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Sensitivity Analysis of Partial Differential Equations with Application to Reaction and Diffusion Processes

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Methods for the computation of sensitivity coefficients for constant, spatially varying, and temporally varying parameters in parabolic partial differential equations characteristic of reaction and diffusion processes are developed. Computational requirements associated with the methods are estimated, and the methods are applied to the sensitivity analysis of atmospheric diffusion.

1. INTRODUCTION

In many branches of science and engineering, descriptions of phenomena lead to differential equations of substantial complexity. The complexity of such models makes it difficult to determine the effect small errors in physical and chemical parameters have on solutions. The analysis of the sensitivity of models to perturbations in parameters is called *sensitivity analysis.* Conceptually the simplest approach to a sensitivity analysis is to solve the system equations repeatedly while varying one parameter at a time and holding the others fixed. This type of analysis soon becomes impractical as the number of parameters subject to variation increases.

When the system states, u_i, $i = 1, 2,..., n$, are dependent on a set of constant parameters, k_j, $j = 1, 2,..., m$, the sensitivity information is embodied in the so-called *sensitivity coefficients*, $\partial u_i/\partial k_j$, $i = 1, 2,..., n$; $j = 1, 2,..., m$. By differentiating the differential equations comprising the model with respect to k_j, a set of differential equations for the nm sensitivity coefficients, $\partial u_i/\partial k_j$, can be derived [1]. These so-called sensitivity equations have the same basic mathematical structure as the original model equations. Given the sensitivity coefficients, the variations in the system states in the neighborhood of a nominal set of parameter values $\bar{k}_j$ are given by

$$u_i(\bar{\mathbf{k}} + \delta\mathbf{k}) = u_i(\bar{\mathbf{k}}) + \sum_{j=1}^{m} \frac{\partial u_i}{\partial k_j}\, \delta k_j\,, \tag{1}$$

where the implicit dependence of the state u_i on the parameter vector, $\mathbf{k} = [k_1, k_2,..., k_m]^T$, is indicated by $u_i(\mathbf{k})$.

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Most of the work on sensitivity analysis has been concerned with calculation of the sensitivity coefficients, $\partial u_i/\partial k_j$, for models described by differential equations [1, 2]. Recently a new sensitivity analysis method has been developed by Shuler *et al.* [3–6]. The method is based on the assignment of periodic functions of a new variable s to each parameter, $k_j(s)$. The periodic functions are related to the distributions assigned to the parameters. Each value of s determines a value for $\mathbf{k}(s)$, for which the solution of the model produces a quasi-periodic function $\mathbf{u}(s)$. Sufficient values of $\mathbf{u}$ must be generated to enable the Fourier amplitudes of $\mathbf{u}(s)$ to be computed. A unique frequency ω_j indicates the dependence of the solution on k_j.

In models consisting of partial differential equations, there may exist, in addition to constant parameters (those independent of time and location, such as chemical reaction rate constants), parameters that are spatially varying or temporally varying. When the parameters are functions of the independent variables of the problem, e.g., space and time, the sensitivity analysis becomes much more complex than when the parameters are constants. Porter [7] was apparently the first to consider this problem. He suggested that the parameters be represented by expansions in orthogonal basis functions, leading to a lumped parameter system, for which the sensitivity equations can be obtained in a straightforward manner. For spatially and temporally varying parameters, the quantities of direct interest in a sensitivity analysis are the *functional derivatives* $\delta u_i(\mathbf{x}', t)/\delta k_j(\mathbf{x})$ and $\delta u_i(\mathbf{x}, t')/\delta k_j(t)$. The functional derivatives are defined such that the change in $u_i(\mathbf{x}, t)$ as a result of perturbations in the parameters $k_j(\mathbf{x})$ and $k_j(t)$ are

$$\delta u_i(\mathbf{x}', t) = \int \frac{\delta u_i(\mathbf{x}', t)}{\delta k_j(\mathbf{x})}\, \delta k_j(\mathbf{x})\, d\mathbf{x} \tag{2}$$

and

$$\delta u_i(\mathbf{x}, t') = \int \frac{\delta u_i(\mathbf{x}, t')}{\delta k_j(t)}\, \delta k_j(t)\, dt, \tag{3}$$

respectively. Thus, for a specific value of the spatial variables, $\mathbf{x}'$, $\delta u_i(\mathbf{x}', t)/\delta k_j(\mathbf{x})$ is the sensitivity of u_i at location $\mathbf{x}'$ to variations in $k_j(\mathbf{x})$ at any time t. Likewise, for a specific value of time t', $\delta u_i(\mathbf{x}, t')/\delta k_j(t)$ represents the sensitivity of u_i at time t' to variations in $k_j(t)$ at any location $\mathbf{x}$.

In certain problems the location of one or more of the boundaries is uncertain. In such a case it is desirable to be able to calculate the sensitvity of the state with respect to the boundary position. A related problem concerned with estimation of the location of the boundary of a petroleum reservoir has been considered by Chen and Seinfeld [8], although the desired functional derivatives were not computed in that work.

The objects of this paper are as follows. First, we wish to organize and present in a unified manner the various approaches to the calculation of sensitivity coefficients for parameters in partial differential equations. Second, we desire to develop methods for the calculation of the functional derivative sensitivity coefficients for spatially and temporally varying parameters and boundary position. Third, we wish to develop guidelines for which method should be used for a given set of circumstances. Finally, we wish to apply the results to the sensitivity analysis of vertical diffusion in the atmosphere.

2. Statement of the Problem

We seek to review and develop techniques for sensitivity analysis of models consisting of sets of nonlinear partial differential equations containing constant, spatially varying, and temporally varying parameters and temporally varying boundary locations. In order to minimize the complexity associated with dealing with a model of such extreme generality, we choose to define a rather specific system for which we will develop all our results. The development is in no way restricted by the class of systems we have chosen, and all results we will obtain can be extended with little difficulty to the more general classes of partial differential equations.

We consider a system governed by the following set of partial differential equations, initial and boundary conditions,

$$\frac{\partial u_i(x, t)}{\partial t} = \frac{\partial}{\partial x}\left(K(x)\frac{\partial u_i}{\partial x}\right) + R_i(u_1, \ldots, u_n; k_1, \ldots, k_m), \tag{4}$$

$$u_i(x, 0) = u_{i_0}(x), \tag{5}$$

$$-K(0)\frac{\partial u_i}{\partial x} = S_i(t), \qquad x = 0, \tag{6}$$

$$\frac{\partial u_i}{\partial x} = 0, \qquad x = H(t). \tag{7}$$

The system of Eqs. (4)–(7) describes, for example, simultaneous diffusion and chemical reaction in a variable domain $(0, H(t))$. The diffusion coefficient $K(x)$ is taken to be a function of position x. The functions R_i are prescribed nonlinear functions of the n state variables, $u_1, u_2, \ldots, u_n$, and of m parameters, $k_1, k_2, \ldots, k_m$. The R_i can arise, for example, from a set of chemical reaction rate equations if the u_i represent species concentrations. In that case the k_j are reaction rate constants. The initial conditions, $u_{i_0}(x)$, are assumed to be known without error. The boundary condition at $x = 0$ expresses that the flux of state variable i is equal to a function $S_i(t)$, whereas at the $x = H(t)$ boundary there is no flux across the boundary but the location of the boundary itself changes with time.

The system of Eqs. (4)–(7) arises naturally in the study of vertical diffusion and chemical reaction in the atmosphere. One-dimensional models of stratospheric chemistry and transport are based on (5)–(8) [9]. In addition, so-called trajectory models describing simultaneous vertical diffusion and chemical reaction of air pollutants in an air parcel adjacent to the ground are based on (4)–(7) [10]. In the air pollution trajectory model application, $H(t)$ represents the elevation of the layer adjacent to the ground within which pollutants are emitted, mixed, and react. Mathematical models for the one-dimensional flow of oil in a porous medium are a special case of (4) [11]. Specifically, for $n = 1$, $u(x, t)$ represents the oil pressure, and $R = 0$, yielding[1]

$$\frac{\partial u(x, t)}{\partial t} = \frac{\partial}{\partial x}\left(K(x)\frac{\partial u}{\partial x}\right). \tag{8}$$

Whereas the system of (4)–(7) is directly relevant to problems in atmospheric transport and chemistry and, in a simplified form, to flow of oil in petroleum reservoirs, it contains all the elements desired for the more general sensitivity analysis problem in partial differential equations, namely, spatially ($K(x)$) and temporally ($S_i(t)$) varying, as well as constant ($k_1, k_2, \ldots, k_m$) parameters and temporally varying boundary location ($H(t)$).

We now wish to review existing methods and, where necessary, develop new methods for the sensitivity analysis of the system (4)–(7) with respect to $K(x)$, $S_i(t)$, k_j, and $H(t)$. In particular, the new methods we will develop are concerned with the computation of the functional derivative sensitivity coefficients, $\delta u_i(x', t)/\delta K(x)$, $\delta u_i(x, t')/\delta S_j(t)$, and $\delta u_i(x, t')/\delta H(t)$.

3. Theory of Sensitivity Analysis

There are three approaches to sensitivity analysis that we will consider—the direct method, the variational method, and the Fourier amplitude sensitivity test (FAST) method of [3–6]. The direct method is based on considering all parameters as constants. In the direct method spatially varying parameters such as $K(x)$ are approximated by a finite number of constant parameters, K_j, $j = 1, 2, \ldots, N$, and then sensitivity coefficients, $\partial u_i/\partial K_j$, $i = 1, 2, \ldots, n$; $j = 1, 2, \ldots, N$, are computed. Likewise, temporally varying parameters such as $S_i(t)$ are represented by a set of constant parameters, S_{ij}, $i = 1, 2, \ldots, n$; $j = 1, 2, \ldots, M$, and the sensitivity coefficients, $\partial u_i/\partial S_{jl}$, $i, j = 1, 2, \ldots, n$; $l = 1, 2, \ldots, M$, are computed. In the variational approach the distributed nature of the parameters is retained, and the sensitivity coefficients are calcalculated based on the introduction of adjoint variables, with a finite-dimensional approximation introduced only at the end to solve the state and adjoint equations. The functional derivative sensitivity coefficients, $\delta u_i(x', t)/\delta K(x)$, $\delta u_i(x, t')/\delta S_j(t)$, and $\delta u_i(x, t')/\delta H(t)$, can be computed only from a variational approach.

We note that the direct and variational methods are linearized theories, strictly valid only for small parameter uncertainties. Thus, in the use of these two methods, it is assumed that the effect of parameter variations on the state variables is small. The FAST method, on the other hand, is applicable for nonlinear sensitivity analysis with respect to large parameter uncertainties. Subsequently, we will compare the computational requirements of the three methods. Such a comparison assumes that

[1] In models for oil flow in porous media $K(x)$ is related to the permeability of the medium. Usually there is also another parameter $\phi(x)$, related to the porosity of the medium, multiplying $\partial u/\partial t$. One is concerned with the sensitivity of the pressure $u(x, t)$ to variations in both $K(x)$ and $\phi(x)$.

all three methods would be used for the same problem. Because of the limitations of the direct and variational methods, such a problem would of necessity involve small parameter variations.

3.1. *Direct Method*

Constant Parameters. The sensitivity coefficients $\beta_{ij}(x, t) = \partial u_i(x, t)/\partial k_j$ are governed by the sensitivity equations,

$$\frac{\partial \beta_{ij}(x, t)}{\partial t} = \frac{\partial}{\partial x}\left(K(x)\frac{\partial \beta_{ij}}{\partial x}\right) + \sum_{l=1}^{n} \frac{\partial R_i}{\partial u_l}\beta_{lj} + \frac{\partial R_i}{\partial k_j}, \tag{9}$$

$$\beta_{ij}(x, 0) = 0, \tag{10}$$

$$\frac{\partial \beta_{ij}}{\partial x} = 0, \qquad x = 0, H(t). \tag{11}$$

The direct approach consists of solving (9)–(11) to obtain the nm sensitivity coefficients, $\beta_{ij}(x, t)$, $i = 1, 2,..., n$; $j = 1, 2,..., m$.

Spatially Varying Parameters. First the partial differential equations are reduced to a set of ordinary differential equations in time by an appropriate spatial approximation such as finite differences. The state $u_i(x, t)$ and the parameter $K(x)$ are thus transformed into finite-dimensional form, $u_{il}(t)$ and K_l, $l = 1, 2,..., N$. The result is a set of coupled ordinary differential equations containing constant parameters. For simplicity, we consider the linear scalar case of (8). After finite-dimensional approximation and inclusion of the boundary conditions (6) and (7), (8) becomes

$$\frac{d\mathbf{U}}{dt} = \mathbf{A}(\mathbf{K})\,\mathbf{U} + \mathbf{S}(t), \tag{12}$$

where $\mathbf{U}(t) = [u_1(t),..., u_N(t)]^T$ and $\mathbf{S}(t) = [S_1(t), 0,..., 0]^T$. Equation (12) is solved subject to $\mathbf{U}(0) = \mathbf{U}_0$. The solution of (12) is

$$\mathbf{U}(t) = e^{\mathbf{A}(\mathbf{K})t}\left(\mathbf{U}_0 + \int_0^t e^{-\mathbf{A}(\mathbf{K})\tau}\mathbf{S}(\tau)\,d\tau\right). \tag{13}$$

Let

$$\boldsymbol{\beta}_j = \left(\frac{\partial u_1}{\partial K_j}, \frac{\partial u_2}{\partial K_j},..., \frac{\partial u_N}{\partial K_j}\right)^T, \qquad j = 1, 2,..., N, \tag{14}$$

be the sensitivity vector for parameter K_j. Next, let us differentiate (12) with respect to K_j. The following vector differential equation is obtained:

$$\frac{d\boldsymbol{\beta}_j}{dt} = \mathbf{A}(\mathbf{K})\,\boldsymbol{\beta}_j + \frac{\partial \mathbf{A}(\mathbf{K})}{\partial K_j}\,\mathbf{U}(t), \tag{15}$$

$$\boldsymbol{\beta}_j(0) = \mathbf{0}, \qquad j = 1, 2,..., N. \tag{16}$$

Equation (15) is again a linear initial value problem of the same type as (12). The solution of (15) is

$$\boldsymbol{\beta}_j(t) = e^{\mathbf{A}(\mathbf{K})t} \int_0^t e^{-\mathbf{A}(\mathbf{K})\tau} \frac{\partial \mathbf{A}(\mathbf{K})}{\partial K_j} \mathbf{U}(\tau)\, d\tau. \tag{17}$$

$\mathbf{U}(t)$ from (13) can be substituted into (17) and integrations can be carried out analytically. (See the Appendix for a discussion of the evaluation of (13) and (17).)

3.2. *Variational Method*

Variations in $K(x)$, $S_j(t)$, and k_j. We consider first the case in which $H(t) = H$ and is not subject to variation. Our object is to derive equations for the functional derivative sensitivity coefficients, $\delta u_i(x', t)/\delta K(x)$ and $\delta u_i(x, t')/\delta S_j(t)$, and the sensitivity coefficients $\partial u_i(x, t)/\partial k_j$.

Consider simultaneous perturbations in $K(x)$, $S_j(t)$, and k_j, $\delta K(x)$, $\delta S_j(t)$, and δk_j, $\bar{K}(x)$, $\bar{S}_j(t)$, $\bar{k}_j$, that lead to perturbations, $\delta u_i(x, t)$, $i = 1, 2,\ldots, n$, about a nominal value $\bar{u}_i(x, t)$. To first order, $\delta u_i(x, t)$ satisfies

$$\frac{\partial\, \delta u_i}{\partial t} = \frac{\partial}{\partial x}\left(\bar{K}(x) \frac{\partial\, \delta u_i}{\partial x}\right) + \frac{\partial}{\partial x}\left(\delta K(x) \frac{\partial \bar{u}_i}{\partial x}\right) + \sum_{j=1}^{n} \frac{\partial \bar{R}_i}{\partial u_j} \delta u_j + \sum_{j=1}^{m} \frac{\partial \bar{R}_i}{\partial k_j} \delta k_j, \tag{18}$$

$$\delta u_i(x, 0) = 0, \tag{19}$$

$$-\bar{K}(0) \frac{\partial\, \delta u_i}{\partial x} - \delta K(0) \frac{\partial \bar{u}_i}{\partial x} = \delta S_i(t), \qquad x = 0, \tag{20}$$

$$\frac{\partial\, \delta u_i}{\partial x} = 0, \qquad x = H. \tag{21}$$

Let us multiply (18) by a sequence of arbitrary functions $\{\psi_i(x, t)\}$ and sum from $i = 1$ to $i = n$,

$$\sum_{i=1}^{n} \psi_i \frac{\partial\, \delta u_i}{\partial t} = \sum_{i=1}^{n} \psi_i \frac{\partial}{\partial x}\left(\bar{K}(x) \frac{\partial\, \delta u_i}{\partial x}\right) + \sum_{i=1}^{n} \psi_i \frac{\partial}{\partial x}\left(\delta K(x) \frac{\partial \bar{u}_i}{\partial x}\right) + \sum_{i=1}^{n} \sum_{j=1}^{n} \psi_i \frac{\partial \bar{R}_i}{\partial u_j} \delta u_j + \sum_{i=1}^{n} \sum_{j=1}^{m} \psi_i \frac{\partial \bar{R}_i}{\partial k_j} \delta k_j. \tag{22}$$

Integration of (22) with respect to x over $[0, H]$ and with respect to t over $[0, T]$, subsequent integration by parts, and use of (19)–(21) lead to

$$\sum_{i=1}^{n} \int_0^H \psi_i(x, T)\, \delta u_i(x, T)\, dx = \sum_{i=1}^{n} \int_0^H \int_0^T \left[\frac{\partial \psi_i}{\partial t} + \frac{\partial}{\partial x}\left(\bar{K}(x) \frac{\partial \psi_i}{\partial x}\right) + \sum_{j=1}^{n} \frac{\partial \bar{R}_j}{\partial u_i} \psi_j\right] \delta u_i\, dt\, dx$$

$$- \sum_{i=1}^{n} \int_0^H \int_0^T \delta K(x) \frac{\partial \bar{u}_i}{\partial x} \frac{\partial \psi_i}{\partial x} \, dt \, dx + \sum_{i=1}^{n} \int_0^T \delta S_i(t) \, \psi_i(0, t) \, dt$$

$$+ \sum_{i=1}^{n} \sum_{j=1}^{m} \int_0^H \int_0^T \psi_i \frac{\partial \bar{R}_i}{\partial k_j} \delta k_j \, dt \, dx$$

$$- \sum_{i=1}^{n} \int_0^T \left[\bar{K}(x) \, \delta u_i \frac{\partial \psi_i}{\partial x} \bigg|_{x=H} - \bar{K}(x) \, \delta u_i \frac{\partial \psi_i}{\partial x} \bigg|_{x=0} \right] dt. \tag{23}$$

If we now specify that $\psi_i(x, t)$ satisfy

$$\frac{\partial \psi_i}{\partial t} = - \frac{\partial}{\partial x} \left(\bar{K}(x) \frac{\partial \psi_i}{\partial x} \right) - \sum_{j=1}^{n} \frac{\partial \bar{R}_j}{\partial u_i} \psi_j , \qquad i = 1, 2, \ldots, n, \tag{24}$$

$$\frac{\partial \psi_i}{\partial x} = 0, \qquad x = 0, H, \tag{25}$$

with the terminal condition as

$$\psi_i(x, T) = \delta_{il} \delta(x - x'), \qquad i = 1, 2, \ldots, n; \; l = 1, 2, \ldots, n, \tag{26}$$

where δ_{il} and $\delta(x - x')$ are the Kronecker and Dirac deltas, respectively, then (23) becomes

$$\delta u_l(x', T) = - \sum_{i=1}^{n} \int_0^H \delta K(x) \int_0^T \frac{\partial \bar{u}_i}{\partial x} \frac{\partial \psi_{il}}{\partial x} \, dt \, dx$$

$$+ \sum_{i=1}^{n} \int_0^T \delta S_i(t) \, \psi_{il}(0, t) \, dt$$

$$+ \sum_{j=1}^{m} \delta k_j \sum_{i=1}^{n} \int_0^H \int_0^T \frac{\partial \bar{R}_i}{\partial k_j} \psi_{il} \, dt \, dx, \tag{27}$$

where ψ_{il} denotes the solution of (24) and (25) subject to (26). The desired functional derivatives are, therefore,

$$\frac{\delta u_l(x', T)}{\delta K(x)} = - \sum_{i=1}^{n} \int_0^T \frac{\partial \bar{u}_i}{\partial x} \frac{\partial \psi_{il}}{\partial x} \, dt, \tag{28}$$

$$\frac{\delta u_l(x', T)}{\delta S_j(t)} = \psi_{jl}(0, t), \tag{29}$$

$$\frac{\partial u_l(x', T)}{\partial k_j} = \sum_{i=1}^{n} \int_0^H \int_0^T \frac{\partial \bar{R}_i}{\partial k_j} \psi_{il} \, dt \, dx. \tag{30}$$

Variation in $H(t)$. Our object is to derive equations for the functional derivative sensitivity coefficients $\delta u_i(x', t)/\delta K(x)$, $\delta u_i(x, t')/\delta S_j(t)$, and $\delta u_i(x, t')/\delta H(t)$, where we now allow H to be a function of time.

We note that for the system of (4) variations in $H(t)$ may be reduced by a suitable coordinate transformation to equivalent variations in $K(x)$. We represent the variation in $H(t)$ as

$$\begin{aligned} H(t) &= \bar{H}\mu(t), \\ \mu(t) &= 1 + \delta H(t), \end{aligned} \tag{31}$$

where $\bar{H}$ is the constant nominal value of $H(t)$. The variation $\delta H(t)$ is dimensionless, i.e., $\delta H(t) = (H(t) - \bar{H})/\bar{H}$. We now define the coordinate transformation $z(x, t) = x/\mu(t)$. Then the system becomes

$$\frac{\partial u_i}{\partial t} = \frac{\partial}{\partial z}\left\{\frac{K(\mu(t)\, z)}{\mu(t)^2}\frac{\partial u_i}{\partial z}\right\} + R_i(\mathbf{u}; \mathbf{k}), \tag{32}$$

$$u_i(z, 0) = u_{i_0}(z), \tag{33}$$

$$-K(0)\frac{\partial u_i}{\partial z} = S_i(t)\, \mu(t), \qquad z = 0, \tag{34}$$

$$\frac{\partial u_i}{\partial z} = 0, \qquad z = \bar{H}. \tag{35}$$

Thus the original system has been transformed into (32)–(35) with the fixed spatial domain $[0, \bar{H}]$.

Using Taylor's theorem to expand $K(\mu(t)\, z)/\mu(t)^2$ around the nominal function $\bar{K}(z)$, and retaining only first-order variation terms, we have

$$\frac{K(\mu(t)\, z)}{\mu(t)^2} = \bar{K}(z) + \delta K(z) + \left[z\frac{\partial \bar{K}}{\partial z} - 2\bar{K}(z)\right] \delta H(t) \tag{36}$$

Similarly, we have the first-order approximation

$$S_i(t)\, \mu(t) = \bar{S}_i(t) + \delta S_i(t) + \bar{S}_i(t)\, \delta H(t). \tag{37}$$

If $\delta H(t) = 0$, then (36) and (37) collapse to $\delta K(z) = K(z) - \bar{K}(z)$ and $\delta S_i(t) = S_i(t) - \bar{S}_i(t)$.

We now resolve the state $u_i(z, t)$ into nominal and variational components, $u_i(z, t) = \bar{u}_i(z, t) + \delta u_i(z, t)$, and obtain the following nominal system equations

$$\frac{\partial \bar{u}_i}{\partial t} = \frac{\partial}{\partial z}\left\{\bar{K}(z)\frac{\partial \bar{u}_i}{\partial z}\right\} + R_i(\bar{\mathbf{u}}, \bar{\mathbf{k}}), \qquad i = 1, 2,..., n, \tag{38}$$

$$\bar{u}_i(z, 0) = u_{i0}(z), \tag{39}$$

$$-\bar{K}(0)\frac{\partial \bar{u}_i}{\partial z} = \bar{S}_i(t), \qquad z = 0, \tag{40}$$

$$\frac{\partial \bar{u}_i}{\partial z} = 0, \qquad z = \bar{H}. \tag{41}$$

For first-order variational equations, we have

$$\frac{\partial\, \delta u_i}{\partial t} = \frac{\partial}{\partial z}\left\{\bar{K}(z)\frac{\partial\, \delta u_i}{\partial z}\right\} + \frac{\partial}{\partial z}\left\{\delta K(z)\frac{\partial \bar{u}_i}{\partial z}\right\}$$
$$+ \frac{\partial}{\partial z}\left\{\left[z\frac{d\bar{K}}{dz} - 2\bar{K}(z)\right]\frac{\partial \bar{u}_i}{\partial z}\right\}\delta H(t)$$
$$+ \sum_{j=1}^{n}\frac{\partial R_i(\bar{\mathbf{u}}, \bar{\mathbf{k}})}{\partial u_j}\delta u_j + \sum_{j=1}^{m}\frac{\partial R_i(\bar{\mathbf{u}}, \bar{\mathbf{k}})}{\partial k_j}\delta k_j, \qquad (42)$$

$$\delta u_i(z, 0) = 0, \qquad (43)$$

$$-\bar{K}(0)\frac{\partial\, \delta u_i}{\partial z} - \delta K(0)\frac{\partial \bar{u}_i}{\partial z} = \delta S_i(t) + \bar{S}_i(t)\,\delta H(t), \qquad z = 0, \qquad (44)$$

$$\frac{\partial\, \delta u_i}{\partial z} = 0, \qquad z = \bar{H} \qquad (45)$$

Let us multiply Eq. (42) by a sequence of arbitrary functions $\{\psi_i(z, t)\}$ and sum from $i = 1$ to $i = n$:

$$\sum_{i=1}^{n}\psi_i\frac{\partial\, \delta u_i}{\partial t} = \sum_{i=1}^{n}\psi_i\frac{\partial}{\partial z}\left\{\bar{K}(z)\frac{\partial\, \delta u_i}{\partial z}\right\} + \sum_{i=1}^{n}\sum_{j=1}^{n}\psi_j\frac{\partial \bar{R}_j}{\partial u_i}\delta u_i$$
$$+ \sum_{i=1}^{n}\psi_i\frac{\partial}{\partial z}\left\{\delta K(z)\frac{\partial \bar{u}_i}{\partial z}\right\}$$
$$+ \sum_{i=1}^{n}\psi_i\frac{\partial}{\partial z}\left\{\left[z\frac{d\bar{K}}{dz} - 2\bar{K}(z)\right]\frac{\partial \bar{u}_i}{\partial z}\right\}\delta H(t)$$
$$+ \sum_{i=1}^{n}\sum_{j=1}^{m}\psi_i\frac{\partial \bar{R}_i}{\partial k_j}\delta k_j. \qquad (46)$$

Integration of Eq. (46) with respect to z over $[0, \bar{H}]$ and with respect to t over $[0, T]$ and subsequent integration by parts yield

$$\sum_{i=1}^{n}\int_0^{\bar{H}}\psi_i(z, T)\,\delta u_i(z, T)\,dz$$
$$= \sum_{i=1}^{n}\int_0^T\int_0^{\bar{H}}\left[\frac{\partial \psi_i}{\partial t} + \frac{\partial}{\partial z}\left\{\bar{K}(z)\frac{\partial \psi_i}{\partial z}\right\} + \sum_{j=1}^{n}\frac{\partial \bar{R}_j}{\partial u_i}\psi_j\right]\delta u_i\,dz\,dt$$
$$- \sum_{i=1}^{n}\int_0^T\left[\bar{K}(z)\,\delta u_i\frac{\partial \psi_i}{\partial z}\bigg|_{z=\bar{H}} - \bar{K}(z)\,\delta u_i\frac{\partial \psi_i}{\partial z}\bigg|_{z=0}\right]dt$$
$$- \sum_{i=1}^{n}\int_0^T\int_0^{\bar{H}}\delta K(z)\frac{\partial \bar{u}_i}{\partial z}\frac{\partial \psi_i}{\partial z}\,dz\,dt + \sum_{i=1}^{n}\int_0^T\delta S_i(t)\,\psi_i(0, t)\,dt$$

$$+ \sum_{i=1}^{n} \int_0^T \delta H(t) \left\{ -\bar{S}_i(t)\, \psi_i(0, t) + \int_0^{\bar{H}} \left[2\bar{K}(z) - z \frac{d\bar{K}}{dz} \right] \frac{\partial \bar{u}_i}{\partial z} \frac{\partial \psi_i}{\partial z} dz \right\} dt$$

$$- \sum_{i=1}^{n} \sum_{j=1}^{m} \int_0^T \int_0^{\bar{H}} \delta k_j \frac{\partial \bar{R}_i}{\partial k_j} \psi_i \, dz\, dt. \tag{47}$$

We now specify that $\psi_i(z, t)$ satisfy

$$\frac{\partial \psi_i}{\partial t} = - \frac{\partial}{\partial z} \left\{ \bar{K}(z) \frac{\partial \psi_i}{\partial z} \right\} + \sum_{j=0}^{n} \frac{\partial \bar{R}_j}{\partial u_i} \psi_j , \tag{48}$$

$$\frac{\partial \psi_i}{\partial z} = 0, \qquad z = 0, \bar{H}. \tag{49}$$

We specify the terminal conditon

$$\psi_i(z, T) = \delta_{il} \delta(z - z'), \qquad i, l = 1, 2, \ldots, n. \tag{50}$$

With this specification, $\delta u_l(z', T)$ is given by

$$\delta u_l(z', T) = - \sum_{i=1}^{n} \int_0^{\bar{H}} \delta K(z) \int_0^T \frac{\partial \bar{u}_i}{\partial z} \frac{\partial \psi_{il}}{\partial z} dt\, dz$$

$$+ \sum_{i=1}^{n} \int_0^T \delta S_i(t)\, \psi_{il}(0, t)\, dt$$

$$+ \sum_{i=1}^{n} \int_0^T \delta H(t) \left\{ -\bar{S}_i(t)\, \psi_{il}(0, t) + \int_0^{\bar{H}} \left[2\bar{K}(z) - z \frac{d\bar{K}}{dz} \right] \frac{\partial \bar{u}_i}{\partial z} \frac{\partial \psi_{il}}{\partial z} dz \right\} dt$$

$$+ \sum_{i=1}^{n} \sum_{j=1}^{m} \delta k_j \int_0^T \int_0^{\bar{H}} \frac{\partial \bar{R}_i}{\partial k_j} \psi_{il}\, dz\, dt, \tag{51}$$

where $\psi_{il}(z, t)$ denotes the solution of (48) and (49) subject to the terminal condition (50).

From (51), we obtain the following functional derivative sensitivity coefficients:

$$\frac{\delta u_l(z', T)}{\delta K(z)} = - \sum_{i=1}^{n} \int_0^T \frac{\partial \bar{u}_i}{\partial z} \frac{\partial \psi_{il}}{\partial z} dt, \tag{52}$$

$$\frac{\delta u_l(z', T)}{\delta S_j(t)} = \psi_{jl}(0, t), \tag{53}$$

$$\frac{\delta u_l(z', T)}{\delta H(t)} = \sum_{i=1}^{n} \left\{ -\bar{S}_i(t)\, \psi_{il}(0, t) + \int_0^{\bar{H}} \left[2\bar{K}(z) - z \frac{d\bar{K}}{dz} \right] \frac{\partial \bar{u}_i}{\partial z} \frac{\partial \psi_{il}}{\partial z} dz \right\}, \tag{54}$$

$$\frac{\partial u_l(z', T)}{\partial k_j} = \sum_{i=1}^{n} \int_0^T \int_0^{\bar{H}} \frac{\partial \bar{R}_i}{\partial k_j} \psi_{il}\, dz\, dt. \tag{55}$$

To compute the sensitivity functions (53)–(55) we need the nominal solution $\{\bar{u}_i(z, t), i = 1, 2,..., n\}$ to (38)–(41) and adjoint function $\{\psi_{il}(z, t), i, l = 1, 2,..., n\}$ to (48)–(50).

3.3. *Fourier Amplitude Sensitivity Test (FAST) Method* [3–6]

The "Fourier amplitude sensitivity test" method essentially involves the computation of the amplitudes of an expansion of the system state. For a frequancy ω_l assigned to the parameter k_l, the corresponding amplitude of the state u_i is given by

$$A_{\omega_l}^{(i)} = \frac{1}{\pi} \int_0^{2\pi} u_i(\mathbf{k}(s)) \sin \omega_l s \, ds. \tag{56}$$

To compute this integral, $\mathbf{u}(\mathbf{k}(s))$ must be evaluated at a set of points in the interval $0 \leqslant s \leqslant 2\pi$. For a system with m constant parameters, the empirical formula for the number of integrals required to compute $A_{\omega_l}^{(i)}$ $(l = 1, 2,..., m)$ is $O[m^{M/2+\gamma}]$, where γ is constant, usually equal to 0.5, and M is an integer defined by a frequency set chosen to be free of interferences. For a spatially varying parameter $K(x)$, we apply finite differences, and $K(x)$ is approximated by a finite number of constant parameters K_j, $j = 1, 2,..., N$.

4. Example. Atmospheric Diffusion

We wish to illustrate the direct, variational, and FAST approaches to sensitivity analysis with a simple example representing vertical diffusion of an inert species in the atmosphere. We have chosen this system because the functional derivative sensitivity coefficients can be obtained analytically. The sensitivity coefficients from the direct and variational methods can be compared to any desired degree of accuracy since exact solutions are available. (The sensitivity information from the FAST method is not amenable to direct comparison with those of the other two methods.)

We consider the system

$$\frac{\partial u}{\partial \tau} = \frac{\partial}{\partial \xi}\left(K(\xi)\frac{\partial u}{\partial \xi}\right), \tag{57}$$

$$u(\xi, 0) = 0, \tag{58}$$

$$-K(0)\frac{\partial u}{\partial \xi} = S(\tau), \qquad \xi = 0, \tag{59}$$

$$\frac{\partial u}{\partial \xi} = 0, \qquad \xi = H(\tau). \tag{60}$$

Our object is to determine the sensitivity of $u(\xi, \tau)$ with respect to variations in $K(\xi)$, $S(\tau)$, and $H(\tau)$. We will apply the direct, variational, and FAST approaches for $K(\xi)$

and only the variational approach for $S(\tau)$ and $H(\tau)$. The nominal values of $K(\xi)$, $S(\tau)$, and $H(\tau)$ are chosen to be constants, $\bar{K}$, $\bar{S}$, and $\bar{H}$.

Using the normalized variables $x = \xi/\bar{H}$ and $t = \tau/T$ where T is the final time, (57)–(60) become

$$\frac{\partial u}{\partial t} = \frac{\bar{K}T}{\bar{H}^2}\frac{\partial^2 u}{\partial x^2}, \qquad t > 0, \tag{61}$$

$$u(x, 0) = 0, \qquad 0 \leqslant x \leqslant 1, \tag{62}$$

$$-\frac{\bar{K}}{\bar{H}}\frac{\partial u}{\partial x} = \bar{S}, \qquad x = 0, \tag{63}$$

$$\frac{\partial u}{\partial x} = 0, \qquad x = 1 \tag{64}$$

4.1. *Direct Method*

Let us divide the x-domain into $(N - 1)$ equal intervals (N grid points) and approximate the spatial derivative in (61) by a central difference scheme for each grid point. Consequently, the following linear matrix differential equation of the form (12) is obtained:

$$\frac{d\mathbf{U}}{dt} = \frac{\bar{K}T}{\Delta x^2\,\bar{H}^2}\begin{pmatrix} -1 & 1 & 0 & & \cdots & 0 \\ 1 & -2 & 1 & 0 & \cdots & 0 \\ 0 & 1 & -2 & 1 & & \vdots \\ \vdots & & & \ddots & & \vdots \\ \vdots & & & & \ddots & 0 \\ 0 & & \cdots & 0 & 1 & -1 \end{pmatrix}\mathbf{U} + \begin{pmatrix} \dfrac{\bar{S}}{\Delta x\,\bar{H}} \\ 0 \\ \vdots \\ 0 \end{pmatrix}. \tag{65}$$

[redacted]

where $\mathbf{P}$ and $\mathbf{\Lambda}$ are defined in the Appendix. More explicitly, the ith element of the state vector $\mathbf{U}$ is given by

$$U_i = \sum_{k=1}^{N-1} P_{ik}\left[\frac{e^{\lambda_k t} - 1}{\lambda_k}\sum_{j=1}^{N} P_{jk}\bar{S}_j + e^{\lambda_k t}\sum_{j=1}^{N} P_{jk}U_{0j}\right]$$
$$+ P_{iN}\left(t\sum_{j=1}^{N} P_{jN}\bar{S}_j + \sum_{j=1}^{N} P_{jN}U_{0j}\right), \qquad i = 1, 2,\ldots, N. \tag{67}$$

The sensitivity equation for the jth parameter K_j has the following form:

$$\boldsymbol{\beta}_j = \frac{\partial \mathbf{U}}{\partial K_j} = \mathbf{P}[\mathbf{Q}\mathbf{P}^T\mathbf{U}_0 + \mathbf{R}\mathbf{P}^T\mathbf{S}], \tag{68}$$

where

$$
\mathbf{Q} = \begin{bmatrix}
g_{11} t e^{\lambda_1 t} & \dfrac{g_{12}}{-\lambda_1 + \lambda_2}(e^{\lambda_2 t} - e^{\lambda_1 t}) & \cdots & \dfrac{g_{1,N-1}}{-\lambda_1 + \lambda_{N-1}}(e^{\lambda_{N-1} t} - e^{\lambda_1 t}) & \dfrac{g_{1N}}{-\lambda_1}(1 - e^{\lambda_1 t}) \\
\dfrac{g_{21}}{-\lambda_2 + \lambda_1}(e^{\lambda_1 t} - e^{\lambda_2 t}) & g_{22} t e^{\lambda_2 t} & \cdots & \dfrac{g_{2,N-1}}{-\lambda_2 + \lambda_{N-1}}(e^{\lambda_{N-1} t} - e^{\lambda_2 t}) & \dfrac{g_{2N}}{-\lambda_2}(1 - e^{\lambda_2 t}) \\
\dfrac{g_{N1}}{\lambda_1}(e^{\lambda_1 t} - 1) & \dfrac{g_{N2}}{\lambda_2}(e^{\lambda_2 t} - 1) & \cdots & & g_{NN} t
\end{bmatrix}, \tag{69}
$$

$$
\mathbf{G} = \mathbf{P}^T \frac{\partial \mathbf{A}}{\partial K_j} \mathbf{P}, \tag{70}
$$

and

$$
\mathbf{R} = \begin{bmatrix}
\dfrac{g_{11}}{\lambda_1}\left[t e^{\lambda_1 t} + \dfrac{1}{\lambda_1}(1 - e^{\lambda_1 t})\right] & \dfrac{g_{12}}{\lambda_2}\left[\dfrac{1}{-\lambda_1 + \lambda_2}(e^{\lambda_2 t} - e^{\lambda_1 t}) + \dfrac{1}{\lambda_1}(1 - e^{\lambda_1 t})\right] & \cdots & \dfrac{-g_{1N}}{\lambda_1}\left[t + \dfrac{1}{\lambda_1} - \dfrac{e^{\lambda_1 t}}{\lambda_1}\right] \\
\dfrac{g_{21}}{\lambda_1}\left[\dfrac{1}{-\lambda_2 + \lambda_1}(e^{\lambda_1 t} - e^{\lambda_2 t}) + \dfrac{1}{\lambda_2}(1 - e^{\lambda_2 t})\right] & \dfrac{g_{22}}{\lambda_2}\left[t e^{\lambda_2 t} + \dfrac{1}{\lambda_2}(1 - e^{\lambda_2 t}\right] & \cdots & \dfrac{-g_{2N}}{\lambda_2}\left[t + \dfrac{1}{\lambda_2} - \dfrac{e^{\lambda_2 t}}{\lambda_2}\right] \\
\cdots & \cdots & \cdots & \cdots \\
\dfrac{g_{N1}}{\lambda_1}\left[\dfrac{(e^{\lambda_1 t} - 1)}{\lambda_1} - t\right] & \dfrac{g_{N2}}{\lambda_2}\left[\dfrac{1}{\lambda_2}(e^{\lambda_2 t} - 1) - t\right] & \cdots & \dfrac{g_{NN} t^2}{2}
\end{bmatrix}. \tag{71}
$$

4.2. *Variational Method*

The adjoint system to Eqs. (57)–(60) is

$$
\frac{\partial \psi}{\partial \tau} = -\bar{K} \frac{\partial^2 \psi}{\partial \xi^2}, \tag{72}
$$

$$
\frac{\partial \psi}{\partial \xi} = 0, \qquad \xi = 0, \bar{H}, \tag{73}
$$

$$
\psi(\xi, T) = \delta(\xi - \xi'). \tag{74}
$$

Letting $x = \xi/\bar{H}$, and $t = \tau/T$, the solutions for $u(x, t)$ and $\psi(x, t)$ are

$$
u(x, t) = \frac{\bar{S}\bar{H}}{3\bar{K}}\left[1 + \frac{3}{2}(x^2 - 2x)\right] + \frac{\bar{S}T}{\bar{H}} t
- \frac{2\bar{S}\bar{H}}{\pi^2 \bar{K}} \sum_{n=1}^{\infty} \frac{1}{n^2} \cos n\pi x \exp\left[-\left(\frac{n\pi}{\bar{H}}\right)^2 \bar{K} T t\right], \tag{75}
$$

$$\psi(x, t) = 1 + 2 \sum_{n=1}^{\infty} \cos n\pi x' \cos n\pi x \exp\left[-\left(\frac{n\pi}{\bar{H}}\right)^2 \bar{K}T(1-t)\right]. \tag{76}$$

The functional derivative sensitivity coefficients (52)–(54) are

$$\begin{aligned}\frac{\delta u(x', t)}{\delta K(x)} &= \frac{4\bar{S}Tt}{\bar{H}\bar{K}} \sum_{n=1}^{\infty} \cos n\pi x' \sin^2 n\pi x\, e^{-(n\pi/\bar{H})^2\bar{K}Tt} \\ &+ \frac{2\bar{S}\bar{H}}{\pi\bar{K}^2}(x-1) \sum_{n=1}^{\infty} \frac{1}{n} \cos n\pi x' \sin n\pi x(1 - e^{-(n\pi/\bar{H})^2\bar{K}Tt}) \\ &+ \frac{4\bar{S}\bar{H}}{\pi^2\bar{K}^2} \sum_{i=1}^{\infty} \sum_{\substack{j=1 \\ (i\neq j)}}^{\infty} \frac{j}{i(j^2-i^2)} \cos j\pi x' \sin i\pi x \sin j\pi x(e^{-(i\pi/\bar{H})^2\bar{K}Tt} - e^{-(j\pi/\bar{H})^2\bar{K}Tt}),\end{aligned} \tag{77}$$

$$\frac{\delta u(x, 1)}{\delta S(t)} = \frac{T}{\bar{H}} + \frac{2T}{\bar{H}} \sum_{n=1}^{\infty} \cos n\pi x\, e^{-(n\pi/\bar{H})^2\bar{K}T(1-t)}, \tag{78}$$

$$\frac{\delta u(x, 1)}{\delta H(t)} = -\frac{\bar{S}T}{\bar{H}} + \frac{2\bar{S}T}{\bar{H}} \sum_{n=1}^{\infty} \cos n\pi x[e^{-(n\pi/\bar{H})^2\bar{K}T(1-t)} - 2e^{-(n\pi/\bar{H})^2\bar{K}T}]. \tag{79}$$

4.3. *Fourier Amplitude Sensitivity Test*

First, the standard finite-difference scheme is applied to (57)–(60) to yield (12). Utilizing the concept of a space-filling search curve, we vary the parameters $\{K_j\}$ according to

$$K_j = \bar{K}_j + \nu_j \sin \omega_j s, \qquad j = 1, 2,..., N, \tag{80}$$

where $0 \leqslant s \leqslant 2\pi$ and $\bar{K}_j$ denotes the nominal value for K_j, and ν_j is a suitable positive constant such that $\bar{K}_j + \nu_j$ and $\bar{K}_j - \nu_j$ are the upper and lower limits, respectively, of the range of K_j. The set of frequencies $\{\omega_j\}$ is incommensurate and a unique frequency ω_j is assigned to jth parameter K_j. Using the parameters generated by (80), we can solve (12) for any time t, the solution of which can be represented as $\mathbf{U}(s)$.

A sensitivity measure utilized in the Fourier amplitude sensitivity test involves the computation of the Fourier spectrum of $\mathbf{U}(s)$. We define the Fourier coefficients

$$A_l^{(i)} = \frac{1}{\pi} \int_0^{2\pi} u_i(s) \sin ls\, ds, \tag{81}$$

$$B_l^{(i)} = \frac{1}{\pi} \int_0^{2\pi} u_i(s) \cos ls\, ds, \tag{82}$$

where the subscript l denotes any frequency. By means of Parseval's theorem, the total variance of the output solution $\mathbf{U}(s)$ can be written as

$$\sigma^2 = \tfrac{1}{2} \sum_{l=1}^{\infty} (A_l^2 + B_l^2), \tag{83}$$

where we have dropped the superscript i for convenience. If the Fourier coefficients (81) and (82) are evaluated for the fundamental frequencies of the transformation (80) or its harmonics, i.e., $l = p\omega_j$, $p = 1, 2, \ldots$, the variance

$$\sigma_{\omega_j}^2 = \tfrac{1}{2} \sum_{p=1}^{\infty} (A_{p\omega_j}^2 + B_{p\omega_j}^2) \tag{84}$$

is the part of the total variance σ^2 that corresponds to the variance of the output solution arising from the jth parameter uncertainty. The ratio of $\sigma_{\omega_j}^2$ to σ^2 is denoted by $S_{\omega_j} = \sigma_{\omega_j}^2/\sigma^2$ and is the partial variance which serves as the sensitivity measure for the Fourier amplitude sensitivity test. We note that S_{ω_j} is a normalized sensitivity measure, allowing ordering of the S_{ω_j} to obtain a ranked list of the parameters with respect to relative importance.

4.4. *Numerical Results*

For numerical simulation the following nominal values are used: $\bar{K} = 1$, $\bar{H} = 1$, $\bar{S} = 1$, and $T = 1$. With this set of nominal values $\bar{K}T/\bar{H}^2 = 1$. For this choice of nominal values the sensitivity coefficients represent both absolute and relative values.

The exact functional derivative sensitivity coefficients $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$ are shown in Fig. 1 for $x = 0$, 0.4, and 1.0. These functional derivatives define the sensitivity of $u(x, 1)$, i.e., the state at time $t' = 1$, with respect to the changes in the system parameters S and H at some past time t. Thus, $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$ indicate, as a function of time, the history of contributions of the changes in the system parameters S and H to the changes in the state $u(x, 1)$. For this reason, we have reversed the time axis in Fig. 1.

Several interesting observations can be made from Fig. 1. At ground level, $x = 0$, the values of $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$ both attain infinite value. This is expected as a result of the boundary condition at the ground level. For $x > 0$, $\delta u(x, 1)/\delta S(t)$ starts from 0 and converges to the final (steady-state) sensitivity of 1. Throughout the entire period of time, $\delta u(x, 1)/\delta S(t) > 0$, indicating that an increase in the surface flux at any past time tends to increase the current concentration. The current concentration is highly sensitive to the past history of the changes in the surface flux. On the other hand, for $x > 0$, the absolute value of $\delta u(x, 1)/\delta H(t)$ attains its maximum value of 2 at time $t = 1$ and decreases to the final sensitivity of 1. This indicates that the current concentration is more sensitive to current changes in the mixing height than past changes. Prior increases in the mixing height tend to decrease the current concentration. On the whole, the sensitivities $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$ have the same order of magnitude for the values of $\bar{K}$, $\bar{S}$, and $\bar{H}$ chosen.

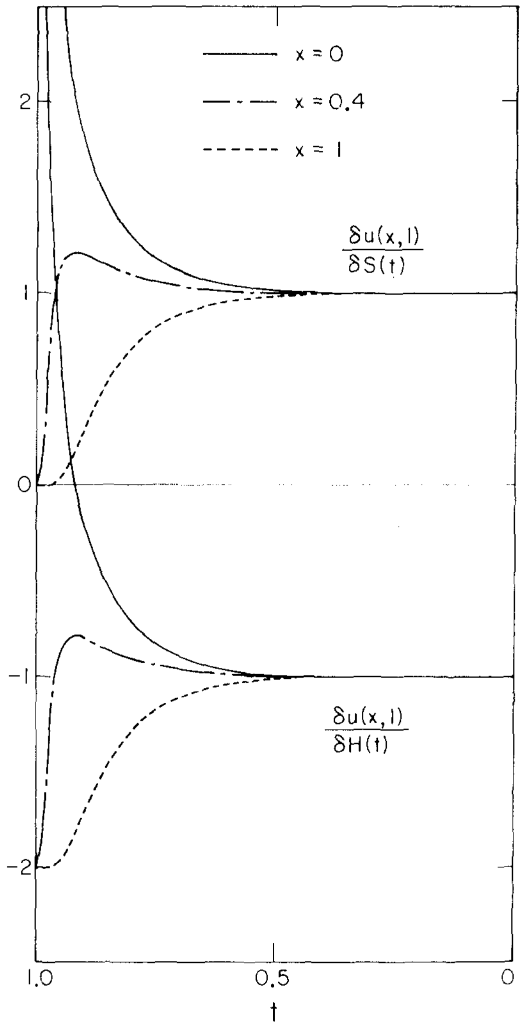

FIG. 1. Functional derivative sensitivity coefficients, $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$, calculated from the exact solution for $x = 0$, 0.4, and 1.0. (Note that the time axis is reversed.)

The spatial distributions of $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$ are plotted in Fig. 2. The sensitivity $\delta u(x, 1)/\delta S(t)$ has its largest value at ground level, $x = 0$, and decreases as the x increases. On the contrary, the sensitivity $\delta u(x, 1)/\delta H(t)$ is largest at the mixing height, $x = 1$, and decreases as x decreases.

The sensitivity $\delta u(x, t)/\delta K(x)$ is plotted in Fig. 3 as a function of time. After a transient period $(0 < t < 0.5)$, $\delta u(x, t)/\delta K(x)$ converges to the final sensitivity. Compared with $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$, the value of $\delta u(x, t)/\delta K(x)$ is smaller by an order of magnitude. The spatial distribution of $\delta u(x', 1)/\delta K(x)$ as a function of x is plotted in Fig. 4 for $x' = 0.1$, 0.2, 0.5, and 0.7. The value of $\delta u(x', 1)/\delta K(x)$

reverses sign at $x = x'$. At some fixed altitude, $x = x'$, the increase in $K(x)$ at lower altitude, $x < x'$, tends to increase the concentration at $x = x'$, while the increase at higher altitude, $x > x'$, tends to decrease the concentration.

From the results obtained above, we may conclude that the surface flux and mixing height are the parameters which influence the concentration most strongly, whereas the effects of changes in the eddy diffusivity are minor.

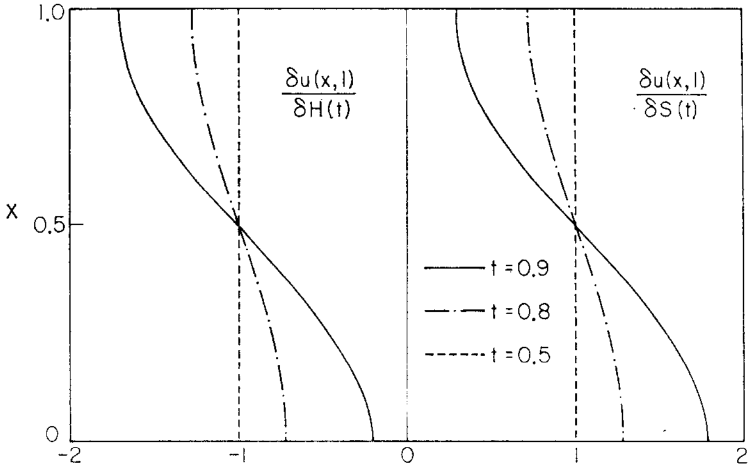

FIG. 2. Functional derivative sensitivity coefficients, $\delta u(x, 1)/\delta S(t)$ and $\delta u(x, 1)/\delta H(t)$, calculated from the exact solution for $t = 0.5$, 0.8, and 0.9.

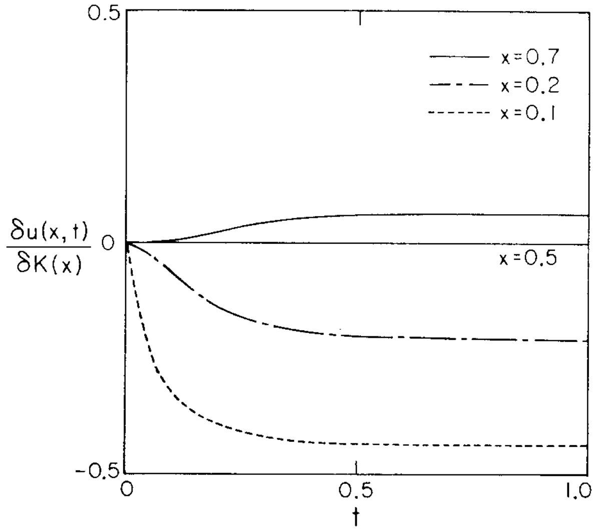

FIG. 3. Functional derivative sensitivity coefficient $\delta u(x, t)/\delta K(x)$ calculated from the exact solution for $x = 0.1$, 0.2, and 0.7.

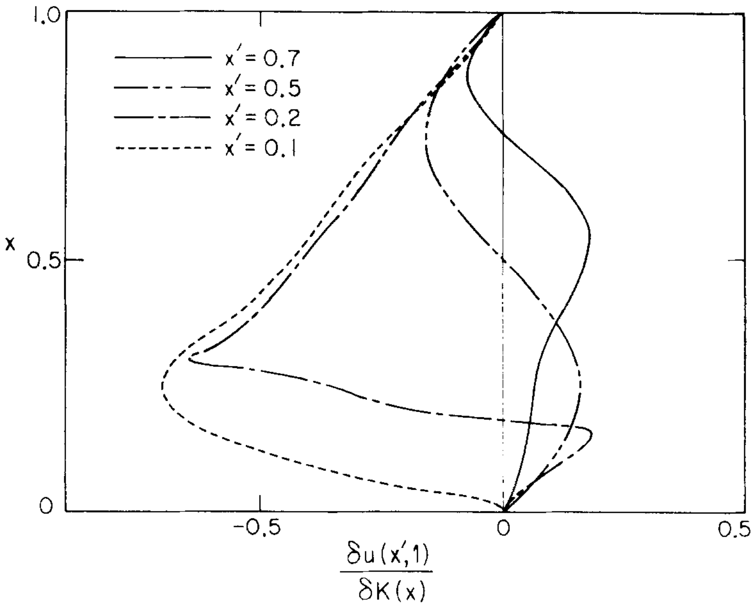

FIG. 4. Functional derivative sensitivity coefficient $\delta u(x', 1)/\delta K(x)$ calculated from the exact solution for $x' = 0.1$, 0.2, 0.5, and 0.7.

Direct and Variational Methods. In order to compare the sensitivity coefficients calculated by the variational and the direct methods, we must establish the relationship between the functional derivatives and the sensitivity coefficients as usually defined. From the definition of the functional derivative, we have the following estimate for $\delta K(x) = \delta K_j = \text{const}(x_j - \frac{1}{2}\Delta x < x \leqslant x_j + \frac{1}{2}\Delta x)$:

$$\frac{\partial U_i(t)}{\partial K_j} \cong \int_{x_j-1/2\Delta x}^{x_j+1/2\Delta x} \frac{\delta u(x_i, t)}{\delta K(x)}\, dx \cong \Delta x \frac{\delta u(x_i, t)}{\delta K(x_j)}, \tag{85}$$

where we have assumed $\delta K(x) = 0$ for the rest of the x-domain. Thus, the functional derivative, $\delta u(x_i, t)/\delta K(x_j)$, and the sensitivity coefficient, $\partial U_i(t)/\partial K_j$, are related as follows

$$\frac{\delta u(x_i, t)}{\delta K(x_j)} \cong \frac{1}{\Delta x} \frac{\partial U_i(t)}{\partial K_j}, \tag{86}$$

where the mesh size Δx is dimensionless.

In the direct approach, we have used the following spatial discretization for the computation: $N = 1$ and $\Delta x = 0.1$.

The sensitivity coefficient, $\partial U_i(t)/\partial K_i$, is shown as a function of time in Fig. 5 for $x = x' = 0.1$, 0.2, 0.5, and 0.7. The spatial profile of $\partial U_i(1)/\partial K_j$ ($j = 1, 2, \ldots, 11$) is shown in Fig. 6 for $x' = 0.1$, 0.2, 0.5, and 0.7. In view of (86), $10(\partial U_i(t)/\partial K_j)$ is shown in the plots. Thus, the curves in Figs. 5 and 6 correspond essentially to those of Figs. 3 and 4. We see that the general trend of $\delta u(x_i, t)/\delta K(x_j)$ agrees quite well

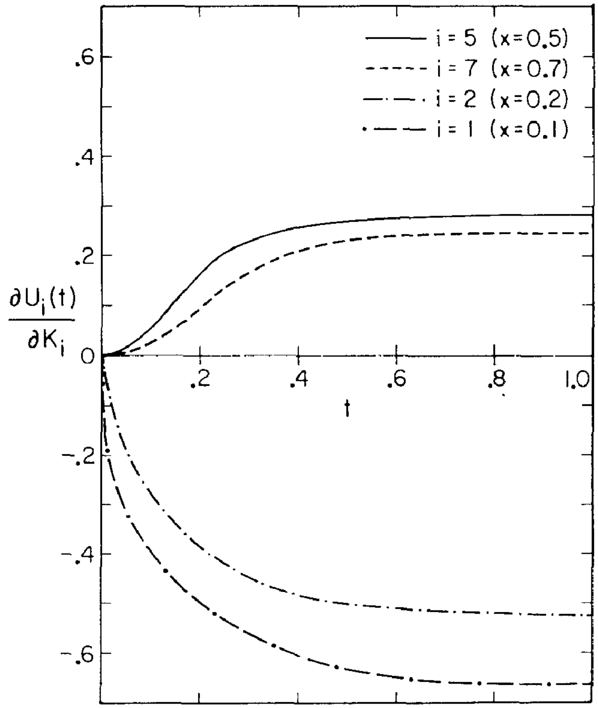

FIG. 5. Sensitivity coefficient $\partial U_i(t)/\partial K_i$ as a function of time for $x = x' = 0.1$, 0.2, 0.5, and 0.7, as calculated by the direct method.

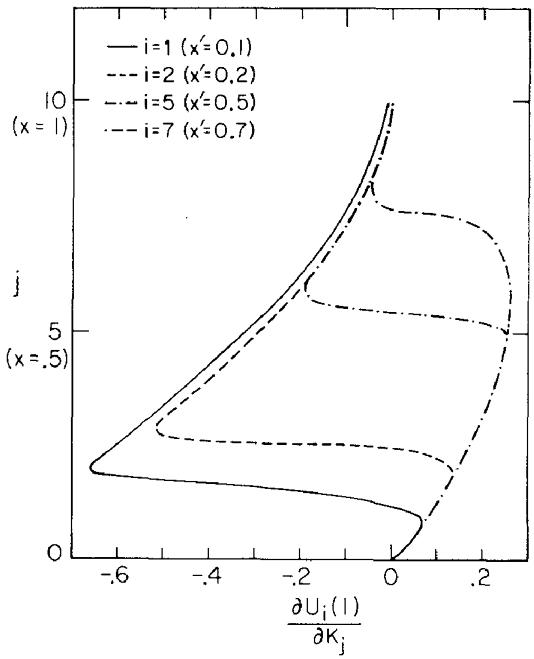

FIG. 6. Sensitivity coefficient $\partial U_i(1)/\partial K_j$ ($j = 1, 2, \ldots, 11$) as a function of x for $x' = 0.1$, 0.2, and 0.7, as calculated by the direct method.

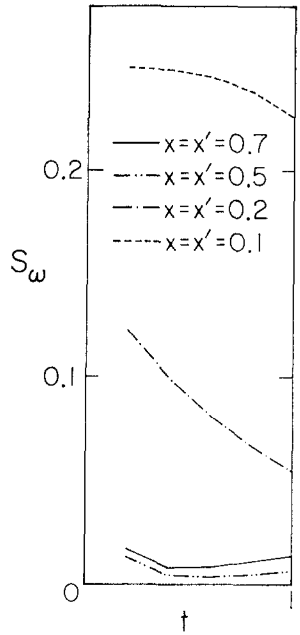

FIG. 7. Partial variance of $u(x', t)$ due to uncertainty in $K(x)$ for $x = x' = 0.1, 0.2, 0.5$, and 0.7, as a function of time, as calculated by the FAST method.

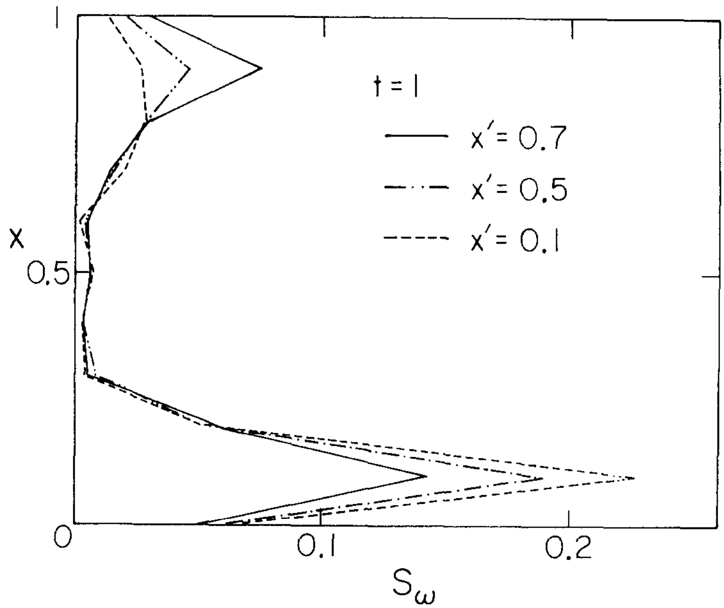

FIG. 8. Partial variance of $u(x', 1)$ due to uncertainty in $K(x)$ for $x' = 0.1, 0.5$, and 0.7, as a function of x, as calculated by the FAST method.

with that of $\partial U_i(t)/\partial K_j$. We repeated the solution using $N = 21$ and $\Delta x = 0.5$. With this refined mesh, it was observed that the sensitivities calculated by the direct and variational methods were closer than for $N = 11$, as expected.

FAST Method. In the application of the FAST method with a finite sample of output solutions, the integrals in (81) and (82) and the summations in (83) and (84) are approximated by finite sums. For $N = 11$, we represent $K(x)$ by 11 independent parameters. For the transformation (80), we used the frequency set $\{\omega_j\} = (41, 67, 105, 145, 177, 199, 219, 229, 235, 243, 247)$ for which the minimal number of evenly spaced dividing points for s, $0 \leqslant s \leqslant 2\pi$, is 974 [6]. In order to keep a realistic profile for the spatial distribution of $\{K_j\}$, the following parameters are used for (80): $\bar{K}_j = 1$ and $\nu_j = 0.1$, $j = 1, 2, \ldots, 11$. Thus, we allow a maximum of 10% variations in K_j. The standard Crank–Nicolson method with $\Delta x = 0.1$ and $\Delta t = 0.05$ is used in the numerical integration of (12).

We carried out the Fourier analysis at $t = 0.2$, 0.4, 0.6, 0.8, and 1. In Fig. 7, the time history of the partial variance of the output function $u(x', t)$ due to the uncertainty in the parameter $K(x)$ is shown for $x = x' = 0.1$, 0.2, 0.5, and 0.7. In Fig. 8, the spatial distribution of the partial variance of the output function $u(x', t)$ due to the uncertainty in $K(x)$ is given as a function of x for $x' = 0.1$, 0.5, and 0.7 at $t = 1$. The concentration is found to be most sensitive to the changes in $K(x)$ near $x = 0$. The concentration is also sensitive to the changes in $K(x)$ near $x = 1$. This indicates that $K(x)$ at the locations where it influences the boundary conditions is most important to a diffusion process. On the whole, the results obtained by the Fourier amplitude sensitivity test agree with those obtained by the variational method and the direct method.

5. Comparison of Approaches

We have explored three approaches to sensitivity analysis of parameters in partial differential equations, the direct method, the variational method, and the FAST method. Any of the three approaches is capable, in principle, of providing the same information concerning the system. A key consideration, therefore, is the relative computational efficiency of the methods.

In the choice of a particular method for the sensitivity analysis, the main considerations are the computational costs incurred and the sensitivity measures obtained. The variational approach can provide a rigorous sensitivity measure that gives a precise interpretation of the results but may require a lengthy computation to solve two sets of partial differential equations, the system and adjoint equations. In principle, these two sets of equations give complete information about the functional derivative sensitivity coefficients for both spatially and temporally varying parameters. The implementation of the direct method requires solution of two sets of ordinary differential equations, the system and the sensitivity equations. Usually, these two sets of equations are coupled and must be solved simultaneously. Thus, storage may become a problem in solving, especially, three-dimensional problems. The Fourier amplitude sensitivity test uses only the system but it requires auxiliary calculations such as the Fourier amplitudes. The solution of the system must be carried out repeatedly, perhaps requiring lengthy computations, depending on the number of parameters tested.

As a basis for comparison of the methods we consider calculation of the sensitivity of the state $u_i(x, t)$ to variations in the constant parameters k_j and the diffusion coefficient $K(x)$. As before, we assume n state variables and N discrete spatial points at which the states are approximated for the direct method. For comparison of the direct and variational methods we assume that $\delta u_i(x', t)/\delta K(x)$ from the variational method is to be evaluated at N values of both x and x'.

In the direct approach the sensitivity equations are solved simultaneously with the system equations using nominal parameter values. Thus, in the cases of k_j, $j = 1, 2,..., m$ and K_i, $i = 1, 2,..., N$, the number of ordinary differential equations to be integrated is $(m + 1)\, nN$, comprising nN system equations and mnN sensitivity equations, and $(N + 1)\, nN$, comprising nN system equations and nN^2 sensitivity equations, respectively. To compare the variational approach with the direct approach we assume that the system and adjoint equations are each approximated by N ordinary differential equations. Then for constant parameters k_j, the variational approach requires nN system equations and n^2N^2 adjoint equations. The number of adjoint equations is governed by the terminal condition (26) (assuming x' is evaluated at N discrete points). The calculation of the sensitivity coefficient by (30) requires evaluation of the n double integrals for each of m parameters at N values of x. For parameter $K(x)$, nN system and n^2N^2 adjoint equations must be solved, and the n integrals in (28) must be evaluated.

APPENDIX: Calculation of Sensitivity Coefficients by the Direct Method

The matrix exponential function required in the calculation of sensitivity coefficients by the direct method can be computed by various techniques. The properties of $\mathbf{A(K)}$ depend on the boundary conditions of the problem. Generally $\mathbf{A(K)}$ is nonsingular for Dirichlet boundary conditions or mixed boundary conditions. For Neumann-type boundary conditions $\mathbf{A(K)}$ becomes singular.

If $\mathbf{A(K)}$ is nonsingular, then $\mathbf{A(K)}$ has N distinct nonzero eigenvalues $\lambda_1, \lambda_2,..., \lambda_N$, and thus N linearly independent eigenvectors $\mathbf{p}_1, \mathbf{p}_2,..., \mathbf{p}_N$ such that

$$\mathbf{PA(K)\, P}^{-1} = \mathbf{\Lambda}, \tag{A1}$$

where

$$\mathbf{\Lambda} = \begin{pmatrix} \lambda_1 & & & 0 \\ & \lambda_2 & & \\ & & \ddots & \\ 0 & & & \lambda_N \end{pmatrix} \tag{A2}$$

and

$$e^{\mathbf{A(K)}t} = \mathbf{P}e^{\Lambda t}\mathbf{P}^{-1} \tag{A3}$$

and where the nonsigular matrix $\mathbf{P} = [\mathbf{p}_1, \mathbf{p}_2, ..., \mathbf{p}_N]$; $\mathbf{p}_i$ is an N-vector. In case of a singular $\mathbf{A(K)}$, one of the eigenvalue, say, λ_1, becomes zero but still a decomposition such as the one defined by (A1) holds. One can directly evaluate $\mathbf{U}(t)$ for a given $\mathbf{S}(t)$. For example, let us assume that $\mathbf{S}(t) = \mathbf{q} = \text{const}$. If $\mathbf{A(K)}$ is nonsingular, from (13),

$$\mathbf{U}(t) = \mathbf{P}e^{\Lambda t}\mathbf{P}^{-1}\mathbf{U}_0 + \mathbf{P}e^{\Lambda t}\mathbf{F}\mathbf{P}^{-1}\mathbf{q}, \tag{A4}$$

where

$$\mathbf{F} = \int_0^t e^{-\Lambda\tau}\,d\tau = \begin{bmatrix} \frac{1}{\lambda_1}(1 - e^{-\lambda_1 t}) & 0 & & \cdots & 0 \\ 0 & 0 & \frac{1}{\lambda_i}(1 - e^{-\lambda_i t}) & 0 & 0 \\ & & & \ddots & \\ 0 & & \cdots & & \frac{1}{\lambda_N}(1 - e^{-\lambda_N t}) \end{bmatrix}. \tag{A5}$$

Equation (A4) can be substituted into (17) to give

$$\boldsymbol{\beta}_j(t) = \mathbf{P}e^{\Lambda t}\left[\int_0^t e^{-\Lambda\tau}\mathbf{G}e^{\Lambda\tau}\mathbf{c}\,d\tau + \int_0^t e^{-\Lambda\tau}\mathbf{G}e^{\Lambda\tau}\int_0^\tau e^{-\Lambda s}\mathbf{d}\,ds\right] \tag{A6}$$

where

$$\mathbf{G} = \mathbf{P}^{-1}\frac{\partial\mathbf{A(K)}}{\partial K_j}\mathbf{P} \tag{A7}$$

is a time-independent constant matrix. The coefficient vectors $\mathbf{c}$ and $\mathbf{d}$ are obtained from the equations $\mathbf{Pc} = \mathbf{U}_0$ and $\mathbf{Pd} = \mathbf{q}$.

The integration in (A7) is carried out analytically and, consequently, a series solution to each sensitivity vector $\boldsymbol{\beta}_j$ is obtained in terms of eigenvalues and eigenvectors (e.g., see the example problem).

The symmetry of $\mathbf{A(K)}$ is of a great advantage in the computations. $\mathbf{A(K)}$ can be made symmetric by imposing an equal-size grid net on the spatial domain. When $\mathbf{A(K)}$ is symmetric, we have $\mathbf{P}^{-1} = \mathbf{P}^T$.

In cases where $\mathbf{A(K)}$ becomes singular due to flux boundary conditions and one desires to use a different technique than decomposition the following procedure is recommended.

Let $\mathbf{A(K)}$ be a singular matrix, which implies that one of the eigenvalues, say, λ_0, is equal to 0. Then

$$\mathbf{PA(K)P}^{-1} = \begin{pmatrix} \lambda_0 & 0 & \cdots & 0 \\ 0 & & & \\ \vdots & & \mathbf{B} & \\ 0 & & & \end{pmatrix}; \tag{A8}$$

Using this property we obtain

$$e^{-\mathbf{A}(\mathbf{K})\tau} = \mathbf{P}^{-1} e^{-\mathbf{P}\mathbf{A}(\mathbf{K})\mathbf{P}^{-1}} \mathbf{P} = \mathbf{P}^{-1} \begin{pmatrix} 1 & 0 & \cdots & 0 \\ 0 & & & \\ \vdots & & e^{\mathbf{B}\tau} & \\ 0 & & & \end{pmatrix} \mathbf{P}, \tag{A9}$$

where $\mathbf{B}$ is an $(N-1) \times (N-1)$ full matrix, and $\mathbf{P}^{-1}$ is an orthogonal similarity matrix, each element in the first column of which is unity. The matrix $\mathbf{P}^{-1}$ can be constructed by Gram–Schmidt orthogonalization or Householder's method.

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