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The literature on turbulent concentration fluctuations in atmospheric smoke plumes is reviewed and new models developed for predicting the statistical characteristics of these fluctuations, including the mean and the variance. A method for estimating the integral time scale for concentration fluctuations is suggested, and it is shown how this time scale can be used to calculate the effects of averaging and sampling times on the total variance. The spectra and cross-spectra of concentration and wind speed fluctuations are investigated using data from several field and laboratory experiments, leading to generalizations regarding spectral shape and phase relations between the concentration and wind speed time series.

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## 1. STATEMENT OF PROBLEM STUDIED

Observed and modeled concentration fluctuations in smoke plumes in the atmosphere are quite large, with the standard deviation  $\sigma_c$  at least as large as the mean  $\bar{C}$ . There is interest in concentration fluctuations because of their importance in assessing environmental and toxicological effects, in evaluating models for predicting mean concentrations, and in determining the response of remote sensors pointed towards smoke plumes. During the past ten years there has been a large increase in research on this subject because of the concerns listed above, because of advances in the development of instruments that can measure short-term concentration fluctuations, and because of increases in speed and storage capabilities of computers that some researchers are using to run models of concentration fluctuations.

The purpose of the research performed under this contract has been to analyze field data and theoretical models of turbulent concentration fluctuations and develop methods to predict the statistical characteristics of these fluctuations (mean, variance, probability density function, time or length scale, shape of energy spectrum). This information has not previously been brought together in a comprehensive fashion, since other researchers have emphasized subsets of the overall statistical problem. Our approach has been to acquire as much data as possible from other investigators, and then to use the complete set of data to draw general conclusions.

## 2. SUMMARY OF THE MOST IMPORTANT RESULTS

This three-year research project can be divided into three tasks.

- Theoretical research on the time and space scales of concentration fluctuations.
- Acquisition of field data on concentration fluctuations.
- Time-series analysis of field data.

The results of our research have been reported in six articles in peer-reviewed journals and four articles in conference proceedings, as listed in Section 3. Copies of the first pages of the journal articles are given below in Section 2.1, and the reader is referred to the original articles for details. Because the final journal article has not yet been published, it is summarized in Section 2.2 and is included in this report as Appendix A.

### 2.1 Reproductions of First Pages of Journal Articles

(see the following five pages)

## SPECTRA OF CONCENTRATION FLUCTUATIONS: THE TWO TIME SCALES OF A MEANDERING PLUME

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(First received 30 September 1985 and in final form 8 November 1985)

**Abstract**—The integral time scale,  $T_C$ , of the concentration fluctuations in a narrow, meandering (intermittent) plume can be much less than the time scale,  $T_E$ , of the turbulence in which the plume is immersed, and is generally of the same order of magnitude as the time required for the plume to pass once over a receptor. However the total concentration fluctuation variance is equally strongly influenced by the larger time scale,  $T_E$ , associated with meandering caused by ambient turbulence. It is shown that observed energy spectra of concentration fluctuations can be fitted by a linear combination of two Markov spectra, one with the time scale of the meandering motions and another with the time scale of the small scale plume motions. The two components are weighted by  $I$  and  $1 - I$ , respectively, where  $I$  is the intermittency or fraction of time that non-zero concentrations occur.

**Key word index:** Concentration fluctuations, spectrum of turbulence, meandering plume, dispersion, time scales of plumes, plume transport.

### 1. INTRODUCTION

For many practical applications it is necessary to know the magnitude of instantaneous plume concentration fluctuations and the time scales of the autocorrelogram or spectrum of these fluctuations. Using this information, the variance of the concentration fluctuations can be expressed as functions of averaging time and position. The effects of thresholds and response time of instruments can also be determined. Many researchers (Fackrell and Robins, 1982; Sykes, 1984; Hanna, 1984; Wilson *et al.*, 1985) assume a two-scale system where the concentration fluctuations are due to:

- (i) in-plume turbulence
- (ii) intermittency; i.e. meandering of the plume, leading to periods of zero concentrations.

This two-scale system is based on Gifford's (1959) fluctuating plume theory, which assumes that a narrow plume with an instantaneous standard deviation  $\sigma_{y1}$  meanders back and forth to give a total standard deviation  $\sigma_{yT}$ . The standard deviation of the meandering motions is  $\sigma_{yM}$ , and the three standard deviations are related by  $\sigma_{yT}^2 = \sigma_{y1}^2 + \sigma_{yM}^2$ . If the source aperture is very small, then  $\sigma_{y1}$  is small compared with  $\sigma_{yM}$  at travel times less than the Eulerian time scale,  $T_E$ , of the ambient turbulence. In other words, close to the source the instantaneous plume is narrow and meanders back and forth a great deal. Because of mesoscale and regional fluctuations in wind velocity,  $T_E$  can be quite large and atmospheric plumes can meander a great deal even at large distances from the sources. During convective conditions the meandering at distances of

1 km or more from the source may be masked by the spotty, highly-turbulent nature of the instantaneous plume, as seen in the lidar cross-sections reported by Uthe (1983).

Some of the concepts in Gifford's (1959) fluctuating plume model were used by Sykes (1984) to develop analytical expressions for the integral time scale  $T_C$  of the concentration fluctuations. Assuming that the autocorrelogram was exponential, he derived the result that the integral time scale  $T_C$  decreased steadily as the source was approached. The research reported in this note arose because of the need to determine the relative influence of the time scale related to meandering and the time scale related to the size of the instantaneous plume. This question is first studied by means of analysis of some arbitrary intermittent plumes generated numerically by a computer, and then by means of analysis of some field data. It will be seen that both the large and small time scales are equally important in estimating the standard deviation of concentration fluctuations,  $\sigma_C$ , and that the observed spectra can be simulated by simple analytical expressions.

### 2. A QUALITATIVE EXAMPLE OF AN INTERMITTENT PLUME

Not far downwind of a source with a small aperture, the instantaneous plume is narrow and acts like a wind vane. Vertical fluctuations of the plume are neglected in this analysis. The resulting time series of concentration observed at a fixed point consists of a few sharp spikes of high concentration separated by relatively long periods of zero concentration. This is shown schematically in Fig. 1. In this example the Eulerian velocity field is dominated by a nearly-sinusoidal

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# THE EFFECT OF LINE AVERAGING ON CONCENTRATION FLUCTUATIONS

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(Received 11 May, 1987)

**Abstract.** The reduction in variance of concentration fluctuations due to line averaging is estimated assuming that the process is influenced by the integral distance scale,  $y_i$ , of ambient turbulence and the scaling width,  $W$ , of the time-averaged plume. An analytical formula is derived for the line-averaged variance for situations where the autocorrelogram is exponential and the point variance decreases exponentially with distance from plume centerline. Predictions of concentration fluctuation variance are compared with water tank and field data, with the result that the decrease of variance with averaging distance is well-simulated if the model parameters  $y_i$  and  $W$  are carefully chosen.

## 1. Introduction

In many practical problems such as the health effects of large emissions of  $H_2S$  or the flammability of LNG gas, instantaneous concentration fluctuations at a point are of interest. Theories have been developed and tested for predicting these fluctuations (e.g., Hanna, 1984; Sawford, 1985; Wilson and Simms, 1985). If the statistics of the concentration time series do not vary with time, then Taylor's (1921) theory can be used to relate the variance of concentration fluctuations at a point, averaged over time  $t_a$ , to the variance averaged over time zero:

$$\sigma_c^2(t_a)/\sigma_c^2(0) = 2(t_i/t_a) (1 - (t_i/t_a) (1 - \exp(-t_a/t_i))), \quad (1)$$

where  $t_i$  is the integral time-scale of the turbulent concentration fluctuations. Hanna (1986) suggests a practical method for estimating  $t_i$ , which is a function of the integral time-scale of velocity fluctuations and the time of passage of a plume over a given receptor.

Spatial averaging of concentration fluctuations is also of interest in many applications, such as the line-integrated concentration observed by the eye or an optical device (e.g., laser), or the volume-integrated concentration appropriate to a single breath or a lidar observation. Equation (1) can be used for the spatial averaging problem also, with averaging interval or line  $dy$  substituted for time  $t_a$ , and integral distance  $y_i$  substituted for integral time  $t_i$ , if the statistics of the concentration fluctuations are spatially homogeneous (i.e., do not vary with space). This condition is satisfied only if the averaging line,  $dy$ , is located deep within the plume of air pollutants at all times, or if the averaging line  $dy$  is exposed to a meandering plume equally over its length (see Figure 1). Note that in both these situations the averaging interval or line  $dy$  is well within the time-averaged plume boundary. In these cases, statistics such as the mean  $\bar{C}$ , the variance  $\sigma_c^2$ , higher moments such as the skewness and kurtosis, and the integral

- Hanna S. R. (1984) Concentration fluctuations in a smoke plume. *Atmospheric Environment* 18, 1091-1106.
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### AUTHOR'S REPLY

Dr. Netterville's interesting comments address the problem of simulating fluctuations in the scalar concentration field by means of various combinations of lateral and vertical terms. He and Csanady have developed one analytical approach and I have suggested another in my paper. In a more recent paper (Hanna, 1984) I recommend yet another simple procedure in which the lateral and vertical components are multiplied rather than added.

Netterville objects to my assumptions because of a "lack of theoretical justifications." Most of the research mentioned in his note has been in wind tunnels, where turbulence tends to be more isotropic than in the atmosphere. My concentration fluctuation models were based more on observations in the atmosphere, where lateral meandering continues to be important long after a plume becomes well-mixed vertically. Also, plumes released near the ground are usually subjected to lateral and vertical turbulence with greatly different integral time scales. Because of this anisotropy in the atmosphere, I feel that it is important to account for the influence on concentration fluctuations of both the large scale lateral meanders and the small scale vertical turbulence. The lateral meanders cause concentration fluctuations by bodily moving a plume back and forth over a monitor, while the small scale vertical turbulence causes concentration fluctuations within the relatively well-mixed vertical cross-section of the plume.

The "one-dimensional model" that Netterville objects to can be tested in the atmosphere if the cross-wind integrated (either vertical or lateral) concentration can be calculated from the observations. For example, the lateral integral,  $C^y = \int C dy$ , removes the influence of lateral meanders and can be used to study the concentration fluctuations caused by vertical motions.

At this point in the study of concentration fluctuations in the atmosphere the data are insufficient to warrant choosing one method of combining the components over another. In the future we will, perhaps, have the luxury of closely examining the bases of the various methods that have been suggested.

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### REFERENCE

- Hanna S. R. (1984) The exponential probability density function and concentration fluctuations in smoke plumes. *Boundary-Layer Met.* 29, 361-375.

### A PRINCIPAL COMPONENT ANALYSIS OF SULFUR CONCENTRATIONS IN THE WESTERN UNITED STATES\*

Ashbaugh *et al.* (1984a) apply principal component analysis to a data set of 40 air samplers to obtain spatial patterns of sulfur concentrations. There are several points they posit concerning the application and interpretation of the eigenanalysis procedure which cause concern. Specifically, there are three problem areas: (i) problems with the terminology of the various eigentechniques and the definitions of orthogonal vs oblique solutions, (ii) the Preisendorfer and Barnett eigenvalue truncation test (Rule N) may be misapplied to the rotated solution and misinterpreted to both unrotated and rotated solutions and (iii) the authors state that "In general, however, an orthogonal rotation is desired" which seems to indicate that they are sanctioning orthogonal rotation for most research.

Problems with the terminology appear throughout the text although Section 2 correctly differentiates the factor model from the component model. Furthermore, empirical orthogonal functions or eigenvectors are also confused with components in several instances, most notably in Figs 2-11 where what actually are PC loadings are referred to as eigenvectors. Since these three models can lead to different solutions, a distinction should be made so that other researchers can reproduce the results presented. Part of the terminology problem may arise from the widespread use of statistical packages for principal component analysis. Many of the packages freely intermix the terms for components and factors, thoroughly confusing users. The precise definitions of these terms can be found in standard texts on factor analysis by Harman (1976), Mulaik (1972), Rummel (1970) and in the report by Lorenz (1956) for empirical orthogonal functions which all clearly delineate the differences.

- (i) Empirical Orthogonal Functions (EOFs) are simply the unit length eigenvectors and corresponding eigenvalues given by the decomposition of a correlation matrix  $R$  as  $RV = VD$  where  $V$  is the orthogonal matrix of eigenvectors and  $D$  is the diagonal matrix of eigenvalues. In the meteorological literature, the term EOF was introduced by Lorenz (1956, p. 20) where he denotes  $V$  as empirical orthogonal functions of space (due to their spatial orthogonality). The corresponding time series,  $X$ , is given by  $Z = XV$  where  $Z$  are the standardized input data (assumed standardized for this discussion but not a necessary condition) and  $X$  are the empirical orthogonal functions of time (also called 'principal axes' in the statistical literature and 'time-dependent amplitudes' in the meteorological literature). It should be noted that the time/space definitions provided by Lorenz are tailored to his specific application where a series of weather stations are related through time for one parameter (sea-level pressure). This defines a S-mode analysis and is only one of six basic ways of interrelating data for an eigenanalysis (Ruchman, 1983a, p. 66). This type of decomposition (S-mode) is the same as Ashbaugh *et al.* who relate one parameter (sulfur concentration) over a series of stations (38 sites) through time (months from September 1979 to September 1980) to yield a correlation matrix relating the sites in an analysis. Under another type of decomposition, where the correlation matrix related the observations (T-mode), the  $V$  would represent EOFs of time while  $X$  would represent EOFs of space.
- (ii) Principal components (PCs) are slightly more elegant mathematically compared to EOFs as the spatial

\*Ashbaugh L. L., Myrup L. O. and Flocchini R. G. (1984a) *Atmospheric Environment* 18, 783-791.

tation of some of the existing National Ambient Air Quality Standards.

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Chock D. P. (1985) Statistics of extreme values of air quality—a simulation study (to be submitted).

## GROUND-LEVEL CONCENTRATION FLUCTUATIONS FROM A BUOYANT AND A NON-BUOYANT SOURCE WITHIN A LABORATORY CONVECTIVELY MIXED LAYER

Deardorff and Willis (1984) and Willis and Deardorff (1983) report on mean plume rise and diffusion and concentration fluctuations observed in buoyant and non-buoyant plumes released in their laboratory's convective tank. Lamb (1982) and Moninger *et al.* (1983) have shown that properly-scaled mean plume observations in the convective tank are similar to observations in the real atmosphere. Appropriate scaling parameters are the mixing depth,  $h$ , and the convective velocity scale,  $w_* = (ghH/c_p\rho)^{1/3}$ , where  $H/c_p\rho$  is the kinematic surface heat flux. In this note it will be shown that a simple analytical model for concentration fluctuations provides a good fit to the convective tank observations.

Probability distribution functions of pollutant concentrations can usually be fitted by a two-parameter analytical formula, such as the clipped-normal (Lewellen and Sykes, 1983), the log-normal with intermittency (Wilson, 1982) or the exponential with intermittency (Barry, 1977; Hanna, 1984). Intermittency,  $I$ , is defined as the fraction of non-zero readings in the time-series of concentrations at a given monitor. In any of the three suggested formulas the full distribution is completely determined once any two of the three parameters ( $I$ ,  $\sigma_c$ ,  $\bar{C}$ ) are specified, where  $\sigma_c$  and  $\bar{C}$  are the standard deviation and the mean of the concentration data. Wilson (1982) and Hanna (1984) show that the exponential distribution is more valid for atmospheric observations around a single point source, where strong variations in wind direction nearly always occur and the resulting concentration time series is highly intermittent. For an exponential distribution function, the following formula can be used to predict the ratio of the standard deviation of the concentration fluctuations to the mean:

$$\sigma_c/\bar{C} = ((2/I) - 1)^{1/2}. \quad (1)$$

The intermittency at a receptor beneath the plume centerline can be estimated from a formula derived by Hanna (1984):

$$I = (\sigma_{y1}\sigma_{z1}/\sigma_{yT}\sigma_{zT}) \exp(-h_e^2/2\sigma_{zT}^2). \quad (2)$$

where subscript I and subscript T refer to the instantaneous and time-mean plume spreads, and  $h_e$  is the elevation of the mean plume centerline above the receptor. The ratio  $\sigma_{y1}/\sigma_{yT}$  is given by a formula suggested by Gifford (1982) and Lee and Stone (1983) based on a solution to the Langevin equation:

$$\frac{\sigma_{y1}^2}{\sigma_{yT}^2} = \frac{\hat{\sigma}_0^2 + T' - (1 - e^{-T'}) - 0.5(1 - e^{-T'})^2}{\hat{\sigma}_0^2 + T' - (1 - e^{-T'})} \quad (3)$$

where  $T' = t/T_L$  and  $T_L$  is the Lagrangian time scale of the velocity fluctuations. The parameter  $\hat{\sigma}_0$  equals  $\sigma_0/\sqrt{2\sigma_{yT}}$ , where  $\sigma_0$  is the source size. Hanna (1984) presents a figure which contains graphical solutions to Equation (3) for several assumed values of  $\hat{\sigma}_0^2$ .

In order to apply these formulas to the convective tank data, the following assumptions are made, based on information in the papers by Deardorff and Willis (1984) and Willis and Deardorff (1983):

$$\sigma_0 = 0.0078 h, T_L = 0.5 h/w_*, \sigma_z = 0.6 w_*$$

$$h_e = 0.15 h \text{ non-buoyant}$$

$$h_e = 0.40 h \text{ buoyant (assumes a receptor height of } 0.086 h)$$

Lamb (1982) has derived the empirical formulas  $\sigma_z/h = X/2$  for  $X < 2/3$  and  $\sigma_z/h = 1/3$  for  $X > 2/3$  from previous experiments in Deardorff's tank, where dimensionless downwind distance  $X$  equals  $xw_*/uh$ . However, Lamb states that this formula is only approximate at  $X$  less than one because of the non-Gaussian shape of the vertical velocity distribution, and is uncertain for  $X$  greater than about one because the vertical distribution becomes nearly uniform beyond that point. (For a uniform distribution  $\sigma_z/h = 0.3$  from the definition of the second moment.) In order to assure that the exponential term in Equation (2) approaches unity, which will better parameterize the effects of a uniform distribution at large  $X$  on concentration fluctuations, the formula  $\sigma_z/h = X/2$  is used for all  $X$  in our application.

It is also assumed that the vertical and lateral turbulence and diffusion are homogeneous in the convective boundary layer, so that Equation (3) applies equally to  $\sigma_y$  and  $\sigma_z$ . Resulting predictions of  $\sigma_c/\bar{C}$  and  $I$  are compared with the observations in Figs 1 and 2, respectively. The predicted lines tend to pass through the observed points on the figures. The biggest disagreements between observations and predictions occur at large  $X$  in Fig. 1, where observed  $\sigma_c/\bar{C}$  drops to less than 0.5 while predicted  $\sigma_c/\bar{C}$  remains at about unity. This disagreement may be due to the constraining effects of the tank walls at large downwind distances, which limit the plume meandering and make the exponential pdf assumption invalid. After sufficient time has elapsed in the tank,  $\sigma_c/\bar{C}$  would approach zero, similar to the fluctuations in cream concentrations in a cup of coffee. In the atmosphere, no such constraints exist (except in valleys) and Gifford (1982) has shown that meandering exists at time scales out to several days.

These calculations have assumed that concentrations are observed in an infinitely small volume. Deardorff and Willis (1984) state that the finite averaging distance (about 0.01  $h$ ) for the concentration measurement in their tank may reduce the effective observed  $\sigma_c/\bar{C}$  by about 10% from what it would be for instantaneous measurements in an infinitesimal volume. This effect can be studied using an equation suggested by Venkatram (1979) and assuming that  $x_A/x_1$  equals  $t_A/t_1$ , where  $x_1$  and  $t_1$  are integral distance and time scales for the concentration fluctuations:

$$\sigma_c(x_A)/\sigma_c(0) = \left(2 \frac{x_1}{x_A} \left(1 - \frac{x_1}{x_A} (1 - \exp(-x_A/x_1))\right)\right)^{1/2}. \quad (4)$$

The parameter  $x_A$  is the averaging distance. As Sykes (1984) has shown,  $x_1$  for concentration fluctuations is not necessarily equal to the integral distance scale for velocity fluctuations ( $uT_L$ ), he suggests a simple formula for calculating the ratio of the integral scale of the concentration fluctuations,  $x_1$ , to the integral scale of the velocity fluctuations,  $uT_L$ , near the center of the plume:

$$x_1/uT_L = 0.5(\sigma_c/\bar{C})^{-2} \ln(1 + 2\sigma_c/\bar{C}) \quad (5)$$

Deardorff and Willis' data show that  $\sigma_c/\bar{C}$  is about 2 at  $X$  equal to 1.0, and Equation (5) predicts that  $x_1/uT_L$  would equal 0.20 at that distance. Consequently the ratio  $x_A/x_1$  is about 0.20 at  $X$  equal to 1.0, and the ratio of the standard deviations  $\sigma_c(x_A)/\sigma_c(0)$  is predicted by Equation (4) to be

# Air Quality Model Evaluation and Uncertainty

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The many advances made in air quality model evaluation procedures during the past ten years are discussed and some components of model uncertainty presented. Simplified statistical procedures for operational model evaluation are suggested. The fundamental model performance measures are the mean bias, the mean square error, and the correlation. The bootstrap resampling technique is used to estimate confidence limits on the performance measures, in order to determine if a model agrees satisfactorily with data or if one model is significantly different from another model. Applications to two tracer experiments are described.

It is emphasized that review and evaluation of the scientific components of models are often of greater importance than the strictly statistical evaluation. A necessary condition for acceptance of a model should be that it is scientifically correct. It is shown that even in research-grade tracer experiments, data input errors can cause errors in hourly-average model predictions of point concentrations almost as large as the predictions themselves. The turbulent or stochastic component of model uncertainty has a similar magnitude. These components of the uncertainty decrease as averaging time increases.

There is increasing interest in the evaluation of air quality models and the assessment of typical values for the uncertainties of these models. The purpose of this paper is to review previous work on this subject, suggest a simplified model evaluation procedure, and provide examples of the application of this procedure. Estimates are made of the contributions of model input errors and stochastic uncertainties to the total model error.

## Background on Air Quality Model Evaluation

### Early Model Evaluation Studies (Pre-1980)

A great increase in the number of air quality model evaluation studies took place about 1980, since U.S. EPA procedures<sup>1</sup> were standardized at that time and many research studies were begun in order to further improve the system. Prior to that time, air quality model evaluations were often conducted on an arbitrary, ad hoc basis. A characteristic of the early studies was a lack of understanding of the expectations for models. For example, it is now recognized that models cannot produce adequate correlations between hourly predictions and observed data paired in time and space. However, models

can simulate the ground-level patterns of the concentrations fairly well, if the center of the pattern is free to move in space. Also, it is clear that models should be applied only to physical situations similar to those used to derive the model. A model derived from near-ground data over a flat agricultural field should not be applied to dispersion from a tall stack in complex terrain. This knowledge has been used over the past five years to develop improved model evaluation procedures.

### U.S. EPA and EPRI Model Evaluation Procedures (Post-1980)

In response to the inconsistent and arbitrary model performance evaluations that were typical of the earlier period, and the need for standardized model evaluation procedures generated by the U.S. Clean Air Act and its amendments, both the Electric Power Research Institute (EPRI)<sup>2</sup> and the U.S. EPA<sup>3</sup> sponsored workshops on air quality model performance evaluations. The aim of both groups was to develop a set of operational procedures for evaluating models for short-range dispersion from industrial plants. In both workshops, concern was expressed about the absolute rather than the statistical nature of air quality standards (e.g., the second-highest concentration is used as a standard,

whereas the 90th percentile of the cumulative distribution would be more robust). It was recognized that data used for evaluation purposes should be independent and should be normalized (i.e., transformed into a variable with a normal or Gaussian distribution).

As a result of the recommendations of the workshops, EPA has produced a guideline on procedures to be followed in evaluating air quality models.<sup>1,4</sup> Lists of recommended statistical measures are given, emphasizing the mean variance and gross variability of the residuals (predicted minus observed concentrations), as well as the correlation. The tables of recommended performance measures are too lengthy to be reproduced here. These measures are intended to be applied to a wide variety of subsets of the original data, such as the maximum concentrations during a given hour observed and predicted anywhere over a monitoring network. It is stated that in order to calculate a single "score" in any given model evaluation application, the various performance measures are to be summed using a weighting system arbitrarily selected based on the major problems at that site. This weighting system must be agreed to by all involved parties (e.g., the state agency, the EPA, and the industry). Generally, the weighting procedure recognizes the desirability of having a model overpredict rather than underpredict.

During the past five years the EPA and EPRI have sponsored several test applications of their recommended procedures. A committee composed of EPA and American Meteorological Society (AMS) representatives oversaw one part of these evaluations and reported some of the results in the *Bulletin* of the AMS.<sup>5</sup> This particular exercise involved ten models under consideration by the EPA for application to dispersion from industrial plants in a flat rural environment, and both a scientific and a statistical evaluation took place. It should be mentioned that all ten models have a Gaussian structure, with slightly different methods of estimating stability and dispersion parameters.

The EPA/AMS committee concluded that none of the models was up-to-

## 2.2 Overview of Results of Time Series Analysis

The journal articles whose first pages are reproduced in Section 2.1 represent the results of research conducted during the first two years of this project. During the past year, emphasis has been on time series analysis of several field data sets. The results of this work have just recently been incorporated in a manuscript submitted to Boundary Layer Meteorology (see Appendix A). An overview of the highlights of this research is given below:

### Purpose of Study

This research project has the purpose of conducting time series or spectral analyses on concentration fluctuation field data sets from six independent experiments (Smoke Week III, Dugway Proving Ground Trials, WSU Palouse Wheat Field Trials, WSU Hanford Trials, Australia Two Source Project, Deardorff-Willis Convection Tank). The slopes of the concentration fluctuation spectra in the inertial subrange are of interest, as well as the time period showing peak spectral energy. Cross-correlograms and cross-spectra between concentration and turbulent wind fluctuations are investigated to determine the magnitude of the correlation and whether there is any particular eddy period that strongly contributes to the cross-correlation. If this were true, then observations of wind speed fluctuations could be used to infer the characteristics of the concentration fluctuations. Clearly the sampling and averaging times and distances are important. In addition, if the concentrations are due to continuous emissions from a point source and the plume has width  $d$  at the downwind distance where a monitor is located, then all turbulent speed oscillations or eddies with scales larger than  $d$  will cause the plume only to meander or wave back and forth. These oscillations will show up in the concentration time record as an intermittent series of non-zero concentrations separated by finite periods with zero concentrations.

### Conclusions

Analyses of concentration fluctuation ( $C'$ ) spectra and co-spectra from field experiments at four separate locations yield results that are more revealing for the spectra than the co-spectra. The  $C'$  spectra from these experiments generally show an inertial subrange with a slope of  $-5/3$  and indicate peak energy at a time periods of about 50 to 100 s. These periods

are a factor of two to five less than those for the peak of the  $u'$  or  $v'$  spectra. A general spectral formula fits normalized spectra from the U.S. and Australia. Cross-spectra indicate little relation between  $C'$  and  $u'$ ,  $v'$ , or  $w'$ . However, most of the correlation that exists is due to periods larger than about 10 to 20 s.

### 3. LIST OF PUBLICATIONS

#### Journal Articles

Hanna, S.R., 1985: Reply to Comments by D. Netterville, Atmos. Environ., 19, 1972.

Hanna, S.R., 1985: Ground-Level Concentration Fluctuations from a Buoyant and a Non-Buoyant Source within a Laboratory Convectively Mixed Layer. Atmos. Environ., 19, 1210-1212.

Hanna, S.R., 1986: Spectra of Concentration Fluctuations: The Two Time Scales of a Meandering Plume. Atmos. Environ., 20, 1131-1137.

Hanna, S.R., 1987: The Effect of Line Averaging on Concentration Fluctuations. Bound. Lay. Meteorol., 40, 329-338.

Hanna, S.R., 1988: Air Quality Model Evaluation and Uncertainty. J. Air Poll. Control Assoc., 38, 406-412.

Hanna, S.R. and E. Insley, 1988: Time Series Analyses of Concentration and Wind Fluctuations. Submitted to Bound. Lay. Meteorol.

#### Conference Proceedings

Yamartino, R.J. and S.R. Hanna, 1985: A Strand Theory of Concentration Fluctuations, Proc. of Seventh Symposium on Turbulence and Diffusion, AMS, Boston, MA, 252-258.

Hanna, S.R., 1985: Time Scales of an Intermittent Plume. Proc. of 1985 Scient. Conf. on Chem. Defense Res., CRDC-SP-86007, 143-147.

Hanna, S.R., 1986: Accuracy of Plume Concentration Fluctuation Statistics Calculated from a Limited Number of Lidar Cross-Sections. Proc. of Smoke/Obscurants Symposium X, Paper B-10, 4 pp.

Hanna, S.R. and E. Insley, 1988: Time Series Analyses of Concentration and Wind Fluctuations. Proc. of Eighth Symposium on Turbulence and Diffusion, AMS, Boston, MA, 119-122.

4. LIST OF MEETINGS ATTENDED

Nov. 20, 1985, Boulder, Co.

A paper on the strand theory of concentration fluctuations was presented by Dr. Yamartino at the AMS Seventh Symposium on Turbulence and Diffusion.

April 16, 1986, and October 14, 1986, Dugway Proving Ground, Utah

Field experiments on concentration fluctuations at Dugway Proving Ground were discussed with J. Bowers and K. Dumbauld. Travel expenses were paid by a separate project but the data were subsequently analyzed as part of our study of concentration and wind spectra.

April 28, 1988, San Diego, CA

A paper on spectral analysis of concentration fluctuations was presented by Dr. Hanna at the AMS Eighth Symposium on Turbulence and Diffusion.

Note: The Smoke/Obscurant and CRDC Symposia have been closed since 1986 to anyone without a security clearance. We published two papers in their proceedings but have not attended any of the conferences because we do not have a security clearance.

5. LIST OF ALL PARTICIPATING SCIENTIFIC PERSONNEL

This research was conducted by Environmental Research and Technology, Inc. (ERT) in Concord, MA, and by Sigma Research Corp. (SRC) in Lexington, MA. The following scientific personnel participated in various stages of the research:

Dr. Steven Hanna (ERT, SRC)

Dr. Robert Yamartino (ERT, SRC)

Mr. Jonathan Pleim (ERT)

Ms. Elizabeth Insley (SRC)

No advanced degrees were awarded during the course of this project.

APPENDIX A

TIME SERIES ANALYSIS OF CONCENTRATION  
AND WIND FLUCTUATIONS

(submitted to Boundary Layer Meteorology)

To be submitted to  
Boundary Layer Meteorology

TIME SERIES ANALYSES OF CONCENTRATION  
AND WIND FLUCTUATIONS

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## Abstract

Analyses of concentration fluctuation ( $C'$ ) spectra from boundary layer smoke plume experiments at six separate locations show that the spectra from these experiments generally exhibit an inertial subrange at high frequencies with a slope of  $-5/3$  and indicate peak energy at a time periods of about 50 to 100 s. These periods of peak energy are a factor of two to five less than those for the peak of the wind speed fluctuation ( $u'$  or  $v'$ ) spectra. A general spectral formula fits normalized spectra from the U.S. and Australia, where the frequency,  $n$ , is made dimensionless by multiplying by the plume dispersion parameter,  $\sigma_y$ , and dividing by the wind speed,  $u$ . Peak energy occurs at a dimensionless frequency of  $n\sigma_y/u$  equal to about 0.15. The Kolmogorov constant in the inertial subrange is estimated from a set of averaged spectra. Cross-spectra indicate little relation between concentration and wind fluctuations. However, most of the correlation that exists is due to periods larger than about 10 or 20 s.

## 1. Purpose of Study

The boundary layer of the atmosphere is characterized by a relatively high degree of turbulence, with fluctuations in one-second averages of variables such as wind speed typically having magnitudes roughly equal to 10% to 100% of the mean value. When pollutants are emitted into this boundary layer velocity field, they are carried about by turbulent eddies and also are observed to exhibit fluctuations of the same order of magnitude as their mean value (Csanady 1973). These fluctuations are of great practical importance when the pollutant is a highly toxic or flammable material. Most dispersion models are capable of predicting the mean pollutant concentration, but provide no guidance on the probability of fluctuations from this mean.

Recently there have been several studies completed of the probability distribution of concentration fluctuations. Sykes (1984), Hanna (1984) and Sawford et al. (1986) agree that the distribution of concentration fluctuations is skewed towards higher values, but disagree on the exact form of the optimum distribution function (exponential, log-normal, or clipped normal). These authors, as well as several other researchers, have suggested analytical formulas for the mean,  $\bar{C}$ , and the standard deviation,  $\sigma_c$ , of the concentration fluctuations. They find that the fluctuation intensity,  $\sigma_c/\bar{C}$ , varies with distance from the source, position within the plume, averaging time, and the time scale of atmospheric turbulence. However, observations or models of the mean, the variance, or the probability distribution can provide no information on the eddy sizes that cause the observed fluctuations.

With the advent of practical instruments for measuring high-frequency (1 Hz or better) variations in concentration (e.g. Hadjitafti and Wilson 1979 and Jones 1983), a few field and laboratory experiments have been completed in which time series of pollutant concentration have been observed. In most cases, concurrent observations of wind and temperature fluctuations have also been observed in these experiments. Consequently, it is now possible to investigate the spectral characteristics of concentration time series, such as the variation of fluctuation intensities with the period of turbulent eddies, and the relation between the wind and concentration time series. We have acquired six of these independent experimental data sets, and have applied

time series analysis procedures to the available concentration and wind fluctuation data.

The purpose of the study is to answer the following questions:

- 1) Can the concentration energy spectrum be represented by a generalized analytical formula?
- 2) Does the concentration energy spectrum have an inertial subrange and, if so, can the constant in the Kolmogorov formula be estimated?
- 3) What is the relation between the periods with peak energy in the concentration and wind spectra?
- 4) What is the shape of the cross-spectrum between concentration and wind fluctuations?

The following sections describe the theoretical basis for concentration fluctuation spectra, the data bases that were analyzed, the time series analysis procedures, and the spectra and cross-spectra results.

## 2. Theoretical Basis for Concentration Fluctuation Spectra

It is assumed that a time series of concentration fluctuations is available with a total sampling length of about one hour and a sampling interval or averaging time on the order of one second (i.e., the record consists of a few thousand data points). The moments of the time series can be calculated (mean, variance, skewness, kurtosis, etc.), but they reveal nothing about the time periods of the variations in concentration. To better estimate toxic response or flammability of hazardous pollutants, it is necessary to know whether the dominant period of the fluctuations is 5 sec, 50 sec, or 500 sec. Furthermore, if the concentration fluctuations are somehow tied to wind speed fluctuations, it may be possible to predict the spectral characteristics of concentration fluctuations knowing only the characteristics of wind speed fluctuations.

The idea for this research was formed while viewing videotapes of smoke plumes from ground level continuous sources at Dugway Proving Ground. During daytime experiments with light winds and strong surface heating, portions of the continuous plume were carried upward by convective eddies so that the plume looked more like a series of misshapen teardrops (see Figure 1). The postulated turbulent wind fluctuation field that would lead to these shapes is drawn on the figure. If this relationship were true, then high values of concentration would be correlated with low values of wind speed and positive values of vertical velocity (i.e., updrafts). As Sawford et al. (1985) point out, the scale of atmospheric wind fluctuations that influence the plume fluctuations will change as the plume size increases. Figure 2 illustrates a plume imbedded in a field of eddies of uniform size, showing that when the plume is smaller than the eddies, the eddies advect (meander) the plume back and forth, but when the plume is larger than the eddies, they no longer advect the whole plume but merely cause minor fluctuations deep within the plume.

Often it is possible to estimate the dominant period in a time series by visual inspection of the time series (i.e. the graph of concentration plotted versus time). But usually there are so many time periods acting that the time series looks like a random set of fluctuations. Spectral analysis can be used

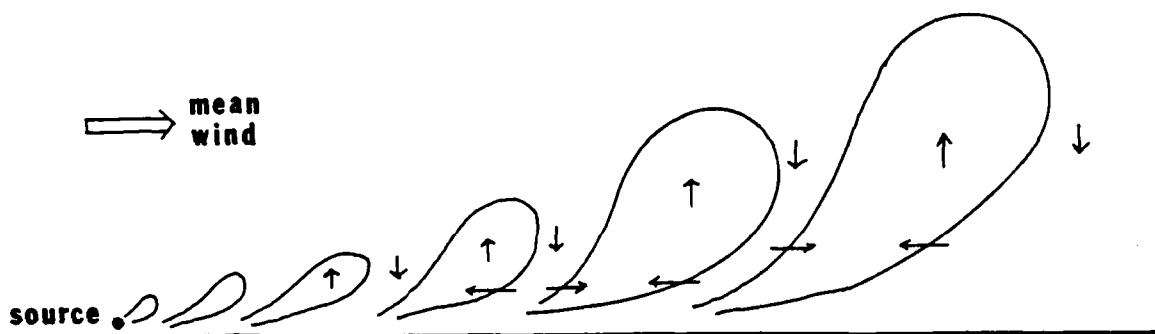


Figure 1. Schematic drawing of the sideview of a smoke plume in a convective atmosphere with  $\sigma_w/u > 1$ , where  $\sigma_w$  is the standard deviation of vertical wind fluctuations and  $u$  is the mean wind speed. The small arrows represent turbulent velocity vectors, as perturbations from the mean wind vector.

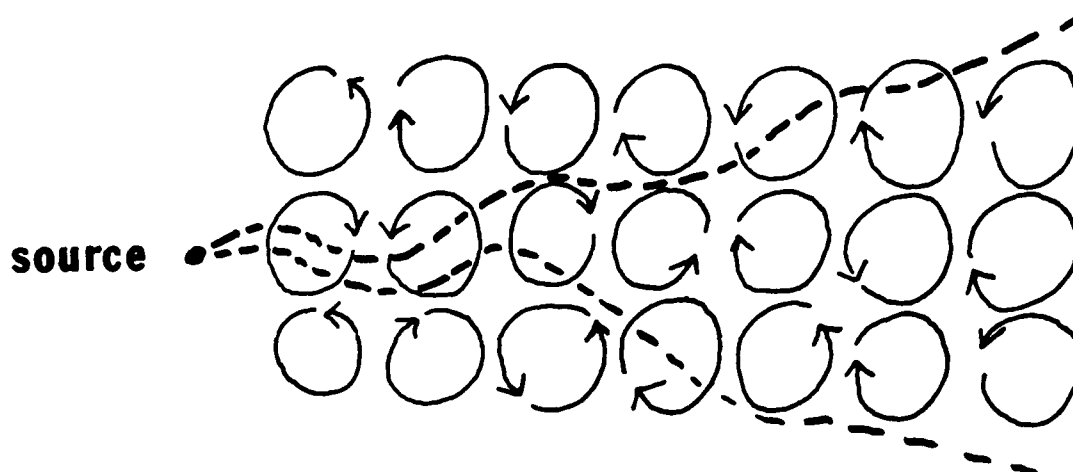


Figure 2. Illustration of the influence of eddies of a given size on a smoke plume. Initially, the eddies transport the plume laterally; later, they diffuse the plume.

to estimate the fraction of the total variance or energy that is due to eddy periods in a certain narrow range. Lumley and Panofsky (1964) introduce the concept of atmospheric spectral analysis, and Panofsky and Dutton (1984, pp. 169-209) provide a detailed discussion of theoretical aspects and give many examples. There is much interest in the inertial subrange of eddies, where energy is being transferred from the larger energy-generating eddies to the smaller energy-dissipating eddies. In the inertial subrange, simple scaling relations may be valid. For example, much work has been done on the inertial subrange of wind speed spectra, which can be expressed in the form:

$$k S_u(k) = a_u \epsilon^{2/3} k^{-2/3} \quad (1)$$

where  $k$  is wavenumber (in cycles/m,  $S$  is wind speed spectral energy per unit wavenumber (in  $m^3/sec^2$ ),  $a_u$  is the Kolmogorov constant (about 0.5), and  $\epsilon$  is the eddy dissipation rate (in  $m^2/sec^3$ ). Taylor's "frozen-turbulence" hypothesis is invoked to convert from wavenumber,  $k$ , to frequency,  $n$ :

$$k = n/u \quad (2)$$

Taylor's hypothesis has been shown to be valid for wind spectra, but has never been demonstrated for concentration spectra.

An inertial subrange can be postulated for fluctuations of any conservative variable,  $x$ , in the atmospheric boundary layer:

$$n S_x(n) = a_x \epsilon_x^{2/3} (n/u)^{-2/3} \quad (3)$$

where  $\epsilon_x$  is the dissipation rate of fluctuations of  $x$  by molecular forces and  $a_x$  is the Kolmogorov constant appropriate for  $x$ . It is desirable to use scaling parameters to normalize equation (3) so that it is universally valid. Monin-Obukhov similarity theory is used for variables,  $x$ , whose statistics are spatially homogeneous. For example, the wind speed fluctuation variance,  $\sigma_u^2$ , the mean wind speed,  $u$ , and the momentum and heat fluxes would be spatially homogeneous (constant) at a height of 2m over a broad grassy field. Other variables that satisfy this criterion are the pressure, the temperature, and the relative humidity. Similarity theory then permits equation (3) to be written in the form:

$$nS_x(n)/\sigma_x^2 = a_x \epsilon_x^{2/3} (n/u)^{-2/3} / \sigma_x^2 \quad (4)$$

where the dissipation rate can be expressed as:

$$\epsilon_x = (X^*/0.4z) \phi_{\epsilon x}(z/L) \quad (5)$$

where  $z$  is elevation above the ground (in m),  $L$  is the Monin-Obukhov length (in m), and  $X^*$  is the scaling value for parameter  $x$ , defined by

$$X^* = F_*/u_* \quad (6)$$

The parameter  $F_*$  is the upward turbulent vertical flux of  $x$  (also defined as  $\overline{w'x'}$ , where  $w'$  and  $x'$  are fluctuations in vertical speed and  $x$  and the overbar indicates a time average) and  $u_*$  is the friction velocity ( $u_* = (-\overline{u'w'})^{1/2}$ ). If  $x$  is vertical velocity, then  $X^* = u_*$  and  $\phi_{\epsilon w}(z/L) = 1 - z/L$  (Panofsky and Dutton, 1984). The generalized stability function  $\phi_{\epsilon x}(z/L)$  is defined so that it equals 1.0 in neutral conditions ( $z/L = 0$ ).

Panofsky and Dutton suggest formulas for the scaling relations given above for scalars such as temperature, relative humidity, and pressure. However, the turbulent flux,  $F_c$ , of pollutant concentration,  $C$  (units  $\mu\text{g}/\text{m}^3$  or ppm), in a smoke plume is spatially homogeneous only if the pollutant is uniformly emitted over a broad area source with area  $1 \text{ km}^2$  or greater. For example, radon may satisfy this criterion, since it is emitted nearly uniformly from the ground. If the pollutant is emitted from a point source, the turbulent flux,  $F_c$ , will be directed radially outward from the plume axis and thus can change sign and magnitude depending on the location of the monitor within the plume. Furthermore, the mean and variance in concentration fluctuations,  $\bar{C}$  and  $\sigma_c^2$ , vary with downwind distance and with crosswind position. Consequently it is unlikely that Monin-Obukhov similarity relations will apply to concentration fluctuation spectra. However, it is possible that some sort of scaling relations will be valid locally. Consider the total variance,  $\sigma_c^2$ , as one scale and the plume dispersion parameter  $\sigma_y$ , as another. Then we can postulate that the following relation is valid:

$$nS_c(n)/\sigma_c^2 = f(n\sigma_y/u) \quad (7)$$

where  $f$  represents a universal function. Thus, if  $nS_c(n)/\sigma_c^2$  is plotted versus  $n\sigma_y/u$ , it is hoped that all observations will fall along some general curve.

Wilson and Simms (1985) and Hanna (1986) have postulated that the simple Markov spectrum may be valid for concentration fluctuations:

$$nS_c(n)/\sigma_c^2 = (2/\pi)nT_{pc}/(1 + (nT_{pc})^2) \quad (8)$$

where  $T_{pc}$  is the period with peak spectral energy. This formula is derived from the exponential autocorrelogram:

$$R(t) = \exp(-t/T_c) \quad (9)$$

where the integral time scale,  $T_c$ , equals  $T_{pc}/2\pi$  for sinusoidal fluctuations. The large- $n$  asymptote of equation (8) is  $nS_c(n) \propto n^{-1}$  rather than the  $nS_c(n) \propto n^{-2/3}$  relation of equation (7). Both equations (7) and (8) will be tested with the field data.

### 3. Description of Data

During the past five years, a number of fast-response concentration fluctuation data sets have appeared in the literature. The data tapes and descriptions of the experiments were requested from each principal investigator. In a few cases the data could not be used because of problems such as poorly formatted data tapes, but in most cases it was possible to analyze the concentration fluctuation time series. In most data sets, concurrent time series of wind speed fluctuations were also available. A summary of the characteristics of the data sets is given in Table 1, and brief descriptions of the data are given below. Except for the Deardorff-Willis data set, all source releases were non-buoyant.

Smoke Week III. The U.S. Army sponsors an annual field experiment in which smoke/obscurant devices are tested and concurrent fast response aerometric and meteorological data are taken. Nearly all of the sources are complicated multiple releases of time-varying emissions. Only a few experiments employ simplified continuous releases of smoke suitable for our analysis. Trial 2 in Smoke Week III took place at Eglin AFB, Florida, and has been analyzed previously by Hanna (1984). The fog oil source was located at a height of 1m at a distance of about 70m upwind of a line of aerosol photometers oriented across the plume. Point measurements of particle concentrations were made by each instrument at a height of 1m. In our analysis, spectra were averaged over the five monitors that were nearest to the center of the plume. A meteorological tower was located at the mid-point of the line. The experiment took place during the daytime with light-to-moderate winds. Only 300 seconds of data were available due to the short duration of the fog oil release.

Dugway Trials. Fog oil was also used as source in field tests at Dugway Proving Ground (Bowers and Black 1985). The source was again located at a height of 1m, but in this case the continuous release extended over a one hour period. Cross-wind integrated concentrations at an elevation of 1m and a distance of 50 m downwind (at the point the observed line crossed the plume centerline) were recorded by a transmissometer looking perpendicular to the plume centerline. A meteorological tower was located close to the plume centerline near the line-of-sight of the transmissometer. All data were available as one second averages. Two trials were analyzed: one during

Table 1

## Characteristics of Experiments

| Test Name                                          | Downwind Distance (m)                  | Number of Runs | Duration of Runs | Averaging Time (sec) | Average Wind Speed (m/s) | Wind Speed Range (m/s) | Stability           |
|----------------------------------------------------|----------------------------------------|----------------|------------------|----------------------|--------------------------|------------------------|---------------------|
| <u>Smoke Week III (fog oil):</u>                   |                                        |                |                  |                      |                          |                        |                     |
| Trial 2                                            | 70                                     | 1              | 300 sec          | 1                    | 3.0                      | 0.8 - 4.6              | Moderately Unstable |
| <u>Dugway Trials (fog oil):</u>                    |                                        |                |                  |                      |                          |                        |                     |
| Trial T091                                         | 50                                     | 1              | 3560 sec         | 1                    | 6.6                      | 2.9 - 11.4             | Nearly Neutral      |
| Trial T011                                         | 50                                     | 1              | 3400 sec         | 1                    | 3.0                      | 0.5 - 6.5              | Moderately Unstable |
| <u>WSU - Palouse Wheat Field Study 1985 (SF6):</u> |                                        |                |                  |                      |                          |                        |                     |
| P3                                                 | 100                                    | 1              | 10 min           | 0.5                  | 11.6                     | 5.2 - 18.7             | Nearly Neutral      |
| P18                                                | 100                                    | 1              | 20 min           | 0.5                  | 2.1                      | 1.1 - 3.0              | Stable              |
| <u>WSU - Hanford Diffusion Grid</u>                |                                        |                |                  |                      |                          |                        |                     |
| Unstable                                           | 192                                    | 1              | 34 min           | 0.5                  | 2.2                      | 0.5 - 4.5              | Unstable            |
| <u>Sawford - 2 Source Project (SF6, P):</u>        |                                        |                |                  |                      |                          |                        |                     |
|                                                    | 25                                     | 23             | 1 hr             | 6                    | 4.8                      | 1.5 - 9.0              | Slightly Unstable   |
|                                                    | 50                                     | 6              | 1 hr             | 6                    | 5.6                      | 4.5 - 7.5              | Slightly Unstable   |
|                                                    | 100                                    | 15             | 1 hr             | 6                    | 4.1                      | 2.0 - 6.5              | Slightly Unstable   |
| <u>Deardorff-Willis Tank</u>                       |                                        |                |                  |                      |                          |                        |                     |
|                                                    | 500                                    | 1              |                  |                      | 5                        |                        | Unstable            |
|                                                    | (Instantaneous cross-section of plume) |                |                  |                      |                          |                        |                     |

nearly-neutral conditions and another during unstable conditions (this trial inspired the schematic drawing in Figure 1).

WSU - Palouse Wheat Field Study. Lamb et al. (1985) discuss an  $\text{SF}_6$  tracer study conducted by Washington State University (WSU) at their Palouse wheat field experimental site. Fast response (20 Hz) point measurements of  $\text{SF}_6$  concentration and wind fluctuations were made at a single site at a distance of about 100 m from the source during several runs. Source height and monitor height were each about 1.5 m. The duration of the runs was on the order of 10 to 20 minutes. The 20 Hz data were averaged to produce a data record consisting of 2 Hz averages. Otherwise the storage limits in the spectral analysis program would have been exceeded. Because there was only one monitor, it was difficult to assure that the plume would blow right over the monitor, and during several runs the sporadic nature of the concentration time series suggested the monitor was off the edge of the time-mean plume. We selected two runs where the data indicated that the time-mean plume was over the monitor.

WSU - Hanford Diffusion Grid. Peterson et al. (1988) discuss the results of another test of the WSU system at the Hanford, WA, diffusion grid. The instrument set-up was the same as above, except that the source and the concentration monitor were at a height of 3.9 m and the monitor was at a distance of 192 m from the source. The 10 Hz data were averaged to produce a data record consisting of 1 Hz averages.

Sawford - 2 Source Projects. Sawford et al. (1985) released  $\text{SF}_6$  and P from two point sources located about 10 m apart at their field site in Australia and studied the relations between the time series of the two tracers observed at monitors located at a distance downwind of 25, 50, or 100 m. Source height and monitor height were both about 1.5 m. Our analysis was limited to the  $\text{SF}_6$  data, which were characterized by an averaging time of 6 sec and a sampling time of 3000 s. Forty-four separate runs were analyzed. Unfortunately, concurrent wind fluctuation data were not available.

Deardorff-Willis Tank. Deardorff and Willis (1987) have used a  $1 \text{ m}^3$  tank of water to conduct several innovative studies of turbulence and dispersion in a convective boundary layer. In this experiment they towed a stack through the

tank and scanned the buoyant plume with a laser at a fixed position relative to the tank to produce nearly instantaneous cross-sections of concentrations at six different downwind distances. The effective averaging distance of the laser was about 10 m (scaled to full size). The ratio of  $\sigma_w/u$  was equal to about 0.75, where  $\sigma_w$  is the standard deviation of vertical velocity fluctuations and  $u$  is the speed that the stack is towed through the tank. The dimensionless plume buoyancy parameter (ratio of heat flux in the plume to the convective heat flux in the boundary layer),  $F_*$ , equaled 0.116, and the downwind distance was 3.57 times the length scale of the dominant large convective eddies. These concentration data were used to calculate space spectra rather than time spectra. Concurrent velocity fluctuation data were not available.

#### 4. Time Series Analysis Procedure

A generalized software package for the analysis of time series using the Fast Fourier Transform procedure was obtained from the National Center for Atmospheric Research (NCAR) (B. Stankov, personal communication, 1986) and was modified by R. Yamartino for use on an IBM PC/AT microcomputer. The text by Bendat and Piersol (1980) was used to develop these modifications. Time series for one to eight concurrent variables can be input to this package. In order to fit in existing storage space in a PC/AT, the total number of data points is limited to 2048 or less if eight variables are studied. The limit is 4096 if five variables or less are studied. Standard procedures such as detrending, smoothing, and tapering can be applied to the data. In this analysis, the time series were detrended (a linear trend removed) and tapered (beginning and ending 10% of the time series tapered to zero). Smoothing was applied only to final graphs. Output consists of tables and graphs for single variables (original time series, autocorrelogram, and spectrum) and for pairs of variables (co-spectrum, quadrature spectrum, phase angle, and coherence). A separate program was used to calculate the cross-correlogram.

Panofsky and Brier (1968) provide derivations and discussions of the interpretation of each of these plots, which are briefly summarized below.

An Autocorrelogram shows the correlation between a variable and itself as a function of time lag. It is dimensionless and ranges between -1.0 and +1.0.

A Spectrum shows the relative contribution of eddies or oscillations of various frequencies to the total energy or mean square variability. It has the units of  $X^2$  per unit frequency, where  $X$  is the units of the primary variable (concentration in, say, ppm, or wind speed in m/s). Frequency is given in units of cycles/s.

A Cross-Correlogram shows the correlation between one variable and another variable as a function of time lag. It is dimensionless and ranges from -1.0 to +1.0.

A Co-Spectrum measures the contribution of oscillations of different frequencies to the total cross-covariance at lag zero between two time series.

It has the units of XY per unit frequency, where X and Y are the units of the two variables being analyzed.

A Quadrature Spectrum measures the contribution of different oscillations to the total covariance obtained when the harmonics of one of the time series are delayed by a quarter period. It has the units of XY per unit frequency, where X and Y are the units of the two variables being analyzed.

The Phase Angle shows the angle by which two time series are out of phase at each harmonic. It ranges from  $-180$  to  $+180^{\circ}$  and is plotted as a function of frequency.

The Coherence shows how good the relationship is between two variables for various frequencies. It varies from 0 to 1.0 and is dimensionless.

## 5. Results of Time Series Analysis

The time series analysis software package is capable of producing large quantities of plots and tables. For example, if five concurrent time series are being analyzed, there are 55 plots produced for all possible types of graphs and combinations of variables. Because little information was contained in many of the plots that were produced in this study, the results that are discussed below are limited to the more interesting plots. Emphasis is on the shape of the concentration spectra, the dominant time scale and the slope of the inertial subrange in the concentration spectra, the magnitude of the cross-correlogram, and the dominant time scale in the cospectra.

### 5.1 Dominant Time Scale of Concentration Spectra

Concentration spectra were calculated for each of the data sets in Table 1. Except for the convective tank data, which are expressed as functions of position, all of the data were taken for similar conditions:

- Continuous point source located near the ground.
- Concentration measurements near the ground at downwind distances on the order of 100m.
- Moderate wind speeds.
- Mostly daytime conditions (a few were during the night, though).

Consequently it is expected that the normalized spectra,  $nS_c(n)/\sigma_c^2$  would exhibit similar shapes when plotted versus frequency,  $n$ . When  $nS_c(n)/\sigma_c^2$  is plotted versus  $\ln n$  the area under the curve should equal 1.0, and the peak of the curve occurs at a frequency associated with maximum energy. In fact it is found that  $nS_c(n)/\sigma_c^2$  reaches a maximum of about 0.3 at a frequency of about 0.01 to 0.02 cycles/sec or a time period of about 50 to 100 sec. Concurrent wind speed spectra have peaks at time periods of about 200 to 600 sec, and vertical velocity spectra have peaks at periods of about 1 to 5 sec. Thus the concentration spectra have shapes similar to the wind speed spectra, but are shifted to higher frequencies by a factor of about five. If the plumes were sampled farther downwind, where they would encompass a larger range of eddy sizes, it would be expected that the time scales of the concentration spectra

would increase, as found by Sawford et al. (1985).

Although individual spectral shapes can be quite variable, a smoother curve is obtained when several similar spectra are combined together. Figure 3 shows the distribution of spectra (after normalization by the area or  $\sigma_c^2$ ) for the six experimental data sets listed in Table 1, where the normalized frequency,  $n\sigma_y/u$ , is plotted on the abscissa. Because the figure would be a mess if all 51 spectra were plotted at once, the median and the range over all the spectra are given at 11 representative frequencies. It is seen that the median points follow a fairly smooth curve with peak energy at a normalized frequency of  $n\sigma_y/u$  equal to about 0.15. Two empirical curves are also drawn on the figure. One represents a Markov spectrum (equation 8) with an integral time scale,  $T_c$ , of  $\sigma_y/u$ . The time scale of peak spectral energy,  $T_{pc}$  is then  $2\pi\sigma_y/u$ .

$$\text{Markov: } nS_c(n)/\sigma_c^2 = 4(n\sigma_y/u)/(1 + (2\pi\sigma_y/u)^2) \quad (10)$$

This curve tends to underestimate the spectral energy on the figure at high and low frequencies.

An alternative empirical equation that has the proper slope in the inertial subrange is the following:

$$nS_c(n)/\sigma_c^2 = 6.5(n\sigma_y/u)/(1 + 54(n\sigma_y/u)^{5/3}) \quad (11)$$

This curve provides a better fit than equation (10) to the points in Figure 3. It is implicit in both these curves that the plume scale,  $\sigma_y$ , is much less than the scale of atmospheric turbulence,  $\Lambda_a$ , and therefore  $\Lambda_a$  does not influence the concentration spectra. In the daytime boundary layer,  $\Lambda_a$  is approximately equal to the mixing depth, which is on the order of 1000 m. Consequently  $\sigma_y$  is two order of magnitude smaller than  $\Lambda_a$  for these experiments. In the case when  $\sigma_y$  is the same order as  $\Lambda_a$ , Hanna (1986) suggests an empirical formula that is a linear combination of two Markov spectra--one with a time scale of  $\sigma_y/u$  and another with a time scale of  $\Lambda_a/u$ .

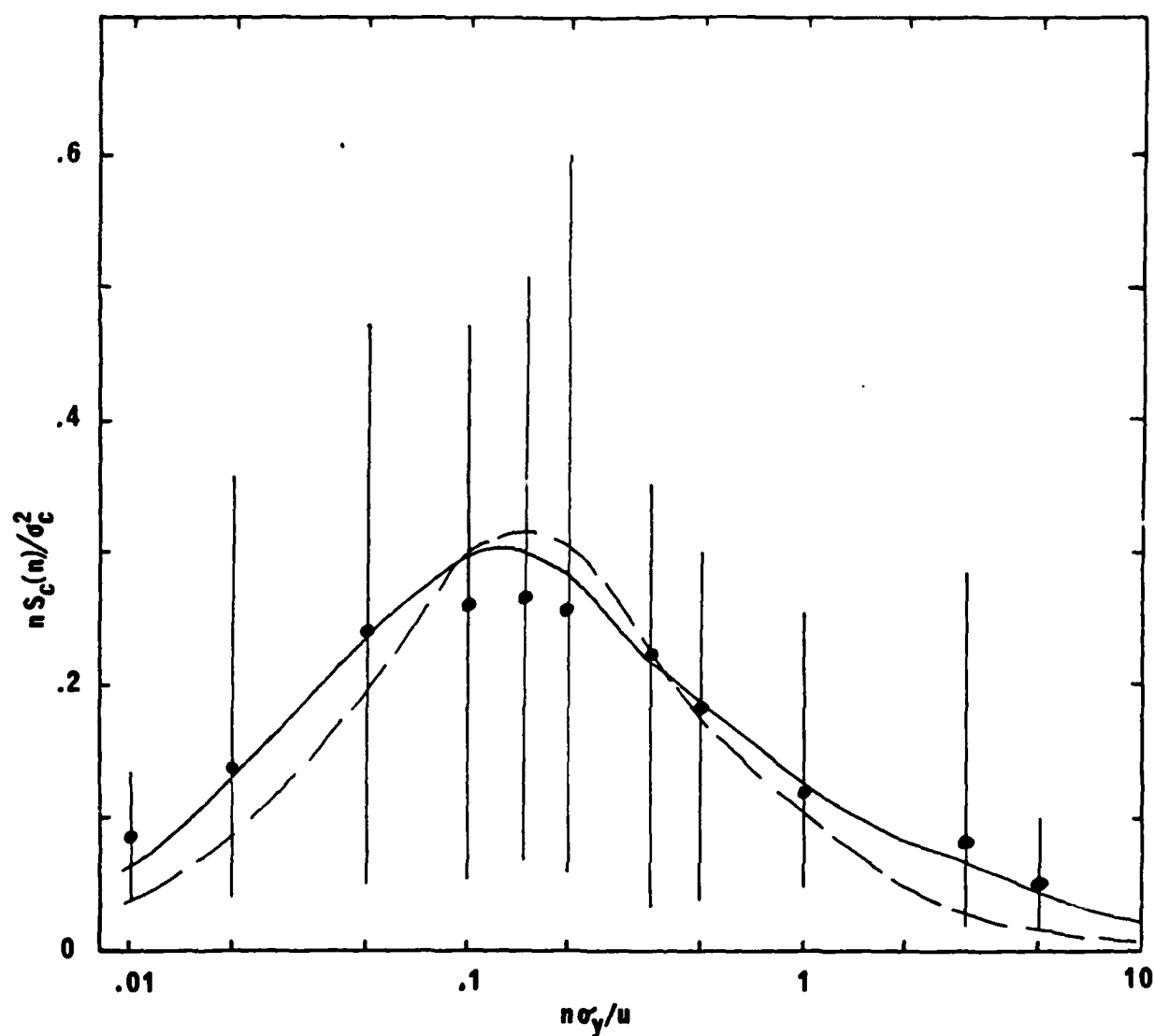


Figure 3. Composite Power Spectrum for Concentration Fluctuations from All Data in Table 1, Plotted in Dimensionless Coordinates as  $nS_c(n)/\sigma_c^2$  versus  $n\sigma_y/u$ . The Median and the Range from 51 Spectra are Given at Representative Frequencies. Analytical Curves from Equation 11(Solid Line) and Equation 10 (Dashed Line) are Drawn.

## 5.2 Characteristics of Inertial Subrange

According to similarity theory, the relation  $S_c(n) \propto n^{-5/3}$  is valid in the inertial subrange of the concentration spectrum (see equation 7). Garvey et al. (1982) verified this relation using earlier U.S. Army Smoke Week data. Each of the spectra calculated from the data in Table 1 were analyzed in an attempt to determine whether this relation was valid. In most cases, the  $-5/3$  power law was evident at frequencies,  $n$ , greater than about 0.05 or 0.10 cycles/sec (i.e., time periods less than 10 or 20 sec). Figure 4 contains an example of the inertial subrange of a spectrum from the Dugway Proving Ground field trials, which exhibits a particularly well-behaved curve. Generally the spectra contained more scatter than is seen in this figure.

The constant,  $a_c$ , in the following equation can be estimated from the data:

$$nS_c(n)/\sigma_c^2 = a_c(n\sigma_y/u)^{-2/3} \quad (12)$$

Figure 3 illustrates that  $nS_c(n)/\sigma_c^2$  equals about 0.12 at a normalized frequency,  $n$ , of unity for the median of 51 spectra. With these values for the parameters in equation (12), the constant,  $a_c$ , would equal about 0.12.

The Deardorff and Willis (1987) convective tank data were included in Figure 3. They can be used independently to derive the constant,  $a_c$ , although it is necessary to rewrite equation (9) as a function of wavenumber,  $k$ , which equals  $n/u$  if Taylor's frozen turbulence hypothesis is valid:

$$kS_c(k)/\sigma_c^2 = a_c(k\sigma_y)^{-2/3} \quad (13)$$

There are not enough runs to generate time series of concentration fluctuations from these data. We calculate a spatial spectrum  $S_c(k)$  using an instantaneous cross-section of the plume taken at a position such that a few hundred observation points are well within the plume. Consequently the concentration distribution can be assumed to be spatially homogeneous over these points. Spectra are calculated over horizontal rows of points well within the plume and then the spectra over 11 rows are combined to give a composite spectrum. It is found that this composite concentration spectrum

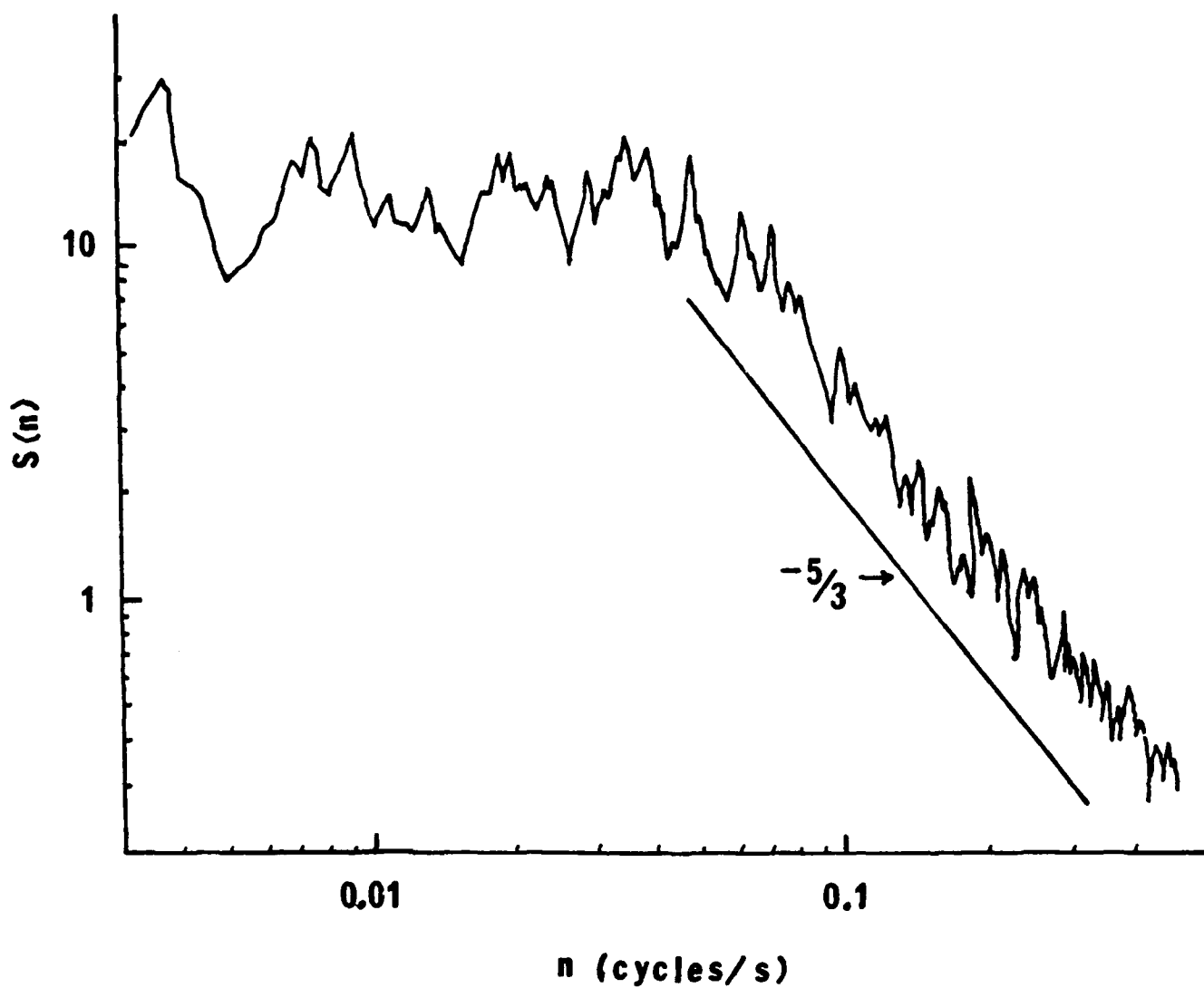


Figure 4. Power Spectrum Plotted Versus Frequency for Dugway Proving Ground Cross-Wind Integrated Concentration Fluctuations in Trial T091. The Curve Follows a  $-5/3$  Slope for Periods Less than about 20s.

exhibits an inertial subrange at wave numbers,  $k$ , greater than about  $5/h$ , (or wavelengths less than  $0.2h$ ), where  $h$  is the mixing depth. Thus the inertial subrange begins at a wavelength equal to about 10% of the wavelength of peak turbulent energy,  $1.5h$  (Kaimal et al. 1982). This composite spectrum from a single cross section of the plume in the convective tank yields a constant,  $a_c$ , of 0.36, which is about a factor of three larger than the value calculated from the median spectrum in Figure 3. It is expected that the calculated  $a_c$  would be larger during convective conditions, since the term  $\phi_{\varepsilon c}$  ( $z/L$ ) in equation (5) has been neglected. However the difference could also be due to the fact that the convective tank data represent only a single realization, and there is large uncertainty in the positioning of the inertial subrange in a single experiment.

### 5.3 Cross-Correlations and Cross-Spectra

Visual impressions from video tapes of smoke plumes would lead to the hypothesis that positive excursions in concentration fluctuations are correlated with positive excursions in turbulent vertical velocity. However, these visual effects are relatively short-lived, and when averaged over an hour, they contribute little to the total correlation. The cross-correlations between concentration fluctuations and turbulent wind speed fluctuations (e.g.  $r_{cu} = \overline{c'u'}/\sigma_c\sigma_u$  or  $r_{cw} = \overline{c'w'}/\sigma_c\sigma_w$ ) at zero time lag are typically about 0.1 or 0.2. At non-zero time lags the cross-correlations vary back and forth about zero, with a magnitude seldom exceeding 0.1. For comparison, the cross correlation between  $u'$  and  $w'$  ( $r_{uw} = \overline{u'w'}/\sigma_u\sigma_w = -u_*^2/\sigma_u\sigma_w$ ) at zero time lag is equal to about -0.4, where  $u_*$  is the friction velocity and it is assumed that  $\sigma_w/u_*$  equals 1.3 and  $\sigma_u/u_*$  equals 2.0. The cross-correlations  $r_{wc}$  and  $r_{vc}$  are proportional to the fluxes of plume material away from the center of the plume, and will change signs depending on which side of the plume the monitor is located. None of the cross-correlation plots as a function of time lag are reproduced here because they are so random and have such small correlations.

The cross-spectra, on the other hand, do yield some useful, although not unexpected results. It is known that small eddies in the atmosphere are isotropic (i.e. have no preferred direction). Consequently the cospectra and quadrature spectra for pairs of the variables  $u'$ ,  $v'$ ,  $w'$ , and  $c'$  are nearly

zero for large frequencies (small time periods). The low frequency limit of this range corresponds roughly to the limit of the inertial subrange (about  $n \sim 0.1 \text{ sec}^{-1}$  or period  $\sim 10 \text{ sec}$ ). This conclusion is well known for  $u' - w'$  cross-spectra (Panofsky and Dutton 1984), since there is observed to be little coherence between  $u'$  and  $w'$  for eddies in the inertial subrange, and the major contribution to the  $\overline{u'w'}$  product (i.e. the momentum flux) comes from eddies with relatively long time periods. Cross-spectra calculated between concentration and wind velocity fluctuations for the data in Table 1 agree with this hypothesis. Figure 5 is an example of a cospectrum from WSU Run P3, illustrating the lack of energy at high frequencies. The area under the curve is proportional to the cross correlation at lag zero. Quadrature spectra (the out-of-phase component of the cross-spectra) yield similar results to those seen in Figure 5.

The ratio of the quadrature to the co-spectrum at a given frequency can be used to calculate the phase angle between the concentration and wind fluctuations at that frequency. These phase angles tend to flip back and forth from negative to positive with no systematic patterns discernible in the data. Coherence plots were also generated from the co-spectrum and the quadrature spectrum, and show the contribution of each frequency to the total cross-correlation between the variables (ranges from 0 to 1). The  $C' - u'$  coherence plots illustrate a weak peak near the dominant meandering period, which tends to be about 100 to 200 s for these data, and yield little correlation at periods less than about 5s.

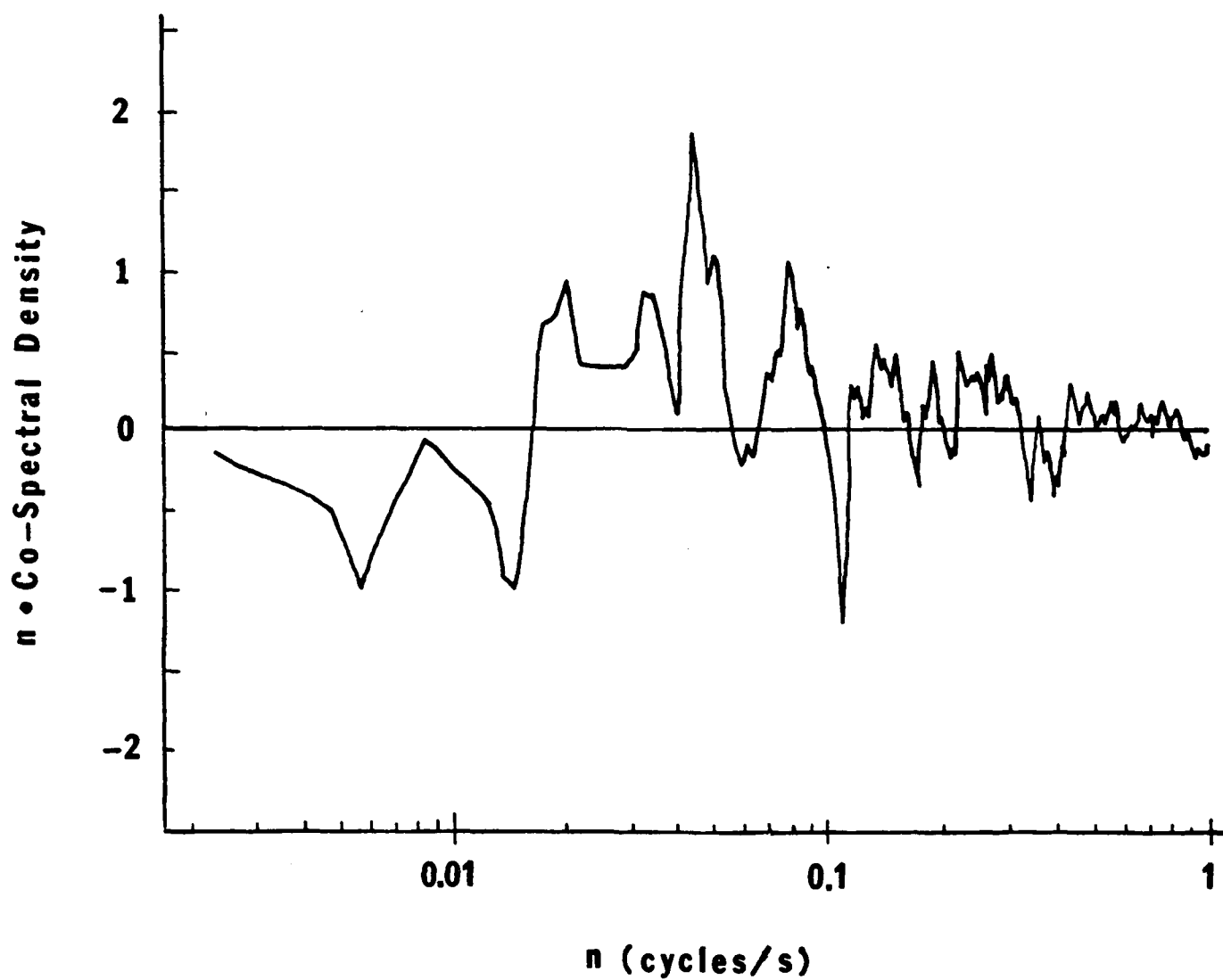


Figure 5. Co-Spectrum between Concentration Fluctuations and Wind Direction Fluctuations for WSU Run P3.

## 6. Conclusions

Field and laboratory data sets containing fast-response measurements of concentration fluctuations were acquired. Of the six separate data sets, four also contained fast response measurements of wind velocity fluctuations. A time series/spectral analysis procedure was modified for use on an IBM PC/AT (copies of this computer program are available from the authors). This procedure calculates autocorrelograms, spectra, cospectra, quadrature spectra, coherence, and phase angle as a function of frequency for up to 2048 data points for each of up to 10 variables.

Nearly all of the field data sets were obtained under similar conditions--continuous point sources near the ground, observations near the ground at a downwind distance of about 100m, and daytime conditions with moderate winds. Sampling times ranged from a few minutes to one hour, and averaging times were used that ranged from 0.5 sec to 6 sec. For these conditions an inertial subrange existed in the concentration spectra for time periods less than about 10 sec. When written in the form  $nS_c(n)/\sigma_c^2 = a_c(n\sigma_y/u)^{-2/3}$ , the Kolmogorov inertial subrange formula is valid when the constant,  $a_c$ , is set equal to 0.12. The entire shape of the observed spectra can be fit by an analytical formula, where the normalized frequency associated with peak spectral energy is about  $n\sigma_y/u = 0.15$ . The time scale of the concentration spectra is typically about a factor of two to five less than the time scale of the concurrent wind speed spectra, and is a factor of 10 to 20 more than the time scale of the concurrent vertical velocity spectra.

A spatial concentration spectrum was calculated using data from an instantaneous cross-section of a plume in a laboratory convective tank. This spectrum exhibited an inertial subrange at wavelengths less than  $0.1h$ , where  $h$  is the mixing depth. The Kolmogorov constant,  $a_c$ , calculated from these data, was a factor of three higher than that calculated from the field data.

Cospectra, quadrature spectra, coherence, and phase angles were calculated between concurrent concentration and wind velocity time series. At most sites, the meteorological and concentration monitoring instruments were colocated. There was much variability in these plots, due to the tendency of a given time series to be dominated by one or two obvious sinusoidal patterns.

The only clear conclusion is that small eddies with time scales less than about 10 sec contribute little towards the relations between the time series.

Despite our attempts to acquire a comprehensive set of all publicly-available concentration fluctuation data sets, it is seen that these data sets are quite limited in their characteristics. In the future, concentration fluctuation data should be collected for a wide range of source heights, monitor positions, downwind distances, and meteorological conditions. The validity of Taylor's frozen turbulence hypothesis should be tested. Furthermore, the cross-correlations are strengthened if the concentration and meteorological instruments are co-located. However, even though the available data sets are limited, the analysis represents a first step towards the development of generalized spectral relations for concentrations fluctuations.

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