volumetric flow rate of spray (cm^3/s)

The drop absorption efficiency, E, in equation 3 is given by (Reference 2),

$$E = 1 - \exp(-6k_g t_e / d(H + k_g/k_l)) \qquad (4)$$

where $k_g = \frac{D_v}{d}(2 + 0.6\,Re^{0.5}Sc^{0.33})$

= gas phase mass transfer coefficient

$k_l = 2\pi D_\ell^2 / 3d$

= liquid phase mass transfer coefficient

t_e = drop exposure time (s)

H = equilibrium partition coefficient

d = diameter of a spray drop (cm)

D_v, D_ℓ = diffusivity of iodine in gas and liquid phases respectively (cm^2/sec)

R_e = Reynolds number $\rho vd/\mu$, where ρ is density of air stream, v is velocity of air stream, and μ is viscosity of air stream.

Sc = Schmidt number, $\mu/\rho D_v$

As soon as the spray removal rate constant, λ_s, is evaluated, the time, τ_{q/q_o}, required to reduce the iodine concentration to any given decontamination factor, q/q_o, can be obtained by taking the natural logarithms of both sides of equation 2, yielding

$$\log_e(q/q_o) = -\lambda_s \tau_{q/q_o} \qquad (5)$$

or $$\tau_{q/q_o} = \frac{-\log_e(q/q_o)}{\lambda_s} \quad (6)$$

Thus, we have all the relevant equations to work with in estimating the transient of the concentration of elemental iodine released into the containment atmosphere after a postulated LOC/LOECC* accident, assuming that spray is available continuously.

2.2 Removal by Deposition

2.2.1 Removal Rate Constant and Deposition Velocity

Elemental iodine is also removed by natural deposition in addition to removal by engineered safety systems. If deposition is the only iodine removal mechanism, the rate is given by :

$$-\frac{dq}{dt} = \lambda_d t$$

or $$q/q_o = e^{-\lambda_d t} \quad (7)$$

where q/q_o = fraction of initial iodine remaining

λ_d = deposition removal rate constant

t = time after release of iodine

The deposition removal rate constant , λ_d , is related to the area to volume ratio, A/V, of the containment volume and an important parameter —— deposition velocity V_g by:

$$\lambda_d = V_g A/V \quad (8)$$

where V_g = deposition velocity (cm/s)

A = area of surface available for deposition (cm^2)

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* Postulated Loss of Coolant with Simultaneous Loss of Emergency Coolant.

V = gas volume of containment (cm^3)

2.2.2 Model of Deposition of Iodine and Deposition Velocity

In estimating V_g, I shall adopt the model developed by Knudson and Hilliard (Reference 3). It is a model in which fission products are considered to be released into a well mixed steam-air atmosphere in the containment volume and the containment wall is covered with a thin flowing film of water. In such a model the deposition velocity, V_g, is practically equal to the gas phase mass transfer coefficient, k_c, of iodine through the gas boundary layer at the walls of the containment.

The gas phase mass transfer coefficient, k_c, is a function of the temperature difference, $(T_b - T_{si})$, between the bulk gas and the containment wall. The value of k_c can be read off a graph of k_c versus $(T_b - T_{si})$, which can be found in reference 4.

Thus, we can evaluate the removal rate constant, λ_d, if we know the details of the temperature difference $(T_b - T_{si})$. In the case that such detail is not known, from a practical standpoint, a lower bound of 1 ^oF for the temperature difference $(T_b - T_{si})$ can be assumed(Reference 5). A conservative estimate of k_c or V_g can then be obtained.

3 Removal of Methyl Iodide

Methyl iodide is removed only very slowly by natural deposition and the removal through this channel can then be neglected. However,methyl iodide may be removed at a reasonable rate by aqueous sprays doped with various reagents.

The removal of methyl iodide by spray is achieved through two channels. First, the iodide is absorbed by spray drops. Second, the spray will wet the containment wall, and the iodide is absorbed into the spray film formed on the wall.

The overall removal rate constant λ is then given by

$$\lambda = \lambda_{\text{wall film}} + \lambda_{\text{drop}} \qquad (9)$$

where λ_{wall} = removal rate constant by wall film absorption

λ_{drop} = removal rate constant by spray drops

Since these two processes are quite independent of each other, each channel will be discussed separately.

3.1 Removal by Spray Drops

In the following discussion, I shall adopt the model developed by Schwendiman, Hasty, and Postma (Reference 2,4,6). In this model, liquid drops are formed due to the break up of liquid sheet or jets. The drop soon comes to its terminal velocity so that the drop exposure time, t, can be approximated by the height of the fall divided by the terminal velocity. While a drop is falling, methyl iodide is absorbed into the drop at a rate determined by the solubility and first order reaction rate of methyl iodide in the falling drop. As methyl iodide dissolves only slightly in and reacts slowly with water, a large reaction rate can only be achieved with the doping of the spray with a suitable reagent at an appropriate concentration. This is expected to help increase the absorption rate of methyl iodide into the drop. At the end

of the fall of the drop, the amount of methyl iodide absorbed by a drop is given by

$$\frac{Q}{DaC^*t} = 8\pi \sum_{n=1}^{\alpha} \frac{\alpha + (\alpha + \beta n^2\pi^2)(1-\exp(-\alpha - \beta n^2\pi^2))}{\alpha + \beta n^2\pi^2} \qquad (10)$$

$$= \frac{4}{3}\pi \frac{E_d}{\beta} \qquad (10a)$$

where Q = mass of methyl iodide absorbed (g)

D = diffusivity of methyl iodide in liquid drop (cm^2/s)

a = radius of drop (cm)

C^* = concentration of solute in liquid at drop interface (g/cm^2)

t = drop exposure time (s)

α = kt

β = Dt/a^2

k = first order reaction rate constant within the drop ($1/s$)

E_d = fraction of saturation

$$= \frac{\text{average solute conc. at the end of fall}}{\text{equilibrium conc. neglecting reaction}}$$

The removal rate constant λ_{drop} is related to the fraction of saturation, E_d, in equation 10 by

$$\lambda_{drop} = \frac{F_s H E_d}{V} \qquad (11)$$

where F_s = spray flow rate (cm^3/s)

H = methyl iodide partition coefficient

E_d = fractional saturation achieved during single pass

V = volume of containment gas space (cm^3)

In order to evaluate E_d, we have to solve the right

hand side of equation 10. Numerical values of equation 10 can be obtained from reference 4 provided we input α and β. Alternatively, a small program can be written to evaluate the right hand side of equation 10. Once the right hand side of equation 10 is evaluated, E_d can be obtained with equation 10a, and then λ_{drop} can be evaluated with equation 11.

The partition coefficient of methyl iodide, H, in equation 11, is given by (Reference 6) :

$$\log H = 4.82 + \frac{1597}{T} \qquad (12)$$

where $\log H$ = logarithm to the base 10 of the partition coefficient H

T = absolute temperature in ^{o}K

The first order reaction rate constant, k, depends on both concentration of reagents in spray solution and temperature. Usually, the so called second order reaction rate K_o for different reagents at 25 ^{o}C can be found in the literature, e.g. reference 6. The first order reaction rate, k, at 25 ^{o}C is thus given by

$$k = K_o C$$

where K_o = second order reaction rate (litre mole^{-1} s^{-1})

C = concentration of reagent in spray solution (mole litre^{-1})

At temperature other than 25 ^{o}C, k can be obtained by

$$\log \frac{k_2}{k_1} = \frac{E_a}{G}\left(\frac{1}{T_1} - \frac{1}{T_2}\right) \qquad (13)$$

where T_2 = temperature other than 25 ^{o}C (^{o}K)

T_1 = 25 °C

E_a = activation energy (kcal $mole^{-1}$)

G = universal gas constant (cal $mole^{-1}$ $^oK^{-1}$)

universal gas constant (cal $mole^{-1}$ $^oK^{-1}$)

The quantity E_a is always found along with K_o in the literature.

3.2 Removal by Wall Film

After the spray is turned on, the containment will soon be covered with a flowing film formed by the spray drops. Methyl iodide can be absorbed into this wall film and thus removed from the atmosphere. From a model discussed in reference 4, the rate of removal of methyl iodide by wall film can be conservatively estimated as the following. The removal rate constant, $\lambda_{\text{wall film}}$, is given by (reference 4) :

$$\lambda_{wall} = \frac{q}{C} \frac{A}{V} \tag{14}$$

where q/C_g = $\frac{\text{absorption rate per unit area}}{\text{gas phase concentration}}$

A = surface area of wall film

V = volume of gas space.

The numerical value of q/C_g is given by (reference 4)

$$q/C_g = H\sqrt{k}D \tanh\left(\frac{\sqrt{k}}{D}\delta\right) \tag{15}$$

where H = partition coefficient

k = first order reaction rate constant

D = diffusivity of methyl iodide in water

δ = thickness of wall film

The film thickness, δ , may be predicted from laminar flow theory and is given by (reference 4) :

$$\delta = \left(\frac{3\nu\Gamma}{g}\right)^{1/3} \tag{16}$$

where δ = film thickness on vertical wall

ν = kinematic viscosity of water film

Γ = film flow rate per unit length of perimeter

g = acceleration due to gravity

Or,
$$\lambda_{\text{wall film}} = \frac{HA}{V}\sqrt{kD}\ \tanh\left(\sqrt{\frac{k}{D}}\,\delta\right) \tag{17}$$

Part II

POPULATION DOSAGE AS A RESULT OF RELEASE OF RADIOACTIVE IODINE AND NOBLE GASES FROM THE REACTOR CONTAINMENT AFTER A SINGLE FAILURE

4 Introduction

In a postulated reator accident, e.g. LOC/LOECC, isotopes of the noble gases (Kryton and Xenon) and iodine are by far the most abundant fission products released to the containment atmosphere. Following such an accident, the pressure in the containment rises due to the entry of steam,* hydrogen and gaseous fission products. Hence, there is a potential for leakage of some fission products to the outside environment. This potential for leakage will continue for many days unless the pressure is reduced by intentional discharge through filters. Thus, although the bulk would be contained, a small portion of these fission products may find its way out of the containment through small leaks. The effluent, after being released to the atmosphere outside the reactor, may be carried away from the reactor by the wind and turbulence. As a result, a person who happens to be in the path of the effluent will receive some radiation dosage.

In this report, I shall discuss briefly applicable models which describe the spread of the effluent under different meteorological conditions. With these models, I

* This is only true for single unit CANDU containments. For multiple unit stations, a vaccuum building is connected to containment, resulting in subatmospheric containment pressures following accidents.

shall also outline the procedure in estimating the population (collective) dosage received by the public. In particular, I shall discuss the procedure for estimating the population dosage due to the intake of isotopes of iodine released, and that due to external gamma irradiation by the radioactive noble gases. In both cases, I have assumed that there is no deposition and no settling of the released fission products although radioacitve decay will be taken into accout. Also, I have assumed that the terrain complexity in the path of the effluent is uniform. With these assumptions, the estimates made will probably be conservative in the sense that the calculation will predict a larger dose than would be observed in experiments.

5 Spread of Effluent

Once a radioactive gas or aerosol becomes air-borne it travels and disperses in a manner governed by its own physical properties and those of the ambient atmosphere into which it is released. The effluent enters the atmosphere with a certain velocity and temperature which are generally different from those of the ambient. The effluent motion has a vertical component because of the combined effect of initial momentum and buoyancy (either positive or negative) until these properties are dissipated. The vertical rise of the effluent due to this motion is called plume rise and has the net effect of changing the effective height of the release.

During and after plume rise the effluent is transported by the wind. The turbulent motion of the atmosphere also

causes random movement of the effluent resulting in its progressive lateral and vertical dispersion and its dilution by mixing with air. This process is called atmospheric diffusion.

The spread of the effluent by these mechanisms can be described by a mathematical model.

5.1 Mathematical Model

Let us consider the idealized case of a single puff of an effluent released to the atmosphere. Because of turbulent diffusion its size will grow with travel time as it moves downwind. The concentration of radioactivity within the puff can be described by (reference 7)

$$\chi(x,y,z,t) = \frac{Q_o}{(2\pi)^{3/2}\sigma_x\sigma_y\sigma_z} \exp\left\{-\frac{1}{2}\left[\frac{(x-x_o-\bar{u}t)^2}{\sigma_x^2} + \frac{(y-y_o)^2}{\sigma_y^2} + \frac{(z-z_o)^2}{\sigma_z^2}\right]\right\} \qquad (18)$$

where Q_o = the quantity of the instantaneous source (Ci)

x_o, y_o, z_o = the co-ordinates of the source of release and with the x-axis in the downwind direction, the y-axis in the crosswind direction and the z-axis in the vertical direction

t = time after release (s)

$\sigma_x, \sigma_y, \sigma_z$ = the standard deviations of the concentration distributions along the three co-ordinate directions at the centroid of the cloud at time t

$\bar{u}$ = mean wind speed (m/s)

If the radioactivity is released continuously rather than in a puff, to sum the contributions from the succession of continuous "puffs", we can integrate equation 18 along the

x-axis when the wind speed $\bar{u}$ is not near zero, using the transformation relation: $x = \bar{u}t$ and assuming that downwind diffusion can be neglected as compared with advective transport (reference 7). In this case we get

$$\chi(x,y,z) = \frac{Q'}{2\pi\bar{u}\sigma_y\sigma_z} \exp\left\{-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2} + \frac{z^2}{\sigma_z^2}\right)\right\} \tag{19}$$

In equation 19, Q' is the rate of release of radioactivity. Also, the co-ordinates of the source, (x_o, y_o, z_o) are set arbitrarity at zero.

It is often important to calculate ground level concentrations arising from elevated sources, such as the release from a stack. We can do this by first shifting the origin of the co-ordinate system in equation 19. Also, assuming that the plume is reflected at the ground, we get

$$\chi(x,y,z) = \frac{Q'}{2\pi\bar{u}\sigma_y\sigma_z}\left[\exp\left(\frac{-y^2}{2\sigma_y^2}\right)\right]\left[\exp\left\{-\frac{(z-H)^2}{2\sigma_z^2}\right\} + \exp\left\{-\frac{(z+H)^2}{2\sigma_z^2}\right\}\right] \tag{20}$$

where H is the effective stack height (see section 5.3 below)

We can then obtain the ground level concentration by setting z=0 in equation 20, yielding:

$$\chi(x,y,0) = \frac{Q'}{\pi u\sigma_y\sigma_z} \exp\left[-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2} + \frac{H^2}{\sigma_z^2}\right)\right] \tag{21}$$

The ground level concentration along a line below the plume centre line is obtained by letting y=0 in equation 4, yielding

$$\chi(x,0,0) = \frac{Q'}{\pi\bar{u}\sigma_y\sigma_z} \exp\left\{-\frac{H^2}{2\sigma_z^2}\right\} \tag{22}$$

The time integral of the concentration (T.I.C.) of radioactive material at a point is required for calculating the related dose. In short term continuous release, a conservative T.I.C. is obtained by replacing Q' in equation 19 with the total quantity of release Q (reference 7), yielding,

$$\frac{\text{T.I.C.}}{Q} = \frac{1}{2\pi\bar{u}\sigma_y\sigma_z}\exp\left\{-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2}+\frac{z^2}{\sigma_z^2}\right)\right\} = \psi(x,y,z) \tag{23}$$

Similarly, equations 21 and 22 respectively become,

$$\frac{\text{T.I.C.}}{Q} = \frac{1}{\pi\bar{u}\sigma_y\sigma_z}\exp\left\{-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2}+\frac{H^2}{\sigma_z^2}\right)\right\} = \psi(x,y,0) \tag{24}$$

$$\frac{\text{T.I.C.}}{Q} = \frac{1}{\pi\bar{u}\sigma_y\sigma_z}\exp\left\{\frac{H^2}{2\sigma_z^2}\right\} = \psi(x,0,0) \tag{25}$$

The left hand side of equation 23, 24, and 25, i.e., T.I.C. per Ci of radioactivity released is defined as specific exposure, ψ, and is an important quantity in estimating the radiation dose per Ci of release received by a person at a point.

5.2 Stability Classes and Determination of σ's

As the diffusion of the radioactivity is highly influenced by turbulence which in turn is determined by atmosphere stability, we can expect the 'measure of spread of the radioactivity' (σ) to be a function of atmospheric stability. Conversely, atmospheric stability can be identified with the dependence of σ's on downwind distance. Pasquill

(reference 9) has developed a method for classifying atmospheric stability. He grouped stability condition into 7 classes A - G, ranging from extremely unstable to extremely stable. Smith-Hosker (reference 8), in parallel to Pasquill, also classified atmoshperic stability into classes A - G, and give the dependece of σ_y and σ_z on stability classes analytically as the following

$$\sigma_y(x) = C_3 x / (1 + 0.0001x)^{1/2} \tag{26}$$

$$\sigma_z(x) = g(x) F(z_o, x) \tag{27}$$

with $$g(x) = a_1 x^{b_1} / (1 + a_2 x^{b_2}) \tag{28}$$

and $$F(z_o, x) = \ell n \left\{ C_1 x^{d_1} (1 + (C_2 x^{d_2})^{-1} \right\} \text{ if } z_o > 10 \text{ cm} \tag{29}$$

or $$F(z_o, x) = \ell n \left\{ C_1 x^{d_1} / (1 + C_2 x^{d_2}) \right\} \text{ if } z_o \leq 10 \text{ cm} \tag{30}$$

In equations 26 - 30, the quantity z_o is roughness length describing the terrain complexity of the land in the path of the plume. The parameters c_3, a_1, b_1, a_2, b_2, c_1, d_1, c_2, d_2 as a function of stability classes A - G are given in Table 1, 2, and 3.

Other authors have published analytic and graphic means of obtaining σ_y and σ_z for all the Pasquill stability categories (reference 9, 10). However, the Smith/ Hosker scheme is adopted in the present analysis.

The knowledge of the dependence of σ_y and σ_z anables the prediction of the specific exposure ψ which depends on

TABLE 1

Parameters for g(x) in Equations 28

Stability Class	a_1	b_1	a_2	b_2
A	0.112	1.060	5.38×10^{-4}	0.815
B	0.130	0.950	6.52×10^{-4}	0.750
C	0.112	0.920	9.05×10^{-4}	0.718
D	0.098	0.889	1.35×10^{-3}	0.688
E	0.0609	0.895	1.96×10^{-3}	0.684
F	0.0638	0.783	1.36×10^{-3}	0.672

TABLE 2

Parameters for $F(z_0,x)$ in Equation 29 or 30

Roughness length	c_1	d_1	c_2	d_2
1 cm	1.56	0.048	6.25×10^{-4}	0.45
4 cm	2.02	0.027	7.76×10^{-4}	0.37
10 cm	e	0.	0.	0.
40 cm	5.16	-0.098	1.86×10	-0.225
100 cm	7.37	-0.0957	4.29×10^{3}	-0.60
400 cm	11.7	-0.128	4.59×10^{4}	-0.78

TABLE 3

Parameters for $\sigma_y(x)$ in Equation 26

Stability Class	A	B	C	D	E	F
c_3	0.22	0.16	0.11	0.08	0.06	0.04

σ_y and σ_z (section 5.1). Knowing ψ we can then proceed with the estimate of dose.

5.3 Effective Stack Height, H and Plume Rise

The effective stack height H in the expressions for χ, and specific exposure ψ is obtained by adding the plume rise to the physical stack height.

5.3.1 Plume Rise for Tall Stacks

Stacks that are at least twice the height of adjacent solid structure are classified as tall stacks. For effluents released from tall stacks, the plume rise, h, can be estimated using the following equations according to weather stability.

(a) neutral stability (reference 8)

$$h = 1.44 D (w_o/\bar{u})^{2/3} (x/D)^{1/3} - C \qquad (31)$$

where h = plume rise (m)

w_o = exit velocity (m/s)

x = distance downwind (m)

$\bar{u}$ = wind speed (m/s)

D = internal stack diameter (m)

and $C = 2D(1.5 - w_o/\bar{u})$

For this case, also use

$$h = 3(w_o/\bar{u})D \qquad (32)$$

The value of h to be adopted is the lower of the two obtained from equation 31 and 32.

(b) Stable Conditions (reference 8)

The results from equation 31 should be compared with the results from the following two equations:

$$h = 4(F_m/S)^{1/4} \tag{33}$$

$$\text{or} \quad h = 1.5\, S^{-1/6} (F_m/u)^{1/3} \tag{34}$$

and the smallest value of h is used. In these equations F_m and S are defined as

$$F_m = w_o^2 (D/2)^2$$

$$S = \begin{cases} 8.7 \times 10^{-4} & \text{for class E stability} \\ 1.75 \times 10^{-3} & \text{for class F stability} \\ 2.45 \times 10^{3} & \text{for class G stability} \end{cases}$$

5.3.2 Plume Rise for Short Stacks

Stacks of height less than twice the height of adjacent solid structures are classified as short stacks. Plume rise h for short stack are estimated according to the $w_o/\bar{u}$ ratio as the following (reference 8).

(a) $w_o/\bar{u} > 5$

We can use equation 31.

(b) $w_o/\bar{u} < 1$

We can equate h to zero for conservative estimates.

(c) $1 \leq w_o/\bar{u} \leq 5$

First estimate an entrainment coefficient E_t given by

$E_t = 2.58 - 1.58(w_o/\bar{u})$ for $1 \leq w_o/\bar{u} \leq 1.5$

$E_t = 0.3 - 0.06(w_o/\bar{u})$ for $1.5 < w_o/\bar{u} \leq 5.0$

Second, consider the release as tall stack release for $100(1 - E_t)$ percent of the time and a ground level release $100\ E_t$ percent of the time. Calculate the concentration for each case and then find the average, weighted with the fraction of that time that each release occurs.

Of course, whenever we don't know the details leading to the estimate of h_1 we can equate it to zero for a conservative estimate (substituting 0 into equation 25).

5.4 Building Wake Effect

Radioactive materials released through leaks in the buildings or from short stacks will be mixed with the turbulent wake created by the ambient air flow around the buildings. This effect results in an air flow around the buildings. This effect results in an increase in both σ_y and σ_z . This building wake effect can be taken account of by modifying σ_y and σ_z according to (reference 11)

$$\Sigma_y = (\sigma_y^2 + CA/\pi)^{1/2} \tag{35}$$

$$\Sigma_z = (\sigma_z^2 + CA/\pi)^{1/2} \tag{36}$$

where A = total area of building contributing to the building wake effect

C = building wake effect constant, usually 0.5

The estimate of χ and ψ can then be done by replacing σ's with Σ's in all the appropriate equations.

With the spread of effluent dealt with, we are in a position to proceed with the estimate of dose.

6 Population Dose due to Intake of Radioactive Iodine

6.1 Individual Dose and Specific Exposure

The critical organ for radioactive iodine is the thyroid (reference 14). The activity can reach the thyroid eventually by inhalation and ingestion by an individual. The amount of intake by an individual situated at a point on the ground is proportional to the time integrated concentration of iodine and thus specific exposure (ψ). Hence, the dose received by the same individual is also proportional to the specific exposure (ψ), that is

$$D = kQ\psi \tag{37}$$

where D = Iodine intake dose (rem/yr)

Q = activity released (Ci)

ψ = specific exposure (s/m^3)

k = dose conversion factor ($rem\text{-}yr^{-1}/s\text{-}m^{-3}$)

The dose conversion factor k varies from isotope to isotope for reasons such as different half-lives for different isotopes. It varies for different age groups even for the same isotope. The iodine intake dose conversion factor for a critical individual (6 month old child) and an average individual for the 5 Iodine isotopes present in the release are tabulated in Table 4 (reference 12 - 14).

6.2 Population Dose

The population dose, PD, is defined in the following way (reference 8):

TABLE 4

Dose Conversion Factors For Iodine

Isotope	Half-Life (s)	Critical Individual (rem-m^3-s^{-1})	Average Individual (rem-m^3-s^{-1})
I - 131	6.947×10^5	814.58	377.30
I - 132	8.136×10^3	16.30	6.34
I - 133	7.38×10^4	283.47	104.79
I - 134	3.120×10^3	3.18	1.33
I - 135	2.398×10^4	56.48	20.44

$$PD = \int_A D(x,y)\,\rho(x,y)\,dA \qquad (38)$$

where PD = collective dose (man-rem/yr)

D(x,y) = dose received by an individual at the point (x,y) (rem/yr)

$\rho(x,y)$ = population density ($1/km^2$)

A = area encircled by plume boundary (km^2)

6.3 Defining the Plume Boundary

On examination of equations 21 and 22, we can see that the activity concentration is continuously decreasing out to an infinite distance in both the y and x directions. To determine the population dose, we must first determine the boundary within which equation 38 to find population dose can apply. We therefore need to set arbitrary limits to the crosswind (y) and downwind (x) dimensions of the plume shaped cloud. The following are 3 possible methods of defining the plume boundary (reference 16).

(a) Method 1: Constant Specific Exposure Boundary

By this method, the plume boundary is a locus such that the specific exposure on each point on this locus has a constant value. Normally, this is taken to be 1 % of that at the station boundary on the centre-line. This is a closed boundary.

(b) Method 2: Constant Specific Exposure Ratio Boundary, (10 %)

By this method, the boundary is such that the specific exposure at a point (x,y,0) is of a constant percent, (usually) 10 %), of that at its projection on the x-axis, i.e. $\psi(x,y,0)$

is $0.1\,\psi(x,0,0)$.

In the x-directions, the boundary is a straight line perpendicular to the x-axis with the specific exposure at the intersecting point equal to a given per cent of that at the station boundary on the centre-line. It is a boundary formed by two paraboloids in the y-direction and closed by a straight line in the x-direction.

(c) Method 3, Constant Angle Boundary, (10 %)

By this method, the boundary is defined in the y-direction by two straight lines radiating from the reactor building and with the x-axis as the angle bisector. Usually, the angle is 10^{o}.

The boundary in the x-direction is similar to that in method 2.

6.4 Correction for Radioactive Decay

The dose received by a person is proportional to the time integrated concentration which in turn is proportional to the activity present (see equation 37). Since the activity of a given isotope decays with time, the effective dose a person receives from this isotope at distance x downwind must be corrected by a factor f_d given by:

$$f_d = \exp(-\lambda_d t)$$
$$= \exp(-\lambda_d x/\bar{u}) \qquad (39)$$

6.5 Methodology for calculating Population Dose

Having introduced all the necessary equations, I shall list the steps to be involved as the following:

(1) Calculate the specific exposure (Ψ_{CL1}) at the plume centre-line at the station boundary ($x = 1$ km) according to equation 25 using appropriate values of Σ_y and Σ_z obtained with equations 35 and 36 for $x = 1$ km. The effective stack height H in equation 25 is set to zero to obtain conservative estimate.

(2) Let the activity of the mixture of Iodine isotopes permitted to be released be Q_o. The activity of a given isotope i in this mixture is given by $Q_i = Q_o f_i$ where Q_i is the fraction of isotope i in equilibrium in the mixture. Note that this f_i varies from station to station. Next, obtain from Table 4 the dose conversion factor for a critical individual at the station boundary on the plume centre-line ($x = 1$ km) contributed by the ith isotope is then $Q_o k_i f_i \Psi(1,0,0)$. The dose received by the critical individual due to all the isotopes is then the sum of all the contributions. Equate this sum to the limit of thyroid dose set for a critical individual. Thus,

$$Q_o \Psi(1,0,0) \sum_{i=1}^{N} f_i k_i = D_{max} \qquad (40)$$

Hence, Q_o is obtained from equation 40 since every quantity except Q_o is known in that equation.

(3) We shall estimate the population specific exposure. We shall limit our attention to half the plume because of

symmetry about the centre-line of the plume. At a point on the centre-line (x,0,0), we shall calculate $\Sigma_y(x)$ and $\Sigma_z(x)$ according to equations 35,36,26, and 27. Calculate the specific exposure $\psi(x,0,0)$.

(4) Determine the y-coordinate for the plume boundary at x in step 3 dependent on the method of defining the plume boundary we have chosen (section 6.3).

(5) Increase y from 0 to Δy. Using equation 22, calculate $\psi(x,\Delta y,0)$

(6) Compare $\psi(x,\Delta y,0)$ with $\psi(x-\Delta x,\Delta y,0)$ which has been previously found. Do the same to $\psi(x,0,0)$ and $\psi(x-\Delta x,0,0)$. The highest of these four values is chosen as the local maximum ψ_{LM} for the mesh with corners at (x,0,0), (x,Δy,0), (x-Δx,Δy,0) and (x-Δx,0,0) and with are $\Delta x\Delta y$.

(7) The population density, $\rho(x)$, is obtained from some population density map. Multiply ψ_{LM}, $\Delta x\Delta y$, and $\rho(x)$ to obtain the 'differential' contribution by this mesh to the population specific exposure per Ci of activity released.

(8) Steps 5 to 7 are repeated with y changed from Δy to $2\Delta y$ and ψ_{LM} for the square with corners (x,2Δy,0), (x-Δx, 2Δy,0), (x-Δx, Δy,0) and (x, Δy,0) obtained in similar manner and so on until the y-coordinate for the plume-boundary determined in step 4 is just exceeded. The contributions from all squares between (x-Δx) and x are added to yield the contribution to population specific exposure by the strip between (x-Δx) and x.

(9) For each ith isotope, determine the correction factor for radioactive decay f_{di} at x according to equation 39. Multiply the strip contribution to population specific exposure obtined in step 8 by f_{di} to yield the decay corrected strip contribution to population specific exposure for the ith isotope. Add this decay corrected strip contribution to its respective subtotal previously determined for the ith isotope.

(10) Steps 3 to 9 are repeated for x increased to $(x+\Delta x)$ and so on until the boundary set for x is reached. At this point, the decay corrected total population specific exposure (half-plume) for each ith isotope is obtained.

(11) For each ith isotope, obtain from TABLE 4 the respective dose conversion factor for an average individual. Multiply this factor with the activity of the ith isotope released as determined in step 2 and its respective decay corrected population specific exposure to obtain the population dose attributive to the ith isotope.

(12) Add all the population dose attributive to each isotope to yield the total population dose (half-plume).

(13) Multiply the result obtained in Step 12 by two to obtain the total population dose.

7 Population Dose due to Whole-Body External Gamma-Irradiation by Noble Gases

7.1 Dose received from a Point Source

Calculations of the Gamma dose from an extended source

such as the cloud of radioactive Noble Gases start with consideration of the radiation received at the receptor from a differential area or volume that can be regarded as a point source. Taking attenuation of gamma photons by air into account, the gamma dose rate, $_{\gamma}D'$, to tissue at distances r from the point source is given by (reference 11)

$$_{\gamma}D' = \frac{0.040\ \mu_a q \bar{E}_{\gamma}(1+k\mu r)\exp(-\mu r)}{r^2} \qquad (41)$$

where $_{\gamma}D'$ = dose rate (rem/s)

μ_a = total absorption coefficient for air (m^{-1})

μ = total absorption coefficient for tissue (m^{-1})

$k = (\mu - \mu_a)/\mu_a$

q = radioactivity of point source (Ci)

$\bar{E}_{\gamma}$ = average gamma energy from decay (Mev)

r = distance between point source and receptor (m)

This dose rate is then integrated over the entire source with account being taken of the geometry of the source, variation in concentration, attenuation by interactions of the photons with matter in the path between source and receptor, and scattering of radiation from material outside the direct path to the receptor.

In the process of determining the gamma dose at a given point on the ground, there are three approaches that we can adopt in order to have a conservative estimate of the gamma dose rate. I shall outline each in the following section.

7.2 Gamma Dose from the Cloud

7.2.1 Infinite Cloud Dose

We shall consider a spherical cloud with a uniform concentration of a gamma emitting isotope and a radius equal to the range of the gamma photon. Such a cloud could be considered an infinite cloud for a receptor at the centre because any additional gamma-emitting material beyond the cloud dimensions would not alter the flux of gamma photons to the receptor. Integrating equation 41 yields the infinite cloud gamma dose as (reference 11)

$$ {}_{\gamma}D_{\infty}(x,y,0) = 0.25\,\bar{E}_{\gamma} Q \psi(x,y,0) \qquad (42) $$

where ${}_{\gamma}D_{\infty}(x,y,0)$ = infinite cloud gamma dose (rem)

$\psi(x,y,0)$ = specific exposure at point (x,y,0) (s/m^3)

$\bar{E}_{\gamma}$ = average gamma ray energy (MeV)

Of course, in reality, the cloud is not infinite and the concentration of radioactivity needs not be constant for a distance equal to the range of the gamma rate. An advantage of this model is the simplicity of the equation and it is ideal for programming.

7.2.2 Finite Cloud Dose

The gamma dose due to a finite cloud can be obtained by replacing q in equation 41 with $\psi(x,y,z,t)dV$ and integrating throughout the entire cloud volume and through all time. This integration will lead to (reference 11)

$$ {}_{\gamma}D(x,y,0) = \frac{0.1616\,\mu\,\mu_a \bar{E}_{\gamma} Q\,(I_1 + kI_2)}{\bar{u}} \qquad (43) $$

with $$I_1 = \frac{\bar{u}}{4(2\pi)^{1/2}\mu\Sigma}\int_0^\infty\int_0^\infty \frac{\exp(-\mu r)}{mr}\left\{\exp\left[\frac{-(m-r)^2}{2\Sigma^2}\right]-\exp\left[\frac{-(m+r)^2}{2\Sigma^2}\right]\right\}dr\,dt \qquad (44)$$

$$I_2 = \frac{\bar{u}}{4(2\pi)^2\mu\Sigma}\int_0^\infty\int_0^\infty \frac{\mu\exp(-\mu r)}{m}\left\{\exp\left[\frac{-(m-r)^2}{2\Sigma^2}\right]-\exp\left[\frac{-(m+r)^2}{2\Sigma^2}\right]\right\}dr\,dt \qquad (45)$$

where m = distance between the receptor and the centre of the cloud (m)

$$\Sigma = (\Sigma_y \Sigma_z)^{1/2} \quad \text{(m)}$$

and $_\gamma D(x,y,0)$, μ, μ_a, $\bar{E}_\gamma$, Q, k, are defined as before in section 7.2.1.

The two integrals I_1 and I_2 can be read off Figures 1 and 2 which are reproduced from reference 11.

It is interesting to note that the ratio of finite cloud dose to infinite cloud dose is given by

$$\frac{_\gamma D(x,y,0)}{_\gamma D(x,y,0)} = \frac{0.1616\,\mu\mu_a\bar{E}_\gamma Q\,(I_1 + kI_2)\,/\,\bar{u}}{0.25\,Q\exp(-y^2/2\Sigma_y)/\pi\bar{u}\Sigma^2}$$

$$= \frac{2.03\,\mu\mu_a\Sigma^2(I_1 + kI_2)}{\exp(-y^2/2\Sigma_y^2)} \qquad (46)$$

$$= f$$

Thus,

$$_\gamma D(x,y,0) = f\,{}_\gamma D_\infty(x,y,0)$$

$$= 0.25\,\bar{E}_\gamma Q\psi(x,y,0)\,f$$

$$= Q\psi F$$

with $$F = 0.25\,\bar{E}_\gamma f$$

An important graph of f versus Σ for $\bar{E}$ of 0.7 MeV is given on page 345 of reference 5. This graph is useful as 0.7 MeV is the average gamma ray energy of Noble Gases released in

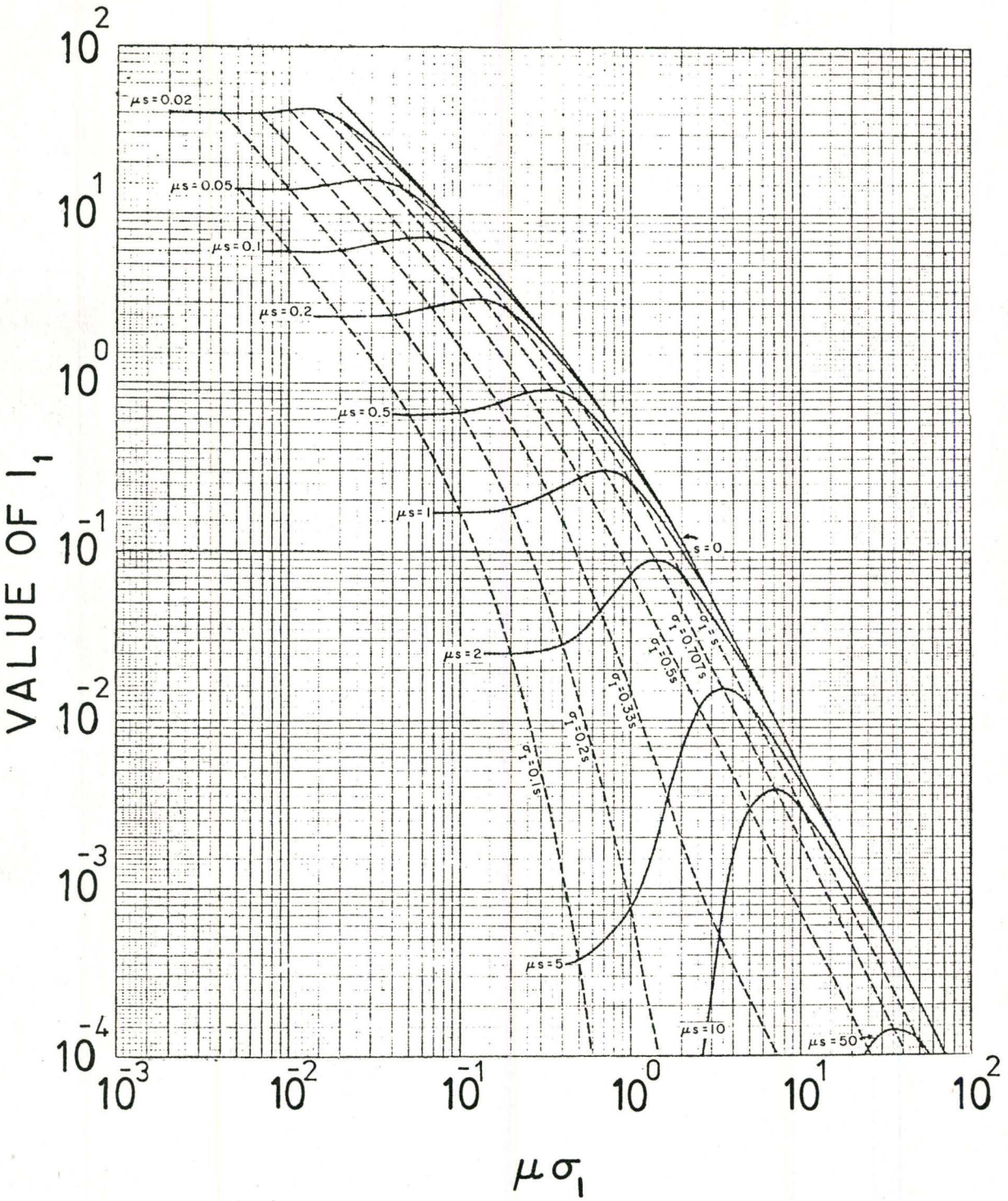

Figure 1. Values of the I_1 integral. Values of σ or σ_I can be used.

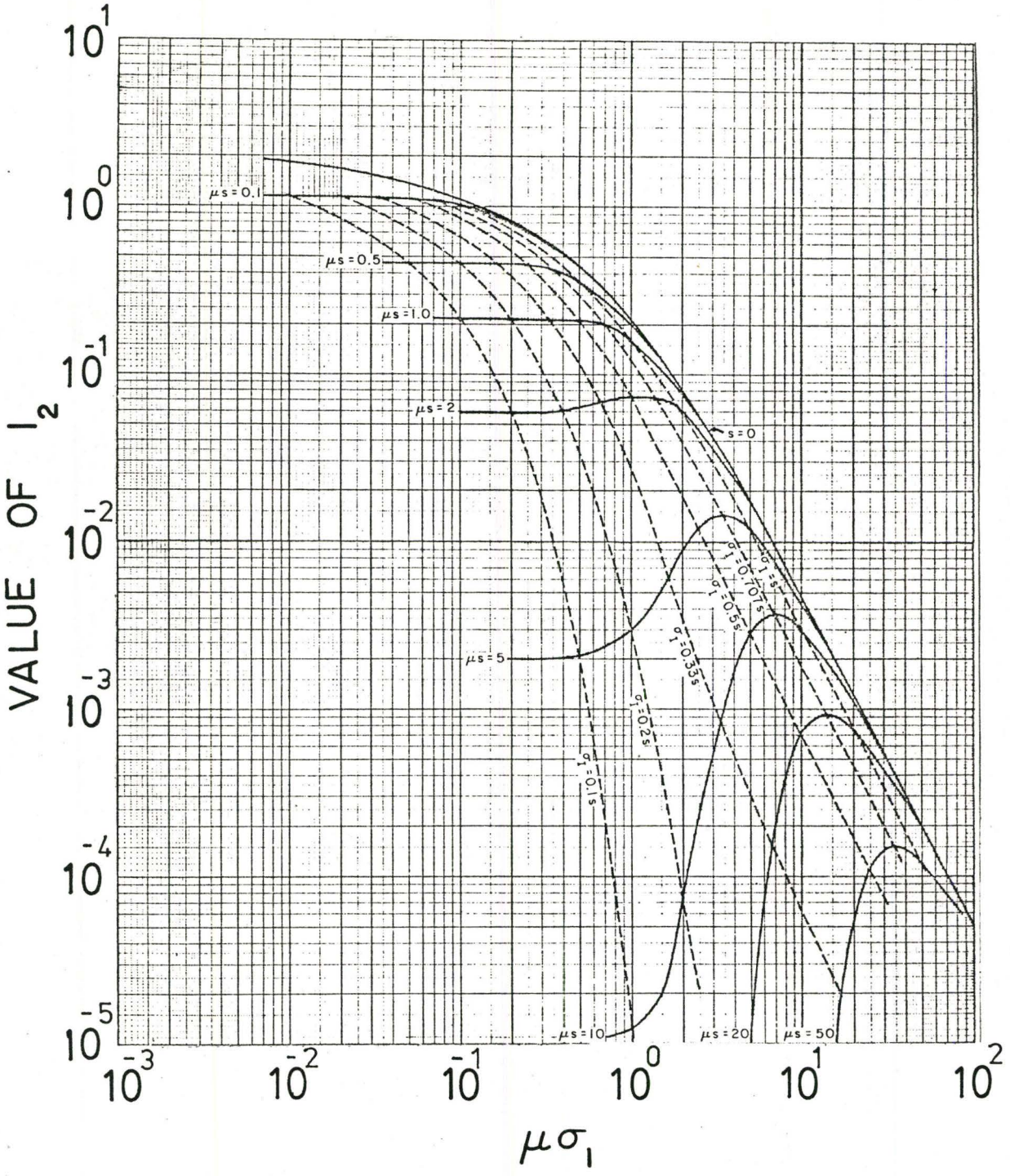

Figure 2. Values of the I_2 integral. Values of σ or σ_I can be used

most reactors.

Thus, to evaluate ${}_{\gamma}D(x,y,0)$, we can first read off f from the appropriate graph and then use equation 33 and then equation 32.

7.2.3 WASH1400 Method

The third approach for finding the gamma dose at a point (x,y,0) is given by (reference 15)

$$ {}_{\gamma}D(x,y,0) = 0.25\bar{E}_{\gamma}Q\psi(x,0,0)F \qquad (49) $$

where ${}_{\gamma}D(x,y,0)$, $\bar{E}_{\gamma}$, Q, $\psi(x,0,0)$, are defined as usual.

The reduction factor F is given in TABLE 5.

This approaches assumes uniform dose along y-direction and is useful only in the estimation of population gamma dose.

7.3 Methodology for Calculating Population Gamma Dose

The procedure of estimating the population gamma dose is very similar to that in the case of iodine intake discussed in section 3.5. The same steps can be followed. However, I would like to point out some differences which must be noted.

First, $\psi(x,y,0)$ has to be corrected with a factor g(x) dependent on the approach adopted. In the infinite cloud approach, $g(x) = 1$. In the finite cloud approach, g(x) has to be read off figures 7.10, 7.11, and 7.14 in reference 11. In the WASH1400 approach, g(x) is $(\psi(x,0,0)/\psi(x,y,0))F(x)$ where F(x) is obtainable in TABLE 5.

Second, unlike in the case of iodine intake, where the dose conversion factor per Ci of activity differs for different

TABLE 5

Correction Factor F for Finite Cloud Dose as listed in WASH-1400

$\Sigma_z(x)$ (m)	Correction Factor, F
$\Sigma_z < 10$	0.1
$10 \leq \Sigma_z < 30$	$0.1 + 0.13 \ell n(\Sigma_z/10)$
$30 \leq \Sigma_z < 300$	$0.24 + 0.33 \ell n(\Sigma_z/30)$
$\Sigma_z \geq 300$	1.0

isotope, the dose conversion factor per Ci of activity depends only on the gamma ray energy given by 0.25 $\bar{E}$.

With these two differences in mind, the steps listed in section 3.5 can be followed closedly.

8 A Brief Review of the program POPDOSE

The estimation of population dose due to iodine intake and gamma irradiation is long winded, especially when the calculation has to be repeated for different boundaries and different approaches of estimating gamma doses. For this reason, I wrote a program code named POPDOSE to handle the computation. I shall very briefly outline what this program can do as the following:

(1) At a distance x in the downwind direction, it determines Σ_y and Σ_z according to equations 26,27,35, and 36 for our choice of stability class and terrain complexity (uniform).

(2) It determines the centre line specific exposure, $\psi(x,0,0)$ according to equation 25 for our choice of effective stack height H and average wind speed $\bar{u}$.

(3) It calculates the y co-ordinate of the plume boundary for our choice of boundary.

(4) It calculates the correction factor f_d at x due to radioactive decay according to equation 39 for every isotope present.

(5) It determines the population density $\rho(x)$ in the strip defined by $(x-\Delta x)$ and x.

(6) Along the y-direction, it calculates the specific exposure ψ for a point $(x, n\Delta y, 0)$ for our choice of Δy according to equation 24.

(7) It compares $\psi(x, m\Delta y, 0)$ with $\psi(x-\Delta x, m\Delta y, 0)$ and determines the higher of the two. This gives the local maximum of ψ for the square with corners at $(x, m\Delta y, 0)$, $(x, (m+1)\Delta y, 0)$, $((x-\Delta x), (m+1)\Delta y, 0)$, and $((x-\Delta x), m\Delta y, 0)$.

(8) It multiplies local maximum of ψ determined in Step 7 by $\rho(x)$ and then by $\Delta x\,\Delta y$ to yield the differential contribution to the population specific exposure.

(9) Step 6 to 8 is repeated for $(x, (m+1)\Delta y, 0)$ and so on until the plume boundary is reached.

(10) It then sums up all the differential contributions to population specific exposure to yield the strip contribution between $(x-\Delta x)$ and x.

(11) It determines the strip contribution according to the WASH1400 approach as described in section 7.2.3 (only if command is given that population gamma specific exposure is to be determined with this approach).

(12) The strip contributions determined in Steps 10 and 11 are corrected for radioactive decay for each isotope we input by multiplying each of the two results for strip contribution with the respective decay correction factor f_d obtained in Step 4.

(13) For each isotope, it adds each of the two decay corrected strip contributions obtained in Step 12 to its previously determined subtotal.

(14) Step 1 to 13 are repeated until our choice of boundary in the x-direction is reached, yielding the half plume total population specific exposure (both approaches) for each isotope.

The program POPDOSE is not so successful in the finite cloud approach for population gamma specific exposure. The reason is that the two integrals I_1 and I_2 in equation 31 determining the ratio of finite cloud dose to infinite cloud dose have to be manually read off the graph. Thus, POPDOSE can be used only up to the point where the local maximum at each mesh point is determined and the correction and summing have to be performed manually.

The program POPDOSE is meant to be a starting point for the possible development of much more sophisticated code in the future. There is plenty of room for improvement. To name a few, a subroutine to determine the correction factor f in the finite cloud approach by interpolation between values available in figures 7.10 and 7.11 in reference 11 for I_1 and I_2 curves in equations 44 and 45. Then, all three approaches for gamma doses can be computerized. Also, the program can be modified to handle non-uniform terrain complexity, non uniform pupulation density in the y-direction, deposition, change of wind direction, etc. However, all these changes are beyond the scope of the present project.

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