Original Article

Phosphorus Flux across the Sediment-Water Interface in a Drainage Ditch Receiving Water from Livestock Farms in Kasaoka, Japan

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ABSTRACT

Internal loading of phosphorus (P) in drainage ditches may be sufficient to enhance eutrophication in aquatic systems under certain conditions. This study aimed to determine the effects of dissolved oxygen (DO) concentrations on P flux across the sediment-water interface. The study was conducted in a drainage ditch that receives discharge from livestock farms. The diffusion *in situ* was estimated according to Fick's first law equation using P concentrations in pore water and the overlying water. The overlying water and intact sediment cores were taken at the same location and time and incubated under anaerobic or aerobic conditions to determine the effects of DO on P fluxes based on the time-course of changes of P concentrations in water columns for 21 days in the laboratory. The diffusion of P across the sediment-water interface in the field was 9.79 ± 0.60 mg/(m² day) from sediment to the overlying water. In the laboratory, the fluxes of P were 1.76 ± 0.48 mg/(m² day) from sediment to overlying water under anaerobic conditions and 1.57 ± 1.05 mg/(m² day) from water into sediment under aerobic conditions. The upward diffusion of P under anaerobic conditions may be related to the reduction of iron-bound P. These results highlighted the role of oxygen in regulating P diffusion at the sediment-water interface. Accordingly, the low oxygen concentration in the drainage ditch was a reason for P release from sediment.

Keywords: flux, oxygen, phosphorus, sediment-water interface

INTRODUCTION

Phosphorus (P) is the most important element in the eutrophication process. The accumulated P in the sediment in agricultural drainage ditches poses a threat to the ecology because it can enhance eutrophication [1]. Drainage from agricultural farming is tightly coupled with downstream aquatic ecosystems. The concentrations of dissolved reactive phosphorus (DRP) and total phosphorus (TP) in a water body are dependent on internal and external loadings. Internal loading leads to a high P concentration in a water column although external loadings are curtailed [2].

Reduction-oxidation (redox) reactions play an important role in the fate of P [3]. When sediment is covered by water, a number of biogeochemical processes occur, resulting in either an increase or a decrease in the concentration of P in

the overlying water [4,5]. Under anaerobic conditions, the reduction of iron oxyhydroxides results in the diffusion of P from sediment into the water column [4]. Decomposition of plant tissues also increases the release of P into the water column. A high water temperature stimulates the activities of microorganisms, and thus promotes the degradation of organic matter (OM), resulting in the release of P [6].

The mechanisms that control P flux at the sediment-water interface are generally related to dissolved oxygen (DO) concentrations [4]. Phosphorus diffusion can be estimated *in situ* by measuring the change in P concentration in the water column inside a benthic chamber [1,7,8] or based on the concentration gradient in the pore-water profile according to Fick's first law [5]. The maximum P flux can be measured in the laboratory by a simple regression analysis of the concentration vs. time curve during incubation of the intact sediment-water column [2,4]. It is important to understand

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Fig. 1 Sampling locations: U (upstream), M (midstream), D (downstream).

how P diffuses across the sediment-water interface and its contribution to downstream aquatic systems. In this respect, there have been numerous studies that measured P flux in lakes [2,4,9,10], in river estuaries [5], and *in situ* wetland ditches [1]. However, measurement of P flux in drainage ditch sediment during both *in situ* and sediment-water column incubation has not been documented as an estimation of P internal loading.

Kasaoka Bay is located in the southwestern part of Okayama Prefecture, Japan. Agricultural activities on the reclaimed land with an area of 1,191 ha were established in 1990. The north area is divided into three sectors: 99 ha of livestock and horticulture (LHA), 176 ha of livestock farms (LA), and 132 ha of grass land (GLA) for dairy cattle. Algae blooms have occurred recently in drainage ditches, especially in the downstream areas and the stabilization pond (Fig. 1) because the concentrations of DRP in water of drainage ditches is as high as 0.30 ~ 1.37 mg/L. Sharpley *et al.* [11] noted that the eutrophication threshold value for

P was 0.02 mg/L. In addition, our surveys on water quality since 2008 have demonstrated that the ditches were contaminated with nitrogen (N) and P throughout the years [12]. In response to the problems, the estimation of internal loading of P is significant for eutrophication management. Determination of P flux from sediment to manage P losses from ditches are needed [13]. So far, although P release from and adsorption onto sediments of drainage ditches have been reported, the estimation of P flux from agricultural drainage ditch sediment have not been investigated. The aim of this study was to assess the P flux at the sediment-water interface of an agricultural drainage ditch *in situ* and to determine the effects of DO concentrations on P diffusion from intact sediment cores.

MATERIALS AND METHODS

Study area and sampling

Soil is classified as gray lowland soil for agriculture in

Kasaoka Bay. The agricultural land is 2 ~ 3 m below the sea level. The present study was conducted in a drainage ditch which receives discharge mainly from livestock farming (**Fig. 1**). The ditch is approximately 1,850 m long, 3 m wide and the water level is 0.35 m deep. The ditch used for this study is connected to the main canal downstream. The supply water is from Takahashi River to the farm pond through the pipelines. The water in the farm pond is redistributed to irrigate farmland areas. All water discharges from the agricultural areas are transferred by the main canal to the stabilization pond. Water in the stabilization pond is pumped out to the sea. The seawater enters into the main canal from the intertidal zone in the east. Concentrations of N and P in the studied ditch were higher than those in LHA and GLA ditches.

Intact sediment cores and overlying water samples were taken within a 1 m radius from the center of the ditch downstream. Nine intact sediment cores were manually obtained to 10 cm depth by inserting acrylic tubes (5.5 cm in diameter) directly in September, 2014. The sampling depth was determined because redox reactions occurred within 10 cm of drainage ditches [1].

Field flux estimation

Phosphorus diffusion was estimated in triplicate from the difference in concentrations of P in pore water and the overlying water, which were sampled and sectioned *in situ* into 0 ~ 2, 2 ~ 4, 4 ~ 6, 6 ~ 8, and 8 ~ 10 cm layers in plastic zipper bags. Inorganic P (IP) in pore water sample fractions and sediment porosities were analyzed in the laboratory. Upward diffusion of P from sediment into the overlying water at the sediment-water interface refers to material transport in response to a chemical potential gradient accorded to Fick's first law of diffusion [14] as Eq. (1):

$$J = -D_s \phi \frac{\partial C}{\partial z} \quad (1)$$

where J is the diffusion [$\text{mg}/(\text{m}^2 \text{ day})$], which is calculated for the top 2 cm of the sediment layer. The points 0 cm (sediment-water interface), -1 cm (below sediment-water interface), and +1 cm (above sediment-water interface) were considered for gradient estimation, D_s is the diffusion coefficient, $D_s = D_0(1 - \ln \phi^2)$, and D_0 is the molar diffusion, which is dependent on sediment porosity and temperature [15]. Its value at 25°C is $8.46 \times 10^{-6} \text{ cm}^2/\text{sec}$, $\partial C/\partial z$ is the concentration gradient of DRP in the vertical direction (concentration per unit distance, $\mu\text{g}/\text{cm}^3$), and ϕ is the porosity of the sediment, which was calculated according to the equation of

Berner [16]:

$$\phi = \frac{W_{pw}}{W_{pw} + W_s / G_s} \quad (2)$$

where W_{pw} is the weight of pore water in sediment expressed in grams, W_s is the weight in grams of dried sediment, G_s is the specific gravity, which is assumed to be a constant of $2.65 \text{ g}/\text{cm}^3$.

Intact sediment core incubation and flux measurement

Six intact sediment cores used for incubation experiments were capped on both sides under water by silicone stoppers and transported to the laboratory within 3 h in a cooler box. The internal loading of P was studied by experiments simulating two different DO concentrations (O_2 saturated and zero) and field temperature incubation (**Table 1**). The experiment was conducted in triplicate to estimate DRP and TP fluxes at the sediment-water interface. After being transported to the laboratory, the overlying water in the cores was siphoned off and replaced with approximately 120 mL of field water (5 cm overlying water) using a syringe without disturbing the sediment surface (**Fig. 2**).

For the anaerobic treatment, the overlying water was purged with N_2 gas (NP) until saturated after every day sampling, while aerobic sediment cores were maintained O_2 saturated with an aquarium pump (LUNG, GX500_N, GEX, Japan) during the experimental period. The P daily flux estimation is representative of real conditions in a ditch because DO concentrations and temperatures in the ditch fluctuate seasonally (**Table 1**). All sediment cores were wrapped in aluminum foil to exclude light in order to diminish algae development, then incubated at room temperature (25°C) for 21 days. Thirty milliliters of the overlying water was removed daily and replaced with field water. The average fluxes of DRP and TP in water columns for 21 days were calculated from the change of P concentration in the water column using the equation (3):

$$F = V \frac{(C_t - C_0)}{At} \quad (3)$$

where F is the net P release (positive values) or retention (negative values) [$\text{mg}/(\text{m}^2 \text{ day})$], C_t and C_0 [$\mu\text{g}/\text{cm}^3$] are concentrations of P on day 21 and day 0, respectively, V is the volume of the overlying water [cm^3], and A is the area of the sediment surface [cm^2].

Table 1 Water quality parameters in the drainage ditch in 2014.

Date/ locations	T [°C]	DO [mg/L]	pH [-]	EC [dS/cm]	TN [mg/L]	NO ₃ -N [mg/L]	NH ₄ -N [mg/L]	TP [mg/L]	DRP [mg/L]
January									
U	7.3	11.45	7.75	5.09	0.88	0.05	0	0.03	0.01
M	7.3	11.39	7.75	3.47	4.40	2.90	0.28	0.71	0.69
D	7.3	0.26	7.58	3.68	11.19	0.03	5.60	1.85	1.47
April									
U	17.8	9.65	8.04	4.96	0.91	0.14	0	0.23	0.18
M	19.3	9.76	8.21	3.95	2.10	2.19	0.01	0.98	1.02
D	18.4	2.97	7.96	3.86	6.34	0.94	2.35	1.46	1.42
September									
U	25.3	5.83	7.96	4.16	2.27	0.05	0.21	0.41	0.38
M	22.7	4.87	8.03	4.33	3.76	0.87	0.25	1.58	1.53
D	24.8	0.61	7.73	2.02	9.15	0	6.19	1.88	1.57
December									
U	7.8	9.30	7.62	2.87	4.20	3.17	0.01	0.17	0.15
M	8.2	11.73	7.59	3.38	14.89	11.91	0.11	1.41	1.29
D	8.1	7.4	7.38	3.40	15.90	11.53	0.45	1.39	1.23

T°C: temperature, DO: dissolved oxygen, EC: electrical conductivity, TN: total nitrogen, NO₃-N: nitrate nitrogen, NH₄-N: ammonia nitrogen, TP: total phosphorus, DRP: dissolved reactive phosphorus, U: upstream, M: midstream, D: downstream.

Fig. 2 Column incubation under different conditions: (a) NP (nitrogen purged) or OP (oxygen provided), (b) vent tubes, (c) sampling ports.

Sample analysis methods

Pore water collection

Sediment of each layer was centrifuged to separate gra-

dients of the P concentrations in pore water and filtered through a 0.2 µm membrane filter for DRP analysis [5,17].

Phosphorus fractionations procedure

The P fractionations was subjected to the sequential extraction scheme of Rydin [18]. The loosely absorbed P (loose-P) was extracted with 1 M NH₄Cl solution for 2 h. Iron-bound P (Fe-P) in the residue was extracted with 0.11 M NaHCO₃/Na₂S₂O₄ for 1 h. Subsequently, aluminum-bound P (Al-P) and organic P (Org-P) were extracted from the residue with 0.1 M NaOH for 16 h. While Al-P was directly analyzed from the extraction solution, another part of the solution was digested with K₂S₂O₈ for 30 min at 120°C, then the product in the form of PO₄-P was analyzed (total NaOH-P). The concentration of Org-P is the difference between the total NaOH-P and Al-P. The last fraction, calcium-bound P (Ca-P), was obtained by extracting the residue with 0.5 M HCl for 16 h. Extracted solutions were centrifuged at 3000 rotations per minute (rpm) for 10 min and the supernatant was filtered through a 0.2 µm membrane filter for fraction analysis.

Sample analysis

Water samples were analyzed according to the method of Murphy and Riley [19]. Water samples were filtered through a 0.2 µm filter (DISMIC-25AS, Advantec, Tokyo, Japan) to determine DRP concentrations. An unfiltered 10 mL water

Fig. 3 P fraction concentrations in sediment ($n = 3$).

sample and 2 mL reagent of $K_2S_2O_8$ were digested at 120°C for 30 min, and the product $PO_4\text{-P}$ was analyzed for TP. Concentration of $PO_4\text{-P}$ was determined by the spectrophotometric method using a continuous flow auto-analyzer (QuAatro 2-HR, Bltec, Tokyo, Japan).

Sediment samples were subjected to measurements of pH and electrical conductivity (EC) at soil: water ratio of 1:5 after shaking for 1 hour at 175 rpm, using a digital pH meter (F-23, Horiba, Kyoto, Japan) and EC meter (DS-14, Horiba, Japan), respectively. The total carbon (TC) and total nitrogen (TN) were determined by the dry combustion method with C/N Coder (CMT-700, Yanaco, Kyoto, Japan). The soil particle-size distributions were analyzed by the pipette method.

A two-tailed t -test was used to evaluate the significance of differences between the fluxes of DRP or TP under anaerobic and aerobic conditions. Statistical comparison tests were conducted at a level of significance of $p = 0.05$.

Fig. 4 Pore water P concentration profiles ($n = 3$).

RESULTS AND DISCUSSION

Water quality parameters of drainage ditch

Table 1 shows the differences in water quality parameters upstream, midstream, and downstream of the drainage ditch on different days in 2014. The concentrations of N and P downstream were higher than those upstream and midstream. Differences in N and P concentrations between sampling days were observed, and the highest concentrations of TP and DRP in drainage water were in September. The P concentration in pore water and its fractions in sediment are shown in Figs. 3 and 4. Iron-P, Ca-P, and Org-P fractions and pore-water P decreased with increasing depth, whereas the topmost loose-P and Al-P fractions were lower than those in the 2 ~ 4 cm layer. The main fraction of P, which was bound to calcium, occupied more than 50% of the total IP. A high concentration of the Ca-P fraction is in the form of $CaHPO_4$ and/or apatite ($Ca_5(OH)(PO_4)_3$) when the con-

Table 2 Physiochemical properties of drainage ditch sediment.

Sediment and water parameters		Concentrations
Sediment		
	TC [%]	4.32
	TN [%]	0.36
	TP [mg/kg]	640
	pH	7.8
	EC [dS/m]	2.5
Particle sizes	Clay [%]	7
	Silt [%]	66
	Sand [%]	27

TC: total carbon, TN: total nitrogen, EC: electrical conductivity.

Table 3 Phosphorus diffusive flux across sediment-water interface.

Samples	ϕ	$\partial C/\partial z$ [$\mu\text{g}/\text{cm}^4$]	J [$\text{mg}/(\text{m}^2 \text{ day})$]
1	0.57	-2.90	9.59
2	0.59	-2.47	9.33
3	0.62	-2.12	10.47
Mean $\pm$ SD	0.59 ± 0.02	-2.50 ± 0.39	9.79 ± 0.60

Fig. 5 Changes in DRP and TP in water columns. NP (nitrogen purged), OP (oxygen provided), DRP (dissolved reactive phosphorus), and TP (total phosphorus). “a” and “b”: Different letters indicate significant differences between the two treatments ($p < 0.05$).

centration of calcium is high in the environment [20]. The second largest P fraction was in organic form. Maximum concentrations of both total IP fractions and pore water P were recorded in the topmost sediment layer, which may be related to the OM concentration [5]. The high concentration of Ca-P in sediment was due to the calcareous recharge from the water inflow from the sea for dilution [21]. In addition, the biological processes induced CaCO_3 precipitation to P retention in calcareous lakes [9].

Phosphorus concentrations of pore water decreased from 7.25 mg/L in the topmost layer to 4.2 mg/L in the deepest layer. This distribution of P in different sediment layers may be related to physical conditions and exterior input of P [22]. Moore *et al.* [4] reported that higher DRP concentration near the surface sediment in summer increases mineralization of organic P and causes more reductive conditions. These indicate that the upward diffusion occurred from the top layer into overlying water and that the downward diffusion is from the top to deeper layers. Loosely-P and Fe-P fractions

were the main sources of P released from sediment to overlying water in drainage sediment in Kasaoka [21]. In addition, Fe-P is considered a redox sensitive form, whereas Al-P is redox insensitive and Ca-P is stable in the environment [2]. Accordingly, under anoxic conditions, Fe^{3+} is reduced to Fe^{2+} as a result of the P release from sediment. **Table 2** illustrates the fundamental characteristics of drainage ditch sediment. The silt accounted for the two-third of fraction distribution. High pH and EC values of the sediment suggest that the drainage sediment was affected by seawater intrusion from inflow (**Fig. 1**).

Phosphorus diffusive flux

The upward flux of P from sediment to the overlying water is described in **Table 3**. The diffusion value was relatively high due to the high concentration of P in pore water in the sediment top layer [23]. The average flux was $9.79 \pm 0.60 \text{ mg}/(\text{m}^2 \text{ day})$. The diffusion of P in the present study was much higher than that in previous studies. For example, the dif-

Table 4 DRP and TP fluxes in incubated columns.

Sediment-water columns	DRP <i>F</i> [mg/(m ² day)]		TP <i>F</i> [mg/(m ² day)]	
	NP	OP	NP	OP
1	1.71	-2.72	2.14	-3.02
2	2.26	-0.68	2.53	-2.35
3	1.31	-1.30	1.79	-1.49
Mean ± SD	1.76 ± 0.48 ^a	-1.57 ± 1.05 ^b	2.15 ± 0.37 ^a	-2.29 ± 0.77 ^b

^{a-b}Significantly different fluxes of DRP or TP between aerobic and anaerobic conditions ($p < 0.05$).

fusive flux in drainage ditches in the Catcott Lows wetland reserve ranged from 0.33 to 1.30 mg/(m² day) and the flux was significantly correlated with Al/Fe-bound P and degree of P saturation of sediment [1]. Likewise, the flux of P from sediment to overlying water in Suma Park, Australia was 0.10 ± 0.07 mg/(m² day) [24], $-14.46 \sim 4.92$ mg/(m² day) in the Pearl River estuary [5], and $0 \sim 6.19$ mg/(m² day) in Massachusetts, USA [6]. The pore water concentration regulated the P flux from sediment into overlying water [23]. The value of upward flux from top layer to the overlying water was much higher than downward fluxes from top layer to deeper layers.

Phosphorus-incubated fluxes

Figure 5 shows the temporal variations of DRP and TP concentrations in water columns under aerobic and anaerobic conditions for 21 days. The concentration of DRP in the water column increased gradually from 1.95 mg/L on day 1 to 2.40 mg/L on day 9, then remained stable until the end of the experiment under anaerobic conditions. In contrast, the concentration of DRP under aerobic conditions increased slightly from 1.65 mg/L on day 1 to 1.82 mg/L on day 8, then decreased dramatically to 0.92 mg/L by day 21. Total P patterns showed a similarity with those of DRP under aerobic conditions. The decline in concentrations of both DRP and TP in water columns was accompanied by oxygenated sediment cores after 8 days of incubation. The changes of DRP and TP concentrations were different depending on the time of incubation. Under anaerobic conditions, the steepest straight line of DRP was found on days 1 and 9, whereas that of TP was found in the third week of incubation. However, under oxygenation, the steepest drop of both DRP and TP were observed in the last week of incubation. In other words, the negative diffusion of P indicates that P came from the water column to the sediment, though P tended to be slightly released in the first week of incubation. Similarly, Zhang *et al.* [5] found that the negative fluxes in most sites in the Pearl

River estuary were attributed to iron hydroxide adsorption of P.

The maximum flux was observed under anaerobic conditions from day 1 to day 9 due to release from the sediment. The result showed that P fluxes quickly reached equilibrium on day 9 under anaerobic conditions and may be due to the high concentration of P in input water, indicating the role of P in the water column in regulating P flux from sediment [4]. Concentrations of DRP and TP in water columns did not change until the end of the experiment [20]. The incubated fluxes of DRP and TP were the differences between the two oxygen conditions ($p < 0.05$) shown in Table 4. Flux rates of DRP and TP were remarkably higher in anaerobic sediment cores, with values of 1.76 ± 0.48 and 2.15 ± 0.37 mg/(m² day), respectively, and anaerobic sediments acted as a source of P during the incubation. On the contrary, aerobic sediment cores acted as a sink of P in this study. On the average, daily rates of DRP and TP adsorptions onto sediment were 1.57 ± 1.05 and 2.29 ± 0.77 mg/(m² day), respectively. The amount of P absorbed into aerobic sediment cores was nearly three times higher than that released from anaerobic sediment cores. According to Hill and Robinson [1], the oxidized surface of the sediment results in reformation of Fe oxides with high capacity to absorb P. In addition, the flux was much greater under anaerobic conditions demonstrating physicochemical conditions, and negative flux was observed under aerobic conditions [4]. These results were comparable to the fluxes of P in Taylor Creek and Fisheating Creek under anaerobic conditions which were 12.3 mg/(m² day) and 3.7 mg/(m² day), respectively, suggesting that the water column DRP concentration can play an important role in P flux from sediment [4]. Under different oxygen levels, transformation between different P fractions occurred due to biological activities [25].

According to Nguyen *et al.* [21], P released from sediment to overlying water in Kasaoka drainage ditch was mainly from loose-P and Fe-P. Assuming that the top 0 ~ 10 cm of

sediment is responsible for P flux, the concentrations of P in these forms are sufficient to release P for a period of 5.4 years without external loading under anaerobic conditions. In fact, the top sediment suffered from high loadings of P from livestock farming. On the other hand, the sediment has a high potential to remove P from drainage water, with a value of $1.57 \pm 1.05 \text{ mg}/(\text{m}^2 \text{ day})$ when the sediment core is oxygenated. These results suggest that oxygen should be maintained in a drainage ditch to diminish P in both drainage water and downstream aquatic systems.

CONCLUSIONS

This study indicates the importance of the superficial sediment in regulating P flux across the sediment-water interface in drainage ditches. The diffusion of P from sediment into the overlying layer was $9.79 \pm 0.60 \text{ mg}/(\text{m}^2 \text{ day})$. This demonstrated that the drainage ditch receives discharge from livestock farming with a high concentration of P. The study emphasizes the importance of the oxygen concentration on P fluxes across the sediment-water interface of agricultural drainage ditches by estimating fluxes in intact sediment-water columns in the laboratory. When a ditch is maintained with an air supply, the sediment can increase P absorption up to $1.57 \pm 1.05 \text{ mg}/(\text{m}^2 \text{ day})$. However, $1.76 \pm 0.48 \text{ mg DRP}/(\text{m}^2 \text{ day})$ from sediment diffused into overlying water under anaerobic conditions. Our study may assist in planning the best technical options for water contamination management by improving P retention capacity in sediment.

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