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CONCENTRATION FLUCTUATION AND PEAK-TO-MEAN
CONCENTRATION RATIOS IN REAL PLUMES*

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INTRODUCTION

There are numerous instances in which time-integrated plume diffusion models are inadequate to solve current air pollution problems. In particular, the models fail when instantaneous concentration predictions are needed. Although Hilst,⁽¹⁾ Davidson⁽²⁾ and Gifford^(3,4) have attempted to model diffusion in instantaneous plumes from a more-or-less theoretical viewpoint, little experimental data is available on which to model the instantaneous plume. In the fall of 1967, however, a series of diffusion tests were conducted at Hanford which provide experimental insight into the concentration fluctuations associated with real plumes. These concentration fluctuations can be ascribed in part to the meandering of the instantaneous plume and in part to random variations of the concentration within the instantaneous plume. This paper is primarily concerned with the description of the magnitude of the fluctuations and the systematic ways in which they vary.

EXPERIMENTAL DESIGN AND DATA DESCRIPTION

During the summer and fall of 1967 a prototype diffusion grid using ⁸⁵Kr** as a tracer, was installed and tested at Hanford by Ludwick and Nickola and their coworkers. The layout of this grid was essentially the

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**⁸⁵Kr is an artificially produced inert gas having a half-life of 10.76 years which decays by beta emission with a predominant (99%) energy of 0.69 MeV.

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same as that described by Barad and Fuquay⁽⁵⁾ for the Green Glow Diffusion Program, although reduced in scale. A total of 64 halogen-quenced Gieger-Mueller sensors were located on concentric arcs 200 and 800 m from the source point as shown in Figure 1, and functioned as independent sequential samplers operated in unison. The output from the samplers was accumulated over 38.4 sec sampling periods with the lost time between sampling periods of the order of a few milliseconds. The source consisted of a cylinder from which the ^{85}Kr was released at a constant rate of about 1 Ci min^{-1} for periods 10 to 20 minutes in duration and had effective height of approximately 1 m. A comprehensive description of the instrumentation has been published by Ludwick, et al.⁽⁶⁾ Meteorological supporting facilities for diffusion tests included: a 125 m tower near the center of the grid instrumented at 15 m intervals, and a 24 m portable tower instrumented at 6 levels.

Four continuous tracer releases were made during periods of relatively constant atmospheric stability; the significant meteorological and diffusion parameters for these releases are summarized in Table 1. Typically, the maximum activity within the plume (averaged over a 38.4 sec sampling period) was of the order of $170 \text{ counts sec}^{-1}$ at 200 m and $25 \text{ counts sec}^{-1}$ at 800 m. Calibration of the sensors indicated that $9.2 \text{ counts sec}^{-1}$ were approximately equal to $1 \mu\text{Ci m}^{-3}$. The count rates were not converted to true concentrations for this analysis. The complete data for the prototype tests are extensive and are contained in a separate data volume.⁽⁷⁾ The standard deviations of the crosswind distribution of tracer in the mean plume (σ_y) were computed for each sampling arc and are included in Table 1. The estimates of the standard deviation of the vertical distribution of the tracer (σ_z) included in Table 1 were obtained using bivariate-normal diffusion equation and the measured values of the centerline concentration, wind speed and σ_y . 100 percent reflection of the plume at ground level was assumed for this computation.

Figure 1. Layout of the ^{85}Kr diffusion grid.

TABLE 1

TEST PARAMETERS

TEST	DURATION OF RELEASE*	Ri**	$\bar{U}^\dagger$	$\sigma_u^\dagger$	$\sigma_v^\dagger$	200M		800M	
						$\sigma_y^\ddagger$	$\sigma_z^\ddagger$	$\sigma_y^\ddagger$	$\sigma_z^\ddagger$
1	15+28	+0.20	1.2	0.22	0.14	20	12	67	360
2	14+15	-0.022	7.7	1.5	1.3	42	10	196	25
3	9+58	-0.31	4.1	1.1	0.93	24	35	64	120
4	19+51	+0.046	2.6	0.60	0.33	25	23	83	46

* DURATION OF RELEASE IN MIN AND SEC

** AVERAGE VALUE OF RICHARDSON NUMBER FOR THE RELEASE PERIOD COMPUTED FOR THE LAYER BETWEEN 1.5 AND 12.2 M

† WIND SPEED AND STANDARD DEVIATIONS IN m sec^{-1} BASED UPON OBSERVATIONS TAKEN AT 5 sec INTERVALS DURING THE PERIOD OF TRACER GENERATION

‡ STANDARD DEVIATIONS OF PLUME CONCENTRATION DISTRIBUTIONS IN m

All crosswind distances were measured from the center of mass of the crosswind concentration distribution and vertical distances were measured from the ground. Positions within each plume were normalized to the standard deviation of the concentration distribution. In the analysis of data from the 1.5 m sensors the normalizing factor was σ_y , while in those cases when the data from all the sensors were analyzed the normalizing factor was $(\sigma_y^2 + \sigma_z^2)^{1/2}$.

A detailed analysis of the data from 1.5 m level samplers has recently been published.⁽⁸⁾ The results of this analysis are summarized in the next two sections. Following this summary in order are the presentation of a peak-to-mean concentration ratio model and a discussion of absolute peak-to-mean concentrations. Finally, the direction of current research at Battelle, Pacific Northwest Laboratories (PNL) is indicated.

ANALYSIS

On the average the concentration fluctuations tend to follow rather well defined patterns. The analysis and discussion to follow are intended as a description of these mean patterns, although in several instances an indication is given of the scatter of the data about the patterns. No significant differences in the concentration fluctuations could be determined between the data from the 200 and 800 m arcs, and 4 plumes do not provide sufficient data from which to attempt to identify stability effects on the fluctuations. Therefore, all data were combined for analysis.

Concentration Fluctuations

One of the characteristics of diffusion in real plumes is the intermittency of the concentration at any fixed position. The frequency of occurrence of a given concentration level at a position is a function location with respect to the mean plume axis. This relationship is clearly shown in the cumulative frequency distributions in Figure 2. In preparation

of this figure the concentrations for each sensor, averaged over 38.4 sec periods, were normalized to the local mean concentration during plume passage. The normalization compensated for variations in the rate of tracer release, atmospheric conditions between tests and for diffusion from 200 to 800 m.

The highly skewed nature of the distribution of short-term concentrations is obvious in Figure 2. It is of particular interest that short-term concentrations exceed the local mean concentration 65 percent of the time near the mean plume axis and about 85 percent of the time near the edge of the mean plume. If it is assumed that for all practical purposes the plume is absent from a position when the short term concentration is less than 1/10 the local mean concentration, then the plume is absent from the mean plume axis more than 35 percent of the time. At the edge of the mean plume this absence increases to about 80 percent.

Peak-to-Mean Concentration Ratios

Frequently interest is expressed in the upper tails of the cumulative frequency distributions in Figure 2. These tails contain the peak-to-mean concentration ratios. The peak concentration is defined as a maximum short-term concentration at a given location. The peak concentrations are intimately related to the smoothing of the concentration data as well as position within the plume. For this reason, peak-to-mean concentration ratios (P/M) are examined in relation to both position in the mean plume and smoothing of the data.

Maximum values of concentrations, smoothed over increasing time intervals by use of running means, were determined for each sampling position. From these maxima and the local mean concentrations, peak-to-mean ratios were calculated. Individual peak-to-mean ratios are indicated as P/M , and mean ratios by an overbar, $\overline{P/M}$. The ratio of the duration of plume passage (T) to the duration of the peak (t) was computed as a measure of the degree of

Figure 2. Cumulative frequency distribution of normalized short-term concentrations.

smoothing of the peak and is called the time ratio (T/t). Large time ratios indicate minimal smoothing. The $\overline{P/M}$ were better related to smoothing when the time ratio was used as the smoothing variable than when the duration of the peak was used. No systematic errors appear to have been induced in the $\overline{P/M}$ versus T/t relationship by combining data from releases varying between 10 and 20 min in duration. The use of peak-to-mean ratios provides the same normalization benefits described for the short-term concentrations, while the time ratio provides a normalization for the variations in duration of releases.

The relationship between $\overline{P/M}$ and the normalized crosswind position is shown in Figure 3. The time ratios in the figure indicate the mean time ratio for the group of $\overline{P/M}$ used to derive each curve. The scatter about each of the curves in Figures 3 and 4 are indicated in Table 2 of Reference 8. At small time ratios the $\overline{P/M}$ ratio is almost independent of crosswind position. As the time ratio increases the relationship between $\overline{P/M}$ and $|y|/\sigma_y$ approaches those described by Singer⁽⁹⁾ and hypothesized by Gifford.⁽⁴⁾ An interesting feature of this figure is that $\overline{P/M}$ ratios approach a definite maximum at the edge of the plume. The relationship between $\overline{P/M}$ and T/t is better examined in Figure 4. The increase of $\overline{P/M}$ with T/t is as expected, however, can not be adequately described by a power law as has previously been suggested.^(4,10,11) Gifford's estimates of the power law relationship between $\overline{P/M}$ and T/t have been included in Figure 4 for comparison. It is particularly significant that the $\overline{P/M}$ observed near the center of the ⁸⁵Kr plumes were higher than would have been anticipated from Gifford's centerline curve. It is also interesting to note that in each case the magnitude of $\overline{P/M}$ ratios is less than or equal to the magnitude of T/t .

A Model for Peak-to-Mean Concentration Ratios

The relationship between $\overline{P/M}$, $|y|/\sigma_y$ and T/t as illustrated in Figures 3 and 4 appeared to be sufficiently well defined to justify a modeling. As the initial data analysis was not based upon a strong theoretical foundation, a strictly empirical model was attempted. The shapes of the curves in Figures 3

Figure 3. Peak-to-mean concentration ratios as a function of normalized crosswind position.

Figure 4. Peak-to-mean concentration ratios as a function of the time ratio.

and 4 were used as a guide in selecting the mathematical functions to be incorporated in the model. Additional inputs were obtained from the following considerations:

1. At $T/t = 1$, P/M must equal 1 for all crosswind positions in the plume,
2. P/M ratios cannot exceed T/t in magnitude,
3. P/M ratios are a minimum at $|y|/\sigma_y = 0$, and
4. Near the edge of the plume P/M ratios should approach T/t in magnitude.

The double curvature in the relationship between $\overline{P/M}$ and $|y|/\sigma_y$ indicate that the $\tanh^2$ of some function of $|y|/\sigma_y$ would be an appropriate factor in the model. Similarly, the relationship between P/M and T/t indicated that the $\ln(T/t)$ would also be an appropriate factor in the model. These factors were combined in a variety of ways in an attempt to satisfy all of the conditions. Regression coefficients were determined for each of the prospective models using the data from which Figures 3 and 4 were derived (approximately 820 points) and least squares techniques. Each model was judged by comparison of lateral and transverse cross-sections of the model with corresponding curves from Figures 3 and 4. Quantitative estimates of the goodness of fit of the models to data were not reliable as the variance of the individual P/M ratios about the $\overline{P/M}$ is not homoscedastic. The best $\overline{P/M}$ model was determined to be:

$$\overline{P/M} = 1.9 Y^2 X^4 + 0.90 X^2 + 1.0 \quad (1)$$

where

$$Y^2 = \tanh^2 \left[\frac{|y|/\sigma_y}{X + 1} \right] \quad (2)$$

and

$$X^2 = \ln(T/t) \quad (3)$$

Figure 5 shows an isometric view of the model. The model is not entirely satisfactory, as the predicted $\overline{P/M}$ ratios exceed T/t near the edge of the plume for small T/t . However, this does not appear to be a serious deficiency.

For lack of more definitive data, it is tempting of the model for prediction of $\overline{P/M}$ ratios beneath elevated plumes. With this prospect in mind, the data from the towers were analyzed for P/M ratios. These P/M ratios were grouped by T/t and normalized position in the plume, where the normalized position is defined by

$$r/\sigma_r = [(y/\sigma_y)^2 + (z/\sigma_z)^2]^{1/2} \quad (4)$$

It was then assumed that r/σ_r could be substituted for $|y|/\sigma_y$ in Equation (2) and $\overline{P/M}$ ratios were predicted for each group using the average time ratio and relative position in the model.

The peak-to-mean ratios predicted by the model under this assumption are reasonably close to the observed values as shown in Figure 6. The model appears to be especially well suited for the prediction of P/M ratios at large T/t and r/σ_r . However, some caution is in order as the vertical extent of the data was limited to $z/\sigma_z \leq 1.1$. The tendency of the initial model to over predict $\overline{P/M}$ ratios at large $|y|/\sigma_y$ and small T/t is evident in Figure 6 as the data tend to drop below the line in the middle of the P/M ratio range. But, the most important feature of Figure 6 is that in no case did the model significantly under predict a $\overline{P/M}$ ratio.

Absolute Peak-to-Mean Concentration Ratios

In many ways it seems advantageous to consider peak concentrations normalized to a common concentration rather than the local mean concentration. The obvious concentration for use in the normalization of peak concentrations would appear to be the average concentration at mean plume axis. To prevent confusion, peak concentrations normalized to this axial mean concentration will be referred to as absolute P/M ratios (P/M_a) as opposed to conventional

Figure 5. Isometric view of the peak-to-mean concentration ratio model.

Figure 6. Correlation of predicted and observed peak-to-mean ratios.

P/M ratios (P/M_L) in which peak concentrations are normalized to the local mean concentration.

An absolute peak-to-mean concentration ratio might be thought of as the product of a conventional P/M ratio and the ratio between the local and axial mean concentrations. Thus, it is to be expected that P/M_a are a function of the same variables as P/M_L . A summary of the results of absolute peak-to-mean ratio is presented in Table 2 by relative crosswind position and time ratio. The central value in each group is the geometric mean P/M_a for the class and the upper and lower values are one geometric standard deviation removed from their respective $\overline{P/M_a}$. The range of values of T/t and y/σ_y for each group is indicated by the arithmetic mean $\pm$ one standard deviation.

The relationship between $\overline{P/M_a}$ and normalized crosswind position is shown in Figure 7. The most striking feature of this figure is that the maximum $\overline{P/M_a}$ ratios occur at the mean plume axis. The relatively flat portion of the curves near the center of the mean plume is the result of plume meander and its extent essentially defines the limit of meandering of the axis of the instantaneous plume. As the edge of the mean plume is approached the rapid decrease in $\overline{P/M_a}$ is caused by the comparison of peak concentrations at the extreme edge of the meandering plume with the axial mean concentrations. A small crosswind displacement with respect to the mean plume σ_y will appear as a significantly larger displacement when compared to the standard deviation of the crosswind concentration distribution of the meandering plume.

The relationship between absolute peak-to-mean concentration ratios and the time ratio is clearly shown in Figure 8. As with conventional P/M, P/M_a monotonically increase with increasing T/t . Unlike conventional P/M, however, the P/M_a do not start from a common point at $T/t = 1$. Rather, P/M_a appear to converge at some large value of T/t . The presentation in this figure has an advantage over that in Figure 7 in that it emphasizes the decrease in P/M_a near the edge of the mean plume.

TABLE 2

AVERAGE VALUES OF ABSOLUTE PEAK-TO-MEAN CONCENTRATION RATIOS

AS A FUNCTION OF THE NORMALIZED CROSSWIND POSITION AND

TIME RATIO

(T/t)	by/σ_y				
	<u>.21 + .13</u>	<u>.70 + .14</u>	<u>1.23 + .16</u>	<u>1.75 + .16</u>	<u>2.31 + .16</u>
1.94 + .08	1.83	2.13	1.83	0.877	0.239
	1.44	1.26	0.988	0.351	0.060
	1.13	0.743	0.535	0.140	0.015
	n=24	n=28	n=25	n=23	n=15
3.98 + .22	2.85	3.48	3.12	1.66	0.474
	2.32	2.03	1.70	0.680	0.097
	1.75	1.19	0.923	0.278	0.020
	n=24	n=28	n=25	n=23	n=16
8.04 + .48	3.65	4.28	3.96	2.51	0.763
	2.81	2.66	2.34	1.09	0.169
	2.16	1.65	1.39	0.475	0.038
	n=24	n=28	n=25	n=23	n=16
20.2 + 3.9	4.33	4.95	4.59	3.70	1.02
	3.53	3.56	3.00	1.81	0.268
	2.88	2.55	1.97	0.883	0.070
	n=22	n=26	n=23	n=19	n=15

Figure 7. Absolute peak-to-mean concentration ratios as a function of normalized crosswind position.

Figure 8. Absolute peak-to-mean concentration ratios as a function of the time ratio.

Current and Future Research

During the past year the ^{85}Kr system has been greatly expanded. The horizontal coverage of samplers on the 200 and 800 m has been increased and an additional arc has been added at 1600 m. The biggest improvement, however, is in the vertical coverage. Two additional towers have been instrumented on each of the 200 and 800 m arcs and the height of top of the sampling array has been doubled on each arc. The electronics in the data collection system have also been improved so that in future tests it will be possible to examine considerably longer releases and at the same time use shorter sampling increments.

Data from future tests will be analyzed in much the same fashion as the present data, but other more fundamental types of analysis are also planned. In particular, an effort will be made to relate the diffusion directly to turbulence measurements. Analysis of the existing data indicate that at least in some plumes it is possible to identify those frequencies in the turbulent spectra which contribute to plume meander. The diffusion in the instantaneous plumes will be examined following identification and removal of plume meander. A partially completed analysis of existing data indicates that the peak-to-mean ratios in the instantaneous plumes may be only 50% of those reported here, and that the scatter of the individual values may also be significantly reduced.

A complete listing of the potentialities of the ^{85}Kr system is impossible, but they go well beyond those mentioned here. It is obvious that this system provides the opportunity to examine details of the diffusion process which until now have been beyond the reach of experimentalists.

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