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Assessment of contaminant flux from heavily polluted benthic sediment of Tongi Khal (canal): an ex-situ approach

Partho Das[a], Muhammad Ashraf Ali[a,*], Kazi Parvez Fattah[b]

[a]*Bangladesh University of Engineering and Technology, Dhaka-1000, Bangladesh,*
Tel. +8801713043325; email: ashraf@ce.buet.ac.bd (M.A. Ali), Tel. +8801724858796, email: partho@iwfm.buet.ac.bd (P. Das)
[b]*Department of Civil Engineering, American University of Sharjah, University City, P.O. Box: 26666, Sharjah, UAE,*
Tel. +97165152495, email: kfattah@aus.edu

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ABSTRACT

Polluted sediment bed can act as a secondary pollution source for the overlying water layer in polluted water bodies. This study investigates contaminant (selected nutrient and heavy metals) flux from the sediment of Tongi Khal (canal) through laboratory batch experiments. The results of this research suggest that accumulated pollutants in bed sediment are important contributors to the deteriorated water quality of Tongi Khal. Significant flux of ammonia (as high as 900 mg/m^2 d) from sediment to water column was observed. However, the ammonia concentration in water has been found to decrease with time together with an appreciable rise in pH, indicating significant algal uptake of ammonia; nitrification was not an important mechanism (for reduction of ammonia concentration) in the anoxic condition of the experimental systems. The flux of other nutrient constituents (e.g., phosphate) was not significant, but enough to support algal growth in the water column. The flux of Pb was insignificant suggesting that lead was strongly bound to the sediment layer in undisturbed condition. Though there was some flux of chromium from sediment to water, this was not very significant. However, results of this research suggest that physical disturbance of sediment could immediately make the overlying water layer anoxic, and promote enhanced release of nutrients (e.g., ammonia), organic matter, suspended solids, and heavy metals. Sustained disturbance, as could be expected during dredging, would severely deteriorate the water quality of Tongi Khal.

Keywords: Contaminant flux; Polluted sediment; Tongi canal; Nutrient flux

1. Introduction

Dhaka the capital city of Bangladesh is suffering from overpopulation, unplanned urbanization, and unregulated industrialization. The city is surrounded by five peripheral rivers, Buriganga, Balu, Tongi Khal, Turag and Sitalakhya, which are burdened with heavy pollution load from domestic and industrial sources. Due to direct discharge of untreated or semi-treated industrial wastewater (due to poor enforcement of environmental regulations) and dumping of untreated domestic sewage through numerous point and non-point sources (due to the absence of functional wastewater management system), ecosystems in all the surface water bodies surrounding Dhaka have become severely degraded [1].

Globally, legacy pollution is considered as one of the major problems in urban catchments. Sediments have long residence time in urban river systems and act as a major non-point source of pollution [2]. In natural aquatic systems, sediment can accumulate both organic and inorganic contaminants discharged into water bodies, and therefore a significant part of the contaminant load in watersheds often

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[*] Corresponding author.

gets accumulated in the sediment bed [3]. Once sediment is contaminated, it can cause lethal and sub-lethal effects in benthic (sediment-dwelling) and other sediment-associated organisms [4]. Researchers have termed contaminated sediment as a 'sleeping source' of contamination for river basins [5]. Sediments can influence the water quality in aquatic systems because under favorable conditions these can act as a secondary source of pollutants for the overlaying water [6]. Both natural and manmade events such as periods of anoxia, severe storms, flood, tidal movement, change in pH after acid rain, introduction of complexing agents, change in surface water composition, physical disturbance events such as dredging, fishing, bioturbation and so on can mobilize the accumulated pollutants from contaminated sediment bed [4,7–9]. However, contaminant fate is not well understood in both disturbed and undisturbed sediments and kinematics of metal release from sediment is also "poorly understood" [7].

The sediments of the peripheral river of Dhaka have turned into a sink of a diverse range of pollutants including domestic and industrial effluent as well as solid wastes [10,11]. Few studies were conducted on contaminant mobilization from sediment bed to upper water layer for the contaminated rivers in Bangladesh [10]. The primary objective of this study was to assess the flux of selected nutrients and heavy metals from benthic sediments of Tongi Khal.

2. Methods

2.1. Study area and sampling

Tongi Khal (canal) is one of the peripheral rivers around Dhaka city, originating from the Turag river and ending in the Balu river. This entire river system (Turag-Tongi-Balu river system) is one of the most polluted river systems in the world [12]. Tongi Khal is tidal and perennial with a discharge of about 35 m^3/s during the low flow months of January – March, and 205 m^3/s during the high flow month of August. This surface water system gets polluted from discharges from the nearby industrial clusters and domestic sources. Industries close to the river system are concentrated in the Tongi BSCIC area, Tongi industrial area, Cherag Ali, Ershad Nagar, Vhadam, Gazipura, Sataish, and Nimtoli areas. Polluting industries in these industrial areas include textile-dyeing, chemical-pharmaceutical, printing-packaging, glass-ceramic factories, food, and other miscellaneous industries [11]. Considering the deteriorated ecological condition in the river system, the Department of Environment (DoE) has designated the Tongi Khal (canal) as an "Ecologically Critical Areas (ECA)" in 2009 [13]. For the present study, sediment samples were collected from two distinct locations (S_1 and S_2, Fig. 1a) of Tongi Khal in December 2017 and January 2018, using Ekman grab sampler and following standard protocol [14,15]. One of these sampling locations (S_2) is located very close to the industrial cluster and the other (S_1) is located upstream of the cluster.

2.2. Experimental method

To estimate the contaminant flux from sediment to water column, two sets of batch experiments were performed. These were conducted in 48 cm × 30 cm (L × W) containers. In each container, a 2.5 cm layer of sediment was kept with a 16 cm layer of fresh groundwater on top (shown in Fig. 1b). Two types of sediment samples, collected from locations S_1 and S_2 of Tongi Khal (referred to in Fig. 1b as Sediment 1 and Sediment 2, respectively) were used in the first set of batch experiments carried out in four containers. In one of the two containers with the same type of sediment sample (i.e., either from S_1 or S_2 location), copper sulfate ($CuSO_4$) at a dose of 1 mg/L was added to the overlying water to restrict the growth of algae [16]. The first copper sulfate dosing was applied at the start of the experiment, and the second copper sulfate dosing was applied on Day 6 when algal growth was visually observed in the system. Water samples were collected daily from the water column in each container and analyzed for selected water quality parameters. The parameters pH, electrical conductivity (EC), redox potential (ORP), dissolved oxygen (DO), temperature, and turbidity were measured in-situ; while ammonia (NH_3–N), nitrate (NO_3–N), phosphate (PO_4^{3-}), lead (Pb), and chromium (Cr) concentrations were measured in the laboratory. The values of these parameters were used to estimate contaminant flux.

The second set of batch experiments was similar to the first set (described above), except in this case the entire system was physically disturbed (mixed) on Day 2 and Day 7 to assess the effect of physical disturbance on contaminant flux.

2.3. Contaminant flux determination

From measured water quality parameters, the contaminant flux across the sediment-water interface for each contaminant/parameter of interest was estimated using the following equation [17]:

$$J = \frac{\Delta M}{A.t}$$
$$\Delta M = \sum V(t)\left\{C(t) - C(t-1)\right\} \quad (1)$$

where J = flux (mass/time/area); t = time; ΔM = total mass change in water at time t; $V(t)$ = total volume of overlying water at time t; $C(t)$ = solute concentration at time t; $C(t–1)$ = solute concentration in sample before time t.

3. Results and discussion

3.1. Contaminant flux in undisturbed sediment-water system

Fig. 2 shows the changes in pH, DO, EC and turbidity in the water layer as a function of time. Fig. 2a shows that the pH increases with time in all systems. The rise in pH was most likely due to the conversion of organic matter to ammonia (referred to as ammonification) and incorporation of ammonia into algal cells. Ammonification involves conversion of nitrogenous organic matter into ammonia, and is accompanied by uptake of a proton (H^+); this results in a rise of pH of water. Algal growth is also associated with a significant rise in the pH of water due to the uptake of the proton during the formation of the algal cell. As noted above, $CuSO_4$ was added to water in some containers of the batch experiments to restrict the growth of algae. The rise in pH

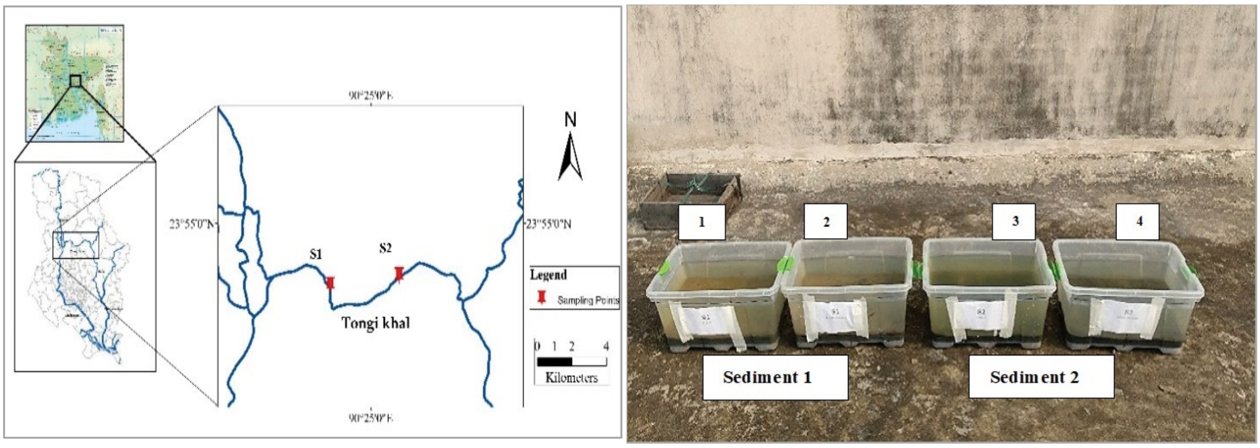

Fig. 1. (a) Sampling points along Tongi Khal and (b) sediment-water system for contaminant release experiment.

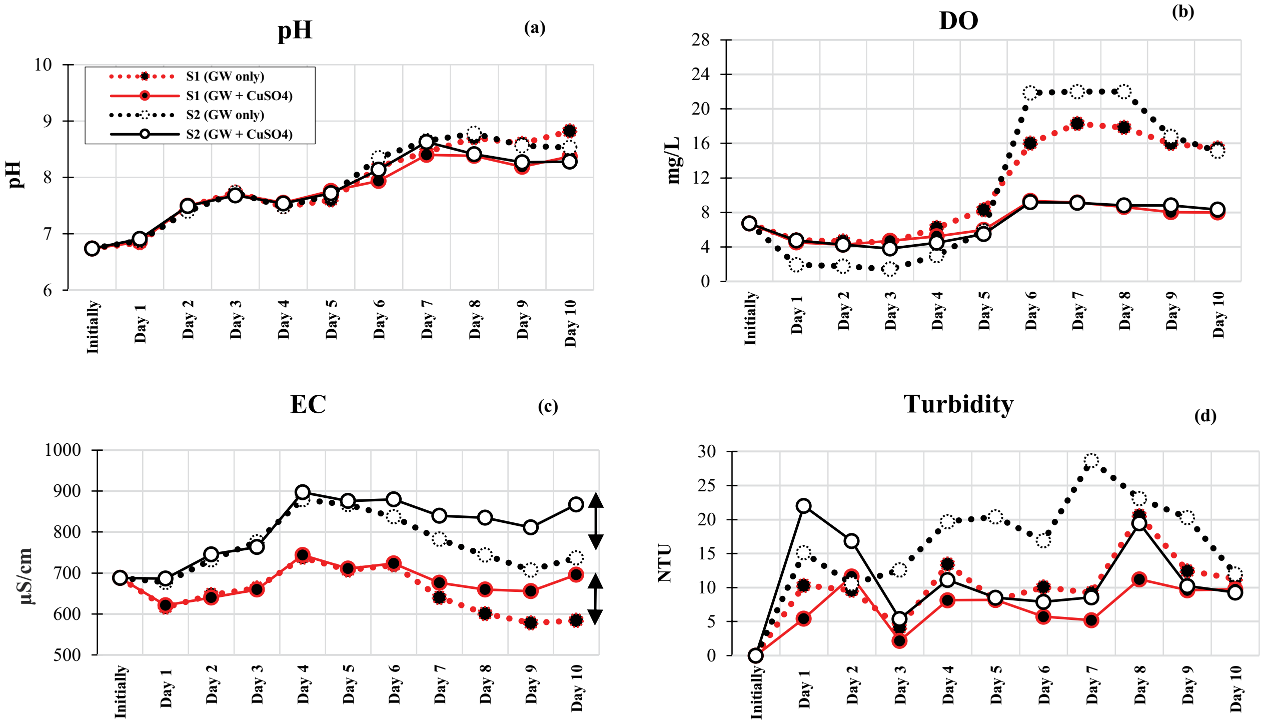

Fig. 2. Change in chemical parameters (a) pH, (b) dissolved oxygen, (c) electrical conductivity, and (d) turbidity of water layer in the experimental setup (system physically undisturbed).

was higher in experimental systems without copper sulfate. It indicates that copper sulfate played a role in restricting the rise in pH by reducing algal growth. It should be noted however that visual observation suggests that algal growth could not be stopped completely with the addition of copper sulfate, but the growth could be significantly reduced. Fig. 2b shows variation of DO in water as a function of time. It very clearly shows that the concentration of DO in water without copper sulfate is much higher than those with copper sulfate. This higher DO appear to come from algal photosynthesis. Fig. 2c shows variation of EC with time. It shows the higher release of dissolved ions from sediment S_2, which was collected from a location close to the industrial belt. This suggests that contaminated sediments would contribute more ions to the overlying water. Fig. 2c also shows that after about 4 days, EC was higher in the systems where algal growth was prohibited using $CuSO_4$. In these water samples, the presence of copper sulfate prevented/limited algal growth, and hence most dissolved C, P, and N remained in solution. On the other hand, in systems where algae could grow (in the absence of $CuSO_4$), the EC was lower due to uptake of dissolved nutrients in the formation of algae cells. Fig. 2d shows variation of turbidity with time. For systems without copper sulfate, turbidity was higher than the systems where algae growth was restrained using $CuSO_4$. This is most likely due to the contribution of algae to turbidity.

Changes in nutrients and heavy metals and associated fluxes in the water layer are shown in Fig. 3. Fig. 3a shows variation of ammonia concentration in water in all experimental systems as a function of time. Ammonia concentration increased in the systems up to about Day 4, most likely due to the ammonification (i.e., conversion of organic nitrogen into ammonia) process; it should be noted that the sediment from Tongi Khal contains high concentration of organic

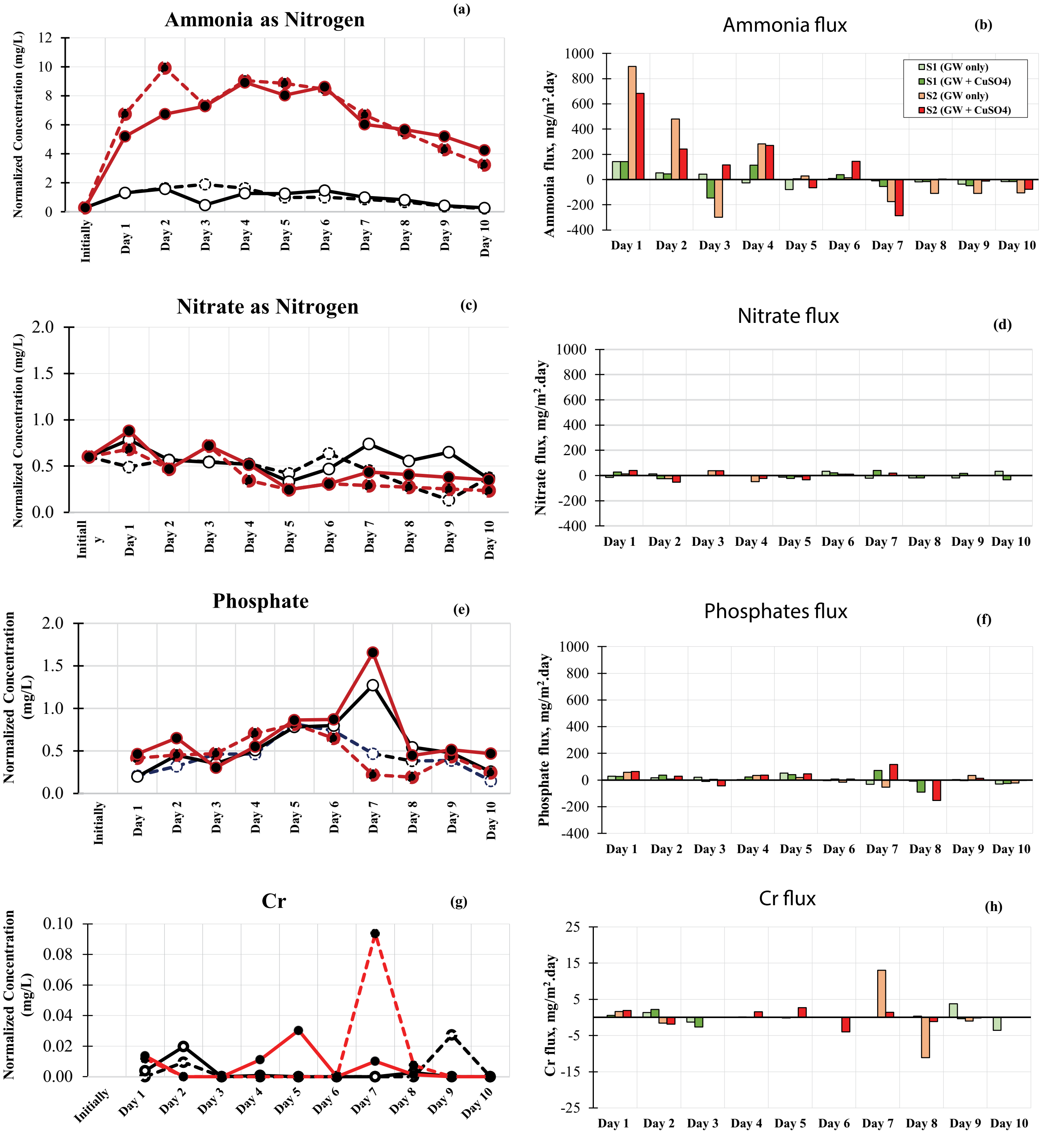

Fig. 3. (a) Change in ammonia concentration, (b) calculated ammonia flux from sediment to water column, (c) change in nitrate concentration, (d) calculated nitrate flux from sediment to water column, (e) change in phosphate concentration, (f) calculated phosphate flux from sediment to water column, (g) change in Cr concentration, and (h) calculated Cr flux from sediment to water column, as a function of time.

matter, ranging from 5.9% to 11.3% (w/w basis) [10]. From Day 5, the ammonia levels dropped most likely due to (a) reduction in ammonification process, (b) incorporation of ammonia into the algal cell, and (c) escape of ammonia to the atmosphere. However, the escape of ammonia to the atmosphere is unlikely to be significant under the pH condition (below 9.0) of the experimental systems. As shown in Fig. 3c, nitrate concentration did not increase during this period, suggesting that nitrification was not responsible for the reduction in ammonia concentration. Fig. 3a, however, does not show a significant difference in ammonia level in experimental systems with and without copper sulfate. Fig. 3b shows a significant positive flux of ammonia during the first four days of the experiment. This was due to ammonification or mineralization of organic matter present in the sediment, and its release into the water column. After that, the negative flux observed was most likely due to the incorporation of ammonia into the algal cell. Fig. 3d shows that nitrate flux was much less significant than that for ammonia, while a negative flux indicates uptake, most likely by algae.

Fig. 3e shows phosphate concentration in water as a function of time. After Day 5, in systems where algae growth was allowed (shown in dotted line), there was a sharp decrease in phosphate level. This is possibly due to the uptake of phosphate in the formation of algal cells. Fig. 3f shows lower fluxes of phosphate (compared to ammonia and nitrate); positive flux indicating release from sediment, while negative flux most likely indicating uptake by algae.

Lead concentration in the overlying water in the experimental systems was below the detection limit of 0.001 mg/L. This suggests that lead is strongly bound to the sediment. Fig. 3g shows Cr concentration in overlying water during the experimental system. Though some chromium was detected in water, the concentration was not significant, and there was no definite trend/pattern of chromium release. Fig. 3h shows that chromium flux was insignificant. Higher chromium flux was found from sediment collected from location S_2, possibly due to its proximity to industrial establishments.

3.2. Contaminant flux in disturbed sediment-water system

Fig. 4 shows changes in pH, DO, and EC in the water layer for the second set of experiments. The yellow arrows in the figure indicate the time/day when the sediment samples in the experimental systems were physically disturbed. Fig. 4a shows changes in pH in the experimental system and the effect of physical disturbance on the pH of the water layer. As observed with the first set of experiments, the pH showed an increasing trend with time due to the formation of algae and ammonification. Rise in pH was observed in both systems (with and without $CuSO_4$), indicating $CuSO_4$ was not very effective in suppressing the growth of algae. Algal growth was visually observed in both systems, though much less in the systems with copper sulfate. Fig. 4a shows that physical disturbance caused a sharp decline in pH. This reduction in pH could be due to sediment resuspension accompanied by the hydrolysis of newly formed ferric iron from ferrous iron oxidation [18,19].

$$Fe^{3+} + 3H_2O = Fe\left(OH\right)_3 + 3H^+ \qquad (2)$$

Fig. 4b shows the effect of physical disturbance on the DO of the water layer. In the system where algal growth was allowed, the DO level was higher due to the photosynthesis of algae. During both disturbance events (Day 2 and Day 7), the DO dropped immediately but increased with time. Fig. 4c

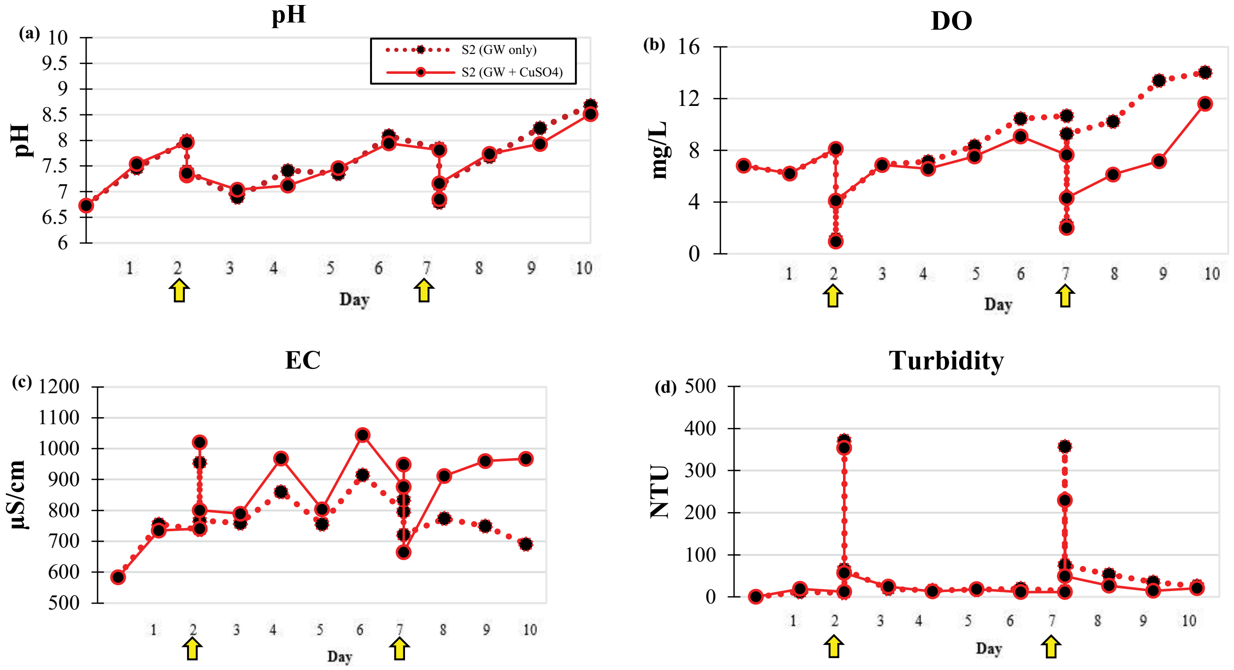

Fig. 4. Change in chemical parameters (a) pH, (b) dissolved oxygen, (c) electrical conductivity, and (d) turbidity of water layer in the experimental systems (systems physically disturbed).

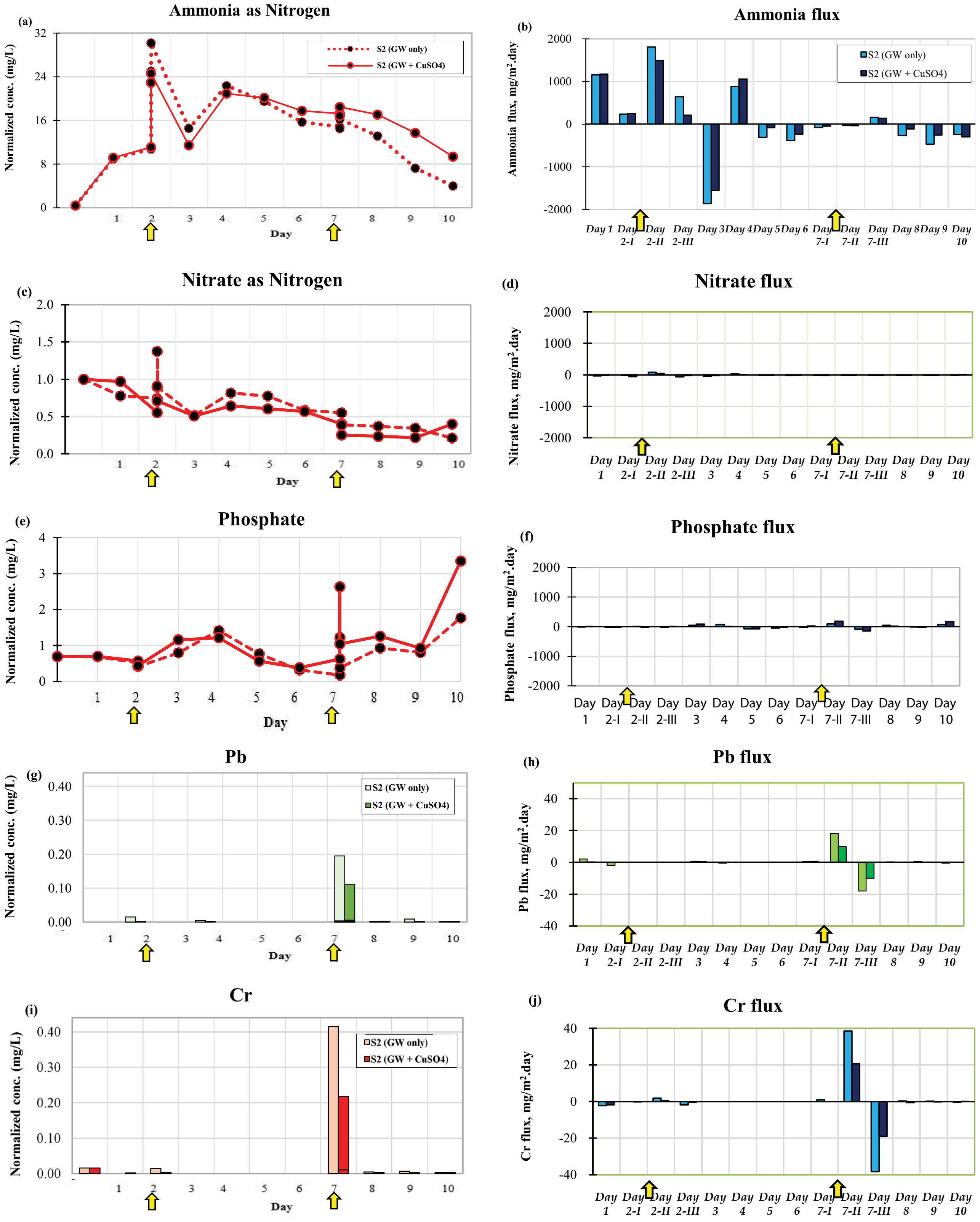

Fig. 5. (a) Change in ammonia concentration, (b) calculated ammonia flux from sediment to water, as a function of time, (c) change in nitrate concentration, (d) calculated nitrate flux from sediment to water, as a function of time, (e) change in phosphate concentration, (f) calculated phosphate flux from sediment to water, as a function of time, (g) change in lead (Pb) concentration, (h) calculated lead (Pb) flux from sediment to water, as a function of time, (i) change in chromium (Cr) concentration, and (j) calculated chromium (Cr) flux from sediment to water, as a function of time.

shows that EC exhibits a rising trend with time in all the systems, indicating that dissolved solids are gradually released into the overlying water. Systems where algae growth was allowed, the EC was found to be lower, due to the consumption of dissolved nutrients in the formation of algal cells. Physical disturbance caused an instantaneous sharp increase in EC due to the release of ion or contaminant from sediment. After the disturbance, EC dropped with time, possibly due to adsorption onto suspended materials and their subsequent settlement.

Fig. 5 shows the effect of physically disturbing the sediment samples on nutrients and heavy metal concentrations and associated fluxes for the second set of experiments; the two yellow arrows indicate the day when the sediment samples were physically disturbed. Fig. 5a shows a change in ammonia in the experimental system and the effect of physical disturbance on ammonia concentration in the water. The resuspension (physical mixing) of the sediment layer (on Day 2 and Day 7) caused an instantaneous increase in ammonia level. The maximum ammonia flux was observed immediately after the disturbance event on Day 2, indicating that the physical mixing of the sediment-water system promoted the release of ammonia from sediment. However, within a few hours, there was a sharp drop of ammonia concentration, followed by a rise; ammonia concentration then gradually declined as observed in the first set of experiments. As explained earlier, this is most likely due to a reduction in the ammonification process and the formation of algae. Fig. 5c shows that nitrate release from sediment was insignificant compared to ammonia release. It also indicates that ammonia-nitrate conversion was not significant. Similarly, phosphates release (shown in Fig. 5e) from sediment was insignificant compared to ammonia release. However, during both the disturbance events (on Day 2 and Day 7), there was a rise in the phosphate level in the water layer. There were some spikes in fluxes of both nitrate and phosphates after the resuspension event. It implies that the resuspension-flux of phosphates is higher than the normal diffusive flux. After 6 h of the resuspension event, there was a drop of phosphate values, possibly due to adsorption onto sediment particles and subsequent settling of the sediment particles.

Figs. 5g and i show changes in two Pb and Cr concentrations, respectively, in the system and effect of physical disturbance on these metal ion concentrations. Compared to the quiet condition, the effect of disturbance on the release of these metal ions was significant. The release of Pb and Cr was accelerated by the physical disturbance of the sediment-water system. However, within a short time after disturbance, the heavy metal concentrations became low, due to removal with settling sediment.

4. Conclusions

This study suggests that the accumulated pollutants in the sediment bed contribute to the pollution of the water of Tongi Khal. Laboratory experiments show significant flux of ammonia from sediment to water column under static condition; conversion of sediment-bound organic matter to ammonia by the ammonification process appears to be the source of ammonia. Ammonia concentration in water has been found to decrease with time together with an appreciable rise in pH, indicating significant algal uptake of ammonia; Nitrification was not an important mechanism in the anoxic condition of the experimental systems. Flux of other nutrient constituents (e.g., phosphate) was not significant, but enough to support algal growth in the water column. Flux of lead and chromium from sediment to water was not very significant. In summary, considering all the fluxes in the sediment-water system, it was found that ammonia flux (from sediment to water) was the most dominant. Phosphates, nitrate and heavy metal fluxes were not very significant in the quite condition under which these experiments were carried out. It was also found that physical disturbance of sediment could immediately make the water anoxic, and promote the release of nutrients (e.g., ammonia), organic matter and heavy metals. Sustained disturbance, as could be expected during dredging, would severely deteriorate the water quality of Tongi Khal.

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