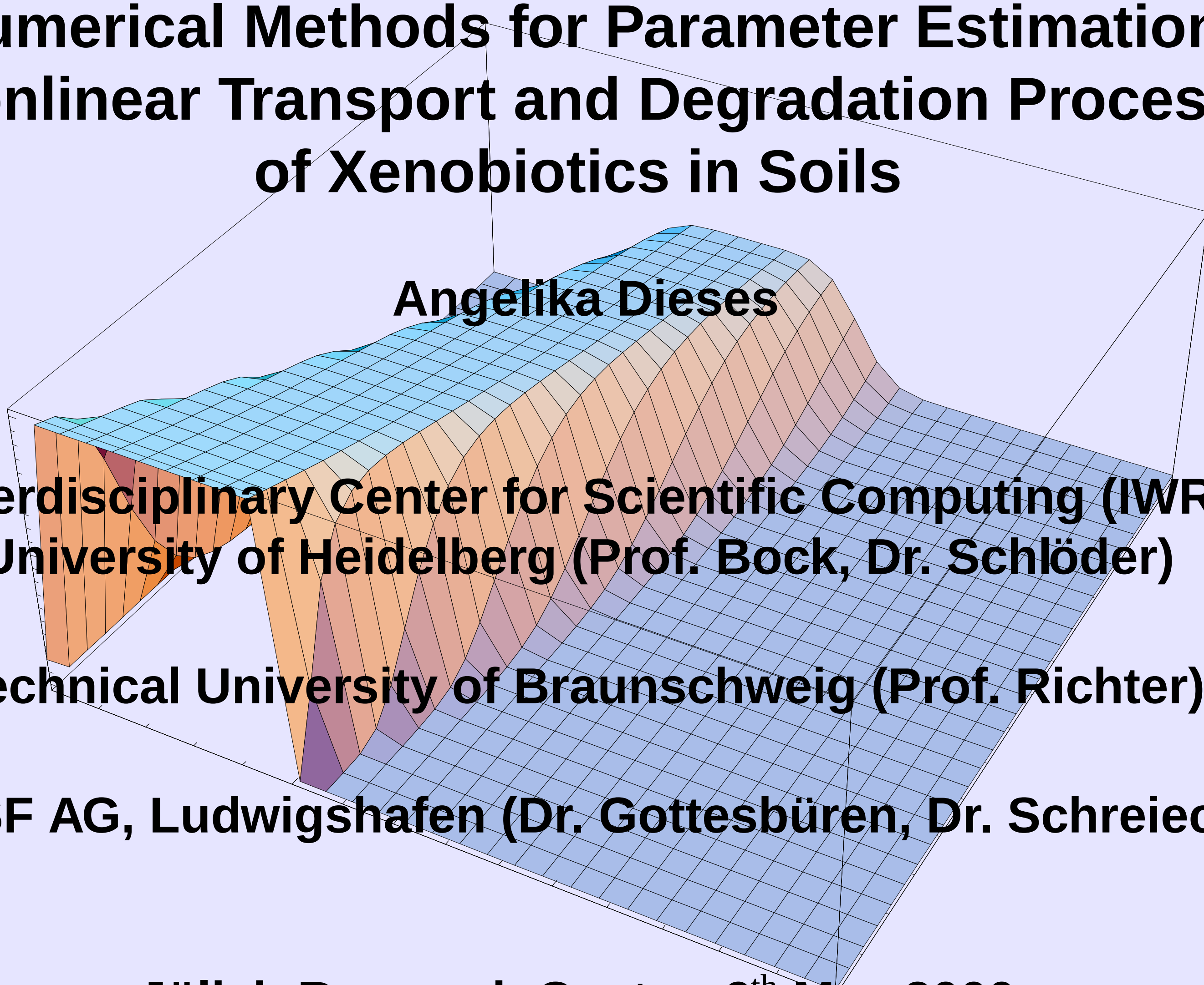
Numerical Methods for Parameter Estimation in Nonlinear Transport and Degradation Processes of Xenobiotics in Soils



Angelika Dieses

Interdisciplinary Center for Scientific Computing (IWR)
University of Heidelberg (Prof. Bock, Dr. Schlöder)

Technical University of Braunschweig (Prof. Richter)

BASF AG, Ludwigshafen (Dr. Gottesbüren, Dr. Schreieck)

Jülich Research Centre, 9th May 2000

angelika.dieses@iwr.uni-heidelberg.de

Contents

Methods

- Formulation of the Inverse Problem
- Discretization in Space
- Multiple Shooting Method
- Generalized Gauss Newton Method
- Reduced Approach
- Accuracy of Derivatives

Applications

- Field Experiment: Estimation of Van Genuchten Parameters (K. Aden)
- Minilysimeter Study: Environmental Fate Parameters in a Parent/Metabolites System (A. Horn, O. Richter)

Formulation of the Inverse Problem

Given: experimental data η_{kij}, $(k = \psi, \theta, c_l)$, $(l = 1, \ldots, n)$, $(i = 1, \ldots, m_1)$, $(j = 1, \ldots, m_2)$

$$\eta_{kij} = b_k\big(t_i, z_j, k(t_i, z_j), p\big) + \varepsilon_{kij}, \qquad \varepsilon_{kij} \sim N\,(0, \sigma^2_{kij})$$

Aim: vector of parameters p and a solution $k(t,z)$, such that

$$\begin{aligned}
\min \|F(\psi, \theta, c_l; p)\|_2^2 &= \min \sum_{k=\psi,\theta,c_l} \sum_{i=1}^{m_1} \sum_{j=1}^{m_2} \sigma_{kij}^{-2}\big(\eta_{kij} - b_k(t_i, z_j, k(t_i, z_j), p)\big)^2 \\
C(\psi; p)\frac{\partial\psi}{\partial t} &= \frac{\partial}{\partial z}\Big(K(\psi; p)\frac{\partial}{\partial z}(\psi - z)\Big) + S_1(\psi; p) \\
\frac{\partial\theta}{\partial t} &= \frac{\partial}{\partial z}\Big(\bar{D}(\theta; p)\frac{\partial\theta}{\partial z} - \bar{K}(\theta; p)\Big) + S_2(\theta; p) \\
\frac{\partial(\theta c_l)}{\partial t} + \frac{\partial(\rho S_l)}{\partial t} &= \frac{\partial}{\partial z}\Big(\theta D_{h_l}(\theta; p)\frac{\partial c_l}{\partial z}\Big) - \frac{\partial}{\partial z}\big(q(p)c_l\big) + Q_l(c_1, \ldots, c_n; p) \\
&+ \text{ initial and boundary conditions}
\end{aligned}$$

Discretization in space and time

Method of lines: Transformation of PDE systems into systems of ODEs

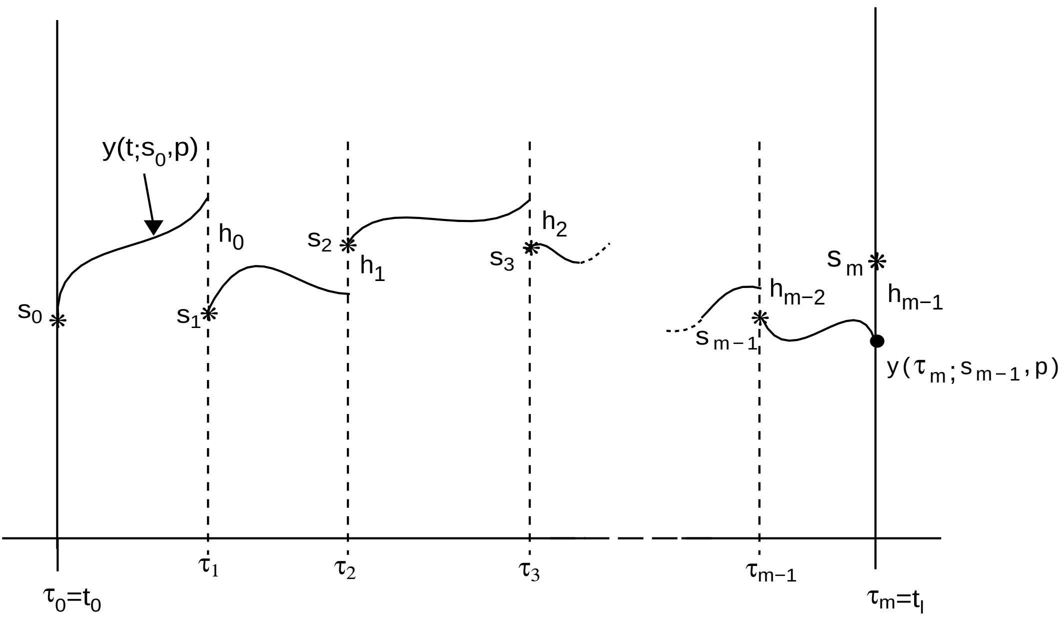
Space discretization: Finite Differences of higher order

- central differences, e.g. for diffusion-dispersion term
- upwind strategies, e.g. for convective term

Time discretization: Multiple Shooting

- time interval $[t_0, t_e]$ is divided into subintervals $[\tau_j, \tau_{j+1}]$ $(j = 0, \ldots, m-1)$
- on each subinterval an initial value problem is introduced: $\dot{y} = f(t, y, p) \qquad y(\tau_j) = s_j$
- additional matching conditions h_j enforce continuity of the final solution

Advantages of Multiple Shooting



Initial trajectory for multiple shooting

- Garantees an initial solution on the complete interval
- Use of prior information
  - Information about the solution
  - Experimental data as initial guesses for s_k
- Reduction of nonlinearity

Discretized Parameter Estimation Problem

Result of space and time discretization:
Large scale, nonlinear, constrained least squares problem in the augmented vector $x = (s_0, \dots, s_m, p)^T$

$$
\begin{aligned}
||r_1(s_0, \dots, s_m, p)||_2^2 &\to \min_x \\
r_2(s_0, p) &= 0 \\
\bar{r}_3(s_0, \dots, s_m, p) &= 0.
\end{aligned}
$$

r_2: initial conditions
$\bar{r}_3 = (r_3, h_0, \dots, h_{m-1})$: other equality conditions including matching conditions

Generalized Gauss Newton method (Bock/Schlöder)

1. Start with an initial guess x_0.
2. Improve the solution iteratively:

$$x_{k+1} = x_k + \lambda_k \Delta x_k,$$

where Δx_k solves the linearized problem

$$\begin{aligned} ||r_1(x_k) + J_1(x_k)\Delta x_k||_2^2 &= \min_{\Delta x_k} \\ r_2(x_k) + J_2(x_k)\Delta x_k &= 0 \\ \bar{r}_3(x_k) + \bar{J}_3(x_k)\Delta x_k &= 0. \end{aligned}$$

λ_k: relaxation factor of a globalization strategy

convergence to solution x^*: $J^+ r(x^*) = 0$

Structure of the Jacobian and the Right Hand Side

$$J = \begin{pmatrix} D_1^0 & D_1^1 & \dots & \dots & D_1^m & D_1^p \\ D_2^0 & 0 & \dots & \dots & 0 & D_2^p \\ D_3^0 & D_3^1 & \dots & \dots & D_3^m & D_3^p \\ G_0 & -I & & & & G_0^p \\ & G_1 & \ddots & & 0 & \vdots \\ & & \ddots & \ddots & & \vdots \\ & 0 & & G_{m-1} & -I & G_{m-1}^p \end{pmatrix} \qquad R = \begin{pmatrix} r_1 \\ r_2 \\ r_3 \\ h_0 \\ \vdots \\ h_{m-1} \end{pmatrix}$$

$$D_i^j = \partial r_i(s_0,\dots,s_m,p)/\partial s_j \qquad D_i^p = \partial r_i(s_0,\dots,s_m,p)/\partial p$$

$$G_i = \partial y(\tau_{i+1};s_i,p)/\partial s_i \qquad G_i^p = \partial y(\tau_{i+1};s_i,p)/\partial p$$

Dimension of the Problem

Parameter estimation should be done on the basis of highly accurate solutions of the PDEs.

$\Longrightarrow$ Sufficiently fine spatial grids (100-1000 space nodes)

$\Longrightarrow$ Large scale optimization problems

Example:

2 PDEs
400 Grid points in space
10 Multiple shooting nodes
} $\Longrightarrow$ 8000 Variables

6 Unknown parameters
40 Measurements
} $\Longrightarrow$ Jacobian with more than 7 mio entries $\Longrightarrow$ 56 MB

$\Longrightarrow$ **New strategies required !**

Reduced Approach (Schlöder)

- Exploitation of initial conditions
- Simultaneous evaluation and decomposition of linear systems
- No explicit computation and storage of D_l^i and G_i

$\Longrightarrow$ Successive evaluation of directional derivatives

$\Longrightarrow$ Only (dim p + 1) directional derivatives required

Result: Essentially, the same computational effort as for the **single shooting** while maintaining the advantages of **multiple shooting**.

Generation of Derivatives

Bad approximations of derivatives $\Longrightarrow$ Wrong parameter estimates

$$\frac{\partial y}{\partial y_0}\Delta y_0 \doteq \frac{y(t;y_0+\varepsilon\Delta y_0)-y(t;y_0)}{\varepsilon}$$

External numerical differentiation (END)

$\Longrightarrow$ 3 digits for derivatives: at least 6 digits for $y(t;y_0+\varepsilon\Delta y_0)$ and $y(t;y_0)$ are required

Internal numerical differentiation (IND)

IDEA: Compute the varied trajectory $y(t;y_0+\varepsilon\Delta y_0)$ with the same step size and order as the nominal trajectory $y(t;y_0)$.

$\Longrightarrow$ 3 digits for derivatives: only 3 digits for $y(t;y_0+\varepsilon\Delta y_0)$ and $y(t;y_0)$ are required

ECOFIT (Dieses, Schlöder, Bock)

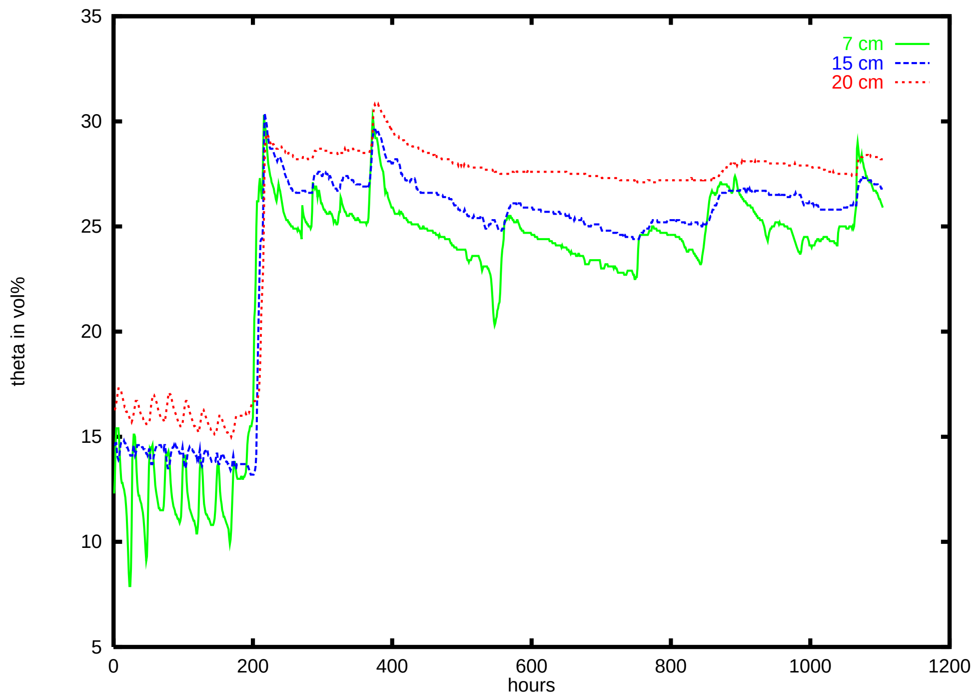
Reduced Generalized Gauss Newton method

- Fine space discretization
- Multiple Shooting (use of prior information)
- Exploitation of structures on several levels
- Efficient computation of derivatives

$\Longrightarrow$ **Efficient solution of large scale parameter estimation problems**

Field Experiment: Estimation of Van Genuchten Parameters (K. Aden)

- Loamy sand without crop cover
- Time-domain reflectrometry (TDR): hourly measurements for the water content θ in 7, 15 and 20 cm depth
- Time period: 28.10.1997-13.12.1997



Modelling: Richards Equation

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial z}\left(D(\theta)\frac{\partial\theta}{\partial z} - K(\theta)\right)$$

$$K(\theta) = K_s\Theta^{1/2}\left[1-\left(1-\Theta^{n/(n-1)}\right)^{1-1/n}\right]^2, \qquad \Theta = \frac{\theta-\theta_r}{\theta_s-\theta_r}$$

$$D(\theta) = K(\theta)\bar{C}(\theta)$$

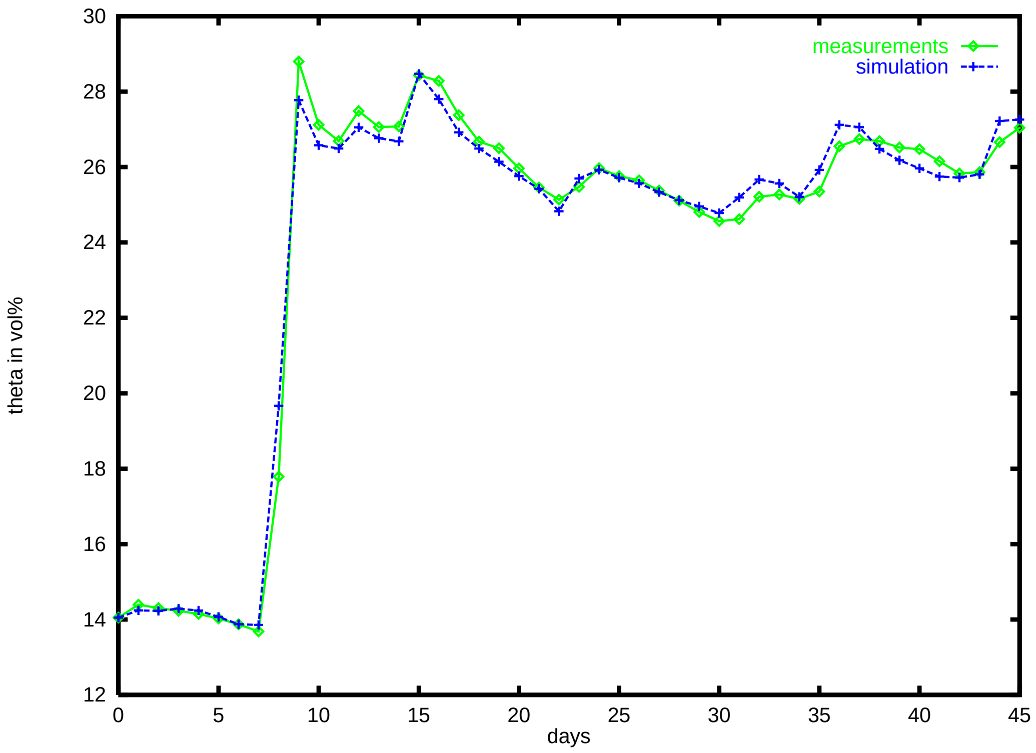
$$\bar{C}(\theta) = \frac{1}{\alpha nm}\left(\Theta^{-1/m}-1\right)^{-m}\Theta^{-1/m}\frac{1}{\theta-\theta_r}, \qquad m = 1-\frac{1}{n}$$

- Initial condition: Linear interpolation of θ_{7cm}, θ_{15cm}, θ_{20cm} at the beginning of the experiment (28.10.97)

- Upper boundary: Dirichlet condition (TDR data in 7 cm depth)

- Lower boundary: Dirichlet condition (TDR data in 20 cm depth)

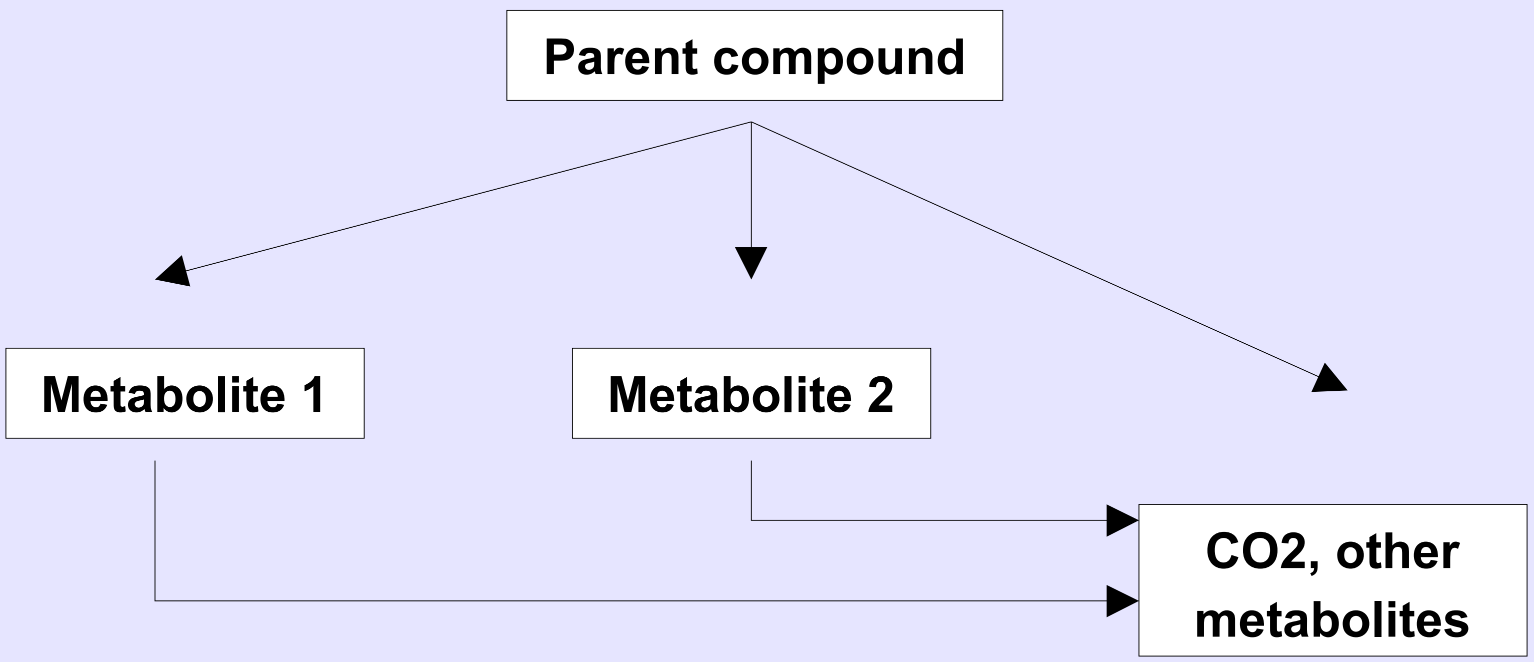
Resultats: Estimation of n, α and K_s



	Guess	Estimate
n	1.5	1.262 ± 0.0024
α	0.05	0.0324 ± 0.0024
K_s	35.0	20.92 ± 1.68

	α	K_s
n	0.14	-0.61
α	-	-0.94

Minilysimeter Study (A. Horn, Prof. O. Richter)



Description of Experiments

- Minilysimeter (30cm)
  1. Control column for water transport (-90 hPa at the lower boundary)
  2. Parent incorporated in the first 5cm of the column
- Exposition: normal climatical conditions (precipitation + irrigation)
- Measurements
  1. Water contents in 5, 15 und 20 cm depths (TDR)
  2. Outflow data every 14 days
- Unknown parameters
  1. Van Genuchten parameters
  2. Degradation rates, K_d-values, dispersion lengths (9 parameters)

Modelling

- **Water transport**
  $\Longrightarrow$ Problems with pressure at the lower boundary
  $\Longrightarrow$ Approximation of the water flux q from precipitation and irrigation data

- **Solute transport**

$$
\begin{aligned}
R_P \frac{\partial c_P}{\partial t} &= \frac{D_s}{\theta}\frac{\partial^2 c_P}{\partial z^2} - \frac{q}{\theta}\frac{\partial c_P}{\partial z} - k_1 R_p c_P - k_2 R_P c_P - k_r R_p c_P \\
R_{M_1} \frac{\partial c_{M_1}}{\partial t} &= \frac{D_s}{\theta}\frac{\partial^2 c_{M_1}}{\partial z^2} - \frac{q}{\theta}\frac{\partial c_{M_1}}{\partial z} + f_1 k_1 R_P c_P - k_{el1} R_{M_1} c_{M_1} \\
R_{M_2} \frac{\partial c_{M_2}}{\partial t} &= \frac{D_s}{\theta}\frac{\partial^2 c_{M_2}}{\partial z^2} - \frac{q}{\theta}\frac{\partial c_{M_2}}{\partial z} + f_2 k_2 R_P c_P - k_{el2} R_{M_2} c_{M_2},
\end{aligned}
$$

with

$$
R_l = 1 + \frac{\rho}{\theta} K_{d,l}, \qquad l = P, M_1, M_2
$$

$$
D_s = \lambda |q| + a \exp(b\theta) D_w.
$$

- Initial conditions

$$c_P(0,z) = \begin{cases} c_0 & x \leq 0.05 \\ 0 & x > 0.05 \end{cases} \qquad c_{M_1}(0,z) = c_{M_2}(0,z) = 0.0, \qquad z > 0.0$$

- Upper boundary condition

$$\nu c_P - \frac{D_s}{\theta}\frac{\partial c_P}{\partial z} = 0.$$

- Lower boundary condition

$$\lim_{z\to\infty} c_l(t,z) = 0, \qquad l = P, M_1, M_2$$

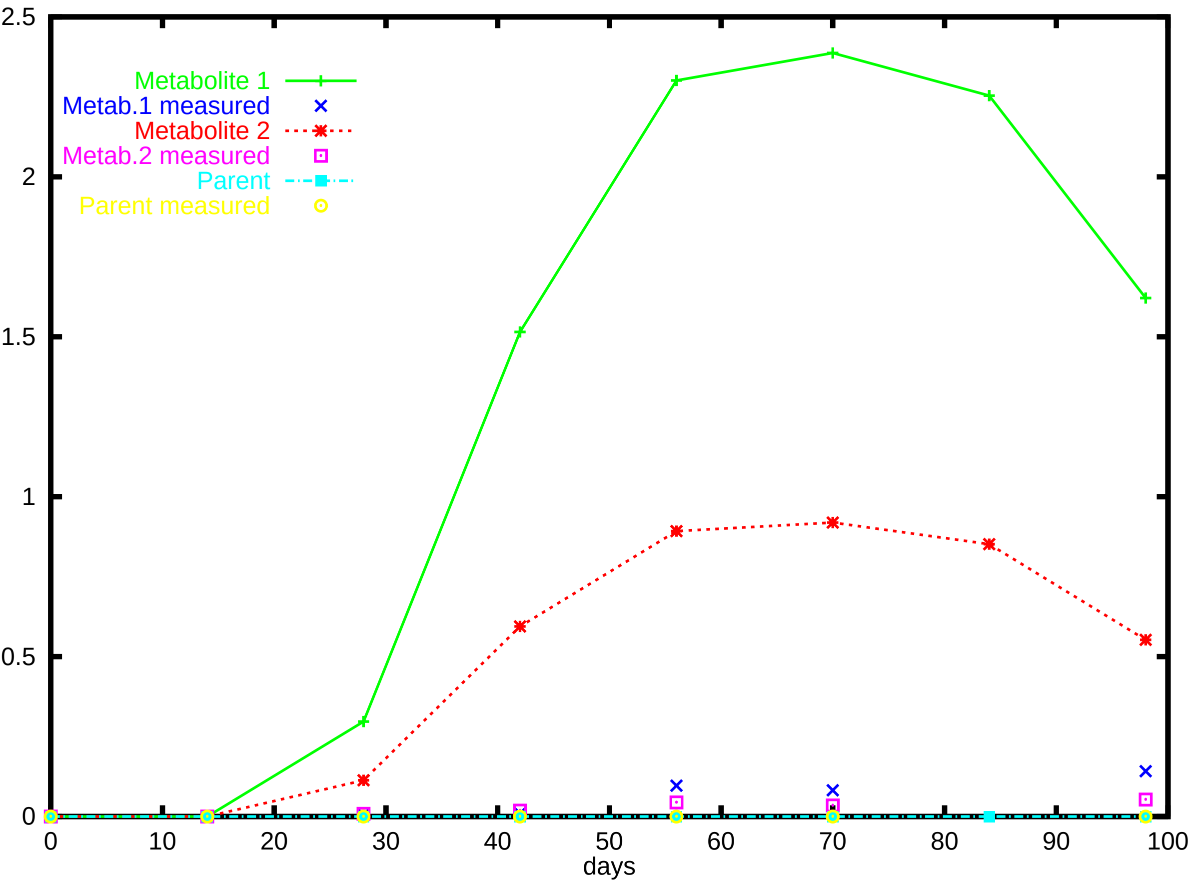
- Flux concentrations

$$c_l^{flux} = c_l - \frac{D_s}{q}\frac{\partial c_l}{\partial z}, \qquad l = P, M_1, M_2.$$

- Measurements: Outflow data for the time interval $[t_i, t_{i+1}]$

$$M_l^i = \int_{t_i}^{t_{i+1}} c_l^{flux}(\tau, L)\,d\tau, \qquad i = 1, \cdots, n$$

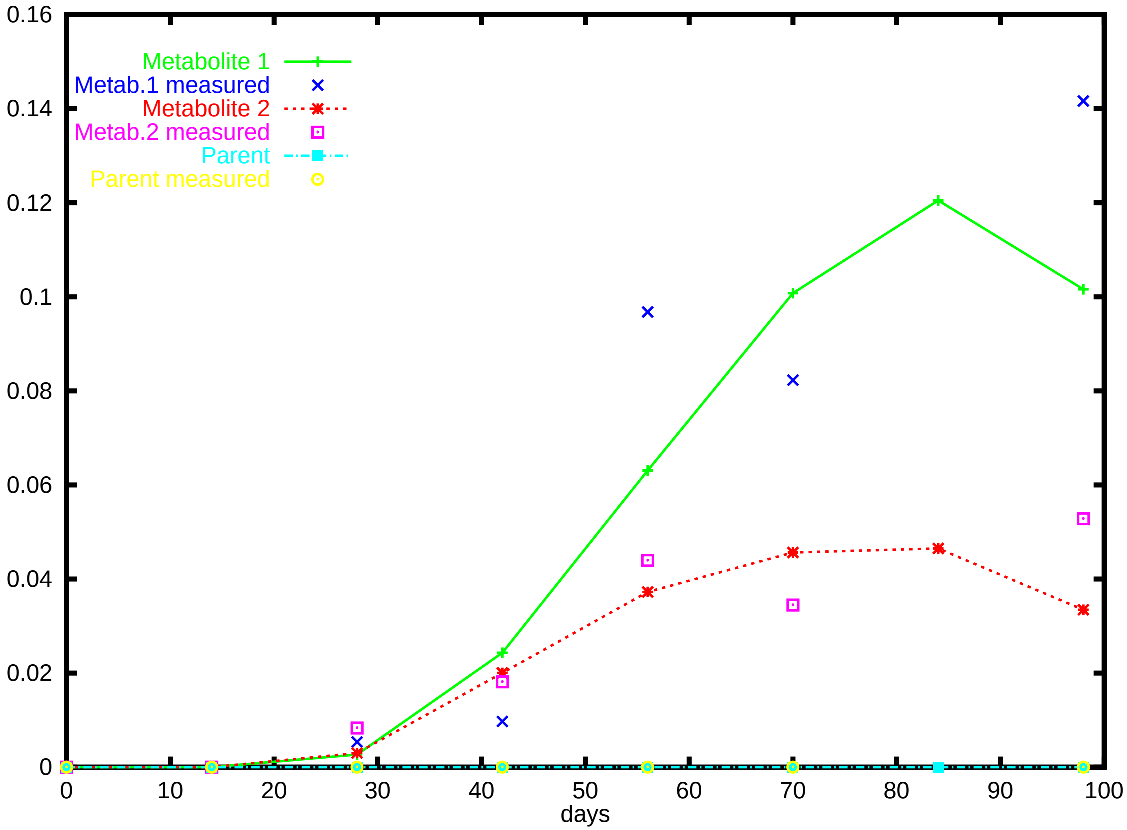
Simulation with Initial Guesses



	Initial Guess	Unit
$K_{d,P}$	2.95D-6	m^3/g
K_{d,M_1}	0.12D-6	m^3/g
K_{d,M_2}	0.14D-6	m^3/g
k_r	3.35D-2	$1/d$
k_1	1.1D-2	$1/d$
k_2	7.85D-3	$1/d$
λ	0.054	m
k_{el1}	5.31D-3	$1/d$
k_{el2}	1.32D-2	$1/d$
b	10.0	-
a	0.005	-

Fitting Resultats

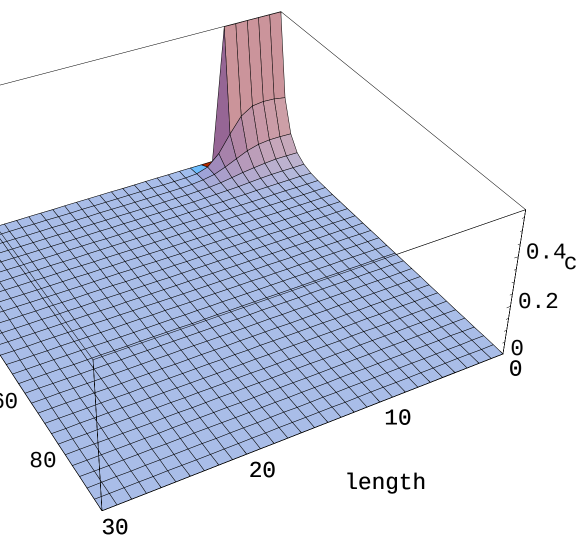
- Singular problem $\Longrightarrow$ simultaneous estimation of all 9 parameters is not possible



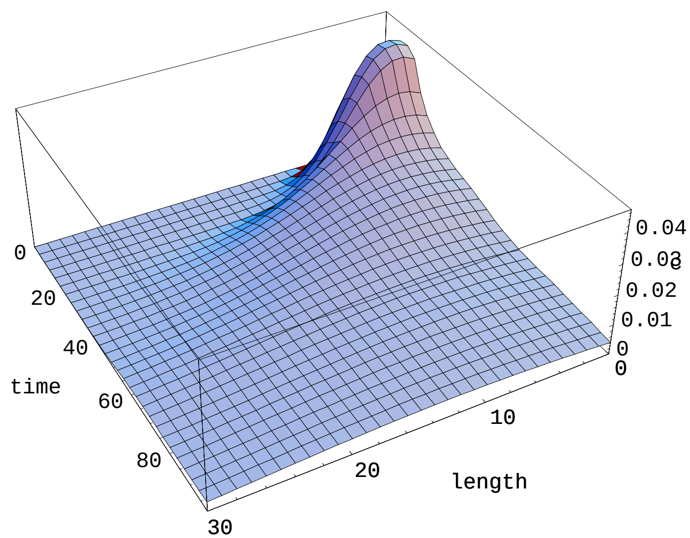
	Initial Guess	Estimates	95 % Conf.
$K_{d,P}$	2.95D-6	fixed	
K_{d,M_1}	0.12D-6	1.18D-6	$\pm$ 0.176D-6
K_{d,M_2}	0.14D-6	0.96D-6	$\pm$ 0.168D-6
k_r	3.35D-2	0.23	$\pm$ 0.06
k_1	1.1D-2	0.89D-2	$\pm$ 0.16D-2
k_2	7.85D-3	6.05D-3	$\pm$ 0.12D-2
λ	0.054	0.144	$\pm$ 0.02
k_{el1}	5.31D-3	5.37D-3	$\pm$ 0.64D-3
k_{el2}	1.32D-2	1.34D-2	$\pm$ 0.15D-2
b	10.0	4.83	$\pm$ 4.89
a	0.005	fixed	

Resident Concentrations

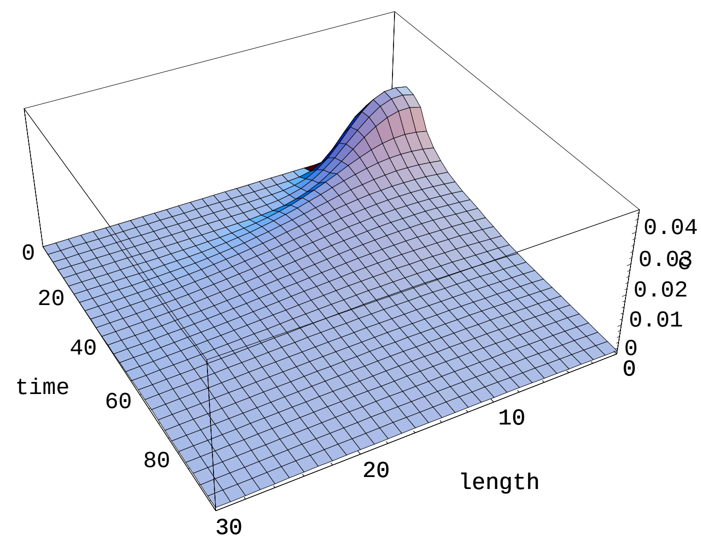
Parent



Metabolite 1



Metabolite 2



Summary and Outlook

State-of-the-art methods for parameter estimation in large scale systems $\Longrightarrow$ **ECOFIT**

Ill-posedness of inverse problems

- Parameters are often insensitive to observed data
- Unsatisfactory results (e.g. large confidence intervals)

$\Longrightarrow$ **OPTIMAL EXPERIMENTAL DESIGN (ECOPLAN)**