THE FLUCTUATIONS MODULE

by

D.J. Thomson, Meteorological Office

Summary

This paper describes the specification of the fluctuations module.

1. Introduction

Fluctuations in concentrations are important in many dispersion problems involving the release of toxic, inflammable or odourous substances. Knowledge of fluctuations is also useful for assessing uncertainties in dispersion models. By using a combination of theory and experimental data it is possible to put together a scheme which should give some useful guidance on the magnitude of fluctuations. However it should be noted that estimates of concentration fluctuations are, given our present understanding, likely to suffer from somewhat larger errors than estimates of mean concentrations; indeed it is hard to estimate how big the errors in the present scheme might be in situations which lie well outside those for which the scheme has been tested (see P13/02). The scheme is restricted to cases where there is an appreciable mean wind with U » σ_u.

2. Output from the fluctuations module

The inclusion of fluctuations in a dispersion model necessitates quite a precise description of what we are actually calculating. In particular the questions `what sort of mean is the mean concentration?' and `what is the output of the fluctuations module intended to represent?' need addressing. In general the results from ADMS are computed with the assumption that the meteorology is approximately constant in time. Averages are then defined to be ensemble averages, where `ensemble average' means an average over a large number of occasions in which the meteorology is the same (and approximately constant) but in which the details of the boundary layer turbulence differ. We adopt the pragmatic assumption that most of the changes on a period of less than 1 hour are due to the boundary layer turbulence while changes on a longer time-scale are due to changing meteorology. This is consistent with the way met data is treated in the met input module (see P05/01). If either the observation period or release period is less than about an hour, the assumption of approximately constant meteorology is generally not too bad and the above concepts are appropriate. If however these periods are both much greater than an hour, the results apply only to those occasions when the meteorology remains roughly constant for some time. ADMS offers two options to overcome this problem, one involving the mean concentration module and one involving the output module. If these options are used the fluctuations module should not be used - the fluctuations module deals only with fluctuations due to the turbulence.

If information on fluctuations is required, the fluctuations module will be run once for each hour that ADMS is considering. The output provided for each hour depends of the type of calculation being carried out - this could fall into one of three possible types.

2.1 The first type of calculation is concerned with continuous releases. Of course no releases actually continue indefinitely, but the continuous release results will also be appropriate to the `plateau' part of a finite duration release. For this case the mean concentration module calculates the ensemble average concentration - this is a function of (x,y,z). The fluctuations module will estimate the probability (over the ensemble under consideration) with which the 'concentration averaged over period t_{av}' exceeds any particular value $\hat{c}$ - this is a function of $(x,y,z,\hat{c})$ - and will also output the variance of this probability distribution. In addition the ensemble average of the p_{dose}th power of the `concentration averaged over period t_{av}' will be calculated if required. This quantity is often used in calculating the effect of exposure to fluctuating concentrations of toxic substances (Griffiths 1990).

Although the averages and probability distributions are defined relative to a hypothetical ensemble they should be close to time averages and frequency distributions over periods of about an hour. This is because an hour should be long enough to give a representative sample of boundary layer turbulence. If t_{av} is much greater than 1 hour, the met conditions would in general change over the averaging period and so the results obtained (which assume constant meteorology) would not generally be relevant to what happens in reality.

2.2 The second type of calculation is concerned with situations where predictions of time-integrated quantities from finite duration releases are required. For this case the mean concentration module calculates the ensemble average of the time-integrated concentration - this is a function of (x,y,z). The fluctuations module will estimate the probability (over the ensemble under consideration) with which the time-integrated concentration exceeds any particular value

$\hat{c}$ - this is a function of $(x,y,z,\hat{c})$ - and will also output the variance of this probability distribution. In addition the ensemble average of the p_{dose}th power of the time-integrated concentration will be calculated if required. This latter quantity is not usually of interest, but, in order to simplify the program structure, it is convenient to allow the possibility of calculating it anyway.

If t_R is much greater than 1 hour, the met conditions would in general change over the release period and so the results obtained (which assume constant meteorology) would not generally be relevant to what happens in reality.

2.3 The third type consists of situations where predictions of instantaneous quantities from finite duration releases are required. For this case the mean concentration module calculates the ensemble average of the instantaneous concentration - this is a function of (x,y,z,t). The fluctuations module will estimate the probability (over the ensemble under consideration) with which the instantaneous concentration exceeds any particular value $\hat{c}$ - this is a function of $(x,y,z,t,\hat{c})$ - and will also output the variance of this probability distribution. In addition the ensemble average of the p_{dose}th power of the instantaneous concentration will be calculated if required. This quantity (or its time-integral) is often used in calculating the effect of exposure to fluctuating concentrations (Griffiths 1990). Note that predictions of fluctuations for this case are likely to be subject to greater errors than in the cases described in §§2.1 and 2.2 above as a result of the very limited experimental data available with which to test the theory.

The structure of the rest of this paper is as follows. §3 describes a scheme used to estimate the variance of instantaneous concentrations for instantaneous releases in idealised homogeneous flows and §4 discusses how the ideas in §3 can be used to model the variance of instantaneous concentrations in more realistic flows. §5 discusses the effect of time-averaging on the variance while in § the modelling of the probability distribution in terms of its mean and variance is presented. §7 then discusses the problem of calculating statistics of time-integrated concentrations for finite duration releases. Finally §8 discusses the interaction between the model and the plume rise, deposition, radioactive decay, complex terrain, coastline, and building effects modules. The appendices summarise the notation and equations used.

3. Variance of instantaneous concentrations for instantaneous releases in homogeneous turbulence

We start by considering the idealised case of stationary homogeneous turbulence with no mean velocity and note that the second moments of the concentration field for an instantaneous source released at time zero are given by

$$\overline{c(\boldsymbol{x}_1,t)\,c(\boldsymbol{x}_2,t)} = \int p(\boldsymbol{y}_1,\boldsymbol{y}_2,0|\boldsymbol{x}_1,\boldsymbol{x}_2,t)\,q(\boldsymbol{y}_1)\,q(\boldsymbol{y}_2)\,d\boldsymbol{y}_1\,d\boldsymbol{y}_2, \tag{1}$$

where $p(\mathbf{y}_1,\mathbf{y}_2,0|\mathbf{x}_1,\mathbf{x}_2,t)$ is the probability density function of the positions $\mathbf{y}_1$ and $\mathbf{y}_2$ that two particles had at time zero given that they were at positions $\mathbf{x}_1$ and $\mathbf{x}_2$ at time t. In applying this result we adopt an approximation introduced by Sawford (1983). Sawford argued that, if we follow the particles backwards in time from time t to time zero, the displacement of the centre of mass of particle pairs is close to Gaussian and independent of the particle separation (for particles that are close at time t). For an instantaneous source centred on the origin this leads to the result

$$\overline{c^2} = Q^2 G_3(\boldsymbol{x}\sqrt{2},\ \boldsymbol{S}_\Sigma + \boldsymbol{S}_0) \int p(\boldsymbol{r},t)\,G_3(\boldsymbol{r},\boldsymbol{S}_0)\,d\boldsymbol{r}. \tag{2}$$

Here we have assumed that the shape of the source distribution is Gaussian (i.e. proportional to $G_3(\mathbf{x},\mathbf{S}_0)$); this assumption is of course unlikely to be accurate in practice but an attempt at a more precise description seems unwarranted in view of the uncertainties associated with source effects. Note that when p is Gaussian, the integral $\int p(\mathbf{r},t)G_3(\mathbf{r},\mathbf{S}_0)d\mathbf{r}$ in (2) is equal to $G_3(0,\mathbf{S}_\Delta+\mathbf{S}_0)$. Theory and random walk simulations show that p is actually more peaked than Gaussian, resulting in an additional contribution to $\overline{c^2}$. We will write $\int p(\mathbf{r},t)G_3(\mathbf{r},\mathbf{S}_0)d\mathbf{r} = \mu G_3(0,\mathbf{S}_\Delta+\mathbf{S}_0)$, where μ is a factor related to the non-Gaussianity of p. In conjunction with (2), it is natural, in homogeneous turbulence, to make a Gaussian assumption for the mean concentration:

$$\bar{c} = QG_3(\boldsymbol{x},\boldsymbol{S}_1 + \boldsymbol{S}_0). \tag{3}$$

We also note that $\mathbf{S}_1$, $\mathbf{S}_\Delta$ and $\mathbf{S}_\Sigma$ are related by

$$\boldsymbol{S}_\Delta + \boldsymbol{S}_\Sigma = 2\boldsymbol{S}_1. \tag{4}$$

By using (2) and (3) and some simple assumptions, $\overline{c^2}$ can be expressed more simply. Our aim here is to express $\overline{c^2}$ in terms of the $\bar{c}$-field and as few extra quantities as possible. The reason for this is to provide a basis for modelling concentration variance in more complex flows, in particular in flows where the mean concentration distribution is non-Gaussian. A possible simplifying assumption is to assume that $\mathbf{S}_1$, $\mathbf{S}_\Delta$, $\mathbf{S}_\Sigma$ and $\mathbf{S}_0$ are isotropic. However we will not assume $\mathbf{S}_0$ is isotropic because we wish to use the results below in a framework where the source size in the y- and z-directions represents the true source dimensions but in which the source size in the x-direction is related to the release duration. Hence we assume $\mathbf{S}_0$ is axi-symmetric about an axis aligned with the x-direction and for simplicity we make the same assumption about $\mathbf{S}_1$, $\mathbf{S}_\Delta$ and $\mathbf{S}_\Sigma$. This leads to

$$\overline{c^2} = \overline{c_m^2}\,(\overline{c_x}/\overline{c_m})^{\chi}(\overline{c}/\overline{c_x})^{\chi_x} \qquad (5)$$

with χ, χ_x and $\overline{c_m^2}$ given by

$$\chi = 2\,\frac{\sigma_1^2 + \sigma_0^2}{\sigma_\Sigma^2 + \sigma_0^2}, \qquad (6)$$

$$\chi_x = 2\,\frac{\sigma_{1x}^2 + \sigma_{0x}^2}{\sigma_{\Sigma x}^2 + \sigma_{0x}^2} \qquad (7)$$

and

$$\overline{c_m^2} = \mu\overline{c}_m^2\,\frac{(\sigma_{1x}^2 + \sigma_{0x}^2)(\sigma_1^2 + \sigma_0^2)^2}{\sqrt{(\sigma_{\Delta x}^2 + \sigma_{0x}^2)(\sigma_\Delta^2 + \sigma_0^2)}\sqrt{(\sigma_{\Sigma x}^2 + \sigma_{0x}^2)(\sigma_\Sigma^2 + \sigma_0^2)}} \qquad (8)$$

We also note that, using (4), σ_Σ and $\sigma_{\Sigma x}$ can be expressed as

$$\sigma_\Sigma^2 = 2\sigma_1^2 - \sigma_\Delta^2 \qquad (9)$$

and

$$\sigma_{\Sigma x}^2 = 2\sigma_{1x}^2 - \sigma_{\Delta x}^2. \qquad (10)$$

Equations (5) to (10) hold without the axi-symmetric assumption is σ_0, $\sigma_{0x} \ll \sigma_\Delta \ll \sigma_1$ (i.e. if t is large enough for initially close particles to have separated to distances much greater than σ_0 and σ_{0x}, but small enough for the particles' motions to be strongly correlated) since the source size does not then affect results and $\mathbf{S}_\Sigma \approx 2\mathbf{S}_1$. They also hold at large times when σ_0, $\sigma_{0x} \ll \sigma_\Delta$ and $\mathbf{S}_1 \approx \mathbf{S}_\Delta \approx \mathbf{S}_\Sigma$ (i.e. if t is large enough for initially close particles to have separated to distances much greater that σ_0 and σ_{0x}, and also large enough for the particles' motions to be independent). Finally, in both of the above cases the condition $\sigma_{0x} \ll \sigma_\Delta$ can be replaced by the requirement that σ_{0x} be very large (so that the source looks like a line source). For simplicity we will assume equations (5) to (10) hold more generally - the use of more complicated expressions seems unwarranted in view of the uncertainties associated with source effects and because of our lack of detailed understanding of the behaviour in more realistic flows at times when σ_Δ is of order σ_1. Note that in reality σ_1 and σ_Δ satisfy $0 \leq \sigma_\Delta \leq \sigma_1$, σ_{1x} and $\sigma_{\Delta x}$ satisfy $0 \leq \sigma_{\Delta x} \leq \sigma_{1x}$ and μ is greater than or equal to unity. Provided these constraints are satisfied, equations (5) to (10) imply that $1 \leq \chi,\chi_x \leq 2$ and that σ_c^2 is non-negative.

In equations (5) to (10), $\overline{c^2}$ is determined by the $\overline{c}$-field, σ_1, σ_{1x}, σ_Δ, $\sigma_{\Delta x}$, σ_0, σ_{0x} and μ and so we will now consider the problem of determining σ_Δ, $\sigma_{\Delta x}$ and μ. Consider first short travel times, where the separation of particle pairs is dominated by inertial subrange eddies. This range of travel times can be characterised by σ_Δ, $\sigma_{\Delta x} \ll \sigma_1$ or, equivalently, $t \ll T_L$. In this region inertial subrange theory predicts that p grows self-similarly and isotropically, with σ_Δ^2 growing in proportion to εt^3. The random walk simulations of Thomson (1990), which show reasonable agreement with measurements of concentration fluctuations indicate that

$$\sigma_\Delta^2 \simeq \varepsilon t^3/3.$$

At large times $\mathbf{S}_1 \approx \mathbf{S}_\Delta$ with both quantities growing linearly with t. Suitable formulae for σ_Δ and $\sigma_{\Delta x}$ which interpolate between the above limiting cases are

$$1/\sigma_\Delta = 1/\sigma_1 + 1/(\varepsilon t^3/3)^{1/2} \qquad (11)$$

and

$$1/\sigma_{\Delta x} = 1/\sigma_{1x} + 1/(\varepsilon t^3/3)^{1/2}. \qquad (12)$$

The selection of a model for μ is more complicated. We will consider a number of limiting cases where we can estimate μ with a degree of confidence. Consider first the case where σ_{0x} is very large, so that the source looks like a line source. For $t \ll T_L$ the simulations indicate that μ equals unity near the source and approaches about 2.8 at longer range, with a transition at the distance where $\sigma_\Delta \sim \sigma_0$ (see figure 1a). A reasonable approximation to μ is

$$\mu = \begin{cases} 1 & \sigma_\Delta/\sigma_0 \le 0.9 \\ 1 + 1.8\log((\sigma_\Delta/\sigma_0)/0.9)/\log(17/0.9) & 0.9 \le \sigma_\Delta/\sigma_0 \le 17 \\ 2.8 & 17 \le \sigma_\Delta/\sigma_0 \end{cases} \qquad (13)$$

(see figure 1a). For $t \gg T_L$, provided $\sigma_0 \ll \sigma_\Delta$, the random walk results for μ can be approximated by

$$\mu = \max(1, 2.8 - 0.6t\varepsilon/\sigma_{vel}^2) \qquad (14)$$

(see figure 1b). A suitable expression for μ which behaves qualitatively correctly for all times and source sizes is given by the minimum of the expressions in (13) and (14). The second case we consider is the case where $\sigma_{0x} = \sigma_0$, i.e. we consider a compact isotropic source, For this case the simulations indicate rather large values of μ with a suitable expression being the minimum of

$$\mu = \begin{cases} 1 & \sigma_\Delta/\sigma_0 \le 1 \\ 1 + 11\log((\sigma_\Delta/\sigma_0)/1)/\log(100/1) & 1 \le \sigma_\Delta/\sigma_0 \le 100 \\ 12 & 100 \le \sigma_\Delta/\sigma_0 \end{cases} \tag{15}$$

and

$$\mu = \max(1, 12 - (11/3)t\varepsilon/\sigma_{vel}^2) \tag{16}$$

(see figure 2a and b). We now need to consider the general case. An appropriate expression for μ with the right qualitative properties can be obtained as follows. First evaluate (i) the minimum of (13) and (14) and (ii) the minimum of (15) and (16) with, in (ii), σ_0 taken to be the maximum of σ_0 and σ_{0x}. Then take the maximum of these two values of μ.

With the above expressions for σ_Δ, $\sigma_{\Delta x}$ and μ, $\sigma_c/\overline{c}$ tends to zero at large times as might be expected in truly homogeneous stationary turbulence.

4. Variance of instantaneous concentrations for finite duration and continuous releases

As usual, the dispersion in time from an instantaneous line source aligned with the x-direction in homogeneous turbulence with no mean flow (i.e. the situation considered in §3 with $\sigma_{0x} = \infty$) can be regarded as an approximation to the downwind dispersion from a continuous compact source in a homogeneous turbulent flow with mean velocity $U(\gg \sigma_u)$. If the line source is finite in length (due to σ_{0x} being finite) then it can be regarded as an approximation to a finite duration release. Hence, for continuous or finite duration releases from compact sources, we can use equations (5) to (16), except that (i) t is replaced by x/U (an approximation to the travel time), (ii) $\overline{c}_m$ is interpreted as the maximum of the ensemble mean concentration over t, y and z with x fixed, (iii) $\overline{c}_x$ is replaced by $\overline{c}_t$, and (iv) σ_{0x} and σ_{1x} are interpreted as U times the spread in release times and as U times the spread in travel times to the downwind distance x.

We will now consider more realistic, inhomogeneous atmospheric flows and construct a model based on the above concepts which were developed for idealised flows. Consider first short travel times, where $\sigma_\Delta \ll \sigma_1$ and the separation of particle pairs is dominated by inertial subrange eddies. In this regime we can use the homogeneous results discussed above (i.e. equations (5) to (16)) with some confidence since the correct scales are represented in the equations. At larger times the homogeneous results are of less help in parametrizing $\overline{c^2}$. In particular inhomogeneity and shear effects, and possibly also microscale eddies, nearly always become important at large travel times and tend to prevent $\sigma_c/\overline{c}$ tending to zero. (Strictly speaking, fluctuations due to mesoscale eddies do not come within the ambit of the fluctuations module; however, in the absence of a pronounced spectral gap it is impossible to draw a clear distinction between fluctuations produced by mesoscale eddies and those produced by the boundary layer turbulence). Observations of near-surface continuous releases in near-neutral conditions (Fackrell and Robins 1983; Mylne and Mason 1990) suggest $\sigma_c/\overline{c}$ at the point where $\overline{c} = \overline{c}_m$ is of order unity at large travel times. Guided by the above we adopt the following pragmatic approach. Equations (5) to (12) are used with the modifications indicated in the previous paragraph. μ is calculated from equations (13) to (16) as indicated in §3 (using x/U for t) with the proviso that μ is not allowed to fall below the value at which the value of $\overline{c_m^2}/(\overline{c}_m)^2$ for a continuous point source (i.e. for $\sigma_0 = 0$ and $\sigma_{0x} = \infty$) equals 2. σ_{vel}^2 is taken to be $(\sigma_u^2 + \sigma_v^2 + \sigma_w^2)/3$ and σ_u, σ_v, σ_w, ε and U are evaluated at the mean plume height $\overline{z}$ rather than at the height at which σ_c is required.

To complete the model we need to consider the values of σ_1, σ_{1x}, σ_0 and σ_{0x}. For the simplified situation considered in §3, σ_1 can be estimated from the $\overline{c}$-field using (3). This leads to

$$\sigma_1^2 + \sigma_0^2 = Q/2\pi\overline{c}_i.$$

Here $\overline{c}_i$ is the maximum over y and z (at some fixed time t) of the integral of $\overline{c}$ in the x-direction. The corresponding result for homogeneous turbulence with a mean velocity U (as considered in the first paragraph of this section) is

$$\sigma_1^2 + \sigma_0^2 = Q/2\pi U\overline{c}_i. \tag{17}$$

where $\overline{c}_i$ is now the maximum over y and z (at some fixed downwind distance x) of the time-integral of $\overline{c}$. For more general flows we propose using (17) with U evaluated at height $\overline{z}$. This is consistent with the general approach of trying to express $\overline{c^2}$ in terms of the $\overline{c}$- field and as few extra quantities as possible, as discussed in §3. It is conceivable that, depending on the scheme used to calculate $\overline{c}_i$, (17) could lead to a negative value for σ_1^2. To avoid this, we impose a minimum value of $10^{-6}\mathrm{m}^2$ on σ_1^2. σ_{1x} is much more difficult to determine from the $\overline{c}$-field. This is because if t_R is large the maximum concentration will depend only very weakly on σ_{1x} (this is especially true if, as would usually be the case, the time-profile of the release has, not a Gaussian, but a top hat shape). Hence the module requires σ_{1x} as an input variable. Let us now consider σ_0 and σ_{0x}. Comparison of random walk results with the data of Fackrell and Robins (1982) (see Thomson (1990)) suggests that, for uniform sources with circular cross-section, σ_0 should be taken to be the source diameter D_s. For complex source geometries, the model will only be able to give a general indication of the magnitude of source size effects and some general estimate of the length-scale of the source would have to be used. σ_{0x} will be taken to be $\max(t_R U, \sigma_0)$ with U evaluated at the mean plume height. For the continuous source case, σ_{0x} is infinite and this results in a considerable simplification of the equations (see Appendix B for details).

5. The effect of time-averaging

Let us first consider the situation discussed in §3 and consider the correlation between the concentration at two points $\mathbf{x}_1$ and $\mathbf{x}_2$ at some time t. We will restrict attention to cases where $\sigma_0 = 0 \ll \sigma_\Delta$ and $\sigma_\Delta \ll \sigma_1$ (i.e. $t \ll T_L$). If $|\mathbf{x}_1 - \mathbf{x}_2| \ll \sigma_\Delta$ then, in following a pair of particles backwards from positions $\mathbf{x}_1$ and $\mathbf{x}_2$ at time t to time zero, the particles will tend to forget their initial separation and so, from (1), $\overline{c(\mathbf{x}_1, t)c(\mathbf{x}_2, t)} \approx \overline{c(\mathbf{x}_1, t)^2}$. If however $|\mathbf{x}_1 - \mathbf{x}_2| \gg \sigma_\Delta$, then this will make a large difference to the scale of the particle separation at time zero, and so the probability that both parties will pass through the source region will be much smaller than the probability for two particles which are close at time t, and we will have $\overline{c(\mathbf{x}_1, t)c(\mathbf{x}_2, t)} \ll \overline{c(\mathbf{x}_1, t)^2}$. This indicates that the length-scale of the concentration fluctuations is of order σ_Δ. If σ_{0x} is small too (i.e. $\sigma_{0x} \ll \sigma_\Delta$), then, using Sawford's (1983) approximation in the forward direction, the correlation between the concentration at two points separated by a vector $\mathbf{r}$ takes the form $p'(\mathbf{r}/\sqrt{2}, t)/p'(0, t)$. The results of the random walk simulations (Thomson 1990) indicate that the integral scale of this correlation function is approximately $0.43\sigma_\Delta$. For larger values of σ_{0x} it is possible that the coefficient of σ_Δ should be a weak function of $\sigma_{0x}/\sigma_\Delta$. At larger times when $\sigma_\Delta \sim \sigma_1$ and t is of order T_L or larger, it is much harder to estimate the length-scale as there are two possible contenders, σ_Δ and the turbulence integral length-scale. This is discussed below in relation to more complex flows.

Let us now consider more complex flows and restrict attention to continuous releases (i.e. $\sigma_{0x} = \infty$) with $\sigma_0 \ll \sigma_\Delta$. For $\sigma_\Delta \ll \sigma_1$, the ideas in the previous paragraph support the use of a length-scale proportional to σ_Δ. At larger times when $\sigma_\Delta \sim \sigma_1$, we retain this assumption, both for simplicity and because it is consistent with the fact that the model for σ_c does not allow $\sigma_c/\overline{c}$ to tend to zero (the fact that $\sigma_c/\overline{c}$ does not approach zero implies the presence of eddies of similar or larger scale to the plume and hence the generation of some fluctuations on a scale comparable to the plume width). In order to achieve reasonable agreement with the data of Mylne and Mason (1990) and Mylne (1992), we choose the length-scale to be, not $0.43\sigma_\Delta$, but $4\sigma_\Delta$ see P13/02. This change in the coefficient is permissible on theoretical grounds because of the idea that the coefficient may be a weak function of $\sigma_{0x}/\sigma_\Delta$. Note that we have assumed a single length-scale at all points in the plume at a given downwind distance and so the module is unable to describe the variation with height observed by Fackrell and Robins (1982) and Mylne (1992). To represent such effects it is probably necessary to account in more detail for the variation of turbulence properties with height.

We expect the time-scale of concentration fluctuations to be determined by the spatial fluctuations being carried past the measurement point and so we assume an integral time-scale T_c of $4\sigma_\Delta/U$ where U is the mean velocity at the height where σ_c is required. As the height of interest tends to zero, U becomes small and $4\sigma_\Delta/U$ tends to infinity. This indicates a failure of the above argument near the ground, In reality vertical mixing is efficient near the ground, with the time-scale for mixing from the ground to the height of interest being less than the time-scale for advection of fluctuations of scale $4\sigma_\Delta$ past the measurement point, Hence the concentration fluctuations change little in the vertical and are in effect advected by the velocity at some height z_b above the surface. This suggests we should, in calculating T_c, evaluate U at the maximum of z_b and the height of interest. In the neutral surface layer the order of magnitude of z_b can be determined by equating $4\sigma_\Delta/U$, where U is evaluated at height z_b, to the time for diffusion between the surface and height z_b, i.e. z_b/ku_* where k is the von Karman constant. This leads to

$$z_b \sim \frac{4\sigma_\Delta k^2}{\log((4\sigma_\Delta k^2 + z_0)/z_0)}.$$

For simplicity we will replace '~' by '=' in this expression and will adopt it more generally in non-neutral conditions. We take k equal to 0.4.

In order to determine the effect of time-averaging we could try to construct a model of the shape of the correlation function. However it is simpler for our purposes to adopt an exponential form with the same integral scale. For an averaging period t_{av} this gives a concentration variance equal to

$$2\sigma_c^2(t_{av} = 0)(\exp(-\hat{t}) - 1 + \hat{t})/\hat{t}^2$$

where $\hat{t} = t_{av}/T_c$.

When σ_0 is not much less than σ_Δ it is harder to estimate the length-scale of the concentration fluctuations. The pragmatic approach adopted here is to take the variance of the (time-averaged) concentration to be the smaller of (i) the variance of the instantaneous concentration and (ii) the variance of the time-averaged concentration due to a point source (i.e. $\sigma_0 = 0$) of the same strength.

6. Model for the concentration p.d.f.

There are now a number of observations (Lewellen and Sykes 1986; Sawford 1987; Dinar et al 1988; Mylne and Mason 1990) that indicate that the so-called clipped-normal distribution is a useful approximation to the p.d.f. of the (possibly time-averaged) concentration at a point, For this distribution, the probability of the (possible time-averaged) concentration exceeding $\hat{c}$ is given by

$$P(\hat{c}) = \begin{cases} \tfrac{1}{2}\left(1 - erf\left(\dfrac{\hat{c}/\sigma - \gamma}{\sqrt{2}}\right)\right) & \hat{c} \geq 0 \\ 1 & \hat{c} < 0 \end{cases}$$

where γ and σ are parameters of the distribution. If we make such an assumption it is possible to calculate γ and σ, and hence $P(\hat{c})$, from $\overline{c}$ and σ_c (for details see Appendix B). The intermittency, defined here as the fraction of time when clean air is observed, can be obtained as 1 - $P(0)$ and is an increasing function of $\sigma_c/\overline{c}$. The ensemble average of any function of the (possibly time-averaged) concentration, such as the p_{dose}th power, can also be deduced. The 'exponential distribution plus intermittency' has also been tested against data and has shown reasonable agreement in some situations (Sawford 1987; Mylne and Mason 1990). However the exponential distribution plus intermittency has the property that $\sigma_c/\overline{c}$ is never less than unity, and so is unable to represent cases where $\sigma_c/\overline{c}$ is small.

At each point at which output is required, it is necessary to specify the values $\hat{c}$ for which $P(\hat{c})$ is to be calculated. These values need to be able to resolve the spectrum of possible concentration values quite well (e.g. one may wish to invert the curve $P(\hat{c})$ to obtain the concentration which is exceeded with a particular probability). Also, if one wishes to combine results from the calculations for different hours (e.g. to obtain an estimate of the fraction of time the 3 minute averaged concentration exceeded a particular level over some 24 hour period), then it is important that the values of $\hat{c}$ are the same for each hour and are not related to the mean concentration. In order to meet these requirements, logarithmical spaced $\hat{c}$-values are used so as to cover a large range without too many values. In order that the $\hat{c}$-values at a particular location are the same for all hours, these values are determined from the source strength and the distance d from the source using
in S.I. units. d_1 determines the overall size of the $\hat{c}$-values, d_2 the number of $\hat{c}$-values per decade and m the total number of $\hat{c}$-values. These values can be specified by the user subject to a maximum of 100 for m. The values m = 73, $d_1 = 10^{-3}$ and d_2 = 8 should be sufficient to cope with all eventualities, but will often provided unnecessarily many points.

$$\hat{c} = \begin{cases} \dfrac{Q' d_1}{d \min(d, 1000)} 10^{j/d_2}, & j = 0, \ldots, m-1 \quad \text{for continuous releases} \\ \dfrac{Q d_1}{d \min(d, 1000)(t_R + d/6)} 10^{j/d_2}, & j = 0, \ldots, m-1 \quad \text{for finite duration releases} \end{cases}$$

7. Statistics of time-integrated concentrations for finite duration releases

The statistics of time-integrated concentrations from a finite duration release can be easily computed from knowledge of the statistics of time-averaged concentrations from a continuous release. This is because, if the release rate is the same in the two cases and $t_R = t_{av}$, then the time-integral of the concentration resulting from a release of duration t_R has the same statistics as t_{av} times the average over a period t_{av} (i.e. the time-integral over a period t_{av}) of the concentration from a continuous release. For this case, the probability of the time-integrated concentration exceeding $\hat{c}$ is calculated for $\hat{c}$-values determined as for continuous releases (see §6) but with Q' replaced by Q.

8. Interaction with the plume rise, deposition, radioactive decay, complex terrain, coastline and building effects modules

When the plume rise, dry or wet deposition, complex terrain, coastline or building effects modules are used, the fluctuations module will simply make use of the mean concentration field as influenced by these modules. It is hoped that plume rise, deposition or complex terrain effects will not greatly change the plume's structure and so, by making use of the mean concentration field as influenced by these phenomena, useful estimates of fluctuation statistics can be made. It is or course true that steep terrain with flow separation and recirculation can have a major effect on the plume structure, but the complex terrain module is not valid anyway for such situations. In the case of the complex terrain module it would be better to use values of U and the turbulence quantities as influenced by the terrain, but for simplicity we have not done this. The situation as regards the coastline and building effects module is rather different. For the coastline module the changes in turbulence and mean flow can be substantial across the coast and would be expected to have a major effect. Hence results should not be regarded as valid if the coastline module is used. Buildings are likely to cause increased mixing and substantial structural changes to plumes in building wakes. As a result the concentration fluctuation statistics should not be regarded as valid where results differ significantly from results in the absence of the buildings. It does seem likely however that results from the module will predict a larger hazard than occurs in reality (increased mixing in the building wake will tend to reduce fluctuations).

The plume rise module includes a prediction for the increased spreading of the instantaneous plume due to turbulence generated by the plumes' buoyancy. It might be beneficial to make use of this in the fluctuations module but, for simplicity, we have not done this. This is likely to lead to a (hopefully small) overprediction of the fluctuation variance and hence of the hazard.

If removal processes are invoked, the effective source strength is reduced as downwind distance increases (See P17/01). In such situations the $\hat{c}$-values are calculated using the unmodified source strength and σ_c and $P(\hat{c})$ are calculated using the source strength and mean concentration field as modified by the removal processes. This is necessary to ensure that (i) $\overline{c}_i$ and the source strength are consistent for use in equation (17), and (ii) the values of $\hat{c}$ are the same for different hours.

When the radioactivity decay module is invoked, mean concentrations are calculated for a (possibly large) number of species. It is not very practical to use the fluctuations module in such a situation and, in any case, it is generally accepted that the presence or absence of fluctuations does not have a large influence on the hazard caused by radioactive substances. In principle the fluctuations module could be used for the primary emissions by calling the fluctuations module once for each radioactive species. The main control program would however require modification to permit this.

Appendix A: Notation

c	instantaneous concentration
$\overline{c}_m$	maximum of $\overline{c}$ over all values of x, y and z for fixed t in the case of the idealised flows discussed in §3 and the maximum over all values of t, y and z for fixed x elsewhere
$\overline{c}_x$	maximum of $\overline{c}$ over all values of x for fixed t, y and z
$\overline{c}_t$	maximum of $\overline{c}$ over all values of t for fixed x, y and z
$\overline{c}_i$	maximum of $\int \overline{c}\,dx$ over all values of y and z for fixed t in the case of the idealised flows discussed in §3 and the maximum of $\int \overline{c}\,dt$ over all values of y and z for fixed x elsewhere
$\overline{c_m^2}$	value of $\overline{c^2}$ at positions where $\overline{c} = \overline{c}_m$
$\hat{c}$	values of the (possibly time-averaged or time-integrated) concentration for which exceedance probabilities are calculated
d	distance from source to receptor (= $\sqrt{x^2+y^2}$)
d_1	scale factor used in defining the $\hat{c}$-values

d_2	number of $\hat{c}$-values per decade
D_s	source diameter
$G_3(\mathrm{x},\mathbf{S})$	the density function of a three-dimensional Gaussian distribution with covariance matrix **S**
k	von Karman's constant
m	number of $\hat{c}$-values
$p(\mathbf{r},t)$	p.d.f. at time zero of $1/\sqrt{2}$ times the separation of two particles which are close at time t
$p'(\mathbf{r},t)$	p.d.f. at time t of $1/\sqrt{2}$ times the separation of two particles which are close at time zero
p_{dose}	power of concentration used in calculating quantities relevant to toxic response
$P(\hat{c})$	probability of the (possibly time-averaged or time-integrated) concentration exceeding $\hat{c}$
q	source distribution
Q	total amount of material released for finite duration releases
Q'	rate of release of material for continuous sources
$\mathbf{S}_1$	covariance matrix of the displacement of a particle
$2\mathbf{S}_\Delta$	covariance matrix of the separation of a pair of particles which are initially close
$\frac{1}{2}\mathbf{S}_\Sigma$	covariance matrix of the displacement of the centre of mass of a pair of particles which are initially close
$\mathbf{S}_0$	covariance matrix of the source distribution
t	time after an instantaneous release or travel time to a point a distance x downwind of a continuous release
t_{av}	averaging period for concentration measurements
t_R	release duration
T_c	integral time-scale of concentration fluctuations
T_L	Lagrangian integral time-scale
U	mean wind speed
x,y,z	Cartesian coordinates in the downwind, crosswind and vertical directions with origin centred on the source for the idealised flows considered in §3 and with origin on the ground below the source elsewhere
x	vector with components x, y and z
$\overline{z}$	mean plume height at given downwind distance
z_0	roughness length
γ,σ	parameters of the clipped-normal distribution
ε	rate of dissipation of turbulent energy
μ	factor related to the non-Gaussianity of p

σ_c^2	variance of the (possibly time-averaged or time-integrated) concentration
$\sigma_u^2,\sigma_v^2,\sigma_w^2$	variance of the *x, y* and *z* components of velocity
σ_{vel}^2	$(\sigma_u^2+\sigma_v^2+\sigma_w^2)/3$

For a number of three-dimensional distributions a certain notation is used to denote the extent of the distributions in the (y,z)-plane and in the x-direction. As an example, consider the distribution of the displacement of a particle. σ_1 is defined by setting σ_1^4 equal to the determinant of the (y,z) sub-matrix of $\mathbf{S}_1$ (e.g. $\sigma_1^2 = \sigma_y\sigma_z$ if $\mathbf{S}_1 = \mathrm{diag}(\sigma_x^2,\sigma_y^2,\sigma_z^2)$) while σ_{1x} is defined by $\sigma_{1x}^2\sigma_1^4 = \det\mathbf{S}_1$ (e.g. $\sigma_{1x} = \sigma_x$ for $\mathbf{S}_1 = \mathrm{diag}(\sigma_x^2,\sigma_y^2,\sigma_z^2)$). σ_Δ, $\sigma_{\Delta x}$, σ_Σ, $\sigma_{\Sigma x}$, σ_0 and σ_{0x} are defined similarly in terms of $\mathbf{S}_\Delta$, $\mathbf{S}_\Sigma$ and $\mathbf{S}_0$. To further illustrate the significance of these definitions, consider a problem involving mean shear. In such a situation the density function of the distribution of particle displacements might be proportional to

$$\frac{1}{(2\pi)^{3/2}\sigma_x\sigma_y\sigma_z}\exp\left(-\frac{(x-az)^2}{2\sigma_x^2}-\frac{y^2}{2\sigma_y^2}-\frac{z^2}{2\sigma_z^2}\right)$$

for some σ_x, σ_y and σ_z. The above definitions then lead to $\sigma_1^2 = \sigma_y\sigma_z$ and $\sigma_{1x} = \sigma_x$. Note that σ_{1x} is equal to the root mean square along-wind spread at given y and z and is, for given σ_1, simply related to the peak of the density function; it is not however so simply related to the overall along-wind spread which is increased by the shear. For cases other than the idealised instantaneous released discussed in §3, the three-dimensional distributions are best interpreted as distribution over (t,y,z) values instead of (x,y,z) values (see discussion at the start of §4).

Appendix B: Summary of input, output, equations used and numerical procedures

Input

The following information is required as input to the fluctuations module (although some of these items are not used by the current version of the module):

Name of log dataset

Name of datasets containing the limits on the number of messages to be produced by the module and the limits within which input variables should lie

The number of the hour being considered (in the sequence of hours given in the met input dataset)

Flag to indicate if the module is being called for the first time

Flag to indicate whether the calculation is of the type defined in §2.1, §2.2 or §2.3

Flag to indicate if statistics involving p_{dose} are required

List of coordinates at which output is required

Functions giving σ_u, σ_v, σ_w, ε and U values

z_0, D_s, m, d_1, d_2, and p_{dose}

The remaining variables depend on the calculation type:

Continuous releases (as in §2.1)

$\overline{c}$, $\overline{c}_m$ and $\overline{z}$ at the locations where output is required
Q' and t_{av}

Time-integrated results from finite duration releases (as is §2.2)

$\int \overline{c}\,dt$, $\overline{c}_i$ and $\overline{z}$ at the locations where output is required
Q and t_R

Finite duration releases (as in §2.3)

$\overline{c}$, $\overline{c}_t$, $\overline{c}_i$, $\overline{c}_m$ and $\overline{z}$ at the locations where output is required
Q, t_R and σ_{1x}

The values of Q and Q' as amended by the removal processes and without amendment are both required. The unamended values are used only for calculating the $\hat{c}$-values.

Output

The following quantities are output:

Flag to indicate invalid input data (not used at present)

σ_c
Ensemble average of the p_{dose}th power of the (possibly time-averaged of time-integrated) concentration - this is set to -999 if the flag instructing the module to calculate this quantity is not set
P(ĉ) for various values of ĉ (the ĉ-values are also returned)

Equations Used

For finite duration releases we use

$$\sigma_c^2(t_{av}=0) = \overline{c^2} - \bar{c}^2,$$

$$\overline{c^2} = \overline{c_m^2}(\bar{c}_t/\bar{c}_m)^{\chi}(\bar{c}/\bar{c}_t)^{\chi_x},$$

$$\chi = 2\frac{\sigma_1^2 + \sigma_0^2}{\sigma_\Sigma^2 + \sigma_0^2}, \quad \chi_x = 2\frac{\sigma_{1x}^2 + \sigma_{0x}^2}{\sigma_{\Sigma x}^2 + \sigma_{0x}^2},$$

$$\overline{c_m^2} = \mu \bar{c}_m^2 \frac{(\sigma_{1x}^2 + \sigma_{0x}^2)(\sigma_1^2 + \sigma_0^2)^2}{\sqrt{(\sigma_{\Delta x}^2 + \sigma_{0x}^2)}\,(\sigma_\Delta^2 + \sigma_0^2)\sqrt{(\sigma_{\Sigma x}^2 + \sigma_{0x}^2)}\,(\sigma_\Sigma^2 + \sigma_0^2)},$$

$$\sigma_\Sigma^2 = 2\sigma_1^2 - \sigma_\Delta^2, \quad \sigma_{\Sigma x}^2 = 2\sigma_{1x}^2 - \sigma_{\Delta x}^2,$$

$$\frac{1}{\sigma_\Delta} = \frac{1}{\sigma_1} + \frac{1}{\sqrt{\varepsilon(x/U)^3/3}}, \quad \frac{1}{\sigma_{\Delta x}} = \frac{1}{\sigma_{1x}} + \frac{1}{\sqrt{\varepsilon(x/U)^3/3}},$$

$$\sigma_1^2 = \max(Q/2\pi U\bar{c}_i - \sigma_0^2,\ 10^{-6}m^2)$$

$$\sigma_0 = D_s, \quad \sigma_{0x} = \max(t_R U, \sigma_0)$$

and

$$\mu = \max(\mu_1, \mu_2)$$

$\mu_1$is determined as the minimum of

$$\mu_1 = \begin{cases} 1 & \sigma_\Delta/\sigma_0 \le 0.9 \\ 1 + 1.8\log((\sigma_\Delta/\sigma_0)/0.9)/\log(17/0.9) & 0.9 \le \sigma_\Delta/\sigma_0 \le 17 \\ 2.8 & 17 \le \sigma_\Delta/\sigma_0 \end{cases}$$

and

$$\mu_1 = \max\left(1, 2.8 - 0.6\,\frac{x}{U}\,\frac{\varepsilon}{\sigma_{vel}^2}\right)$$

subject to the condition that μ_1 must be greater than the value at which $\overline{c_m^2}/\overline{c}_m^2$ for $\sigma_0 = 0$ and $\sigma_{0x} = \infty$ equals 2. μ_2 is determined as the minimum of

$$\mu_2 = \begin{cases} 1 & \sigma_\Delta/\sigma_{0x} \le 1 \\ 1 + 11\log((\sigma_\Delta/\sigma_{0x})/1)/\log(100/1) & 1 \le \sigma_\Delta/\sigma_{0x} \le 100 \\ 12 & 100 \le \sigma_\Delta/\sigma_{0x} \end{cases}$$

and

$$\mu_2 = \max\left(1, 12 - \frac{11}{3}\,\frac{x}{U}\,\frac{\varepsilon}{\sigma_{vel}^2}\right)$$

For continuous duration releases these equations simplify to

$$\sigma_c^2(t_{av} = 0) = \overline{c^2} - \overline{c}^2,$$

$$\overline{c^2} = \overline{c_m^2}(\overline{c}/\overline{c}_m)^\chi,$$

$$\chi = 2\,\frac{\sigma_1^2 + \sigma_0^2}{\sigma_\Sigma^2 + \sigma_0^2},$$

$$\overline{c_m^2} = \mu \bar{c}_m^2 \frac{(\sigma_1^2 + \sigma_0^2)^2}{(\sigma_\Delta^2 + \sigma_0^2)(\sigma_\Sigma^2 + \sigma_0^2)},$$

$$\sigma_\Sigma^2 = 2\sigma_1^2 - \sigma_\Delta^2,$$

$$\frac{1}{\sigma_\Delta} = \frac{1}{\sigma_1} + \frac{1}{\sqrt{\varepsilon (x/U)^3/3}},$$

$$\sigma_1^2 = \max(Q'/2\pi U\bar{c}_m - \sigma_0^2, 10^{-6} m^2)$$

and

$$\sigma_0 = D_s.$$

μ is determined as the minimum of

$$\mu = \begin{cases} 1 & \sigma_\Delta/\sigma_0 \le 0.9 \\ 1 + 1.8\log((\sigma_\Delta/\sigma_0)/0.9)/\log(17/0.9) & 0.9 \le \sigma_\Delta/\sigma_0 \le 17 \\ 2.8 & 17 \le \sigma_\Delta/\sigma_0 \end{cases}$$

and

$$\mu = \max\left(1, 2.8 - 0.6\, \frac{x}{U}\, \frac{\varepsilon}{\sigma_{vel}^2}\right)$$

subject to the condition that μ must be greater than the value at which $\overline{c_m^2}/\bar{c}_m^2$ for $\sigma_0 = 0$ equals 2.

For both finite duration and continuous releases σ_{vel}^2 is taken to be $(\sigma_u^2 + \sigma_v^2 + \sigma_w^2)/3$. Throughout the above, σ_u^2, σ_v^2, σ_w^2 and ε and U are evaluated, not at the height where σ_c is required but at the mean plume height $\bar{z}$.

The variance of the time-averaged concentration for a continuous release is taken to be the minimum of $\sigma_c^2(t_{av} = 0)$ and

$$2\sigma_c^2(\sigma_0 = 0, t_{av} = 0)(\exp(-\hat{t}) - 1 + \hat{t})/\hat{t}^2$$

where $\hat{t} = t_{av}U/4\sigma_\Delta$ with U evaluated at the higher of the height of interest and the height z_b defined by

$$z_b = \frac{4\sigma_\Delta k^2}{\log((4\sigma_\Delta k^2 + z_0)/z_0}$$

k is taken to be 0.4.

The probability distribution of the (possibly time-averaged) concentration is given by

$$P(\hat{c}) = \begin{cases} ½\left(1 - erf\left(\frac{(\hat{c}/\sigma - \gamma)}{\sqrt{2}}\right)\right) & \hat{c} \geq 0 \\ 1 & \hat{c} < 0 \end{cases}$$

with γ and σ determined from

$$\frac{\overline{c}}{\sigma} = \frac{\gamma}{2}(1 + erf(\gamma/\sqrt{2})) + \frac{1}{\sqrt{2\pi}}\exp(-\gamma^2/2)$$

and

$$\frac{\sigma_c^2 + \overline{c}^2}{\sigma^2} = \frac{1}{2}(1 + \gamma^2)(1 + erf(\gamma/\sqrt{2})) + \frac{\gamma}{\sqrt{2\pi}}\exp(-\gamma^2/2).$$

To avoid possible problems with precision, σ_c^2 is not allowed to fall below $10^{-5}\overline{c}^{\,2}$ for the purpose of calculating γ and σ. $P(\hat{c})$ is output for values of $\hat{c}$ given, in S.I. units, by

$$\hat{c} = \begin{cases} \frac{Q'd_1}{d\min(d,1000)}10^{j/d_2}, & j = 0,\ldots,m-1 \text{ for continuous releases} \\ \frac{Qd_1}{d\min(d,1000)(t_R + d/6)}10^{j/d_2}, & j = 0,\ldots,m-1 \text{ for finite duration releases.} \end{cases}$$

For the statistics of the time-integrated concentration from a finite duration source, the above equations for a continuous release are used, but reinterpreted as indicated in §7. In fact the same code is used for both cases with the input and output variables being interpreted as indicated in the input and output lists given above.

Numerical Procedures

Here we describe the methods used to calculate γ, σ and the ensemble average of the p_{dose}th power of the (possibly time-averaged or time-integrated) concentration. γ is calculated from $(\sigma_c^2 + \bar{c}^2)/\bar{c}^2$ using three different approaches according to the size of $(\sigma_c^2 + \bar{c}^2)/\bar{c}^2$. For small values (corresponding to $\gamma > 3.5$) the equation for γ has an asymptotic form which enable an explicit form for γ to be derived, while for large values (corresponding to $\gamma < -3.5$) another asymptotic form exists which can be solved by a rapidly converging iteration. For immediate values a bisection approach is used. σ is calculated by expressing it in terms of $\bar{c}$ and γ and, as in calculating γ, asymptotic limits are used for large positive and negative γ. The reason for using these asymptotic limits (for both γ and σ) is that they are much faster for large positive γ and much more accurate for large negative γ than using the full equations. The full equations suffer from precision problems for large negative γ due to the taking of differences of nearly equal terms; this can lead to errors of orders of magnitude or even of sign. The ensemble average of the p_{dose}th power of the (possibly time-averaged or time-integrated) concentration is calculated by numerical integration using Simpson's rule.

References

Dinar N. et al 1988 'Characterisation of concentration fluctuations of a surface plume in a neutral boundary layer', *Boundary-Layer Meteorol.*, **45**, 157-175.

Fackrell J.E. and Robins A.G. 1982 'Concentration fluctuations and fluxes in plumes from point sources in a turbulent boundary layer', *J. Fluid Mech.*, **117**, 1-26.

Griffiths R.F. 1990 'The use of probit expressions in the assessment of acute population impact of toxic releases', *J. Loss Prev. Process Ind.*, **4**, 49-57.

Lewellen W.S. and Sykes R.I. 1986 'Analysis of concentration fluctuations from Lidar observations of atmospheric plumes', *J. Clim. Appl. Met.*, **25**, 1145-1154.

Mylne K.R. 1992 'The vertical profile of concentration fluctuations in near-surface plumes', Submitted to *Boundary-Layer Meteorol.*

Mylne K.R. and Mason P.J. 1991 'Concentration fluctuation measurements in a dispersing plume at a range of up to 1000m', *Q. J. R. Meteorol. Soc.*, **117**, 177-206.

Sawford B.L. 1983 'The effect of Gaussian particle-pair distribution functions in the statistical theory of concentration fluctuations in homogeneous turbulents', *Q. J. R. Meteorol. Soc.*, **109**, 339-354.

Sawford B.L. 1987 'Conditional concentration statistics for surface plumes in the atmospheric boundary layer', *Boundary-Layer Meteorol.*, **38**, 209-223.

Thomson D.J. 1990 'A stochastic model for the motion of particle pairs in isotropic high Reynolds number turbulence, and its application to the problem of concentration variance', *J. Fluid Mech.*, **210**, 113-153.

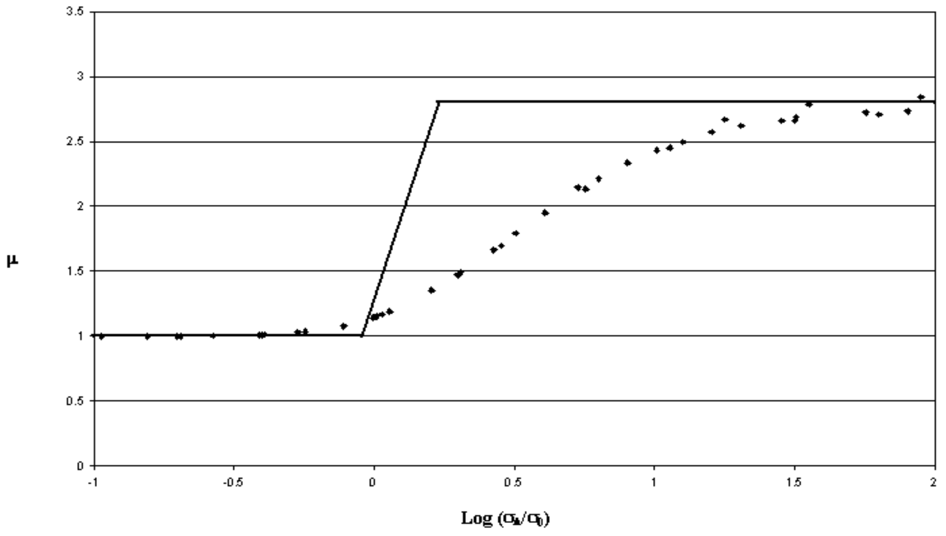

Figure 1a Values of μ for $\sigma_{0x}=\infty$. μ as a function of σ_Δ/σ_0 for t « T_L. (♦ indicates the random walk results (Thomson 1990), while the solid line denotes equation (13)

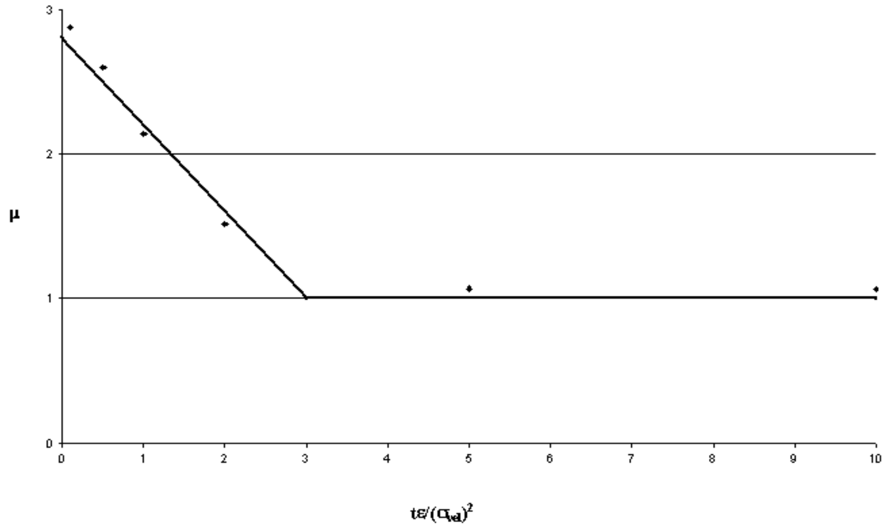

Figure 1b Values of μ for $\sigma_{0x}=\infty$. μ as a function of $t\varepsilon/(\sigma_{vel})^2$ for σ_0 « σ_Δ for t « T_L. (♦ indicates the random walk results (Thomson 1990), while the solid line denotes equation (14)

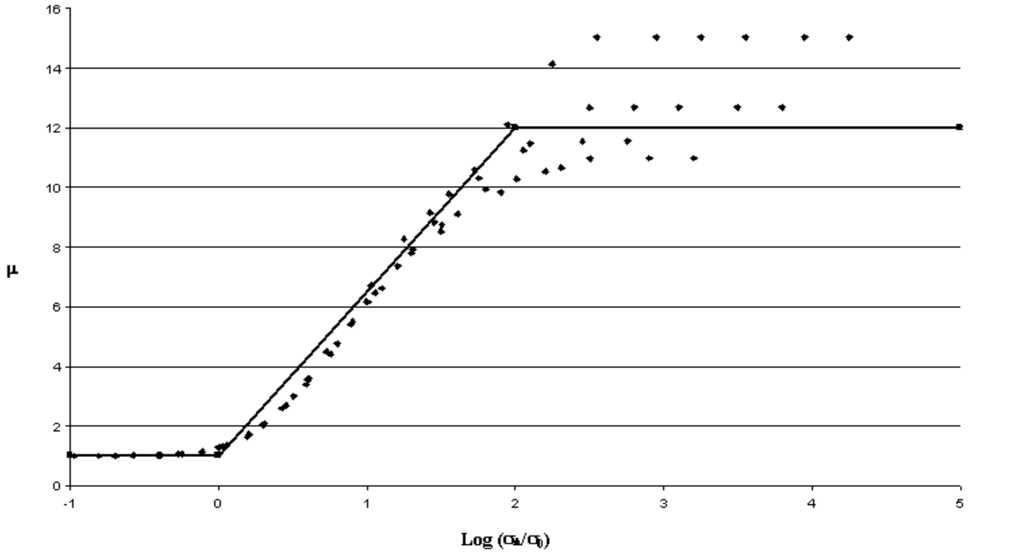

Figure 2a Values of μ for $\sigma_{0x} = \sigma_0$. μ as a function of σ_Δ/σ_0 for $t \ll T_L$. (♦indicates the random walk results (Thomson 1990), while the solid line denotes equation (15)

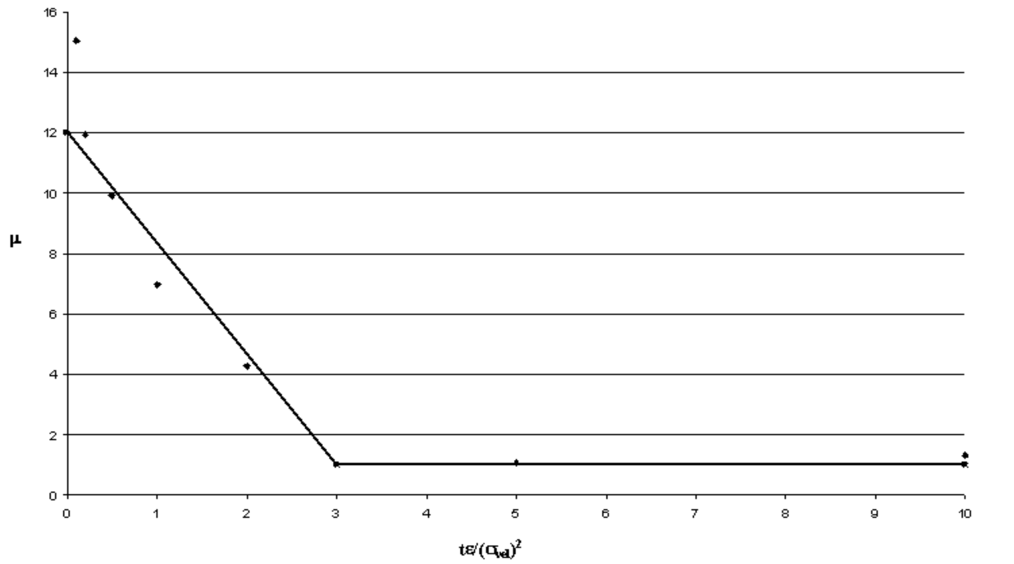

Figure 2b Values of μ for $\sigma_{0x}=\sigma_0$. μ as a function of $t\varepsilon/(\sigma_{vel})^2$ for $\sigma_0 \ll \sigma_\Delta$ for $t \ll T_L$. (♦indicates the random walk results (Thomson 1990), while the solid line denotes equation (16)