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ON COMPARTMENTAL MODELLING OF MIXING PHENOMENA

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Abstract: In this paper the problem of modelling partial mixing phenomena, mostly relevant in environmental and reactors modelling practice, is considered. The ultimate modelling goal is to find *identifiable, finite-dimensional state-space models*, which are *physically interpretable*, *realisable* and which describe partial mixing. Hence, realization theory will be linked to prior physical systems knowledge to answer the question which mixing models are good candidates in environmental/reactor systems modelling. The starting point is compartmental systems modelling with backflows. It appears however that only a limited set of low-dimensional structures is identifiable. From the real world example given in this paper it appears that an appropriate, physically interpretable and realisable model within this class of models cannot be easily found. *Copyright © 2002 IFAC*

Keywords: mixing, linear systems, identifiability, physically-based model structures

1. INTRODUCTION

Mixing plays a key role when substances are transported by air or water flow. In the modelling of the hydraulics in reactor systems two approaches prevail. The first one is based on the assumption of an ideally mixed liquid leading to a CSTR model and in the second approach one assumes no mixing at all, so that a pure plug flow results. Clearly, in practice most often *partial mixing* will appear.

Due to the increasing computational possibilities and the availability of simulation packages a growing interest in advanced modelling of the reactor's hydraulics, in addition to the biochemistry, can be observed. A physically-based modelling approach, in which partial mixing plays a key role, leads to a convection-diffusion (CD) equation (see e.g. Makinia and Wells, 2000). For implementation a common approach is to discretise this so-called infinite-dimensional system description using an appropriate numerical scheme. In general, this discretisation step leads to a set of ordinary differential/difference equations of high dimension, which may cause computational problems. Alternatively, in the past active and dead zone (ADZ) models or finite stage models (see Ferrara and Harleman, 1981; Beer and Young, 1983) have also been proposed. Recently, an approach using the results of CFD (computational fluid dynamics) analysis, providing static flow fields, has been developed. Subsequently, on the basis of these flow fields high-dimensional compartmental models with backflows, parallel and/or circular flows are derived (see e.g. Alex *et al*, 1998; Hunze *et al*, 2000). Again a high-dimensional system results. The main problem associated with these conceptual modelling approaches is how to choose the transfer coefficients between and the volumes of the different compartments. If I/O (input/output) data becomes available one can try to estimate these coefficients from the data. But then the structural identifiability property of the postulated model, including output equations relating the sensor outputs to the states, is crucial!

The objective of this paper is now to analyse the system theoretic properties of some very elementary compartmental models with different flow paths that

are candidate models for describing the partial mixing phenomenon. As far as the authors are aware no such study linking realization theory and prior physical/hydraulic systems knowledge has been performed in the past. And, thus the question which are good candidate models for describing partial mixing cannot be answered simply. This study is therefore relevant to the hydraulic modelling of reactors and environmental systems. As mentioned before, the emphasis will be on the identifiability properties of these *a priori* postulated models (see also Young and Lees (1993) for a discussion on this topic that starts from the experimental data). For simplicity, we restrict ourselves to non-reactive reactor systems, so that linear models result. From that point of view extension to models with zeroth- or first-order reaction terms is straightforward (see e.g. Dötsch and van den Hof (1996) for the non-linear case). Our approach is as follows: (i) list appropriate low-dimensional system configurations allowing input/output splitting, circular and parallel flow patterns, (ii) provide a state-space model of the system configuration, (iii) test whether a minimal realisation (*i.e.* system is observable and controllable) has been obtained, otherwise calculate a minimal realisation, (iv) give the I/O model using Laplace transformation (*i.e.* determine the transfer function $G(s,\vartheta)$), and finally (v) test structural identifiability of the given system configuration.
In the next section some preliminaries about elementary compartmental models and associated identifiability issues will be presented first. Then, in section 3 some system theoretic properties of compartmental models will be presented and illustrated to some prespecified models. The results will be discussed in the section 4. Finally the paper ends with some concluding remarks

2. PRELIMINARIES

A compartmental model consists of a finite number of compartments, which are assumed to be ideally mixed. In general, compartmental models are dominated by the law of conservation of mass. Each compartment with volume V_i is subject to two or more possible flows, as illustrated in Fig. 1 (see Bellman and Aström (1970), but also van den Hof (1996) for a recent treatment of compartmental systems).

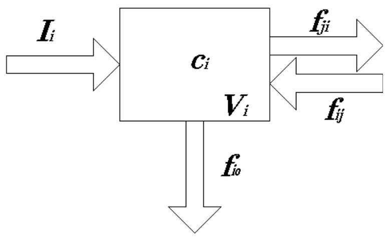

Figure 1: Compartment with possible flows.

In what follows, it is assumed that each compartment has constant volume. Hence, for compartment *i* the following volume, under constant specific density, balance equation holds,

$$\frac{dV_i}{dt} = \sum_{j\neq i}(-f_{ij} + f_{ji}) + I_i - f_{i0} = 0 \qquad (1)$$

It is further assumed that $f_{ij} = k_{ij}\,Q$, where Q in m^3/s is a positive constant flow and $k_{ij} \geq 0$, which are called the *fractional transfer coefficients*. Let $I_i = Q$, then it can be easily verified from Fig. 1 that, for instance, $k_{i0} = 1 + k_{ij} - k_{ji}$, so that algebraic relationships appear. In addition to this volume balance (1), the mass balance equations for the inert substance with concentration c_i in kg/m^3 will be added,

$$\frac{dc_i}{dt} = \frac{1}{V_i}\left(\sum_{j\neq i}(-f_{ij}c_i + f_{ji}c_j) + I_i c_{in} - f_{i0}c_i\right) \qquad (2)$$

where c_{in} is the input concentration in the inflow from outside the system. Notice that (2) can be written as the general linear system: $\dot{x}(t) = \Phi x(t) + \Gamma u(t)$, where the system matrix Φ and input matrix Γ are introduced here for later use. Furthermore, it is assumed that the observed output can be expressed as a linear combination of the concentrations, *i.e.*

$$y = \mathrm{H}c \qquad (3)$$

where $c = [c_1, c_2, \ldots, c_n]^T$ is the state vector, *i.e.* the vector containing all concentrations in the *n*-compartmental system, also called the state vector. In what follows, for simplicity and without loss of generality, only the single output case is considered. Note that the output is *not* necessarily related to the outflow of the compartmental system.

Compartmental systems can also be represented by a directed graph, in which each compartment is represented by a vertex or node and the flows by a directed arc. For describing mixing phenomena three elementary classes of compartmental systems will be distinguished in the sequel (see Fig. 2). The first class contains so-called *catenary*, if $f_{1n} = f_{n1} = 0$, or *circular* systems, if $f_{1n} \neq 0$ and/or $f_{n1} \neq 0$. A typical example of a catenary system is a tanks-in-series model with backflows (see Fig. 3A), while such a model of, for instance, an oxidation ditch or carrousel leads to a circular system description (see *e.g.* Abusam *et al.*, 2000). The second class is the so-called class of *mammilary* systems, which all have a central or *mother* compartment (see Figs. 2B-3B). The third class contains *interconnected parallel* systems (see Figs. 2C-3C).

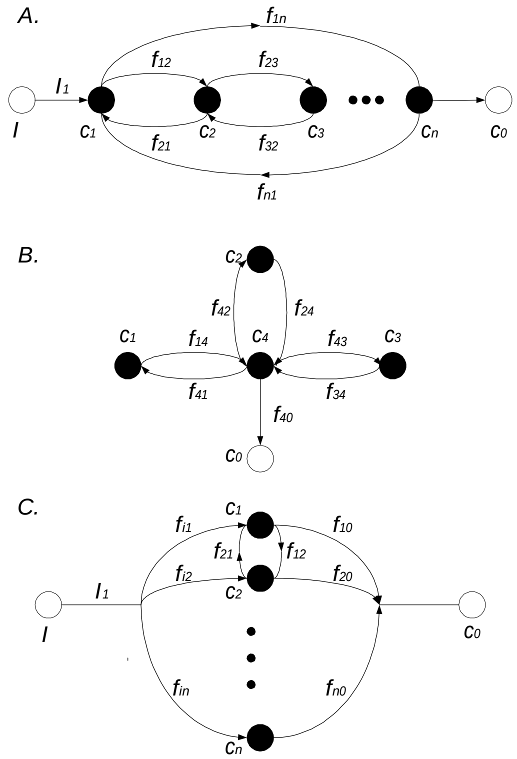

Figure 2: Elementary compartmental systems: A. circular/catenary, B. mammilary and C. interconnected parallel.

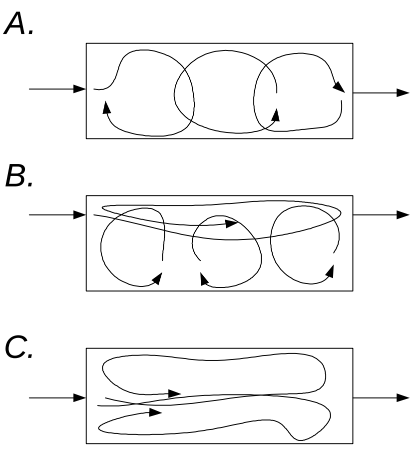

Figure 3: Typical side-viewed mixing patterns: A. catenary, B. mammilary and C. interconnected parallel.

In the next section, each of these compartmental systems will be subject to a structural identifiability analysis in order to obtain *physically-based, structural identifiable* state-space models of the form (2-3) for describing partial mixing.
But, let us first recall the structural identifiability test in a very pragmatic way. That is, let $G(s,\vartheta)$ be a parameterised transfer function of the system, where ϑ is a p-dimensional parameter vector. The model structure is said to be *globally identifiable* at ϑ^* if the equations in ϑ that arise from the equivalence

$$G(s,\vartheta) \equiv G(s,\vartheta^*) \quad \forall s$$

has the solution: $\vartheta = \vartheta^*$

Hence, the Laplace transform of the linear system (2-3), providing the parameterised transfer function $G(s,\vartheta)$, is needed for this analysis. Furthermore, if the model is structural identifiable, for practical use relationships between the physically interpretable parameters (k_{ij} , V_i) and the transfer function parameters g_k for k = 0, 1, 2, … , given the empirical transfer function $G(s)$, should also be provided. For the problem of how to obtain $G(s)$, either directly or via transformation of a time series model, from I/O data we refer to classical system identification books as Norton (1986) and Ljung (1987).

3. SYSTEM THEORETIC PROPERTIES OF TWO-COMPARTMENTAL MIXING MODEL

Let us as a first example consider the following system configuration (see Fig. 4). The introduction of the backflows can be motivated as follows. Starting from the convection-diffusion (C-D) equation and after semi-discretisation with step Δz one obtains,

$$\frac{dc_i(t)}{dt} = \frac{D}{\Delta z^2} c_{i+1}(t) - \left(\frac{2D}{\Delta z^2} + \frac{\nu}{\Delta z}\right) c_i(t) + \left(\frac{D}{\Delta z^2} + \frac{\nu}{\Delta z}\right) c_{i-1}(t) \qquad (4)$$

where i indicates the compartment index, D is the diffusion coefficient and ν the velocity. Hence (4) describes the concentration in the ith compartment as a function of the concentrations in the compartment i-1 to i+1, and thus backflows naturally appear. Instead of starting the modelling from this C-D equation the key idea here is to find a physically interpretable mixing model structure from the data.
As mentioned before fixed constant volumes are assumed, so that the outflow (Q) equals the inflow.

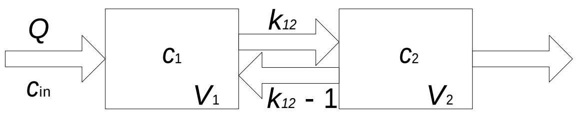

Figure 4: Two-compartmental model with backflow.

Furthermore, the positive mixing parameter k_{12} is introduced, such that the flow from compartment 1 to 2 is equal to k_{12} Q in order to allow a backflow. Hence, the flow from compartment 2 to 1 is equal to (k_{12}-1) Q. Notice then that backflow occurs for k_{12} >1, which puts an additional constraint on the fractional transfer coefficient. And thus, in this particular case the following mass balances for an inert substance with concentration $c_i(t)$ for i =1, 2 can be derived for each ideally mixed compartment:

$$\frac{dc_1(t)}{dt} = -k_{12}\frac{Q}{V_1}c_1(t) + (k_{12}-1)\frac{Q}{V_1}c_2(t) + \frac{Q}{V_1}c_{in}(t) \quad (5)$$

$$\frac{dc_2(t)}{dt} = k_{12}\frac{Q}{V_2}c_1(t) - k_{12}\frac{Q}{V_2}c_2(t) \quad (6)$$

where $c_{in}(t)$ is the control input, that is the known incoming concentration. Furthermore, if only the concentration in compartment 2, $c_2(t)$, is measured, the following output equation can be added:

$$y(t) = c_2(t) \quad (7)$$

In matrix-vector notation:

$$\frac{d}{dt}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} = \begin{bmatrix} -k_{12}Q/V_1 & (k_{12}-1)Q/V_1 \\ k_{12}Q/V_2 & -k_{12}Q/V_2 \end{bmatrix}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} + \begin{bmatrix} Q/V_1 \\ 0 \end{bmatrix} c_{in}$$

$$y = \begin{bmatrix} 0 & 1 \end{bmatrix}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} \quad (8)$$

It can be easily verified, using the Matlab symbolic toolbox and the controllability and observability functions *ctrb* and *obsv*, that (8) is a *minimal state-space realisation* (*i.e.* system is controllable and observable, see *e.g.* Kwakernaak and Sivan, 1972) for almost any parameter value. The transfer function of this system configuration is given by

$$G(s,\vartheta) = \frac{\frac{k_{12}Q^2}{V_1V_2}}{s^2 + k_{12}Q(\frac{1}{V_1}+\frac{1}{V_2})s + \frac{k_{12}Q^2}{V_1V_2}} \quad (9)$$

where $\vartheta := [k_{12}\ V_1\ V_2]^T$. Hence, it can be easily verified that only two parameter combinations, for instance $\alpha_1 := \frac{k_{12}Q^2}{V_1V_2} = q_0 = p_0$ and $\alpha_2 := k_{12}Q(\frac{1}{V_1}+\frac{1}{V_2}) = p_1$ can be identified from a given empirical transfer function $G(s)$, where

$$G(s) = \frac{q_n s^n + ... + q_1 s + q_0}{s^n + p_{n-1}s^{n-1} + ... + p_1 s + p_0} \quad (10)$$

Consequently, only two out of the three physically interpretable parameters can be determined. Select V_2 such that $V_2 < V$, the total reactor volume. Then, from (9) the following estimates are obtained:

$$\hat{V}_1 = \frac{(p_1Q - q_0V_2)}{q_0} \quad (11)$$

$$\hat{k}_{12} = \frac{(p_1Q - q_0V_2)*V_2}{Q^2} \quad (12)$$

The estimated total volume is found from $\hat{V}_1 + V_2 = \frac{p_1}{q_0}Q$, which may appear to be smaller than the actual volume, indicating that the active mixing volume is smaller than the total reactor volume. A remarkable result is found, when V is considered to be exactly known *a priori*. Then after substituting $V_2 = V - V_1$ in (9) only one identifiable parameter (k_{12}) remains! It can be further shown, that the model for any $k_{12} > 1$ will always give a non-oscillatory and stable response.

However, if the sensor is placed in the first compartment, that is H = [1 0], the following transfer function is obtained:

$$G(s,\vartheta) = \frac{\frac{Q}{V_1}s + \frac{k_{12}Q^2}{V_1V_2}}{s^2 + k_{12}Q\left(\frac{1}{V_1}+\frac{1}{V_2}\right)s + \frac{k_{12}Q^2}{V_1V_2}} \quad (13)$$

From $Q/V_1 = q_1$ one can estimate V_1 and thus one extra equation is available. Hence all three parameters, k_{12}, V_1 and V_2, can be identified from I/O data. Consequently, the sensor location is crucial.

From this example the following results, which can also be easily proven using elementary algebra and matrix theory for the general case, are deduced:

Result I: for physical, mass conserving compartmental systems without by-passes, as *e.g.* (9) and (13), (in mathematical terms: strict proper systems with unit gain) of order n at most $2n$-1 parameters are structural identifiable.

Result II: locating the sensor in the "input" compartment will lead to the maximally possible structural identifiable parameters.

In order to appreciate the minimal realisation condition, mentioned before, consider the following example, where $\Phi = \begin{bmatrix} 0 & k_1 \\ 0 & k_2 \end{bmatrix}$, $\Gamma = \begin{bmatrix} 1 \\ 0 \end{bmatrix}$ and $H = \begin{bmatrix} 1 & 0 \end{bmatrix}$. One can easily verify that this system is *un*controllable, but it is observable for $k_1 \neq 0$! Consequently, this is a non-minimal realisation. Calculation of the transfer function gives: $G(s) = H(sI - \Phi)^{-1}\Gamma = 1/s$. Hence, both k_1 and k_2 can never be identified from I/O data. It should be noted that minimality is not sufficient for identifiability (see example above), but non-minimality leads in general to unidentifiability!

It should be noted that this paper focuses on structural identifiability, implicitly assuming that all transfer function coefficients q_k and p_k, for k = 0, 1, 2, …, are available. In practice this means that the system should be persistently excited, so that these coefficients can be estimated from I/O data.

4. RESULTS AND DISCUSSION

In this section alternative compartmental model structures, such as mammilary and interconnected parallel systems of order two and three, are also evaluated. In the sequel only the case with optimally chosen sensor location will be considered, that is with sensor in the "input" compartment.

For a second-order mammilary system with inflow and sensor in the first compartment the following state-space model, where $k_{21} = k_{12}$, can be derived

$$\frac{d}{dt}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} = \begin{bmatrix} -(1+k_{12})Q/V_1 & k_{12}Q/V_1 \\ k_{12}Q/V_2 & -k_{12}Q/V_2 \end{bmatrix}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} + \begin{bmatrix} Q/V_1 \\ 0 \end{bmatrix} c_{in} \tag{14}$$

$$y = \begin{bmatrix} 1 & 0 \end{bmatrix}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix}$$

So the transfer function becomes,

$$G(s,\vartheta) = \frac{\frac{Q}{V_1}s + \frac{k_{12}Q^2}{V_1V_2}}{s^2 + \left[k_{12}\left(\frac{1}{V_1}+\frac{1}{V_2}\right)+\frac{1}{V_1}\right]Qs + \frac{k_{12}Q^2}{V_1V_2}} \tag{15}$$

Again, for $k_{12} \geq 0$ non-oscillatory, stable responses appear, the model is a minimal realisation and all three parameters k_{12}, V_1, and V_2 are identifiable. The estimates are found from

$$\hat{V}_1 = \frac{Q}{q_1} \tag{16}$$

$$\hat{V}_2 = -Q\frac{(q_0 + q_1^2 - q_1 p_1)}{q_0 q_1} \tag{17}$$

$$\hat{k}_{12} = -\frac{(q_0 + q_1^2 - q_1 p_1)}{q_1^2} \tag{18}$$

Finally a second-order interconnected parallel system with inflow splitting coefficient k_{in} can be represented as

$$\frac{d}{dt}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix} = \begin{bmatrix} -(k_{in}+k_{21})Q/V_1 & k_{21}Q/V_1 \\ k_{12}Q/V_2 & -(1-k_{in}+k_{12})Q/V_2 \end{bmatrix} * \begin{bmatrix} c_1 \\ c_2 \end{bmatrix} + \begin{bmatrix} k_{in}Q/V_1 \\ (1-k_{in})Q/V_2 \end{bmatrix} c_{in} \tag{19}$$

$$y = \begin{bmatrix} k_{in} - k_{12} + k_{21} & 1 - k_{in} + k_{12} - k_{21} \end{bmatrix}\begin{bmatrix} c_1 \\ c_2 \end{bmatrix}$$

Due to space limitations the transfer function related to (19) is not given here. It should be noted that $k_{in} \in [0, 1]$ and $0 \leq k_{in} - k_{12} + k_{21} \leq 1$. Given these restrictions no unstable behaviour can be expected. Notice that the model contains five unknowns, while only three equations can be deduced (see also Result II) . Hence, 10 combinations have to be evaluated. It appears, however, that only the parameter sets $\{k_{12}, k_{21}, V_1$ or $V_2\}$ can be uniquely identified. Because of the complexity of the solutions these are not presented here. Consequently, all combinations with k_{in} are unidentifiable. This can be easily understand because of the symmetry in the model structure (see Fig. 2C). The three combinations with both V_1 and V_2 lead to even three or four solutions. If the sensor is placed in the first compartment instead of in the outflow the identifiability properties significantly increases: all combinations with k_{12}, except $\{k_{12}, k_{21}, V_2\}$ are identifiable! For the case the sensor is placed in the second compartment similar results are obtained. Of course, also special cases where *e.g.* $k_{12} = 0$, $k_{12} = k_{21} = 0$ or $k_{12} = k_{in} + k_{21}$ can be evaluated. Only in the last case, where $k_{12} = k_{in} + k_{21}$, identifiable parameters, in particular $\{k_{in}, k_{21}, V_1\}$, are found.

Extension to third-order model structures lead to the following conclusion. Only a catenary system with sensor in the "input" compartment lead to a fully identifiable set, *i.e.* the set $\{k_{12}, k_{21}, V_1, V_2, V_3\}$. In all other cases symmetry appear and thus in general two or more solutions will be found. Hence, unique representations will not be found. Addition of sensors in the different compartments will certainly help to improve the identifiability of the system.

Let us finally demonstrate the theory to a practical example, which can be found in Young and Lees (1993). In this example, the modelling of solute transport in an experimental soil column from experimental data is considered. Young and Lees identified a discrete-time transfer function of order [3, 3, 0] (standard order notation), which gave an almost perfect fit and which for our case has been converted, using a zero-order hold approximation, to the following continuous-time transfer function:

$$G(s) = \frac{1.436s^2 + 0.117s + 0.2202}{s^3 + 0.624s^2 + 0.1788s + 0.01787} \tag{20}$$

On the basis of this empirical transfer function we will try to find an identifiable, physically interpretable state-space realisation. Notice first that the static gain of $G(s)$, setting $s \equiv 0$, is equal to 12.32 and not 1.0 as expected. Hence, in what follows this factor is assumed to be originated from the conversion from input to measurement.

Let us start with a third-order catenary system with unknowns k_{12}, k_{23}, V_1, V_2 and V_3. It appears, however, that the unique relationships between these unknowns and the transfer function coefficients result in negative values for both $k_{23} = -367.4$ and $D_3 = V_3/Q = -104.9$. Hence, this type of realisation is physically *unfeasible*. If a by-pass from compartment 1 to 3 with fractional transfer function k_{13} is added, it can be shown that either the estimate of k_{13} or V_2 is negative. And thus, again an *unfeasible* solution appears. Let us try next a mammilary system with three compartments. From symmetry of the problem we know that in this case two solutions can be expected, which are interrelated. However, the result is that except for V_1 all other parameter estimates take imaginary values. Consequently, a mammilary system representation is *unfeasible*, too. Investigation of parallel systems without interaction lead to the conclusion that only exponential decaying responses result, and thus no appropriate fit can be obtained. At last, mixed catenary/parallel system have been investigated. First of all no explicit relationships between five physical system parameters and the transfer function coefficients could be obtained using Maple symbolic software. Direct fitting of the impulse response generated from (20) gives one reasonable approximation using a parallel interconnected system (compartments 1 and 3) with in the upper path an extra compartment (2). For this structure the following six parameters have been found: k_{01} = 0.16, k_{12} = 0.86, V_1/Q = 0.21, V_2/Q = 0.21, V_3/Q = 1.0 and k_{13} = 0.0 with mean square error of $4*10^{-5}$. However, an exact fit using the mixing theory of section 2 could not be realised. A model structure in line with the one suggested by Young and Lees, containing four compartments gives k_{01} = 0.84 (related to input of slow path), $V_1/Q = V_2/Q = V_3/Q$ = 0.31 and V_4/Q = 4.81 with MSE equal to $1.4*10^{-5}$. Typical fits are presented in Fig. 5.

In conclusion, the expected exact fit could not be realised due to lack of identifiability of the mixing models, complexity of the analytical solution and the existence of imaginary solutions. Furthermore, the numerical search is hampered by the existence of local minima. Hence, for this specific case the conjecture of parallel flow (as suggested by Young and Lees) with a fast first-order system and a slow third-order system could not be further worked out along the line of section 2.

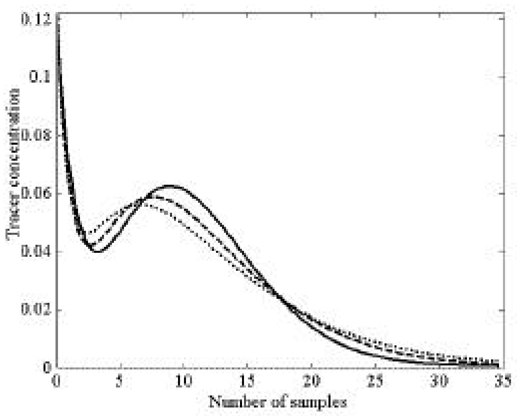

Figure 5. Typical impulse response fits of 3rd-order (...) and 4th-order system (---) with data from Young and Lees (1993).

5. CONCLUDING REMARKS

In the paper a number of low-dimensional system configurations with catenary/circular and interconnected parallel flows have been evaluated using elementary Laplace transform analysis. It appears that for full identifiability from given I/O data the number of parameters, and thus the number of different compartments and flow patterns, should be limited, unless additional sensors are placed.

Given the empirical transfer function found from solute transport data (see Young and Lees, 1993) no physically interpretable (in line with the mixing theory of section 2) and feasible state-space realisation could be found. Catenary, circular and mammilary structures led to models with negative or even imaginary fractional transfer coefficients. Inclusion of parallel pathways was not successful either due to lack of identifiability and the existence of imaginary solutions and of local minima.

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