Modeling dynamics of organic carbon and nitrogen removal during aeration interruption in aerated horizontal flow treatment wetlands

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ABSTRACT

Despite recent developments in process-based modeling of treatment wetlands (TW), the dynamic response of horizontal flow (HF) aerated wetlands to interruptions of aeration has not yet been modeled. In this study, the dynamic response of organic carbon and nitrogen removal to interruptions of aeration in an HF aerated wetland was investigated using a recently-developed numerical process-based model. Model calibration and validation were achieved using previously obtained data from pilot-scale experiments. Setting initial concentrations for anaerobic bacteria to high values (≈ 35–70 mg L−1) and including ammonia sorption was important to simulate the treatment performance of the experimental wetland in transition phases when aeration was switched off and on again. Even though steady-state air flow rate impacted steady-state soluble chemical oxygen demand (CODs), ammonia nitrogen (NH4−N) and oxidized nitrogen (NOx−N) concentration length profiles, it did not substantially affect corresponding effluent concentrations during aeration interruption. When comparing simulated with experimental results, it is most likely that extending the model to include mass transfer through the biofilm will allow to better explain the underlying experiments and to increase simulation accuracy. This study provides insights into the dynamic behavior of HF aerated wetlands and discusses assumptions and limitations of the modeling approach.

Key words | constructed wetland, nature-based solution, nitrification, reactive transport modeling, resilience, system analysis

INTRODUCTION

Wastewater treatment using nature-based solutions such as treatment wetlands (TW) has gained increased attention in recent years (WWAP 2018). TW are man-made ecosystems that mimic natural occurring processes to treat wastewater. TW can be equipped with air pumps to move air into the wetland basin. Such systems are termed aerated wetlands. These systems have been shown to produce lower effluent concentrations and more stable treatment performance than conventional (non-aerated) horizontal flow (HF) wetlands (Wallace et al. 2008) and lower effluent concentrations of total nitrogen (TN) and pathogens than non-aerated vertical flow (VF) wetlands (Dotro et al. 2017; Nivala et al. 2019). Aerated wetlands have been successfully applied to treat both municipal and industrial wastewater (Ilyas & Masih 2017). However, little is known about the dynamic behavior of such systems; especially the response to potential interruptions of aeration induced by air pump failures or power shortages. To the best of our knowledge, only two experimental studies on this topic exist. Boog et al. (2018) & Murphy et al. (2016) have shown that aeration interruptions of several days substantially deteriorate the treatment efficacy for organic carbon and nitrogen. Indeed, the aerated wetlands in these two studies recovered treatment efficacy within several days after the aeration was switched back on. Nevertheless, in order to estimate the associated risks of a loss in treatment efficacy and to design aerated wetlands that are more resilient towards aeration interruptions, it is important to improve the understanding of the treatment...
processes involved during aeration failure. This requires extensive experimentation and/or the application of process-based modeling and simulation.

Process-based modeling has been applied to study the dynamic behavior and associated functioning of different types of TW (Samsó & García 2013; Morvannou et al. 2014; Pálfy & Langergraber 2014; Rizzo & Langergraber 2016; Boano et al. 2018). However, process-based modeling has not yet been applied to investigate the dynamic response of horizontal flow (HF) aerated wetlands to interruptions in aeration. Murphy et al. (2019) analyzed the dynamic response of a VF aerated wetland to aeration interruption using a process-based model. Their model accounted for diffusive transport of NH₄-N, nitrate nitrogen (NO₃-N) and dissolved oxygen (DO) through the biofilm and nitrification inside the biofilm. However, this model left out the important phenomena of water flow and advective-dispersive mass transport. Recently, Boog et al. (2019) developed a reactive transport model for HF aerated wetlands. This model was calibrated and validated with data of pilot-scale HF aerated wetlands at variable air flow rates and steady-state operation. The model’s validity to simulate the dynamic behavior during an aeration interruption remains unknown. Moreover, the studies of Boog et al. (2018) and Murphy et al. (2016) have shown that aeration interruptions of several days triggered a change in process conditions inside the wetland from aerobic to anaerobic. Although the model by Boog et al. (2019) includes formulations to simulate the behavior of anaerobic bacteria and associated pollutant removal reactions, it is not clear how initial concentrations of anaerobic bacteria affect a simulation. Additionally, removal processes that are hidden during steady-state behavior of HF aerated wetlands may emerge when treatment performance dynamically changes in a short period of time, such as during an aeration interruption.

This study aimed to model a six-day-long aeration interruption in a pilot-scale HF aerated wetland. Our previously developed process-based model (Boog et al. 2019) was extended with a process for ammonia adsorption to catch the dynamic response of NH₄-N effluent concentrations during aeration interruption. The extended model was calibrated and validated with data of the aeration interruption experiments conducted by Boog et al. (2018) using a pilot-scale HF aerated wetland. Initial simulation conditions for anaerobic bacteria were investigated in order to elucidate their role in the dynamic loss of treatment performance during aeration interruption. A scenario analysis was carried out to investigate whether air flow rate affects the response to aeration interruption. Additionally, the experiment from Boog et al. (2019) that was used to calibrate the original model was simulated using the extended model to test if the extension with ammonia adsorption affected the original simulation fit. This study provides insights into the dynamic behavior of HF aerated wetlands and discusses assumptions and limitations of the modeling approach.

METHODS

Experiments

Experimental site and system description: All experiments were conducted using pilot-scale HF aerated wetlands at the research facility in Langenreichenbach, Germany described by Nivala et al. (2013) (see also supplementary information Figure S1, available with the online version of this paper). Boog et al. (2018) performed two aeration interruption experiments in the wetland HA, while Boog et al. (2019) performed steady-state experiments in the wetlands HMc and HM. Here, steady-state is referred as operation of the wetland with continuous aeration (24 h per day). The design of the three wetlands were identical (Figure S2, available online), except that HA (Figure 1)
was unplanted, while HM and HMc were planted with *Phragmites australis*. All three wetlands were filled with gravel (medium and coarse), equipped with a similar type of aeration system and loaded with primarily treated domestic sewage at a hydraulic loading rate of 0.567 m$^3$ d$^{-1}$. This corresponds to an organic loading rate of 46.1 ± 7.8 g COD, m$^{-2}$ d$^{-1}$ during the experiments in HA and 48.0 ± 5.4 g COD, m$^{-2}$ d$^{-1}$ during the experiments in HM and HMc. HM and HMc started operation in September 2014. HA started operation in August 2010. It is to be noted that from August 2012 to August 2014, the electric air pumps in HA were replaced by a mechanical wind-driven air pump. This resulted in a DO shortage that turned process conditions in the wetland from aerobic into anaerobic, impeded treatment efficacy to that of a conventional HF system and clogged most of the aeration lines (Boog et al. 2016). Aeration lines were cleaned and electric aeration was restarted in November 2014; initial treatment efficacy recovered by April 2015 (data not shown).

**Aeration interruption experiments by Boog et al. (2018):** During June to August 2015, Boog et al. (2018) conducted one aeration interruption experiment in the wetland HA; the second experiment was conducted from December 2015 to February 2016. Both experiments consisted in monitoring steady-state operation for 42 days, then aeration was interrupted for six days; the wetland was monitored for additional eight days after switching aeration on again. Samples were taken from the influent, effluent as well as from pore water at fractional lengths of 0.13, 0.25, 0.38, 0.5 and 0.75. All samples were analyzed for temperature (T), electric conductivity, pH, redox potential, dissolved oxygen (DO), dissolved organic carbon (DOC), dissolved nitrogen (DN), ammonia nitrogen (NH$_4^-$-N), nitrite nitrogen (NO$_2^-$-N) and nitrate nitrogen (NO$_3^-$-N) as reported in Boog et al. (2018). Total organic carbon (TOC) and total nitrogen (TN) were measured for the second experiment (January to February 2016) only. Therefore, TOC for the influent and effluent of the first experiment were estimated by regression on measured DOC concentrations. Total chemical oxygen demand (COD$_T$) and soluble chemical oxygen demand (COD$_S$) values were imputed by regressions on TOC and DOC; corresponding regression equations were obtained from Boog et al. (2019). Air flow rates for the aeration front and back grids were measured to 1.2 and 1.0 m$^3$ h$^{-1}$ (equal to 426 and 354 L m$^{-2}$ h$^{-1}$, respectively).

This study considered additional unpublished data from the routine monitoring of HA that was obtained before and between the two aeration interruption experiments (May 2015 to February 2016). Routine monitoring consisted of weekly sampling of the influent and effluent. Corresponding samples were analyzed for the aforementioned wastewater quality parameters according to the procedures in Nivala et al. (2013). Additionally, COD$_T$ was analyzed using test kits (LCK 514 and LCK 314 Hach–Lange) and a spectrophotometer (DR5900 Hach–Lange). Further information is presented in the supplementary information (Section S1) (available online).

**Steady-state experiments by Boog et al. (2019):** Boog et al. (2019) monitored the two HF aerated wetlands HM and HMc during steady-state operation from August 2017 to April 2018. The air flow rate at 0.0–0.4 fractional length in HM was reduced step-wise from approximately 700–72 L m$^{-2}$ d$^{-1}$ with the intention to provide data to calibrate their simulation model. HMc was left unchanged as control and used as cross-validation data set.

**Simulation model**

The simulation model applied here was based on the one-dimensional reactive transport model developed by the same others (Boog et al. 2019). This model simulates water flow using a dual-permeability formulation, convective-conductive heat transport, advective-dispersive mass transport of soluble and particulate wastewater pollutants, biodegradation using extended formulations of the Constructed Wetland Model No.1 (CWM1; Langergraber et al. 2009) and oxygen mass transfer through aeration. CWM1 describes the growth and decay of six bacterial groups (heterotrophic XH, autotrophic nitrifiers XA, fermenters XFB, methanogenic bacteria XAMB, sulfide oxidizers XSOB and sulfate reducers XASRB) through monod-type equations. The biofilm or suspended flocs are not explicitly considered as physical structures. Therefore, processes such as mass transfer through the biofilm or floc and/or biofouling are not described. Plants were not modeled, as plant presence in similar HF aerated wetlands designs at the same site was reported as insignificant with respect to the removal of organic carbon and nitrogen (Nivala et al. 2019). Therefore, the use of planted and unplanted experimental wetlands for model calibration and validation does not introduce any bias. Further details and equations are given in Boog et al. (2019). To simulate the dynamic response of NH$_4^-$-N concentrations to aeration interruption, the model was extended with a formulation of a kinetic ammonia adsorption and desorption process using a Freundlich isotherm (Equations (1)–(2)); the subscripts $m$ and $im$ denote...
the mobile and immobile (adsorbed) phase. Corresponding parameters are listed in Table 1.

\[ r_{ads,SNH} = k_{ads,SNH} (S_{NH,im} - S_{NH,im}^*) \]  
\[ S_{NH,im} = a_{ads} S_{NH,m}^* \]  
\[ \text{Parameter Description Unit} \]
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<td>( S_{NH} )</td>
<td>Actual adsorbed ammonia nitrogen concentration</td>
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Data set partition: The model was used to simulate the aeration interruption experiments by Boog et al. (2018) and the simulation scenarios by Boog et al. (2019). For the partitioning and processing of the experimental data of Boog et al. (2019), see the corresponding publication. The experimental data from Boog et al. (2018) and the routine monitoring were partitioned into two sets: 1) one set covering the aeration interruption experiment from June to August 2015 (42 days steady-state, six days interruption and eight days recovery) termed the Calibration set; 2) one set covering the steady-state period between the two experiments (121 days) and the aeration interruption experiment in December 2015 to February 2016 (42 days steady-state, six days interruption and eight days recovery) termed the Validation set.

Influent fractionation: As the biodegradation reactions of the simulation model are based on chemical oxygen demand (COD), measured TOC and DOC concentrations were converted to total COD (COD\(_t\)) and soluble COD (COD\(_s\)), respectively. Influent and effluent COD\(_t\) were imputed by regression models that were calibrated using measurements for COD\(_t\) of the routine monitoring during January to December 2015 (data not shown); corresponding COD\(_t\) were imputed by combining ratios of TOC/DOC and COD\(_t\)/COD\(_t\) from measurements obtained by Boog et al. (2019). Similarly, COD\(_t\) and COD\(_s\) of pore water samples were imputed using ratios of COD\(_t\)/TOC and COD\(_t\)/DOC computed by Boog et al. (2019). Biodegradable COD\(_t\) (BCOD\(_t\)) was estimated from CBOD\(_5\) measurements of the routine monitoring according to the procedures in Roeleveld & Loosdrecht (2002) using the value of \( k_{BOD} \) from Boog et al. (2019). Details are given above. Imputed COD\(_t\), COD\(_s\), and BCOD\(_t\) were then converted into model input components as described in Boog et al. (2019).

Model discretization and parametrization: For all simulations, a one dimensional finite-element mesh with 94 elements (element size of 0.05 m) was considered and regular time-stepping schemes with step sizes between 1,800–7,200 s were applied. The same parameter set as in Boog et al. (2019) was used except for the additional parameters of ammonia adsorption and the oxygen transfer coefficient \( k_{La,20} \). The parameter values of the Freundlich isotherm in Equation (2) \((a_{ads} = 1.63 \text{ and } b_{ads} = 0.548)\) were taken from Sikora et al. (1995); the ammonia adsorption rate coefficient \( k_{ads,SNH} \) was adjusted during Calibration. The oxygen transfer coefficients \( k_{La,20} \) for the front and back aeration grids were estimated to be 2.875 and 2.75 h\(^{-1}\), respectively, using the equation \( k_{La,20} = 0.511 \cdot \log(\text{air flow rate}) \) (Boog et al. 2019) and air flow rates for front and back aereration grid of 426 and 354 L m\(^{-2}\) h\(^{-1}\) (converted from 1.2 and 1.0 m\(^3\) h\(^{-1}\) and an aereration grid size of 2.82 m\(^2\) each; Boog et al. 2018), respectively.

Initial and boundary conditions: Initial water pressure in the entire model domain was set to 9,810 Pa (Figure 1). The initial temperature was set to the mean of the measured water temperature of the in- and effluent at start-up of the Calibration data set. Initial concentrations of wastewater components and bacteria were 0.1 and 1.0 mg L\(^{-1}\), except for bacteria \( X_{AMB} \) and \( X_{SRM} \) that were adjusted during calibration. The boundary condition for water flow at the influent point was defined as a source term of 6.667e-6 m\(^3\) s\(^{-1}\) (converted from a hydraulic loading rate of 0.576 m\(^2\) d\(^{-1}\)). Measurements of influent concentrations were used to define time-dependent Dirichlet boundary conditions at the influent point. As sulfur compounds were not measured but were considered in the biodegradation model, corresponding boundary conditions for H\(_2\)S-S (S\(_{H_{2}S}\)) and SO\(_4\)-S (S\(_{SO4}\)) at the influent point were set to 8.6 and 56.6 mg L\(^{-1}\), respectively (same values as used by Boog et al. 2019). For heat transport, time-dependent Dirichlet boundary conditions based on measured water temperature were defined at both the influent and effluent point. The simulation conditions at the end of the simulation of the Calibration set were used as initial conditions for the Validation set; the boundary condition types in the Validation set were similar.
Calibration, validation and scenario analysis: For calibration using the Calibration data set, the adsorption rate coefficient $k_{\text{ads,SOI}}$ and initial concentration of acetotrophic methanogenic bacteria $X_{\text{AMB}}$ and acetotrophic sulphate reducing bacteria $X_{\text{ASRB}}$ were adjusted to obtain a reasonable fit of the effluent concentrations for NH$_4$–N and COD. The starting value for $k_{\text{ads,SOI}}$ was 2 h$^{-1}$ (Pálfy & Langergraber 2014); for initial bacteria concentration it was 1.0 mg L$^{-1}$. Assessing the model fit was achieved by visually comparing measured and simulated effluent concentrations and calculating the modified coefficient of efficiency ($E_1$, also termed modified Nash–Sutcliffe Efficiency). The fit of the model for the Validation data set was assessed by calculating the $E_1$. Finally, the impact of $k_{\text{La,20}}$ on the response to aeration interruption was analyzed by simulating the Calibration scenario at $k_{\text{La,20}}$ of 1–4 h$^{-1}$. All simulations were executed as serial runs on the high-performance computing cluster EVE (Dell PowerEdge R630, Intel Xeon E5-2690 v4 CPUs and/or Intel Xeon E5-2670 v2 CPUs). Post-processing and visualization was done in the statistical software R (R Core Team 2014).

RESULTS AND DISCUSSION

Model calibration and validation

During the 42 days of steady-state operation before the aeration was switched off, effluent concentrations showed typical effluent concentrations for HF aerated wetlands (high DO, low COD and NH$_4$–N and moderate NO$_x$–N) (Ilyas & Masih 2017). When the aeration was switched off, DO depleted and inhibited further removal of COD and nitrification of NH$_4$–N; consequently, the production of NO$_x$–N ceased (Figure 2).

After restarting aeration, pore water and effluent concentrations recovered to values of the steady-state operation within 3–8 days. The trends in effluent concentrations during Calibration were described by the model with a reasonable degree of accuracy $E_{1_{\text{cal}}} = 0.42$ (Figure 2). The fit of pore water concentration at a fractional length <0.5 was less accurate as a result of the estimated oxygen transfer coefficient $k_{\text{La,20}}$ using the data from Boog et al. (2019). To fit COD, effluent concentrations, the initial concentrations of acetotrophic methanogenic bacteria $X_{\text{AMB}}$ and acetotrophic sulphate reducing bacteria $X_{\text{ASRB}}$ were adjusted to obtain a reasonable fit of the effluent concentrations for NH$_4$–N and COD. The starting value for $k_{\text{ads,SOI}}$ was 2 h$^{-1}$ (Pálfy & Langergraber 2014); for initial bacteria concentration it was 1.0 mg L$^{-1}$. Assessing the model fit was achieved by visually comparing measured and simulated effluent concentrations and calculating the modified coefficient of efficiency ($E_1$, also termed modified Nash–Sutcliffe Efficiency). The fit of the model for the Validation data set was assessed by calculating the $E_1$. Finally, the impact of $k_{\text{La,20}}$ on the response to aeration interruption was analyzed by simulating the Calibration scenario at $k_{\text{La,20}}$ of 1–4 h$^{-1}$. All simulations were executed as serial runs on the high-performance computing cluster EVE (Dell PowerEdge R630, Intel Xeon E5-2690 v4 CPUs and/or Intel Xeon E5-2670 v2 CPUs). Post-processing and visualization was done in the statistical software R (R Core Team 2014).
methanogenic bacteria $X_{AMB}$ and acetotrophic sulphate reducing bacteria $X_{ASRB}$ were adjusted to relatively high values of 35 and 70 mg L$^{-1}$, respectively. To fit NH$_4$–N effluent concentrations, the rate coefficient for ammonia adsorption ($k_{ads,S_{NH4}}$) was adjusted to 0.05 h$^{-1}$ during the Calibration scenario. The simulation of the Validation scenario was less accurate for the response of COD$_s$ and COD$_e$ effluent concentrations, despite an overall similar value of $E_1$ ($E_{val} = 0.48$). The resimulation of the two scenarios of Boog et al. (2019) with the extended model, resulted in a similar fit than the original simulation ($E_{cal,Boogetal,2019} = 0.01$, $E_{cv,Boogetal,2019} = -0.41$). These results show that the simulation model was able to simulate the dynamic system behavior during aeration interruption, however, indicate that ammonia adsorption and initial concentrations of anaerobic bacteria $X_{AMB}$ and $X_{ASRB}$ were quite important.

Impact of initial concentrations of anaerobic bacteria $X_{AMB}$ and $X_{ASRB}$ on COD$_e$ effluent concentration

By assuming low initial bacteria concentrations (1 mg L$^{-1}$), which correspond to a newly built and non-commissioned TW, the simulated peak concentrations of COD$_e$ during aeration interruption were too high compared to measured ones (Figure 3). During aeration interruption, COD$_e$ effluent concentrations were mainly composed of acetate ($S_A$) produced by fermenting bacteria $X_{FB}$ at the inlet zone. Samso & García (2013) also reported the dominance of $S_A$ in simulated COD effluent concentrations of a conventional (non-aerated) HF wetland.

In the biodegradation model, $S_A$ is consumed by methanogenic bacteria $X_{AMB}$ and sulphate reducing bacteria $X_{ASRB}$. The growth rates of these bacterial groups are comparably low and both bacterial groups are assumed to be strictly anaerobic. This prevented their growth during the steady-state phase when aeration was switched on and translated into slow growth when aeration was switched off. As a result, the acetate ($S_A$) produced by fermenting bacteria ($X_{FB}$) accumulated during aeration interruption.

By increasing initial concentrations of both methanogenic $X_{AMB}$ and sulphate reducing bacteria $X_{ASRB}$ to 35 and 70 mg L$^{-1}$, COD$_e$ effluent concentrations were well fitted (Figure 3). Due to the assumption that the initial concentrations were spatially uniform, the accuracy of the simulated spatial COD$_e$ patterns when aeration was interrupted was inaccurate. Nevertheless, the dependency on high initial concentrations for $X_{AMB}$ and $X_{ASRB}$ indicates that the assumption of a newly-built and non-commissioned TW (initial bacteria concentration of 1 mg L$^{-1}$) was not appropriate to simulate dynamic performance of a five year old HF aerated wetland. It also indicates that anaerobic bacteria $X_{AMB}$ and $X_{ASRB}$ must have been established prior to the aeration interruption experiments. In fact, from 2012 to 2014 the experimental wetland HA was not aerated properly, which deteriorated treatment efficacy to that of a conventional HF wetland (Boog et al. 2016). During that time, the aeration system of the wetland clogged and had to be repaired in order to recover treatment efficacy when the aeration was switched back on in 2014 (Boog et al. 2016). It is highly likely that communities of anaerobic bacteria $X_{AMB}$ and $X_{ASRB}$ had developed and formed within a biofilm during the non-aerated period. Anaerobic methanogenic and sulphate reducing microorganisms are common in conventional HF wetlands (Jahangir et al. 2016; López et al.)
2019; Wu et al. 2013). For instance, Samsó & García (2013) simulated the bacterial community dynamics in a conventional HF wetland and reported established communities of methanogenic and sulphate reducing bacteria within two years. Therefore, these bacterial communities could have been established by the end of the two-years-long non-aerated period in the experimental wetland HA. Most likely, overloading during this period as reported by Boog et al. (2016) had intensified bacterial development and biofilm formation.

Methanogenic and sulphate reducing bacteria are reported to survive in biofilms even under anoxic to aerobic and nutrient limiting conditions (Stewart & Franklin 2008; Angel et al. 2012). It is highly likely that the established anaerobic bacterial communities survived the subsequent aerated periods until the aeration-interruption experiments started. In the simulations, high DO concentrations during the aerated phases eliminated the methanogenic and sulphate reducing bacterial communities (XAMB and XASRB) as a protective biofilm was not considered. This die-off resulted in an increased accumulation of SA during the Validation scenario and produced high COD efluent concentrations (Figure 3). Therefore, initial simulation conditions must be critically evaluated when simulating dynamic behavior in HF aerated wetlands.

Impact of $k_{ads,SNH}$ on NH4–N effluent concentration

The model extension with a kinetically controlled ammonia adsorption process improved the fit to the NH4–N effluent concentrations during aeration interruption for both the Calibration and Validation scenarios (Figure 4). Excluding ammonia adsorption ($k_{ads,SNH} = 0$) resulted in an overestimation of measured NH4–N effluent concentrations. The adsorption of ammonia to sand and gravel was reported by Sikora et al. (1995) & Wen-Ling et al. (2011), as well as being applied in TW modeling studies using gravel and/or sand as media (Morvannou et al. 2014; Pálffy & Langergraber 2014; Rizzo et al. 2014). As gravel was used in this study, ammonia adsorption is highly likely to explain the ongoing NH4N removal during aeration interruption. An explanation by other nitrogen removal processes such as anaerobic ammonia oxidation (Anammox) or ammonia oxidation using ferric iron as electron acceptor (Feammox; Wu et al. 2013) is less likely. Anammox was probably inhibited at the CODt/NH4–N ratios (3–4) of this study as it was reported to be inhibited at C/N ratios above 1.7 (Cao et al. 2017). Assuming that influent iron concentrations in this study were similar as the 0–2 mg L$^{-1}$ reported from other domestic wastewaters (Kadlec & Wallace 2009), and, considering that these concentrations are one to two orders of magnitude lower than measured influent NH4–N concentrations, the Feammox process is unlikely to have substantially contributed to NH4–N removal during aeration stop. Thus, the adsorption of ammonia is likely to happen in HF aerated wetlands but will be only of importance during operational disruption such as aeration interruption. The fitted rate coefficient $k_{ads,SNH}$ of 0.05 h$^{-1}$, however, was considerably lower than values of 2.0 and 6.2 h$^{-1}$ reported by Pálffy & Langergraber (2014) and Wen-Ling et al. (2011), respectively. It is likely that the differences were caused by differences in gravel substrate. Additionally, the high amount of organic matter accumulation related to biofilm growth in the experimental wetland HA could have altered adsorption sites availability. Adsorption sites on the gravel matrix were reduced while additional sites were provided.

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Figure 4 | Measured and simulated nitrogen effluent concentration before, during and after aeration interruption. Duration of zero represents time when aeration was switched off.
on the biofilm, however, the biofilm probably has different characteristics than the gravel.

Adsorbed NH$_4$–N amplified the peak of simulated NO$_x$–N during the recovery of the wetland after aeration was switched back on (Figure 4). The mass of NH$_4$–N that caused the peak originated in the mass of NH$_4$–N that adsorbed during aeration interruption, then desorbed during recovery and finally was nitrified to NO$_x$–N. The accumulated organic matter in the experimental wetland HA enabled, most likely, a simultaneous denitrification of produced NO$_x$–N during recovery by providing the required carbon sources. This would explain the absence of a peak in measured NO$_x$–N effluent concentrations. Besides high initial concentrations of anaerobic bacteria $X_{AMB}$ and $X_{ASRB}$, additional accumulated organic matter was not assumed at simulation start-up as this would have been quite difficult with the current model formulations. For example, additionally accumulated organic matter at start-up could have been realized by defining high initial concentrations of slowly biodegradable particulate COD ($X_{S,im}$), however, $X_{S,im}$ would have been rapidly hydrolyzed and consumed by heterotrophic bacteria $X_{H}$ before aeration was switched off. Another explanation for the absence of a peak in measured NO$_x$–N effluent concentration could be that heterotrophic bacteria in the experimental wetland used organic carbon stored in the biofilm for denitrification. For instance, Murphy et al. (2016) highlighted the importance of the diffusion of NH$_4$–N and NO$_x$–N in and out of biofilms during aeration interruption in a VF aerated wetland – this could be similar for COD$_s$. Further research is necessary to elucidate the importance of biofilm formation for microbial community development and associated dynamic behavior of aerated wetlands.

### Influence of $k_{La,20}$ on spatial concentration patterns

Porewater concentration gradients for DO, COD$_s$, NH$_4$–N and NO$_x$–N of the steady-state phase before stopping aeration (Calibration) were approximately fit at an oxygen transfer coefficient ($k_{La,20}$) of 2.875 and 2.75 h$^{-1}$ for the front and back aeration grid, respectively (Figure 5). This shows that the internal concentrations gradients of an HF aerated wetland can be approximately simulated using a model that was calibrated on data from another HF aerated wetland; however, accuracy lacked, especially with respect to the DO concentration pattern. On one hand, this could have been caused by estimating $k_{La,20}$ from the equation $k_{La,20} = 0.511 \cdot \log \text{(air flow rate)}$ proposed by Boog et al. (2019). On the other hand, this could also be explained by a lack of the simulation model that ignores the mass transfer resistance of biofilms.

In the current model, DO concentration only depends on the oxygen transfer rate by aeration and oxygen uptake rates.
Validation of the change in the experimental system. Measured porewater compared to the Calibration vated sludge plants (Manser adsorption was found to be important for modeling the dynamic for the impeded treatment ef state operation air NOx lengths greater than 0.6 (Figure 5). Consequently, $k_{L_a,20}$ did not alter the simulated effluent concentrations during aeration interruption. This means that during aeration interruption in HF aerated wetlands, treatment efficacy for organic carbon and nitrogen is likely to deteriorate and recover in a similar manner regardless of the air flow rate during steady-state operation.

Pore water concentrations during steady-state operation of the Validation scenario were fit with higher accuracy as during the Calibration scenario. This was grounded in a change in the experimental system. Measured porewater concentration gradients during Validation shifted upstream compared to the Calibration scenario; simulated pore water gradients stayed almost the same. This indicates that the recovery of the water quality patterns in the experimental wetland HA from the period without aeration in 2012–2014, which was associated with intense accumulation of organic matter, was still occurring. This shows that it can take a long time (in the order of a year) for an HF aerated wetland to recover from operational disruptions that occur for a long period of time (in the order of months or years). For instance, Boano et al. (2018) simulated the effect of sudden increase in hydraulic and organic loads on conventional HF wetlands and reported an adaptation period of treatment performance of approximately one year. This further highlights potential difficulties to model TW that have been in operation for many years prior to the start of a modeling study.

**CONCLUSION**

This study modeled treatment performance before, during and after aeration interruption in an horizontal flow aerated wetland. The process-based simulation model was able to simulate the dynamic response of DO, COD$\_a$, NH$_4$-N and NO$_3$-N pore water and effluent concentrations. The steady-state operation air flow rate did not play a substantial role for the impeded treatment efficacy for COD$\_a$, NH$_4$-N and NO$_3$-N during aeration interruption. In contrast, ammonia adsorption was found to be important for modeling the dynamic behavior of HF aerated wetlands when the aeration is interrupted. Furthermore, the presence of anaerobic bacteria can be crucial for organic carbon removal during aeration interruption, especially for HF aerated wetlands that have been operated for many years and/or have undergone severe operational disruptions. Thus, defining the right initial conditions is critical for simulating dynamic behavior of such systems. The current model lacked accuracy in simulating porewater concentration profiles for DO, NH$_4$-N and NO$_3$-N as well as COD$\_e$ effluent concentrations during the second aeration interruption experiment, which was most likely caused by ignoring mass transfer through the biofilm. Therefore, future modeling studies on aerated wetlands should focus on including an explicit description of mass transfer through the biofilm and its impact on microbial community dynamics and dynamic system behavior.

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