

Modelling of sediment nutrient fluxes for a pulsed organic load

Abstract: A sediment model has been developed to simulate the Sediment Oxygen Demand (SOD) and sediment nutrient fluxes after a sudden input of Particulate Organic Matter (POM). Compared with previous sediment models, the main model advancements are the use of a continuous function to represent the kinetics of POM diagenesis and inclusion of the biomass content in POM diagenesis. Laboratory experiments also were carried out to investigate the SOD dynamics and sediment nutrient fluxes after an input of fish food pellets into the sediment. The simulation results of SOD and nutrient fluxes compared fairly well with the experimental measurements.

Keyword: Ammonia nitrogen, dissolved oxygen (DO), nutrients, sediment oxygen demand (SOD), sediment flux, water quality modelling.

1. Introduction

Sediment is regarded as a sink of particulate materials in natural waters. However, the physical, chemical and biological processes that take place in sediment may have a considerable impact on the quality of the overlying water body (DiToro, 2001). The degradation of settled organic matter causes a high sediment oxygen demand (SOD), which may lead to summertime oxygen depletion in lakes and estuaries (Floderus and Hakanson, 1989; McComb, 1995; DiToro, 2001). Furthermore, the dissolved organic matter and nutrients that are produced by diagenesis in the sediment can be returned to the water phase

and reused by the microbial community, resulting in water pollution and eutrophication. Therefore, material exchange and dissolved oxygen (DO) consumption at the sediment-water interface are of importance to the quality of surface waters. There is a need to fully incorporate the impact of sediment fluxes into the modelling and assessment of water quality in natural waters (Thomann and Mueller, 1987; Lee et al., 2000; DiToro, 2001).

Organic transformation in sediment is a complex process that is dependant upon the background nature of the sediment and the level of sediment organic load (Lung, 1993; McComb, 1995; DiToro, 2001). The parameterisation of the current sediment diagenetic model is primarily based on a few major studies in temperate waters with clean or slight polluted natural sediment (Thomann and Mueller, 1987; Lung, 1993; DiToro, 2001). However, in tropical and sub-tropical regions, sediment may receive a sudden load of rich particulate organic matter (POM) from major surface runoffs, the addition of fish food from mariculture activities and the settlement of biomass after a massive algal bloom. As a heavy organic load in warmer sediments is likely to promote more dynamic sediment activities and sediment fluxes, it is essential to investigate the kinetics of organic diagenesis and degradation in the sediment and the resulting SOD and material fluxes from the sediment.

The present study was carried out to develop a sediment model for the SOD dynamics, POM diagenesis and sediment fluxes after a pulsed input of organic into the sediment. The main improvements over previous sediment models (DiToro, 2001, Wang et al., 2006) include a multi-layer structural model for the sediment, a simple mathematical function to represent the kinetic features of sediment organic diagenesis and a detailed analysis of biological reactions and material transport in the sediment. In addition, the modified model takes into account the role of biomass content in POM diagenesis and biotransformation in the sediment. Laboratory experiments were carried with fish food pellets in the SOD

chambers to validate the results of the model simulation of the SOD dynamics and sediment fluxes following an input of rich organic matter.

2. Materials and methods

2.1 SOD apparatus and experiments:

SOD chambers were used to measure the SOD and organic and nutrient fluxes from the sediment after a POM load. The SOD chambers were fabricated from a Perspex cylinder with an internal diameter of 14.2 cm and a height of 14 cm (Lee et al., 2000). For each SOD experiment, surface marine sediment that was obtained from the Mai Po Nature Reserve in Hong Kong was placed in an SOD chamber to form a 3-cm sediment layer. A pre-determined amount of fish food pellets – either 0.5, 1.0 or 2.0 g – was then mixed into the sediment to simulate a load of rich organic matter. The corresponding POM loads were 31.3, 62.5 and 125 g/m² for the three load cases. Other experimental details can be found in Wang et al. (2006). The water in a SOD chamber was replaced regularly by flushing with DO-saturated fresh seawater. The average SOD value during a sampling period of one or two days can be determined using $SOD = \frac{(DO_0 - DO_\theta)V}{A\theta}$, where DO_0 and DO_θ are the DO concentrations in the water at the beginning and the end of the sampling period (θ), respectively, V is the water volume (2.3 L) and A is the area of the sediment surface (0.016 m²) of the SOD chambers. Similarly, the sediment fluxes of total organic carbon (TOC), J_{TOC} , ammonia-nitrogen, J_{NH} , and nitrogen-nitrogen, J_{NO} , can be determined following the chemical analysis of the water from the SOD chambers.

2.2 Model Formulation

2.2.1 *Multi-layer sediment model*: A multi-layer approach, rather than the double-layer sediment configuration, is adopted in the sediment model. In the multi-layer model, the sediment and the overlaying diffusive boundary layer (DBL) are divided into a series of fine layers with a thickness of 0.1 mm, or $\Delta H = 0.1$ mm. A uniform concentration is assumed for each chemical in a fine layer, and the mass transfer between conjunctive layers is carried out by molecular diffusion at a rate of $-D(C_i - C_{i-1})/\Delta H$, where the subscript i signifies a sediment or DBL layer, C is the chemical concentration and D is the diffusivity of the chemical (Lopez, 2001).

2.2.2 *Diagenesis of particulate organic matter (POM)*: Instead of the conventional 3- or 4-G approach (DiToro, 2001; Wang et al., 2006), the POM may be divided into many more fractions to form a more realistic and continuous profile of the rate constants for sediment POM diagenesis. In view of the importance of biological reactions, the biomass content also should be included in POM diagenetic kinetics. Without loss of generality, the following cosine-based function is proposed for the rate constant of POM diagenesis.

$$k_j = \begin{cases} \frac{1}{2} \left(\frac{\log X}{15} + k_0 \right) \left(1 + \cos \frac{f_j}{f_a} \pi \right) & f_j < f_a \\ 0 & f_j \geq f_a \end{cases}, \quad (1)$$

where j ranges from 1 to n specifying the POM classes, f is the corresponding POM mass fraction, k_0 is the maximum diagenetic rate constant of the POM, and the POM fractions of $1-f_a$ are apparently non-reactive to biological diagenesis. The flux of soluble organic carbon into the sediment layer from POM diagenesis can be then written as $J_c = \sum_j k_j f_j (POM) \Delta H$, (DiToro, 2001).

Diagenesis converts the POM into soluble organic matter that is further utilised as substrates by microorganisms in the sediment. The rate of soluble organic carbon (SOC) C is supposed to consist of two portions, or $C = S + C_{NS}$, where S is the biodegradable organic carbon (BDC) and C_{NS} is the non-biodegradable organic carbon (NBDC). Assuming that BDC and NBDC are produced following the same ratio of $f_a/(1-f_a)$, the rate of BDC production from POM diagenesis can be written as

$$\left(\frac{dS}{dt}\right)_d = \sum_{j=1}^n f_a k_j (POM)_j \quad (2a)$$

According to the composition of POM, the rate of ammonia production from POM diagenesis can be related to the SOC production in $\left(\frac{dNH}{dt}\right)_d = \left(\frac{f_N}{f_C}\right)\left(\frac{dC}{dt}\right)_d$, where f_N and f_C are the mass fractions of carbon and nitrogen in the POM. Thus,

$$\left(\frac{dNH}{dt}\right)_d = \frac{f_N}{f_C} \sum_{j=1}^n k_j (POM)_j \quad (2b)$$

2.2.3 Material transformation and mass balance: One-dimensional (y-axis) mass balance equations for the rate of change in organic carbon that results from diffusion in the DBL and reaction and diffusion inside the sediment can be written as follows (Wang et al., 2006).

$$\frac{\partial C}{\partial t} = D_{C,w} \frac{\partial^2 C}{\partial y^2} \quad (\text{DBL}), \quad (3)$$

$$\frac{\partial C}{\partial t} = D_{C,s} \frac{\partial^2 C}{\partial y^2} + \left(\frac{f_a}{1-f_a}\right)\left(\frac{dS}{dt}\right)_d - R_s \quad (\text{Sediment}), \quad (4)$$

where $D_{C,w}$ and $D_{C,s}$ are the diffusion coefficients of the soluble organic carbon in water and the sediment, respectively. The rate of organic carbon degradation in sediment R_s can be

related to the growth of heterotrophic biomass X in $R_S = \frac{1}{Y_S} \left(\frac{dX}{dt} \right)_G$, where Y_S is the yield coefficient of the biomass resulting from organic carbon utilisation (Bailey and Ollis, 1986).

Similarly, the overall mass balance of ammonia in each layer can be described as

$$\frac{\partial NH}{\partial t} = D_{N,W} \frac{\partial^2 NH}{\partial y^2} \quad (\text{DBL}), \quad (5)$$

$$\frac{\partial NH}{\partial t} = D_{N,S} \frac{\partial^2 NH}{\partial y^2} + \left(\frac{dNH}{dt} \right)_d - R_{NH} - R_{Biogrowth} + R_{Biolysis} + R_{Adsorp} \quad (\text{Sediment}), \quad (6)$$

where $D_{N,W}$ and $D_{N,S}$ are the diffusion coefficients of ammonia in water and sediment, respectively. The rate of ammonia oxidation in the sediment R_{NH} can be related to the growth

of ammonia oxidisers X_{NH} in $R_{NH} = \frac{1}{Y_{NH}} \left(\frac{dX_{NH}}{dt} \right)_G$, where Y_{NH} is the yield coefficient of the

biomass resulting from ammonia oxidation (Bailey and Ollis, 1986). $R_{Biogrowth}$ signifies the loss of ammonia due to the heterotrophic synthesis from the utilisation of organic matter, i.e. $R_{Biogrowth} = \gamma_n \left(\frac{dX}{dt} \right)_G$, where γ_n is the stoichiometric coefficient of nitrogen content in the

biomass. $R_{Biolysis}$ means the production of ammonia due to endogenous respiration by active biomass, or $R_{Biolysis} = \gamma_n f_d (k_d X + k_d X_{NH})$, where f_d is the biodegradable fraction of the

biomass and k_d is the decay coefficient (Rittmann et al., 2002). R_{Adsorp} is for the reversible adsorption of ammonia on the sediment solids. It is assumed that adsorption happens at a rate significantly faster than the vertical transport in the sediment; hence, adsorption equilibrium is always present. The amount of ammonia adsorption on the sediment solids is found to be proportional to ammonia concentration in the surrounding pore waters, i.e., $R_{Adsorp} = K \times NH$,

where K is the ammonia adsorption constant (Mackin and Aller, 1984; Berg et al., 2003).

Aerobic heterotrophic growth, ammonia oxidation and biomass maintenance are the three main biological processes that consume DO in the sediment. The total DO consumption rate in the aerobic layers can be written (Bailey and Ollis, 1986; Rittmann et al., 2002) as

$$R_o = \frac{1}{Y_o} \left(\frac{dX}{dt} \right)_G + \frac{1}{Y_{o,NH}} \left(\frac{dX_{NH}}{dt} \right)_G + m(X + X_{NH}), \quad (7)$$

where Y_o and $Y_{o,NH}$ are the effective yield coefficients from the utilisation of oxygen for heterotrophic and ammonia oxidising biomass, and m is a DO-based maintenance coefficient.

2.2.4 Biomass dynamic: The biomass dynamics can be described by the Monod equation for different growth limiting factors. Under aerobic conditions

$$\frac{dX}{dt} = \left(\mu_{\max,O} \frac{O}{K_o + O} \frac{S}{K_{s,O} + S} - k_{d,O} \right) X, \quad (8a)$$

and under anaerobic conditions

$$\frac{dX}{dt} = \left(\mu_{\max,A} \frac{S}{K_{s,A} + S} - k_{d,A} \right) X, \quad (8b)$$

where μ_{\max} is the maximum specific growth rate of the microbes for the growth condition indicated, and K_o and K_s are the half-saturation coefficients for oxygen and carbon utilisation, respectively (Bailey and Ollis, 1986; DiToro, 2001; Rittmann et al., 2002; Wang et al., 2006).

The ammonia oxidising biomass also can be described by Monod equation with two limiting factors, i.e.,

$$\frac{dX_{NH}}{dt} = \left(\mu_{\max,NH} \frac{O}{K_o + O} \frac{NH}{K_{s,NH} + NH} - k_{d,NH} \right) X \quad (9)$$

where $\mu_{\max,NH}$ is the maximum specific growth rate of ammonia oxidisers, K_{NH} is the half-saturation coefficient for ammonia utilisation and $k_{d,NH}$ is the decay coefficient of ammonia oxidisers.

2.2.5 Coefficients and parameters: A set of values for the bioreaction kinetic coefficients and other parameters used in simulation are given in Table 1. Most of the coefficients that are reported in literature have been converted to TOC-based values. The thickness of the diffusive boundary layer in the water column, δ_D , is estimated based on the hydraulic study of Lee et al. (2000) on the same SOD apparatus with internal circulation at a pumping rate of 4 L/h. $D_{C,W}$ for soluble organic matter with a high molecular weight is assumed to be a half of $D_{O,W}$ in water. The initial biomass in the sediment is assumed to be 100 mg/L in accordance with Higashino and Stefan (2005), and the initial TOC concentration in the sediment is 10 mg/L. Numerical simulations on the SOD, organic and nutrient fluxes for a pulsed sediment organic load were performed using a programme that was written in Turbo C programming language and run on a PC in the Windows XP environment. The time step of the simulation was set to 1×10^{-5} days.

3. Results and discussion

3.1 SOD dynamics

The SOD dynamics obtained from the laboratory tests and numerical simulations for 3 different fish food loads into the sediment are given in Figure 1. A sudden organic input resulted in a higher SOD, and the initial SOD value increased significantly with the amount of fish food loaded. The SOD for 2 g of fish food could be as high as $1.1 \text{ g O}_2/\text{m}^2\text{-d}$ according to the measurement, in comparison to the initial SOD of less than $0.4 \text{ g O}_2/\text{m}^2\text{-d}$ for the case of 0.5 g fish food. During the first three days, the difference in SOD for 2.0 and 1.0 g inputs was only $0.2 \text{ g O}_2/\text{m}^2\text{-d}$; thereafter, the difference increased to $0.5 \text{ g O}_2/\text{m}^2\text{-d}$. This was likely

due to a faster biomass growth for the high loading (2 g) case, which subsequently led to a higher rate of POM diagenesis and DO consumption. The SOD for the lowest load fluctuated around 0.4-0.5 g O₂/m²-d. In general, the simulation results compare fairly well with the SOD measurements, particularly for data of the initial loads of 1.0 and 2.0 g.

3.2 Nutrient fluxes

The diagenesis of POM, or fish food pellets in this study, produced soluble materials that either were utilised by the biomass in the sediment or diffused to the overlying water as sediment fluxes. The simulation and laboratory results of TOC and ammonia fluxes are showed in Figure 2 and Figure 3. TOC flux reached a high level within the first two days during the experiments and decreased gradually afterwards. With an initial load of 2 g, the TOC flux was about 500 mg/m²-d, which decreased to around 200 mg/m²-d after 9 days. For a low initial load of 0.5 g, the TOC flux decreased from 100 to about 10 mg/m²-d. In general, the simulation results are comparable to the laboratory TOC flux measurements (Figure 2).

For the sediment ammonia flux, however, a different trend of variation with time was observed. During the early phase of the tests, the ammonia flux increased continuously for all of the loading cases. The fluxes reached the maximum after about 4 days and then began to decrease gradually. For the load of 2 g, a peak ammonia flux of 700 mg/m²-d was detected on day 4, and the flux dropped to a level below 300 mg/m²-d after 10 days. It is likely that the trend of change in ammonia flux with the testing time was caused partially by the adsorption of ammonia in the sediment. With the POM diagenesis, both soluble TOC and ammonia were produced in the sediment. However, ammonia would be adsorbed initially by the sediment solids (Mackin and Aller, 1984; Berg et al., 2003). Only after the saturation of ammonia adsorption was achieved at a high level in the sediment had the ammonia flux from the

sediment become significant. As a result, for a fresh SOD test, the ammonia flux would increase gradually in the first a few days. Afterwards, with a reduction in ammonia from POM diagenesis and the utilisation of ammonia for biomass growth, the ammonia flux from sediment would decrease with time. The simulation results on ammonia flux were not fully consistent with the experimental results. However, the trend of ammonia flux variation was well predicted. In addition, the comparison in ammonia flux between the three loading cases was comparable to the laboratory observations (Figure 3).

3.3 TOC and ammonia concentration profiles in the sediment

The organic and ammonia concentration profiles in the sediment after a POM load were simulated (Figures 4 and 5). With the POM diagenesis, soluble TOC increased dramatically in the sediment, particularly in the deeper sediment. For the 2 g load, TOC increased from 10 to 300 mg/L at the sediment depth of 30 mm after 4 days. For the 0.5 g load, TOC increased to 35 mg/L at the same position. However, there was a much lower TOC accumulation in the surface layer (top 5 mm) for all of the loading conditions. This was likely caused by the diffusion of soluble organics from the top sediment into the water phase. In addition, a more intensive biological activity was expected for the top sediment layers, which would result in a faster organic degradation. Nonetheless, the accumulation of soluble TOC did not increase continuously in the sediment. Biodegradation of the organic became dominant after day 4, and for both loading cases the TOC content in sediment was reduced to a low level after 10 days (Figure 4).

The ammonia concentration profiles in the sediment were similar to those of soluble TOC (Figure 5). However, compared to the TOC profiles, the ammonia concentration decreased at a lower rate after the maximum being achieved. For example, for both loads of

2.0 g and 0.5 g, the ammonia concentrations after 10 days at different sediment depths were still much higher than those after 1 day or 2 days. This was probably caused by the fact that organics could be degraded and consumed in both aerobic and anaerobic sediment layers, while ammonia could not be consumed in the thick anaerobic layers. It would take a longer time for ammonia to diffuse from the bottom to the top layers, where ammonia could be oxidised by nitrification or diffuse into the overlaying water.

4. Conclusions

A sediment model has been developed to simulate SOD dynamics and related sediment material transport and fluxes following the input of a pulsed load of rich organic matter. The main modelling advancements are the provision of a multi-layer model in place of a double-layer sediment configuration and a continuous function, rather than the conventional 3-G classes, to represent the kinetics of POM diagenesis. In addition, the role of biomass content in POM diagenesis, organic degradation and nitrification are included in the model and simulation. The model is able to simulate the SOD in a dynamic system following a sudden organic load, and can predict the organic and ammonia fluxes from the sediment into the water column. The numerical results indicate a higher SOD and a greater sediment nutrient flux as the POM load increases. The results of numerical simulations of the SOD, TOC and ammonia fluxes compare well with the experimental measurements. The model developed in this study can be used to evaluate and assess the effect of organic pollutants in sediment on the water quality of surface waters.

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Table 1. Coefficients and parameters used in the model and simulations.

Parameter	Value	Reference
δ_D (mm)	0.5	Lee et al. (2000)
$D_{O,W}$ (cm ² /d)	2.0	DiToro (2001)
$D_{C,W}$ (cm ² /d)	1.0	
$D_{NH,W}$ (cm ² /d)	1.7	DiToro (2001)
f_a	0.9	Estimation based the biodegradation test of this study.
f_d	0.8	Rittmann et al. (2002)
ϕ	0.7	Measurement of this study.
k_o (1/d)	0.2	DiToro (2001)
$k_{d,O}$ (1/d)	0.06	Rittmann et al. (2002)
$k_{d,A}$ (1/d)	0.03	Rittmann et al. (2002)
K_O (mg/L)	0.1	Rittmann and McCarty (2001)
$K_{S,O}$ (mg/L)	10	Metcalf and Eddy (2004)
$K_{S,A}$ (mg/L)	30	Metcalf and Eddy (2004)
$K_{NO,O}$ (mg/L)	0.2	Reddy, 1998
K_{NO} (mg/L)	0.2	Reddy, 1998
$K_{NH,O}$ (mg/L)	0.5	Reddy, 1998
K_{NH} (mg/L)	1.5	Reddy, 1998
$K_{NH,AD}$ (mL/g)	1.6	Mackin and Aller, 1984; Berg et al, 2003
m (1/d)	0.2	Rittmann et al. (2002), Bailey and Ollis (1986)
Y_O	0.4	Bailey and Ollis (1986)
$Y_{C,O}$	1.0	Bailey and Ollis (1986)
$Y_{C,A}$	0.4	Bailey and Ollis (1986)
Y_{NH}	0.33	Reddy, 1998
μ_{max} (1/d)	8.0	Rittmann et al. (2001)
$\mu_{max,A}$ (1/d)	0.2	Rittmann et al. (2001)
$\mu_{max,NH}$ (1/d)	0.2	Reddy, 1998

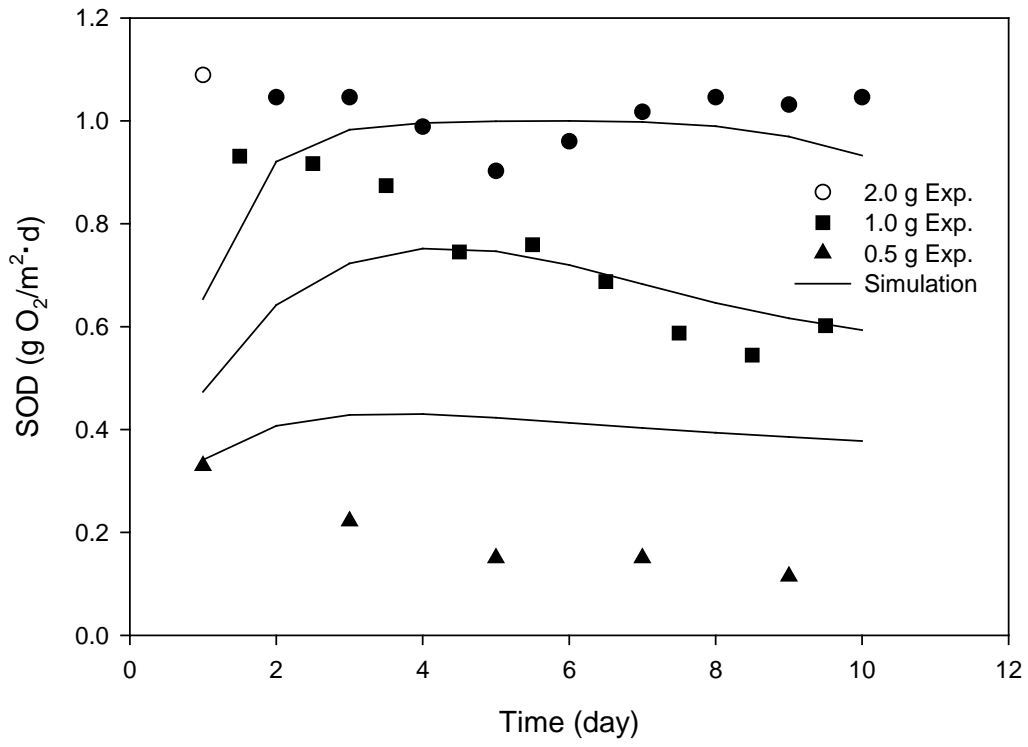


Figure 1. Experimental SOD results in comparison with the simulations.

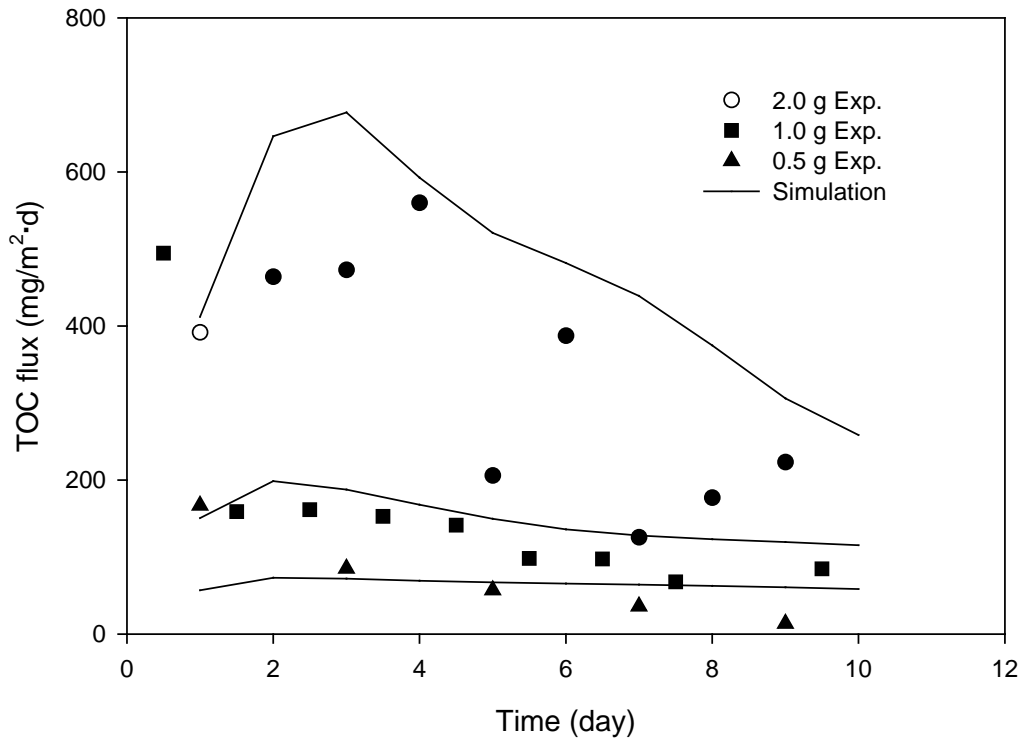


Figure 2. Experimental organic flux results in comparison with the simulations.

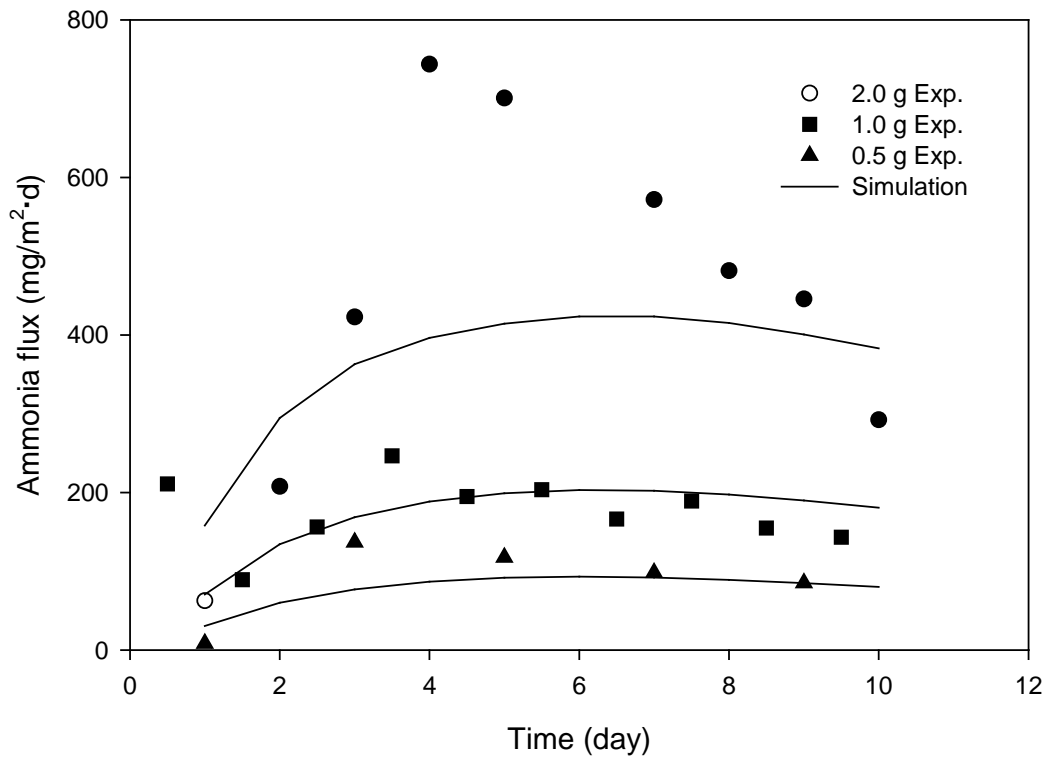


Figure 3. Experimental ammonia flux results in comparison with the simulations.

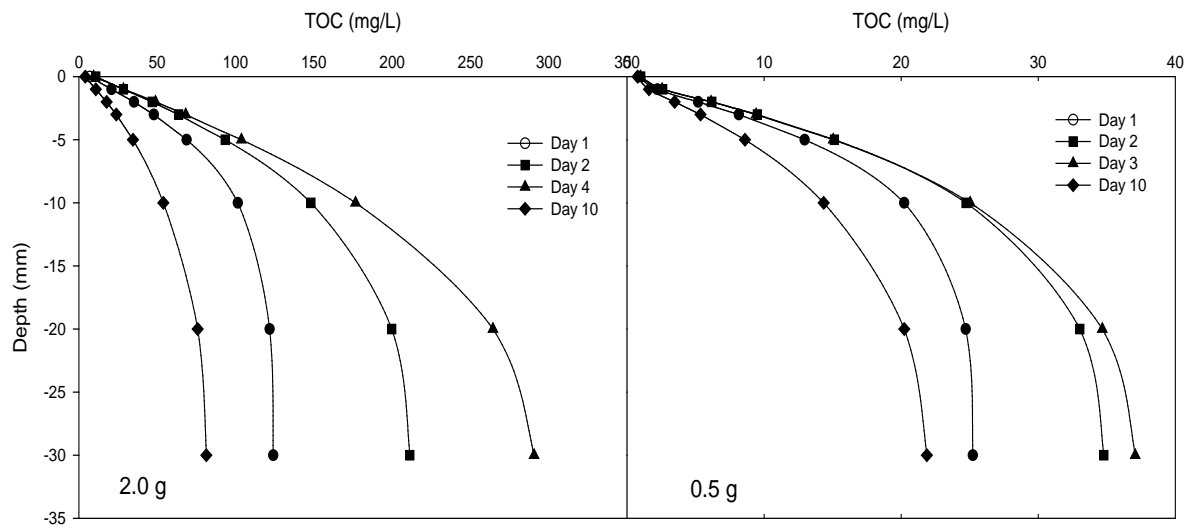


Figure 4. Soluble organic concentration profiles in the sediment for the fish food loads of 2.0 (left) and 0.5 g (right).

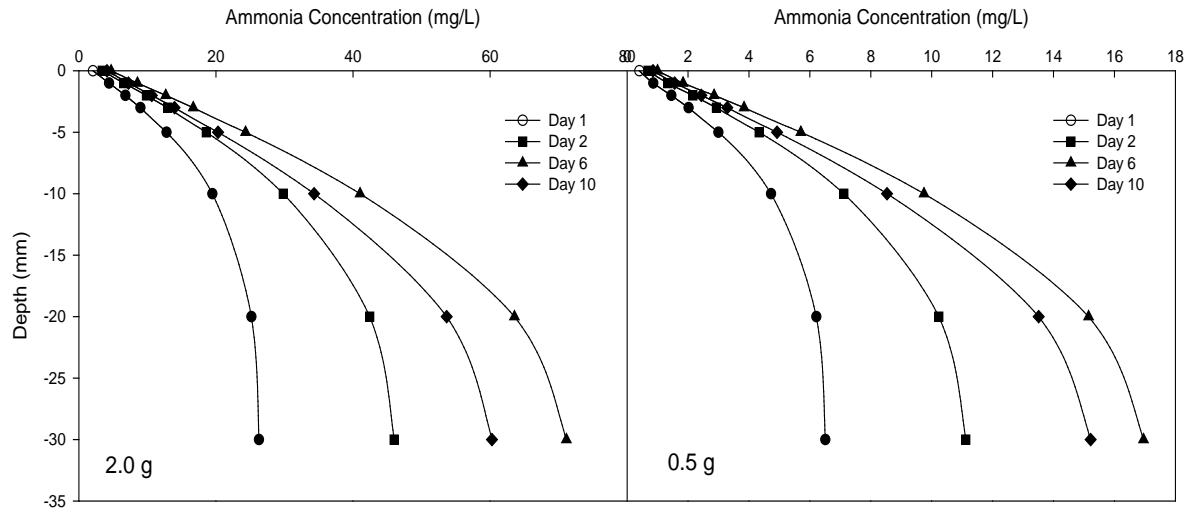


Figure 5. Ammonia concentration profiles in the sediment for the fish food loads of 2.0 (left) and 0.5 g (right).