COMPARING METHODS FOR MODELLING DISPERSION IN FLOWING MEDIA

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ABSTRACT. There are several available methods for the modelling of dispersion in flowing media, including analytical represeentations of the diffusion equation, numerical representations of the diffusion equation and compartmental models involving the use of lumped parameter ordinary differential equations. A technique is presented for comparing solutions generated by these different methods. By equating the low order moments of the first-passage time of the diffusing substance in the different methods, relationships are obtained between the parameters in the different models. The relationships are shown to be a good indication that the solutions generated by the different methods are indeed closely similar.

1. *Introduction.*

There are a number of different methods available for the modelling of dispersion in flowing media. The standard approach is to represent the movement and spreading of the flow or of a tracer by a diffusion equation. Solving the diffusion equation analytically is occasionally possible (Crank, 1956; Carslaw and Jaeger, 1960), but usually a numerical solution is required. Most commonly used methods of solution are finite differences (see Richtmyer, 1957; Forsythe and Wasow, 1960), variational methods (Price et al., 1968; Guyman et al., 1970), and Monte Carlo methods (Haji-Sheikh and Sparrow, 1967; Sklarew et al., 1972). A good review of these and many other possible approaches is given by Boley (1972).

Another approach to modelling dispersion in flowing media involves the use of multiple lumped parameter ordinary differential equations, each representative of behaviour in a fixed spatial compartment or cell of the flowing media (Buffham and Gibilaro, 1970; Whitehead and Young, 1975); once again these equations are usually solved numerically.

The existence of two apparently unrelated analytical formulations for modelling dispersion - the diffusion equation and the lumped parameter model - as well as numerical formulations which are different from the analytical formulations in significant ways, provides the motivation for

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the determination of relationships between parameters in some of the different models for dispersion in flowing media. The following approach will be used. For each model considered, the low order moments of the first-passage time of the tracer will be determined at a particular place downstream from a pulse input.* By equating corresponding low order moments for solutions generated by the different methods, relationships between the parameters used in the different models often can be obtained. If the distribution of first-passage times generated by the dispersion process is characterised by its low order moments, then solutions obtained using different methods with appropriately corresponding parameters will be closely similar.

Not all numerical methods are tested and compared here. For some of the more complex models, the analysis would be extremely difficult. The rationale for limiting the analysis here is twofold. First, the simple models used give an indication of the general character of the relationships between parameters that may be obtained. Second, by demonstrating the comparison procedure, more elaborate models can be analysed when the need arises.

2. *Moments of the Analytical Solution.*

Consider the concentration $c(x,t)$ of a diffusing substance in one dimension, as a function of the distance x and time t. In differential form the generalised diffusion equation may be written

$$\frac{\partial c}{\partial t} = \sum_{i=1}^{\infty} \frac{(-1)^i}{i!} M_i \frac{\partial^i c}{\partial x^i} . \tag{1}$$

M_i describes the change in the i^{th} order spatial moment of the solution to (1). That is, given an impulse source at $x = 0$, $t = 0$, the i^{th} spatial moment of the solution to (1) at time t is $M_i t$. Only an impulse source is considered here, and there are no spatial or temporal variations in the M_i.

In the most common cases in which the diffusion equation is used, only M_1 and M_2 are nonzero. In more conventional notation, $M_1 \equiv u$, the mean velocity of the medium, and $\frac{1}{2}M_2 \equiv D$, the diffusion coefficient, so that (1) becomes

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - u \frac{\partial c}{\partial x} . \tag{2}$$

The object here is to calculate the moments of the first-passage time at position $x = x_0$ from a point source at $x = 0$. Multiplying each side of (1) by t^n and integrating over all time t, and noting that

$$\mu'_n = \int_0^{\infty} c(x,t)\, t^n \, dt \tag{3}$$

is the n^{th} non-central moment at position x, (1) becomes

$$-n\, \mu'_{n-1} = \sum_{i=1}^{\infty} \frac{(-1)^i}{i!} M_i \frac{\partial^i \mu'_n}{\partial x^i} . \tag{4}$$

Given μ'_n as a known function of x, (4) can be used to calculate all higher moments. For an absorbing barrier at any point x,

$$\mu'_0 = 1 . \tag{5}$$

In other words, all diffusing particles are at position x exactly once (i.e. when they are absorbed). Using (4) and (5), noting that by the initial condition $\mu'_n = \delta_{n0}$ for $x = 0$, writing $M_1 = u$ and $\frac{1}{2}M_2 = D$, and converting to central moments, it is easy to determine that

$$\mu'_1 = \frac{x_0}{u} , \tag{6}$$

$$\mu_2 = \frac{2Dx_0}{u^3} , \tag{7}$$

$$\mu_3 = \frac{x_0}{u^5} (12D^2 - uM_3) . \tag{8}$$

* The first-passage time for an infinitesimal element of the diffusing substance is the time it takes for that element first to reach a particular place. The first-passage time for the tracer as a whole is the average of the first-passage times for the infinitesimal elements composing it (Feller, 1968). The first-passage time can easily be obtained from the solution to the diffusion equation in the presence of an absorbing barrier; since such solutions exist, first-passage times are obviously compatible with the property of the simple diffusion equation - with no boundaries - which indicates that a signal is transmitted to infinity instantaneously.

Note that it is not necessary to know the analytical solution to (1) in order to obtain expressions (6)-(8).

3. Moments of a Numerical Solution.

Numerical solutions to the diffusion equation (1) may not have the same moments as the analytical solution. This is due to various inaccuracies, such as spurious diffusion which results from finite difference approximations of the term in $\partial c/\partial x$. Therefore it is useful to calculate the moments of a numerical solution to see if they are the same as those of the analytical solution. If they are not, it may be possible to adjust certain parameters in the numerical algorithm to make the corresponding moments equal, and thereby improve the accuracy of the numerical solution.

Here the moments for only one general type of numerical solution are calculated in terms of the numerical parameters. Divide the x-axis into a set of boxes, with box $i = 0$ centred at $x = 0$ and box $i = N$ centred at $x = x_0$. The length of each box then is $\Delta x = x_0/N$. In a time interval of duration Δt, let a fraction p of the mass in box i move to box $i-1$, a fraction q of the mass in box i move to box $i+1$, and the remaining fraction $r = 1-p-q$ of the mass remain in box i. The fractions p, q, and r thus are transition probabilities for the mass in box i for the time interval $(t, t+\Delta t)$.

Depending on the values of p, q, and r, this numerical representation is equivalent to some variational method representations of the diffusion equation, and to explicit finite difference representations of the diffusion equation. This can be seen readily by writing these representations in transition probability form. For example, for the diffusion equation (2), a standard explicit finite difference approximation using a centred difference for the term in $\partial c/\partial x$ gives

$$p = \frac{D\Delta t}{(\Delta x)^2} - \frac{1}{2}\frac{u\Delta t}{\Delta x} , \tag{9}$$

$$q = \frac{D\Delta t}{(\Delta x)^2} + \frac{1}{2}\frac{u\Delta t}{\Delta x} , \tag{10}$$

$$r = 1 - 2\frac{D\Delta t}{(\Delta x)^2} . \tag{11}$$

The moments of the numerical solution resulting from the transition probability representation are the moments of the first-passage time at box N for a unit mass starting at box 0. These moments may be calculated via the generating function for the arrival time at box N. Using the technique in Feller (1968, pp. 349ff), one can determine that the generating function for the first-passage time of the random walk is

$$U(s) = \left(\frac{1-rs-(1-2rs-(4pq-r^2)s^2)^{\frac{1}{2}}}{2ps} \right)^N . \tag{12}$$

The un-normalised and non-central moments of the first-passage time are given in terms of derivatives of $U(s)$, as follows.

$$\left(U^{(n)}(1) \equiv \frac{d^n U}{ds^n}\Big|_{s=1} \right):$$

$$\mu_0 = U(1) , \tag{13}$$

$$\mu_1' = U^{(1)}(1) , \tag{14}$$

$$\mu_2' = U^{(2)}(1) + U^{(1)}(1) , \tag{15}$$

$$\mu_3' = U^{(3)}(1) + 3U^{(2)}(1) + U^{(1)}(1) . \tag{16}$$

Evaluating (13) - (16) and converting to central moments, it is found of course that $\mu_0 = 1$ (all diffusing particles are absorbed exactly once at x_0) and that

$$\mu_1' = \frac{N}{q-p} , \tag{17}$$

$$\mu_2 = N\frac{B}{(q-p)^3} , \tag{18}$$

$$\mu_3 = N\left(\frac{3B^2}{(q-p)^5} + \frac{2B-r}{(q-p)^3} \right) , \tag{19}$$

where

$$B = q+p - (q-p)^2 . \tag{20}$$

Using these expressions, one can determine the moments of the solution generated by a number of simple numerical algorithms, for example from explicit finite difference formulations. These moments can then be compared to the moments (6) to (8) of the analytical solution. (The sets of expressions (6) to (8) and (17) to (19) are identical when Δt and Δx approach 0, at least for any numerical approximation - such as (9) to (11) - consistent with the partial differential equation being modelled.)

Alternatively, one may equate corresponding moments of the analytical and numerical solutions, and determine expressions for p, q, and r. Equating corresponding moments μ_1' from (6) and (17) and μ_2 from (7) and (18) and solving for p and q, it is found that

$$q = \frac{D\Delta t}{(\Delta x)^2} + \tfrac{1}{2}\frac{u\Delta t}{\Delta x}\left(1 + \frac{u\Delta t}{\Delta x}\right), \tag{21}$$

$$p = \frac{D\Delta t}{(\Delta x)^2} + \tfrac{1}{2}\frac{u\Delta t}{\Delta x}\left(-1 + \frac{u\Delta t}{\Delta x}\right), \tag{22}$$

$$r = 1 - \frac{2D\Delta t}{(\Delta x)^2} - \left(\frac{u\Delta t}{\Delta x}\right)^2 . \tag{23}$$

The solution generated by the numerical algorithm with transition probabilities p, q, and r determined as above will have the same moments μ_1' and μ_2 as the analytical solution. Since reasonably well-behaved distributions are well characterised by their low order moments, it is expected that a numerical solution generated with this procedure will be more accurate than other similar numerical solutions. This expectation is tested and confirmed in the next section.

4. *Comparison of Numerical Solutions.*

To illustrate how different numerical algorithms may be compared by inspecting the moments of the numerical solutions, presented here are results of some tests of selected representations. First, the representations are described, and then the particular tests and the results obtained. For illustrative purposes and convenience, the analytical solution is assumed to be a solution to the diffusion equation (2).

Three numerical representations are tested which are based on the differential form of the diffusion equation, and which utilise finite differences. Grid points separated by the distance Δx are assigned to the x-axis, with one point at the origin. The change in the concentration at each grid point is calculated with each application of the specified algorithm.

First write the diffusion equation (2) in the form

$$\frac{\partial c}{\partial t} = F(t,c) . \tag{24}$$

Using simple finite difference approximations for the spatial derivatives in (21), the operator $F(t,c)$ becomes

$$F(t,c) = \frac{D}{(\Delta x)^2}\left(c(x+\Delta x,t) - 2c(x,t) + c(x-\Delta x,t)\right) - \frac{u}{2\Delta x}\left(c(x+\Delta x,t) - c(x-\Delta x,t)\right) . \tag{25}$$

Algorithms I, II, and III result from the following approximations for $\partial c/\partial t$ in (24):

I. Point-slope formula:

$$c(t+\Delta t) = c(t) + \Delta t\, F(t,c(t)) . \tag{26}$$

II. Crank-Nicolson implicit method formula:

$$c(t+\Delta t) = c(t) + \tfrac{1}{2}\Delta t\,\left(F(t,c(t)) + F(t+\Delta t,\, c(t+\Delta t))\right) . \tag{27}$$

III. Fully implicit method formula:

$$c(t+\Delta t) = c(t) + \Delta t(F(t+\Delta t,\, c(t+\Delta t))) . \tag{28}$$

Note that Algorithm I may be cast into an integral representation of the diffusion equation, with transition probabilities given by (9) - (11).

By specifying the following transition probabilities, an algorithm based on the integral form of the diffusion equation is obtained:

(IV)

$$p = \frac{D\Delta t}{(\Delta x)^2} + \max\left(0, -\frac{u\Delta t}{\Delta x}\right) . \tag{29}$$

(30) $$q = \frac{D\Delta t}{(\Delta x)^2} + \max\left(0, \frac{u\Delta t}{\Delta x}\right),$$

(31) $$r = 1 - \frac{2D\Delta t}{(\Delta x)^2} - \left|\frac{u\Delta t}{\Delta x}\right| .$$

This algorithm guarantees that no negative masses are produced due to subtracting the advection term $u\Delta t/\Delta x$. It is also equivalent to the use of an uncentred upstream difference scheme to approximate $\partial c/\partial x$, with the point-slope approximation for the time derivative $\partial c/\partial t$.

Algorithm V is one that produces the same zeroth, first, and second moments as the analytical solution at the sample point specified. The equations from which the transition probabilities may be obtained are given above in (21) - (23). These transition probabilities are identical to those obtained by Martin (1975) by equating the changes in the low-order *spatial* moments of the solution $c(x,t)$, such changes being obtained from the diffusion equation and the numerical algorithm. Equations (21) - (23) also correspond to a numerical algorithm obtained by using the Lax-Wendroff approximation for advection and the simple centred difference scheme for diffusion (Runca, 1976).

The final algorithm is a simple one based on a lumped parameter compartmental model.

To compare the numerical solutions, the low-order moments of the first-passage time up to μ_3 are calculated from a numerical simulation. (In cases where the moments could be calculated analytically by the technique in Section 2 above - all algorithms except II and III - these agreed with the moments obtained by numerical simulation.) Since the low order moments do not completely characterise a distribution*, it is revealing (and convincing) to provide a measure of goodness of fit of the analytical and numerical solutions in the time domain. A simple and straightforward measure is used here: the sum of the square of the differences between the analytical and numerical solutions, calculated at the final box or grid point N. This sum of squares is denoted L.

* Low order moments usually will characterise well a non-bumpy distribution; two separated delta functions, for example, would not be well characterised. However, the numerical procedures tested here will be applicable as long as the distribution is reasonably smooth *within* boxes.

Table 1

Values of the moments of the first-passage time and values of a measure of goodness of fit for several selected solutions to a dispersion problem with

$$\frac{D\Delta t}{(\Delta x)^2} = 0.2, \quad \frac{u\Delta t}{\Delta x} = 0.2, \quad N = 10.$$

	μ_0	μ_1'	μ_2	μ_3	L
Analytical solution	1.000	50.0	500.	15000.	-
Numerical solutions					
I	1.000	50.0	450.	12300.	.000041
II	1.000	50.5	499.8	13720.	.000032
III	1.000	51.0	550.	15300.	.00011
IV	1.000	50.0	700.	30300.	.00039
V	1.000	50.0	500.	15300.	.0000015
Lumped parameter model solution	1.000	50.0	500.	10000.	.00023

Table 2

Values of the moments of the first-passage time and values of a measure of goodness of fit for several selected solutions to a dispersion problem with

$$\frac{D\Delta t}{(\Delta x)^2} = 0.2, \quad \frac{u\Delta t}{\Delta x} = 0.4, \quad N = 10.$$

	μ_0	μ_1'	μ_2	μ_3	L
Analytical solution	1.000	25.0	62.5	469.	-
Numerical solutions					
I	1.000	25.0	37.5	150.	.0021
II	1.000	25.5	62.2	300.	.00051
III	1.000	26.0	87.5	525.	.0016
IV	1.000	25.0	100.0	1370.	.0018
V	1.000	25.0	62.5	525.	.000049
Lumped parameter model solution	1.000	25.0	62.5	312.	.00025

Table 3

Values of the moments of the first-passage time and values of a measure of goodness of fit for several selected solutions to a dispersion problem with

$$\frac{d\Delta t}{(\Delta x)^2} = 0.2, \quad \frac{u\Delta t}{\Delta x} = 0.4, \quad N = 50.$$

	μ_0	μ_1'	μ_2	μ_3	L
Analytical solution	1.000	125.	312.5	2340.	-
Numerical solutions					
I	1.000	125.	188.	750.	.00089
II	1.000	125.5	312.2	1500.	.000044
III	1.000	126.	438.	2620.	.00039
IV	1.000	125.	500.	6840	.00062
V	1.000	125.	312.5	2620	.0000025
Lumped parameter model solution	1.000	125.	312.5	1560.	.000018

Sample results are presented in Tables 1-3 for different values of D, u, and N. It is readily seen that Algorithm V which gives the same low order moments as the analytical solution, gives the highest accuracy of the set of algorithms tested.

There are several other ways in which numerical representations may be evaluated: ease of generation and computation, stability, non-negativity of concentrations, and mass conservation. These criteria, and the degree to which the different algorithms satisfy them, are discussed in Martin (1975). For example, algorithms IV and V are stable if p,q, and r are non-negative and have a unit sum.

5. *Moments of Lumped Parameter Compartmental Solutions.*

Buffham and Gibilaro (1970) present a unified compartmental "time-delay" model for dispersion in flowing media. This unified model contains five parameters, and for special choices of these reduces to a number of simpler models, referred to by Buffham and Gibilaro. Therefore it is appropriate to compare the moments calculated by Buffham and Gibilaro for the solution generated by their model with the moments

(6) - (8) of a diffusion equation solution. But first a description of their unified "time delay" model is called for.

The cell form of the model is presented in Figure 1.

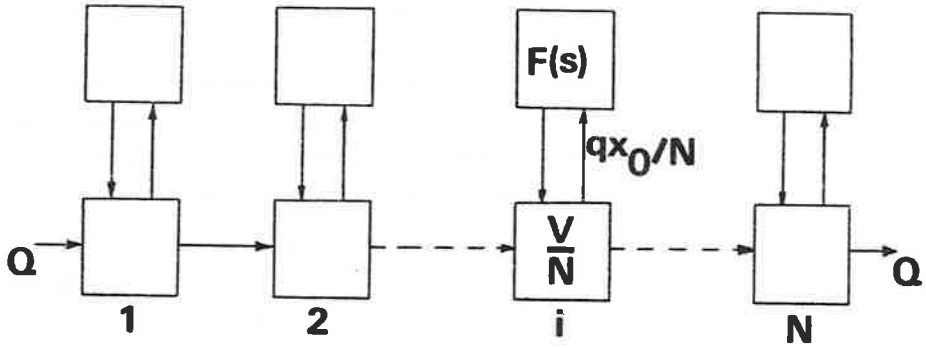

Figure 1. Compartmental model.

The main flow path is represented by N well-mixed cells. Lateral flow is at a rate of qx_0/N per box, and the distribution of delay times in the lateral zones is characterised by the transfer function F(s), where s is the Laplace variable. The throughput flow rate is Q and the total volume of the main flow path is V.

As a most general delay time distribution Buffham and Gibilaro suggest the gamma distribution with transfer function

$$F(s) = \left(\frac{t_D s}{m} + 1\right)^{-m} . \tag{32}$$

They show that for this distribution the moments of the arrival time are

$$\mu_1' = t_0 + \alpha x_0 t_D, \tag{33}$$

$$\mu_2 = \alpha x_0 \left(1 + \frac{1}{m}\right) t_D^2 + \frac{1}{N}(t_0 + \alpha x_0 t_D)^2 , \tag{34}$$

$$\mu_3 = \alpha x_0 \left(1 + \frac{1}{m}\right)\left(1 + \frac{2}{m}\right) t_D^3 + \frac{3}{N}\left(1 + \frac{1}{m}\right) \alpha x_0 (t_0 + \alpha x_0 t_D) t_D^2 + \frac{2}{N^2}(t_0 + \alpha x_0 t_D)^3 , \tag{35}$$

where $t_0 = V/Q$ and $\alpha = q/Q$.

Given a particular model of this type, with parameters t_0, α, N, t_D, and m, it is easy to calculate the parameters M_1, M_2,... of the diffusion equation whose solution has the same impulse response, by equating the corresponding moments. Working in the other direction is not so easy. Because Buffham and Gibilaro's model has 5 parameters, moments up to the fifth order would need to be determined in order to specify all of them. Therefore it is useful to consider a special case of their model, obtained by eliminating the lateral zones in Figure 1. This model is equivalent to solving a set of N input-output mass conservation equations of the form

$$\frac{V}{N}\frac{dc_i}{dt} = -Qc_i + Qc_{i-1}, \tag{36}$$

where c_i is the output concentration, i.e. the concentration in a given cell, and c_{i-1} is the input concentration, i.e. the concentration in the cell upstream from the given cell. This model is equivalent to a set of continuous stirred tank reactors (CSTRs) (see Whitehead and Young, 1975). Each input-output equation of the form (36) does not explicitly include mixing. Dispersion in the solution arises from the exponentially time delayed distribution c_i that results from each input c_{i-1}.

The moments of the arrival time at position x_0 of the solution to N equations of the form (36) are simply determined from (33) - (35) by setting $\alpha = 0$. This gives

$$\mu_1' = t_0 , \tag{37}$$

$$\mu_2 = \frac{t_0^2}{N} , \tag{38}$$

$$\mu_3 = \frac{2t_0^3}{N^2} . \tag{39}$$

Comparing these to (6) - (8) it is found that the solution to (36) has the same low order moments as a solution to the diffusion equation (1) which has parameters

$$u = \frac{x_0}{t_0} , \tag{40}$$

$$D = \frac{x_0^2}{2t_0N} , \tag{41}$$

$$M_3 = \frac{x_0^3}{t_0N^2} . \tag{42}$$

It is also simple to determine the parameters for a CSTR model which will result in a given mean velocity and dispersion. From (40) and (41),

$$t_0 = \frac{x_0}{u} , \tag{43}$$

$$N = \frac{ux_0}{2D} . \tag{44}$$

In many problems it is useful to use the CSTR model, since other processes may be simply included and certain types of analysis easily made. (43) and (44) tell one the values of t_0 and N that should be used if one wishes to model a certain mean flow and dispersion. Similarly, other such compartmental models may be superior to diffusion formulations in certain circumstances (Buffham et al.,1970).

The lumped parameter model tested in the previous section is a CSTR model with parameter t_0 and N taken from (43) and (44). The agreement, indicated by L, between the solution to the diffusion equation and the lumped parameter model solution is good, indicating that relationships such as (40) - (42) or (43) - (44) will usually imply a close similarity between the solutions generated by the different models. Lumped parameter compartmental models can be considered to be independent mathematical models for the physical processes involved in flowing media; they are not necessarily intended to be approximations to diffusion equations. Therefore the L values in the previous section might just as well represent the divergence of the solution to the diffusion equation from the lumped parameter model solution.

Although the agreement is good between the solution to the diffusion equation and the lumped parameter model solution with corresponding parameters, this will not necessarily be true for numerical solutions to the lumped parameter model; for such numerical solutions, new parameter relationships might be obtained via the

technique in Section 3. The basic point here is that whenever a different method for modelling diffusion is introduced, whether via a different analytical model or a numerical solution, it is valuable to look at the relationships between the models in terms of the low order moments of the solutions which they generate. Such relationships can be used to translate one analytical model into another, to investigate the accuracy of a numerical approximation, and in some cases to generate an algorithm specially suited for the problem at hand.

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