NILU OPPDRAGSRAPPORT NR 32/78 REFERANSE: 25077 DATO: SEPTEMBER 1978

DISPERSION OF HEAVY GAS CLOUDS IN THE ATMOSPHERE

15

BY K.J. EIDSVIK

NORWEGIAN INSTITUTE FOR AIR RESEARCH P.O. BOX 130, 2001 LILLESTRØM NORWAY

LIST OF CONTENT

			Page
S	UMMARY		5
l	INTRO	DUCTION	7
2	A DIS	PERSION MODEL FOR A HEAVY GAS CLOUD	8
	2.1	Idealized source model	8
	2.2	Idealized vertical representation	10
	2.3	Gravity induced velocity	12
	2.4	Equation of state	13
	2.5	Heat transfere from the surface	13
	2.6	Entrainment of air	15
	2.6.1	Convective entrainment	15
	2.6.2	Mechanical entrainment	18
	2.6.3	Entrainment of air	19
	2.7	Enthalpy equation	21
3	APPLIC	CATION AND DISCUSSION	22
	3.1	Propane cloud	23
	3.1.1	Instantaneous release	23
	3.1.2	Continuous release	28
	3.2	Methane cloud	31
	3.3	Sensitivity analysis	35
4	CONCLU	JDING REMARKS	36
	REFERI	ENCES	39

SUMMARY

A simple model for the dispersion of heavy and cold gas clouds is developed. The horizontal dimension of the cloud is assumed to increase only due to the effects of gravity. The cold cloud is heated from below and from air entrainment at its upper boundary. The entrainment of air is estimated as for atmospheric inversions and density interfaces in laboratory flows.

The dispersion is predicted to be strongly dependent on environmental conditions, particularly the roughness of the underlying surface and the mean wind speed. Under unfavourable conditions a heavy gas cloud from a major release may be hazardous for hours and at distances as large as ten's of kilometers from the source.

0.131

DISPERSION OF HEAVY GAS CLOUDS IN THE ATMOSPHERE

1 INTRODUCTION

The production, transportation and storage of large quantities of heavy, explosive or poisonous gases may introduce hazards of unusual proportions to the public. A cloud of methane, propane or butane may be flammable even if the mean volume concentration is as low as 1%. A cloud of chlorine may be poisonous at much lower concentrations. If accidental release occur in unfavourable atmospheric flows, such clouds could be hazardous far away from the source.

The gas is often stored in liquified form. If this liquid is exposed to normal environments, the gas will boil off from a liquid pool at a boiling temperature that may be much lower than the environmental temperature. In this phase the gas density will be significantly higher than that of the atmosphere, and the gas cloud will spread horizontally due to gravity. As it does so, it may be heated from below, and turbulence will entrain air into the heavy cloud. After some time the gas mixture may obtain the same density as the air while still having a much lower temperature (methane). The heating from below will then continue. For other gases (butane, propane, chlorine) the density is higher than air at atmospheric temperature. For these, the heating from below will stop when atmospheric temperature has been obtained. Eventually, turbulence will dilute the gas so much that it becomes non-hazardous.

Numerous investigations have been done towards estimating the spread of such clouds. The following is a representative selection of reports: Fay (1), Fanneløp (2), van Ulden (3), te Riele (4), Germeles and Drake (5) and (6). Authors claim that their models explain experimental data, but they usually

- 7 -

have many adjustable coefficients available to fit curves to incomplete, sparce, and highly stochastic data. Experiments involving the release of heavy gases are described by Burges *et al.* (7), Feldbauer *et al.* (8), (9), and in (3). The quantitative physical understanding of the dispersion of nonpassive gases is modest.

2 A DISPERSION MODEL FOR A HEAVY GAS CLOUD

In principle, the problem of how a heavy and cold gas cloud behaves in the atmosphere is a question of the dynamics and irreversible thermodynamics of one turbulent flow in another. A reasonably complete and satisfactory description of this process would probably be extremely complicated. The aim is only to model the characteristics of the bulk properties of the process for the simplest environmental conditions. Some of the approximations used to obtain a simple description may seem unneccesarily crude. Their validity should, however, be judged relative to other more implicit approximations. The uncertainty of an estimate produced by any model for heavy and cold gas dispersion is most probably of the same order of magnitude as the estimate itself.

2.1 Idealized source models

If the characteristic time required to release most of the gas is t_r , the transverse (cross wind) dimension of the cloud at time t_r will be $r_r \simeq U_g t_r$. Here U_g is a characteristic radial speed during the release process. With a transport velocity U_a , which may be somewhat less than the atmospheric wind, the longitudinal (parallel to the wind) dimension of the cloud at the time t_r becomes: $(U_a+U_g)t_r$.

- 8 -

If it is assumed that physical processes important for the gas spreading, such as the heating of the cloud, have time scales significantly larger than t_r and that the cloud is approximately circular at time t_r , i.e. $U_g >> U_a$, the source is considered to be instantaneous. The instantaneous source model is thus more realistic under weak mean wind conditions.

When t_r is large or the transverse dimension r_r is much smaller than the longitudinal, the most important spread direction will be the longitudinal. For finite t_r the condition for this is $U_g << U_a$. In this case the source is said to be continuous. The continuous source model is thus the more realistic at high mean wind conditions. As this continuous source model only takes into account the spread due to the mean wind transport in the longitudinal direction, it overestimates the gas concentration of a release over a finite time interval.

Of all ways to release a given amount of gas, the instantaneous release results in the largest hazard distances, while a constant, continuous release over the time t_r results in the smallest hazard distances. The reason is that the transverse and vertical spread will be of the same order for the two clouds, while the longitudinal spread will be much larger for the continuous release. In this respect the instantaneous source idealisation overestimates and the continuous source idealisation under-estimates the hazard of a realistic release.

From a different point of view it may be stated that when the amount of gas and t_r are given, increasing atmospheric wind U_a makes the release less hazardous by making the source more like a continuous release.

2.2 Idealized vertical representation

It is assumed that the underlying surface is flat with roughness elements which are small compared to the vertical dimension of the gas mixture.

- 9 -

A general experimental experience is that the transition layer between a light fluid over a more dense fluid has a very small vertical dimension and is very persistent, even in the case of a significant mean velocity shear across the layer. It is therefore assumed that the flow adjusts its gradient Richardson number over the interface so that Kelvin Helmholz instability is almost always avoided.

The heavy gas cloud spreads horizontally by gravity. Because of the mean velocity shear and convection due to heating from below, this flow is probably a well developed turbulent flow. The turbulence is likely to produce a nearly homogeneous vertical distribution of most variables inside the cloud. This implies that the vertical dimension of the cloud, h, is well defined. The scale of the energy containing eddies in the bulk layer, is assumed to be of the same order as h both in the vertical and the horizontal direction.

As the horizontal dimension of the cloud, r, is much larger than h, it is sufficient to consider a representative vertical cross section of the flow. The characteristics of such a cross section are illustrated in Figure 2.1.



Figur 2.1: Assumed, representative vertical profiles of the heavy gas flow.

h Ul	-	characteristic	vertical dimension . mean velocity vector
ρ	-	19	mean density
T	-	11	mean temperature
Θ	-	19	fluctuating temperature
W	-	11	" vertical velocity

It is noticed that the atmospheric mean flow velocity, U_a, is likely to introduse horizontal gradients in the mean velocity of an instantaneously released gas cloud. Approximate symmetry with respect to the center of such a cloud is retained if either the atmospheric mean velocity and/or the momentum exchange across the interface is small.

If these concepts of the flow are realistic, the dispersion can not be realistically estimated by using K-theories of turbulence as done in references (4) and (6). In the bulk cloud there will be large vertical turbulent transports in spite of negligible mean gradients, and across the interface the turbulent transport may be small in spite of very large mean gradients. It would also be difficult to use numerical models of the Reynolds equations for mean flow computations as done in reference (6), because the vertical resolution of the grid would have to be very small, both near the underlying surface and near the density interface.

With the assumption of a well defined upper boundary of the gas mixture a definition of the mass, M(t), of the cloud can be given.

$$M(t) = M_{q} + M_{a}(t)$$
 (2.1)

Here M_g is the mass of gas released and $M_a(t)$ is the mass of entrained air. When the gas cloud is assumed to be approximately sylindrical and the characteristic entrainment velocity is denoted $\frac{dh_d}{dt}$ one has

$$\frac{dM_{a}(t)}{dt} = \pi \sigma_{a} r^{2} \left(\frac{dh_{d}}{dh} \right)$$
(2.2)

Equation (2.1) may then be written

$$\pi \rho(t) r^{2}(t) h(t) = M_{q} + M_{a}(t)$$
(2.3)

The corresponding equations for a continuous source release in a stationary flow field are obtained by considering the flux of gas mixture at a distance x from the source.

$$U\rho(x)2r(x)h(x) = \frac{dM_g}{dt} + \frac{dM_a}{dt}$$
(2.4)

The transverse dimension of the plume is 2r, $\frac{dM_g}{dt}$ is the source strength and $\frac{dM_a}{dt}$ is the entrained air over the distance x. We have

$$\frac{dM_{a}}{dt} = 2\rho_{a} \int_{0}^{x} r(x) \left(\frac{dh_{d}}{dt}\right) dx$$

or

$$\frac{d}{dx}\left(\frac{dM_{a}}{dt}\right) = 2\rho_{a}r(x)\left(\frac{dh_{d}}{dt}\right)$$
(2.5)

2.3 Gravity indused velocity

The frontal speed of a heavy gas cloud has been estimated by several authors (1,2,3,5), to be

$$\frac{\mathrm{d}r}{\mathrm{d}t} = U_{\mathrm{g}} = \alpha_1 \left(\mathrm{gh} \frac{\Delta\rho}{\rho} \right)^{\frac{1}{2}}$$
(2.6)

Here α_1 is a coefficient $\alpha_1 \in (1,\sqrt{2})$, g is the gravity acceleration and $\Delta \rho = \rho - \rho_a$. When there are no horizontal variations of h and ρ , the radial gravity induced horizontal speed will vary linearly with the distance from the center of both a circular cloud and a plume. The average gravity speed over a circular cloud and a plume then becomes respectively, $\frac{2}{3} U_g$ and $\frac{1}{3} U_g$. By choosing $\alpha_1=1$, the velocity given by equation (2.6) is thus representative for the gravity induced flow of the bulk cloud. This small value of α_1 will result in a small underestimate of r, the area of the upper surface of the cloud and thus the entrainment of air.

2.4 Equation of state

Initially, the heavy, cold gas mixture is not in thermodynamic equilibrium. With all the inaccuracies involved in this problem, we nevertheless assume that it obeys the laws of an ideal gas. The equation of state for the gas mixture is thus:

$$\rho = \frac{p}{RT}$$
(2.7)

The pressure, p, is asumed to be constant. When the universal gas constant is R_* the "gas constant" for a gas with molecular weight m_i is $R_i = R_*/m_i$. For a mixture of two ideal gases we have, for an instantaneous source.

$$R(t) = \frac{M_{g}R_{g} + M_{a}(t)R_{a}}{M_{g} + M_{a}(t)}$$
(2.8)

For a continuous source it is

$$R(x) = \frac{\frac{dM}{dt}g_{R} + \frac{dM}{dt}(x)R_{a}}{\frac{dM}{dt} + \frac{dM}{dt}(x)}$$
(2.9)

2.5 Heat transfer from the surface

Since there is an imposed gravity flow field, U_g , we assume that the heat transfer from the underlying surface may be approximated by the laws of forced convection heat transfer, as described by for instance Welty *et al.* (10). The heat transfer per unit area is proportional to the difference between the surface temperature, set equal to T_a , and the gas temperature.

$$\frac{\delta q}{dt} = \kappa \left(T_{a} - T \right) \tag{2.10}$$

$$\kappa = \mathrm{St} \cdot \rho \cdot c_{\mathrm{p}} \cdot \mathrm{U} \tag{2.11}$$

U is the free stream velocity.

The Reynolds analogy, stating that the coefficients of heat and momentum transfer are approximately equal, implies that the Stanton number can be roughly estimated as shown in reference (10).

St
$$\simeq \frac{\frac{1}{2}c_{f}}{1+5(\frac{1}{2}c_{f})^{\frac{1}{2}}(Pr-1)}$$
 (2.12)
 $\simeq \frac{1}{2}c_{f}$

Here c_f is the drag coefficient, given as twice the quadratic ratio of the friction and the free stream velocities.

$$c_{f} = 2\left(\frac{u_{k}}{U}\right)^{2}$$
(2.13)

Kitaigorodskii (11) has estimated $c_f \simeq 2 \cdot 10^{-3}$ to be a representative value over water. Over a smooth land surface it is significantly larger, say $2 \cdot 10^{-2}$.

The heat transfer from the surface must be equal to the turbulent enthalpy flux near the surface so that

$$(\overline{\Theta W})_{0} = \frac{1}{2} c_{f} U(T_{a} - T)$$
(2.14)

For heat transfer and entrainment estimation, the free stream velocity is approximated as, for instantaneuous releases

$$U = \max \begin{cases} U_{g} \\ U_{a} \end{cases}$$
(2.15)

and for continuous releases

$$U = U_{a}$$
(2.16)

For the purpose of this report, U_a is the mean wind velocity, considered to be constant with height.

2.6 Entrainment of air

A characteristic feature of the initial flow of a heavy gas cloud seems to be a vortex ring at its front. This vortex is probably essential to the initial entrainment of air near the peripherial boundary. After some time, however, most of the entrainment must occur at the large upper surface of the cloud. This is most probably the entrainment which eventually makes the cloud non-hazardous, and the only entrainment considered in the present model.

2.6.1 Convective entrainment

Tennekes (12) and Heidt (13), among others, have discussed the entrainment rate across an atmospheric inversion above a free convection layer. Tennekes assumes that the downward enthalpy flux near the inversion $\rho c_p(\overline{\theta w})_h$ is equal to the enthalpy loss of the newly entrained air. For a sufficiently large water vapor mixing ratio χ_W , condensation will release the latent heat of evaporation, L. For large temperature differences the latent heat of sublimation may be released. The enthalpy loss of the newly entrained air is then

$$\begin{bmatrix} \sigma_{2a}(T_a - T) + L\chi_w H(T_{da} - T) \end{bmatrix} \rho_a \frac{dh_d}{dt}$$

This gives

$$\begin{bmatrix} c_{pa}(T_{a}-T) + LX_{w}H(T_{da}-T) \end{bmatrix} \rho_{a} \frac{dh_{d}}{dt} = \div \rho c_{p}(\overline{0w})_{h}$$
(2.17)

 $H(T_a-T)$ is the Heavyside generalized unit function. T_{da} is the dew point temperature. This is a closure equation. For simplicity, the difference between $\rho_{a}c_{pa}$ and ρ_{c} is neglected in this connection. Equation (2.17) is then reduced to

$$(\overline{Ow})_{h} = \div \left[(T_{a}-T) + (c_{pa})^{-1} LX_{w} H (T_{da}-T) \right] \frac{dh_{d}}{dt} (2.18)$$

The mixing ratio of water vapour is obtained from the Clausius-Clapeyron equation

$$X_{\rm W} \simeq 3.7 \cdot 10^{-3} \exp\left[\frac{L}{R_{\rm W}} \left(\frac{1}{273} - \frac{1}{T_{\rm da}}\right)\right]$$
 (2.19)

Approximately, $L/c_{pa} \simeq 2.5 \cdot 10^3$ deg. Although χ_w is normally very small, the moisture term can not generally be neglected in equation (2.18).

In estimating the turbulent velocities, the effects of latent heat release near the upper boundaries of the gas cloud is not explicitely taken into account. The convective entrainment velocity $\left(\frac{dh_d}{dt}\right)_T$ is estimated with formally the same equations as used for dry convection. By considering the turbulent energy

equation in the neighbourhood of the inversion, Tennekes (14) estimate that

$$-\left(\overline{\Theta w}\right)_{h} = \alpha_{2} \frac{T}{g} \frac{w_{T}^{3}}{h}$$
(2.20)

where α_2 is a coefficient of the order one and $w_{\rm T}$ is a characteristic convective turbulent velocity for the bulk of the layer

$$w_{\rm T}^3 = \alpha_3 \left(\overline{\Theta w}\right)_0 \frac{gh}{T}$$
(2.21)

 α_3 is a coefficient of the order 0.1. Equation (2.21) illustrates the difficulty in defining a well posed problem when the gas mixture is cold and has the same density as air. In this case $(\overline{\theta w})_0$ is finite, while it is difficult to assign a value to h.

For a well defined h, equation (2.20) and (2.21) give

$$-(\overline{\Theta w})_{h} = \alpha_{2}\alpha_{3}(\overline{\Theta w})_{0}$$

The convective entrainment rate is then, as obtained from equation (2.18) with $\chi_{\rm w}=$ 0.

$$\left(\frac{dh_{d}}{dt}\right)_{T} = \alpha_{4} \left(T_{a} - T\right)^{-1} \left(\overline{\Theta w}\right)_{0}$$
(2.22)

The coefficient $\alpha_4 = \alpha_2 \cdot \alpha_3$ is approximately equal to 0.2 (12).

Accepting the inconsistency of estimating the entrainment rate as if the flow were a free convection flow, and estimating the heat transfer as if it were governed by forced convection, the equations (2.14) and (2.22) give:

$$\left(\frac{dh_{d}}{dt}\right)_{T} = \alpha_{4} \left(\frac{1}{2}c_{f}\right) \cdot U$$
$$= \alpha_{4} \left(\frac{1}{2}c_{f}\right)^{\frac{1}{2}} \cdot u_{*}$$

This is the same type of entrainment equation as used in reference (5). However, their "entrainment coefficient" of 0.1 is much higher than the present $(\alpha_4 \ \frac{1}{2}c_f)$. Their choice of "entrainment coefficient" and Stanton number (10^{-3}) implies that the characteristic turbulent velocity at the interface is higher than in the bulk cloud. This seems to be physically inconsistent with the existence of a density interface.

2.6.2 Mechanical entrainment

When there is little heat transfer, the entrainment rate is estimated as for density interfaces in laboratory flows. In these flows the turbulence is of mechanical origin. Different authors, as Kato and Phillips (15), Crapper and Linden (16), Wu (17) and Long (18,19), estimate the entrainment somewhat differently, but the following relation appears to be representative

$$\left(\frac{dh_{d}}{dt}\right)_{m} = \alpha_{5} \frac{w_{m}^{3}}{(gh\frac{\Delta\rho}{\rho})} = \alpha_{5} \alpha_{1}^{2} \left(\frac{1}{2}c_{f}\right)^{3/2} \frac{U^{3}}{U_{g}^{2}}$$
(2.24)

The coefficient α_5 is estimated to $\alpha_5 \in (0.2, 2.5)$. As there probably will be a large velocity shear across the density interface, a large entrainement coefficient $\alpha_5 = 2.5$ is chosen. w_m is a characteristic mechanical turbulent velocity, in the present case the friction velocity.

 $w_{\rm m} = (\frac{1}{2}c_{\rm f})^{\frac{1}{2}}U$ (2.25)

(2.23)

It is noted that if the molecular weight of the two gases were equal and if the characteristic turbulent velocity w_m is set equal to w_T in equation (2.20), the expressions (2.23) and (2.24) become identical.

2.6.3 Entrainment of air

The velocity scales relevant to the mixing inside the cloud are w_m and w_T . In the case of small wind velocity, the atmospheric turbulence level is supposed to be small relative to the largest of these two velocities. In the case of higher wind velocities w_m is set equal to the atmospheric friction velocity. The ratio of the two velocity scales is obtained from the equations (2.14), (2.21) and (2.25).

$$\frac{w_{\rm T}}{w_{\rm m}} = \frac{\left(\alpha_3 \frac{1}{2}c_{\rm f} U \, {\rm gh} \, \frac{\Delta T}{T}\right)^{1/3}}{\left(\frac{1}{2}c_{\rm f}\right)^{\frac{1}{2}} U}$$

Assuming, in this connection, a constant molecular weight gives:

$$\frac{w_{\rm T}}{w_{\rm m}} \simeq \alpha_1^{-\frac{2}{3}} \alpha_3^{-\frac{1}{3}} (\frac{1}{2}c_{\rm f})^{-\frac{1}{6}} \left(\frac{U_{\rm g}}{U}\right)^{\frac{2}{3}}$$
(2.26)

With $\alpha_1 = 1$, $\alpha_3 = 0.2$ and $c_f = 2 \cdot 10^{-3}$ this becomes

$$\frac{W_{\rm T}}{W_{\rm m}} \simeq 2 \left(\frac{U_{\rm g}}{U}\right)^{2/3}$$
(2.27)

For small wind velocity, U_a , equation 2.15 gives $U=U_g$ so that $w_T > w_m$. For larger wind velocities, however, $U>U_g$, so that the two velocity scales may be more equal.

The ratio of the two entrainment velocities is analogously obtained from the equations (2.23) and (2.24).

$$\frac{\left(\frac{dh_{d}}{dt}\right)_{T}}{\left(\frac{dh_{d}}{dt}\right)_{m}} \simeq \alpha_{1}^{2} \alpha_{4} \alpha_{5}^{-1} \left(\frac{L}{2}C_{f}\right)^{-\frac{1}{2}} \left(\frac{U}{U}\right)^{2}$$
(2.28)

With $\alpha_1=1, \alpha_4=0.2, \alpha_5=2$ and $c_f=2\cdot 10^{-3}$ this becomes

$$\left(\frac{dh}{dt}\right)_{T} \simeq 3\left(\frac{U}{U}\right)^{2}$$
(2.29)

Again for small wind velocities $U=U_g$ so that $\left(\frac{dh_d}{dt}\right)_T > \left(\frac{dh_d}{dt}\right)_m$ For larger wind velocities the inequality is reversed.

The existence of more than one velocity scale is a difficulty. It seems physically reasonable that the mean wind shear and mechanical turbulence will destroy the organized vertical plumes of termal convection and thus make the entrainment less effective. However, little quantitatively is known about this. While waiting for a satisfactory theory on the interaction between convective and mechanical entrainment, the entrainment velocity is, in this report, estimated as follows.

(2.30)

When $\Delta \rho$ or h becomes small enough, the vertical dispersion is assumed to behave as vertical diffusion with a finite velocity described by Monin and Yaglom (20). The transition from entrainment dominated vertical dispersion to dispersion dominated by "normal" atmoshperic turbulence given in equation (2.30), may be written

$$\left(\frac{\mathrm{d}\mathbf{h}_{\mathrm{d}}}{\mathrm{d}\mathbf{t}} \right)_{\mathrm{m}} \geq \mathbf{w}_{\mathrm{m}}$$

$$U_{g} < \alpha_{1} \alpha_{5}^{\frac{1}{2}} (\frac{1}{2}c_{f})^{\frac{1}{2}} U_{a} \simeq u_{*}$$
 (2.31)

In the horizontal direction, it is the energy content of the atmospheric eddies of the same dimensions as the gas cloud that can contribute most to the lateral dispersion of the cloud. The effect is significant only if there is a sufficient vertical momentum exchange across the density interface. U_g is automatically a velocity associated with the most efficient scale. Transition from gravity dominated horizontal spread to horizontal dispersion dominated by atmospheric turbulence, should also in this case be a relation between U_g and the turbulence, not the mean wind as commonly assumed. The condition (2.31) should therefore be reasonable for transition to dispersion dominated by atmospheric turbulence is dispersion dominated is usually not fulfilled in the computations. I.e. The spread is, in the present model, dominated by gravity effects over the most interesting parts of the phase space.

2.7 Enthalpy equation

Provided that there is no turbulent dissipation, there is no heat transfer to a material particle inside the cloud. As the pressure is assumed to be constant, the individual time derivative of the instantaneous temperature, (T + 0), must therefore be zero.

$$\frac{D(T+\theta)}{dt} = 0 \tag{2.32}$$

With a nearly incompressible turbulent flow and small variations of mean values along horizontal coordinates, this gives the usual equation for mean temperature variations

$$\frac{\partial \mathbf{T}}{\partial t} = -\frac{\partial}{\partial z} (\overline{\mathbf{0}\mathbf{w}})$$
$$= \frac{1}{h} \left[(\overline{\mathbf{0}\mathbf{w}})_0 - (\overline{\mathbf{0}\mathbf{w}})_h \right]$$
(2.33)

We now have a closed set of equations to describe the state of the heavy gas cloud.

3 APPLICATION AND DISCUSSION

The equations developed are ordinary, but nonlinear differential equations expected to be integrable as long as $\rho(t) > \rho_a$ and $T \leq T_a$. We have not been able to find analytical solutions and have used the Runge-Kutta-Mersion method for integration. The method is described by, for instance, Skjeldestad (21). The integration is carried out as long as $\Delta \rho / \rho > 10^{-3}$.

The following variables are kept constant: $g = 10 \text{ ms}^{-2}$ $m_a = 29 \text{ g/mol}$; $\rho_a = 1.3 \text{ kg m}^{-3}$; $T_a = 283$; $R_* = 8.3 \text{ J/mol} \cdot \text{deg}$; $L = 2.5 \cdot 10^6 \text{ J/kg}$. When not explicitely stated differently, the experimental coeffecients are chosen as: $\alpha_1 = 1$, $\alpha_4 = 0.2$, $\alpha_5 = 2.5$, the friction coefficient as $c_f = 2 \cdot 10^{-3}$ and the moisture as $\chi_w = 0$. Differences of the dispersion caused by varying initial cloud shape is predicted to vanish rapidly. We therefore present results only for r(0) = 2h(0).

- 22 -

3.1 Propane cloud

With a molecular weight of $m_g = 45$ g/mol and initial temperature T(0) = 230, the model should describe the development of a propane cloud released at boiling temperature.

3.1.1 Instantaneous release

The state of an instantaneously released cloud is given by the equations (2.2, 2.3, 2.6, 2.7, 2.8, 2.12 2.14, 2.15, 2.18, 2.19, 2.23, 2.24, 2.25, 2.30 and 2.33).

As the instantaneous source model is most realistic under low wind conditions, U_a is set equal to a representative, small atmospheric wind velocity of 0.5 ms⁻¹. The general properties of the solution are shown in Figure 3.1.

The cold cloud falls rapidly to a very small vertical dimension. The time which corresponds to minimum height is approximately equal to $t = t(T=T_a)$. At this state there is still a significant density difference between the gas mixture and the air. However, the gravity fall is here balanced and later dominated by air entrainement.

The figure shows that as the mixing ratio, χ , approaches $10^{-2} = 1$ % we have approximately

$$\begin{bmatrix} \Delta T \\ T \end{bmatrix} \begin{pmatrix} \Delta \rho \\ \rho \end{pmatrix}, \chi \end{bmatrix} \propto \begin{cases} t^{-(5/2^{+})} & \text{for } M_{g} = 5 \cdot 10^{2} \text{kg} \\ t^{-(3/2^{+})} & \text{for } M_{g} = 5 \cdot 10^{6} \text{kg} \end{cases}$$
(3.1)

and over most of the time interval

$$r(t) \propto t^{\frac{1}{2}}$$
 (3.2)

Here 5/2+ means a number which is slightly larger than 5/2. Both r(t) and $\chi(t)$ vary with time much like the dimension and concentration of passive scalar clouds in atmospheric turbulence. The rapid increase of h as $\chi \rightarrow 1$ % is consistent with equation (3.2) and (2.24). As $\chi \rightarrow 1$ %, U=U_a so that equation (2.24) may be written

$$\left(\frac{dh_{d}}{dt}\right)_{m} \qquad \propto U_{a}^{3} \left(\frac{dr}{dt}\right)^{-2} \qquad \propto U_{a}^{3} t^{1}$$

Integration then gives

$$h_{d}(t) \propto U_{a}^{3} t^{2}$$
(3.3)



Figure 3.1: The development of an instantaneously released propane cloud. Heavy curves: $M_g = 5 \cdot 10^6 kg$. Thin curves: $M_g = 5 \cdot 10^2 kg$; $U_a = 0.5 ms^{-1}$, $c_f = 2 \cdot 10^{-3}$. $\alpha_4 = 0.2$, $\alpha_5 = 2.5$.

If the vertical spread had been governed by $w_{\rm m}$ in equation (2.30), one would have had

$$h_{d}(t) = (\frac{1}{2}c_{f})^{\frac{1}{2}} U_{a}t$$
 (3.4)

The lower flammable limit, LFL, is located at a volume concentration of approximately 5%. With turbulent concentration fluctuations of the order of 5-10 superposed on the mean value, there is a reasonably large probability of ignition even at a mean mixing ratio of 1%. This mean mixing ratio or mass concentration is taken as representative of the outer boundary of ignition hazard. Figure 3.2 shows the estimated time to and radius of a propane cloud at $\chi = 1$ %. In a real cloud there will be areas with less motion relative to the ground or air than in the bulk cloud. These areas will experience less entrainment than the rest of the cloud and thus have a high x. The horizontal mixing inside the cloud is probably slow because the eddies only have a horizontal dimension of the order of h. This probably implies that the time of potential hazard is underestimated by $t(\chi=1\%)$. The same reasons suggest that $r(\chi=1\%)$ overestimates the hazard radius at the time t(x=1%).

As seen from figure 3.2 the estimated hazard time and radius increase with released mass approximately like

$$t(\chi=1\%) \propto M_{q}^{1/3}$$
 (3.5)
 $r(\chi=1\%) \propto M_{q}^{2/5}$ (3.6)

(3.6)

rough estimate of the cloud height at the state,
$$\chi = 1$$
°,

is obtained from the definition of χ and equation (2.3)

Λ

$$h(\chi=1\%) = \frac{10^2 M_g}{\pi \rho(\chi=1\%) r^2(\chi=1\%)}$$

- 25 -



Figure 3.2: The time until and radius at 1% mixing ratio. Instantaneous release of propane. Heavy curves: $c_f = 2 \cdot 10^{-3}$. Thin curves: $c_f = 2 \cdot 10^{-2}$; $U_a = 0.5 \text{ ms}^{-1}$, $\alpha_{4} = 0.2$, $\alpha_5 = 2.5$.

or in conventional units

$$h(\chi=1\%) \simeq 25 \frac{M_g}{r^2(\chi=1\%)}$$
 (3.7)

With the use of equation (3.6) this gives

$$h(\chi=1%) \propto M_{g}^{1/5}$$
 (3.8)

The height of the cloud at $\chi = 1$ % is thus predicted to increase very slowly with the mass released. Both $t(\chi=1\%)$, $r(\chi=1\%)$ and $h(\chi=1\%)$ vary significantly with c_f which characterize the state of the underlying surface. Figure 3.3 show that $t(\chi=1\%)$ and $r(\chi=1\%)$ also vary significantly with the wind velocity. The maximum distance to ignition hazard, $x(\chi=1\%)$, is of the order



Figure 3.3: "Hazard" time $t(\chi=1\%)$ radius, $r(\chi=1\%)$ and distance $x(\chi=1\%)$ for an instantaneous release of $M_g = 5 \cdot 10^4 \text{kg}$ propane as functions of wind speed. $c_f = 2 \cdot 10^{-3}$, $\alpha_4=0.2$, $\alpha_5=2.5$.

Figure 3.3 indicates that $x(\chi=1\%)$ reaches a maximum at small transport velocities. This appears to be physically reasonable. Also $x(\chi=1\%)$ decreases rapidly with increasing values of c_{f} .

A water vapour content characterized by a dew point depression of 1 deg at a temperature of 10° C is predicted to affect the above results very little. The temperature rise of the cloud is somewhat faster, but it is fast also without moisture. It

has been indicated that the moisture must be important in certain equations over some time intervals. However, for the dispersion (which is an integral process) over a long time interval, it is predicted not to be important at atmospheric temperatures below 10° C.

3.1.2 Continuous release

When the variables are expressed as functions of $x=U_at$, the state of a continuously released propane cloud is described by the equations (2.4, 2.5, 2.6, 2.7, 2.9, 2.12, 2.14, 2.16, 2.18, 2.19, 2.23, 2.24, 2.25, 2.30 and 2.33). As shown in Figure 3.4, the general properties of the solution is analogous to the instantaneous source case.

The gravity fall of the plume dominates only at distances from the source that are not significantly larger than the transverse horizontal dimension of the cloud. After a short distance, the height of the cloud increases rapidly. As $x \rightarrow x(\chi=1\%)$ one has approximately

$$\left[\frac{\Delta T}{T}, \frac{\Delta \rho}{\rho}, X\right] \propto x^{-(2+)}$$
(3.10)

Over most of the interesting downwind distance, the transverse dimension obeys the approximate relation.

 $r(x) \propto x^{2/3+}$ (3.11)

The variation of χ and r with distance from the source thus appears to be almost similar to the variation of the maximum concentration and dimension of passive scalar plumes in atmospheric turbulence.



Figure 3.4: The downwind development of a continuously released propane cloud. $\frac{dM}{dt} = 50 \text{ kg s}^{-1}$, $U_a = 5 \text{ ms}^{-1}$, $c_f = 2 \cdot 10^{-3}$, $\alpha_4 = 0.2$, $\alpha_5 = 2.5$.

The rapid increase of h as $\chi \rightarrow 1\%$ is again to be understood by means of the entrainment equation (2.24), which for a continuous release reads

$$\frac{dh_{d}}{dx} = \alpha_{5} \alpha_{1}^{2} \left(\frac{1}{2} c_{f} \right)^{3/2} \left(\frac{dr}{dx} \right)^{-2}$$
(3.12)

With a r(x)-variation as given by equation (3.11). This gives

$$h_{a}(x) \propto x^{5/3}$$
; (3.13)

- 29 -

The maximum hazard distance is shown in Figure 3.5. The distance, $x(\chi=1\%)$, and the transverse dimension, $r(\chi=1\%)$, increase with released mass approximately like

$$x(\chi=1\%) \propto \left(\frac{dM_{g}}{dt}\right)^{\frac{1}{2}+}$$
(3.14)
$$r(\chi=1\%) \propto \left(\frac{dM_{g}}{dt}\right)^{\frac{2}{3}+}$$
(3.15)



Figure 3.5: Distance to and transverse dimension at 1% mixing ratio. Continuous release of propane. $c_f = 2 \cdot 10^{-3}$, $\alpha_4 = 0.2$, $\alpha_5 = 2.5$. $\dots : x(\chi=1\%)$, $\dots : r(\chi=1\%)$,

Suppose that $5 \cdot 10^{\circ}$ kg were released over a time interval of $t_r = 5 \text{ min}$, giving a source strength of $1.7 \cdot 10^2$ kg/s. Figure (3.3) and (3.5) may then be used to compare the hazard distances for instantaneous and continuous releases. It is observed that the instantaneous release gives the highest hazard distance, as it should. However, the difference is not remarkably large. As the hazard distance from any release over the time t_r must lie between the two, it does not seem important, for the purpose of hazard distance estimation, to model the release process in great detail.

3.2 Methane cloud

With a molecular weight of $m_g = 16$ g/mol and initial temperature T(0) = 113 the model should describe the development of a methane cloud released at boiling temperature. The general properties of an instantaneously released cloud is shown in Figure 3.6.

The development of a methan cloud is very rapid. At the stage when $\rho = \rho_a$, the temperature of the gas mixture is still as cold as approximately $-100^{\circ}C$.

The time when the densities of cloud and atmosphere become equal, $t(\rho=\rho_a)$, and the cloud radius at this stage, $r(\rho=\rho_a)$, are shown as functions of released mass in Figure 3.7. At the temperature $T_a = 10^{\circ}C$, these curves are not much affected by water vapour.



Figure 3.6: The state of an instantaneously released methane cloud. Heavy curves: $M_g = 5 \cdot 10^6 \text{kg}$. Thin curves: $M_g = 5 \cdot 10^2 \text{kg}$ $U_{\alpha} = 0.5 \text{ ms}^{-1}$, $c_f = 2 \cdot 10^{-3}$, $\alpha_4 = 0.2$, $\alpha_5 = 2.5$.

It is observed that the relationships between $t(\rho=\rho_a)$, $r(\rho=\rho_a)$ and M_q are approximately

 $t(\rho = \rho_a) \propto M_q^{1/5}$ (3.16)

$$r(\rho = \rho_a) \propto M_g^{1/3}$$
 (3.17)

 $t(\rho=\rho_a)$ varies remarkably little with released mass. It varies much with the mean wind. For a wind of 5 ms⁻¹, a friction coefficient of $2 \cdot 10^{-3}$ and a mass of $5 \cdot 10^{-4}$ kg, it turns out that $t(\rho=\rho_a)$ is as small as 45 sec.



Figure 3.7: The time for an instantaneously released methane cloud to reach atmospheric density and the cloud radius at this instant. Heavy curves: $c_f = 2 \cdot 10^{-3}$. Thin curves: $c_f = 2 \cdot 10^{-2}$; $U_a = 0.5 \text{ ms}^{-1}$, $\alpha_u = 0.2$, $\alpha_5 = 2.5$.

The gas mixture is still hazardous at this stage. It is obvious that the model is not applicable for the subsequent cloud development. As the cloud is very cold, there is still convective activity and the further development of the cloud may not even be a well posed problem (equation 2.21). It seems reasonable to assume that the dilution of the cloud will be more rapid than turbulent atmospheric dispersion of pasive scalars from an instantaneous area source with a source strenth given as $M_g/\pi r^2 (\rho = \rho_a)$ or an "initial" concentration distribution $\chi(\rho = \rho_a)$. This dispersion has been discussed in the classic literature on turbulent diffusion. For the purpose of a rough outline of this, the mixing ratio is estimated to be of the order

$$\chi(t) = \frac{M_g}{\pi \rho_a r^2 (\rho = \rho_a) \left[h(\rho = \rho_a) + w \cdot (t - t(\rho = \rho_a))\right] - M_g}$$
(3.18)

Here w is the characteristic vertical turbulent velocity. As the computations suggest that $M_g >> M_a(\rho = \rho_a)$ and $h(\rho = \rho_a) << w [t(\chi=1\%) - t(\rho = \rho_a)]$, equation (3.18) may be written

$$\chi(t) \simeq \frac{h(\rho = \rho_a)}{w \cdot [t - t(\rho = \rho_a)]} \text{ for } \chi(t) = 0(1\%)$$
(3.19)

The hazard time is thus of the order

$$t(x = 1\%) \approx t(\rho = \rho_a) + 10^2 \frac{h(\rho = \rho_a)}{w}$$
 (3.20)

Most of the experiments on the spread of "heavy" gases have been done with methane (7, 8, 9). The mass released has always been small. If the present model is realistic, the time or distance along the wind until $\rho = \rho_a$ is then very small. With the coarse spatial resolution of the recording locations, it may then be more relevant to consider these experiments as "convective" dispersion experiments from a large area source (line source for continuous releases). The large concentration fluctuations observed in these experiments could be an indication of the same.

3.3 Sensitivity analysis

The variation with α_5 is, as indicated in section 2.6.3, expected to be most important for $U_a>U_g$ (continuous source). The most dominant variation appears in the state variable $\chi(x)$. Figure 3.4 shows that the hazard distance may increase a factor of approximately 2 when α_5 is decreased a factor of 10. The variation with α_4 is, as indicated in section 2.6.3, expected to be most important for $U_a < U_g$ (instantaneous source). Again $\chi(t)$ is the state variable that show the most dominant effect. Figure 3.6 indicates that a methane cloud remains denser than air somewhat longer when α_4 is increased by a factor of two.

Table 3.1 indicates that the variation of the gas dispersion with the coefficient α_1 is also reasonably small. As expected, $t(\chi=1\%)$ decreases somewhat with increasing values of α_1 .

Table 3.1: Variation of hazard variables with the coefficient α_1 . Instantaneous release of $5 \cdot 10^4$ kg propane. $U_a = 2 \text{ ms}^{-1}, c_f = 2 \cdot 10^{-2}.$

α1	t(χ=1%)	x(χ=1%)	r(x=1%)	h(χ=1%)
1.0	370 s	l.l km	380 m	8 m
1.4	290 s	1.0 km	400 m	8 m

It thus seems that uncertainties about numerical coefficients are not essential for estimation of the spread of heavy gases.

The variation with the initial shape of the cloud was investigated for an instantaneous release of $5 \cdot 10^4$ kg propane. It turns out that differences between two clouds characterizized by r(0)=2h(0) and r(0) = 20 h(0) vanished very rapidly. The hazard parameters $t(\chi=1\%)$, $r(\chi=1\%)$ are predicted to be approximately independent of the initial shape of the cloud.

The dispersion of different heavy gases instantaneously released at their boiling temperature is illustrated in Figure 3.8. The figure suggests that the dispersion will only vary slightly from one heavy gas to another. - 36 -





The large variation of gas spread occurs with $c_{f'}$ which parameterizes the state of the underlying surface and with U_a , which parameterizes the state of the atmosphere. Varying environmental conditions are thus predicted to affect the spread of heavy gases significantly.

4 CONCLUDING REMARKS

One has tried to describe the dynamics of the most essential state variables of a heavy gas cloud as simply as possible. Different refinements of some approximations are obvious, but the design of a consistent and significantly more realistic model seem to introduce complications. It turns out that some aspects of the model appear to be unrealistic i): The initial development of a cloud may be so rapid compared to the time scale, t_r , of a realistically generated cloud that the two processes, release and spreading, should be modelled simultaneously. ii): At its minimum, the height of the cloud is not significantly larger than the typical vertical dimension of a realistic density interface and the dimension of the lowest surface layer characterized by large mean flow gradients and surface obstacles. In a realistic release and initial spreading process there will probably be larger air entrainment than in the present model. Surface obstacles, such as sea waves, of a height comparable to the cloud height would probably also increase the entrainment. It appears, therefore, that the height of a heavy gas cloud is underestimated in this model.

Otherwise, the model appears to be attractive: The results do not depend critically upon uncertainties about numerical coefficients. It the opposite were true, the model would only provided a complicated computation of an uncertain result. This, and the opinion that the model is based on reasonable physics, suggests that if it, or a refined analogous model, could explain comprehensive experimental data on small releases of gas, it would probably serve to predict quite accurately the spread of large (accidental) gas releases too. The dependence between state variables of particular relevance to potential hazard, t(x=1%), $x(\chi=1\%)$ and $r(\chi=1\%)$, is predicted to increase less than linearly with the mass released.

The potential hazard is predicted to vary significantly with the state of the underlying surface and atmosphere. The statistics of hazard is therefore closely related to the statistics of environmental states. In particular will the accuracy of time prediction of the potential hazard following an accidental release of gas be highly dependent upon the accuracy of time prediction of actual environmental conditions. The large variation of gas dispersion with the state of the environments and in addition the fact that these states are highly stochastic, suggest that a sufficient number of comprehensive diffusion experiments on heavy gas dispersion would be very expensive. It therefore seems that hazard estimation, at least in the next decade, must be based on poorly verified, incomplete theoretical models of heavy gas spread. However, the more critical aspects of our model could probably be verified, modified or rejected by means of reasonably inexpensive experiments. The aspects are the assumption of approximate uniform spatial distribution of concentration inside the cloud and the cloud height variation with time and downwind distance. 5 REFERENCES

(1) Fay, V.A. Unusual fire hazard of LNG tanker spills. Combust. Sci. Technol., 7, 47-49 (1973) (2) Fanneløp, T.K. Faremomenter ved sjø-transport av LNG. Trondheim, Institutt for aero- og gassdynamikk NTH, 1974. (in Norwegian). (3) van Ulden, A.P. On the spreading of a heavy gas released near the ground. I: 1st international loss prevention symposium. The Hague/Delft, Netherlands 28-30 May 1974. Amsterdam, Elsevier, 1974, pp. 221-226, 431-439. (4) te Riele, P.H.M. Atmospheric dispersion of heavy gases emitted at or near ground level. I: 2nd international loss prevention symposium. Heidelberg 6-9 Sept. 1977, pp 347-357. Gravity spreading and atmospheric (5) Germeles, A.E. Drake, E.M. dispersion of LNG vapour clouds. I: The 4th int. symposium on transport of hazardous cargoes by sea and inland waterways. Jacksonville, Florida, 1975, pp. 519-539. LNG terminal risk assessment study (6) for point conception, California. La Jolla, Calif., Science Application, 1976. Hazards of spillage of LNG into (7) Burges, D. water. Pittsburgh, Pa. Bureau of Biordi, J. Mines, 1972. (PMSRC Rep.no 4177). Murphy, J. Spills of LNG on water -"Vaporization (8) Feldbauer, G.F. & downwind drift of combustible Heigl, J.J. Mc Queen, W. mixtures". Mag, W.G. Florham Park, Esso Research & Engineering Company, 1972. Report no EEGIE-72.

(9)		LNG safety program interim report on phase II work. Columbus, Ohio, Battelle, 1974. (American Gas Association Project IS-3-1).
(10)	Welty, J.R. Wilks, C.E. Wilson, R.E.	Fundamentals of momentum, heat and mass transfer. New York, Wiley, 1969.
(11)	Kitaigorodskii, S.A.	The analysis of air-sea interaction. Jerusalem, Israel program for scientific translations, 1973.
(12)	Tennekes, H.	A model for the dynamic of the inversion above a convective boundary layer. J.Atm. Sci. 30, 558-567 (1973).
(13)	Heidt, F.D.	The growth of the mixed layer in a stratified fluid due to penetrative convection. Boundary-Layer Met. <u>12</u> , 439-461 (1977).
(14)	Tennekes, H.	Free convection in the turbulent Ekman layer of the atmosphere. J. Atmos.Sci. 27, 1027-1034 (1970).
(15)	Kato, H. Phillips, O.M.	On the penetration of a turbulent layer into a stratified fluid. J. Fluid Mech. 37, 643-655 (1969).
(16)	Crapper, P.E. Linden, P.F.	The structure of density interfaces. J. Fluid Mech. 65, 45-63 (1973).
(17)	Wu, J.	Wind - indused turbulent entrainment across a stable density interface. J. Fluid Mech. <u>61</u> , 257-287 (1973).
(18)	Long, R.K.	Some aspects of turbulence in geophysical systems.

- (19) Long, R.K. A theory of mixing in a stably stratified fluid. J. Fluid Mech. <u>84</u>, 113-124 (1978).
 (20) Monin, A.S. Statistical fluid mechanics;
- Yaglom, A.M. mechanics of turbulence. Vol.1. Cambridge, Mass., The MIT Press, 1971.
- (21) Skjeldestad, K. DAREP Users Manual, Kjeller 1977. (NDRE. Teknisk notat S-465). (In Norwegian).
- (22) Pasquill, F. Atmospheric Diffusion. 2nd Edition. New York, Wiley, 1974.



NILU

TLF. (02) 71 41 70

NORSK INSTITUTT FOR LUFTFORSKNING

(NORGES TEKNISK-NATURVITENSKAPELIGE FORSKNINGSRÅD) POSTBOKS 130, 2001 LILLESTRØM ELVEGT. 52.

RAPPORTTYPE Oppdragsrapport	RAPPORTNR. OR 32/78	ISBN82-7247-040-3		
DATO	ANSV.SIGN.	ANT.SIDER OG BILAG		
JULI 1978	O.F. Skogvold	41		
TITTEL Disporsion of b		PROSJEKTLEDER Y. Gotaas		
the atmosphere	eavy gas clouds in	NILU PROSJEKT NR		
ene acmosphere	25077			
FORFATTER(E)		TILGJENGELIGHET **		
Koml T. Didensil		A		
Karl J. Eldsvik	Karl J. Eidsvik			
OPPDRAGSGIVER				
NTNF				
3 STIKKORD (á m	aks.20 anslag)			
tunge gasser	spredning i luft	eksplosive gasser		
REFERAT (maks. 300 anslag, 5-10 linjer) Rapporten beskriver en modell for spredningen av en tung og kald gass-sky i atmosfæren, hvor de essensielle tilstandsvariable er beskrevet så enkelt som mulig. Resul- tatene er ikke kritisk avhengige av usikkerheter i numeriske koeffisienter. Potensielle fareavstander varierer betydelig med underlagets beskaffenhet og atmosfæreforhold.				
TITTEL Spredning av tunge gassskyer i atmosfæren.				
ABSTRACT (max. 300 characters, 5-10 lines)				
The report describes a model for the dispersion of heavy and cold gas clouds in the atmosphere, where the essential state variables are described as simple as possible. The results do not depend critically upon un- cerntaincies about numerical coefficients. Potential hazard distances are predicted to vary significantly with the state of the underlying surface and atmosphere.				
**Kategorier: Å	pen - kan bestilles fra	a NIT.U A		

Må bestilles gjennom oppdragsgiver B Kan ikke utleveres