Modelling of biofilters for ammonium reduction in combined sewer overflow

M. Henrichs, A. Welker and M. Uhl

ABSTRACT

Biofiltration has proved to be a useful system to treat combined sewer overflow (CSO). The study presented uses numerical simulation to detect the critical operating conditions of the filter. The multi-component reactive transport module CW2D was used for the simulation study. Single-event simulations of lab-scale-column experiments with varying boundary conditions regarding the throttle outflow rate were carried out. For the calibration of the CW2D model measurement results of four experiments in two lab-scale columns were used. The model was validated by simulating four events of two further columns filled with the same filter material. These columns were operating with higher throttle outflow rates than the columns used for calibration. For ammonium (NH_4-N) a good fit between measured and simulated data could be achieved. However, the comparison of simulated and measured effluent concentrations of nitrate (NO_3-N) showed that there is a need for further investigations mainly due to the uncertainties in the degradation process during dry periods between the loadings.

Key words | ammonium, biofilters, CSO, models, reactive transport simulation

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INTRODUCTION

Biofiltration approved to be a useful technology for decreasing the impact of combined sewer overflows (CSOs) on receiving waters. Main objectives for enhanced CSO treatment by biofiltration are (1) reduction of suspended solids and associated substances by filtration, (2) reduction of soluble pollutants by sorption and subsequent biological degradation and (3) detention and reduction of peak flows (e.g. Uhl & Dittmer 2005). The combination of the essential sedimentation tank and biofiltration unit is also termed as subsurface vertical flow constructed wetlands or retention soil filter (Dittmer *et al.* 2005). The stochastic nature of rainfall leads to extreme conditions characterized by long inundation times after runoff events and partly very long dry periods in between. Soluble and particulate pollutants are sorbed or filtrated during an inundation event and degraded in dry periods under aerobic conditions. The filter materials as well as the filter velocity influence the elimination of degradable pollutants like COD, ammonium and phosphorous (Dittmer 2006; Henrichs *et al.* 2006; Wozniak 2006). To avoid acidic conditions a minimum content of carbonate in the filter material is required (Uhl & Dittmer 2005).

This study focuses on ammonium elimination by biofiltration. Due to the possible fish toxicity when converting into ammonia, ammonium proved to be one of the most important parameters in CSO.

Numerical simulation will be used to increase the understanding of the coupled processes sorption and degradation which have been regarded to be the important processes during ammonium elimination. The main objectives of this study are the detection of critical operating conditions of the filter regarding (i) maximum inundation times without re-aeration, (ii) maximum ammonium loads before a breakthrough takes place and (iii) first estimates on the regeneration time a filter needs for degradation of the sorbed pollutants under aerobic conditions.

doi: 10.2166/wst.2009.397

METHODS

Experimental setup

For the simulation lab-scale column (diameter 0.2 m) experiments were chosen. Columns were filled with a 0.75 m sand layer (0–2 mm) above a drainage layer of 0.25 m fine and coarse gravel (2–8 mm and 8–16 mm). A 5 cm layer of gravel (2–8 mm) above the sand protects its surface. Figure 1 shows a photograph of the experimental setup. Peristaltic pumps were used to control each outflow with constant rates between 2×10^{-5}–4×10^{-5} $m^3\,s^{-1}\,m^{-2}$. The column experiments were carried out as batch loadings with a hydraulic load of $0.5\,m^3\,m^{-2}$ and $2.5\,m^3\,m^{-2}$, respectively. For the simulations only experiments with a hydraulic load of $2.5\,m^3\,m^{-2}$ were used. The regeneration time between two experiments was one week.

The columns were loaded with artificial CSO with constant inflow concentrations for COD ($60\,mg\,L^{-1}$) and NH_4-N ($6\,mg\,L^{-1}$). After a first phase with several loadings for inoculation, the outflow of the next 2 loadings was observed. The samples were taken as 2 h composite sample (7 samples per loading). For the simulations the four columns were filled with the same filter material. Because of degradation processes in the supernatant water level only events with inflow and infiltrate samples were measured and considered in the simulation. The infiltrate concentration varied for ammonium nitrogen (NH_4-N) between 4.7–5.4 $mg\,L^{-1}$ and for COD between 29.0–50.0 $mg\,L^{-1}$. The nitrate nitrogen (NO_3-N) concentrations were near the detection limit of $0.2\,mg\,L^{-1}$. The two columns used for calibration were throttled to a filter velocity of $2 \times 10^{-5}\,m\,s^{-1}$ and the two columns used for validating had a filter velocity of $3 \times 10^{-5}\,m\,s^{-1}$ and $4 \times 10^{-5}\,m\,s^{-1}$, respectively.

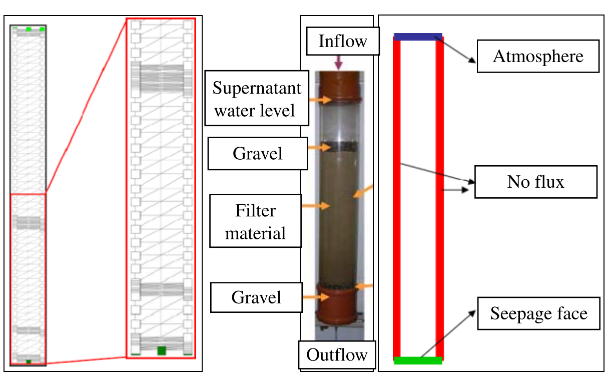

Figure 1 | Visualization of the finite element mesh (left), a photo of the lab-scale columns (middle), and the corresponding boundary conditions of a model (right).

Tracer experiment

After saturation of the soil column a tracer (uranin) was added to the supernatant water level above the surface. The effluent concentrations of uranin were measured by optical fibres technology (MKT 2 from Sommer Measurement and System techniques).

The multi-component reactive transport module CW2D

The multi-component reactive transport module CW2D (Langergraber 2001) was applied to model the behaviour of the experiments. CW2D was developed to model transport and reactions of the main constituents of municipal wastewater in subsurface flow constructed wetlands and is able to describe the biochemical elimination and the transformation processes for organic matter, nitrogen and phosphorus. Organic matter is modelled by separation in three fractions: Readily degradable (CR), slowly degradable (CS) and inert (CI). CW2D is incorporated into the HYDRUS-2D variably-saturated water flow and solute transport model (Langergraber & Simunek 2005). The unsaturated hydraulic properties are modelled using the soil hydraulic functions of van Genuchten (1980).

CW2D considers two types of bacteria: heterotrophic and autotrophic bacteria. Heterotrophic bacteria (XH) are modelled as all-rounder bacteria responsible for hydrolysis, aerobic degradation of organic matter and denitrification. Autotrophic bacteria (2 species, XANs and XANb) are responsible for the two-step nitrification process (Langergraber 2001).

CW2D/HYDRUS-2D had to be modified to model a throttled effluent (constant filter velocity) and impoundage for considering storage volume (Dittmer *et al.* 2005; Henrichs *et al.* 2007). For simulation of CSO treatment by biofiltration the standard CW2D parameters are used. In addition sorption for COD was introduced. The standard CW2D parameter set (Langergraber 2001) considers sorption only for ammonium and inorganic phosphorous.

According to Simunek *et al.* (1999) adsorption in HYDRUS-2D is described by

$$s_i = \frac{k_{s,i} \cdot c_i^{\beta_i}}{1 + \eta_i \cdot c_i^{\beta_i}} \quad (1)$$

where $i = 1, \ldots, N$; N = total number of components; c_i = concentration in the aqueous phase [$\mathrm{mg_i\,dm_w^{-3}}$]; s_i = concentration in the solid phase [$\mathrm{mg_i\,kg_s^{-1}}$]; and $k_{s,i}$, β_i, η_i = empirical coefficients. With $\beta_i = 1$ Equation (1) becomes the Langmuir equation, when $\eta_i = 0$ it becomes the Freundlich equation, and when both $\beta_i = 1$ and $\eta_i = 0$ Equation (1) is a linear sorption isotherm. Sorption of COD is modelled by a chemical non-equilibrium adsorption of CS (slowly biodegradable COD) with a linear sorption isotherm (i.e. in Equation (2) the parameter $\beta = 1$ and $\eta = 0$).

The time dependency of adsorption is described by the concept of two-site sorption. Sorption is instantaneous on one part of the exchange sites whereas on the remaining sites it is considered to be time-dependent. The two-site sorption is described by (Simunek *et al.* 1999)

$$\frac{ds_i^k}{dt} = \omega_i \cdot \left[(1 - f) \cdot \frac{k_{s,i} \cdot c_i^{\beta_i}}{1 + \eta_i \cdot c_i^{\beta_i}} - s_i^k \right] \quad (2)$$

where s_i^k = sorbed concentration of component i at the fraction of sites assumed to be time-dependent [$\mathrm{mg_i\,kg_s^{-1}}$]; ϖi = first-order exchange rate for component i [$\mathrm{h^{-1}}$]; and f = fraction of exchange sites assumed to be in equilibrium with the solute phase [−]. It is assumed that all sorption sites are time-dependent, i.e. $f = 0$.

Simulation approach

According to the objectives of this study a calibrated and validated model is needed. The calibrated parameter set should be validated by using a split sample test. This means that the parameters are transferred to other data sets with (i) the same boundary conditions or (ii) different boundary conditions concerning influent concentrations, loadings and filter velocities.

Calibration and validation were run in a stepwise approach as follows:

1. Calibration for a single-event with measurements from the column experiments
   (a) Calibration of the flow and single-solute transport model using measured data from tracer experiments.
   (b) Calibration of CW2D using measured effluent concentrations of two single-events from two columns.
2. Validation of the calibrated parameter set for four other single-events with different filter velocities.
3. Use of calibrated and validated model for prediction purposes.

RESULTS

Model setup and boundary conditions

For the simulations a 2D finite element mesh with 282 nodes und 372 elements was generated (Figure 1, left). The vertical discretization had to be very fine because of mainly vertical sorption and degradation processes. The supernatant water level (storage volume) was modelled by a virtual layer with a pore volume of 100% and a residual water content of 0%. On the top of the virtual layer an atmospheric boundary condition was applied (see Figure 1, right). At the bottom of the drainage layer a throttled boundary condition that is based on a seepage face boundary with maximal outflow and minimal pressure head was used. For the simulations the maximal outflow was set to 5×10^{-5}–$1 \times 10^{-5}\,\mathrm{m\,s^{-1}}$, and the minimal pressure head was set to minus 1 m.

Table 1 | Input and calibrated model data for the flow and transport model

	Residual soil water content (−)	Saturated soil water content (−)	Longitudinal dispersivity (m)	Saturated hydraulic conductivity (m/s)
Model input data	0.1275	0.352	0.06	8.745
Calibrated model data	0.1343	0.355	0.048	8.64

Calibration of the flow and transport model

The hydraulic model was calibrated by comparison of the hydrographs of the simulated and measured tracer effluent concentrations. The soil parameters for flow and transport were altered in an iterative process until a good match between simulated and measured effluent concentrations was archieved.

Table 1 lists the initial and calibrated model parameters for the hydraulic model. Especially the longitudinal dispersivity is a sensitive parameter with major changes during the calibration process (Uhl & Henrichs 2005).

Figure 2 shows the simulated and measured tracer effluent concentrations after a best fit calibration of the model. For the calibration of the flow and transport model acceptable results were achieved. The simulated hydrograph shows a higher maximum filter velocity in comparison to the measured curve. According to the peak the absolute values are comparable but the measured peak occurs earlier. After 12 h the simulated effluent concentration increases because of a split-up of uranin concentrations in the filter. This effect is caused by the simulation due to the analogous model for the supernatant water level. The characteristics of the cumulative effluent loads show approximately a parallel gradient (Figure 2).

Calibration and validation results

It is generally agreed that the amount of microorganisms decreases with the depth of the filter (Wozniak 2006). The biological active layer has a height of approx. 30 cm. For the simulations of the lab-scale experiments initial concentrations for the autotrophic bacteria were chosen

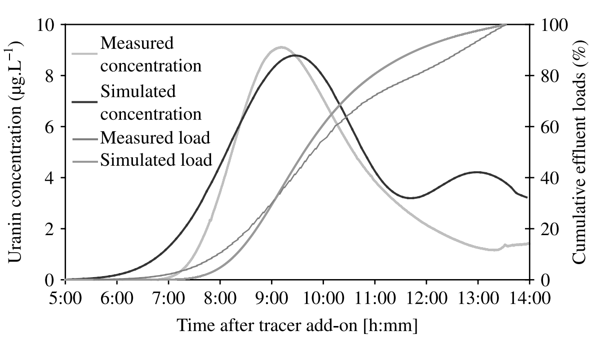

Figure 2 | Calibration results for the hydraulic model; comparison of measured and simulated tracer effluent concentrations and effluent loads.

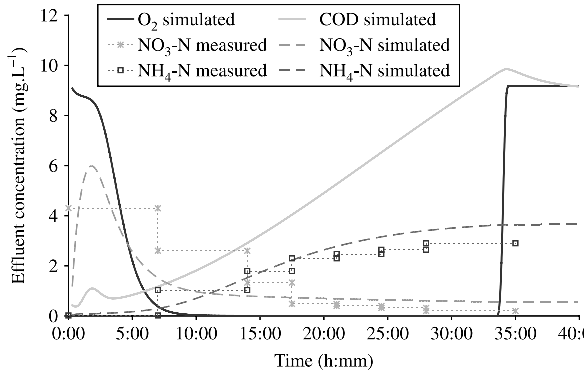

Figure 3 | Effluent concentrations (column S-9, experiment 2) of dissolved oxygen O_2 (simulated), COD (simulated), ammonium NH_4-N (simulated and measured), and nitrate NO_3-N (simulated and measured) for one calibration event.

according to Uhl & Henrichs (2005). Reasonable calibration results are achieved in a concentration range between $15-50\,mg\,kg^{-1}$ COD (XANs) and $15-45\,mg\,kg^{-1}$ COD (XANb). During the calibration process for the sorption parameters of ammonium values of $k_S = 1.1\,kg\,L^{-1}$, $\beta = 0.95$ and $\varpi = 1.1\,h^{-1}$ were obtained.

Figure 3 shows simulated effluent concentrations of COD, dissolved oxygen O_2, ammonium NH_4-N (simulated and measured), and nitrate NO_3-N (simulated and measured) for one calibration event (column S-9, experiment 2). The inundation time of the sand layer is about 32.5 h. Because of the composite samples the measured effluent concentrations are visualized as step functions.

The increasing O_2 concentrations after 34 h indicate the re-aeration of the soil column. The peak in the NO_3-N effluent concentration occurred because of the wash out of nitrified ammonium during dry periods. During the

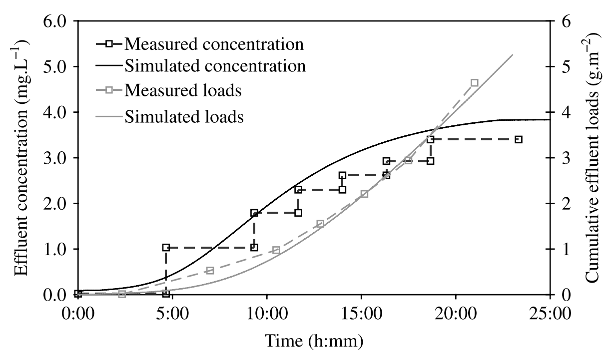

Figure 4 | Validation results for NH_4-N; comparison of measured and simulated effluent concentrations and cumulative effluent loads for column S-5, experiment 1.

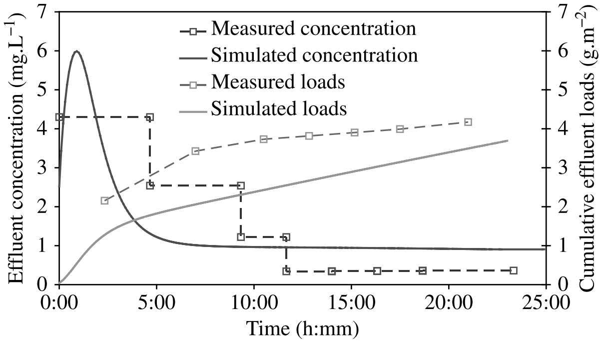

Figure 5 | Validation results for NO_3-N; comparison of measured and simulated effluent concentrations and cumulative effluent loads for column S-5, experiment 1.

absence of O_2 after 8 h oxygen consuming processes like respiration and nitrification stop and the effluent concentration of ammonium and COD increase. From this point on COD and ammonium are only eliminated by sorption processes. Sorption is modelled as a time dependent process. This leads to a breakthrough in the simulated COD and ammonium effluent concentrations after 5 h (Figure 3). After re-aeration the COD effluent concentrations decline because of aerobic degradation processes.

In Figure 4 a comparison of measured and simulated NH_4-N effluent concentrations and cumulative effluent loads is illustrated. Because of the 5 h composite samples the measured effluent concentrations are visualized as a step function. The simulated effluent concentrations as well as the cumulative loads show a good fit to the measured values.

Figure 5 shows the measured and simulated effluent concentrations and cumulative loads for NO_3-N. At the beginning of an event the simulation underestimates the measured loads. The parallel translation between the simulated and measured cumulative loads is caused by the simulation of the wash out effect of nitrate at the beginning of the experiment. After 10 h a constant deviation between measurement and simulation is obvious. This overestimation leads to comparable total effluent loads. The simulation shows on the one hand a faster wash out effect and on the other hand higher nitrate effluent concentration occurred by superior nitrification rates.

Table 2 lists the measured and simulated effluent loads of ammonium and nitrate and the relative deviation of all events used for calibration and validation. The time between the experiments 1 and 2 was 1 week. The nitrate influent loads are about 0.5 g m^{-2} and the influent loads of ammonium vary between 11.5–13.8 g m^{-2}. For the calibration events ammonium effluent loads between 4.4–5.1 g m^{-2} and for the validation events between 4.9–6.3 g m^{-2} were modelled. For ammonium only positive deviations which indicate an overestimation of the simulation are reached. The relative deviations for ammonium vary between 3–56% (calibration events) and 11–36% (validation events).

Table 2 | Comparison of measured and simulated effluent loads and the relative deviations for calibration and validation events

Column	Experiment	Purpose	Throttle outflow rate	NH_4-N				NO_3-N		
				In	Eff	Eff		Eff	Eff	
				Meas	Meas	Sim	Rel. Dev	Meas	Sim	Rel. Dev
			(L s^{-1} m^{-2})	(g m^{-2})			(–)	(g m^{-2})		(–)
S 9	1	Calibration	0.02	11.5	4.3	4.4	0.03	4.2	5.1	0.22
S 9	2	Calibration	0.02	12.3	3.2	5.0	0.56	4.1	3.3	−0.20
S 12	1	Calibration	0.02	11.7	3.7	4.7	0.25	4.4	3.2	−0.28
S 12	2	Calibration	0.02	12.5	3.3	5.1	0.54	4.8	3.2	−0.33
S 5	1	Validation	0.03	12.2	4.6	5.2	0.11	4.2	3.7	−0.12
S 5	2	Validation	0.03	11.8	3.9	4.9	0.26	4.0	3.7	−0.08
S 10	1	Validation	0.04	13.4	5.3	6.0	0.15	2.1	3.3	0.58
S 10	2	Validation	0.04	13.8	4.6	6.3	0.36	4.5	3.3	−0.26

In = influent loads; Eff = effluent; Meas = measured effluent loads; Sim = simulated effluent loads; Rel. Dev = relative Deviation = (meas − sim)·meas^{-1}.

Table 3 | Comparison of the input and output data of the simulated scenarios

Scenario	Loading	Throttle outflow rate	Inundation time	Concentration		Loads		Efficiency
				In	Out	In	Out	
	($m^3 m^{-2}$)	($L s^{-1} m^{-2}$)	(h:mm)	($mg L^{-1}$)		($g m^{-2}$)		(%)
A1	2.5	0.02	35:00	3	1.0	7.5	2.6	65.3
A2	2.5	0.02	35:00	4	1.6	10	3.9	61
A3	2.5	0.02	35:00	6	2.6	15	6.5	56.7
A4	2.5	0.02	35:00	8	3.7	20	9.3	53.5
A5	2.5	0.02	35:00	10	4.8	25	12.1	51.6
B1	2.5	0.02	35:00	6	2.6	15	6.5	56.7
B2	2.5	0.03	23:20	6	2.7	15	6.8	54.7
B3	2.5	0.04	17:30	6	2.8	15	6.9	54
B4	2.5	0.05	14:00	6	2.8	15	7.1	52.7
C1	1	0.02	14:00	6	0.6	6	0.6	90
C2	1.5	0.02	21:00	6	1.4	9	2.1	76.7
C3	2	0.02	28:00	6	1.1	12	4.2	65
C4	2.5	0.02	35:00	6	2.6	15	6.5	56.7
C5	3	0.02	42:00	6	3.0	18	8.9	50.6

In = influent; Out = effluent.

Operation scenarios

The calibrated and validated model is used for simulating different scenarios. The 12 simulated scenarios are listed in Table 3. The table shows only average concentrations. Detailed distributions of the effluent concentrations are shown in Figure 6. In scenario type A the influent NH_4-N-concentrations are varied between 3–10 mg l^{-1}, in type B the throttle outflow rates are altered in a range of 0.02–0.05 $l s^{-1} m^{-2}$ and in type C the hydraulic loads are changed from 1–3 $m^3 m^{-2}$. The shifting boundary conditions lead to varying inundation times (14–42 h) and inflow loads (7.5–25 $g m^{-2}$). Increasing influent loads as well as increasing throttle outflow rates lead to declining efficiencies (Table 3). The inundation time has only a minor influence on simulated effluent loads.

Figure 6 shows the ammonium effluent concentrations for the scenario type A with varying influent concentrations between 3–10 mg L^{-1} (cp. Table 3). With rising influent concentration the effluent concentrations also increase. The ammonium breakthrough in the effluent concentrations takes place between 4–8 h. The increasing influent loads lead to declining efficiencies.

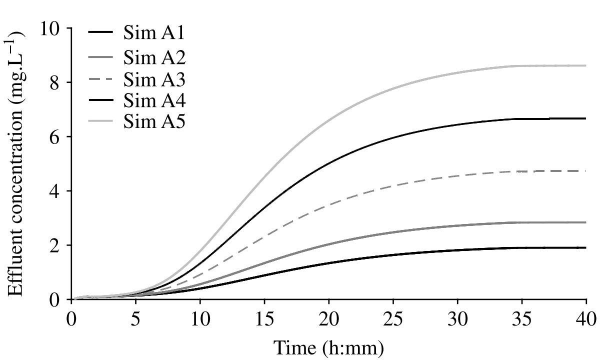

Figure 6 | Hydrographs of the NH_4-N effluent concentrations for scenarios with varying influent concentrations between 3–10 mg L^{-1} (scenario type A).

CONCLUSIONS

It is generally agreed that ammonium is retarded by sorption during an event and nitrified during dry periods in biofilters for enhanced CSO treatment (Uhl & Dittmer 2005; Henrichs *et al.* 2007). The simulation results also showed that nitrification takes place during saturation of the filter. Still, sorption processes play a major role in removing ammonium from CSO by biofiltration.

The simulations scenarios with different inflow concentrations, loadings and throttle outflow rates show a plausible reaction to the input loads. The study indicates that with detailed investigations of model input data a successful calibration and validation could be achieved. After the split sample test the model can be used for simulating varying boundary condition. The simulations showed that input loads up to 13 g m^{-2} ammonium of single events can be retended by a fully regenerated sand filter. Similar values (up to 15 g m^{-2} ammonium) were mentioned by Grobe *et al.* (2005) who carried out lysimeter-scale experiments (diameter 1 m). Higher loads but also smaller concentrations with longer loadings lead to lower efficiencies. The efficiency is also influenced by the throttle outflow rate. If the residence time is lowered (higher throttle outflow rate) the efficiencies decline. This is caused by smaller nitrification rates and the lowered time for adsorption during the percolation phase.

The experimental data indicate that regeneration times of at least 144 h are sufficient for full regeneration of the filters. Minimum values can be investigated by further experiments.

However, the comparison of simulated and measured effluent concentrations of nitrate showed that there is a need for further investigations mainly due to the uncertainties in the degradation process during dry periods between the loadings. Further work will concentrate on long-term simulations with alternating events and dry periods.

Calibrated and validated models can be regarded as very useful tools for further planning of experimental setups in lab-scale studies as well as full scale investigations.

ACKNOWLEDGEMENTS

The study was funded by the Ministry for the Environment and Conservation, Agriculture and Consumers Protection, North Rhine Westphalia, FRG.

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