Effects of Temporal Error Correlation on Quantification of Predictive Uncertainty in Groundwater Reactive Transport Modeling

Dan Lu[1], Ming Ye[1,], Gary P. Curtis[2], Xiaoqing Shi[1], Xufeng Niu[3], Philip D. Meyer[4], and Steve B. Yabusaki[4]

[1]Department of Scientific Computing, [3]Department of Statistics, Florida State University; [2]USGS; [4]PNNL

INTRODUCTION

- Quantifying uncertainty of reactive transport simulations in groundwater can be conducted by using multiple conceptual models because groundwater flow and reactive processes are complex and subject to multiple interpretations.
- Consideration of alternative models results in broader but more realistic estimates of predictive uncertainty because the alternatives capture different plausible conceptual uncertainties.
- When quantifying predictive uncertainty it is important to realize the existence of model structural error besides measurement error because any alternative conceptual model is a simplification of reality.
- Model structural error is likely to present a high degree of temporal correlation for breakthrough data collected sequentially along time.
- It has been long recognized that the error correlation may affect parameter estimation and predictive uncertainty quantification.
- Methods for accurately describing the correlation structure of the errors and to incorporate it into groundwater reactive transport modeling is an open question.
- In conventional groundwater modeling, the errors are assumed to be multivariate Gaussian with zero mean and independent with a diagonal covariance matrix by considering only variances of measurement errors.
- This assumption has been found invalid in reactive transport modeling, and may lead to significant underestimation of predictive uncertainty.
- This is particularly true in multimodel analysis when alternative reactive transport models are considered. Use of a diagonal covariance matrix of the measurement errors in the calibration can cause one model to have an overwhelmingly high model probability (even 100%), which cannot be justified by the available data and knowledge.
- In this study, we developed a statistical method to identify the temporal correlation structure using time series theories.
- The method considers both measurement errors and model structural errors. Unlike the measurement errors, the model structural errors present a high degree of temporal correlation. Therefore, unlike the conventional assumption, the correlation structure of the total errors is characterized by a full covariance matrix instead of the diagonal one.
- The full covariance matrix is obtained by simulating the correlated errors with autoregressive models and is incorporated into groundwater modeling by an iterative method with two stages of parameter estimation.
- We applied this method to a set of synthetic and real-world surface complexation models developed to simulate uranium transport based on a series of column experiments.

THEORETICAL BACKGROUND

One popular method of multimodel analysis is model averaging. An averaged prediction is a weighted average of predictions produced by individual models M_k.

$$\hat{\bar{y}} = \sum_{k=1}^{K} w_k \hat{y}_k$$

$\hat{\bar{y}}$ Averaged prediction;
$\hat{y}_k$ Prediction of individual models;
w_k Averaging weight;

Predictive uncertainty is measured by a linear confidence interval, $\hat{y} \pm t_{1-\alpha/2} \times s_y$, where s_y is standard deviation of the prediction.

The variance of averaged prediction is:

$$Var(\hat{\bar{y}}) = \sum_{k=1}^{K} w_k Var(\hat{y}) + \sum_{k=1}^{K} w_k \left(\hat{y} - \hat{\bar{y}}\right)^2$$

where $Var(\hat{y})$ is predictive variance of individual model calculated by

$$Var(\hat{y}) = \mathbf{Z}^T(\mathbf{X}^T \mathbf{C}_\mathbf{e}^{-1} \mathbf{X})\mathbf{Z}$$

where **Z** is sensitivity matrix of predictions to model parameters, **X** is sensitivity matrix of observations to parameters; $\mathbf{C_e}$ is covariance matrix of error.

The averaging weight is usually estimated based on model selection criteria (*IC*).

$$w_k \approx \frac{\exp(-\Delta IC_k/2)}{\sum_{l=1}^{K}\exp(-\Delta IC_l/2)}$$

The commonly used model selection criteria are *AIC*, *AICc*, *BIC* and *KIC*. They have a common term called negative log likelihood function (*NLL*)

NLL, measure of model fit — N_k, measure of model complexity

$$AIC_k = -2\ln\left[L(\hat{\boldsymbol{\theta}}_k|\mathbf{D})\right] + 2N_k$$

$$AICc_k = -2\ln\left[L(\hat{\boldsymbol{\theta}}_k|\mathbf{D})\right] + 2N_k + \frac{2N_k(N_k+1)}{N-N_k-1}$$

$$BIC_k = -2\ln\left[L(\hat{\boldsymbol{\theta}}_k|\mathbf{D})\right] + N_k \ln N$$

$$KIC_k = -2\ln\left[L(\hat{\boldsymbol{\theta}}_k|\mathbf{D})\right] - 2\ln p(\hat{\boldsymbol{\theta}}_k) + N_k \ln(N/2\pi) + \ln|\bar{\mathbf{F}}_k|$$

For a alternative model $f(\boldsymbol{\beta}_k)$ with model structure error $\boldsymbol{\eta}_k$, a value of measurement data, **D**, collected sequentially along time with measurement error $\boldsymbol{\varepsilon}$ is expressed by

$$\mathbf{D} = f(\boldsymbol{\beta}_k) + \boldsymbol{\eta}_k + \boldsymbol{\varepsilon}$$

Assume sum of model structural error and measurement error, e_k, follow multivariate Gaussian distribution with covariance matrix, then

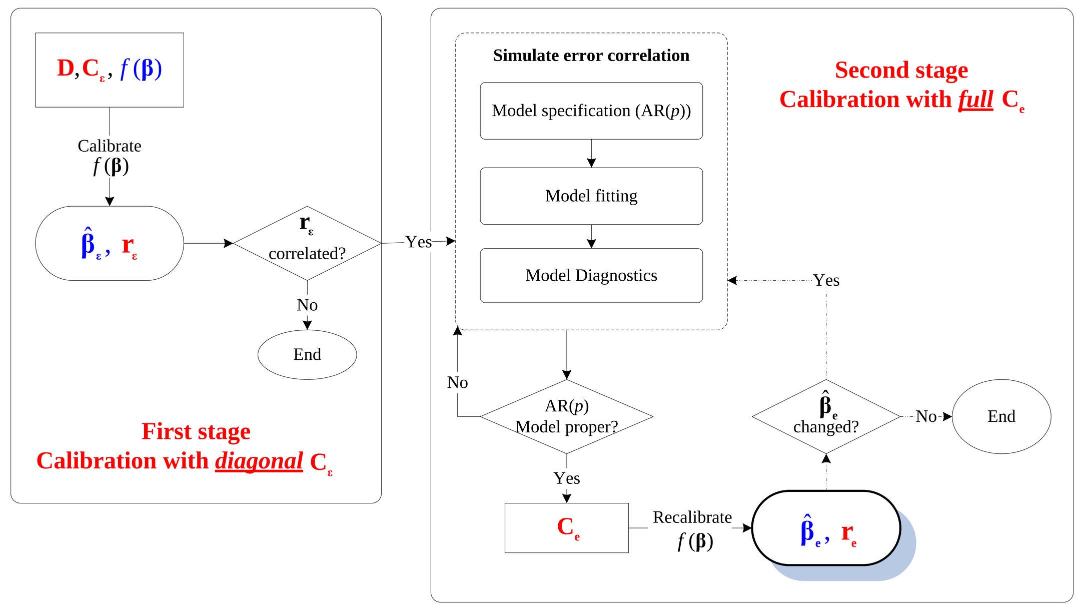
$$NLL = -2\ln\left[L(\boldsymbol{\theta}_k|\mathbf{D})\right] = N\ln(2\pi) + \ln\left|\mathbf{C}_{e_k}\right| + \mathbf{e}_k^T \mathbf{C}_{e_k}^{-1}\mathbf{e}_k$$

where C_{e_k} is a full covariance matrix, because model structural error is likely to show a high degree of temporal correlation for measurement data collected sequentially along time.

- In practice, the error correlation is generally disregarded and the diagonal covariance matrix of measurement error is usually used to evaluate the *NLL* in model selection criteria.
- The miscalculation in *NLL* misrepresents the information content of data, and may lead to incorrect estimation of model selection criteria, model averaging weights and averaged predictive performance.
- To correct the miscalculation it is necessary to reflect error correlation in model calibration.

ITERATED TWO-STAGE PARAMETER ESTIMATION

The full covariance matrix is obtained by simulating the correlated errors with autoregressive time series models (AR(*p*)) and is incorporated into groundwater modeling by an iterative method with two stages of parameter estimation.



CONCLUSIONS

❖ Disregarding error correlation, model uncertainty is underestimated, and predictive uncertainty bound is narrow hardly covering the true values.

❖ Considering error correlation, model averaging weights become more realistically and evenly distributed among the models and give better averaged predictive performance.

REACTIVE TRANSPORT MODELING

- *Kohler et al.* (1996) developed seven SCMs to simulate uranium transport based on eight column experiments data.
- This study considers four SCMs;
- True model consider reactions highlighted by red;
- Models are calibrated by Expt.1, 2, 8 with 120 data generated by true model.
- Predict Expt. 3

Surface Complexation Models (SCMs)

Model	Reactions	N_k
C3	$S_1OH+UO_2^{2+}+H_2O=S_1OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}=S_2OUO_2^+ +H^+$	3
C4	$S_1OH+UO_2^{2+}+H_2O=S_1OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}+H_2O=S_2OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}=S_2OUO_2^+ +H^+$	4
C5	$S_1OH+UO_2^{2+}+H_2O=S_1OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}+H_2O=S_2OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}=S_2OUO_2^+ +H^+$ $S_3OH+UO_2^{2+}+H_2O=S_3OUO_2OH+2H^+$	5
C6	$S_1OH+UO_2^{2+}+H_2O=S_1OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}+H_2O=S_2OUO_2OH+2H^+$ $S_2OH+UO_2^{2+}=S_2OUO_2^+ +H^+$ $S_3OH+UO_2^{2+}=S_3OUO_2^+ +H^+$	5

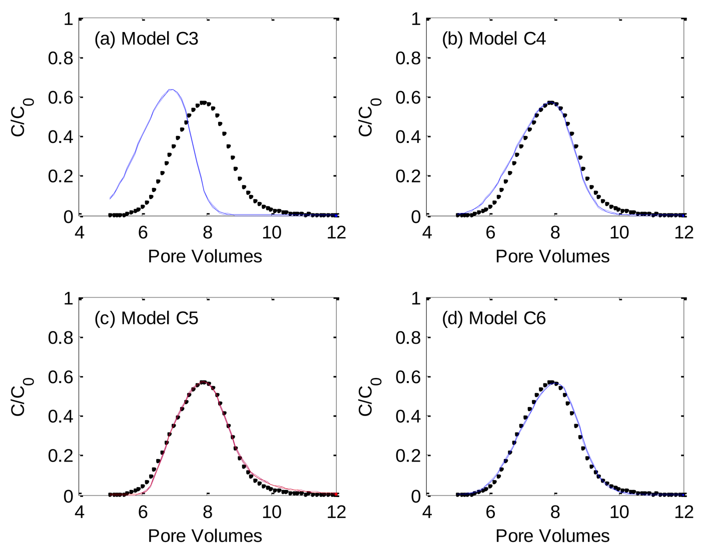
Two calibration cases:
Case I: Disregard error correlation; measurement error with standard deviation around 10^{-3};
Case II: Considering error correlation.

Averaging Weight and Predictive Performance

	Case I Disregarding error correlation				Case II Considering error correlation			
	C3	C4	C5	C6	C3	C4	C5	C6
w_{AICc} (%)	0.0	0.0	100.0	0.0	0.0	0.0	72.4	27.6
w_{KIC} (%)	0.0	0.0	100.0	0.0	0.0	0.0	71.5	28.5

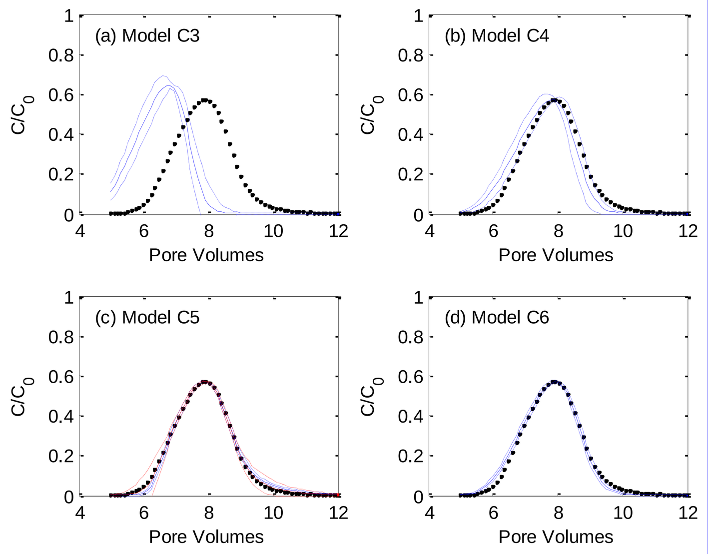
Case I

- Blue: Mean and 95% linear confidence intervals (CI) of single models
- Red: Mean and 95% CI of model averaging
- Uncertainty bounds are extremely narrow due to the small uncertainty of measurement errors.
- Predictive performance of model averaging is the same with C5.



Case II

- Uncertainty bounds of single models become larger due to consideration of error correlation.
- Uncertainty bounds of model averaging become even larger due to consideration of model uncertainty.



Coverage (%)	C3	C4	C5	C6	MLBMA
Case I	0.0	6.7	25.0	15.0	25.0
Case II	3.3	38.3	43.3	43.3	100.0