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Dispersion of Denser-than-Air Gases

by

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LERAMY / BULLIOTH COM ATANAOPTICAL ENGLIGENT SETAMAT SINDE DE L'ESTINE ANDER DE ATENNE SETURE GOS PUT DIALETTA TIDE STUMPEN, ONTAILS, Coverage and sie February, 1980

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ABSTRACT

The problems associated with accidental spills of toxic and also flamable denser-than-air gases have gained new importance in recent years with the greater quantities of these substances being produced and transported. In particular, the chlorine spill in Mississauga, Ontario in November 1979 has resulted in renewed interest in environmental assessment of the hazard of such spills.

This report reviews knowledge in this area and presents recommendations for AES application and research initiatives. An annotated bibliography is included.

1. Introduction

Problems caused by the release of denser than air gases have been a major motivating factor in the study of air pollution dispersion since early in this century. Indeed one can date the era of modern air pollution studies from work in the U.K. during and following World War 1 where chlorine and phosgene played such a devastating role in trench warfare. Strangely though, those early studies seemed to ignore the density property and it was not until the late 60's and 70's that new impetus for such studies developed - this in connection with problems of accidental spills during transportation. An indication of importance of this problem is given by the following compilation of recent chlorine accidental spills.

> July 13, 1972: Port Alice, BC Narrow escapes; rupture of chlorine transfer hose between rail tank car and mill.

Sept. 7, 1973: Greensburg, Pa.

57 injured; valve jammed on 55 ton chlorine rail tank car.

Dec. 15, 1975: Niagara Falls, NY

4 dead, 77 injured, 73 hospitalized from shopping centre 3 miles from accident; chlorine rail tank car explosion.

1976: Baton Rouge, La.

10,000 evacuated; storage tank rupture, 42 mile long cloud formed.

Feb. 27, 1978: Youngstown, Fla.

8 dead, 100 hospitalized, 1250 evacuated; derailment of chlorine tank car, impacts associated with gas accumulation in low ground.

Sept. 25, 1978: Vancouver, BC 32 hospitalized; 5 chlorine canisters fell off truck.

Nov. 11, 1979: Mississauga, Ont. 1 chlorine hospitalization, 240,000 evacuated;

Gases in the atmsophere may be denser than air for two reasons. Firstly, their molecular weight may be greater than air. While air has a molecular weight of 29, chlorine is 71, and phosgene 99. Secondly, they may be colder than air. Ammonia gas has a molecular weight of 17 and in accidental spills can be emitted at its boiling point -33° C. If mixed 10% by volume with air at this temperature, a not untypical spill situation, the mixed gas is \sim 10% more dense than ambient air. Similar considerations apply for LNG and LPG spills. Often both factors are important. For chlorine, accidental spills are usually cold augmenting the density from the high molecular weight, however, sulphur dioxide (molecular weight 64) is usually released as a hot chimney gas giving a net density considerably less than ambient air.

All of the above noted heavy gases are either acutely toxic (chlorine, phosgene, ammonia) or flamable (LNG, LPG). Being denser than air they tend to hug the ground and thus pose major hazards to the environment, including human life, in the vicinity of releases. This ground hugging

action is manifested in that:

- (i) it causes very rapid initial gravity driven spreading so that even locations somewhat upwind of the spill can be affected;
- (ii) residual gas tends to pool in hollows, under viaducts, within vegetation and can persist there for some while.

Studies of the dispersion of heavy gas spills are presently underway in the U.S. and U.K. The U.S. work is primarily motivated by problems of accidental spills of LNG, LPG. Theoretical numerical modelling studies are being conducted by Science Applications Inc. (San Rafael, California) and at Lawrence Livermore Laboratory, wind tunnel simulations by Colorado State University (Fort Collins, Colorado), and field studies at the Nevada Test Site. Much of the above work is cooperative with the U.S. Department of Energy funding and Battelle Northwest Laboratory (Richland, Washington) being a prime coordinating agency. In addition to the above Briggs (personal communication) indicates that there has been quite extensive proprietary work by multi-national oil/chemical companies.

In the U.K. important theoretical, numerical and physical modelling studies have been undertaken at Cambridge University. Government sponsored studies include those by the Atomic Energy Authority, the Warren Spring Laboratory and the Health and Safety Executive. This latter organization has sponsored a program of small-scale field trials at the Chemical Defence Establishment (Porton Down) and some larger scale releases are planned.

The October, 1979 NATO/CCMS meeting in Rome had seven papers on the program relating to this problem, this shows a considerable increase in activity in recent years. Scientists from The Netherlands, The Federal

Republic of Germany, and Norway presented papers as well as those of the United States and United Kingdom.

2. <u>Definition of the Problem</u>

Within the general scope of this type of problem one may be concerned with different types of source configurations and different types of hazard.

The source configuration may be highly irregular as it will likely result from a rupture in the containing vessel. Ruptures may be of various sizes, above or below the fluid level in the container and the fluid propelled out may be propelled anywhere from directly up to directly down. McQuaid (1979) identified this source configuration problem as an important research area. However, he also noted from an examination of actual incidents that " ...catastrophic failure in which a large amount of material was released in a short time was the most important". "It appears to be a characteristic of catastrophic failures that, irrespective of the conditions that prevail immediately upon loss of containment, a cloud forms very quickly around the container". He goes on to recommend separating the problem into that of the formation of the cloud and that of the subsequent evolution of the cloud so formed.

The other aspect that needs to be borne in mind in defining the problem is the nature of the hazard of concern. In the case of a toxic gas release, is the hazard best considered in terms of the instantaneous short term concentration encountered, a longer term dose, or some other characteristic? These characteristics will be quite different from those important when say, flamability is the hazard.

Consider the specific problem of a chlorine release. The following information on effects of chlorine is given by Sax (1975):

"A concentration of 3.5 ppm produces a detectable odor; 15 ppm causes immediate irritation of the throat. Concentrations of 50 ppm are dangerous for even short exposures. 1,000 ppm may be fatal, even when the exposure is brief".

If the objective is to protect the human population, say by evacuation, what is the criterion which says a particular location should be evacuated? These criterion may be even more complex when one realizes that a person in a well insulated home or in upper levels of a high-rise can be subject to considerably smaller concentrations than for an individual outdoors at ground level.

3. The Processes

With the simplifications outlined above and taking the case of a chlorine tank-car accident for concreteness it is reasonable to approximate the gas behaviour in four phases, following the discussion of Kaiser and Walker (1978), hereinafter referred to as KW.

- 1. Formation of the Source Cloud
- 2. Slumping
- 3. Ground Hugging
- 4. Ambient Atmospheric Turbulent Dispersion

A second good reference on the physics of the interacting processes is Briggs (1975).

3 a) Formation of the Source Cloud

As noted before there are many different possible source configurations. KW suggest that the important aspects of source cloud formation for gas escaping from pressurized containers are:

(i) Flash-off: vaporization of gas with latent heat drawn from the liquid remaining. The liquid is cooled to its boiling point. For chlorine the boiling point is -35°C and (as for ammonia) about 20% of the tank contents can be evaporated in this manner.
(ii) Entrainment of Liquid Drops: a proportion of the liquid may be thrown out of the container by mechanical action. The proportion can vary from 0 to 80% depending on the source configuration. Many of these entrained drops are evaporated due to

(iii) Entrainment of Air: because of the violent nature of the release a large mass of air also becomes involved in the source cloud. An air-chlorine ratio 5:1 is needed if all the chlorine mass is to be evaporated at its boiling point using heat from the liquid and entrained air. KW infer a ratio 10:1 for an historical ammonia accidental release and refer to a similar ratio found for a Freon-12 release.

From the above two important properties of the source cloud are evident. Firstly, it is dense and cold, so that the subsequent evolution will be strongly dominated by these properties. Secondly, it is formed by the entrainment of a large mass of air. By conservation of momentum one might expect that the initial cloud centre of gravity motion will closely follow the ambient wind.

3 b) <u>Slumping</u>

This phase is dominated by gravitational effects, the height of the cloud decreases and its lateral extent increases. There is a growing literature devoted to this regime and in particular the structure and dynamics of the expanding edge. Some references are given later. A simple approach is to assume the cloud is a cylinder of radius R, height h and density ρ in an ambient atmosphere of density ρ_a . Dimensional arguments indicate

 $\frac{dR^2}{dt} = 2c \left(\frac{g(\rho-\rho_a)V}{\pi\rho}\right)^{1/2}$

where $V = \pi R^2 h$, c is a constant, g is the acceleration of gravity and t is time. Theory suggests that c is in the range 1.0 to 1.4. Integrating

the above and assuming volume is conserved gives

$$R^{2}(t) = R_{0}^{2} + 2c \left(\frac{g(\rho-\rho_{a})V}{\pi\rho}\right)^{1/2} t$$

KW show that slumping proceeds, relatively independently of ground heating and additional air entrainment effects, in a well defined radial growth phase. The cloud so formed is much broader than that resulting from turbulent dispersion of a neutrally buoyant cloud.

As slumping continues the radial growth slows. Eventually it will fall below that one would expect for a passive pollutant due to atmospheric turbulence. This is one definition for the end of the slumping phase. Others are noted in KW.

3 c) Ground Hugging

On termination of slumping the cloud is of a broad but shallow form, still cold and dense compared to the ambient atmosphere, and having only slightly less chlorine concentration than initially. Lateral turbulent dispersion proceeds at a rate appropriate for a passive pollutant, but vertical dispersion is inhibited owing to the stable stratification at cloud top. It is possible to develop a formulation for vertical growth at this stage based on similarity with the deepening daytime mixed layer. During this phase, chlorine concentrations will decrease, although not as rapidly as for a neutrally buoyant cloud, and the density difference will decrease owing to air entrainment and ground heating.

3 d) Ambient Turbulent Dispersion Phase

When density difference effects become negligible, dispersion will continue at a rate appropriate for a passive pollutant cloud. At this stage the cloud will be broader and not as deep as an equivalent passive pollutant cloud of the same age as the chlorine cloud.

The above considerations are summarized in the following two diagrams which show <u>schematically</u> the evolution of the horizontal and vertical dimensions of a chlorine cloud (solid line) and comparative evolution for passive turbulent dispersion from a point source (dash line) and cloud source (dash-dot line).



Note that for vertical spread, except close to the source, a passive cloud model overestimates the spread while horizontal spread is underestimated by the passive cloud model. It has been suggested that roughly, these are compensating effects and that reasonable estimates of downwind maximum concentration can be obtained using spreads for the neutral buoyancy pollutant model. In similar vein, Meroney and co-workers at Colorado State University have interpreted their wind tunnel studies of LNG dispersion to imply

"The effect of negative buoyancy on the behaviour of a ground source is primarily multiplicative as the (concentration) decay relationship is not changed in form. Large specific gravities produce only moderate percentage increases in downstream concentration values rather than order of magnitude changes. Negative buoyance causes larger lateral and smaller vertical plume dimensions than are observed in cases of neutral buoyancy".

4. Effects of Topography

Slope is known to have a marked influence on local airflow over cooled topographic features (slope and valley flows). Similar effects have been noticed in the Hall et al (1974) wind tunnel studies and in the accident investigated by Booij (1979). One might expect these to be most important for low prevailing wind speeds. No available model appears to consider topographic effects.

Perhaps a more important topographic effect is that of trapping of dense gas in hollows. This was the cause of the most major effect of the incident at Mississauga. This type of persistence does not appear to have been studied and would seem to offer a fruitful and useful area for study.

5. <u>Present Assessment Models</u>

As a general comment two rather conflicting aspects to present assessment models emerge:

- In the Mississauga incident two models based on the Gaussian formulation were employed as input to the evacuation decision. One was computed by the Ontario Ministry of the Environment, the other by Dow Chemical Company. The radius for evacuation indicated by the two models apparently agreed to within a couple of kilometers, about 17 km.
- 2) KW note a study by Havens (1977) which reviewed the kindred problem of assessing the distance a cloud of LNG vapour would need to disperse for concentrations to fall below the lower flamable limit. Apparently this distance varied from 1 km to 50 km between the various models. It is noted that the dominant reason for this magnitude of discrepancy was differences in the rate of entrainment of ambient air.

A description and some comments on specific present assessment models are presented below;

5.1 The DND Model

Information on this model was supplied by Dr. S. B. Mellsen of the Chemistry Section at Suffield Defence Research Establishment. Downwind dosage is computed from

 $C = q / \pi \sigma_z \sigma_y \bar{u}$

where

C is the dosage (mg min m^{-3})

q is instantaneous point source strength (mg)

 σ_v is concentration cross-wind standard deviation (m)

z is concentration vertical standard deviation (m)

 \bar{u} is the mean wind speed affecting the pollutant (m min -1)

The values of the sigmas needed are developed from the standard Pasquill-Gifford curves. A graph was supplied and is reproduced on the following page showing the estimated dosage as a function of downwind distance for an instantaneous release of 50 tons of chlorine or phosgene. DND recommended that the neutral stratification curves be applied for built-up areas, presumably to account for increased surface roughness and anthropogenic heat emission effects. The dosages are to be calculated pro-rata for other than 50 ton releases.

This model has the great virtue of simplicity. It requires only a look-up on a graph and a simple arithmetic adjustment for different source strengths. A technique such as this is probably indispensible, even if more sophisticated models are available.

A sample calculation for a 90 ton chlorine spill, and using the 10 minute intolerable chlorine dosage criterion of 1000 mg min m⁻³, indicates this occurs at a distance 10.5 km using the D stability but near 100 km using the F curve.

Dosage as a Function of Downwind Distance for the DND Model



5.2 The Dow Chemical Model

Information on this model was obtained from Mr. Dorton of the company Midland Michigan facility.

The equation used is the same as for the DND model, except that the release is considered to be continuous so that

C is concentration $(mg m^{-3})$

q is continuous point source strength (mg min⁻¹).

The model is computerized so that concentration isopleths are produced on a CRT.

Considerable study of release rates likely from in-plant accidents has been undertaken. For off-site accidents, particularly tank-car accidents, uncertainties of release rate and meteorology are thought to dominate over errors arising from the model simplicity.

5.3 Van Ulden's Model

This seems to have been the first of a number of multi-stage models. For an instantaneous source the model predicts the dimensions of a cloud cylinder, height h and radius r. The three stages are:

1) The source phase. Initial values of r and h (r_o and h_o) as well as cloud density ρ_0 are specified. This includes and allowance for initial entrainment. The initial volume is $V_0 = \pi r_0^2 h_0$.

The density spread phase. The equations are: 2) $r^{2} - r_{0}^{2} = 2 c \left[\frac{a(p_{0} - p_{a}) V_{0}}{\pi p_{0}} \right]^{1/2} t$

 $\frac{h}{h_0} = \left(\frac{r}{r_0}\right) \qquad 2\alpha - 2$

Van Ulden used small scale field trials to determine that to a good approximation $\alpha = 0$. Thus, the cloud volume and pollutant concentration does not change during this phase. The value c is chosen as one. By assumption this phase terminates when the radial velocity.

$$U_{f} = \frac{c}{r} \left[\frac{g (p_{o} - p_{a}) V_{o}}{\pi p_{o}} \right]^{1/2} = 2u_{*}$$

where u_{\star} is the friction velocity, approximately one-tenth the surface wind speed.

3) The turbulent spread phase. With cloud radius r_u and height h_u at the end of the preceeding phase an area source Gaussian model is employed to predict the subsequent evolution. The values of the radius and height are predicted from,

 $r = 2.14 \sigma_y (x - x_{oy})$ h = 2.14 $\sigma_z (x - x_{oz})$

with

$$r_u = 2.14 \sigma_y (x_u - x_{oy})$$

 $h_u = 2.14 \sigma_z (x_u - x_{oz})$

used to determine the offsets x_{oy} and x_{oz} . It is not clear from Van Ulden's paper which set of sigma curves he prefers.

By way of example consider a 90 ton chlorine spill mixed initially with five times the mass of air. At -35° C this gives an initial volume of 3.29 x 10^{5} m³, and initial density of 1.64 Kg m⁻³, an initial chlorine concentration of 2.74 x 10^{5} mg m⁻³ and, assuming the initial cloud height and radius are equal $r_{0} = h_{0} = 47m$. If u_{\star} is assumed to be 0.5 m s⁻¹ and the ambient air density 1.29 Kg m⁻³, then $r_{u} = 468$ m and $h_{u} = 0.48m$. This occurs after $t_{u} = 232$ s. During this time the concentration is unaltered and the centre of mass of the cloud has moved downwind to $x_{u} = 1.2$ km. Moving to the turbulent diffusion phase $r_{u} = 468$ m implies $\sigma_{z} = 219$ m and $h_{u} = 0.48m$ implies $\sigma_{z} = 0.22m$. Accepting for the moment that the Turner Workbook sigma curves are appropriate, then for all stabilities only the effect of the time delay needs to be included in assigning the offset x_{07} . Thus,

 $h = 2.14 \sigma_{z} (x - 1.2)$

For the x_{oy} offset both the impact of time delay and the enlarged gravity driven horizontal spread need accounting for so, assuming D stability,

$$r = 2.14 \sigma_v (x + 2.3)$$

Again, assuming that the chlorine is uniformly mixed within the cylinder at 10 km the cylinder has radius 1412m, height 321m and the concentration is 45 mg m⁻³. At 20 km the radius is 2568m, height 417m and concentration 10 mg m⁻³.

Making a subjective allowance of a safety factor ten in concentration to allow for non-uniform mixing in the cylinder and for short term fluctuations, then a distance of 20 km would appear to be just sufficient to ensure that the concentration had fallen below the intolerable chlorine level specified for the DND model above. It is worth noting that in this model the distance calculated differs little from that one would compute assuming a instantaneous point source of a neutrally buoyant gas. Certainly the difference in this case is within the error of the inherent assumptions.

5.4 The Germeles and Drake Model

In essence this model is formulated in a similar way to that of van Ulden, that is, it is a multi-stage model with matching between the stages and uniform mixing within a cylinder. One significant difference is that it is formulated for the problem of LNG spills. Thus, there is an explicit treatment of the formation of the initial cloud based on evaporation of a circular pool of LNG which can be tied to a model for the spread of the liquid on the surface. A second significant difference is that there is a specific accounting of air entrainment and heating of the cloud, both from entrainment and contact with the surface and including the effect of water latent heat, during the gravity spreading phase. The effects are incorporated in the form of interacting prognostic differential equations which need to be solved numerically. A final difference is that the termination of the gravity spreading phase is assumed to occur either at neutral buoyancy. for the no-wind condition or when the speed of the cloud periphery equals the ambient wind speed.

The model of Kaiser and Walker builds on these concepts for an ammonia release with somewhat more sophisticated treatment of the entrainment and heating processes. Eidsvik attempts to combine the denser-than-air and neutrally buoyant phases into one so that no matching needs to be done. Each of these models needs investigating before an assessment model for field application is selected.

6. <u>Suggestions for Application Initiatives</u>

- (i) Ensure that an operational instantaneous point source neutral-buoyancy gaussian model is available for field services use. Conduct training of AES personnel in its use and emphasize that the results will not be valid close to the source during the slumping phase either for concentration or cloud dimensions. The model becomes asymptotically correct at larger distances and probably gives a reasonable first guess at distances for which danger exists.
- (ii) Complete the evaluation of present assessment models, recommend and implement a model, or models, for practical assessment application. Chlorine, LNG and LPG spills should be priorities.
- (iii) Attempt to better define the nature and strength of the sources likely to be encountered.
- (iv) Clarify the "criteria for evacuation" for pollutants likely to be encountered.
- (v) Prepare an assessment guide and conduct training of AES personnel to aid them in assisting emergency authorities.

7. <u>Suggestions for AES Research Activities</u>

It would be imprudent at present, without having access to the more recent studies, particularly those at Porton, to make firm research recommendations for AES. One gets the distinct impression that considerable advances are in progress in this field.

It is clear that field studies of these phenomena require a major field research facility, such as the Suffield site, a major committment of funds and of man-years.

It is also clear that the dispersion of a heavy gas is asymptotically dominated by that of a neutrally buoyant gas. Even this simpler case is not well quantified and I personally urge we research the more fundamental case.

The major aspect of heavy gas dispersion which is not well understood <u>and</u> does not appear to now be under active study is that of the dispersion of residual gas from topographic hollows. I urge this as a prime candidate for AES research in this area.

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