Supporting Information

Full-Scale Modeling Explaining Large Spatial Variations of Nitrous Oxide Fluxes in a Step-Feed Plug-Flow Wastewater Treatment Reactor

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Table S1. The definition and units of model components

Variable	Description	Unit
S_{O2}	Dissolved oxygen concentration	mg-O_2/L
S_{NH3}	Ammonia concentration	mg-N/L
$S_{NO_2^-}$	Nitrite concentration	mg-N/L
$S_{NO_3^-}$	Nitrate concentration	mg-N/L
S_{NO}	Nitric oxide concentration	mg-N/L
S_{N2O}	Nitrous oxide concentration	mg-N/L
S_{NH2OH}	Methanol concentration	mg-N/L
S_{N2}	Nitrogen concentration	mg-N/L
S_{Mred}	Reduced form of electron carrier concentration	mmol/g-VSS
S_{Mox}	Oxidized form of electron carrier concentration	mmol/g-VSS
S_S	Readily biodegradable COD concentration	mg-COD/L
X_S	Slowly biodegradable COD concentration	mg-COD/L
X_{AOB}	Active AOB biomass concentration	mg-COD/L
X_{NOB}	Active NOB biomass concentration	mg-COD/L
X_H	Active heterotrophic biomass concentration	mg-COD/L

Table S2. Process matrix related to N_2O dynamics in ammonia oxidizing bacteria (AOB)

Process	S_{O2}	S_{NH3}	S_{NH2OH}	$S_{NO_2^-}$	S_{NO}	S_{N2O}	S_{Mox}	S_{Mred}	Kinetic rate expressions
1. Ammonia oxidation	-1	-1	1				1	-1	$r_{NH3,ox} \frac{S_{O2}}{K_{O2,NH3}+S_{O2}} \frac{S_{NH3}}{K_{NH3}+S_{NH3}} \frac{S_{Mred}}{K_{Mred,1}+S_{Mred}} X_{AOB}$
2. NH_2OH oxidation			-1		1		-3/2	3/2	$r_{NH2OH,ox} \frac{S_{NH2OH}}{K_{NH2OH}+S_{NH2OH}} \frac{S_{Mox}}{K_{Mox}+S_{Mox}} X_{AOB}$
3. NO oxidation				1	-1		-1/2	1/2	$r_{NO,ox} \frac{S_{NO}}{K_{NO,ox}+S_{NO}} \frac{S_{Mox}}{K_{Mox}+S_{Mox}} X_{AOB}$
4. NO reduction					-1	1	1/2	-1/2	$r_{NO,red} \frac{S_{NO}}{K_{NO,red}+S_{NO}} \frac{S_{Mred}}{K_{Mred,2}+S_{Mred}} X_{AOB}$
5. Oxygen reduction	-1/2						1	-1	$r_{O2,red} \frac{S_{O2}}{K_{O2,red}+S_{O2}} \frac{S_{Mred}}{K_{Mred,3}+S_{Mred}} X_{AOB}$
6. Nitrite reduction				-1		1	1	-1	$r_{NO_2^-,red} \frac{S_{NO_2^-}}{K_{NO_2^-}+S_{NO_2^-}} \frac{S_{Mred}}{K_{Mred,4}+S_{Mred}} X_{AOB}$
7. Electron carriers									$S_{Mred}+S_{Mox}=C_{tot}$

Table S3. Process matrix related to N_2O dynamics in heterotrophic denitrifiers

Process	S_{O2}	S_S	S_{NH3}	$S_{NO_2^-}$	$S_{NO_3^-}$	S_{N2O}	S_{N2}	X_S	X_H	Kinetic rate expressions
8. Hydrolysis		1	i_{NXS}					-1		$k_H \frac{X_S / X_H}{K_X + X_S / X_H} X_H$
9. Aerobic growth	$-\frac{1-Y_H}{Y_H}$	$-\frac{1}{Y_H}$	$-i_{NBM}$						1	$\mu_H \frac{S_{O2}}{K_{OH1}+S_{O2}} \frac{S_S}{K_{S1}+S_S} X_H$
10. Nitrate reduction		$-\frac{1}{Y_H}$	$-i_{NBM}$	$\frac{1-Y_H}{1.14Y_H}$	$-\frac{1-Y_H}{1.14Y_H}$				1	$\mu_H \eta_{H1} \frac{K_{OH2}}{K_{OH2}+S_{O2}} \frac{S_{NO_3^-}}{K_{NO_3^-}^{HB}+S_{NO_3^-}} \frac{S_S}{K_{S2}+S_S} X_H$
11. Nitrite reduction		$-\frac{1}{Y_H}$	$-i_{NBM}$	$-\frac{1-Y_H}{1.14Y_H}$		$-\frac{1-Y_H}{1.14Y_H}$			1	$\mu_H \eta_{H2} \frac{K_{OH3}}{K_{OH3}+S_{O2}} \frac{S_{NO_2^-}}{K_{NO_2^-}^{HB}+S_{NO_2^-}} \frac{S_S}{K_{S3}+S_S} X_H$
12. N_2O reduction		$-\frac{1}{Y_H}$	$-i_{NBM}$			$-\frac{1-Y_H}{0.57Y_H}$	$\frac{1-Y_H}{0.57Y_H}$		1	$\mu_H \eta_{H3} \frac{K_{OH4}}{K_{OH4}+S_{O2}} \frac{S_{N2O}}{K_{N2O}^{HB}+S_{N2O}} \frac{S_S}{K_{S4}+S_S} X_H$

Table S4. Kinetic and stoichiometric parameters of the integrated N_2O model

Parameter	Definition	Values	Unit	Source
Stoichiometric parameters				
Y_{AOB}	yield coefficient for AOB	0.150	mg-COD/mg-N	(1)
Y_H	yield coefficient for HB	0.6	mg-COD/mg-COD	(2)
i_{NBM}	nitrogen content of biomass	0.07	mg-N/mg-COD	(2)
i_{NXS}	nitrogen content of X_S	0.04	mg-N/mg-COD	(2)
Ammonium oxidizing bacteria (AOB)				
$r_{NH3,ox}$	Specific maximum ammonia oxidation rate	0.19 ± 0.01	mg-N/(mg-COD*h)	(3)
$r_{NH2OH,ox}$	Specific maximum NH_2OH oxidation rate	0.23	mg-N/(mg-COD*h)	(4)
$r_{NO,ox}$	Specific maximum NO oxidation rate	0.23	mg-N/(mg-COD*h)	(4)
$r_{O2,red}$	Specific maximum oxygen reduction rate	1.42 ± 0.16	mg-O_2/(mg-COD*h)	(3)
$r_{NO_2^-,red}$	Specific maximum nitrite reduction rate	0.041 ± 0.006	mg-N/(mg-COD*h)	(3)
$r_{NO,red}$	Specific maximum NO reduction rate	$(2.3 \pm 0.2) \times 10^{-4}$	mg-N/(mg-COD*h)	(3)
$K_{O2,NH3}$	O_2 affinity constant for ammonia oxidation	0.48 ± 0.03	mg-O_2/L	(3)
K_{NH3}	NH_3 affinity constant for ammonia oxidation	2.4	mg-N/L	(4)
K_{NH2OH}	NH_2OH affinity constant for its oxidation	0.7	mg-N/L	(4)
$K_{NO,ox}$	NO affinity constant for NO oxidation	0.0084	mg-N/L	(4)
$K_{O2,red}$	O_2 affinity constant for oxygen reduction	0.06	mg-O_2/L	(4)
$K_{NO_2^-}$	Nitrite affinity constant for nitrite reduction	0.14	mg-N/L	(4)
$K_{NO,red}$	NO affinity constant for NO reduction	0.0084	mg-N/L	(4)
K_{Mox}	S_{Mox} affinity constant for NO oxidation	$1\times10^{-2}\times C_{tot}$	mmol/g-VSS	(4)
$K_{Mred,1}$	S_{Mred} affinity constant for ammonia oxidation	$1\times10^{-3}\times C_{tot}$	mmol/g-VSS	(4)
$K_{Mred,2}$	S_{Mred} affinity constant for NO reduction	$1\times10^{-3}\times C_{tot}$	mmol/g-VSS	(4)
$K_{Mred,3}$	S_{Mred} affinity constant for oxygen reduction	6.9×10^{-2}	mmol/g-VSS	(4)
$K_{Mred,4}$	S_{Mred} affinity constant for nitrite reduction	1.9×10^{-1}	mmol/g-VSS	(4)
C_{tot}	The sum of S_{Mred} and S_{Mox}, a constant	1×10^{-2}	mmol/g-VSS	(4)

Heterotrophic bacteria (HB)				
k_H	maximum hydrolysis rate of HB	0.125	1/h	(2)
K_X	hydrolysis saturation constant	1.0	mg-COD/mg-COD	(2)
μ_H	maximum growth rate of HB	0.26	1/h	(5)
η_{H1}	anoxic growth factor for nitrate reduction	0.28	—	(5)
η_{H2}	anoxic growth factor for nitrite reduction	0.16	—	(5)
η_{H3}	anoxic growth factor for N_2O reduction	0.31 ± 0.04	—	(3)
K_{OH1}	S_{O2} affinity constant for aerobic growth	0.1	mg-O_2/L	(5)
K_{OH2}	S_{O2} inhibit constant for nitrate reduction	0.1	mg-O_2/L	(5)
K_{OH3}	S_{O2} inhibit constant for nitrite reduction	0.1	mg-O_2/L	(5)
K_{OH4}	S_{O2} inhibit constant for N_2O reduction	0.1	mg-O_2/L	(5)
K_{S1}	S_S affinity constant for aerobic growth	20	mg-COD/L	(5)
K_{S2}	S_S affinity constant for nitrate reduction	20	mg-COD/L	(5)
K_{S3}	S_S affinity constant for nitrite reduction	20	mg-COD/L	(5)
K_{S4}	S_S affinity constant for N_2O reduction	40	mg-COD/L	(5)
K_{NO3}^{HB}	S_{NO3} affinity constant for HB	0.2	mg-N/L	(5)
K_{NO2}^{HB}	S_{NO2} affinity constant for HB	0.2	mg-N/L	(5)
K_{N2O}^{HB}	S_{N2O} affinity constant for HB	0.05	mg-N/L	(5)

Source: (1) Wiesmann, 1994; (2) Henze et al., 2000; (3) Estimated with standard errors by fitting the experimental data in this work; (4) Ni et al., 2014; and (5) Hiatt and Grady, 2008.

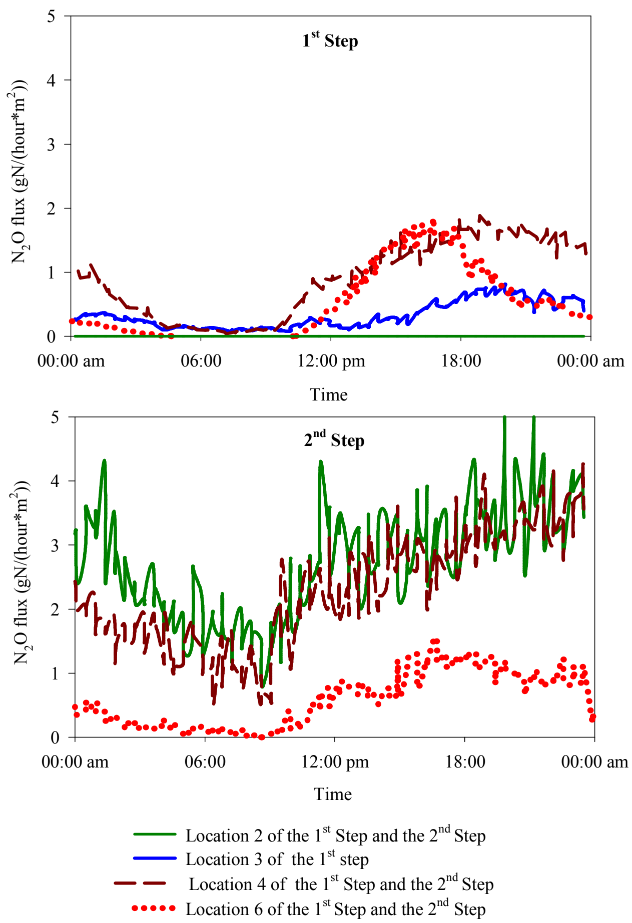

Figure S1. The typical spatial patterns of N_2O flux profiles from the First (upper) and Second (lower) Steps of the step-feed reactor (Pan et al., 2015).

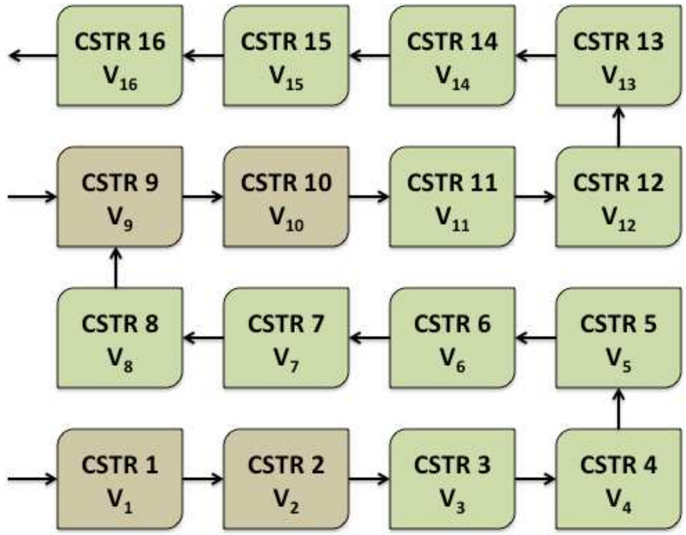

Figure S2. The 16 CSTRs configuration used in this work for describing the full-scale plug-flow reactor.

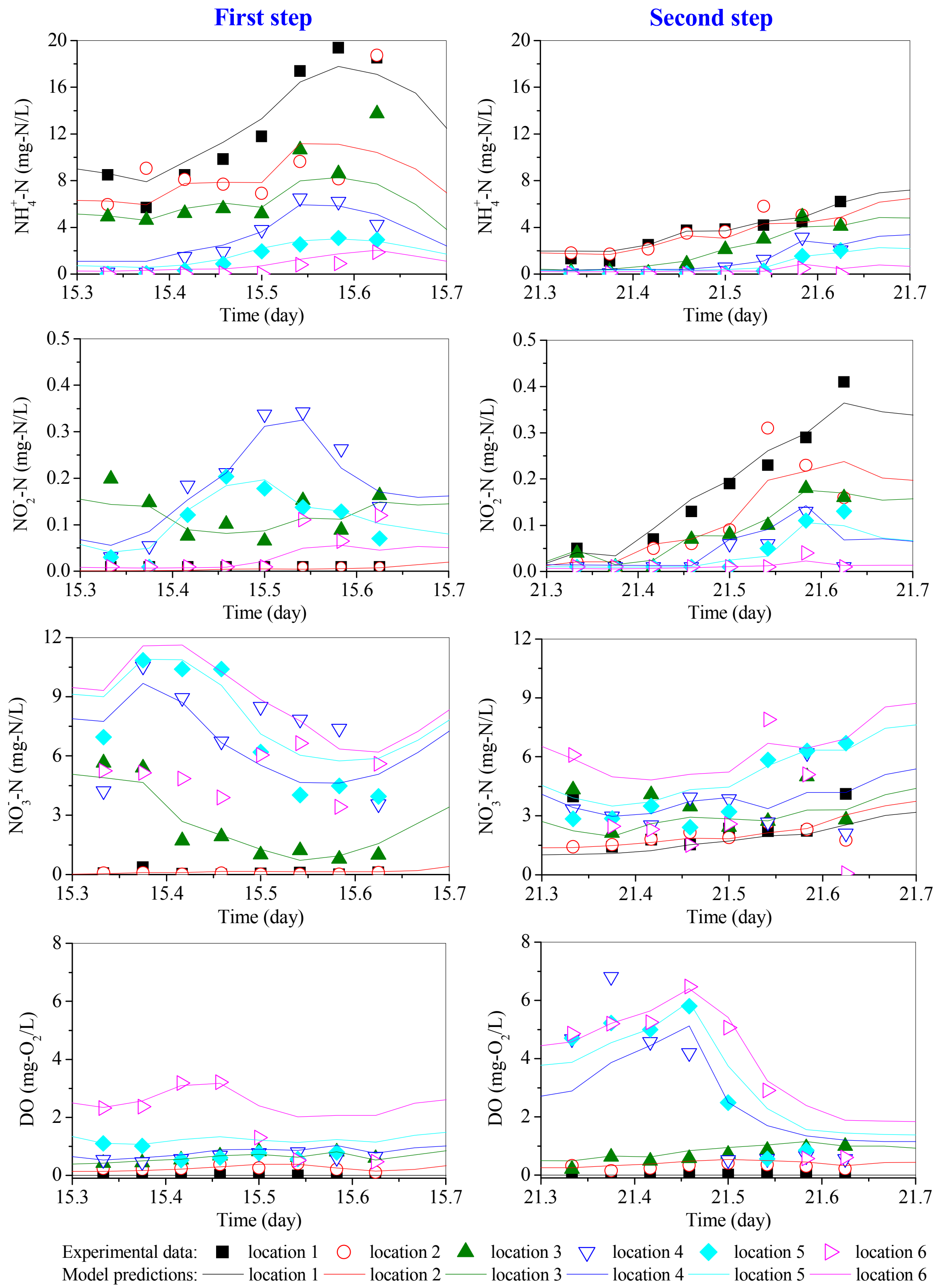

Figure S3. Model calibration results for nitrogen conversion using the 1-day intensive monitoring data (day 15) at different locations from the First Step (left panel), as well as 1-day intensive monitoring data (day 21) at different locations from the Second Step (right panel) (real data: symbols; model predictions: lines): ammonium, nitrite, nitrate, and DO profiles.

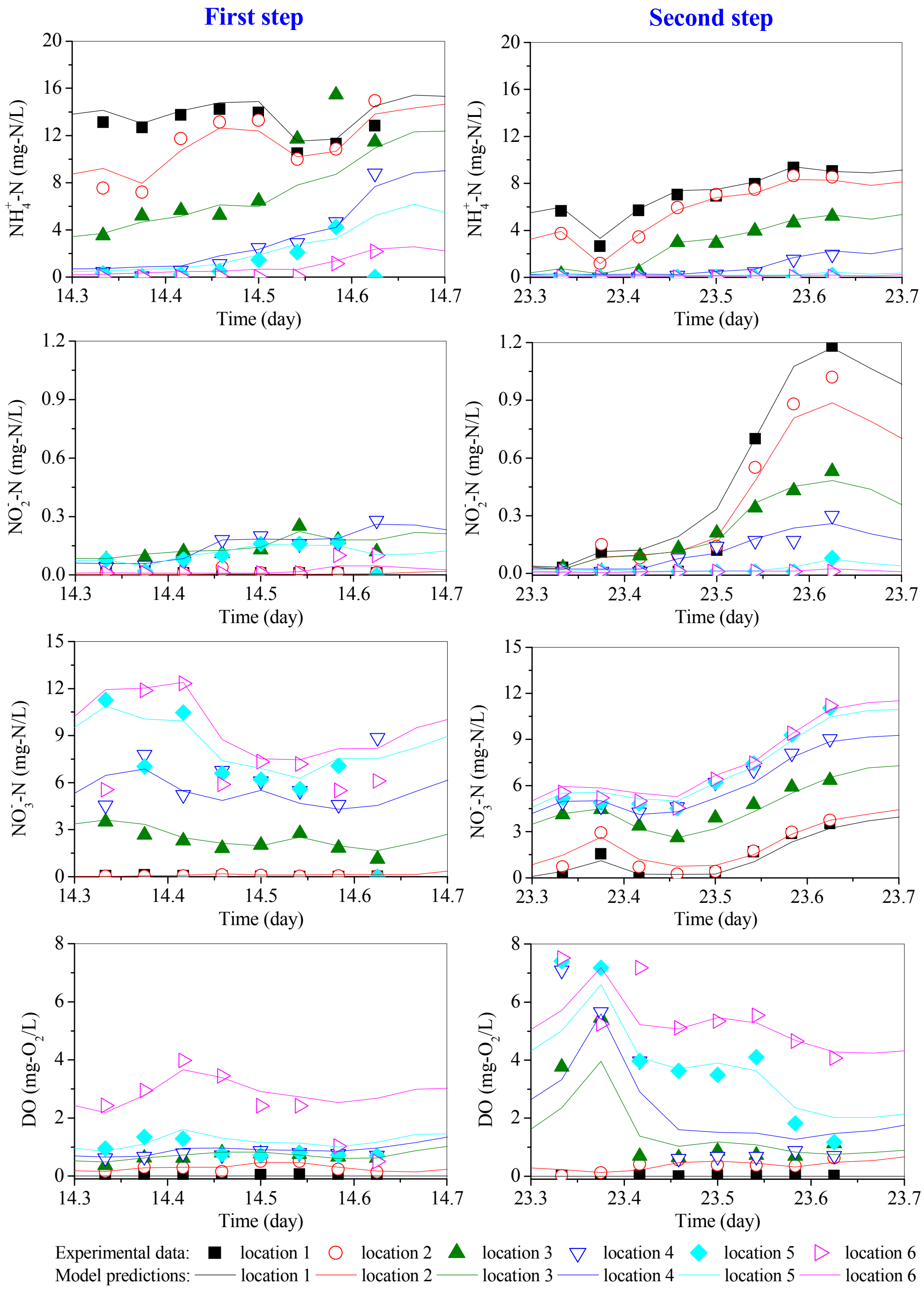

Figure S4. Model validation results for nitrogen conversion using the other 1-day intensive monitoring data (day 14) at different locations from the First Step (left panel), as well as the other 1-day intensive monitoring data (day 23) at different locations from the Second Step (right panel) (real data: symbols; model predictions: lines): ammonium, nitrite, nitrate, and DO profiles.

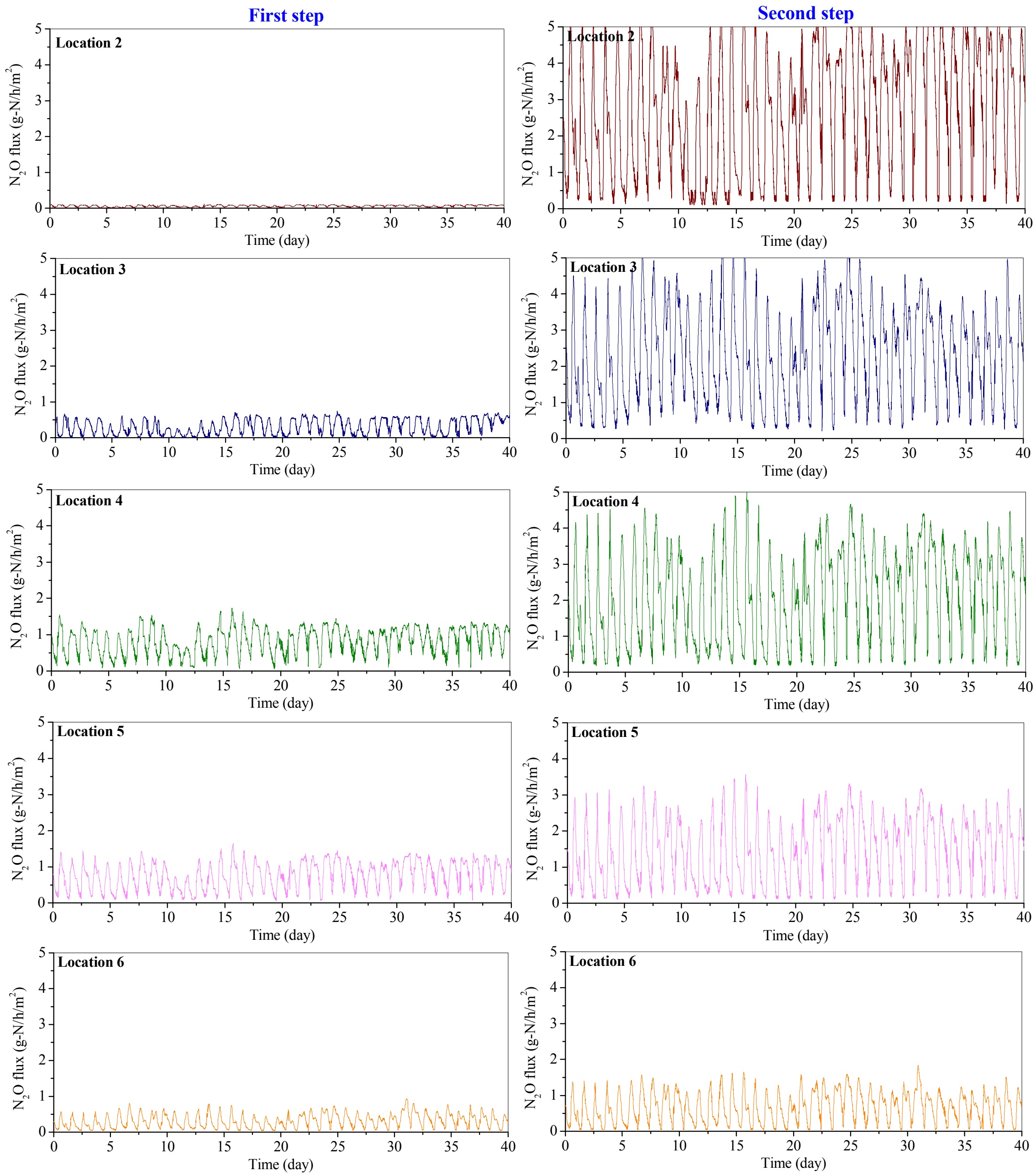

Figure S5. Model predicted N_2O fluxes during the 40-days operation of the step-feed reactor at five different locations from the First Step (left panel) and the Second Step (right panel).

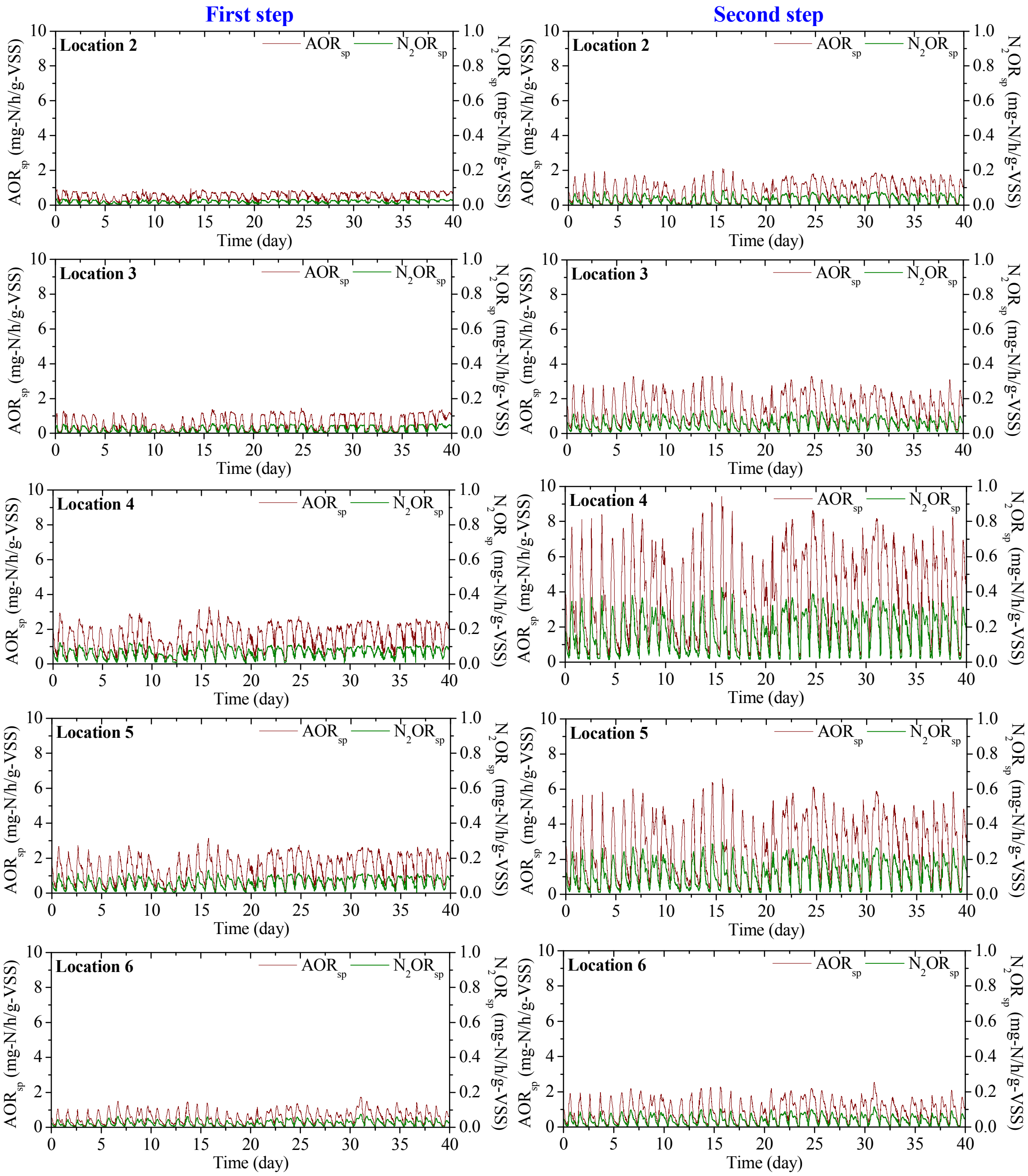

Figure S6. Model predicted biomass specific ammonia oxidation rate (AOR_{sp}) and biomass specific N_2O production rate (N_2OR_{sp}) during the 40-days operation of the step-feed reactor at different locations from the First Step (left panel) and the Second Step (right panel).

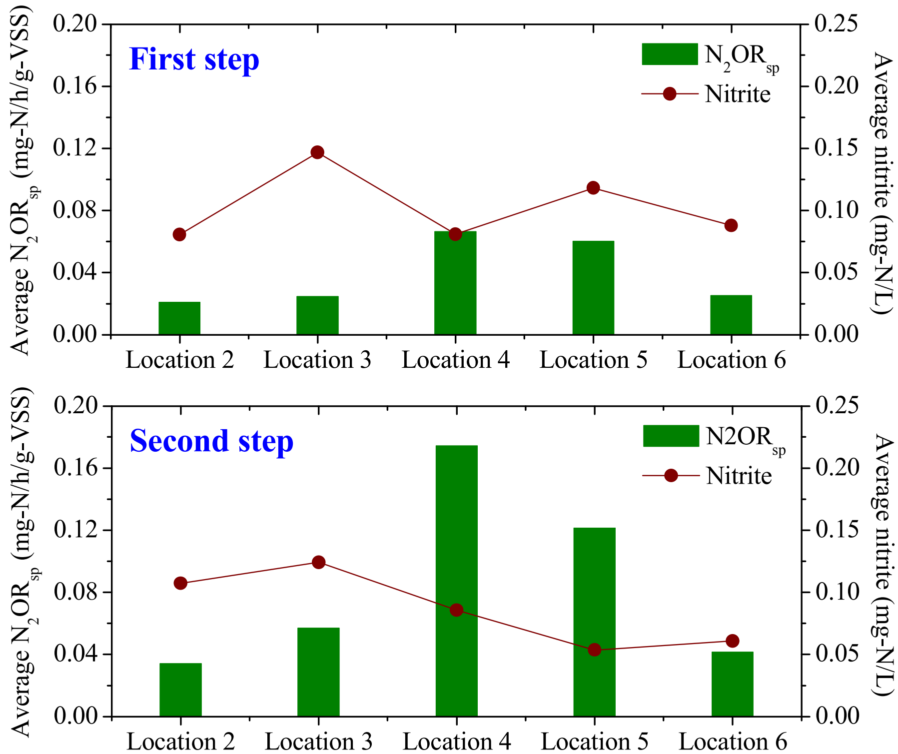

Figure S7. The comparison between the variations of average N_2OR_{sp} and average nitrite concentrations at the different locations of both steps.

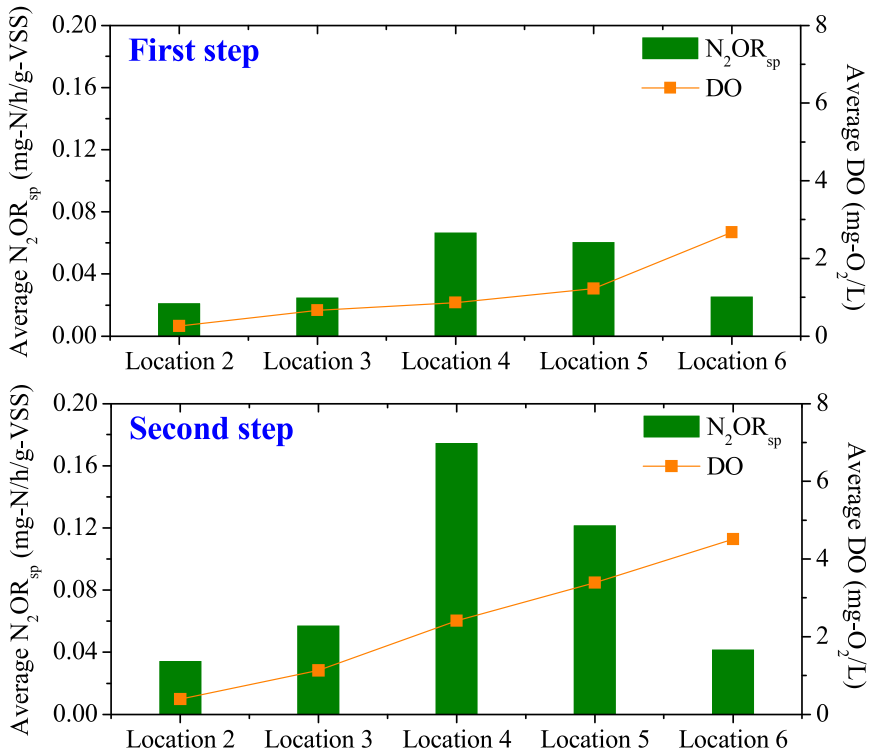

Figure S8. The comparison between the variations of average N_2OR_{sp} and average DO concentrations at the different locations of both steps.