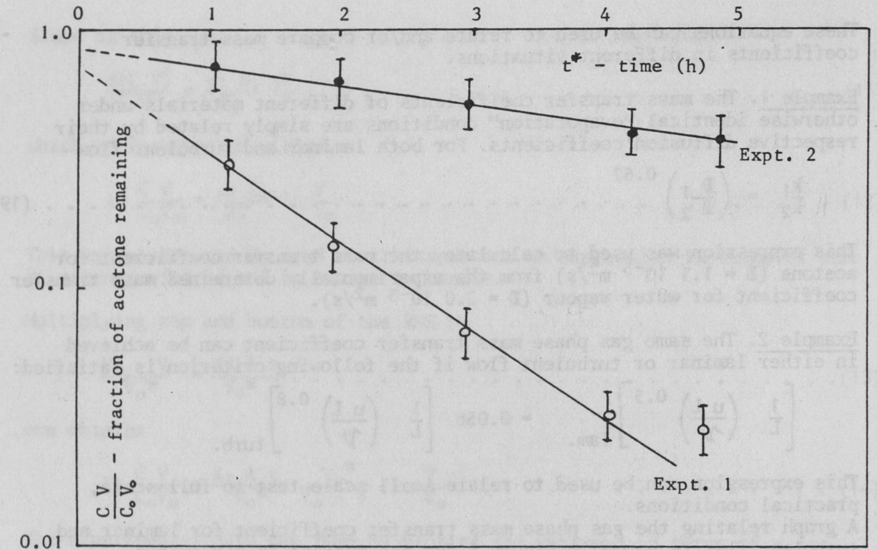

FIGURE 2: Experimental acetone evaporation results.

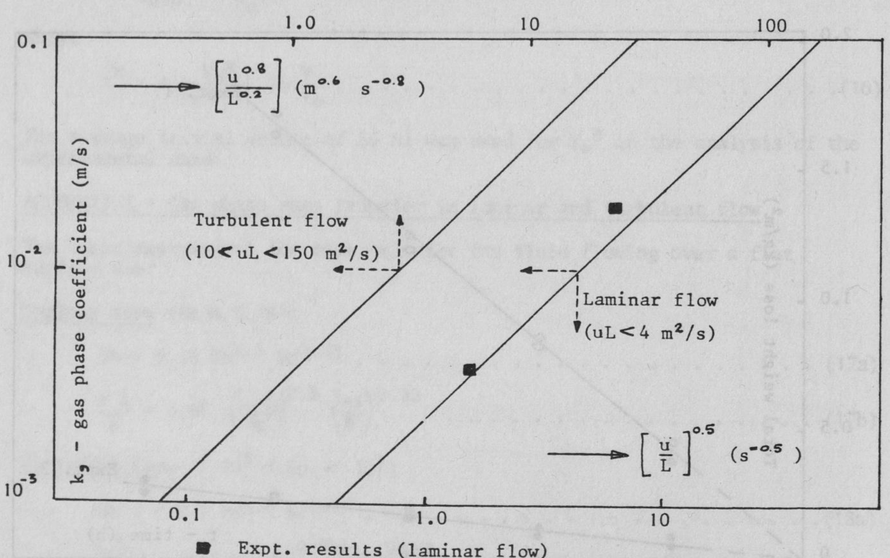

FIGURE 3: Gas phase coefficient (acetone-in-air) for flow over a flat surface

SPREADING AND DISPERSION OF DENSE VAPOURS AND GASES

D. J. Gunn*

The emission of dense vapour at ground level from a vessel or pipe failure spreads by gravity and forms a cloud of dense gas in the neighbourhood. Spreading is controlled by fluid velocities and lateral pressure gradients set up by density difference. Momentum and continuity relations for the cloud are derived from basic equations. The rate of dispersion into the atmosphere and the rate of emission into the cloud are boundary conditions. The description is compared with other models presented in the literature.

INTRODUCTION

The consequences of a major leak of volatile and flammable liquid, or the emission of a flammable vapour in a major hazard plant may be simulated if models of the emission, vapour dispersion and blast propagation are available. The failure of a pipe or vessel is invoked, and the period and rate of leakage are estimated from details of the nature of the failure and the inventory of the vessel or connecting vessels. The intention of the simulation is to examine the consequences of a major loss of containment in which the whole contents of a vessel are expelled, for example.

The concentration of vapour is followed by a dispersion calculation with the object of estimating the extent of the flammable environment as a function of time. If there is a significant possibility of ignition, the consequences of the subsequent explosion and blast may be examined to estimate the extent of plant and neighbourhood damage.

The object of this paper is to consider the importance of velocity, turbulence and other transport processes in the dispersion of vapour into the atmosphere and to examine the effect of density upon dispersion particularly when the density of the emitted vapour is greater than the density of air.

Methods that are available at present for dispersing gas clouds are related to this background. The development of an alternative method is suggested. The discussion is related to the spreading and dispersion of non-buoyant gases.

Transport Processes in the Atmosphere

When an efflux of dense gas takes place at a rate that is large enough to affect the local atmospheric distribution of velocity, a shallow cloud of gas

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is formed around the point of efflux that spreads under gravity and is dispersed into the atmosphere, while the concentration of the dense component is reduced by entrainment of air into the upper surface of the cloud. The cloud is sustained by continued flow of dense gas so that both the rate and duration of the dense gas emission should be known.

The concentration of the dense gas component falls off at least exponentially with horizontal and vertical distance from the cloud. Because of this distribution a flammable environment may be present at some distance, and therefore calculations are required both for the cloud and for its dispersive environment.

As the density of the cloud gas approaches that of air, gravitational spreading of the gas cloud is reduced, and when the rate of emission into the cloud falls off, the cloud thins and contracts. In the final stages of dispersion the boundary contracts as the cloud is dispersed into the atmosphere.

The disturbance of the velocity field near the point of emission may be illustrated by the superposition of a source and uniform flow in an inviscid fluid; the distribution of streamlines in a flow field when the source and flow are steady is shown in Fig. 1. However if the source is that of a gas denser than air the greater density causes an extension of the boundary as gravitational spreading.

Gravitation spreading of a dense fluid under a less dense has been studied, mainly theoretically and an account has been given by Turner (1). From this discussion it may be established from a comparison between fluid pressures at the boundary and under the main fluid that the velocity of spreading is

$$u_s = \sqrt{\left[2\left(\frac{(\rho-\rho_a)}{\rho}\, gH\right)\right]} \qquad (1)$$

where H is the depth of the dense fluid, ρ its density and ρ_a the density of the less dense fluid.

An equation of this type has been used by van Ulden (2) and Cox and Roe(3) in the form

$$\frac{dL}{dt} = \left(kgH\,\frac{\Delta\rho}{\rho_a}\right)^{1/2} \qquad (2)$$

van Ulden compared this equation with the results of some large scale experiments and found good agreement when k was set to 1, and this value was also adopted by Cox and Roe. However it will be noticed that the denominators in equations (1) and (2) differ, and that the form of equation(2) will require k to be less than 2, and equal to $2\rho_a/\rho$.

van Ulden considered edge mixing of the cloud. Cox and Roe, however, found that edge mixing was always small compared to mixing through the top surface. Mixing through the top surface was described by an equation attributed to Ellison and Turner (4), and Thompson (5)

$$u_e = \alpha u_1 (R_i)^{-1}, \qquad R_i = (g\ell/u_1^{\,2})\Delta\rho/\rho_a \qquad (3)$$

where u_e is the entrainment velocity, u_1 is the turbulence velocity and R_i

is the Richardson number. The value of α was estimated from experiments on spills of liquefied natural gas to be 1.5. The value of u_1 was estimated following a suggestion by Monin (6), who considered that the ratio of u_1 to the friction velocity depended mainly on atmospheric stability and suggested the values: 3.0 (very unstable), 2.4 (neutral), 1.6(very stable). The ratio of the friction velocity to wind velocity, a function of the roughness of the terrain was estimated from relationships provided by Sutton (7) and the scale of turbulence ℓ, a function of height and stability, was taken from the results of Taylor et al (8).

The cloud was divided into slices normal to the wind velocity vector and gravitational spreading entrainment and adiabatic mixing were obtained by integrating the ordinary differential equations in time for each cloud slice. The cloud slices were assumed to advect with wind velocity and direction.

A quite different model was suggested by te Riele (9) at the same symposium. He considered dispersion from a homogeneous rectangular area into the atmosphere, with concentration distribution described as a similarity profile of the Gaussian type. However, the Gaussian profile was modified to take into account a power law dependence of velocity upon height with the exponents in the similarity profile and the crosswind dispersion coefficient as functions of the velocity profile. Spreading of the gas was determined by the relationship between takeup of gas in the atmosphere and the rate of gas emission; the extent of the heavy gas layer increased until the rate of dispersion into the atmosphere was equal to the rate of gas emission.

Even without a consideration of the physical accuracy of the individual features it is clear that the emphasis on the separate parts of the dispersion process is quite different in the two models. That of van Ulden,and Cox and Roe concentrates on the gravitational spreading of the gas cloud, with an estimation of the rate of entrainment, but without calculating the atmospheric concentration field; the extent of the atmospheric concentration field is estimated in the model of te Riele, but the spreading of the cloud is not related to the fluid-dynamic equation of motion.

Van Ulden, Cox and Roe, and te Riele all tested their different theories against sets of experimental results and all found satisfactory agreement. The agreement means that the major differences between the models are not reflected in major differences in predictions, probably because some of the parameters of the theories have been found from similar experiments so that parameters have been adjusted to give the observed experimental response. In the next section of the paper we describe the gravitational and dispersive characteristics of a heavy gas emission, and there will be occasion for further comment on the models.

Gravitational Spreading and Dispersion of Dense Gas

We consider the emission of a dense gas at ground level. The momentum equations for the x and y directions are written for a flow in which momentum changes in the vertical direction may be neglected in comparison with momentum changes in the other directions. Fig. 2 illustrates such a flow in which ground level is $z = h_1(x,y)$ with respect to the datum $z = 0$, while the upper surface of the cloud is $z = h(x,y,t)$. At a particular point x,y, the equation of motion in the z direction for such a flow reduces to

$$\frac{\partial p}{\partial z} = \rho g \qquad (4)$$

The equations of motion in the x and y directions reflect the change of pressure at the upper surface of the cloud due to the change of h with x and y. The equations are

$$\rho \frac{\partial u}{\partial t} + \rho u \frac{\partial u}{\partial x} + \rho v \frac{\partial u}{\partial y} + \rho w \frac{\partial u}{\partial z} + (\rho-\rho_a) g \frac{\partial h}{\partial x} + \frac{\partial}{\partial z} \tau_x = 0 \tag{5}$$

$$\rho \frac{\partial v}{\partial t} + \rho v \frac{\partial v}{\partial x} + \rho v \frac{\partial v}{\partial y} + \rho w \frac{\partial v}{\partial z} + (\rho-\rho_a) g \frac{\partial h}{\partial y} + \frac{\partial}{\partial z} \tau_y = 0 \tag{6}$$

while the equation of continuity neglecting rapid changes of density with time, is

$$\frac{\partial \rho u}{\partial x} + \frac{\partial \rho v}{\partial y} + \frac{\partial \rho w}{\partial z} = 0 \tag{7}$$

The equations are now integrated with respect to z over the depth of the cloud. Thus equation (5) becomes

$$\int_{h_1}^{h} \frac{\rho \partial u}{\partial t} dz + \int_{h_1}^{h} \frac{\partial \rho u^2}{\partial x} dz + \int_{h}^{h} \frac{\partial}{\partial y} \rho uvdz + \int_{h_1}^{h} \frac{\partial}{\partial z} \rho uwdz + (\rho-\rho_a) g(h_1-h) \frac{\partial h}{\partial x} + \tau_x(h) - \tau_x(h_1) = 0 \tag{8}$$

where equation (7) has been multiplied by u and added to (5). The first term may be rearranged:

$$\int_{h_1}^{h} \rho \frac{\partial u}{\partial t} dz = \underset{\delta t \to 0}{\mathrm{Lt}} \left[\left(\int_{h_1(t+\delta t)}^{h(t+\delta t)} \rho u(t+\delta t) dz - \int_{h_1(t)}^{h(t)} \rho u(t) dz \right) \Big/ \delta t - \frac{1}{\delta t} \int_{h(t)}^{h(t+\delta t)} \rho u(t+\delta t) dz + \frac{1}{\delta t} \int_{h_1(t)}^{h_1(t+\delta t)} \rho u(t+\delta t) dz \right] \tag{9}$$

and therefore

$$\int_{h_1}^{h} \rho \frac{\partial u}{\partial t} dz = \rho \frac{\partial}{\partial t} \int_{h_1}^{h} u \, dz - \rho uw(h) + \rho uw(h_1) \tag{10}$$

Since

$$h = h(x,y,t) \tag{11}$$

so that

$$\frac{dh}{dt} = u \frac{\partial h}{\partial x} + v \frac{\partial h}{\partial y} + \frac{\partial h}{\partial t} = w(h) \tag{12}$$

Similarly

$$\frac{dh_1}{dt} = u \frac{\partial h_1}{\partial x} + v \frac{\partial h_1}{\partial z} = w(h_1) \tag{13}$$

With the substitutions set out above the momentum equations and the equation of continuity become,

$$\rho \frac{\partial}{\partial t} \int_{h_1}^{h} udz + \frac{\partial}{\partial x} \int_{h_1}^{h} \rho u^2 dz + \frac{\partial}{\partial y} \int_{h_1}^{h} \rho uvdz + (\rho-\rho_a) g(h-h_1) \frac{\partial h}{\partial x} + \tau_x(h) - \tau_x(h_1) = 0 \tag{14}$$

$$\rho \frac{\partial}{\partial t} \int_{h_1}^{h} vdz + \frac{\partial}{\partial x} \int_{h_1}^{h} \rho uvdz \quad \frac{\partial}{\partial y} \int_{h_1}^{h} \rho uv^2 dz + (\rho-\rho_a) g(h-h_1) \frac{\partial h}{\partial y} + \tau_y(h) - \tau_y(h_1) = 0 \tag{15}$$

$$\rho \frac{\partial h}{\partial t} + \frac{\partial}{\partial x} \int_{h_1}^{h} \rho udz + \frac{\partial}{\partial y} \int_{h_1}^{h} \rho vdz = 0 \tag{16}$$

Equations (14) to (16) give the momentum and continuity relationships for the cloud specified as a fluid of density ρ without other specification of composition. The emission of dense gas into the cloud will add a source term on the right-hand side of (16), while the loss of dense gas by entrainment into the atmosphere will have an effect that is less certain if air is entrained into the cloud.

If air is not entrained into the cloud the required form of equation (16) is

$$\rho \frac{\partial h}{\partial t} + \frac{\partial}{\partial x} \int_{h_1}^{h} \rho udz + \frac{\partial}{\partial y} \int_{h_i}^{h} \rho vdz = q_1(x,y) - q(x,y) \tag{17}$$

where $q_1(x,y)$ is the rate of gas emission per unit area into the base of the cloud, while $q(x,y)$ is the rate of dispersion into the atmosphere.

If air is entrained into the cloud, the volume of the cloud is determined by the net effect of entrainment of air less the atmospheric loss of the dense component. Some entrainment of air takes place, but there is very little evidence of the relative magnitudes of the rate of cloud entrainment into the atmosphere and the rate of air entrainment into the cloud. If they are of equal magnitude, but opposite in direction, the continuity equation for the cloud is

$$\rho \frac{\partial h}{\partial t} + \frac{\partial}{\partial x} \int_{h_1}^{h} \rho udz + \frac{\partial}{\partial y} \int_{h_1}^{h} \rho vdz = q_1(x,y) \tag{18}$$

and the continuity equation for the dense component is

$$c \frac{\partial h}{\partial t} + \frac{\partial}{\partial x} \int_{h_1}^{h} ucdz + \frac{\partial}{\partial y} \int_{h_1}^{h} vcdz = q_1(x,y) - q(x,y) \tag{19}$$

where c is the concentration of dense component. If the mean quantities are now introduced,

$$U = \frac{1}{h-h_1}\int_{h_1}^{h} u\,dz, \quad \rho U = \frac{1}{h-h_1}\int_{h_1}^{h} \rho U\,dz, \text{ etc. the continuity equations become,}$$

for the cloud

$$\rho \frac{\partial h}{\partial t} + \frac{\partial}{\partial x}(\rho UH) + \frac{\partial}{\partial y}(\rho VH) = q_1(x,y) \tag{20}$$

for the dense component

$$C \frac{\partial h}{\partial t} + \frac{\partial}{\partial x}(UCH) + \frac{\partial}{\partial y}(VCH) = q_1(x,y) - q(x,y) \tag{21}$$

while the equations of momentum in terms of mean quantities are

$$\frac{\partial U}{\partial t} + U\frac{\partial U}{\partial x} + V\frac{\partial U}{\partial y} + \left(\frac{\rho-\rho_a}{\rho}\right) g \frac{\partial h}{\partial x} + \frac{1}{H}\left(\tau_x(h) - \tau_x(h_1)\right) = 0 \tag{22}$$

$$\frac{\partial V}{\partial t} + U\frac{\partial V}{\partial x} + V\frac{\partial V}{\partial y} + \left(\frac{\rho-\rho_a}{\rho}\right) g \frac{\partial h}{\partial y} + \frac{1}{H}\left(\tau_y(h) - \tau_y(h_1)\right) = 0 \tag{23}$$

Continuity equations (20) and (21), and momentum equations (22) and (23) define the change of cloud shape, velocity and concentration with time from an appropriate initial condition. It is assumed that the source term $q_1(x,y)$ and the sink term $q(x,y)$ do not cause significant gain or loss of momentum. The rate of emission from the source into the cloud $q_1(x,y)$ is the rate of vapour flow into the cloud from a vapour leak, or the rate of vaporisation of liquid. For liquids an additional thermal balance giving the rate of heat gain from earth and atmosphere will be required.

The rate of loss of the dense component $q(x_1y)$ into the atmosphere is determined by conditions of atmospheric stability, turbulence, wind velocity and the extent of the cloud.

The continuity and momentum equations, when integrated, will give the extent of the cloud including the effect of changes in terrain and buildings and other erected structures.

Dispersion into the atmosphere

Because of the change of air density with temperature the vertical temperature gradient has a very important effect upon conditions for atmospheric dispersion. If temperature increases with distance from the ground, the upper air is less dense than the lower giving stable conditions, but if temperature decreases with height the upper air is more dense, and such unstable conditions are accompanied by erratic air movements. There is now general recognition that atmospheric stability as well as vertical velocity gradient, terrain roughness and turbulence intensity affect dispersion from a source into the atmosphere(10).

The description of dispersion may be based upon similarity theory, in which composition profiles, for example, are assumed to be similar, by statistical theory, or by gradient transfer theory in which the rate of dispersion is taken to be proportional to concentration gradient with the eddy

diffusivity as the constant of proportionality. It is interesting to note that the average eddy diffusivity may vary by a factor of 100 between moderately stable and moderately unstable conditions.

The form of concentration dependence in the atmosphere upon distance may be expressed for a continuous point source (10)

$$C(x,y,z) = A \exp(-(b|y|^r + d|z|^s))$$

where the parameters b and d depend upon the dimensions of the dispersing cloud, A is a function of b and d and the exponents r and s lie between 1 and 2. The parameters and exponents reflect atmospheric conditions, surface roughness and the vertical wind velocity profile and are obtained by fitting the formulas to describe experimental data.

For dispersion from a gas cloud it might be expected that short range dispersion up to about 1 km is more likely to be of importance than longer range. Pasquill (10) has recommended that formulas below for the estimation of concentration at short range from a ground level continuous point source in neutral conditions.

$$C(x,y,z) = \frac{0.67Q'}{\pi^{\frac{1}{2}}\,\bar{u}\sigma_y\sigma_z} \exp\left[\frac{-y^2}{2\sigma_y^2} - \left(\frac{z}{1.16\sigma_z}\right)^{1.5}\right] \tag{24}$$

where Q' is the amount emitted in init time, x is the downwind co-ordinate, y is the horizontal crosswind coordinate and x is the vertical coordinate. T is the time since the start of emission.

$$\sigma_y = \sigma_\theta x \tag{25}$$

$$\sigma_z = 0.208x\left[\ln\left(\frac{0.6\sigma_z}{1.3z_o}\right) - 1\right] = \sigma_\gamma x \tag{26}$$

Here σ_θ is the standard deviation of wind direction in radians and z_o is the roughness parameter.

There is an immediate difficulty in applying these formulas to dispersion from the cloud in that the rate of emission Q is required by the formula, but not provided by the dispersing cloud; the cloud concentration only is given.

We may retain the form of equation, provided that the relationship between the rate of emission and the surface concentration can be established. For this purpose we consider a rectangular area extending from X to X + ΔX and Y to Y + ΔY. If the rate of emission Q' is characteristic of this area and Q' is set to QΔXΔY where Q is the rate of emission from unit area of surface eqn.(2) may be rearranged:

$$Q = \frac{1.5\sqrt{T}\,\bar{u}\sigma_y\sigma_z}{\Delta Y \Delta X} \exp\left(\frac{+y^2}{2\sigma_y^2} + \left(\frac{z}{1.16\sigma_z}\right)^{1.5}\right) c(x,y,z) \tag{27}$$

where x, y and z are measured from an origin within the rectangle.

According to equation (25) σ_y is proportional to x, and σ_z increases almost linearly with x, and within the element the distance of down wind dispersion of x is ΔX at maximum. Thus within limitation of representing an area source by a point source Q may be taken as

$$Q = 1.5\sqrt{\pi}\,\bar{u}\,\sigma_\theta \sigma_\gamma c(0) \tag{28}$$

where c(O) is the cloud concentration $\bar{u}$ is the average wind speed, σ_θ and σ_γ are coefficients that depend upon atmospheric conditions (10). For a given cloud concentration the rate of atmospheric dispersion is proportional to the mean wind velocity close to the cloud surface, and will also increase with, for example, intensity of turbulence and roughness of the terrain.

Equation (28) now furnishes the upper boundary condition for the cloud of dense gas given as a proportionality between the rate of loss and cloud concentration. This boundary condition together wtih the surface conditions of emission into the cloud will now allow integration of the momentum and continuity equations for the cloud over the terrain, so that the growth and decay of the cloud may be followed.

The concentration field may be found by considering the concentration at (x,y,z) due to the point source is

$$dC(x,y,z) = \frac{0.67Q\,dx'dy'}{\pi^{\frac{1}{2}} u\sigma_\theta(x-x')\sigma_z[x-x']} \exp\left(\frac{(-u-y')^2}{2\sigma_\theta^2(x-x')^2} - \left\{\frac{z-z'}{1.16\sigma_z[x-x']}\right\}^{1.5}\right) \tag{29}$$

where $\sigma_z[x-x']$ indicates that (x-x') replaces x in equation (26). The concentration at (x,y,z) due to the whole of the cloud is then given by

$$C(x,y,z) = \int_{Y_1}^{Y_2}\int_{X}^{x} \frac{0.67Q}{\pi^{\frac{1}{2}}\bar{u}\sigma_\theta(x-x')\sigma_z[x-x']} \exp\left[\frac{(-u-y')^2}{2\sigma_\theta^2(x-x')^2} - \left(\frac{z-z'}{1.16\sigma_z[x-x']}\right)^{1.5}\right] dx'dy' \tag{30}$$

with $(x-X) < \bar{u}T$

Note that the integration within the inner integral is taken only as far as x since formula (24) shows that sources down wind of x do not contribute to the concentration at x. The limiting condition on the lower limit of the inner integral is that the time interval since the start of emission has to be large enough to allow wind to carry the vapour component to x.

The effect of obstacles on the terrain

The analysis of the spreading of the dense cloud leading to equations (20) to (23) will accommodate the cloud to the terrain since the field of flow will exclude obstacles such as buildings etc. because there is no gas cloud when $h_1 > h$. When the obstacles are sufficiently high to exclude the dispersion of vapour from these regions an extension of the method of images will allow vapour to be excluded. Sources are distributed over the real cloud and also over an image cloud formed by creating the image of the cloud in the surface excluding the vapour. Figure 3 illustrates sources due to the cloud and image. In considering the concentration at a point on the allowed side of the barrier, the integration is taken over both real and image sources. This will have the effect of satisfying the condition $\partial c/\partial x = 0$ at the barrier.

The image of the cloud in a more complicated excluded region will increase

the complexity of the analysis, and it may become intractable. Such an increase in complexity is clearly not justified in view of other uncertainties. However, if obstacles to vapour dispersion are present in the field the practice of distributing image sources over the excluded region will in a measure satisfy the boundary condition of vapour exclusion.

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SYMBOLS

a,b	Constants
c	concentration dense component, kg/m^3
C	concentration integration over z in the cloud
d	constant
g	acceleration due to gravity, N
h	height of gas cloud relative to datum, m
h_1	height of terrain relative to datum, m
H^1	$(h-h_1)$
K	constant in eqn. (2)
ℓ	turbulence scale
L	position of interface, m
p	pressure, N/m^2
q	rate of dense gas emission into the base of cloud, $kg/(m^2s)$
q_1	rate of dense gas loss to atmosphere, kg/m^2s
R_i	Richardson number defined by eqn. (3)
t	time, s
u,v	velocities, m/s
$\bar{u}$	average wind velocity, m/s
u_1	turbulence velocity, m/s
u_e	entrainment velocity, m/s

U,V	velocities integrated over z in the cloud
x,y,z	coordinates, m

Greek

α	constant in eqn. (3)
ρ	dense gas density, kg/m^3
ρ_a	air density,
σ	variance
$\sigma_x,\sigma_y,\sigma_z$	variance in x,y,z directions
$\sigma_\theta,\sigma_\gamma$	variance defined by eqns.(25), (26)

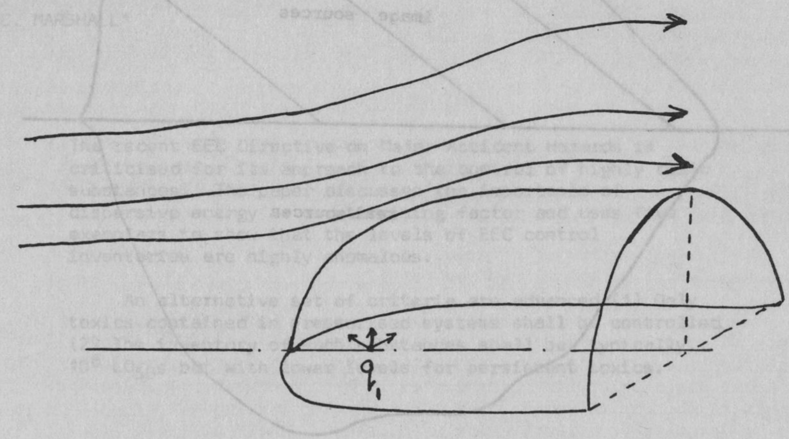

Figure 1 Disturbance of wind flow due to neutral source.

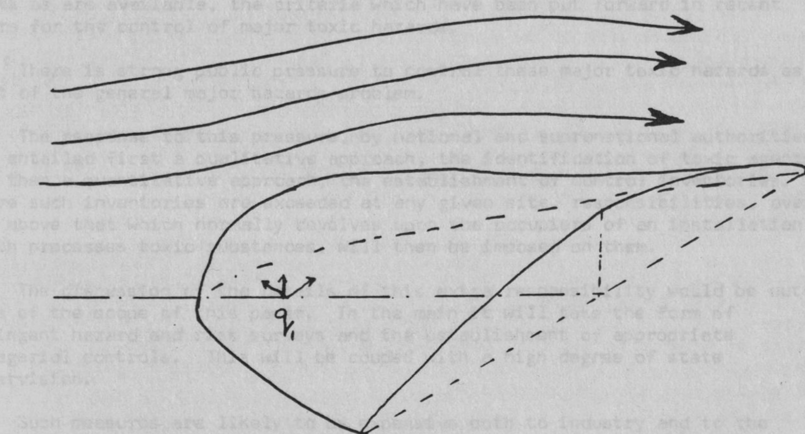

Figure 2 Gravitational spreading of dense cloud.

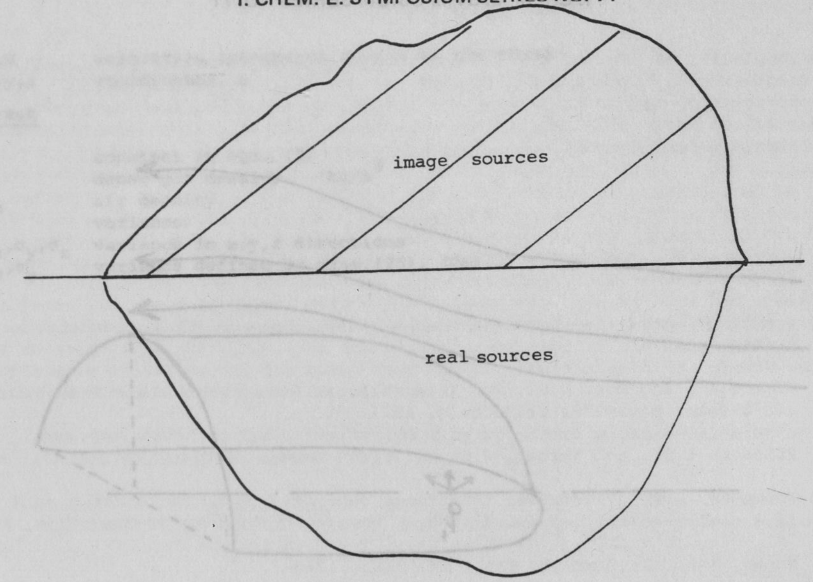

Figure 3 Real and Image sources in cloud

QUANTIFICATION AS A MEANS OF CONTROL OF TOXIC HAZARDS

V. C. MARSHALL*

The recent EEC Directive on Major Accident Hazards is criticised for its approach to the control of highly toxic substances. The paper discusses the importance of dispersive energy as a determining factor and uses five exemplars to show that the levels of EEC control inventories are highly anomalous.

An alternative set of criteria are advanced (1) Only toxics contained in pressurised systems shall be controlled (2) The inventory of such substances shall be, typically, 10^8 LD_{50}s but with lower levels for persistent toxics.

THE PURPOSE OF THE PAPER

The purpose of the paper is to examine, in the light of such quantitative tests as are available, the criteria which have been put forward in recent years for the control of major toxic hazards.

There is strong public pressure to control these major toxic hazards as part of the general major hazards problem.

The response to this pressure, by national and supranational authorities, has entailed first a qualitative approach, the identification of toxic agents, and then a quantitative approach, the establishment of control inventories. Where such inventories are exceeded at any given site, responsibilities, over and above that which normally devolves upon the occupiers of an installation which processes toxic substances, will then be imposed on them.

The discussion of the details of this extra responsibility would be outside of the scope of this paper. In the main it will take the form of stringent hazard and risk surveys and the establishment of appropriate managerial controls. This will be coupled with a high degree of state supervision.

Such measures are likely to be expensive both to industry and to the state and should be implemented only in situations of true major hazard. Reducing the level of the control inventory will increase the cost of the exercise and the law of diminishing returns will apply. To treat every chemical works as a major hazard would be to dissipate the resources which ought to be devoted to really serious problems.

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