

# UNCERTAINTY ANALYSIS IN ATMOSPHERIC DISPERSION MODELING

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## **ABSTRACT**

The concentration of a pollutant in the atmosphere is a random variable that cannot be predicted accurately, but can be described using quantities such as ensemble mean, variance, and probability distribution. There is growing recognition that the modeled concentrations of hazardous contaminants in the atmosphere should be described in a probabilistic framework. This paper discusses the various types of uncertainties in atmospheric dispersion models, and reviews sensitivity/uncertainty analysis methods to characterize and/or reduce them. Evaluation and quantification of the range of uncertainties in predictions yield a deeper insight into the capabilities and limitations of atmospheric dispersion models, and increase our confidence in decision-making based on models.

**Keywords:** Atmospheric Dispersion Models, Concentration Prediction, Uncertainty Analysis, Stochastic Uncertainty, Probabilistic Framework, Regulatory Modeling

## **1. INTRODUCTION**

Atmospheric dispersion models are routinely used to assess the impact of emission sources on air quality for varying meteorological conditions, source configuration, and topography. In addition, these models are also used at nuclear and chemical plants to estimate exposure to hazardous contaminants accidentally released into the atmosphere. A risk assessment is undertaken to quantify the potential hazard to exposed populations and to evaluate response or remediation measures. An uncertainty analysis is recommended as an integral part of any risk assessment to quantify the degree of confidence in the estimate of risk (IAEA, 1989).

Atmospheric dispersion is a stochastic phenomenon and, in general, the concentration observed at a given time and location downwind of a source cannot be predicted precisely (Chatwin, 1982). Since concentration is a random variable, it should be described statistically, using quantities such as ensemble mean, variance, and probability distribution (Csanady, 1973; Lewellen and Sykes, 1989). It was long recognized that uncertainties in atmospheric dispersion models must be studied as part of any comprehensive model performance evaluation (e.g., Fox, 1984). There is a growing trend to move away from the simple deterministic predictions of ensemble mean concentrations and move towards development of probabilistic results which seek to describe a range of likely events and their associated probabilities (Dabberdt and Miller, 2000; Hogrefe and Rao, 2001). This paper addresses the analysis and quantification of various types of uncertainties associated with the prediction of concentrations from atmospheric dispersion models.

## **2. UNCERTAINTY IN DISPERSION MODELS**

A concentration estimate from an atmospheric dispersion model typically represents an ensemble-average of numerous repetitions of the same event at a specific site. The event is characterized by measured values or “known” parameters that are input to the model, e.g., wind speed, mixing layer depth, atmospheric stability, source conditions, etc. However, in addition to the known

parameters, there are unmeasured or unknown variations in the conditions of this event, e.g., unresolved details of the atmospheric flow or the subgrid-scale atmospheric processes. Thus, even with a “perfect” model that predicts the correct ensemble-mean, there are likely to be deviations from the observed concentrations in individual realizations of the event, because of unknown variations in the conditions. In general, both observations and model predictions are uncertain. Therefore, meaningful model verification requires not only the average values but also the probability distributions of target variables.

Uncertainty in atmospheric dispersion model predictions is associated with: (a) “data” or “parameter” uncertainty resulting from errors in the data used to execute and evaluate the model, uncertainties in empirical model parameters, and initial and boundary conditions; (b) “model” or “structural” uncertainty arising from inaccurate treatment of dynamical and chemical processes, approximate numerical solutions, and internal model errors; and (c) “stochastic” uncertainty, which results from the turbulent nature of the atmosphere as well as from unpredictability of human activities related to emissions. The uncertainties associated with (a) and (b) can be minimized by making better (more accurate and more representative) measurements and improving model dynamics, parameterization schemes, and numerical methods, but it may not be feasible to reduce them beyond a certain level. The stochastic uncertainty term (c), arising from the natural variability of the atmosphere, can be expressed by the mean square concentration fluctuations:

$$\sigma_c^2 = \overline{(C^{(r)} - \bar{C})^2}, \quad (1a)$$

where  $\sigma_c$  is the standard deviation and the overbar denotes the ensemble average.  $C^{(r)}(\vec{x}, t)$  is the concentration in one ( $r$ -th) realization of the experiment at spatial location  $\vec{x}$  and time  $t$ , and  $\bar{C}(\vec{x}, t)$  is the ensemble-average concentration defined as

$$\bar{C}(\vec{x}, t) = \lim_{n \rightarrow \infty} \left[ \frac{1}{n} \sum_{r=1}^n C^{(r)}(\vec{x}, t) \right]. \quad (1b)$$

The stochastic uncertainty in modeled and observed concentrations cannot be eliminated, but it can be quantified in a statistical sense. Understanding the various uncertainties and their causes is required to correctly interpret monitoring data and modeling results.

From the differences (residuals) between the observed and predicted concentrations,  $C_o$  and  $C_p$ , the bias (model error),  $\bar{d}$ , and the variance (total model uncertainty),  $\overline{(C_o - C_p)^2}$ , can be expressed (Rao and Hosker, 1993) as

$$\bar{d} = \overline{C_{oa}} - \overline{C_{pa}}, \quad (2)$$

$$\overline{(C_o - C_p)^2} = \underbrace{\overline{(\delta C_p)^2}}_{(a-1)} + \underbrace{\overline{(\delta C_o)^2}}_{(a-2)} + \underbrace{(\bar{d})^2}_{(b)} + \underbrace{\sigma_c^2}_{(c)}, \quad (3)$$

where  $\overline{C_{oa}}$  is the actual ensemble-average observed (without instrument errors) concentration for a given set of external conditions,  $\overline{C_{pa}}$  is the corresponding prediction based on error-free input data,  $\delta C_o$  is the error in the observation of  $C_o$ ,  $\delta C_p$  is the error in  $C_p$  due to input data errors, and  $\sigma_c^2$  is the stochastic uncertainty, defined in Eq. (1), resulting from atmospheric turbulence.

The various components of the total model uncertainty, identified by (a-1), (a-2), (b), and (c) on the RHS of Eq. (3), are discussed above. The sum of the first two terms, (a-1) and (a-2), represents the data errors term (a). The third term on the right in Eq. (3), which is the square of model bias defined in Eq. (2), represents the model errors (e.g., in parameterizations, physics, coding) term (b). The last term (c) is the stochastic uncertainty.

The likely variations of the uncertainty terms in Eq. (3), versus the number of meteorological parameters in the model, are depicted in Fig. 1. We hope that the errors in the model physics can be reduced (as shown) by increasing the number ( $N$ ) of meteorological parameters in the model. This is the primary justification for using complex prognostic 3-D mesoscale meteorological models. The stochastic uncertainty should also decrease as  $N$  increases, as more and more of

the atmospheric variability is “explained” by the model. On the other hand, the data errors are likely to increase monotonically with  $N$ . It can be seen in Fig. 1 that the data errors term is a major contributor to total model uncertainty. Note that the finite value of this error at  $N = 0$  is due to instrument errors in the observed concentrations. Though the variations depicted in Fig. 1 are not directly based on data, they are qualitatively supported by available work in the literature. For example, Lewellen and Sykes (1989) showed that the meteorological input data uncertainty accounts for more than half of the total uncertainty in predicting the observed 1-hr ground level concentrations (GLCs) using a second-order closure plume model, which had improved model physics compared to the usual Gaussian plume model. It is clear that, for any given application, there is an optimum model complexity (defined here by the value of  $N$ ) that minimizes the total uncertainty. This optimum level of complexity cannot be defined *a priori*, but can be determined by trial and error based on experience.

The data errors curve in Fig. 1 represents uncertainties in all measurements such as emissions, meteorology, concentrations, and other input parameters. Even if an instrument is accurate, there can be large uncertainties in the data due to unrepresentative siting. For example, data from a single meteorological tower cannot represent the flow over a large area or over a moderately complex terrain. Even in research-grade field measurements, there are significant uncertainties in the various measured parameters (Hanna, 1988). The total data error is not a simple sum of the errors in individual data components. Uncertainties in input data such as wind speed, dispersion parameters, and plume rise can be propagated through the model and the output concentration distributions can be analyzed through sensitivity/uncertainty analyses using analytical or numerical methods discussed in the next section.

It is important to distinguish between uncertainty and variability. The latter refers to the true heterogeneity that is observed in nature, whereas uncertainty characterizes our lack of complete knowledge of a specific quantity of interest. Variability is a property of the system being studied,

whereas uncertainty is considered a property of the analyst. Different analysts, with different states of knowledge or access to different data sets, may have different levels of uncertainty in the predictions they make for a quantity of interest. Morgan and Henrion (1990) suggest that variability is usually described by frequency distributions, while uncertainty is described by probability distributions. In exposure assessments, common sources of variability are due to differences in characteristics such as intake rates and activity patterns between individuals. However, there may also be uncertainty in the characteristics of specific individuals in the population, due to lack of data or measurement errors. Cullen and Frey (1999) discuss the differences between uncertainty and variability, and suggest that it is desirable to characterize them separately for problems where these distinctions are important. Under specific conditions, it is possible for variability to be interpreted as uncertainty. For example, the natural variability of the atmosphere leads to stochastic uncertainty, as discussed above, in the prediction of short-term air pollutant exposure for a given individual. This is sometimes referred to as *Type A uncertainty* to distinguish it from the *Type B uncertainty* resulting from lack of knowledge about the appropriate mathematical models or parameter values (IAEA, 1989).

### **3. DATA UNCERTAINTY ANALYSIS METHODS**

The relative importance of the uncertainties in input parameters to model outputs can be determined using sensitivity or uncertainty analyses. The objectives of these analyses are to: (1) study the robustness of the model with respect to potential changes in inputs and parameters; and (2) provide quantitative estimates of the overall uncertainty incorporated in model predictions.

Sensitivity analysis is the systematic study of the behavior of a model over ranges in variation of inputs and parameters. Often, the response of the model output to very small changes in a given uncertain parameter are studied, while all the other parameters are fixed. This gives only the local gradient of the model response surface with respect to that parameter. Uncertainty analysis

is the quantitative assessment of how the uncertainties in model physics and input data, as well as the random variability in input parameters, propagate through the model to give a single measure of uncertainty in the model results. The collective uncertainty in all the parameters is studied, yielding more complete information on uncertainty propagation with respect to the multidimensional model response surface. The uncertainty in an atmospheric dispersion model is generally presented as a 90% or 95% confidence interval for the predicted concentrations. While sensitivity analysis is fairly widely used in the atmospheric dispersion model evaluation, parametric uncertainty studies are much less common.

Both sensitivity analysis and uncertainty analysis attempt to rank order the parameters according to their contribution to overall model error. Sensitivity analysis approaches the problem by taking the partial derivatives of model equations with respect to individual parameters. Results are usually stated as the change in a model prediction to be expected from a small change in parameter values. A differential analysis provides good local information about the inputs, but does not extend well to global interpretation. Sensitivity analysis assumes that errors in model predictions can be approximated by examining small perturbations in the parameters, and the error contribution of each parameter can be examined separately. Thus, higher-order effects resulting from simultaneous errors in many parameters are often ignored.

In contrast, uncertainty analysis considers each parameter as a random variable. Assumptions in uncertainty analysis involve the statistical distributions of parameters and their means, variances and covariances. Uncertainty in predictions is estimated by Monte Carlo simulation with each parameter selected either from independent distributions or from multivariate distributions specified by a covariance matrix. The contribution of each parameter to model uncertainty is determined by statistical analysis of simulation results. The calculation of correlation coefficients (between each input parameter and the model output) from Monte Carlo simulations is the most relevant approach for ranking the parameters (Gardner *et al.*, 1981).

### 3.1 Sensitivity Analysis

A sensitivity study examines the way a particular model responds to variations in values of input variables or internal parameters. The results of the study are not directly related to model accuracy and may not correspond to physical reality, but can identify the relative importance of the input variables. This is helpful in indicating how accurately the input variables must be measured. Sensitivity studies are also useful for checking if the model is performing as expected. If it is not, then additional verification and/or development may be necessary.

Sensitivity analysis quantitatively estimates the relationships between changes in the input variables and the resulting changes in the model output. This relationship, described by sensitivity coefficients (SC), will be fairly accurate in a weak response regime where the output is linearly related to the inputs for a *base case* with a specific value for each input variable. For a model with the  $j$ -th output given by  $C_j(t_i)$ , where  $t_i$  ( $i = 1, 2, \dots, n$ ) are independent input variables, the SC ( $S_{ij}$ ) are defined as

$$S_{ij} = \partial C_j / \partial (\ln t_i) , \quad (4)$$

where the derivatives are evaluated for the base-case values of the inputs. The change in the  $j$ -th model output,  $\Delta C_j$ , due to a newly modified input variable set can be calculated then by summing over all the input variable changes from the base case:

$$\Delta C_j = \sum_{i=1}^n [S_{ij} \cdot \Delta(\ln t_i)] . \quad (5)$$

For small finite changes in variables,  $\Delta(\ln t_i) = \ln t_i^* - \ln t_i = \ln(t_i^*/t_i)$ , where  $t_i$  is the base case value and  $t_i^*$  is the new value of the  $i$ -th input variable. Equation (5) assumes that the relationship between the changes in the input variable set,  $\Delta(\ln t_i)$ , and the resulting change in the model output,  $\Delta C_j$ , is linear. The new estimated model output is

$$C_j^* = C_j + \Delta C_j . \quad (6)$$

The standard deviation  $\sigma_j$  of the  $j$ -th model output can be estimated as

$$\sigma_j = \left[ \sum_{i=1}^n \{S_{ij} \cdot \ln(R_i)\}^2 \right]^{1/2}, \quad (7)$$

where  $R_i = (t_i + \sigma_i)/t_i$  and  $\sigma_i$  is the standard deviation of the  $i$ -th input variable. This equation provides an efficient method to estimate the model output uncertainties when the input variable uncertainties are small.

Many of the commonly used sensitivity methods have been developed for use on simple models and are impractical for large models. However, sophisticated sensitivity analysis techniques, which can be applied to the governing equations of multidimensional models describing dynamical and chemical behavior in the atmosphere, are described by Dunker (1981), Rabitz *et al.* (1983), and others.

### 3.2 Uncertainty Analysis

Once the input parameters to be studied are identified, uncertainty analysis is a two-step sequential procedure. The first step involves assigning probability distribution to each key input parameter either by its cumulative distribution function (CDF), or by its probability density function (PDF) which is the derivative of the CDF with respect to the parameter. These distributions should include measurement errors as well as random variations, and should also account for dependencies and correlations among various parameters. When sufficient data are available, these are used to generate the distribution for each parameter. Uniform, triangular, and normal distributions are among those widely used, which are shown in Fig. 2. As long as the mean and variance are held constant, the exact shape of the distribution of a parameter has minimal effect on the distribution of the model prediction (Hoffman and Hammonds, 1992). When the range of a parameter extends over one or more orders of magnitude, logarithmic form of the distribution is used. For some problems, discrete or custom probability distributions may be needed to describe the inputs, as shown in Fig. 2.

The second step in an uncertainty analysis involves propagating the joint probability distribution of the uncertain input parameters through the model to generate a probability distribution of model prediction. Based on this distribution, one can formulate a quantitative description in the form of confidence intervals (CIs) in which the unknown *true* prediction should lie. These CIs are referred to as *subjective* when the input probability distributions of the uncertain model parameters are derived using subjective judgement in the absence of data.

### **3.2.1 Expert Judgement**

An uncertain input parameter should not be treated as a constant simply because data are unavailable to define a range and distribution. Professional judgement is often employed in the absence of data. There has been remarkable progress over the past thirty years in the understanding of how both experts and laypersons make judgements on variables and events in the face of uncertainty, and in the development of techniques for elicitation of professional judgement and encoding of probabilities (Morgan and Henrion, 1990).

The assessment of the range and statistical distribution for each uncertain parameter is the most difficult task in quantitative uncertainty analysis, and requires a high level of expertise. The dependence on expert opinion in this exercise can be reduced in proportion to the amount of theoretical and experimental information directly relevant to the parameter that exists. When there is a large body of information about a parameter, the experts should not differ substantially in their opinions. In situations where information is scarce, experts must rely on experience, theoretical insight and ability to extrapolate information from one situation to another (IAEA, 1989). In this case, the opinion of different experts is expected to differ considerably. Therefore, it is necessary to have a survey of expert opinion when model predictions provide input to important decisions.

The knowledge that the expert (assessor) has about the parameter to be assessed is referred to as *substantive expertise*. A substantive expert should on the average assign high probabilities to

those events that turn out to occur, and low ones to those that do not. The skill of the expert in expressing his or her beliefs in probabilistic form is referred to as *normative expertise*. An expert is said to be well calibrated (reliable) if the assessed probability of events corresponds with their empirical frequency of occurrence. The expert's power to discriminate between different levels of probability is referred to as *resolution*.

The assumptions and sources of information used by the expert must be documented in the elicitation process. When multiple experts are used, the probability distributions elicited from each expert are combined to form an aggregate distribution for each elicitation variable. Hora and Iman (1989) outlined a formal approach for eliciting information from experts. Harper *et al.* (1994) and Cook *et al.* (1994) discussed this approach, application, and results for the dispersion and deposition uncertainty assessment in the probabilistic uncertainty analysis of radiological accident consequences. Hanna *et al.* (1998) utilized an informal elicitation method based on responses from ten experts to specify the uncertainties in 109 input parameters, including those related to emissions, meteorology and boundary conditions for a photochemical grid model.

### 3.3 Uncertainty Analysis Methods

There are two main classes of uncertainty analysis methods: analytical and numerical. The choice of a specific method will depend on the complexity of the model, amount of information desired from uncertainty analysis, effort and time required, and costs for obtaining this information. In general, numerical methods are necessary for analysis of complex, nonlinear physical systems. Monte Carlo methods (MCMs) involve simple random sampling (e.g., Iman and Conover, 1980) from the probability distributions of input parameters and successive model runs with each parameter set until a statistically significant distribution of the output is obtained for uncertainty analysis. A partial rank correlation analysis between the output variable and the input parameters is performed, with the underlying premise that the greater the correlation for a

parameter, the more influence that parameter has in dictating the model response. MCMs have been used for many years in diverse fields such as finance, engineering, and physical sciences.

### 3.3.1 Analytical Methods

For relatively simple models such as the Gaussian plume dispersion model, when the number of input parameters to be varied is small and their relationship to the model prediction can be expressed as an algebraic equation, uncertainty analysis can be performed using analytical methods. Among the analytical methods are moment matching (IAEA, 1989) and variance propagation (Hoffman and Hammonds, 1992). Moment matching permits the derivation of subjective confidence intervals by identifying a distribution function with the same mean, variance, and third and fourth moments as the subjective PDF of the model prediction. Thus, moment matching requires these four moments of the unknown output PDF to be obtained from known moments of the input parameters. Variance propagation can be easily applied to simple additive models, where the mean and the variance of the output distribution are equal to the sum of the means and variances, respectively, of the input parameters (e.g., Morgan and Henrion, 1990).

In the case of a simple multiplicative model of the form:

$$X = p \times q \times r , \quad (8)$$

a logarithmic transformation is applied to put it in an additive form:

$$Y = \ln(X) = \ln(p) + \ln(q) + \ln(r) . \quad (9)$$

The distribution of  $X$  will tend to be approximately lognormal even when the parameters  $p$ ,  $q$ , and  $r$  are assigned distribution shapes other than lognormal (Hoffman and Hammonds, 1992). The median value (or geometric mean,  $X_g$ ) of the lognormal distribution can be obtained by adding the means of the logarithmic terms on the RHS of Eq. (9) and exponentiating the sum ( $\bar{Y}$ ):

$$X_g = e^{\bar{Y}} . \quad (10)$$

The standard deviation  $S_x$  of the distribution is obtained by adding the variances of the terms on the RHS of Eq. (9) and taking the exponential of the square root of the sum ( $S_y$ ):

$$S_x = e^{S_y} . \quad (11)$$

$S_x$  is often referred to as the geometric standard deviation of the lognormal distribution. The upper and lower confidence limits for a 90% subjective CI are then calculated as

$$X_{95} = X_g S_x^{1.65} = \exp(\bar{Y} + 1.65 S_y) , \quad (12a)$$

$$X_5 = X_g / S_x^{1.65} = \exp(\bar{Y} - 1.65 S_y) . \quad (12b)$$

The upper and lower confidence limits for a 95% subjective CI are calculated as

$$X_{97.5} = X_g S_x^{1.96} = \exp(\bar{Y} + 1.96 S_y) , \quad (13a)$$

$$X_{2.5} = X_g / S_x^{1.96} = \exp(\bar{Y} - 1.96 S_y) . \quad (13b)$$

Freeman *et al.* (1986) used an error propagation formula, which involved expanding the atmospheric concentration  $C$  in terms of independent input variables in a Taylor series and retaining only terms of second order or less. This can be written as follows:

$$C = f(t_1, t_2, \dots, t_n) , \quad (14)$$

$$S_c^2 = \sum_{i=1}^n \left( \frac{\partial f}{\partial t_i} \right)^2 S_{t_i}^2 + \frac{1}{2} \sum_{i=1}^n \left( \frac{\partial^2 f}{\partial t_i^2} \right)^2 S_{t_i}^4 + \sum_{i=1}^n \sum_{j=1}^n \left( \frac{\partial^2 f}{\partial t_i \partial t_j} \right)^2 S_{t_i}^2 S_{t_j}^2 . \quad (15)$$

This equation expresses  $S_c$ , the uncertainty in the predicted value of  $C$ , as a function of the uncertainties  $S_{t_i}$  in the input variables. For a Gaussian dispersion model, Freeman *et al.* (1986) showed that  $S_c$  calculated from this approach will be generally within 25% of the uncertainty of model-predicted  $C$  values calculated from Monte Carlo simulation of a randomly perturbed input data set for all stability cases and distances up to 15 km from the source.

For a Gaussian plume dispersion model, the concentration  $C$  can be expressed as

$$C/Q = p(y, \sigma_y) q(z, \sigma_z; H) r(U), \quad (16)$$

where  $Q$  is emission rate,  $U$  is mean wind speed,  $H$  is effective plume height,  $y$  and  $z$  are horizontal crosswind and vertical distances, and  $\sigma_y$  and  $\sigma_z$  are plume dispersion parameters,  $r = 1/U$ , and  $p$  and  $q$  are the horizontal and vertical probability density functions given by:

$$p(y, \sigma_y) = \frac{1}{\sqrt{2\pi}\sigma_y} \exp\left\{-\frac{y^2}{2\sigma_y^2}\right\}, \quad (17)$$

$$q(z, \sigma_z; H) = \frac{1}{\sqrt{2\pi}\sigma_z} \left[ \exp\left\{-\frac{(z-H)^2}{2\sigma_z^2}\right\} + \exp\left\{-\frac{(z+H)^2}{2\sigma_z^2}\right\} \right]. \quad (18)$$

Using a logarithmic transformation of Eq. (16), one can express it in a form identical to Eq. (9), with  $X = C/Q$ . From the above equations, we can determine the standard deviation  $S_y$  for the function  $Y = f(U, \sigma_y, \sigma_z, H)$ , for specified probability distributions in input parameters:  $U, \sigma_y, \sigma_z$ , and  $H$ . The partial derivatives required in Eq. (15) are derived by differentiating the functions  $p, q$ , and  $r$  with respect to these variables.

The analytical method described above has been applied by the present author to assess the effects of uncertainties in the input parameters for off-site radiological consequence estimates from hypothetical nuclear power plant accidents (see Harper *et al.*, 1994, Vol. 2). Typical uncertainties in wind speed  $U$  range from 0.1 m/s (for research grade data) to 1 m/s (for routine monitoring data); these errors arise due to poor calibration and maintenance of anemometers, and use of wind data unrepresentative of the plume transport, especially at night, because of mesoscale or terrain variability and wind shear. Uncertainties in  $\sigma_y$  and  $\sigma_z$  are difficult to estimate, but probably range from 10% to 40%; in addition to measurement uncertainty, they result from differences between the experiment site and the application site, and errors in the stability classification. Uncertainty in the effective plume height  $H$ , arising primarily due to errors in plume rise estimation, usually range from 10% to 30%. For simplicity, it is assumed that

$U$ ,  $\sigma_y$ ,  $\sigma_z$ ,  $H$  are uncorrelated and normally distributed, with means given by the input values and standard deviations ( $S_u, S_{\sigma_y}, S_{\sigma_z}, S_H$ ) by the magnitudes of their respective uncertainties. The model-calculated value of the relative concentration  $\overline{C/Q}$  is taken to be the median or 50th percentile value; this determines  $\overline{Y} = \ln(\overline{C/Q})$ . Once  $\overline{Y}$  and  $S_y$  are calculated, Eqs. (12) and (13) can be used to estimate the 90% and 95% subjective CIs, respectively. Note that these CIs are based only on uncertainties in input parameters, and do not include the model errors and stochastic uncertainty.

### **3.3.2 Numerical Methods**

Given the PDFs of input parameters, MCMs can be used to obtain the probability distributions of model response (output) variables. MCMs have been applied to atmospheric dispersion models ranging from simple Gaussian plume models (Kocher *et al.*, 1987; Irwin *et al.*, 1987) to complex photochemical models (*e.g.*, Hanna *et al.*, 1998).

In practice, the large dimensionality of multiple uncertain parameters in complex models is a major problem in uncertainty analysis. The regular MCM, based on a random sampling of the entire input parameter space, is computationally expensive for nonlinear coupled models with a large number of uncertain input parameters. In modified Monte Carlo methods such as Latin hypercube sampling (LHS; McKay *et al.*, 1979; Iman and Helton, 1988) and the Fourier Amplitude Sensitivity Test (FAST; Cuckier *et al.*, 1978; McRae *et al.*, 1982), sampling from the probability distribution is stratified across the range of each input variable and accomplished in an efficient manner so that the number of necessary solutions (model runs) is greatly reduced, compared to the simple MCM. The main idea of the secondary model techniques such as the “response surface” method (RSM; Downing *et al.*, 1985; Iman and Helton, 1988) is the construction of a comparatively simple approximation (secondary model) of the original (primary) model; the latter is then replaced by the simpler model in subsequent analysis of relations between input and output uncertainties.

LHS is a stratified MCM, designed mainly for reducing the variance of some statistics of the response variables. In this method, the number of model runs is equivalent to the number of intervals  $M$  for each input parameter range between its high and low values. The  $M$  intervals are formed such that each interval contains an equal area under the parameter distribution. In *standard* LHS, a single value is then randomly sampled from each interval, which is equivalent to assuming a uniform distribution over that interval. In *mid-point* LHS, the median value of each interval is selected. This is repeated for all the uncertain input parameters. A scenario is generated by selecting one value at random for each of the input parameters, but without replacement, from the  $M$  sample values for each parameter. This finally yields  $M$  scenarios, with each value for each input parameter being used only once.

The mean and variance of the sample in LHS represent the parameter distribution more accurately than in unstratified random sampling. Using the CDF of the model output resulting from the LH sample of the inputs, one can obtain estimates of the percentile points of the output. LHS can reduce the number of runs required to obtain a given variance typically by a factor of 10 compared to the simple MCM. Iman and Shortencarier (1984) described a computer program for the generation of LH samples. Iman and Helton (1988) recommended the use of LHS method, because it is easy to use and is applicable to many different modeling situations and gives reliable results. However, it is harder to compute the statistics for the LHS method, using standard tests for estimating the precision of the results, than for the MCM (Morgan and Henrion, 1990, Hanna *et al.*, 1998).

FAST uses continuous probability distributions of model input parameters as data, and determines the relative contributions of each input parameter to the variances of model outputs. All input parameters are varied simultaneously through their ranges of possible values following their given PDFs. Each input parameter is assigned a different frequency, which determines the number of times that the complete range of the parameter is traversed. With each input parameter

oscillating at a different characteristic frequency, each model run has a different set of input parameter values such that every value is used only once. The mean and variance for model output parameters, characterizing the uncertainty due to the variability of the input parameters, are then calculated. Fourier analysis of each output for all model runs is used to separate the response of the model to the oscillation of particular input parameters. Summation of those Fourier coefficients corresponding to a particular input parameter frequency and its harmonics determines the contribution of that parameter to the model output variances.

The FAST method provides information on the model sensitivity to particular input parameters. Uliasz (1988) and Collins and Avissar (1994) applied FAST to atmospheric problems. They concluded that the efficiency of FAST is comparable to LHS, with the number of sampling points equivalent to the number of values chosen for each input parameter. Nevertheless, FAST may not generate sufficiently accurate joint PDF of the response variables without requiring a large number of statistical points (Tatang et al., 1997).

In RSM, the original (primary) model is replaced by a comparatively simple approximation (secondary model). The latter is then used as a surrogate for the primary model in the subsequent analysis of relations between the input and output uncertainties. The method consists of screening the original model to determine the subset of important input parameters, fitting a response-surface (usually a polynomial) to the model in terms of these inputs, obtaining moments of the response surface (secondary) model, and fitting a Pearson or Johnson distribution to the moments to obtain a statistical model of the proxy to the output distribution. The output CDFs, the variances of the output variables, and ranking of parameters by their contributions to variances are then derived. There are several pitfalls in this approach. The selection of the most important variables, especially when interactions among parameters cannot be neglected, is a major problem. The fitting of a RSM is not straightforward. In general, highly nonlinear

mathematical models cannot be adequately approximated with a response surface except over a very limited range, and this yields misleading results from uncertainty analysis.

Tatang *et al.* (1997) described the probabilistic collocation method (PCM) which approximates model response surfaces using orthogonal polynomials, whose weighting functions are the PDFs of the input parameters. This PCM method was shown to be potentially a factor of 25 to 60 times faster than the MCM for parametric uncertainty analysis, converging exponentially with increasing orders of polynomial expansions. Isukapalli *et al.* (1998) described the application of stochastic response surface methods (SRS) for uncertainty characterization in environmental and biological systems. These methods, which can be thought of as “models of a model”, are computationally efficient when the number of degrees of freedom is not very large.

### **3.3.3 Other Methods for Uncertainty Assessment**

Fisher (2003) discussed the application of fuzzy set theory to the air pollution problem. Methods based on fuzzy sets usually rely on a judgement of the degree of uncertainty, or fuzziness, associated with the output of an air quality model. The comparison between the output from the model and an air quality objective is similar to the comparison between two fuzzy numbers. Fuzzy aggregation provides a way of combining or comparing fuzzy quantities. These techniques, similar to MCM, rely on expert judgement to set the range of uncertainty, and their predictions have some measure of uncertainty attached to them. Fisher suggested that the fuzzy method may be more economical in terms of computing, since functions of fuzzy numbers can be used to express the uncertainty in models without the need for the large number of replications required by MCM.

In Bayesian methods (Box and Tiao, 1973), which differ from the classical or frequentist methods discussed above, one starts with approximate *prior* distributions for a set of uncertain parameters, representing the available information. The likelihood of the data, given the model, is then used

to derive the *posterior* distributions of the set of parameters by applying Bayes theorem. Since the posterior distributions are guided by the data, they should have less uncertainty than the prior distributions. The Real-time Online Decision Support system (RODOS; Ehrhardt *et al.*, 1993) of the Commission of European Communities (CEC), which deals with off-site management of nuclear accidents, is based on Bayesian methods (French, 1997). Bayesian Monte Carlo analysis (Bergin and Milford, 2000) provides a means of combining subjective prior distributions developed by MCM with the information about the agreement between model outputs and field observations. The resulting posterior uncertainty estimates reflect the model's performance, as well as subjective judgements about uncertainties in the model inputs and parameters.

#### 4. MODEL PHYSICS UNCERTAINTY

A scientific assessment of the basis and physics of the model formulations should be the first step in a model evaluation. The next step is an evaluation of model performance by comparing its predictions with suitable data from laboratory and/or field experiments. A good model should be based on sound physical principles and give “good” predictions for the “right” reasons. This gives the model user faith in model predictions beyond the range of available data, and confidence in modeling new situations with different dispersion climatologies (Weil *et al.*, 1992). However, it is important to limit model applications and evaluations only to situations for which the model was designed (Rao and Hosker, 1993).

The agreement between the observed and predicted concentrations,  $C_o$  and  $C_p$ , is studied using statistics and scatter plots. The statistics include performance measures of bias and error, such as fractional bias ( $FB$ ) and normalized mean square error ( $NMSE$ ), defined (Hanna, 1988) as

$$FB = 2(\overline{C_o} - \overline{C_p})/(\overline{C_o} + \overline{C_p}), \quad NMSE = \overline{(C_p - C_o)^2}/(\overline{C_o} \overline{C_p}). \quad (19)$$

The figure of merit in space ( $FMS$ ) is a statistical coefficient of the space analysis used for evaluating the plume footprint predicted by the model.  $FMS$  is calculated at a fixed time for a

fixed concentration level, and is defined (e.g., Mosca *et al.*, 1998) as the percentage of overlap of the observed ( $A_o$ ) and predicted ( $A_p$ ) plume contour areas divided by their sum:

$$FMS = [(A_o \cup A_p)/(A_o \cap A_p)] \cdot 100 . \quad (20)$$

A high value of  $FMS$  indicates a good model performance, but a low value need not necessarily correspond to a bad model performance. Two areas very similar in shape but shifted in space (due to errors in mean wind direction, for example) may have a low  $FMS$ . In this case, a simple rotation of the predicted area may increase the  $FMS$  value significantly. For this reason, Mosca *et al.* (1998) suggest evaluation of the  $FMS$  value together with a graphical representation of the observed and predicted contour areas. However, based on the experience of the present author, simple rotation of the predicted contours may not be possible or useful sometimes in improving the  $FMS$  values, especially in cases of complicated plume behavior (e.g., for significant wind shear, with plume direction change).

For hazard prediction models, it may be useful to delineate the overlap, over- and under-prediction regions of plume footprint for a given threshold concentration value. A *false negative* area ( $A_{fn}$ ) is where hazard is observed but not predicted, and a *false positive* area ( $A_{fp}$ ) is where hazard is predicted but not observed. Warner *et al.* (2001) plot a measure of effectiveness ( $MOE$ ) for which the  $x$ -axis corresponds to the ratio of overlap area to observed area, calculated as  $(1 - A_{fn}/A_o)$ , and the  $y$ -axis corresponds to the ratio of overlap area to predicted area, calculated as  $(1 - A_{fp}/A_p)$ . As  $x$  increases from 0 to 1, the false negative area fraction ( $A_{fn}/A_o$ ) decreases from 1 to 0. Similarly, as  $y$  increases from 0 to 1, the false positive area fraction ( $A_{fp}/A_p$ ) decreases from 1 to 0. The  $MOE$  plots are an alternate form of  $FMS$  representation for model evaluation.

The scatter plots of predicted versus observed concentrations are widely used for model evaluation. If  $R$  is the correlation coefficient of the regression fit between  $C_o$  and  $C_p$ , then

$R^2$  indicates the fraction of the variance that is explained by the model. An effective method of assessing model physics uncertainty term (b) in Eq. (3) is to plot residuals  $d = C_o - C_p$  against each of the key model variables separately. Ideally, the points should be symmetrically distributed about  $d = 0$  and show no trend (Venkatram, 1982). If a clear trend is evident in the plot, then there is a need for modifying the model's physics or parameterizations. Often, it is useful to divide the points into a finite number of intervals of each key variable and calculate the mean  $\bar{d}$  and its uncertainty (estimated as 95% confidence limits of a normal distribution using the standard deviation  $S_d$  for the points in each interval,  $\bar{d} \pm 1.96 S_d$ ) and plot them. If the uncertainty limits of  $\bar{d}$  in each interval include zero, then the model error is not statistically significant. Otherwise, the model physics and parameterizations should be improved, and the new results should be plotted until this condition is satisfied. This method can also be used with residual plots of the form  $\ln(C_p/C_o)$  versus each of the key variables, where the 95% confidence limits in each interval are determined using the geometric standard deviation. This was illustrated by Weile *et al.* (1992) and Rao and Hosker (1993). The model physics error analysis based on scatter and residual plots is an iterative process in which the identified model deficiencies are corrected, and the modified model is re-evaluated until the uncertainty is reduced to acceptable limits.

## 5. STOCHASTIC UNCERTAINTY

The stochastic uncertainty,  $\sigma_c^2$  in Eq. (3), is caused by atmospheric turbulence. It arises because of the variability in the details of the velocity field in each realization of the turbulent flow, and the finite averaging-time of the measured concentration.  $\sigma_c$  decreases with an increase in the averaging time. In addition to varying in space and time,  $\sigma_c$  also depends on the meteorological and source conditions defining the ensemble (Chatwin, 1982). It is difficult and expensive to conduct an ensemble of dispersion experiments in the atmosphere. The conditions in the ensemble do not repeat with sufficient frequency because of the inherent variability of the atmosphere. Hence, concentration fluctuations have been often determined from laboratory

experiments (e.g., Fackrell and Robins, 1982; Deardorff and Willis, 1984) or models (e.g., Panwar *et al.*, 1994). However, field studies of concentration fluctuations in full-scale atmospheric experiments facilitate proper representation of large-scale eddy motions, which contribute to plume meander, and the full range of atmospheric stability conditions. Among such field studies are those by Sawford (1987), Mylne and Mason (1991), and Yee *et al.* (1993).

For many dispersion problems in the atmosphere, the concentration fluctuation intensity  $\sigma_c/\overline{C_o} \sim 1$  for averaging times of about 1 h and distances less than 30 km. Laboratory data on tall stack plumes show that  $\sigma_c/\overline{C_o}$  can be as large as 6 near the surface for averaging times of a few minutes. Nevertheless, most atmospheric dispersion models presently used in regulation and safety assessment predict only the ensemble-mean concentration, and generally ignore the concentration fluctuations because of the difficulty in making reliable estimates of  $\sigma_c$  for general atmospheric conditions and arbitrary sources.

The concentration fluctuations include contributions both from in-plume fluctuations (relative diffusion) due to small-scale “inertial subrange” turbulence, and from plume meandering due to large-scale “energy containing” eddies in a turbulent flow (Gifford, 1959; Csanady, 1967). Meandering causes plume intermittency, *i.e.*, the concentration measured at a fixed sampler essentially varies between “in-plume” peaks and a zero value in the environment. Meandering dominates the fluctuations from small sources for downwind travel times  $t < \tau_L$ , the Lagrangian integral time scale. The in-plume component dominates the fluctuations from small sources at large travel times. For a surface release, vertical meandering is small, but lateral meandering can lead to significant concentration fluctuations.

Models for the probability distributions of  $C$  or  $\sigma_c$  are useful in predicting the expected exceedance of threshold concentration value (TCV) for a toxic contaminant dispersing in the atmosphere. Such methods transform the deterministic model result into a probabilistic form. The PDF,

$p(\theta; \vec{x}, t)$ , of the concentration  $C(\vec{x}, t)$  of a hazardous pollutant dispersing in the atmosphere can be defined (Chatwin, 1982) as

$$p(\theta; \vec{x}, t) \delta\theta = \text{prob} [\theta \leq C(\vec{x}, t) < \theta + \delta\theta] . \quad (21)$$

The ensemble mean concentration and variance are then given by

$$\bar{C} = \int_0^{\infty} \theta p d\theta , \quad (22)$$

$$\sigma_c^2 = \int_0^{\infty} (\theta - \bar{C})^2 p d\theta . \quad (23)$$

Measurements of the frequency distribution of fluctuating plume concentrations have revealed that the PDF is strongly skewed to the right with an upper tail that is heavier than that of the Gaussian form (Yee and Chan, 1997). Several models are proposed for the PDF of concentration in the literature over the past two decades. Among them are the lognormal distribution (Csanady, 1973) and the exponential distribution (Hanna, 1984). Lewis and Chatwin (1997) proposed a three-parameter model based on a weighted sum of exponential and generalized Pareto distributions, which was shown to fit the concentration data over different atmospheric conditions. Schopflocher and Sullivan (2002) proposed a double Beta distribution for the PDF of a scalar diffusing in a turbulent flow, which emphasizes the representation of the underlying physical structure of the concentration field, as well as accurate modeling of the high-concentration tails of the distribution.

Lewellen and Sykes (1989) described an advanced modeling approach which generates confidence limits for the very high but rare concentrations, which are important for the atmospheric dispersion of hazardous substances. This method involves predicting the variance of concentration fluctuations from a second-order closure model, and estimating the probability distribution from the mean and the variance using a truncated Gaussian (also referred to as “clipped normal”) distribution, which replaces any unphysical negative tail (i.e., negative

concentrations) in the Gaussian with a delta function at zero concentration. Yee and Chan (1997) showed that a clipped gamma PDF, derived from the same two model-predicted parameters, fits the concentration data better and is more flexible in fitting the full range of observed PDF behavior in a dispersing plume.

The SCIPUFF model (Sykes *et al.*, 1989), which uses this probabilistic approach, is one of the few atmospheric dispersion models that can predict a concentration probability density function as a function of time and space. This PDF can be used to specify probabilities of interest, such as the probability of exceeding a TCV for a given time period at a given location. The expected CDF and associated confidence bounds can be estimated directly by random sampling of the predicted PDF of concentration at each location and time. Model evaluation then consists of checking if the observed concentration samples are consistent with what could be expected from a single realization of the range of CDFs estimated from the concentration PDF; see Lewellen and Sykes (1989) and Sykes *et al.* (1989) for details and examples. Though physically realistic, this method has not been widely used since very few dispersion models are capable of predicting the concentration PDF. In addition, the probabilistic evaluation approach is difficult to interpret.

Hanna and Davis (2002) applied this method for the evaluation of a photochemical grid model by estimating the PDF of predicted hourly concentrations at 66 ozone monitors from 100 MCM runs, assuming that the PDF was completely determined only by the uncertainties in input variables. Using the set of observed ( $C_o$ ) and predicted ensemble-mean ( $\overline{C_p}$ ) concentrations and the estimated PDF, the CDF of observed model residuals ( $C_o - \overline{C_p}$ ) at each location was calculated (by integrating the PDF from 0 to the concentration of interest) and plotted, as shown in Fig. 3. Since this curve fell completely within the 95% confidence bounds of the CDFs of predicted model residuals determined from the 100 MC runs (each MC run gives a new CDF), it was concluded that the model was performing as well as expected. Though this evaluation accounts for the uncertainties in the input variables, it did not account for the model physics errors,

stochastic fluctuations in concentrations, and errors in observed concentrations. Therefore, the actual concentration PDF would likely have a larger variance than the PDF used in this work.

### 5.1 Meteorological Data Errors

Atmospheric dispersion models such as SCIPUFF are intended for use over a wide range of distances from short-range (30 km) boundary layer scale to long-range (3000 km) mesoscale applications, and are generally driven by diagnostic (mass-consistent) wind field models for emergency response applications. The output quality of a diagnostic wind-field model depends directly on the quantity and quality of the input wind data. The uncertainty in the predicted concentration field also critically depends on the detail of the available wind field.

The SCIPUFF model accounts only for the stochastic uncertainty arising from the atmospheric turbulence. However, the model results will also depend on the uncertainties in input data and parameters, as well as on model physics errors. The errors in meteorological inputs result from issues such as the differences among various types of data (e.g., tower, aircraft, rawinsonde, wind profiler, etc), averaging times, and representativeness. Lewellen and Sykes (1989) showed that the meteorological input uncertainty accounts for more than 50% of the total uncertainty in predicting the observed 1-hr ground level concentrations (GLCs) with a second-order closure plume model. They found that the meteorological input error was dominated by the horizontal wind variance. In general, uncertainty in the transport wind direction (Weil *et al.*, 1992) is a major contributor to the total model error even at short distances over flat terrain (Hanna, 1988). Simulation of the effective path of mean transport of pollutants is even more difficult and uncertain in situations involving complex terrain (e.g., Banta *et al.*, 1996) and regional or long range transport (Kahl and Samson, 1986; Stohl, 1998).

## 6. UNCERTAINTY AND REGULATORY AIR QUALITY MODELING

Air quality models used for regulation are generally designed to estimate the ensemble average concentrations, while the air quality standards focus on the extreme values of the distribution of observed pollutant concentrations, *i.e.*, the rare event is more significant than the common event. Thus, there is an inherent uncertainty in using the modeling results in an absolute sense for attainment demonstrations. Hogrefe and Rao (2001) described a probabilistic approach for use in decision-making related to compliance with National Ambient Air Quality Standards (NAAQS) for ozone pollution. They utilized extreme value statistics (Roberts, 1979) and bootstrap resampling techniques (Efron, 1982) to estimate the probability of exceeding the NAAQS for both 1-hr and 8-hr ozone concentrations. Exact theory of extreme values can be applied to calculate the CDF of a specified order statistic (Gumbel, 1958), *e.g.*, fourth-highest value in a sample of 1-hr ozone concentrations over a consecutive 3-year period. Instead of thinking of attainment process in a pass/fail mode in the regulatory framework, such an approach helps policy-makers in assessing the probability that a certain emission control strategy would lead to compliance with the NAAQS. Much of the uncertainty work in the literature has dealt with the characterization of the effects of uncertainties in the data input to regulatory models based on Gaussian plume or puff dispersion. Freeman *et al.* (1986) used the analytical method, Eq. (15), to propagate the input data errors in a plume model. Uncertainties in emission rate and height, wind speed and direction, dispersion parameters, and mixing depth were considered for several stability classes and downwind distances. Irwin *et al.* (1987) used MCM to relate the error bounds of meteorological data input to a Gaussian plume dispersion model to the uncertainty in the estimates of the maximum concentration and its downwind distance from the source.

Rao *et al.* (1985) applied a probabilistic approach to air quality model performance evaluation by determining the model's ability to simulate the tails of the CDF of the observed concentrations. Extreme value statistics and bootstrap resampling techniques were applied to develop confidence

intervals for each percentile value of the CDF of observed tail concentrations, and then the CDF of predicted concentrations was superimposed over it. Air quality model performance evaluation consisted of checking whether the model predictions lie within the confidence intervals of observed concentrations at each probability point. The resampling technique was also applied to the differences between the observed and predicted concentrations to estimate the model uncertainty.

## **7. CONCLUSIONS**

Atmospheric Dispersion is a stochastic phenomenon, and it is necessary to consider the concentration as a random variable that must be described in terms of probabilistic quantities such as ensemble mean, variance and probability distribution. The predicted distribution can be used to specify the probabilities of interest to the user, such as the probability of exceeding a TCV for a particular time period at a given location. It is now widely recognized that uncertainty analysis should be an essential part of modeling the dispersion of flammable and/or hazardous substances in the atmosphere. This paper discussed the various uncertainties in atmospheric dispersion model predictions and outlined available techniques to quantify or reduce them. While this review does not claim to be thorough, an attempt is made to provide a comprehensive discussion of key issues and methods with relevant references.

We have to make decisions in the face of uncertainty. Identification and estimation of uncertainties in predictions yields a deeper insight into the capabilities and limitations of atmospheric dispersion models, and increase our confidence in decision-making based on these models. It provides more credibility to the environmental risk assessment process, and indicates a proper direction for action or further investigation. Uncertainty analysis will enable the assessor to rank the atmospheric contaminants (and their pathways) more accurately, and to derive a subjective probability distribution about which CIs can be formed to represent the uncertainty

in the risk. This information can be used to guide decisions. For example, if a 5% (lower) CI concentration value is above a regulatory standard or threshold value of concern, then appropriate remedial measures (*e.g.*, emission controls, evacuations, etc.) are probably needed. If the 95% (upper) CI is below the standard, remedial action is probably not required. If the 95% upper CI is above the standard but the 50th percentile is below the standard, further study should be recommended on the key parameters that dominate the overall uncertainty. If the 50th percentile is also above the standard, further study might still be recommended, but one might also proceed with cost-effective remedial measures for risk reduction. Thus, incorporation of uncertainty analysis into atmospheric dispersion modeling provides a valuable tool for decision-making and optimal use of resources.

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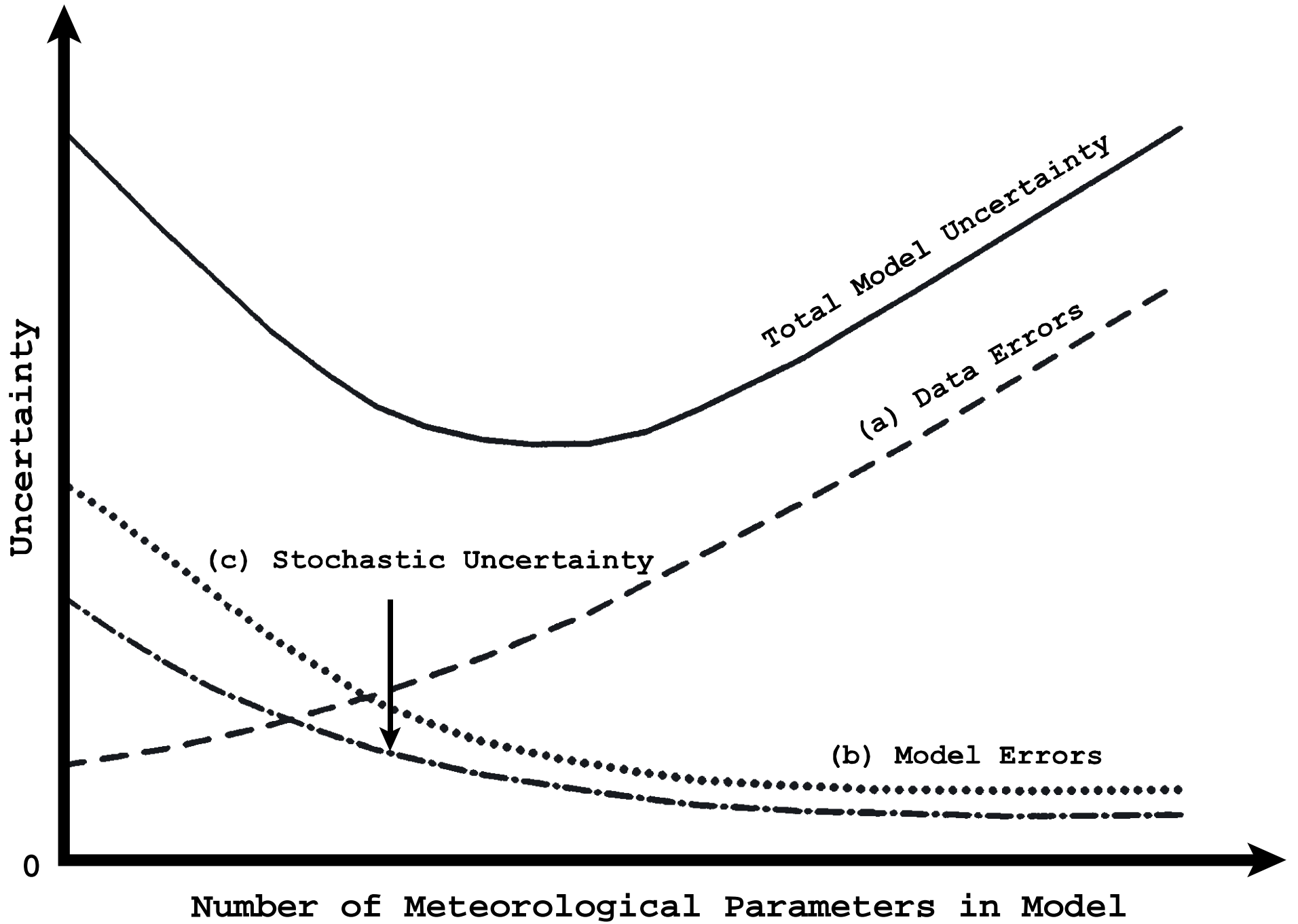
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## FIGURE CAPTIONS

Figure 1. Variations of total model uncertainty and its components versus the number of meteorological parameters in the model input. The various uncertainty components are identified in Eq. (3) and discussed in the text. These conceptual variations are approximate and not drawn to scale.

Figure 2. Probability distributions of input parameters used in uncertainty analysis. Continuous probability distributions are shown by solid figures. Discrete probability distributions are denoted by vertical bars.

Figure 3. CDF for model ozone concentration residuals,  $C_o - \overline{C_p}$ , for 66 monitoring sites in the model domain for 10 July 1995.  $C_o$  is observed concentration and  $\overline{C_p}$  is predicted mean hourly averaged ozone concentration (ppb), and represent maximum values for that day at that monitoring site. Diamonds represent the actual observations. Solid lines denote the 95% confidence bounds, defined as the range of 100 alternate CDFs determined from predictions of 100 Monte Carlo runs. (From Hanna and Davis, 2002).



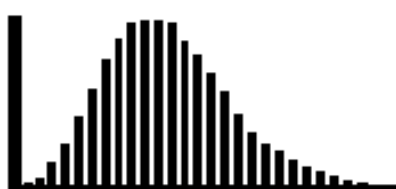
Normal



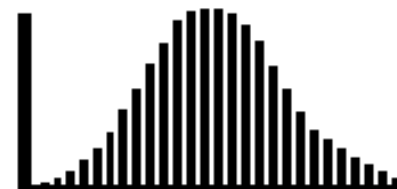
Triangular



Poisson



Binomial



Lognormal



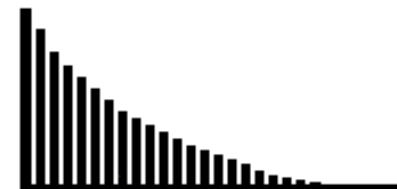
Uniform



Exponential



Geometric



Weibull



Beta



Hypergeometric



Custom

