

Solute Transport Processes in Wetlands – Application of Data Based Mechanistic and Transient Storage Models

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Abstract

The aim of this paper is the analysis of solute transport processes in Upper Narew River based on the results of tracer experiment. Data Based Mechanistic and transient storage models were applied to Rhodamine WT tracer observations. We focus on the analysis of uncertainty and the sensitivity of model predictions to varying physical parameters, such as dispersion and channel geometry. The study is based on a combined Global Sensitivity Analysis (GSA) and Generalized Likelihood Uncertainty Estimation (GLUE). The breakthrough curves for the chosen cross-sections are compared with those simulated with 95% confidence bounds. Apart from the predictions of the pollutant transport trajectories, two ecological indicators are also studied (time over the threshold concentration and maximum concentration). These indicators show an interesting multi-modal dependence on model parameters.

1. Introduction

The present study has been motivated by the need to understand the dynamics of the spread of pollutants in a unique river system situated within the Narew National Park. The River Narew reach chosen for the study has recently been identified as an anastomosing river, which is regarded as a separate group to braided, meandering and straight rivers.

A widely accepted method to understand the fate of solutes in streams is to perform a tracer study, in which a known mass of usually conservative solutes is released into the stream. The study consists of a recording at downstream stations of concentration versus time curves of the artificially released dye and of fitting appropriate models.

The first model considered was advection-dispersion with dead zones that can adequately describe the process of transport of pollutants in a single-channel river with multiple storages. As an alternative to that transient storage model, the Data Based

Mechanistic (DBM) approach introduced by Young (1974) was tested. In this approach, the model is identified and the parameters are estimated from the collected time series data using system identification techniques (Young 1984). These techniques also provide estimates of the modelling errors and the uncertainties of the model parameters.

The applied transient storage model is deterministic; it assumes that observations are without errors and the model structure perfectly describes the process of transport of conservative pollutants. In order to take into account the model and observation errors, an uncertainty analysis is required. In this study we applied a combination of the Generalized Likelihood Uncertainty Estimation technique (GLUE) of Beven and Binley (1992) and the variance based Global Sensitivity Analysis (GSA). The combination is straightforward as the same samples (Sobol samples) were generated for GLUE analysis and for sensitivity assessment. Additionally, the results of the sensitivity analysis were used to specify the best parameter ranges and their prior distributions for the evaluation of predictive model uncertainty using the GLUE methodology.

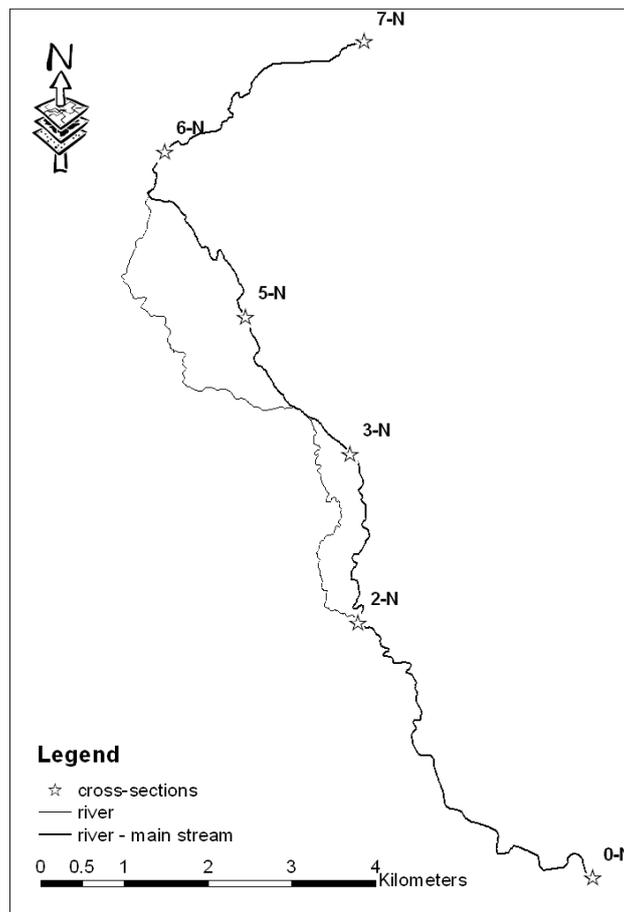


Fig. 1. Map of the experimental reach of upper River Narew.

2. Description of the experiment and case study

The present paper is based on a tracer test performed in a unique multi-channel system of the River Narew reach within the Narew National Park in northeast Poland (Fig. 1). A description of the experiment is presented in Rowiński *et al.* (2003a, b). The dye consisted of 20 liters of 20% solution Rhodamine WT injected at cross-section 0-N. Concentrations were measured in the River Narew at five transects, 2-N, 3-N, 5-N, 6N, and 7N, corresponding to flow distances of 5.75 km, 8.34 km, 10.62 km, 13.58 km, and 16.83 km, respectively. The dye was detected using the field fluorometer Turner Design with a continuous flow cuvette system. Water samples were also collected at sampling points.

3. Methodology

3.1 Applied models

Distributed transient storage model

The transport of a conservative soluble pollutant along a uniform channel is described by the well-known Advection-Dispersion Equation. To apply this equation to a practical scenario, a dispersion coefficient for the reach is required. Since a dispersion coefficient cannot be measured *in situ* directly from a simple individual measurement, it has to be estimated (optimized) on the basis of available experimental data. The One-dimensional Transport with Inflow and Storage (OTIS) model introduced by Bencala and Walters (1983) was applied in this study. The OTIS model is formed by writing mass balance equations for two conceptual areas: the stream channel and the storage zone. The stream channel is defined as that portion of the stream in which advection and dispersion are the dominant transport mechanisms. The storage zone is defined as the portion of the stream that contributes to transient storage, i.e., stagnant pockets of water and porous areas of the streambed. Water in the storage zone is considered immobile relative to water in the channel. The exchange of solute mass between the channel and the storage zone is modelled as a first-order mass transfer process. Conservation of mass for the stream channel and storage zone yields (Bencala and Walters 1983, Runkel and Broshears 1991):

$$\frac{\partial C}{\partial t} = -\frac{Q\partial C}{A\partial x} + \frac{1}{A} \frac{\partial}{\partial x} \left(AD \frac{\partial C}{\partial x} \right) + \alpha(C_S - C) + \frac{q_{LIN}}{A} (C_L - C) \quad (1)$$

$$\frac{dC_S}{dt} = \alpha \frac{A}{A_S} (C - C_S) \quad (2)$$

where: C is the solute concentration in the stream [g/m^3], t the time [s], Q is the flow discharge [m^3/s], A is the main channel cross-sectional area [m^2], x is the distance downstream [m], D is the coefficient of longitudinal dispersion [m^2/s], C_S is the concentration in the storage zone [g/m^3], α is the exchange coefficient [1/s], A_S is the storage zone cross-sectional area [m^2], q_{LIN} is the lateral volumetric inflow rate [m^2/s], and C_L is the solute concentration in lateral inflow [g/m^3].

Aggregated Dead Zone (ADZ) model

As an alternative to the transient storage model described by means of partial differential equations (Eqs. 1-2), a Data-Based Mechanistic (DBM) approach was introduced (Beer and Young 1983, Beven and Young 1998, Wallis 1989, Young and Lees 1993). In this approach, a so-called aggregated dead zone (lumped) model is formulated based on the observed time series data using system identification techniques (Young 1984). In the ADZ model the change of solute concentration in a river reach is described as:

$$C_{out_k} = \frac{B(z^{-1})}{A(z^{-1})} C_{in_{k-\delta}} \quad (3)$$

$$C_{obs_k} = C_{out_k} + \xi_k$$

where C_{in_k} is the concentration at the upstream end of the river reach at time k , the C_{out_k} is the estimated concentration at the downstream end of the river reach, C_{obs_k} is the measured concentration at the downstream end of the river reach, z^{-1} is the back-shift operator, δ is an advection time delay, A and B are the polynomials of the back-shift operator of the order m and n , respectively, and ξ_k represents the combined effect of all stochastic inputs to the system, including measurement noise.

$$B(z^{-1}) = b_0 + b_1 z^{-1} + b_2 z^{-2} + \dots + b_m z^{-m} \quad (4)$$

$$A(z^{-1}) = 1 + a_1 z^{-1} + a_2 z^{-2} + \dots + a_n z^{-n} \quad (5)$$

The order of the ADZ model describing the transport of solute in the river reach is described by triad $[n, m, \delta]$ and is determined in a statistical time series analysis technique using the recursive-iterative simplified, refined instrumental variable (SRIV) method (Young 1984) available in Captain Toolbox developed at the University of Lancaster. The best model structure is identified using three criteria. The first, coefficient of determination, R_r^2 , shows how much of the data variation is explained by the model output. The second measure is the Young Information Criterion (YIC), which is related to the fit and error on parameter estimates and takes into account the problem of over-parameterization. A low value of YIC indicates a well defined model. The third measure is the Akaike Information Criterion. AIC has a component related to the simulation fit but is penalized by the number of parameters in the model. A low value of AIC indicates a well defined model.

3.2 Sensitivity and uncertainty assessment

In the case of over-parameterized environmental models, a unique solution of the inverse problem is not available due to a limited amount and/or poor quality of the data. That results in equifinality problem, i.e. an application of different parameter sets leads to similar results. The Generalised Likelihood Uncertainty Estimation (GLUE) technique was introduced by Beven and Binley (1992) to deal with the problem of the non-uniqueness of the solution of environmental models. This technique is based on

multiple Monte Carlo runs of a deterministic model with parameters chosen randomly from a specified *a priori* distribution. The posterior distribution of the model predictions is obtained using a likelihood measure conditioned on observations. The choice of the likelihood measure should reflect the purpose of the study. There are many different likelihood measures based on the variance of errors. One of them is the Nash-Sutcliffe coefficient of determination. A second, often applied measure is inverse error measure with a shaping factor suggested by Box and Tiao (1992). This measure was applied by Beven and Binley (1992). An exponential transformation of sum of the square errors is also encountered in the literature (Romanowicz *et al.* 1994), where a formal definition GLUE was introduced.

In the case of measures based on error variance there are practical and theoretical limitations. They give unbiased and statistically efficient estimates of model parameters when errors between predictions and observations are normally distributed and they are not correlated. These assumptions are not often met in hydrological modelling but the approach is still used when violation of the assumptions is not large. In the present paper the non-formal GLUE approach presented by Romanowicz and Beven (2006) is applied. The likelihood function has the form of an exponent to the minus of the sum of square errors between simulated and observed concentrations, divided by the reduced (here multiplied by 0.1) mean error variance (Romanowicz and Beven 2006).

$$L(C|\theta) \sim \exp\left(-\sum_{t=1}^T (C_{OBS} - C_{SIM}(\theta))^2 / 0.1\sigma_T^2\right) \quad (6)$$

where C_{OBS} are the measured concentrations, C_{SIM} are computed concentrations, and σ_T^2 is the estimate of the variance of prediction error.

This likelihood measure was used to evaluate the posterior probability of model predictions and to define the 0.95 confidence bounds of these predictions.

Sensitivity analysis is “The study of how the uncertainty in the output of a model (numerical or otherwise) can be apportioned to different sources of uncertainty in the model input” (Saltelli *et al.*, 2004). In this work we have used the variance based Global Sensitivity Analysis approach introduced by Archer *et al.* (1997), described in more detail in Kiczko *et al.* (this issue). That approach does not require an assumption of additivity or monotonicity of model components. The variance of an output Y depending on the variable input set X_i , is based on estimating the fractional contribution of each input factor to the total variance $V(Y)$ of the model output Y .

The direct sensitivity of output Y to the input X_i represents the Sobol first order sensitivity index S_i , which takes the following form:

$$S_i = \frac{V[E(Y|X_i = x_i^*)]}{V(Y)} \quad (7)$$

where $V[E(Y|X_i = x_i^*)]$ is the variance of estimated output Y with X_i parameter fixed, and the other parameters varying, and E denotes expectation operator. Therefore, the

first order sensitivity index represents the average output variance reduction that can be achieved when X_i becomes fully known and is fixed.

The model sensitivity to the interactions among subsets of factors, so-called higher order effects, is investigated using the Sobol total sensitivity indices: S_{Ti} . They represent the whole range of interactions which involve X_i and are defined as:

$$S_{Ti} = \frac{E[V(Y|X_{-i} = x_{-i}^*)]}{V(Y)} \quad (8)$$

where $E[V(Y|X_{-i} = x_{-i}^*)]$ is the estimated variance in the case where all parameters are fixed, except X_i which is varying.

The use of total sensitivity indices is advantageous because there is no need for the evaluation of a single indicator for every possible parameter combination. On the basis of the two indices, S_i and S_{Ti} , it is possible to trace the significance of each model parameter in an efficient way.

4. Discussion of the results

4.1 OTIS

Since it is not possible to estimate solute transport parameters reliably from hydraulic variables and channel characteristics, an application of the transient storage model (Eqs. 1-2) requires an estimation of model parameters for each particular river reach, 2N-3N, 3N-5N, 5N-6N and 6N-7N (Fig. 1), based on data from tracer experiment including measurements of lateral inflow and discharge. Estimation of model parameters, namely D , A , A_s and α , was performed by minimizing the residuals between the simulated and observed concentrations. A general least squares objective function and the Nelder-Mead minimization algorithm were used in this study. The results of the estimation procedure are given in Table 1. They are analogous to that obtained by Rowiński *et al.* (2004) for a similar model but a different numerical scheme.

Table 1
Estimated parameters of transient storage models

Parameters	Sections			
	2N-3N	3N-5N	5N-6N	6N-7N
D [m ² /s]	10.31	1.65	6.96	1.59
A [m ²]	9.71	34.70	11.29	25.02
A_s [m ²]	6.13	22.62	4.46	7.05
α [1/s]	4.82e-006	1.7863e-005	1.2913e-005	6.5701e-005

Note that values of the parameters differ between reaches. These big differences result from a high variability of geometric and hydraulic conditions between reaches.

A comparison of observed and simulated data together with 0.95 confidence bounds obtained from the application of GLUE technique is presented in Fig. 2.

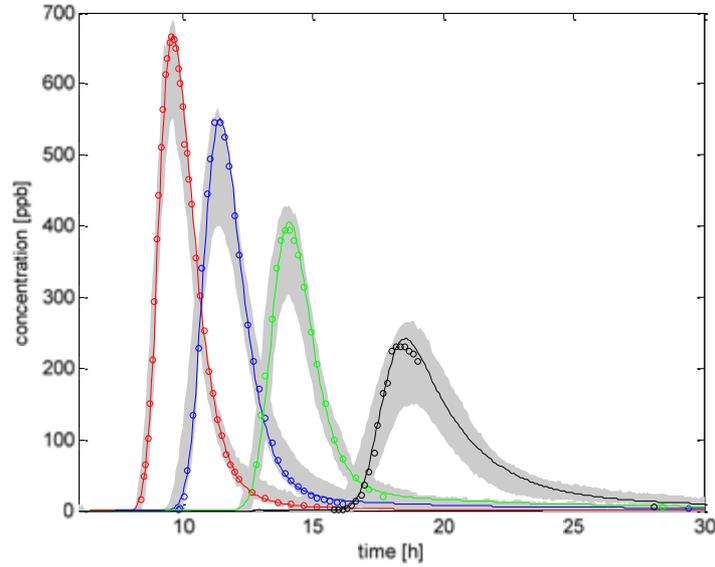


Fig. 2. Comparison of observed (dots) and simulated (solid line) concentrations of Rhodamine WT at cross-section 3N, 5N, 6N and 7N with 95% confidence bounds shown as shaded areas.

4.2 ADZ

Identification of model structure and estimations of parameters of the transfer functions models were conducted independently for every river section using the SRIV method of recursive estimation in the Captain toolbox. The values of coefficient of determination (R_T^2), Young Information Criterion (YIC) and Akaike Information Criterion (AIC) for all analyzed cross-sections are given in Table 2. Analysis shows that the second-order models are the most parametrically efficient model structures, accurately describing the observed solute transport in these reaches.

Table 2

Results of identification of Aggregated Dead Zone model

	Sections			
	2N-3N	3N-5N	5N-6N	6N-7N
Model $[n, m, \delta]$	[2, 2, 29]	[2, 2, 19]	[2, 2, 30]	[2, 2, 50]
R_T^2	0.9996	0.9994	0.9970	0.9934
YIC	-17.454	-18.476	-17.455	-6.201
AIC	0.854	1.017	1.032	3.084

The second order models can be decomposed into a parallel connection of two first order ADZ transfer functions in the following form:

$$\begin{aligned} \text{Fast Component: } y_{q,k} &= \frac{\beta_q}{1 + \alpha_q z^{-1}} C_{in_{k-\delta}} \\ \text{Slow Component: } y_{s,k} &= \frac{\beta_s}{1 + \alpha_s z^{-1}} C_{in_{k-\delta}} \end{aligned} \quad (9)$$

where $\alpha_s, \alpha_q, \beta_s, \beta_q$ are parameters derived from (3) and the concentration is the sum of these two components and a model error, i.e. $C_{out_k} = y_{s,k} + y_{q,k} + \xi_k$. The associated residence times (time constants) (T_s, T_q), steady state gains (G_s, G_q), and partition percentages (P_s, P_q) are given by the following expressions:

$$T_i = \frac{\Delta t}{\log_e(\alpha_i)}; i = s, q \quad G_i = \frac{\beta_i}{1 + \alpha_i}; i = s, q \quad P_i = \frac{100G_i}{G_s + G_q}; i = s, q$$

In practice, the residence times of two parallel connections are of quite different magnitude and they are denoted as quick and slow processes in the above equation (subscript q and s respectively). The most plausible physical explanation is that the pure time delay allows for the flow-induced pure advection of the solute. The quick-flow parallel pathway then represents the ‘main stream-flow’ that is relatively unhindered by vegetation, while the slow-flow pathway represents the solute that is captured by heavy vegetation and so dispersed more widely and slowly before rejoining the main flow and eventually reaching the main channel. The resulting residence times, steady state gains and partition percentages for all analysed reaches are given in Table 3.

Table 3

Parameters of slow and fast components of ADZ models

	Sections			
	2N-3N	3N-5N	5N-6N	6N-7N
T_s [h]	0.27	13.76	13.35	1.54
T_q [h]	0.11	0.28	0.27	0.06
G_s	0.7381	0.1225	0.1003	0.6596
G_q	0.2209	0.8896	0.7698	0.2494
P_s [%]	76.97	12.11	11.534	72.56
P_q [%]	23.03	87.89	88.47	27.44

Using calibrated transfer function models, a SIMULINK system describing the transport of solutes in the Upper Narew was built. A block diagram of a full semi-

distributed system is presented in Fig. 3. Every section is modelled as a second-order transfer function model; the simulated output from a previous section becomes an input to the next section.

Figure 4 presents a comparison of measured concentrations of Rhodamine WT and those simulated using the ADZ model at four cross-sections. In the case of the cross-sections 3N, 5N and 6N, a very good fit is observed. The worst results are for cross-section 7N, caused by a gap in measurements which makes the correct identification of model parameters impossible.

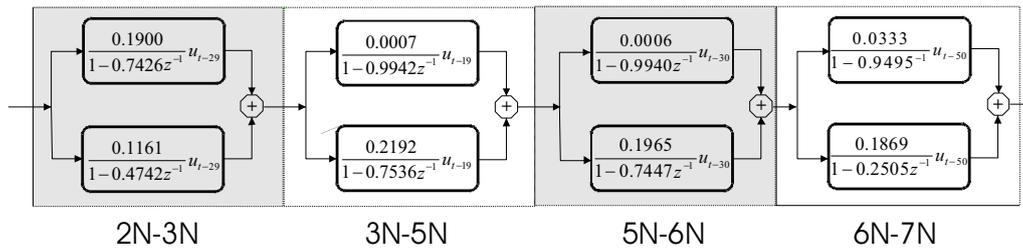


Fig. 3. Transfer function block diagram of the whole analyzed river reach model.

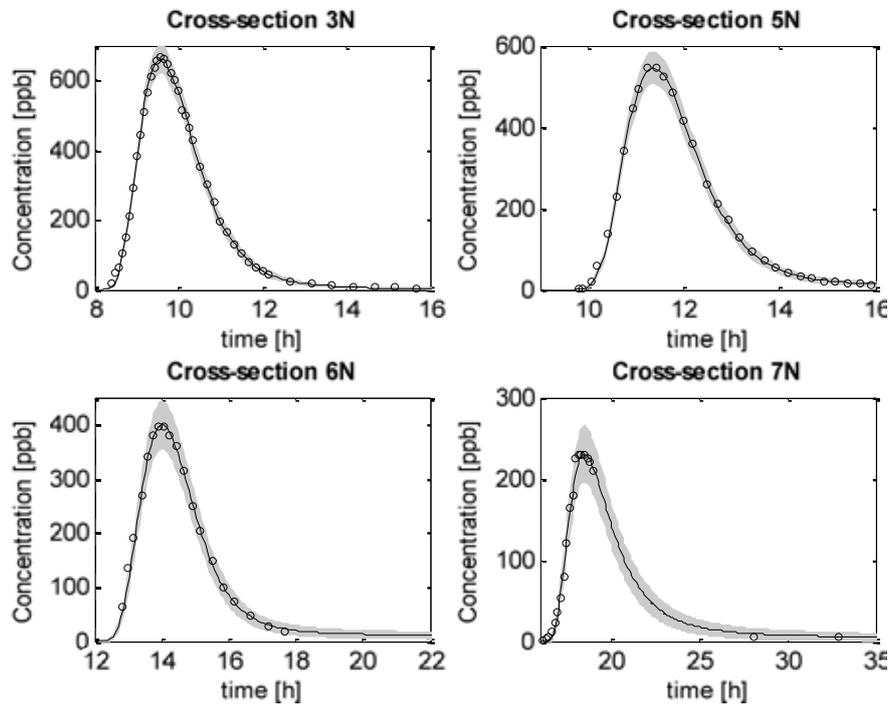


Fig. 4. ADZ model results – application to the whole river reach; open circles represent measurements and shaded areas denote 95% confidence bounds.

4.3 Comparison of ADZ and mechanistic modelling (virtual reality)

To validate the ADZ model for the whole river reach, another tracer experiment is required. Unfortunately, there was just one tracer test in this part of the river, so we used the mechanistic OTIS model to simulate *virtual reality describing solute transport* in the Upper Narew for a different input than that used in the calibration. The results were then compared with those obtained by means of the previously calibrated ADZ model. They are depicted in Fig. 5.

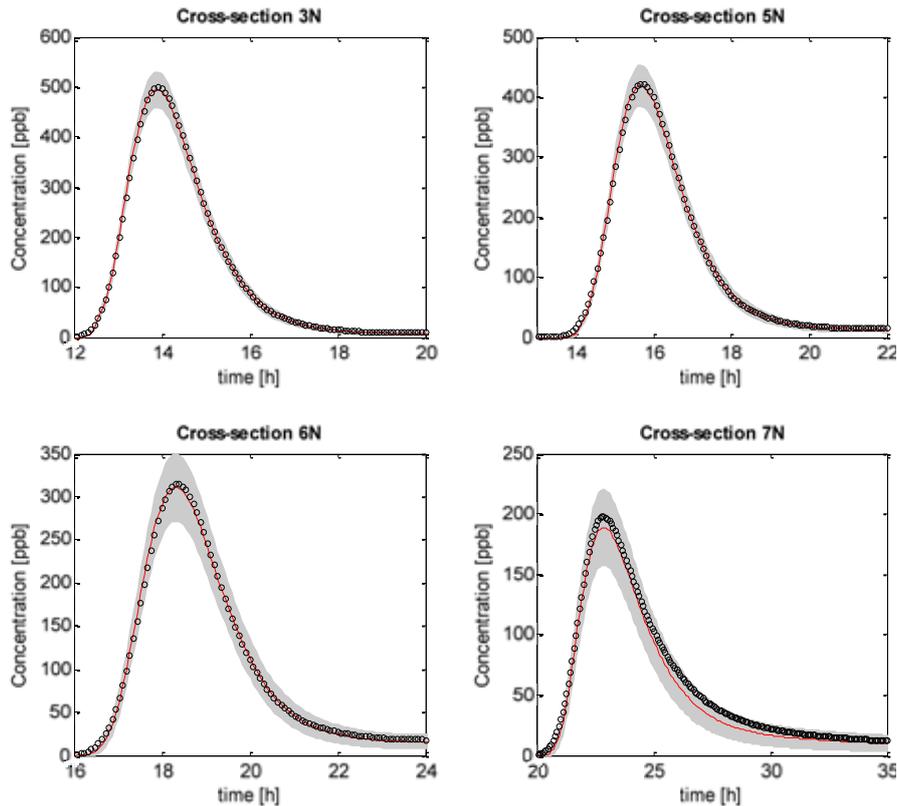


Fig. 5. Validation of active mixing volume model. Open circles represent virtual reality simulated by mechanistic model, red solid line – ADZ model; shaded areas show 95% confidence bounds obtained from the ADZ model.

The results show a good similarity, with R_T^2 equal to 0.9995, 0.9994, 0.9991 and 0.9938 for cross-sections 3, 5, 6, and 7, respectively.

4.4 Sensitivity analysis

In this paper we have analyzed the sensitivity of model output (simulated concentrations and ecologically related measures) to model parameters and model input. The parameters of the model were sampled from a uniform distribution with lower and upper bounds based on a physically justified variation of these parameters. These

bounds are: coefficient of longitudinal dispersion (D) 0.01-20 [m^2/s], the storage zone cross-sectional area (A_S) 0.01-30 [m^2], the exchange coefficient (α) 0.0000001-0.0001 [$1/\text{s}$], the main channel cross-sectional area (A) 5-45 [m^2], and $\pm 10\%$ errors of the measured model input (described as a multiplier in range from 0.9 to 1.1).

Sobol first order and Sobol total order sensitivity indices for the parameters of the OTIS model predictions and uncertainty of observations are shown in Fig. 6. The main channel cross-sectional area (A) has the largest influence on the output of all analyzed river reaches. The exchange coefficient (α), the storage zone area (A_S) and the dispersion coefficient (D) have smaller values of indices, indicating a smaller influence on the model output and a smaller identifiability of these parameters. The order of these three parameters varies in different river reaches. For the three last reaches the longitudinal dispersion coefficient has higher values of sensitivity indices than parameters describing the effect of dead zones. The lowest values of Sobol first and total sensitivity indices are obtained for $\pm 10\%$ variation of the model input.

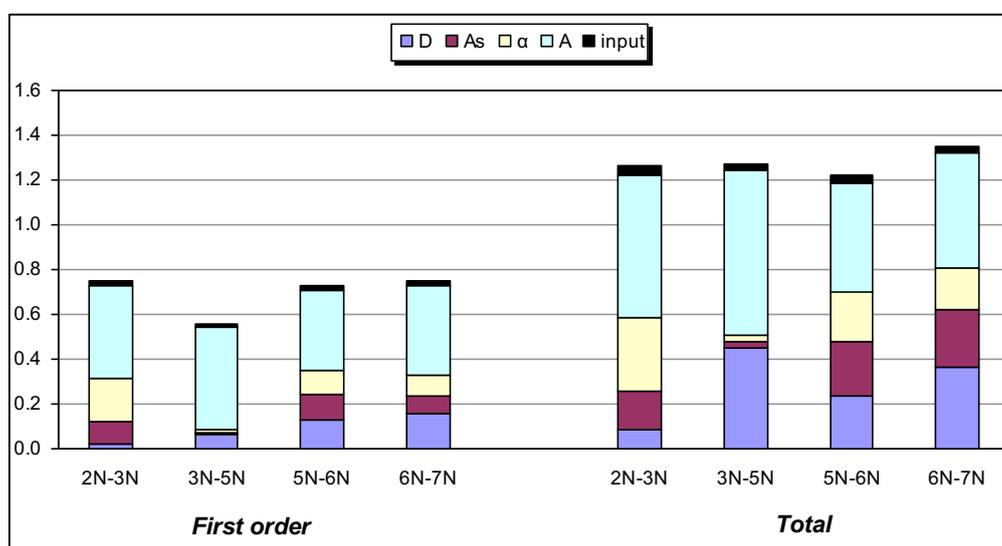


Fig. 6. Sobol first order and total sensitivity indices for OTIS model predictions for all cross-sections.

The sum of first order sensitivity indices for the analyzed river reaches is less than 1, which indicates a non-additive model.

5. Ecologically related criterion

The richness and uniqueness of the flora and fauna in the Narew National Park were the reasons for including ecologically related measures in our sensitivity analysis. The first is the maximum value of concentration, the second the length of time periods with the concentration over a specified threshold. These two variables are used in toxicological assessments.

Sensitivity indices for the maximum concentration of the tracer in relation to the model parameters are shown in Table 4 and Fig. 7. The values of the sensitivity indices are similar for all four analyzed river reaches and they resemble the results obtained for model predictions. There is a relationship between the area of the main channel and the values of maximum concentration.

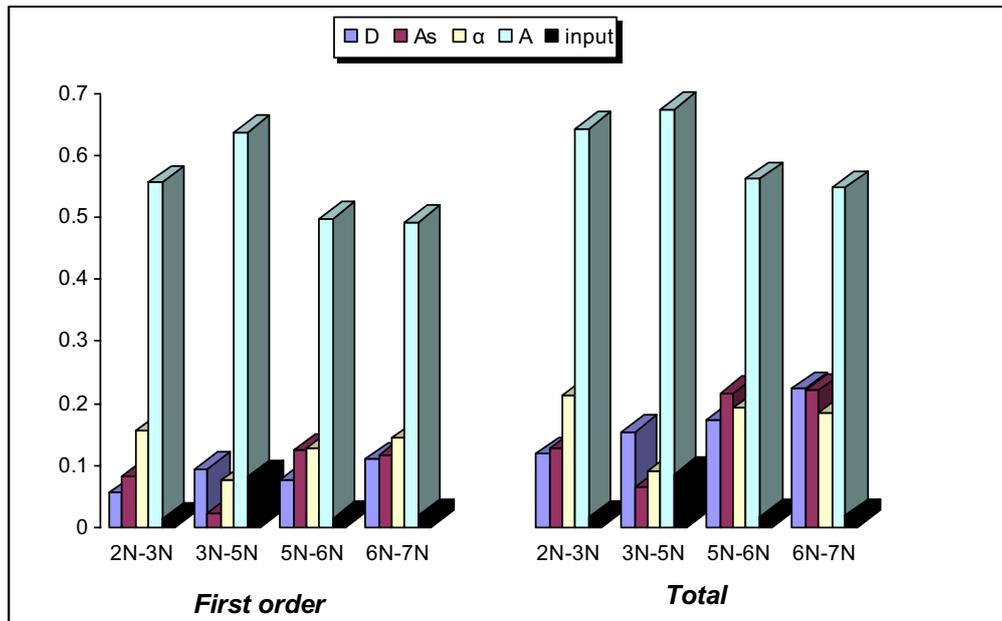


Fig. 7. Sobol first order and total sensitivity indices for OTIS model predictions on maximum concentration for all cross-sections.

Table 4

Results of the GSA sensitivity analysis on maximum concentration

River reach	S_i					S_{Ti}				
	D	A_S	α	A	input	D	A_S	α	A	input
N2-N3	0.0555	0.0827	0.1560	0.5556	0.0133	0.1187	0.1266	0.2143	0.6425	0.0166
N3-N5	0.0940	0.0227	0.0755	0.6372	0.0816	0.1542	0.0645	0.0909	0.6739	0.0857
N5-N6	0.0779	0.1236	0.1280	0.4982	0.0146	0.1744	0.2169	0.1922	0.5614	0.0183
N6-N7	0.1100	0.1178	0.1437	0.4921	0.0190	0.2238	0.2208	0.1854	0.5470	0.0194

The highest values of indices are for the second river reach (0.6372 and 0.6739 for first and total order respectively) and the lowest are for the last river reach (0.4921 and 0.5470 for the first and total order respectively). In the case of the exchange coefficient and the storage zone area this relationship is weaker (Fig. 8). The dispersion coefficient shows a small influence on maximum concentrations. Values of sensitivity indices for this parameter vary between 0.0555 and 0.1100 for first order and between

0.1187 and 0.2238 for total effect. The lowest values of sensitivity indices are obtained for a 10% input measurements error.

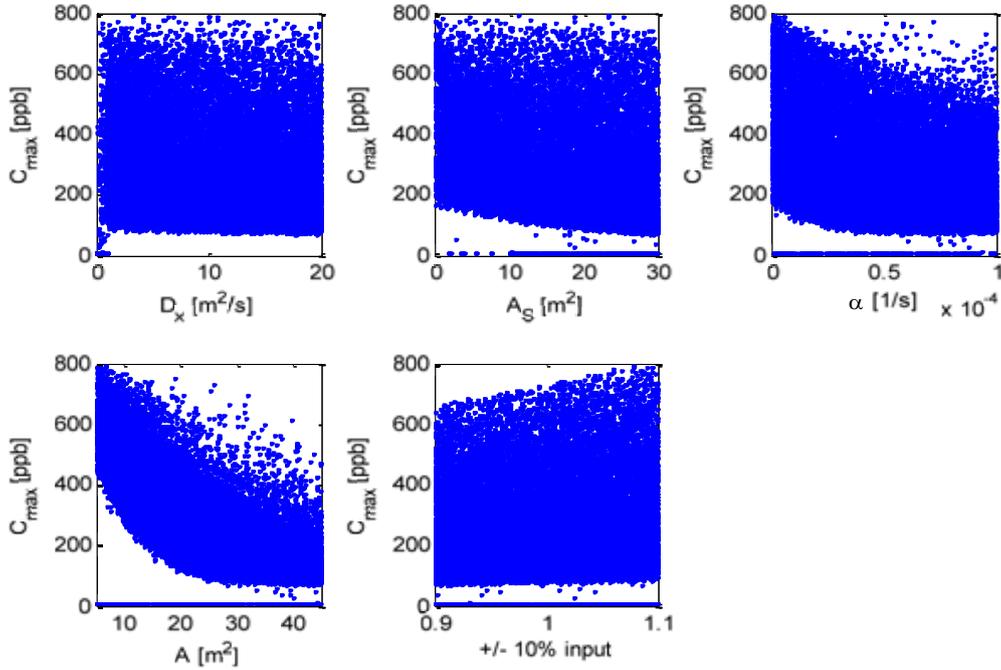


Fig. 8. Relationship between values of maximum simulated concentration and values of parameters for river reach 2N-3N.

Results of the sensitivity analysis for the “over the threshold” period depend on the threshold value. Figure 9 presents the first order and total sensitivity indices of the OTIS parameter variations as a function of the threshold value. It is interesting to note that the sensitivity of the “over the threshold” period for small and large threshold values shows different behaviour, shown in Fig. 9 as multiple minima/maxima. This behaviour is the result of two different processes. One is the direct influence of parameters on different parts of the dynamic response of the system. The other is the dependence of the maximum peak concentration at each cross-section on the parameter values, i.e. for high threshold values, the number of realisations with a non-zero “over the threshold” period decreases.

In order to explain this behaviour, we shall analyse the projections of the response surface for the parameter A_S for four different values of the threshold, 10, 60, 100 and 200 ppb, (Fig. 10 a-d). For small threshold values (Fig. 10a), the storage zone area influences the number of time periods over the threshold due to its influence on the tails of the dynamic response of the system (Wagener *et al.* 2002). This influence decreases with an increase of the threshold value, resulting in the minimum index value at a threshold of about 60 ppb (Fig. 10b). Above this threshold, due to the dependence of maximum concentration on the storage zone area A_S for higher values of this para-

meter, there is an increasing number of realizations for which the threshold concentration of 100 ppb is not reached (Fig. 10c).

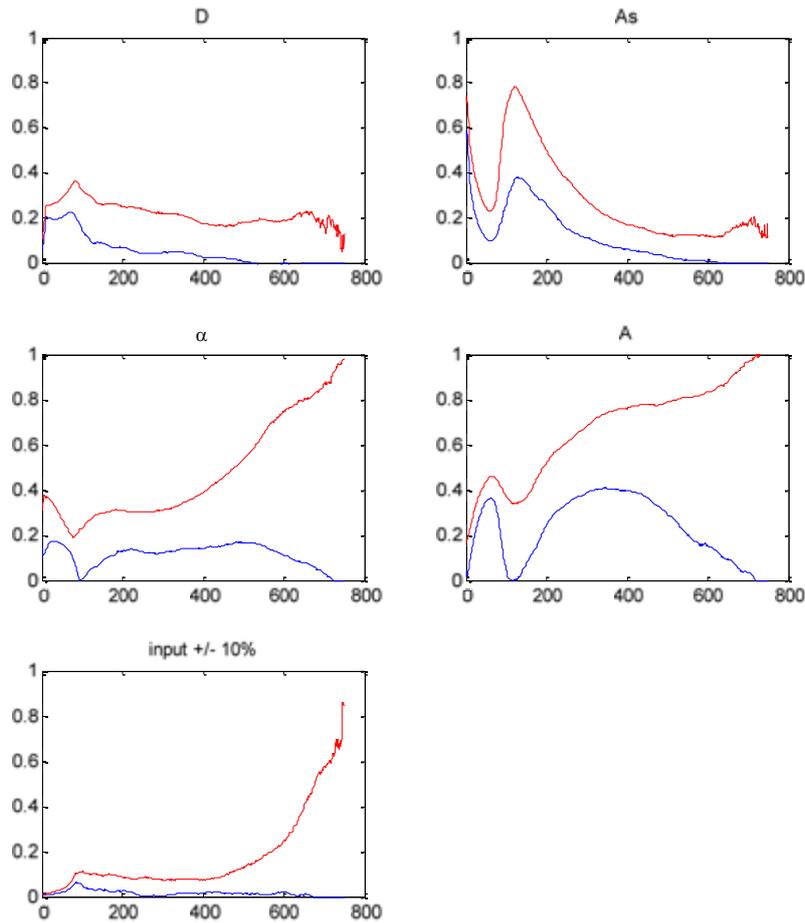


Fig. 9. Sensitivity indices for “over the threshold” period to OTIS parameters variations as a function of the threshold value for river reach 2N-3N. Blue and red lines denote Sobol first order and total sensitivity indices, respectively.

As a result, the number of realizations with decreasing or equal to zero “over the threshold” periods increases, giving a rise of the sensitivity index for this parameter. With a further increase in the threshold value, the number of realizations with “over the threshold” period stabilizes, as there are zero-length “over the threshold” periods over the whole parameter range (Fig. 10d). It is interesting to note nearly opposite relationship for parameter A (main channel cross-sectional area), shown in detail in Fig. 11 a-d for threshold values equal to 10, 60, 100 and 200 ppb, respectively. This parameter influences the higher parts of the dynamic response of the system, giving a rise of the sensitivity index with an increase of the threshold value (Fig. 11 a and b). With a further increase in the threshold values, zero periods appear that counteract the increase of the number of over the threshold periods, thus decreasing the sensitivity

index (Fig. 11c). This influence is limited to the higher values of that parameter, which results in a subsequent rise of the sensitivity index for values of the threshold higher than 100 ppb (Fig. 11d).

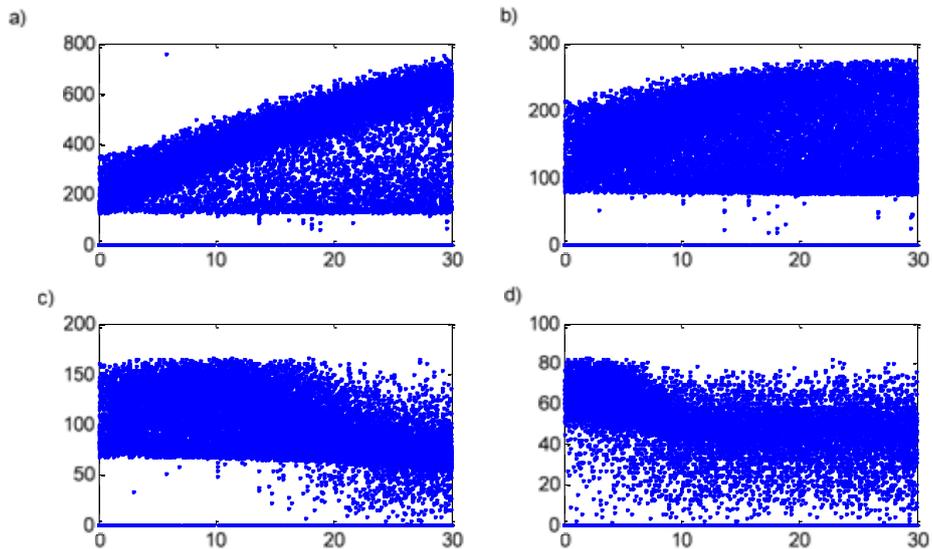


Fig. 10. Sensitivity analyses for the “over the threshold” period for the 2N-3N river reach. Dotted plots a, b, c and d show the projection of the response surface (number of time steps with concentration over the threshold) into the parameter A_S dimension for four threshold values: 10, 60, 100 and 200 ppb, respectively.

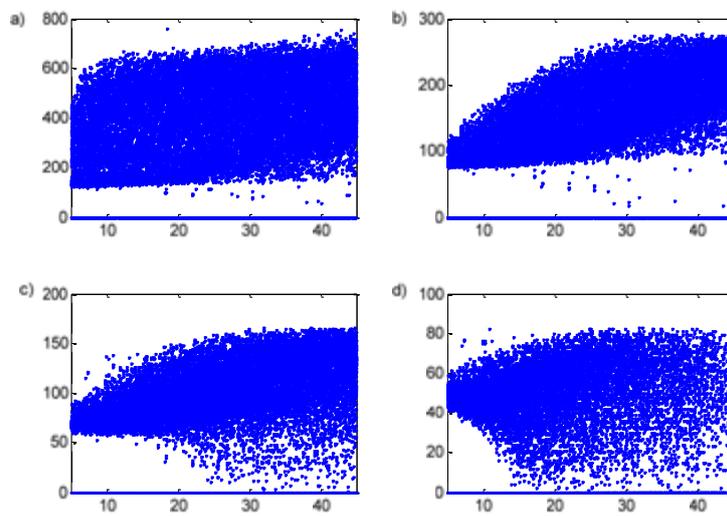


Fig. 11. Sensitivity analyses for the “over the threshold” period for the 2N-3N river reach. Dotted plots a, b, c and d show the projection of the response surface (number of time steps with concentration over the threshold) into the parameter A dimension for four threshold values: 10, 60, 100 and 200 ppb, respectively.

6. Conclusions

We have presented the application of a deterministic, mechanistic model (OTIS) and a stochastic ADZ model to describe dispersion processes in wetlands, based on tracer experiment data from the reach of the River Narew within the Narew National Park. We applied a sensitivity analysis using GSA (Saltelli *et al.* 2004) to define the most sensitive parameters and their ranges. Maximum concentrations and the length of time period with concentrations exceeding the specified threshold were used as ecologically related model outputs together with the concentration trajectories. In particular, the sensitivity analysis of the length of "over the threshold" period shows an interesting relationship for threshold values below 200 ppb. The results of this analysis were used to specify the best parameter ranges and their prior distributions for the evaluation of predictive model uncertainty, using the GLUE technique of Beven and Binley (1992). The stochastic ADZ model was used as an alternative to the mechanistic model. Due to its stochastic nature, the uncertainty of the model predictions is included in the model output. The dynamics of the dispersion process identified by the ADZ model are of second order, indicating the existence of slow and fast dispersion components in the studied River Narew reach. Due to the lack of a validation data set, we applied the mechanistic OTIS model output for a time period different from that used in the calibration stage, as a virtual reality, in order to validate the ADZ model. The results show a good fit with 99% of the variation of the OTIS model output explained.

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