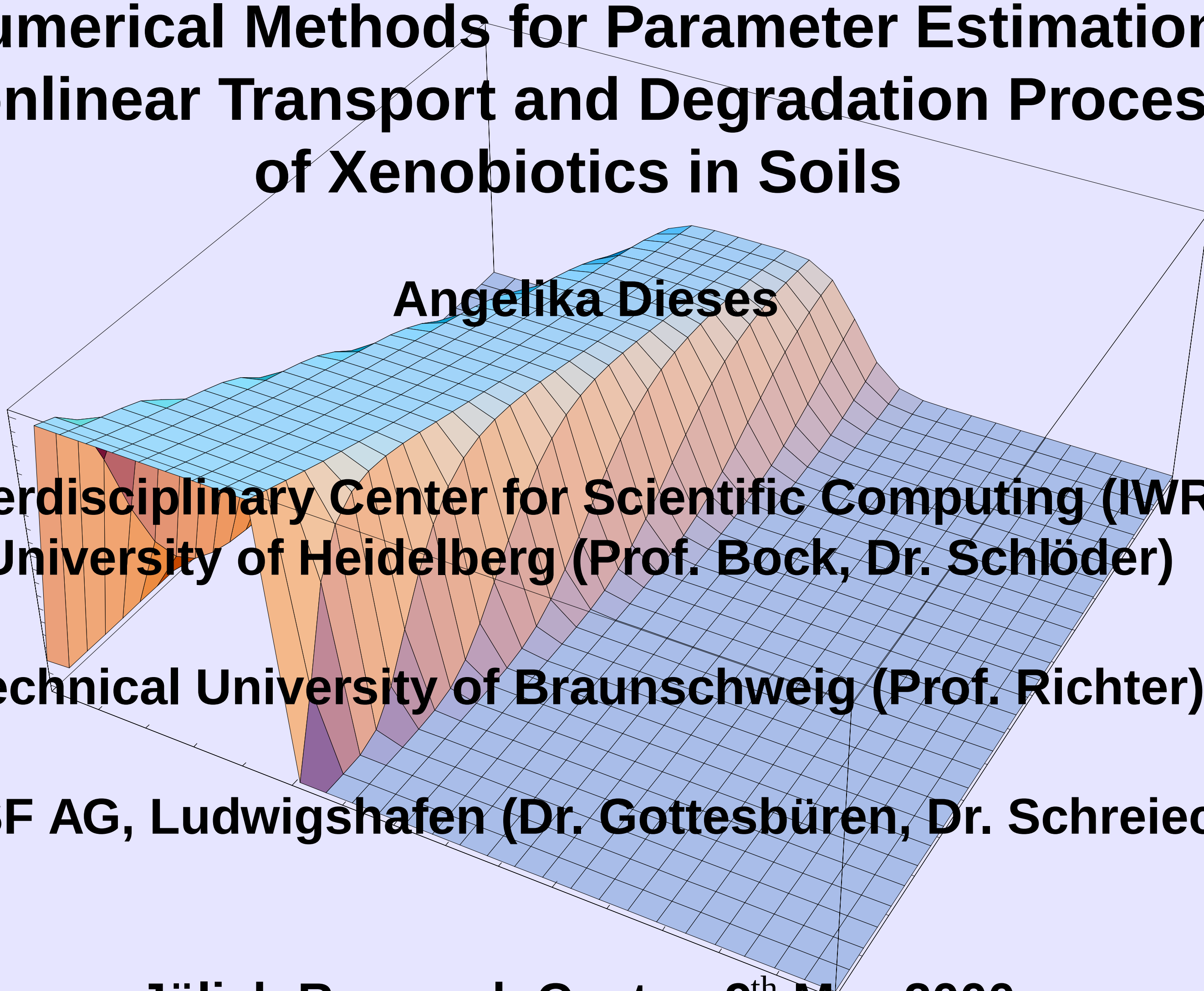
Numerical Methods for Parameter Estimation in Nonlinear Transport and Degradation Processes of Xenobiotics in Soils



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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

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

Discretization in space and time

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

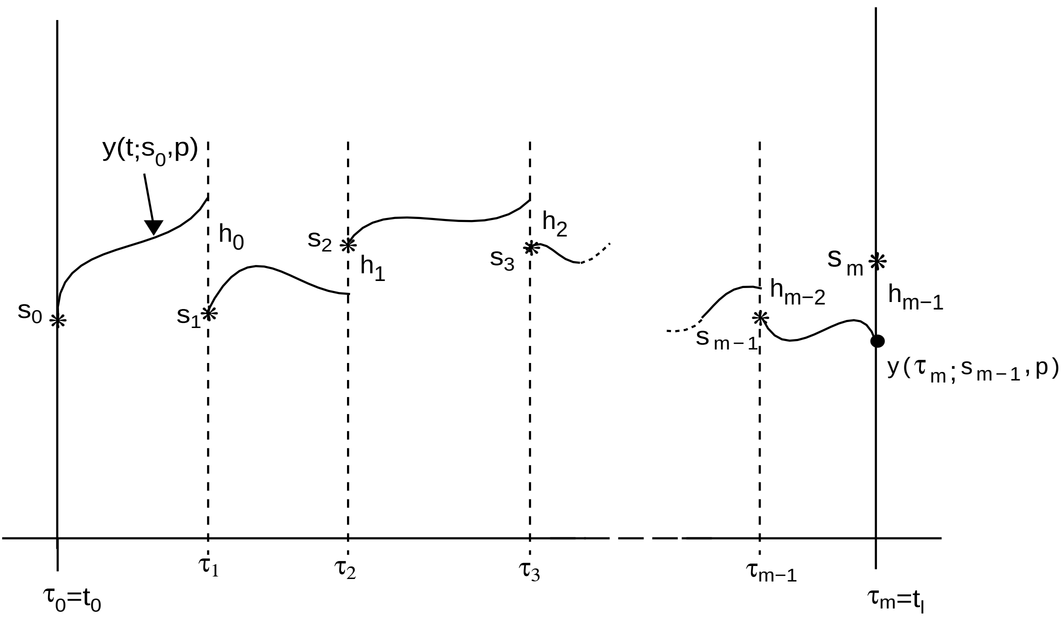
Space discretization: Finite Differences of higher order

- 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 $x = (s_0, \ldots, s_m, p)^T$

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

r_2: initial conditions
$\bar{r}_3 = (r_3, h_0, \ldots, h_{m-1})$: 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)

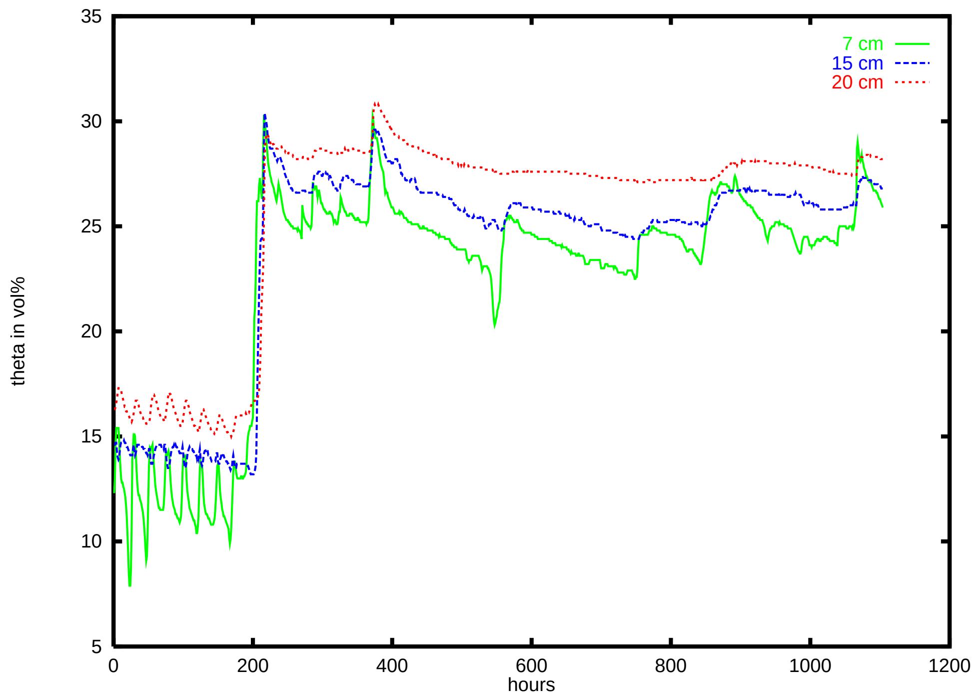
Reduced Generalized Gauss Newton method

- 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

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

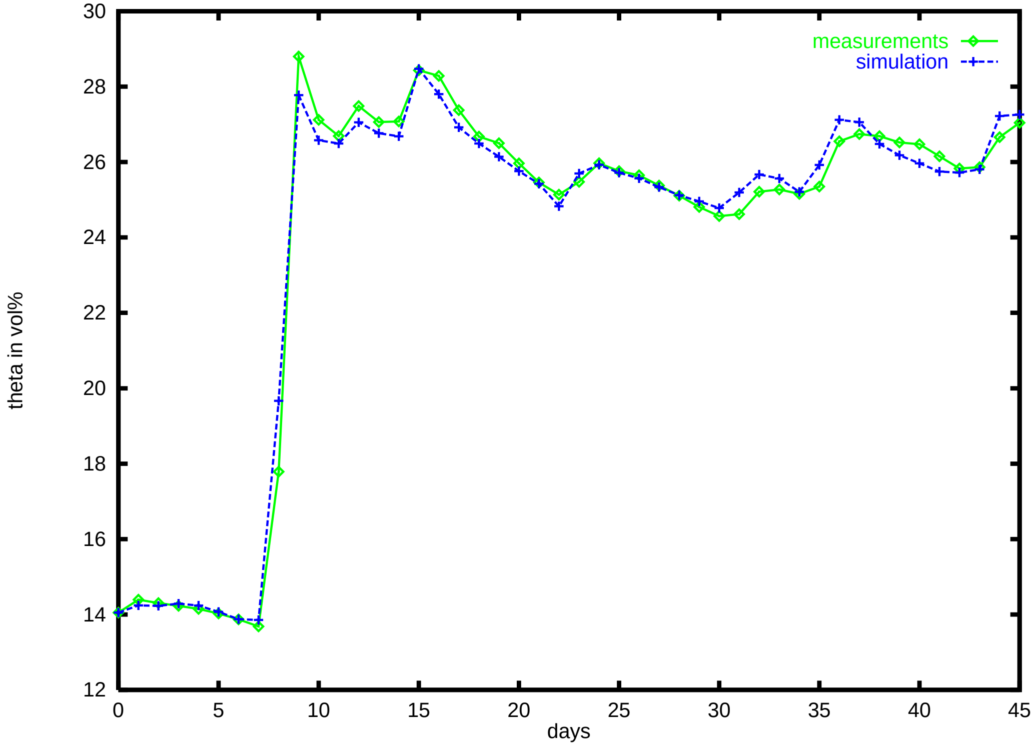
$$K(\theta) = K_s \Theta^{1/2}\left[1-\left(1-\Theta^{n/(n-1)}\right)^{1-1/n}\right]^2, \qquad \Theta = \frac{\theta-\theta_r}{\theta_s-\theta_r}$$

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

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

- 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)

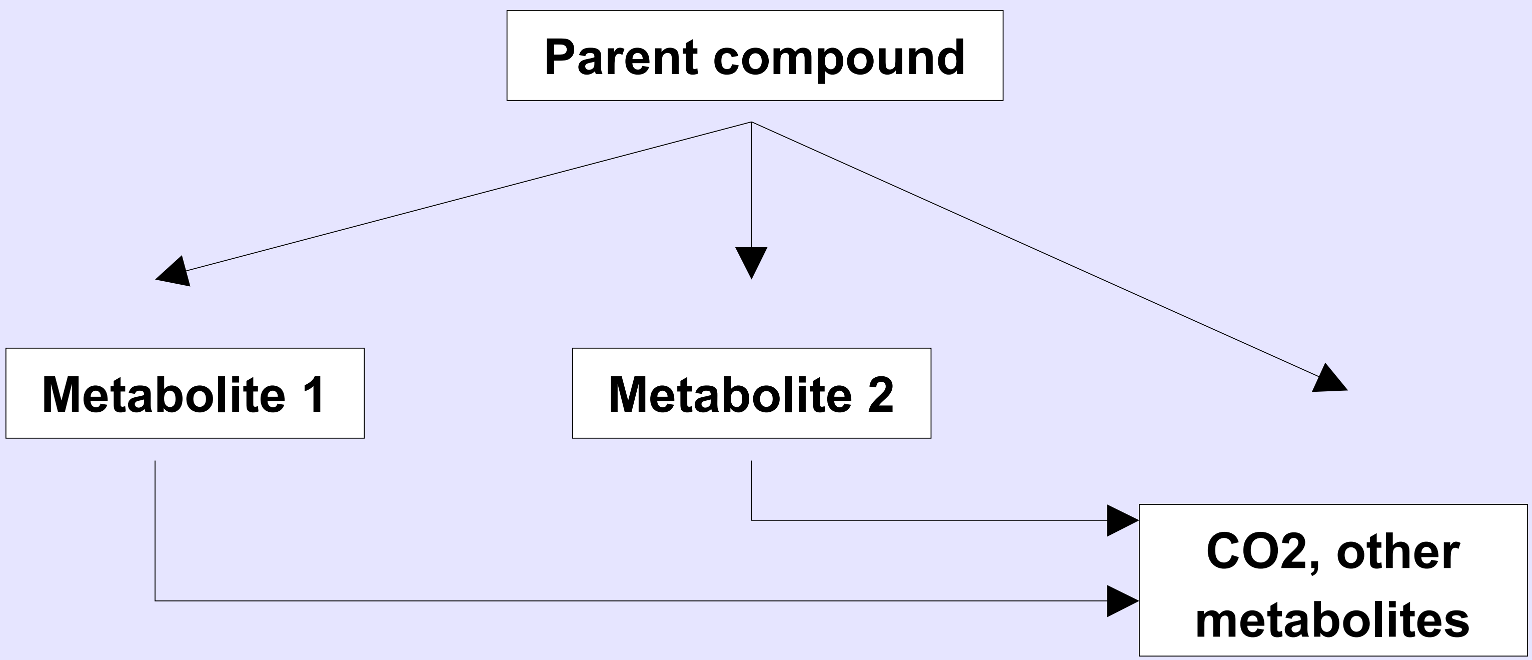
Resultats: Estimation of n, α and K_s



	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**

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

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

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

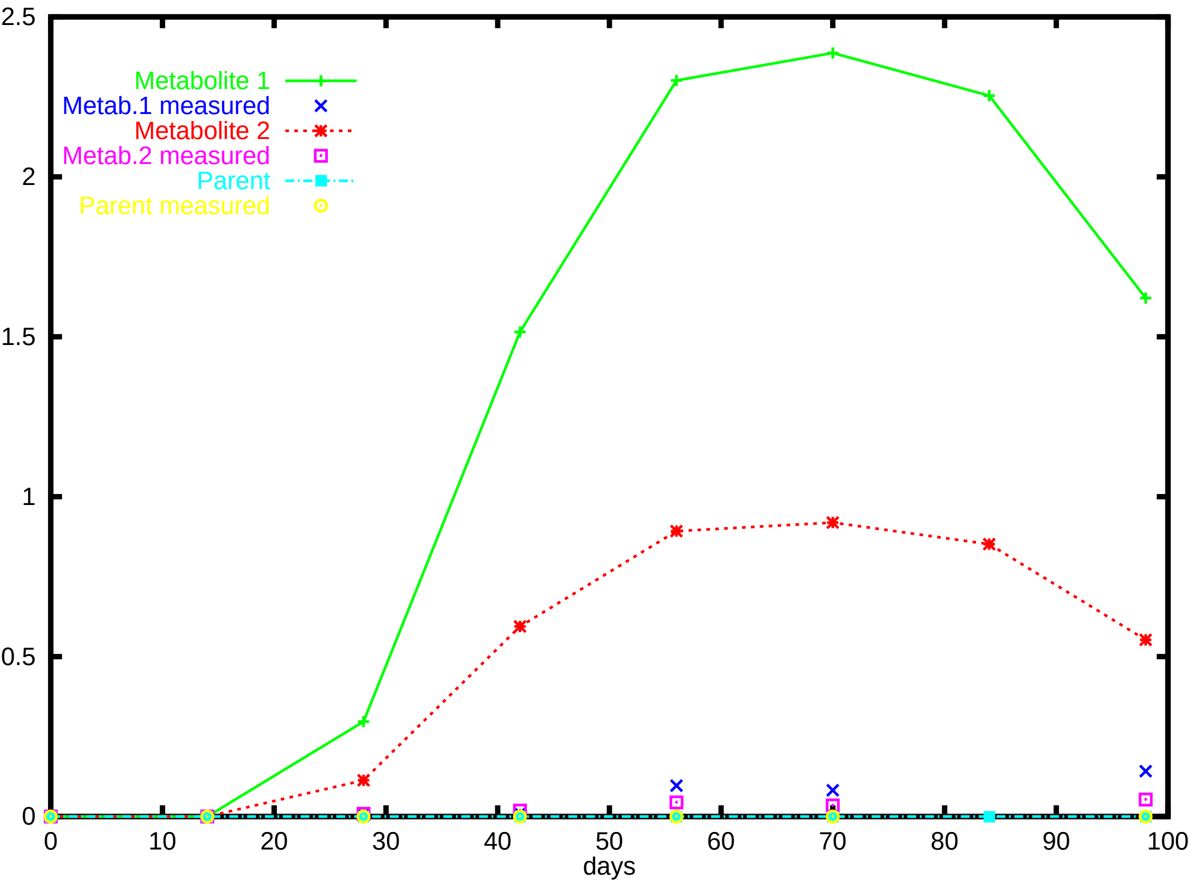
- Flux concentrations

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

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

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

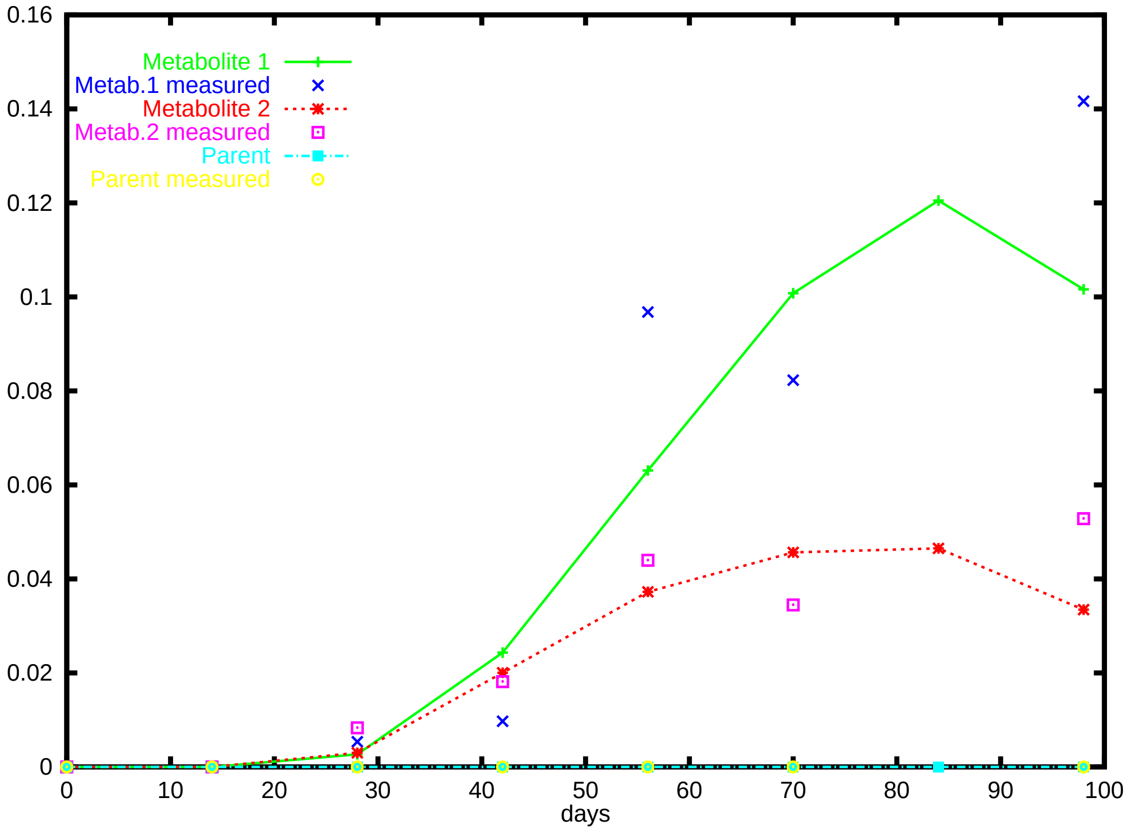
Simulation with Initial Guesses



	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

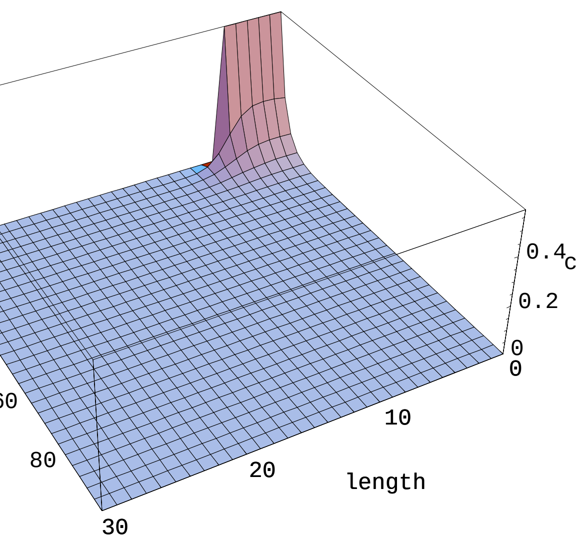
- Singular problem $\Longrightarrow$ simultaneous estimation of all 9 parameters is not possible



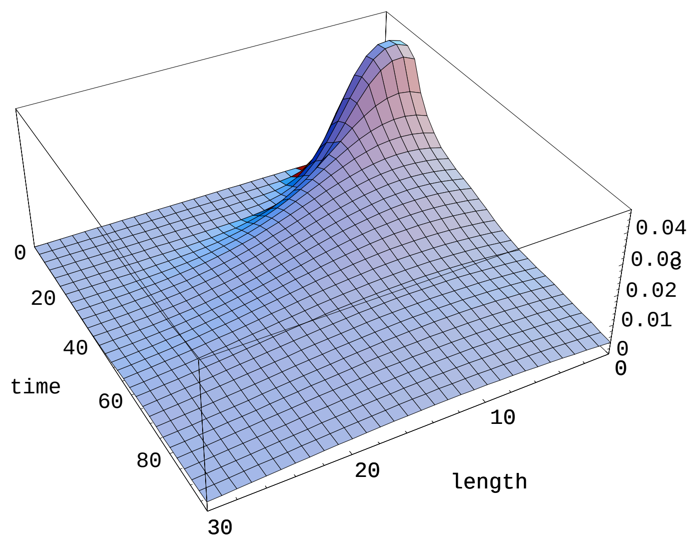
	Initial Guess	Estimates	95 % Conf.
$K_{d,P}$	2.95D-6	fixed	
K_{d,M_1}	0.12D-6	1.18D-6	± 0.176D-6
K_{d,M_2}	0.14D-6	0.96D-6	± 0.168D-6
k_r	3.35D-2	0.23	± 0.06
k_1	1.1D-2	0.89D-2	± 0.16D-2
k_2	7.85D-3	6.05D-3	± 0.12D-2
λ	0.054	0.144	± 0.02
k_{el1}	5.31D-3	5.37D-3	± 0.64D-3
k_{el2}	1.32D-2	1.34D-2	± 0.15D-2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

Resident Concentrations

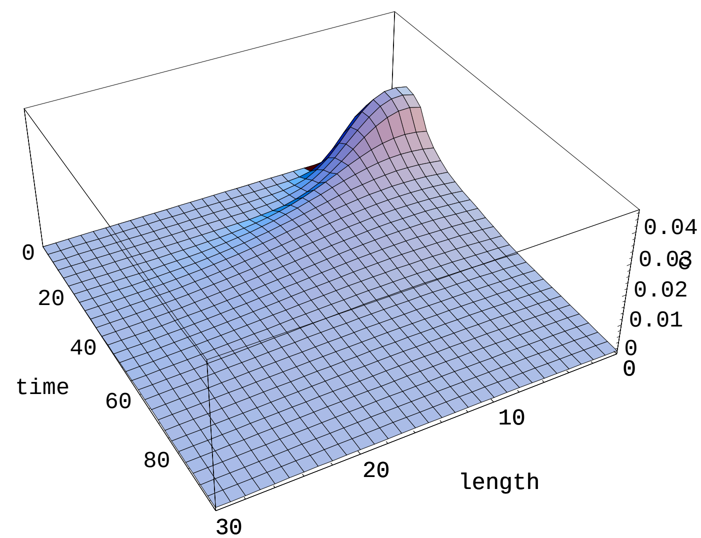
Parent



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)**