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SMR/760-49

**"College on Atmospheric Boundary Layer  
and Air Pollution Modelling"  
16 May - 3 June 1994**

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Dispersion II  
"Atmospheric Dispersion of Dense Gases"

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## DISPERSION II

The Atmospheric Dispersion of Dense Gases

# ATMOSPHERIC DISPERSION OF DENSE GASES

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## 1. INTRODUCTION

Public concern over the risks posed by the use of hazardous materials has grown markedly over the past decade. The dioxin release in Seveso (Italy) in 1976, that of methyl isocyanate in Bhopal (India) in 1984, and the liquefied petroleum gas explosions in Mexico City in the same year emphasized the possible scale of the tragedies that may accompany activities involving hazardous materials.

The development of appropriate regulatory measures to achieve an acceptable balance between economic benefit and potential harm accompanying such activities requires quantitative assessment of the consequences of the accidental release of material into the environment.

It is commonly the case that hazardous industrial materials, be they flammable or toxic, produce a cloud, upon release into the atmosphere, that is denser than the environment.

The information on dense-gas dispersion that is of interest to the hazards analyst is contained in the distribution of concentration as a function of the spatial coordinates and time. Very often, this information is required only in summary form, such as

1. the distance to a given concentration (for example, the lower flammability limit);
2. the size, composition, and shape of the cloud. These are needed for thermal radiation estimates in the event of burning or as input to methods of estimating explosion propagation;
3. the mass of gas in the cloud between the upper and lower flammability

- limits. This is often regarded as the appropriate mass to be used in estimating the TNT-equivalence of a flammable cloud; and
4. the concentration and its time history at a given distance, needed to define toxic effects on human and nonhuman biota.

The relatively recent research on dense—that is negatively buoyant—gas dispersion (in the open literature) should be contrasted with the far more extensive and detailed study of the dispersion of neutrally and positively buoyant pollutants.

The quantitative assessment of the dispersion of dense gases is quite different to conventional dispersion problems for the following reasons:

1. unlike covenanted chimney emissions, the modes of release are very diverse in terms of geometry and source specification;
2. because the released material is typically stored in a liquid phase, the volumes of gas released may be very large;
3. the release may be a gas/liquid mixture;
4. the release is usually transient;
5. the formation of the gas cloud typically involves phase changes; and
6. there may be heat and/or mass transfer with the underlying surface.

In addition, the dispersing gas forms a low-level cloud that is sensitive to the effects of both man-made and natural obstructions and topography.

These complications indicate that the task of predicting the consequences of an accident will not be simple. Further, the rapid development of the field has restricted specific study of the various, relevant fluid-mechanical phenomena.

Nevertheless, very many (of order 100) models for dense-gas dispersion have been developed over the past decade. Few, if any, of these have been subject to a formal model validation. This must incorporate a satisfactory scientific understanding of the flow or phenomenon that is being modeled. Validation is typically restricted to a limited comparison of a few concentrations, a technique that, as we shall see, is rather inadequate. Of course, the accuracy required in an operational dispersion model should be comparable to that of the other aspects of a complete hazards appraisal, which in the case of human toxicology, for example, may be quite poor. This said, many papers show an unwarranted confidence in the relevance or validity of the model used.

This review addresses the fluid mechanics of the atmospheric dispersion of dense gases, rather than assessing the validity of the various models purportedly describing this phenomenon. As a consequence, there is little to be gained from a comprehensive, historical development of the subject. Instead, a limited review is offered that reflects the current state of understanding. Much of the source material resides in unpublished or internal

reports or in the very numerous conference proceedings on this subject. Where possible, I have preferred to quote from articles in refereed journals; however, these articles will refer the reader to the more extensive source material.

## 2. FORMATION OF DENSE-GAS CLOUDS

The density of the cloud<sup>1</sup> results not only from the properties of the material released, but also from the methods of storage and of release. Most cases of interest are covered by the following broad categories:

1. materials with a high molecular weight compared with that of air (e.g. chlorine);
2. materials with low molecular weight that may be at a low temperature [e.g. cold methane evolving from the boiling of refrigerated liquefied natural gas (LNG) following a spill onto a warmer surface];
3. materials with low molecular weight and whose vapor at the boiling temperature is less dense than the environment, but which, as a result of the release type, produce a cloud including material droplets. The cloud-borne droplets increase the cloud density, as does the cooling resulting from their subsequent evaporation [e.g. ammonia (see Griffiths & Kaiser 1982); and
4. materials in which a chemical transformation takes place as a result of reaction with water vapor in the ambient atmosphere [e.g. nitrogen tetroxide ( $N_2O_4$ ), hydrogen fluoride (HF) (see Clough et al. 1987)].

Some typical storage arrangements and conceivable release mechanisms are shown in Figure 1 (taken from Fryer & Kaiser 1979). The releases may be categorized as coming from either pressurized or nonpressurized (typically refrigerated) storage conditions. Releases that result from a catastrophic vessel failure are idealized as instantaneous, whereas those from a pipe break would be continuous.

The source specification for pressurized releases requires the consideration of the amount of liquid carried into the cloud, and the amount of air entrained into the cloud, as a result of the release type. This is relatively straightforward for a single-phase gaseous release (Ooms 1972), although even here the plume interaction with the underlying surface is not treated. For liquid or two-phase releases there is less agreement on source-term models (Hanna & Drivas 1987). Released liquid may also fall to the underlying surface and boil or evaporate over a prolonged period.

The evolution of a dense-gas cloud arising from the release of a fully refrigerated, nonpressurized liquid is determined by heat transfer to the

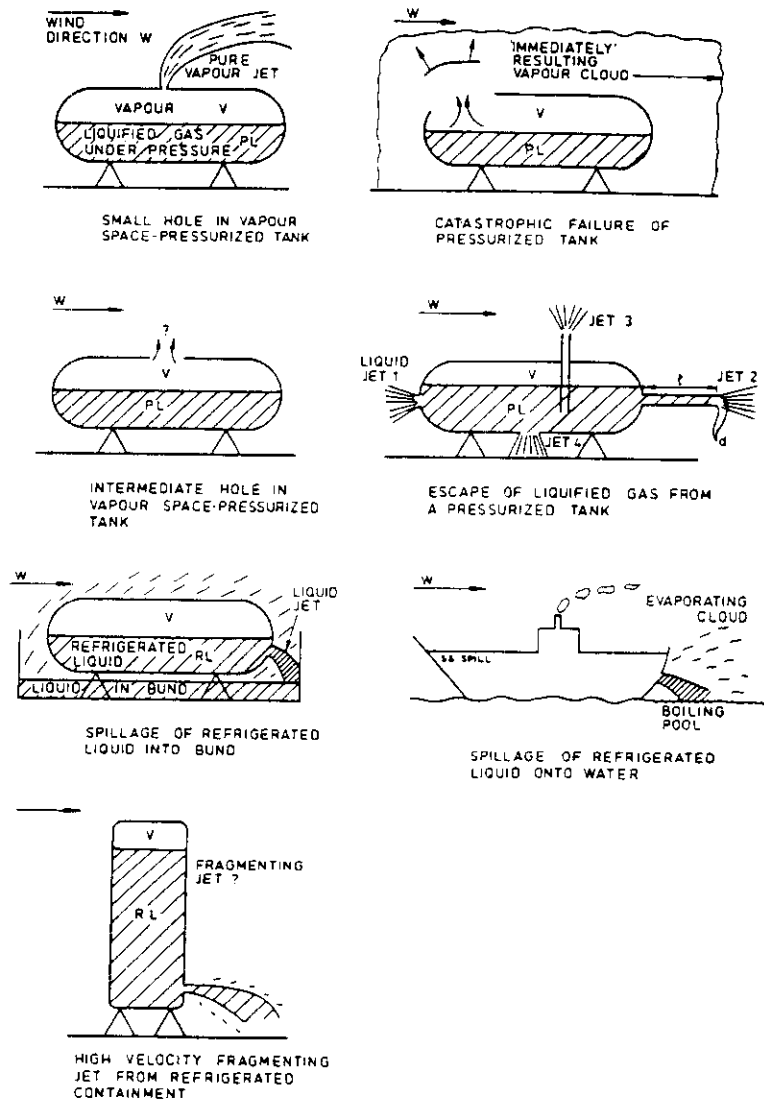


Figure 1 Some conceivable release mechanisms (reprinted with permission from Fryer & Kaiser 1979).

<sup>1</sup> Cloud is used here as a general descriptive term; plume refers to a continuous release, whereas puff refers to an instantaneous release.

liquid. Liquid release into a banded region will initially produce a large source flow that will decay with time as heat transfer from the underlying surface decreases. In the absence of a bund or other liquid-constraining structure, the liquid will spread over an increasing area, producing a source flow rate of gas that increases with time until most of the liquid has boiled off.

For clarity the following discussion only addresses the atmospheric dispersion of an isothermal dense gas from a well-defined source.

### 3. PHYSICAL PROCESSES IN DENSE-GAS DISPERSION

The density difference between the released material and its environment introduces four major effects with regard to dispersion problems.

The velocity field produced by the horizontal density difference, in a gravitational field, is an additional transport mechanism to that provided by the ambient flow. This self-generated flow produces a cloud with an increased horizontal, and reduced vertical, extent when compared with a similar release having no density difference. In addition, the self-generated component of the motion is predominantly deterministic, not random; as a result, profiles of concentration in the lateral direction are frequently quite uniform.

The velocity shear introduced by this velocity field may lead to a gross intermingling of the two fluids and eventually to turbulence generation and consequential turbulent mixing and cloud dilution. This mechanism of dilution is of primary importance when the self-generated velocities are large compared with the mean environmental velocity. In addition, turbulence generated from this flow near rigid boundaries provides a mechanism for cloud dilution.

These processes also appear in the context of thermal discharges (Harleman & Stolzenbach 1972).

Frequently it is the ambient turbulence that is responsible for cloud dilution, be it locally generated or advected from upstream. The variation of density in the vertical direction will, in a gravitational field, be stably stratified, and turbulence and turbulent mixing can be significantly reduced or entirely inhibited (Turner 1973). This effect can extend to the atmospheric turbulence in the windflow over the cloud, as well as to the cloud itself. This fundamental problem, concerning the growth of a dynamically active internal boundary layer, has not been fully addressed within the context of the dispersion of dense gases (though see Brighton 1988).

The inertia of the released material is directly dependent upon the density of the material. However, when the density difference is small compared

with either density, the influence of the *density difference* on the inertia is small and may be neglected. This may not be valid close to the source, but cloud dilution will eventually allow this assumption. Under these conditions, the density difference frequently appears as  $g' = g[(\rho - \rho_a)/\rho_a]$ , where  $g$  is the acceleration due to gravity, and  $\rho$  and  $\rho_a$  are the density of the cloud and of the ambient fluid, respectively.

These effects emphasize the difference between dense-gas dispersion and the dispersion of nondense, or "passive," pollutants. It is obvious that the density difference is not the sole variable determining whether the release behaves as a dense gas. A very small release or release rate into a strong wind, or alternatively a release over a large source area, may be considered effectively passive. A continuous source of volume flow rate  $q_0$ , with source density difference characterized by  $g'_0$  may be considered effectively passive when

$$(g'_0 q_0 / D)^{1/3} / U \leq 0.15,$$

where  $D$  is the source dimension and  $U$  is the ambient velocity (Britter & McQuaid 1988).

For an instantaneous source of release volume  $Q_0$  and implied source dimension  $Q_0^{1/3}$ , the criterion becomes

$$(g'_0 Q_0^{1/3})^{1/2} / U \leq 0.2.$$

The form of these criteria emphasizes the importance of the ambient velocity in describing the flow. In the latter criterion, a halving of the wind speed is equivalent to a 64-fold increase in  $Q_0$ .

### 4. CONTINUOUS RELEASES

Continuous releases from ground-level sources or near-ground-level sources have been the subject of specific study. Britter & McQuaid (1988) have summarized available data from laboratory experiments using idealized area sources with low momentum. More complicated source configurations of dense fluids will eventually lead to a dense plume at ground level to which the idealized source experiments may be relevant (Meroney 1982).

Field experiments have also considered more realistic, less idealized sources, such as liquefied-gas spills on water and the (horizontal) momentum jets of single- and two-phase fluid resulting from the opening of a pressurized vessel (Puttock et al. 1982, Koopman et al. 1988).

Observations in the laboratory and in the field show a wide, flat plume downwind of the source. This is clearly demonstrated by the discharge of

liquefied natural gas shown in Figure 2. For large release rates, low ambient wind speeds or large density differences, the plume extends upwind of the source and can be wider than the physical source size at source position.

For a release in calm conditions, Britter (1979) was unable to obtain a similarity solution but found an approximate solution for the plume radius, namely

$$r \propto (g'_0 q_0)^{1/4} t^{3/4}$$

with a coefficient of order unity. This solution was consistent with experimental observations. Grundy & Rottman (1980) found that there was no similarity solution for this problem in which the source flow varied with  $t^\alpha$ ,  $\alpha \neq 0$ . Some loss (apart from the gravity-current head) is apparent in the solution, and this presumably manifests itself as a breakdown of the simple solution has been questioned, as has the interpretation of the experimental results (Ivey & Blake 1985). Little mixing between the spreading gravity current and environment is observed in these laboratory experiments.

An imposed ambient flow will limit upwind spread of all the source material is eventually carried downwind, in which this flow reversal is attained is uncertain. For a flow reversal may occur solely through an inertial, mixing process.

and ensure that the mechanism by which uniform flow, the mixing process.



Figure 2 A wide, shallow plume resulting from the discharge of liquefied natural gas (courtesy of UK Health and Safety Executive).



Figure 3 A narrow, deep plume resulting from the discharge of liquefied natural gas (courtesy of UK Health and Safety Executive).

This and similar flows are frequently studied in the context of positively buoyant discharges released near a free surface.

However, the flow interaction is less clear when the ambient flow is a turbulent boundary layer. Simpson & Britter (1980) studied a similar two-dimensional problem and were able to determine what magnitude of external flow was able to halt a gravity-current head. It appeared that the flow interaction was unrelated to mixing between the fluids (except in a secondary way). In Simpson & Britter's experiments the boundary-layer height was small or similar to the height of the gravity-current head. A more uniform velocity shear over the complete depth of the gravity-current head may lead to a collapse of the gravity-current head and the formation of a "salt-wedge"-type flow in which the interfacial stress (presumably resulting from small-scale interfacial mixing) is a significant process.

For a gravity current spreading under a turbulent flow, the near-source region may involve both an inertial interaction and a scouring or detrainment of the fluid near the source by the ambient flow. Observation of laboratory experiments would favor the latter of these mechanisms.

Thus, one or both of these mechanisms allow the plume to travel upwind and laterally at the source position prior to being advected downwind.

Various attempts have been made to correlate the near-source flow. Britter (1980) argued that the plume upstream extent was  $2L_b$  and that the cross-stream extent  $L_{H0}$  was  $8L_b$ , where  $L_b = (g'_0 q_0)/U^3$ . There was, however, substantial uncertainty as to what the reference velocity  $U$  should be. Meroney (1982) obtained coefficients of 2.5 and 10.

Progress is more readily made when considering the plume downwind from the source. The plume width increases as a result of a lateral buoyancy-driven motion and diffusion by atmospheric turbulence, whereas the plume depth decreases as a result of the lateral spreading and increases as a result of diffusion by atmospheric and self-generated turbulence. The plume width increases with  $g'_0$  and  $q_0$  and increases markedly as the ambient velocity  $U$  is decreased. This can lead to plumes with very small aspect ratio and a ground-level coverage quite distinct from those observed in conventional dispersion experiments. For example, field experiments at Thorney Island, UK (see McQuaid 1987), produced a plume about 250 m wide and 3 m deep, 200 m downstream from the source.

A simple analysis by Britter (1980) based on downwind advection and lateral buoyancy-driven spreading found that a characteristic plume width is given, as a function of downwind distance  $x$ , by

$$L_H = L_{H0} + AL_b^{1/3} x^{2/3}.$$

There has been some confirmation of this correlation (Britter 1980, Neff & Meroney 1982), with an uncertainty in the coefficient as a result of poor

definition of the velocity  $U$  and of the plume width  $L_H$ . Britter & Snyder (1988) obtained a coefficient of 0.50 when the advection velocity was taken as that at the centroid  $\bar{z}$  of the vertical concentration distribution and the plume width was characterized with the second moment of the lateral, ground-level concentration distribution. The visible plume width is about five times larger.

The plume has sharp, well-defined edges (Figure 3a) when  $(g'h)^{1/2} \gg u_*$ ,

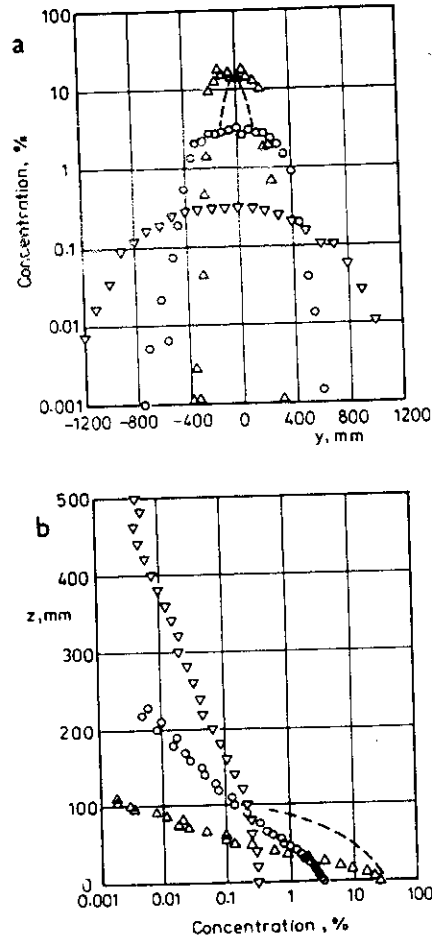


Figure 3 Profiles of mean concentration in a  $\text{CO}_2$  plume (Britter & Snyder 1988) at downstream positions where  $(g'h)^{1/2}/u_* = 2.2(\Delta)$ ,  $1.4(\circ)$ , and  $0.78(\nabla)$  for the (a) lateral and (b) vertical directions. The dashed lines are for a passive plume at the same position.

and the lateral concentration profile approaches a Gaussian shape when  $(g'h)^{1/2} \ll u_*$ , where  $g'$  and  $h$  characterize the plume density difference and depth. They are defined such that  $g'h = \int_0^\infty g[(\rho - \rho_a)/\rho_a] dz$ . The criterion  $(g'h)^{1/2}/u_* \approx 1$  apparently demarcates buoyancy-dominated flows from those dominated by turbulent diffusion (as regards lateral growth). Note that  $u_*$  is the friction velocity for the turbulent boundary layer incident upon the plume.

In contradiction to an often-made assumption, there is no evidence in laboratory or field measurements that dense-gas plumes appear as a well-mixed layer surmounted by a sharp density interface. In fact, quite the opposite is observed, with the vertical profiles of mean concentration having a near-exponential variation (Figure 3b). The height of the plume centroid is substantially reduced when the plume is dense, which is a result of the buoyancy-driven flow and the reduced vertical mixing. The growth rate of the plume centroid is still inhibited even when  $g'h/u_*^2 < 1$ . Density-stratification effects were found to be negligible when  $g'h/u_*^2 \approx 1$  for a similar but two-dimensional experiment (see Britter 1988), for which  $g'h/u_*^2$  is constant. Thus, the three-dimensional flow appears to be still influenced by the substantially larger values of  $g'h/u_*^2$  closer to the source. Near to the source, the increased plume width  $L_{H0}$  will act to restrict  $g'h/u_*^2$  to a value less than about 50 (Britter 1988). Conservation of the negative buoyancy of the plume ensures that  $g'h$  is nearly inversely proportional to the plume width and thus decreases as  $x^{-2/3}$ .

The density difference will reduce mixing between the plume and the environment, but the larger surface area of the plume across which mixing takes place will enhance plume dilution. Observations from Britter & Snyder (1988) and W. H. Snyder (unpublished) show that these two effects can often produce a decay of the maximum, ground-level concentration very similar to that for neutrally buoyant passive plumes (Figure 4). Other laboratory studies (Meroney 1982) have also found that the ground-level concentration is not as strongly influenced by the density difference as is the plume shape. Britter & McQuaid (1988) were able to correlate laboratory and field data, finding that

$$x_i / \left( \frac{q_0}{U} \right)^{1/2} = f \left( \left( \frac{q_0 g_0'^2}{U^5} \right)^{1/5} \right),$$

where  $x_i$  is the distance downwind to the concentration  $i$ . Only a weak dependence on  $g_0'$  was found.

No definitive data exist on the development of the velocity field within and near the plume. The intensity of all components of the velocity fluctuation are markedly reduced within and above the plume (Mercer &

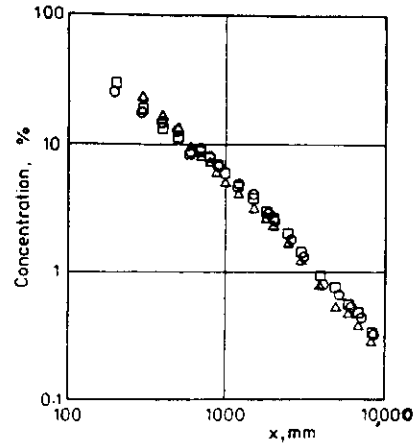


Figure 4 Comparison of the ground-level concentration from three plumes with similar  $q_0$  and  $U$ . The source gas is air ( $\Delta$ ),  $\text{CO}_2$  ( $\square$ ), and  $\text{SF}_6$  ( $\circ$ ).

Davies 1987, Koopman et al. 1988). Concentration fluctuations, as a ratio of the maximum mean concentration, are reduced when the plume is dense (Stretch 1986).

## 5. NEAR-INSTANTANEOUS RELEASE

The near-instantaneous release of material giving rise to a dense cloud may arise from the catastrophic failure of a storage vessel. This produces a rapidly expanding, entraining cloud whose description and quantification are still unclear.

Shortly after the release, this self-generated motion weakens and the cloud collapses toward the surface and spreads horizontally while being advected and diluted by the ambient flow. This latter collapse and subsequent motion have been extensively studied. The initial geometrical configuration is typically an aspect ratio (height to diameter) of about unity. The principal independent dimensionless parameter describing the flow is  $\{g_0 Q_0^{1/3} / U^2\}^{1/2}$ . The flow under calm conditions ( $U = 0$ ) is instructive. The similarity solution for this flow, if we assume no mixing between the fluids, shows that the radius of the cloud increases as (see, for example, Britter 1979)

$$r \propto (Q_0 g_0)^{1/4} t^{1/2}$$

The same equation may be obtained from a simple analysis using a collapsing flat-topped cloud. This description of the radial growth appears

quite robust. Field and laboratory experiments produce coefficients within the range  $1.1 \pm 0.1$  (Puttock 1988a). Most of these experiments were with an initial source density ratio of nearly two. However, as we note later, the flows are subject to substantial dilution, and much smaller density ratios prevail over times for which data were taken. Much smaller initial density ratios and smaller aspect ratios provide similar results (Huppert & Simpson 1980). The above coefficient is commonly related to the dynamics of a *steady* gravity-current head. An estimate of  $1.1 \pm 0.1$  is consistent with, though slightly smaller than, that found for the *steady* gravity-current head (Simpson & Britter 1980). More complicated integral models provide for global momentum and energy constraints and possibly include a simple turbulence model (van Ulden 1988); however, much of the influence of such models may be accounted for by replacing the period of cloud acceleration with an effective time delay of the order of several  $(Q_0^{1/3} / g_0)^{1/2}$ .

It is surprising how uninfluenced the spreading-rate formula is by mixing between the fluids, which must provide an effective drag on the spreading fluid. Mixing, however, does not affect the excess hydrostatic head, which would be responsible for the gravity-current flow.

The robustness of the spreading-rate formula limits its general usefulness as an indication of the correctness of any particular model. The excellent agreement between results for the leading-edge position as a function of time and predictions from some simple integral models has resulted in uncritical acceptance that these models are physically correct.

Observations (Spicer & Havens 1985; see also Figure 6) show that after the cloud collapse, much of the cloud material is contained within a toroidal vortex formed from the roll-up of the vorticity generated by the nonvertical density gradient at the cloud edge. This horizontal propagating vortex ring (or sometimes rings) is stabilized by vortex stretching and produces intense mixing of the cloud with the environment. Some of the mixed fluid is left behind the advancing vortex to provide a substantially diluted cloud. Eventually the leading-edge vortex weakens and adopts the classical, gravity-current-head form. It is only after this stage that the previous similarity analysis could be valid. A simplified analysis based on the shallow-water approximation (Rottman et al. 1985a), which neglects mixing between the fluids and the strong vertical velocities in the leading-edge vortex, is, however, of interest because it also clearly shows a concentration of the dense fluid in a narrow expanding ring. This expanding ring forms in a time of approximately  $4-6r_0/(g_0 h_0)^{1/2}$  and is dissipated in a time of  $40-60r_0/(g_0 h_0)^{1/2}$ , where  $r_0$  and  $h_0$  are the initial radius and height, respectively; these times were approximately confirmed in the field trials at Thorney Island (McQuaid & Roebuck 1985). The formation time is consistent with the delay time required to account for cloud acceleration.

The agreement between models relying on traditional gravity-current arguments and these observations should be considered fortuitous (possibly arising from dimensional considerations alone). The flow might be better represented as an expanding vortex ring near a wall, with the vorticity resulting from the initial collapse (Fannelop & Zumsteg 1986). The similarity analysis can only be appropriate for  $t \geq 40-60r_0/(g'h_0)^{1/2}$ .

This point may seem pedantic. However, the lack of identification of the relevant physical phenomenon does lead to difficulties of some consequence.

A very substantial dilution of the cloud is observed in laboratory (Spicer & Havens 1985) and field experiments (Brighton & Prince 1987). The cloud dilution is a direct consequence of the strong leading-edge vortex and is nearly an order of magnitude larger than the mixing associated with a gravity-current head (Puttock 1988a).

The same radial spreading formula is applicable, apart from the initial acceleration phase, for smaller aspect ratios, but for these cases the cloud dilution is reduced (Havens & Spicer 1985, Webber & Wheatley 1987, van Ulden 1988).

Releases in ambient flows, both in the laboratory and in the field, show a cloud spreading under its own buoyancy and being advected downwind. Multipoint data from large-scale field experiments (Brighton et al. 1985) confirm that the growth rates of cloud area are similar to those in calm conditions. The clouds are slightly longer than they are wide as a result of longitudinal dispersion. The movement downwind results from mixing between the cloud and the ambient flow (Rootman et al. 1985a, Wheatley & Prince 1987) rather than from any "form drag."

The cloud dilution, as measured by the area-averaged, ground-level concentration (Figure 5; from Brighton 1985, Brighton & Prince 1987), is initially similar to that under calm conditions. As a consequence, the distance downwind to a given concentration increases with the ambient wind speed. Later, a more rapid dilution rate is observed, and this is ascribed to mixing over the complete cloud area due to the ambient flow. This latter regime occurs earlier the smaller the initial value of  $\{g'_0 Q_0^{1/3}/U^2\}^{1/2}$ , or, effectively,  $g'_0 h_0/u_*^2$ . A passive dispersion phase is anticipated when  $g'h/u_*^2 \simeq 1$ .

## 6. DISPERSION MODELS FOR DENSE GASES

There are probably in excess of 100 analytical or numerical models currently available that purport to describe the dispersion of dense gases. Recent reviews (Blackmore et al. 1982, Wheatley & Webber 1985, Hanna & Drivas 1987) describe those models that are widely used. The reviews

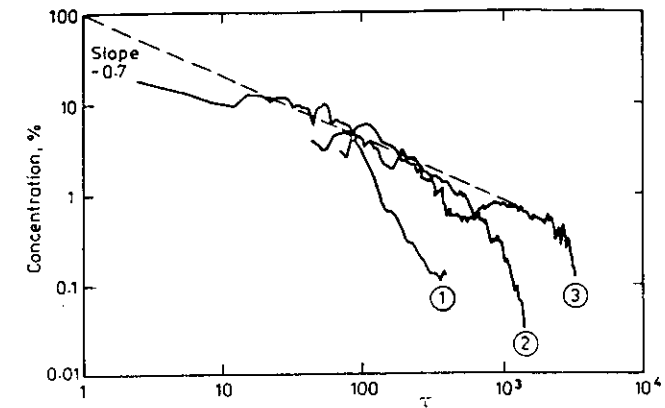


Figure 5 The area-averaged ground-level concentrations from field trials of instantaneous releases. Curves 1, 2, and 3 correspond to  $\{g'_0 Q_0^{1/3}/U^2\}^{1/2} = 1.4, 3.7, \text{ and } 7.3$ , respectively. The nondimensional time scale  $\tau$  is proportional to  $t g'_0^{1/2}/Q_0^{1/6}$ .

do not, however, provide a technical assessment of the models. Such an assessment, both in terms of the incorporated physics and an objective validation of the several relevant model outputs, is still lacking (Mercer 1988).

An early approach to modifying conventional Gaussian dispersion models was found to be inadequate when experimental results became available in the 1970s. Their use had, in part, led to uncertainty in predictions of nearly two orders of magnitude (Havens 1980). Subsequent model development has been along two distinct lines.

The first approach, referred to as three-dimensional models, addresses the Reynolds-averaged, three-dimensional, time-dependent conservation equations. The most common of these use empirical  $K$ -theory for turbulent closure. Havens et al. (1987) compared the four best-known models and highlighted the numerical difficulties associated with the transient, variable aspect-ratio problem (thus rejecting two models) and the uncertain application of the turbulence submodels used (thus rejecting a third). The remaining model (see Chan 1983) showed some agreement with a limited data base. The vertical diffusivity profile is being modified in order to improve agreement (Ermak & Chan 1988). Recent use in the modeling of field experiments of  $\text{NH}_3$  and  $\text{HF}$  spills (Chan et al. 1987b) shows gross qualitative and quantitative differences, though these may be due to inappropriate thermodynamics rather than poor fluid mechanics. Chan et al. (1987a) noted that the modeling of a few minutes of real time took several hours of CRAY-1 computer time.



Incorporation of a more advanced turbulence model ( $k-\epsilon$ ) has been attempted. The future of these models is uncertain. Deaves (1985) concludes that for three-dimensional modeling, "there will be very few cases in which its expense could be justified," and Betts & Haroutunian (1988) stress the "severe numerical problems that can be encountered."

The second and simpler approach is basically an integral formulation, with any variations of the cloud or plume in the vertical or lateral direction integrated out and, if appropriate, later reincorporated through empirically determined profiles; this is a common approach for many applied problems in fluid mechanics. These models are referred to as box models. Though limited in their flexibility, they have only a small number of adjustable constants, whose effect may be easily interpreted physically. They are also computationally inexpensive. Hanna & Drivas (1987) list over 40 models.

A comparison of the output from 11 integral models prior to the field experiments at Thorney Island, UK (McQuaid & Roebuck 1985), produced a variation in predicted concentration greater than one order of magnitude (Mercer 1988). Subsequent modification of these integral models due to recent results from several laboratory and field experiments has substantially reduced this uncertainty.

The integral models incorporate three specific effects:

1. the cloud spreads horizontally under its own negative buoyancy,
2. there is dilution of the cloud by mixing with the ambient environmental flow, and
3. the cloud is advected by the ambient flow.

Horizontal spreading is modeled with a gravity-current-head formula (Simpson 1982), such that the edge velocity is  $U_r = K(g'h)^{1/2}$  with the coefficient  $K$  given a value near unity. The initial acceleration phase may be modeled using an overall momentum argument, for example (Spicer & Havens 1987), though this may not be of consequence except very close to the source.

The buoyancy-driven flow will eventually be altered by the ambient flow and turbulence. Typically, this is anticipated to be when  $U_r = u_*$ , i.e.  $(g'h)^{1/2}/u_* = 1$ .

Subsequent horizontal spreading is modeled as a passive release (though possibly from a finite-length line source).

The dilution of the cloud is modeled using an entrainment velocity approach (see Turner 1986), such that the volume growth rate of the puff or plume section is given by

$$\frac{dV}{dt} = u_e A_e + u_t A_t,$$

where  $u_e$  and  $u_t$  are edge and top entrainment velocities, respectively, and  $A_e$  and  $A_t$  are the relevant edge and top entrainment areas, respectively.

The edge entrainment velocity is typically scaled with  $U_r$ , and the edge entrainment coefficient  $u_e/U_r$  used in models is typically quite large (0.6–0.9) (Puttock 1988a). For example, the model results given by the dashed line in Figure 5 are based on a coefficient of 0.7. These large values are based on results from field and laboratory experiments of unity aspect ratio, and their applicability to other situations has already been questioned.

The increased horizontal area of clouds with time produces a large surface area over which top entrainment takes place. Various correlations have been used (Hanna & Drivas 1987) of the form

$$\frac{u_e}{u_*} = \frac{a}{1 + b \left( \frac{g'h}{u_*^2} \right)}.$$

Coefficients of  $a = 0.40$  (= von Karman's constant) can be deduced from laboratory experiments (Britten 1987). This equivalent correlation has been successfully used in meteorological models (e.g. Colenbrander & Puttock 1983, Spicer & Havens 1987). The use of an incident, and therefore independent, correlation is a simple but superficial treatment. The local  $u_*$  will depend on the stratification locally and immediately upwind.

The modeling of the cloud movement downwind is based either on entrained momentum or, more simply (and valid only for releases from the source), on the ambient wind speed at a height representative of the cloud depth.

Although further development is required, modeling based on the above approach, together with a suitable profile description, can satisfactorily reproduce many aspects of field and laboratory experiments (Spicer & Havens 1987, Puttock 1987a,b).

Incorporation of heat transfer and other gross buoyancy effects is straightforward.

Few models are able to treat transient releases, particularly the near-source region, where the released gas may form a wing cloud at the source until the release rate is equal to that carried away by the wind. Progress is limited by uncertainty about the relevant mixing mechanisms.

Spicer & Havens (1987) and Havens et al. (1987) consider more complicated near-source flows in order to provide input to an integral dense-gas dispersion model.

Integral models are, in general, not suitable for situations in which

ies, respectively, and areas, respectively.

with  $U_r$ , and the edge entrainment coefficient  $u_e/U_r$  used in models is typically quite large (0.6–0.9) (Puttock 1988a). For example, the model results given by the dashed line in Figure 5 are based on a coefficient of 0.7. These large values are based on results from field and laboratory experiments of unity aspect ratio, and their applicability to other situations has already been questioned.

time produces a large surface area over which top entrainment takes place. Various correlations have been used (Hanna & Drivas 1987) of the form

and  $b = 0.125$  may be deduced from laboratory experiments (Britten 1987). This equivalent correlation has been successfully used in meteorological models (e.g. Colenbrander & Puttock 1983, Spicer & Havens 1987). The use of an incident, and therefore independent, correlation is a simple but superficial treatment. The local  $u_*$  will depend on the stratification locally and immediately upwind.

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Spicer & Havens (1987) and Havens et al. (1987) consider more complicated near-source flows in order to provide input to an integral dense-gas dispersion model.

Integral models are, in general, not suitable for situations in which

topography or buildings are present. As three-dimensional models may also be unsatisfactory a pragmatic alternative would be a shallow-water model, i.e., integration in the vertical alone. Although frequently proposed, no shallow-water models of general applicability are available.

## 7. THE INFLUENCE OF BUILDINGS AND OBSTACLES

Recent reviews (e.g. Portelli 1985) restate earlier requests (Britter & Griffiths 1982) for information on more realistic release scenarios. In particular, the release of toxic or flammable materials is unlikely to occur in the absence of some source structure, adjacent buildings, or buildings between the source and the receptors of interest. Nearby structures may be used as release mitigation devices. Air flow near individual buildings or groups of buildings is extremely complex. Turbulence will disperse the released material in a complicated manner dependent upon source configuration and building geometry. It is often difficult to generalize results for such flows, and the problem might best be addressed using specific physical modeling.

Whether the release is dense or not, the presence of buildings or obstacles will act to divert the mean-flow streamlines and to alter, and in general to increase, the turbulence. There is a reduction of velocity ahead of an obstacle with an increase at the sides or over the top. With bluff obstacles, the flow separates from the obstacle and produces a region of reverse velocity in the lee and a large increase in turbulence. Material entering this region will be retained there and released gradually, thereby extending in space or time a transient release.

Downstream of the region of reverse flow, both the magnitudes of the mean-velocity deficit and of the turbulence decrease and their spatial extents increase. A further complication is the downstream persistence of vortices aligned with the flow and with a rotational sense that typically produces a mean downward velocity along the axis. When the release is dense, there will be a reduction in any dilution caused by the obstacle-generated turbulence and also a specific interaction between the buoyancy-influenced cloud and the structure.

In summary, the principal effects of structures are

1. to divert the cloud or plume by altering the background, ambient flow and by interacting with the buoyancy-influenced cloud,
2. to enhance dilution as a result of increased levels of turbulence, and
3. to produce a time lag for the dispersion of material entering the near-wake of the structure.

The above list, in the absence of density effects, is comprehensively treated in the review by Hosker (1984).

The limited data base available for studying the influence of structures on the dispersion of dense gases is frequently of a specific rather than generic nature (e.g. Dirkmaat 1981, Kothari & Meroney 1982, Krogstad & Pettersen 1986, McQuaid 1986, Davies & Inman 1987), and one is left to interpret, and possibly quantify, the various observations in terms of the physical mechanisms listed above. Rottman et al. (1985b) have considered the interaction of steady and unsteady dense-gas flows with obstacles by using shallow-water equations. They obtained qualitative descriptions of various phenomena anticipated or observed in the field and the laboratory. These included two-dimensional solid and porous obstacles and three-dimensional solid obstacles. It was apparent that the shallow-water equations were an appropriate tool for addressing the problem of diversion of the cloud by obstacles.

Typically the relevant cloud variables are a Froude number  $U^2/g'h$  (where  $U$ ,  $g'$ , and  $h$  characterize the velocity, density difference, and depth of the dense material, respectively),  $H/h$  (where  $H$  characterizes the structure depth), and various other geometrical scale ratios. For example, inviscid analysis shows that a steady two-dimensional flow will be blocked by a two-dimensional fence when

$$U^2/g'h < \frac{1}{2} \frac{(H/h - 1)^2(H/h + 1)}{H/h}$$

but will otherwise clear the fence, although an upstream jump may occur.

Experiments by Britter (1987) broadly confirm this description but also show that various mechanisms of turbulent mixing upstream of a fence will eventually allow the dense cloud to surmount the fence. This simple example allows direct assessment of the usefulness of fences and barriers in mitigating the effect of accidental releases by altering the motion of the cloud.

The use of the shallow-water equations to describe the flow near three-dimensional obstacles is less common (Baines 1987), though see Lamb & Britter (1984).

Mitigation is also provided by cloud dilution as a result of structure-generated turbulence. Advanced turbulence models might be applied to flows with simple geometries, but these are expensive to use. Alternatively, a simple energy argument may be used that allows a small fraction ( $\alpha$ ) of the structure-generated turbulence to increase the cloud height, i.e. the use of an integral Richardson flux number (Linden 1980). Using the two-dimensional plume, two-dimensional fence example, one can easily show,

provided that  $g'h/U^2 \leq \alpha$  and that the plume can clear the obstacle, that there is ample production of turbulence to raise the center of gravity of the plume above the fence height. That is, the dense plume will behave as if passive in the immediate lee of the fence, and thus existing passive analyses can be used, although reverting to a dense plume farther downwind. Experiments (Britter 1986) confirm this behavior. Very marked dilution is observed when a three-dimensional plume (for example, from an area source) encounters a two-dimensional fence. The plume may be blocked by the fence, widen upwind, and then surmount the fence with subsequent dilution in the lee; this is a combined mechanism that produces increases in both the lateral and vertical plume dimensions.

A similar energy argument may be made for the continuous release of a dense fluid into the lee of a structure with side dimensions  $H \times H$ . Concentrations near the end of the separated region are comparable to those without any density difference for  $g'_0 q_0 / U^3 H \leq 0.1$ , where  $q_0$  is the volume release rate of dense fluid. Brighton (1986) has provided a more detailed, though basically a bulk, description of such a wake release.

In general, it is difficult to envisage situations in which the obstacles result in ground-level concentrations in excess of those observed in the absence of the obstacle, although the position of the maximum may not be on the downwind centerline. Possible exceptions to the above general statement would be when the lateral spreading of the plume is inhibited by obstacles (e.g. for flow along a "street canyon") when longitudinal vortices capture release material and are not broken up by the environmental turbulence, or where the obstacle results in the gas being diverted into regions where it would not go in the absence of the obstacle (e.g. the possible increased upwind spreading where a fence is very close to the source or the widening of a plume along the line of a two-dimensional fence).

These exceptions aside, we anticipate a reduction of the maximum ground-level concentration (on a radius centered on the release position) to be produced by the presence of one or more obstacles near or removed from the source. There are, however, very limited data to allow quantitative estimates to be made.

## 8. THE INFLUENCE OF TOPOGRAPHY

Variations in the elevation of the underlying surface will influence the buoyancy-generated motion of the dense gas. Topography, in the form of general slope, isolated hills, or more complex terrain, will alter or divert the cloud or plume. The topography may enhance plume dilution and divert the plume away from regions of elevated terrain. Alternatively, the

dense plume may be channeled into valleys or low-lying areas and then be protected from the diluting influence of the ambient flow. There is, of course, extensive treatment of the interaction of topography with buoyancy-influenced flows in the geophysical literature, but little use has been made of this information source.

Topographic features that are small compared with the size of the release may be considered in much the same way as buildings or structures but without any substantial flow separation unless the topography is very abrupt. The shallow-water equations are useful here (e.g. Lamb & Britter 1984, Lee & Meroney 1988). For example, provided that, for a steady plume,

$$U^2/g'h \leq 2 \left( \frac{h_c}{h} - 1 \right),$$

the plume (or at least the bulk of it) is unable to reach an elevation  $h_c$ . This simple result may be reinterpreted as a criterion for the necessary dilution, or reduction in  $g'$ , for the plume to be able to reach an elevated receptor (Heinold et al. 1987).

When a topographic feature is large compared with the scale of the release, the topography reduces to a local slope. Somewhat surprisingly, the downslope velocity of a dense fluid released on a slope under calm conditions is not a strong function of slope. Hopfinger (1983) summarizes results for instantaneous, continuous, and starting plume flows and finds that the flow has velocities such that  $(g'h)^{1/2}$  is typically between 1 and 2 for slopes between  $0^\circ$  and  $90^\circ$ . Steeper slopes lead to increased entrainment and dilution, with the entrained fluid acting as an effective drag on the downslope flow.

Little information is available concerning three-dimensional flows under calm conditions. Unlike for two-dimensional flows, the release is able to spread across the slope, and the Reynolds number based on the flow depth, may decrease with distance from the source (Fietz & Woodcock 1967). As a consequence these flows, in the laboratory, are frequently influenced by viscosity, which reduces the downslope velocity, the width, and the dilution.

Picknett (1981) found that instantaneous releases under noncalm conditions on a slope of 1/13 were influenced by the slope for very low-wind-speed conditions. Hall et al. (1974) and Meroney et al. (1977) observed that slopes of 1/12 and 1/50, respectively, altered the continuous-plume results.

Broadly, three characteristic velocities are relevant

1. the ambient wind velocity,

2. the buoyancy-generated velocity found on flat terrain, and
3. the buoyancy-generated velocity found on slopes (the downslope flow).

The latter two velocities both scale on  $(g'h)^{1/2}$ . As the coefficient in the expression for the slope flow is only a weak function of slope, the slope will have an effect on clouds for which any buoyancy-generated velocities are relevant. However, the flow-development times may differ. For example, an instantaneous release might initially spread radially, then develop a bulk downslope flow, before finally being diluted and swept upslope by an ambient flow.

One criterion for slope influence, provided by Fay & Ranck (1983), is that  $u_*^2/g'h \leq \Theta$  (where  $\Theta$  is the slope) has not been validated.

When the wind is upslope, the cloud widens and its dilution is enhanced. When the wind is downslope, the cloud is narrower and the dilution is decreased. The variation of the lateral growth of the plume results from an effective summation of the wind and the buoyancy-induced motion down the slope. The entrainment is influenced by the velocity shear and will therefore be enhanced by an upslope wind and reduced by a downslope wind. The ambient velocity required to reverse a downslope flow of a plume or cloud is a weak function of slope and is typically twice the downslope flow under calm conditions (Turner 1973). In the case of cross winds, Hall et al. (1982) found that the dilution is not greatly affected, although their conclusion is based on a single wind-tunnel experiment. Further discussion of these points is available in Britter (1982).

A distinctly different topographic influence occurs when the topography alters the velocity field within which the cloud is dispersing. Britter & Snyder (1988) found this to be more important than the direct effect of a slope on the cloud.

Ermak et al. (1982) provide results of field experiments showing the effect of more complicated topography, with the plume moving to low-lying areas.

The scouring of dense gases from low-lying areas by the ambient flow has not been addressed, though Bell & Thompson (1980) consider a similar problem.

## 9. PHYSICAL MODELING OF THE DISPERSION OF DENSE GASES

Physical modeling, using wind tunnels or water flumes, is a particularly attractive technique for the study of the dispersion of dense gases, but there are some specific constraints upon its applicability. The technique of physical modeling incorporates a model of the fluid mechanics superior to

any current mathematical model, provides unequalled spatial and temporal resolution, and is ideally suited to flows influenced by buildings, obstacles, or variable terrain. A particular advantage of physical modeling is that it is possible to obtain information on the variability between members of an ensemble of transient releases (Meroney & Lohmeyer 1984); available mathematical models only provide results for the ensemble alone. However, the physical modeling of multiphase and other thermodynamic effects, including heat transfer to the underlying surface, is generally not possible (Britter 1987).

The physical modeling of the atmospheric dispersion of neutrally and positively buoyant pollutants has been well demonstrated for downwind distances from the source of up to about 10 km, though there is less experience for atmospheric flows that are not neutrally stratified.

The relevant dimensionless groups required for modeling, with a characteristic length scale  $L$ , an instantaneous (or steady) release of a dense gas are  $U^2/g'_0L$ ,  $Q_0^{1/3}/L$  (or  $q_0/UL^2$ ),  $\rho_0/\rho_a$ , various geometrical ratios and boundary conditions, and two groups dependent upon fluid properties ( $\nu$ , kinematic viscosity;  $\mathcal{D}$ , diffusivity)—the Reynolds number  $UL/\nu$  and the Peclet number  $UL/\mathcal{D}$ . A transient source flow rate  $q_0(t)$  would be  $q_0(tU/L)/UL^2$  in the model. Pragmatic fluid modeling relies on the insensitivity of turbulent diffusion and dispersion to the Reynolds and Peclet numbers, provided that these numbers exceed some critical values. Dense-gas effects at full scale are most in evidence for small ambient wind speeds. This, combined with the need to model  $U^2/g'_0L$ , requires much reduced model wind speeds. This reduction, essentially in the Reynolds number, produces practical difficulties in maintaining controlled turbulent flows (Snyder 1981).

The very strong density gradients associated with dense-gas clouds act to inhibit turbulence and render the flow even more prone to the influence of the Reynolds number. As a result, fluid modeling of a dense-gas plume is generally more contentious than a comparable positively buoyant release.

Broadly speaking, an inadequate Reynolds number will make itself obvious by causing the cloud to laminarize, an obvious invalidation of the modeling attempt. If cloud dilution due to entrainment becomes very small in a fluid model, the plume dilution will be influenced by molecular diffusion. This may be significant when  $u_*^3/g'\mathcal{D} \leq 2$  (Meroney 1987). However, in practice, molecular diffusion is unlikely to be of consequence in a model unless the plume has laminarized and the entrainment has fallen to zero. In order to ensure suitable velocities in physical-modeling facilities, some distortion of the source density difference is possible (Davies & Inman 1987, Spicer & Havens 1985). There is still uncertainty about a

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permissible magnitude for the distortion, and it may depend upon the type of release being modeled.

Cloud dilution near the source due to nearby structures or as the result of an initial collapse of a dense cloud also reduces the tendency of the flow to laminarize. Model cloud laminarization is currently detected by observation or experience. Meroney (1987) discusses these and other points relevant to physical modeling and provides a summary of specific examples of physical modeling.

Photographs (Figure 6; from Hall & Waters 1985) of a 1:90 scale model of an instantaneous release support the usefulness of physical modeling, as do the concentration records (Figure 7; from Davies & Inman 1987) measured downwind of a continuous (though a limited duration of about 450 s) release near obstacles for model scales of 1:40 to 1:250.

Comparison is made difficult because both the field experiment and the model are each individual members of an ensemble, and it is the ensemble mean fields that are directly comparable. The concentration records in Figure 7 are typical of the variation found between members of an ensemble for a fixed scale ratio. Davies & Inman (1987) provide statistical analyses comparing wind-tunnel observations with field experiments. The physical-modeling results can broadly be characterized as satisfactory, particularly when substantial dilution is caused either by the initial potential energy of the release or by nearby sharp-edged bluff bodies.

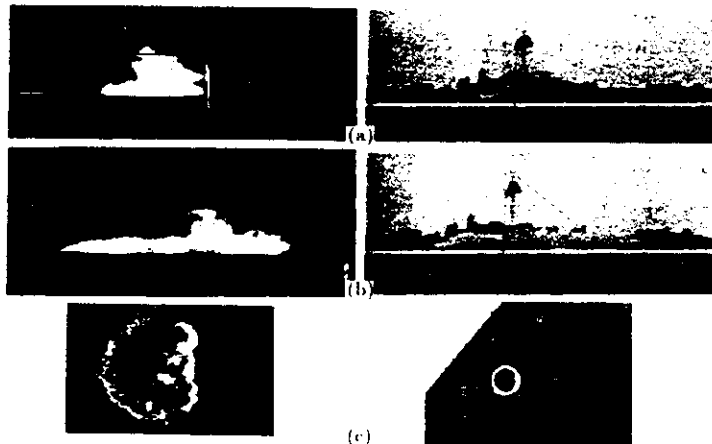


Figure 6 Physical modeling in a wind tunnel of an instantaneous release of  $2000 \text{ m}^3$  of a dense gas. (a) Photographs 3 seconds after release and (b, c) 10 seconds after release (reprinted with permission from Hall & Waters 1985).

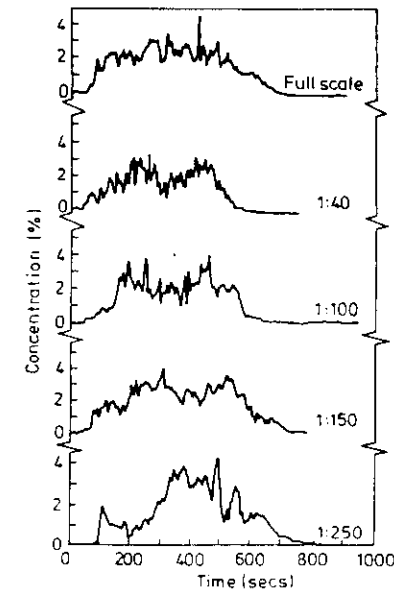


Figure 7 Physical modeling in a wind tunnel of a continuous (though limited duration) dense-gas release near obstacles (reprinted with permission from Davies & Inman 1987).

## 10. FURTHER CONSIDERATIONS

### 10.1 Surface Roughness and Atmospheric Stability

There are no specific studies concerning the influence of surface roughness or atmospheric stability on the dispersion of dense gases. From available data, Britter & McQuaid (1988) conclude that the effects of surface roughness and atmospheric stability are less, as regards downwind concentrations, than for a similar passive release. Both effects reflect the link between  $u_*$  and  $U$ . Thus, the various model formulations will, by introducing correlations in terms of  $u_*$  and/or  $U$ , produce roughness and atmospheric-stability effects that may be quite spurious.

### 10.2 Elevated Releases

The model of Ooms (1972) is frequently used for elevated jet releases. Interaction of the plume with the underlying surface is not addressed. Meroney (1982) summarizes various laboratory experiments, including plume touchdown position and concentration at touchdown.

### 10.3 Concentration Fluctuations

Griffiths & Megson (1984) emphasize the importance of concentration fluctuations in assessing toxic load. Similarly, the ignitability, and sus-

tained burning, of flammable materials will be influenced by a local concentration rather than the mean. Chatwin (1982) has pointed out the limitations of models that only address time-mean or ensemble concentrations. Meroney & Lohmeyer (1983) consider the statistics of instantaneous releases, while Stretch (1986) provides data for continuous releases. Carn & Chatwin (1985) address the variability between nominally identical instantaneous releases. Hanna & Drivas (1987) note that there is often uncertainty as to what averaging times are implied in model results. A related problem is the optimization of model constants using limited data from realizations with different initial conditions, which is often the case with field experiments (Sherrel 1987).

#### 10.4 Major Research Activities

Despite some of the misgivings presented in this review, there is a growing consensus that our ability to predict the atmospheric dispersion of dense gases in an idealized situation is now significantly better than that for other related problems. Thus there is currently active research on

1. the influence of obstacles and topography;
2. the prediction of the source term, in the form of (a) the quantity released, (b) the local mixing mechanisms arising from single and multiphase jets (i.e. the rate of dilution near the source), and (c) the liquid fraction entrained and the size distribution of the aerosol cloud after a pressurized release;
3. concentration fluctuations;
4. model assessment techniques; and
5. possible mitigation procedures.

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## Update to 'The Atmospheric Dispersion of Dense Gases'

In this section I list various recent developments in the subject of the dispersion of dense gases. These will be elaborated on in the presentation, particularly in response to the interest shown in each of the various topics.

1. The 'Workbook on the Dispersion of Dense Gases' by Britte and McQuaid (1988) has found widespread use in Europe, the USA and elsewhere. The workbook approach has performed well in independent evaluation exercises (Hanna et al. 1993) and has been incorporated into the US Environmental Protection Agency's screening model TOXSCREEN.

2. Three important publications are:

Hanna S.R., Drivas P.J. (1987)

Guidelines for the use of vapor cloud dispersion models

CCPS/AIChE

345 E. 47th St., New York, NY 10017.

Hanna S.R., Strimaitis D.G. (1989)

Workbook of test cases for vapour cloud source dispersion models

CCPS/AIChE

345 E. 47th St., New York, NY 10017.

Hanna S.R., Chang J.G. and Strimaitis D.G. (1993)

Hazardous gas model evaluation with field observations

*Atmosphere Environment*, 27A, 15, pp.2265-2285.



3. The Commission of the European Communities has continued to fund research and development work on the dispersion of dense gases under the Environment Programme. The appropriate contact is

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4. There have been attempts to extend integral models to more realistic scenarios including buildings and topography (see Britter, R.E., Cleaver I.P. and Cooper M.G. (1991) Development of a simple model for the dispersion of denser-than-air vapour clouds over real terrain, British Gas Midlands Research Station Report No. MRS E 622).

5. There has been great concern, particularly in the US, on the problems of

- (i) whether integral models can be applied to real industrial sites by increasing the surface roughness length  $z_0$ ;
- (ii) what averaging times are explicit or implicit in model predictions, are these valid;
- (iii) application of integral models to the more realistic transient or finite duration release.

6. It appears that most integral models consistently predict vapour clouds that are too wide. There have been attempts to remedy this possible defect.

7. The model HGSYSTEM, which is an extension of the Shell model HEGADAS, has been developed for application to releases of hydrogen fluoride and includes the appropriate complex chemistry. Other materials can only be treated as perfect gases but there is current work to extend its applicability.

8. There has been growing interest in the application of computational fluid dynamics to the problem of the atmospheric dispersion of dense gases. One approach is to use conventional three-dimensional CFD; the other is to exploit the large horizontal to vertical ratio of scales and thereby use a shallow-layer analysis.