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A COMPARISON BETWEEN DENSE-GAS AND PASSIVE TRACER DISPERSION ESTIMATES FOR NEAR-SOURCE TOXIC EFFECTS OF CHLORINE RELEASES

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Summary

In this paper we examine the assumptions made in a recent study by Withers and Lees concerning the use of reconstructions of gas warfare releases of chlorine in order to obtain corroborative information on acute inhalation toxicity effects. Specifically, we examine the need to include density effects, ingnored in Whithers and Lees, by comparing dense-gas and passive tracer dispersion estimates obtained using an implementation of the dispersion model CRUNCH, and comparing these in turn with the estimates used in Withers and Lees. We find that the inclusion of density effects removes certain substantial inconsistencies that arise from the use of a passive tracer description of the releases considered, and that if CRUNCH is used in passive tracer mode the concentration estimates agree with those given in Withers and Lees.

An important difference between dense-gas and passive tracer dispersion is predicted by the use of CRUNCH to model releases of line sources of chlorine that are of the same linear strength but of different total length. When used in the passive tracer mode, CRUNCH yields concentrations vs distance that are effectively independent of line source length over the distances of interest in the releases modelled here. These concentrations agree closely with those obtained in Withers and Lees using an infinite line source description, which of course yields no line source length dependent effects. However, the inclusion of lateral spread due to density effects using CRUNCH produces higher concentrations for longer line sources. As far as we are aware this effect has not previously been identified, and we offer an explanation of the mechanism in terms of the scaling behaviour of air entrainment via the top surface of the plume in relation to the contaminant mass flux in the wind direction. It is suggested that this predicted behaviour should be tested experimentally.

We conclude that it is inappropriate to ignore density effects in estimating the near-source dispersion behaviour of chlorine and other dense gas releases, and that the conclusions concerning chlorine toxicity obtained by Withers and Lees are accordingly not robust.

Introduction

Risk assessment of hazardous installations involves quantification of the probabilities of occurrence and magnitude of consequence for a spectrum of accident scenarios. Substantial uncertainties attach to both elements in characterising the risk, and it has become a matter of some contention as to

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whether it is necessary to seek a greater degree of reliability in the consequence-magnitude estimates than can be obtained in the probability-of-occurrence estimates. The requirements of land use planning in relation to major hazards, and of emergency response planning under the CIMAH regulations [1], for example, may place an emphasis in practice on the reliability of the hazard range estimate for releases of hazardous materials, implying a need to estimate consequence magnitude to the best attainable degree of reliability even if the probability of occurrence is relatively more uncertain. An important topic in this context is the dispersion of dense gases, which has been the subject of very substantial research progress over the last decade, as evidenced, for example, in the special issues of this journal [2–4] from 1982 to 1987, and many precursors going back to the important pioneering experiment of van Ulden [5] in 1974.

A specific area of application of dense gas dispersion models is concerned with the estimation of hazard ranges for toxic irritant gases. Although the recent advances in the reliability of dispersion modelling attributable to various field experiments have been very substantial, their main impact has been on dispersion distances appropriate to flammable gases, i.e. on concentrations in the few percent range, rather than on the longer dispersion distances of concern for toxic irritant gases, where the concentrations of importance extend down to much smaller values, typically in the 10 to 100 ppm range for materials of importance in major hazards.

In this sector there remain at least two important aspects requiring validation, namely the dispersion behaviour at longer distances of initially dense releases, and the exposure–response relationships appropriate to the various materials, particularly for human subjects. The literature on risk assessment reveals a wide range of opinion as to the inhalation toxicity of chlorine, expressed as a statement specifying what level of exposure in terms of concentration and duration will result in a given percentage of fatalities. This question was examined by Griffiths and Megson [6], who took several published statements of chlorine and ammonia toxicity and calculated the corresponding hazard ranges for a number of notional releases under a variety of meteorological conditions, but using a single dense gas dispersion model so as to eliminate other differences in the estimation method. A typical result showed hazard ranges to the 50% fatality level for chlorine varying from 1 to 5 km depending on the toxic response relationship used. This degree of uncertainty is quite large in terms of planning needs, and therefore it would be desirable to reduce it. Validation experiments on human subjects in order to ascertain toxicity are clearly ruled out, and thus there is a premium on interpreting human toxic response from forensic examination of accidental or other uncontrolled exposures such as the documented use of toxic gases in warfare. Such a reconstruction was carried out by Nussey et al. [7], who were able to achieve compatibility between one such toxic response statement, the predictions of a particular group

of dense gas dispersion models [8–10] and the reported effects of accidental and warfare releases of chlorine. Withers and Lees [11] subsequently carried out a similar exercise using three 1st World War cases of gas attacks using chlorine, of which one was the same as analysed by Nussey et al. [7]. However, Withers and Lees although acknowledging that such releases would give rise to a dense gas mixture have used a passive tracer dispersion model appropriate to releases of zero density difference with the ambient air. In this paper we have examined the implications of this choice, and have explored the differences in estimated dispersion behaviour that result from the use of a dense gas dispersion model.

Problems arising from ignoring density effects

In attempting to justify their use of a passive dispersion model for chlorine Withers and Lees assert that such a release will start as a dense gas but will undergo transition to passive behaviour within a very short distance. (Withers and Lees use the term *neutral density* for the latter class of dispersion behaviour. We use the more conventional description passive or passive tracer dispersion, since we consider that *neutral density* is an ambiguous substitution for zero-density-difference, and is likely to be confused with the related dispersion concept of neutral stability. It should be noted that passive tracer releases are also quite commonly described as being of neutral buoyancy). The assumption that the transition to passive behaviour would occur within a distance that is short compared to the distance to the nearest troop locations is untested, and needs to be examined, especially since the nearest locations used in the three reconstructions are in the range 50 to 100 m.

In judging the validity of reconstructions of gas releases based on eye-witness observations we are dealing with evidence that is fragmentary and necessarily much different in character than the measured values obtained in field experiments; there will be a correspondingly wide scope for judgement as to what constitutes reasonable agreement between the modelled estimates and the observations. However, a minimum requirement must be that unless the observations themselves are contradictory the reconstruction should only be regarded as valid if it is consistent with the observations. The reconstruction by Withers and Lees has two quite major inconsistencies in this respect, one discussed explicitly and the other implicit. The first concerns the high degree of survival of troops in Trench 43 in the Hill 60 attack; these troops were ordered to mount the firing platform, and so were higher up in the plume than those remaining in the trenches. Withers and Lees dismiss the explanation that their enhanced survival was due to the extra height at which they stood, because the calculated concentrations at 2 m height and on the ground differ by very little (the ratio being 0.9 at 100 m and 0.99 at 300 m). They offer speculative explanations to try to deal with this inconsistency. We will exam-

ine the alternative possibility that this inconsistency arises from an inappropriate choice of dispersion model.

The second inconsistency is implicit, and concerns the observation of plume height in the Langemarck attack as a wall of gas about 5 m high (p. 313 in [11]).

In seeking an interpretation of this observation in relation to the passive dispersion model predictions we refer to the conventional method of matching the visible boundary of a plume to the dispersion coefficient, which involves the approximation that the visible boundary corresponds to the position at which the Gaussian concentration profile falls to 10% of its axial value. The basis for this widely adopted convention is discussed by Slade ([12], p. 101). This yields the relationship $H = 2.14\ \sigma_z$ where H is the plume depth for a ground level release. The values of σ_z used by Withers and Lees are taken from the first report of the UK Atmospheric Dispersion Modelling Working Group [13]. The values of σ_z in Class C conditions (as used for the Langemarck reconstruction) are 8 m at 100 m distance, and 14 m at 200 m, reading from Figure 9 in [13]. The corresponding visible plume heights are thus 17 m and 30 m, which are so much larger than the observed value that one must question the validity of applying the model in this situation. This inconsistency is also examined here in the context of an alternative dispersion model allowing for density effects, which are known to inhibit vertical development during the dense phase of the dispersion process.

Alternative modelling including density effects

In order to examine the influence of density effects on the dispersion scenarios we have utilised an implementation of the code for continuous releases known as CRUNCH [9, 10]. The implementation we have used extends the scope of the basic model described in Refs. 9 and 10 by the inclusion of a sub-code which allows for the effects associated with plumes consisting of a mixture of vapour and liquid aerosol of the released material. As this plume entrains ambient air the density and temperature of the resultant mixture are determined by heat balance between the three components. The user may specify the degree of initial air entrainment associated with the turbulent nature of the release itself, such as a jet of material issuing from a pressure vessel. For the releases considered here the conditions as described by Withers and Lees ([11], p. 304) are such that the chlorine emerges from the cylinders as a superheated liquid, a fraction of which rapidly flashes to vapour to produce a vapour/liquid aerosol mixture at the boiling point temperature of $-34.6\,^{\circ}C$. In defining the initial properties of the release we have given emphasis to this scenario (no.1), assuming an ambient temperature of $15\,^{\circ}C$ in order to calculate the vapour flash fraction. In specifying the initial mixing ratio of air to chlorine for the jet entrainment we follow the example of Pape and Nussey [14] in estimating

effects of chlorine releases from pressurised containment in the context of the U.K. Health and Safety Executive's assessment work for major hazard sites, and take a mixing ratio of air-to-chlorine of 10:1 by mass. It should be noted that this is approximately twice the quantity of dry air required to provide the heat input to the plume necessary to just evaporate the remnant liquid aerosol given a vapour flash fraction of 0.14. The initial conditions of the plume are thus determined by the corresponding heat balance. This working assumption should not be taken to imply that the plume would not still contain aerosol due to the presence of moisture in the admixed air. These characteristics are appropriate for the release from chlorine cylinders as described, but it is instructive to examine other cases in order to elucidate the sensitivity of the modelled dispersion behaviour to the assumptions concerning the initial conditions. Accordingly we consider two further sets of release conditions: scenario 2; in which the chlorine is released all as a gas with no aerosol present at the same temperature as the ambient air and with appropriate density, and scenario 3; similar to scenario 2, but with the density difference between pure chlorine gas and the ambient air artificially set to zero so that the dispersion regime is that of a passive tracer from the outset. Scenario 3 thus corresponds to the passive dispersion behaviour assumed by Withers and Lees, and this release serves as a test of the degree to which our model agrees asymptotically with that used by Withers and Lees when density effects are negligible. The mass mixing ratio of 10 (air) to 1 (chlorine) is used in all three scenarios. The characteristics of the three cases are summarised in Table 1. Concerning the need to use a dense-gas dispersion model for certain releases, we note here the Richardson number

TABLE 1

Plume parameters for the release scenarios considered

Initial plume conditions	Scenario 1	Scenario 2
Vapour flash fraction	0.14	1.00
Mass mixing ratio of air to chlorine	10:1	10:1
Height (m)	0.77	0.82
Temperature (°C)	−12	15
Density (kg/m^3)	1.41	1.28
Chlorine concentration (kg/m^3)	0.128	0.116
	Langemarck	Hill 60
Other parameters		
Line source strength of chlorine (kg/m s)	0.058	0.058
Front width (m)	6000	400
Total source strength of chlorine (kg/s)	349	23.3

Ambient air temperature and density: 15°C and 1.21 kg/m^3 Weather Conditions: Pasquill Class C with windspeed of 2 m/s, and roughness length 0.1 m.
In Scenario 3 the conditions are as for scenario 2 but with the initial plume density artificially set to be equal to the ambient air density.

criterion suggested by Puttock et al. [15], who recommended that if the value of $Ri = g' H_0 / u_*^2$ was less than 10, then density effects would not be important. In this expression H_0 is the initial vertical depth of the release, u_* is the friction velocity, and g' is the modified acceleration due to gravity as defined in eqn. (1) below. For the low windspeeds applying here, a value of u_* of about 0.17 m/s is appropriate, yielding values of Ri of about 41 for scenario 1, and 15 for scenario 2, confirming the view that density effects would have an influence for these releases.

These cases have been used here as the basis for modelling the releases for the Langemarck and the Hill 60 gas attacks, using the implementation of CRUNCH referred to above. In this model [9, 10] the initial plume dimensions are specified in terms of a vertical rectangular cross-section perpendicular to the mean wind direction. In the earlier version [9] the rectangle half-width and height were equal, and the user specified the half-width as input data. This was made more flexible in the next version [10], in which the relative dimensions of the rectangle were chosen by the user in specifying both height and half-width as input data. During the dense phase of dispersion the plume is assumed to travel with the wind at a speed equal to that of the ambient wind at half the cloud height, assuming the conventional logarithmic vertical profile of mean windspeed. A finite crosswind line source can thus be modelled by CRUNCH if the user specifies the length of line source as equal to the width of the above-mentioned rectangle, along with a height that is consistent with the volume flux of the release. This is the scheme used here to model the Langemarck and Hill 60 releases, using the plume widths given by Withers and Lees together with total mass rates of release of chlorine corresponding to their estimated line source rate.

It should be noted here that no adaptation is required in order to use CRUNCH for a line source, and that the implication of the statement on p. 307 of Withers and Lees that "the heavy gas models available are for instantaneous or continuous *point* sources" is not true of CRUNCH [9, 10], nor of the rapid release counterpart DENZ [8]. Both of these models specify the source as an entity of finite dimensions to be set by the user.

Results and Discussion

Figure 1 (*a*, *b* and *c*) shows selected results of our calculations for the scenarios specified in Table 1. These graphs display concentration, normalised plume width and plume height for our treatments of the Langemarck and Hill 60 releases, together with corresponding values of concentration and implied cloud height (i.e. 2.14 σ_z) from [11]. All of the results are for class C stability conditions with a mean 10 m height windspeed of 2 m/s, following the established convention in specifying mean windspeed. For scenarios 1 and 2, where the release is initially denser than the ambient air, the concentration values

are plotted out to the distance at which the model undergoes transition to passive dispersion behaviour. As described in [9] and [10] the transition takes place if either one of two conditions is met, these being i) the lateral growth of the plume due to the density effect has become smaller than that due to the ambient turbulence alone, and ii) the density difference has become smaller than a specified value. In all the cases reported here the transition was triggered by the latter condition, the critical value of the relative density difference being set at 10^{-3}. For scenario 3, with no initial density difference, the values are plotted out to 1500 m along with those from [11]. The distance of the trench locations used by Withers and Lees are also shown.

From Fig. 1*a* it is readily seen that the scenario 1 calculations produce concentration estimates that are significantly higher than those obtained by Withers and Lees, whilst the corresponding values for the (less-likely) scenario 2 lie in between. An important feature is that the distances at which transition to passive behaviour occurs are such that the trench locations are wholly (scenario 1) or partly (scenario 2) within the range in which density effects are important, in contradiction to the assumption made by Withers and Lees.

Additionally, the inclusion of density effects leads to some important differences in behaviour for line sources of different length even though they have the same source strength per unit length. These differences are apparent in Fig. 1*a* in two respects: first, the transition to passive dispersion occurs at a greater distance for the larger line source, and second, the concentrations at a given distance are bigger for the longer line source. The latter contrasts strongly with the behaviour of the infinite passive tracer line source approximation used in [11], which clearly produces no such variation with length. The scale of the differences involved is very substantial: in the scheme used by Withers and Lees the short line source correction (as indicated on p. 328 of [11] is stated to be no more than 5% at a distance of 400 m, whereas in our model the difference is a factor of 5 in concentrations seen at the same distance in the case of the scenario 1 release conditions. The reason for this dependence on line source length is not obvious, and some explanation is required.

The expression governing the rate of lateral growth due to density effects is

$$\frac{dL}{dx} = k(g' \dot{V}/2Lu_p^3)^{\frac{1}{2}} \tag{1}$$

where $g' = g(\rho_p - \rho_a)/\rho_a$

L is the plume half width

x is distance in the mean wind direction

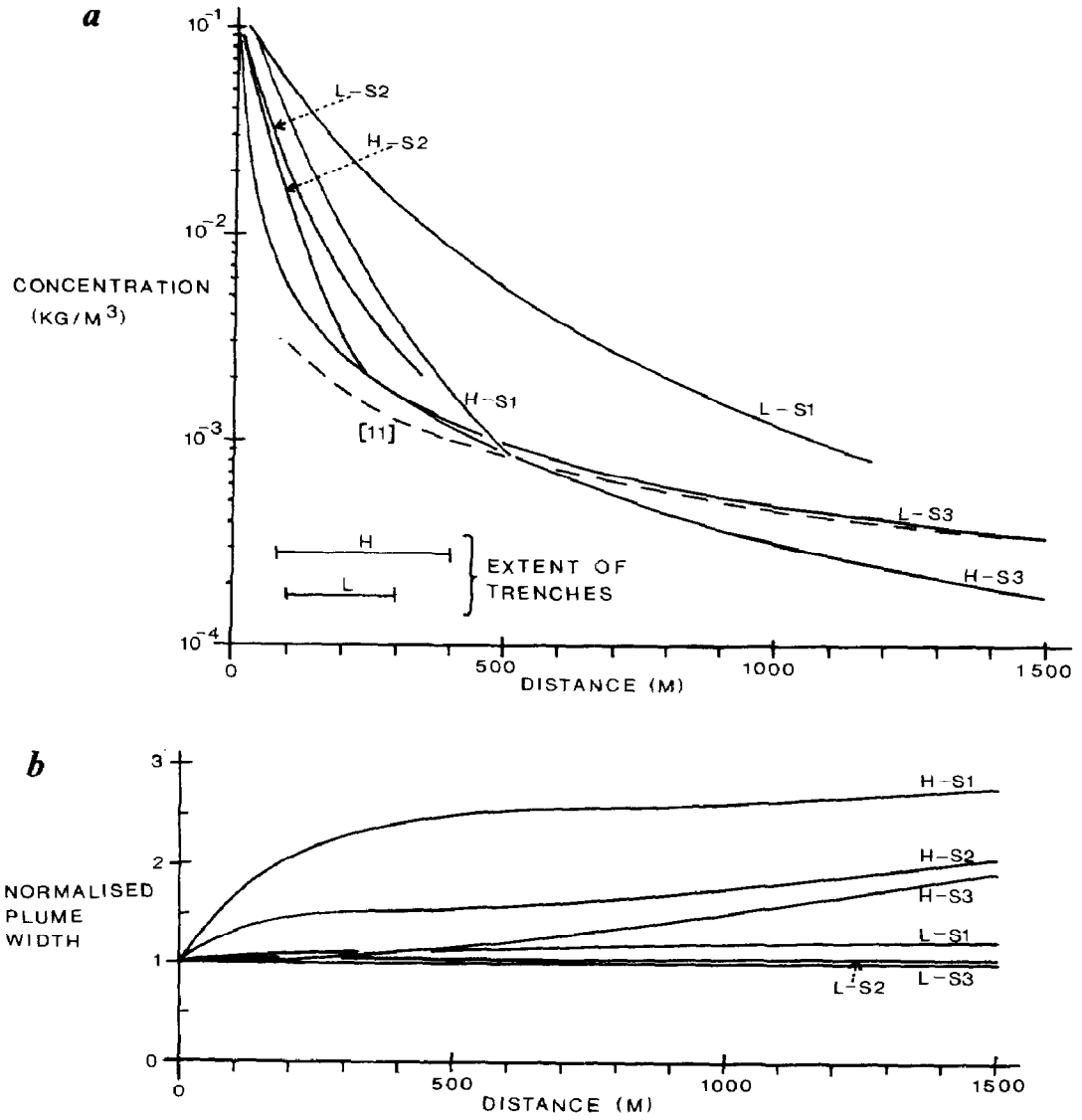
k is a constant

g is the acceleration due to gravity

$\dot{V}$ is the total volume flux of the plume in direction x

ρ_p is the density of the plume
ρ_a is the density of the surrounding air
u_p is the speed of travel of the plume in the mean wind direction

For a given set of release conditions such as scenario 1 this yields an initial value of dL/dx that is independent of L, because at the source the total volume flux $\dot{V}$ is linearly proportional to $2L$, other variables having been specified. At first, then, the two plumes will experience the same absolute increment in half-



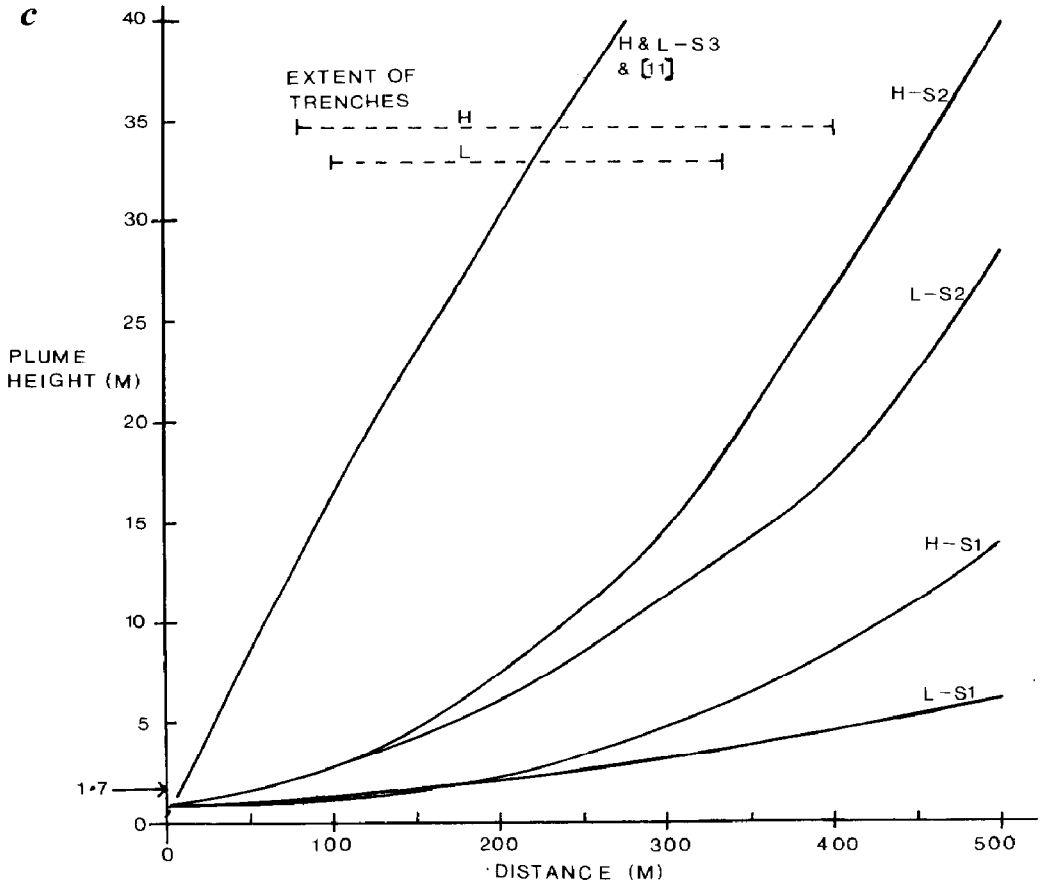


Fig. 1*a*. Concentration vs. distance for scenarios 1, 2 and 3 (see Table 1 and text) compared with those from [11]. The range of trench locations is also shown. In the case of the dense gas releases, scenarios 1 and 2, the distance to the transition to passive dispersion is shown in that the concentrations are plotted only out to that point.

b. Normalised plume width vs. distance.

c. Plume height vs. distance for scenarios 1, 2 and 3, and the implied plume height 2.14 σ_z from [11]. Note the expanded distance scale used in this part of the figure.

The key indicates as follows: S1, S2, S3, scenarios 1, 2 and 3; L, Langemarck; H, Hill 60; [11], from reference [11].

width, $\triangle L$, for a given travel distance, so that the growth relative to the initial value, L_o, is proportionately bigger for the shorter line source. Now the model specifies the mass flux of air entrained through the top surface of the plume via an entrainment velocity description, so that the total mass rate of entrainment through the top is proportional to the horizontal surface area of the interface between the plume and the ambient air, and is thus proportional to the plume width $2(L_o + \triangle L(x))$ or $2L(x)$ at a given distance. (It should be noted at this point that for these releases there is negligible entrainment via the

plume edge, because the plume height-to-width ratio is so small.) However, the total mass flux of contaminant in the mean wind direction is constant, and proportional to the initial plume width $2L_o$ for a specified line source strength. Thus the ratio of the mass-rate-of-air-entrainment to the contaminant-mass-flux scales as the quantity $1+(\triangle L(x)/L_o)$ or $L(x)/L_o$, the normalised plume width. It is readily seen from the form of this expression that the initial occurrence of the same absolute incremental growth in plumes of different line source length will result in a greater relative degree of air entrainment into the shorter line sources, with associated effects on plume parameters such as height and concentration. Values of the normalised plume width for the four dense gas cases are plotted in Fig. 1*b*, and it is seen that their relationship is consistent with the above argument, and with the indications in Fig. 1*a*. The corresponding values for scenario 3 are also shown, taking the plume half-width as $2.14\sigma_y$, i.e. using the same interpretation of plume boundary location as for the implied plume height.

The values of plume height are shown in Fig. 1*c*, where it is seen that from one scenario to another lower concentrations are associated with greater plume height, as one would expect given the previous considerationsof entrainment behaviour (it should be noted here that CRUNCH, in common with other simple box or slab type models, describes the initial dense phase of dispersion in terms of a uniform vertical distribution of contaminant within the plume so that the plume height simply corresponds to the vertical height of the horizontal interface between the plume and the overlying ambient air). Also plotted in Fig. 1*c* are the implied plume height 2.14 σ_z using the σ_z values from [13] employed by Withers and Lees, and the corresponding heights from the scenario 3 releases. The three sets of heights for the passive case are practically identical and are plotted as a single line. This serves as a good test of the convergence of results from our own model running in the passive mode, and those obtained from the passive line source scheme in [11]. This close agreement is further confirmed in the associated concentration estimates from our scenario 3 cases, which are very close to those given by Withers and Lees, as shown in Fig. 1*a*. The divergence in concentrations between the Hill 60 and Langemarck releases evident at the longer distances in scenario 3 arises because of the different rates of lateral growth associated with the differing initial plume widths.

The noteworthy feature of Fig. 1*c* is the relationship between predicted plume height and the typical height of a soldier, say 1.7 m. The values of plume height for scenario 1 are less than this out to about 160 m, thereby permitting the possibility of a substantial difference in concentration between ground level and head height. This feature is consistent with the reported enhanced survival of the troops standing on the firing platform in Trench 43 of the Hill 60 attack, since by a suitable choice of conditions one can readily 'tune' the results of scenario 1 to suppress somewhat the estimated plume height out to the distance indicated of 200 to 250 m. To demonstrate this we have re-run this case

with an initial air-to-chlorine mass mixing ratio of 5.2, which is just slightly in excess of the amount needed to evaporate the remnant aerosol for a vapour flash fraction of 0.14. The initial plume height calculated is then 0.53 m growing to 1.7 m at a distance of 220 m. The existence of any significant difference in concentrations experienced at head height and on the ground is clearly precluded in the passive dispersion estimates, for which the plume heights are much greater than head height. The dense gas scenarios are also consistent with the observation of a plume about 5 m high reported in the Langemarck attack, although the distance at which that occurred is unknown.

The concentrations from the dense gas scenarios are generally higher than those obtained using a passive dispersion model, and considerably so for scenario 1. At first sight this suggests that the effects reported might be associated with higher levels of gas concentration than estimated in [11], and therefore that the toxic response relationship used therein is an over-estimate of the toxicity of chlorine. Given that the response model used lies towards the less toxic end of the spectrum of opinion on chlorine toxicity reported in the literature (for example see the compilation in [6]) this is somewhat surprising. However, there are two factors which militate against such an interpretation. Firstly, there is the consideration of plume height in scenario 1, and the consequent possibility of higher concentrations being experienced on the ground than at head height for a person standing upright on the ground, due to the fact that in the latter case the breathing zone may be above or on the the plume boundary rather than entirely engulfed. Secondly, field experiments on dense gas dispersion reveal considerable variation in concentration over a vertical extent of about 2 m near ground level. For example in the Thorney Island trials special issue [3] Puttock and Colenbrander (p. 383) show concentration profiles taken at distances of about 100 m range in the Thorney Island and the Maplin Sands experiments. For the Thorney Island test the gas concentration measured at 0.4 m height is a factor of about 5 times larger than that at 2.4 m height, and a similar relationship is shown in the Maplin Sands test, with a factor of about 4 between measurements at heights of 0.6 and 2.3 m. Taken together these considerations strongly support the view that there would be a significant difference in exposures between ground level and head height for the Langemarck and Hill 60 attacks. In terms of the toxic response relationship used by Withers and Lees a factor of 5 in concentration is highly significant. For example, taking the 50% mortality level of 433 ppm for 10 minutes at the standard level of activity, a factor of 5 increase in concentration would have an associated mortality of 99.8%, whilst a factor of 5 smaller would yield 0.2%.

Conclusions

Withers and Lees' reconstructions of the Langemarck and Hill 60 chlorine gas attacks using a passive tracer line source model produce features that are

inconsistent with the observations. Alternative modelling procedures that incorporate density effects are available and we have shown that the model CRUNCH [9,10] produces results in which two particular inconsistencies are removed, namely concerning the observed plume height and the associated question of explaining the enhanced survival of troops who were standing compared to those whose breathing zones were essentially at ground level. We have shown also that the distance to transition to passive dispersion behaviour in the dense gas model is such that the density effects should not be ignored.

Use of CRUNCH for the line source releases appropriate in these cases has revealed some important effects that depend on line source length, given a fixed line source strength per unit length, and this behaviour has been explained in terms of the lateral spread due to density, and its effect on entrainment behaviour. This difference in concentration fields for the two gas attacks leads to substantially altered arguments compared to those enforced by the equality of concentration that is a feature of the passive dispersion model. These modelling predictions should be tested experimentally by examining the dispersion behaviour for line sources of the same linear strength but of different total length.

The toxic response relationship for chlorine proposed by Withers and Lees is one of several in the literature, and tends towards the "less toxic" end of the spectrum of opinion. Their crosschecks with gas warfare observations are based on a dispersion model which we argue is inappropriate because it ignores density effects. In consequence neither their dispersion modelling results nor the toxic response relationship they propose can be regarded as particularly robust. This is particularly the case where values for crucial factors such as the number of cylinders used, and the duration of release, have of necessity been assumed. However, we do not suggest that it would be possible to produce a more robust interpretation of toxic response based on reconstructions of releases such as considered here, given the many factors that remain unknown in specifying the necessary conditions. Accordingly, we conclude that an equivalent reconstruction of these cases using a dense gas model might as readily produce support for a more toxic or as less toxic interpretation of the effects of chlorine, depending on the balance chosen between those factors that mitigate the estimated exposure, and those that enhance it.

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