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ORIGINAL ARTICLE

Quantitative evaluation of treatment processes and mechanisms of organic matter, phosphorus, and nitrogen removal in a multi-soil-layering system

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Abstract

The water treatment performance of multi-soil-layering (MSL) systems has been mainly evaluated by measuring differences in the quality of wastewater and treated water. Changes in water quality inside the system have not been assessed together with the water movement. In this study, therefore, quantitative evaluation of the treatment processes inside the MSL system was conducted using laboratory-scale MSL systems, which were set up in 10 cm (depth) × 50 cm (width) × 23–73 cm (height) acrylic boxes enclosing "soil mixture blocks" alternating with permeable zeolite layers. Six MSL systems consisting of one to six soil mixture layers (SML) were constructed, and treated water from the SML and the permeable layers between the SML (PLb) was collected separately at the bottom of each system. In order to determine the treatment processes inside the system, wastewater, with mean concentrations of biological oxygen demand (BOD) 28.1 mg L^{-1}, chemical oxygen demand (COD) 65.7 mg L^{-1}, total nitrogen 9.8 mg L^{-1}, total phosphorus 1.0 mg L^{-1}, was introduced into the system at a hydraulic loading rate of 1000 L m^{-2} day^{-1}.
Concentrations of both BOD and COD in the SML were lower than those in the PLb. As the flow rate in the SML decreased and the rate in the PLb increased, concentrations increased in the PLb in each system. Although the removal rate of BOD always exceeded 80% in the systems with more than three layers, six layers were required for COD to be reduced to the same extent as BOD. Phosphorus concentrations were also lower in the SML than in the PLb due to the adsorption of phosphorus by the soil and mixed iron particles. Therefore, phosphorus removal efficiency was strongly influenced by flow rate in the SML. In this study, the ammonium adsorption and nitrification were almost completed down to the third layer of the MSL system. However, the removal of nitrogen did not proceed much below the fourth layer due to low denitrification efficiency.

Key words: multi-soil-layering system, nitrogen and phosphorus removal, organic matter removal, processes of wastewater treatment, quantitative evaluation.

INTRODUCTION

In recent years, various engineering techniques have been applied to mitigate water related environmental

problems (such as eutrophication, and heavy metal and pathogen pollution) at local, regional and global levels. However, conventional environmental engineering (such as physical and chemical treatment and centralized sewage systems) is often energy and materials consuming and thereby causes its own environmental stresses (Etnier and Guterstam 1997). With these points as background, ecological engineering and ecotechnology have offered alternative ecologically sound and cost-effective methods.

Ecological engineering, or its synonym ecotechnology, is defined as "the design of sustainable ecosystems that integrate human society with its natural environment for the benefit of both" (Etnier and Guterstam 1997; Mitsch 1998; Day *et al.* 2003; Mitsch and Jorgensen 2003). Water purification technology based on soil ecology is part of ecological engineering/ecotechnology. These include land treatment systems, natural and constructed wetlands, reed beds, sand filters, and septic tank and trench systems (Reed *et al.* 1995; Etnier and Guterstam 1997; Crites and Tchobanoglous 1998; Van Cuyk *et al.* 2001; USEPA 2002; Rodgers *et al.* 2004, 2005; Davison *et al.* 2006; Mariappan and Viraraghavan 2006). The physical, chemical, and biological properties of soil have been used for wastewater treatment systems world-wide for many years.

To maximize the water purifying function of soil, our group has investigated multi-soil-layering (MSL) systems. The MSL systems consist of soil units (soil mixture layers, SML) arranged in a brick-like pattern surrounded by permeable layers (PL) of zeolite or alternating particles of homogeneous sizes that allow a high hydraulic loading rate (HLR). The MSL system is effective for the prevention of clogging and shortcut water flow which are major constraints in conventional soil-based wastewater treatment systems.

In domestic wastewater treatment using the MSL system in Japan, Wakatsuki *et al.* (1993) and Luanmanee *et al.* (2001) showed that mixing metal iron and jute pellets with SML was effective for removing phosphate and nitrate. Furthermore, the purification capacity could be controlled by adopting adequate aeration, even after 10 years (Sato *et al.* 2002a). Likewise, river, dairy effluent, and cafeteria wastewater treatments have been carried out in Japan, Thailand, and Hawaii using locally available materials, such as soil, iron particles, jute, sawdust, charcoal, and zeolite or alternative materials (Luanmanee *et al.* 2001, 2002a; Unno *et al.* 2003; Sato *et al.* 2005a; Pattnaik *et al.* 2008). In addition to such demonstrations, several laboratory scale tests have been carried out in order to understand water purification capacities and characteristics. Masunaga *et al.* (2007a) examined characterization of wastewater treatment using a MSL system at two levels of wastewater contamination with mean biological oxygen demands (BOD) of 30 and 70 mg L^{-1}, respectively, and at HLRs of 500 to 2000 L m^{-2} day^{-1}. Despite fluctuations in the wastewater contamination levels, BOD and total phosphorus (T-P) concentrations in the treated waters remained consistently lower than those in the wastewaters. However, total nitrogen (T-N) concentrations in the treated water fluctuated, being influenced by levels in the wastewater. Chen *et al.* (2007b) investigated the relationship between the size of the

SML and the removal efficiency of selected contaminants. At HLRs of 1000 and 2000 L m^{-2} day^{-1}, systems with larger SML surface area had higher removal rates for suspended solids (SS), BOD, chemical oxygen demand (COD) and T-P, probably because of the enhanced contact efficiency between wastewater and the SML. In addition, various basic researches such as effects of aeration position and timing, greenhouse gas emissions, and removal of colored substances have been demonstrated (Luanmanee *et al.* 2002b; Sato *et al.* 2002b; Boonsook *et al.* 2003; Chen *et al.* 2007a; Masunaga *et al.* 2007b).

Although a large number of basic and applied studies have been undertaken as noted earlier, the quantitative processes involved in the wastewater treatment in the MSL system remain unclear. So far, the water treatment performance has been mainly evaluated from differences in the quality of wastewater and treated water, and changes in water quality inside the system have not been assessed together with the water movement. Therefore, parameters such as the number of layers and the apparatus size and height have been established empirically. Although experiments in which water was collected inside the MSL system during wastewater treatment have been conducted (Wakatsuki *et al.* 1993; Sato *et al.* 2005b), only semi-quantitative evaluation of the treatment processes has been achieved. This is because only partially treated water was collected from several parts of a MSL system. Quantitative elucidation of the wastewater treatment processes will enable an improved understanding of the relationship between the water purification capacity and the structure and material composition of MSL systems, and thereby contribute to improving their water treatment efficiency. In the planning stage of design, quantitative evaluation is required in order to set a target for treated water quality against wastewater conditions such as contaminant concentrations and the HLR. In this study, therefore, we considered that it was necessary to investigate the whole quantity and quality of treated water inside a MSL system for quantitative evaluation, including relationships between the number of layers and changes in treatment performance. Additionally, to investigate the detailed treatment processes in the MSL system, wastewater treatment characteristics in the SML and PL between the SML (PLb) were studied.

Six laboratory-scale MSL systems consisting of SML with one to six layers were constructed for the purpose of quantitatively evaluating the treatment processes. Treated water from the SML and the PLb was collected separately at the bottom of each system. An attempt was made to investigate the wastewater treatment mechanisms in each layer by measurement of both the quantities and qualities of treated water from the SML and the PLb.
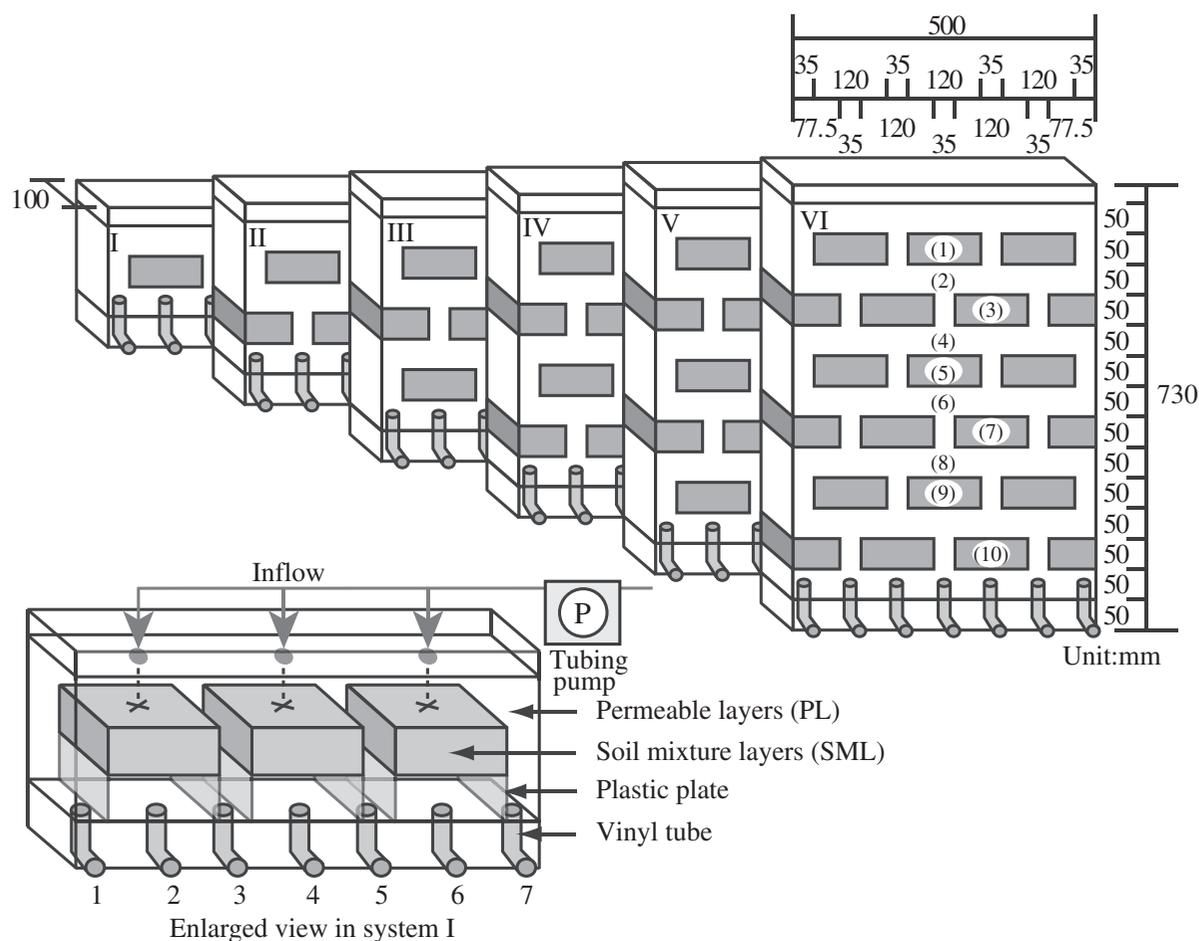
Figure 1 Structures of multi-soil-layering (MSL) systems for studies on quantitative evaluation of treatment processes inside the systems. Each system and outflow position are labeled as I–VI and 1–7, respectively. Numbers (1)–(10) in system VI show positions of the Eh electrodes.

MATERIALS AND METHODS

The multi-soil-layering systems used in this study

Figure 1 shows the structure of the laboratory-scale MSL systems used in this study. They were constructed using acrylic boxes with dimensions of 10 cm (depth) × 50 cm (width) × 23–73 cm (height), forming an alternate brick layer-like pattern with zeolite and the SML. The height of both the PL and the SML was 5 cm and the six MSL systems consisted of one to six SML. To collect the outflow water from the SML and the PLb separately, plastic divider plates were installed at the bottom part of the systems as shown in Fig. 1. A 1.2 cm diameter hole was made at the bottom of each partition part and a vinyl tube was attached to each hole. The SML consisted of a volcanic ash soil rich in organic matter and classified as an Andisol (United States Department of Agriculture 1999), sawdust, approximately 1 mm diameter granular iron, and charcoal in ratios of 70%, 10%, 10%, and

10%, respectively, on a dry weight basis. The Andisol was obtained from the A horizon of a soil at Mt Sanbe in Shimane, Japan, and had total carbon and T-N contents of 5.5% and 0.3%, respectively. The bulk density of the SML was $0.84\,\mathrm{g\,mL^{-1}}$. The void spaces (PL) between each block and block side were filled with zeolite particles 3–5 mm in diameter. Each system was numbered to show the number of layers in the system (Fig. 1). In addition, oxidation–reduction potential (Eh) inside the system was measured using 10 platinum electrodes installed from the side in system VI, as shown in Fig. 1. The resulting oxidation–reduction potentials are presented in a report by Sato *et al.* (2011, Fig. 6).

Quantitative evaluation of wastewater treatment processes in the mult-soil-layering systems

Domestic wastewater from a nearby community disposal plant was diluted three times with well water and introduced into these systems for quantitative evaluation

of the wastewater treatment processes. The wastewater was diluted in a 1000 L storage tank approximately every three days. Average characteristics of the diluted wastewater were: pH 7.71, SS 14.9 mg L^{-1}, BOD 28.1 mg L^{-1}, COD 65.7 mg L^{-1}, T-N 9.8 mg L^{-1} and T-P 1.0 mg L^{-1}. The HLR was set at 1000 L m^{-2} day^{-1}. The inflow water was divided three ways using a peristaltic pump. The three points were positioned right above the centers of the top layer of the three SML of the systems, as shown in Fig. 1. The experiment was started on August 12, 2005 and terminated on December 22, 2005 (132 days). The outflow water was collected from each vinyl tube and sampled five times on August 15 (Day 3), August 30 (Day 18), September 29 (Day 48), November 9 (Day 89), and December 7 (Day 117). A sufficient amount of water for analysis (≥ 500 mL) was collected for 30 minutes to 42 hours from each tube.

Wastewater and each outflow water sample (treated water) from tubes under the SML and the PLb were analyzed for the following: BOD using a dissolved oxygen (DO) meter, COD using the potassium dichromate method, ammonium nitrogen (NH$_4$-N) concentration using the Nesslerization method, nitrate nitrogen (NO$_3$-N) and nitrite nitrogen (NO$_2$-N) concentrations using ion chromatography (DIONEX DX-120), T-N concentration using potassium peroxodisulfate digestion and the ultraviolet absorption method, phosphate phosphorus (PO$_4$-P) concentration using the ascorbic acid method, and T-P concentration using potassium peroxodisulfate digestion and the ascorbic acid method. Mean values of the concentrations in the SML and the PLb of each system are shown later in Figs 3, 5 and 7.

RESULTS

Characteristics of water movement inside the systems

Figure 2 shows changes in the proportion of water flow volumes in the SML of each system during wastewater treatment. In system I, the proportion sharply decreased from 100% at Day 0 to approximately 30% at Day 3, and then gradually decreased with time. The decrease probably resulted from proliferation of microorganisms and the overgrowth of biofilms in the SML. Water which could not flow into the SML would then flow into the PLb and subsequently down into the next SML in the systems with more than two layers. The structure of the MSL system makes possible more high-speed treatment as conventional soil-based systems cannot continue to treat wastewater because of permeability and clogging limitations. It took a longer time before the flow rate in the SML started to decrease with increasing numbers of layers. In other words, the total volume of water
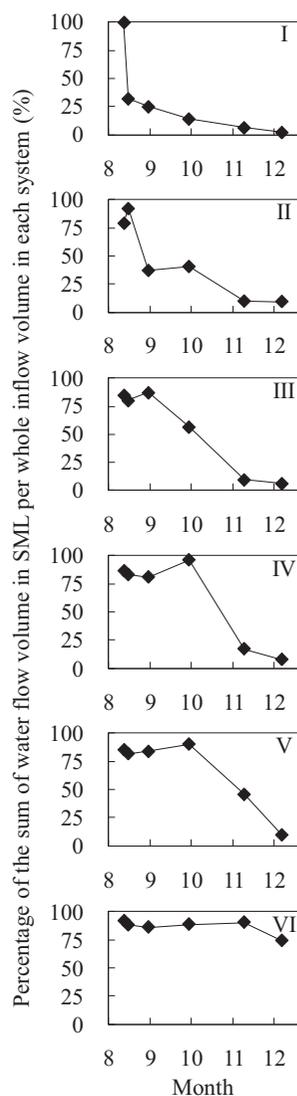
Figure 2 Changes in proportion of water flow volume in soil mixture layers (SML) of each system during wastewater treatment. Day 0 data show the percentages when well water flowed into the systems at a hydraulic loading rate of 1000 L m^{-2} day^{-1}. I–VI show the number of the system.

flowing in the SML gradually increased towards the lower layers. Detailed characteristics of water movement inside the systems in this study were reported by Sato et al. (2011).

Organic matter (biological oxygen demand and chemical oxygen demand) removal

In the SML, BOD concentrations were extremely low (nearly 0 mg L^{-1}) except in system I (Fig. 3). In contrast, the COD in the SML decreased with an increase in the number of layers. For both BOD and

Figure 3 Changes in mean concentrations of (a) biological oxygen demand (BOD) and (b) chemical oxygen demand (COD) of the w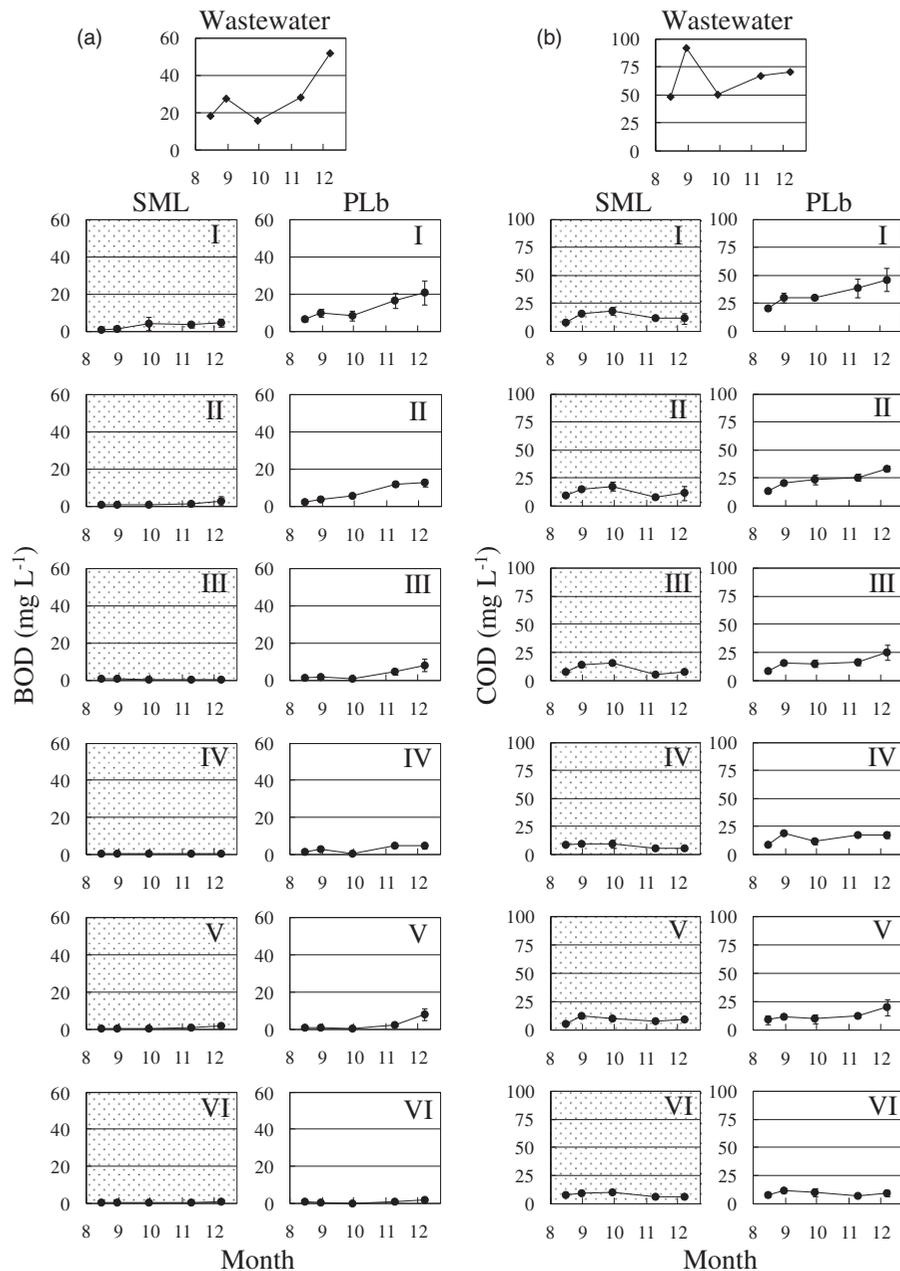astewater and treated water in each system. The error bars show the standard deviations. I–VI show the number of the system. SML, soil mixture layers; PLb, permeable layers between SML.

COD, their concentrations in the SML were usually lower than those in the PLb in each system. Additionally, as the flow rate in the SML decreased and the rate in the PLb increased, the BOD and COD in the PLb increased in each system (Figs 2 and 3). The trends of those concentrations in the PLb, especially in systems I and II, showed a similar pattern to those of fluctuations in the wastewater.

As the number of layers increased, removal rates of BOD and COD increased (Fig. 4). However, the removal rates on Day 117 in system V were lower than those in system IV. The removal rate of BOD was always more than 80% in the systems with more than three layers, and reached around 100% in system VI. In contrast, the removal rate of COD was usually lower than that of BOD in each system, and six layers
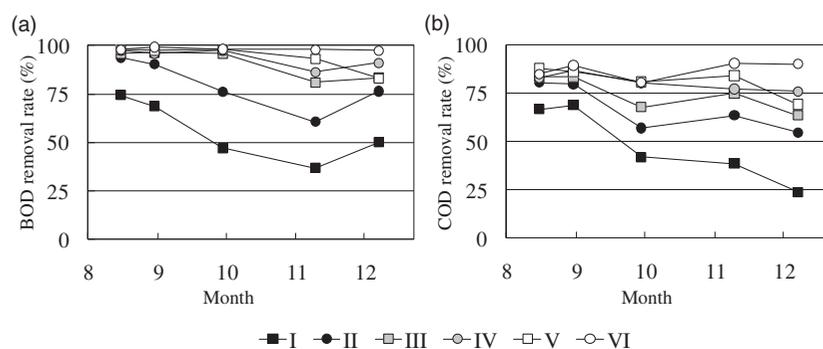
Figure 4 Changes in removal rate (%) of (a) biological oxygen demand (BOD) and (b) chemical oxygen demand (COD) in each system. I–VI show the number of the system.

were required for COD to be reduced more than 80% as the BOD removal.

Phosphorus removal

Phosphate phosphorus and T-P concentrations in the SML were lower and fluctuated less than those in the PLb (Fig. 5). Total phosphorus concentrations in the PLb tended to fluctuate, especially in systems I and II, which showed a similar pattern to that of fluctuations in the wastewater. Furthermore, PO_4-P and T-P concentrations in the PLb clearly increased with a decrease in the flow rate in the SML of each system (Figs 2 and 5).

Meanwhile, T-P concentrations were higher than PO_4-P concentrations in the SML and the PLb of system I and the PLb of system II. The differences in PO_4-P and T-P concentrations were small except in those layers of these two systems. These results suggest that organic phosphorus decomposed in the first and second layers and that phosphorus was mainly present as PO_4 below the third layer. Additionally, concentrations in the SML of systems I–III showed a tendency to increase with time.

Although the removal rate of T-P increased with increasing numbers of layers, the rate was significantly influenced by the water flow rate in the SML (Figs 2 and 6). The trends in removal rate were quite similar to that of the proportions of water flow volume in the SML in each system.

Nitrogen removal

Ammonium nitrogen concentrations were significantly low in every SML and PLb of the systems with more than three layers (Fig. 7). In the PLb of systems I and II, however, concentrations showed an increasing trend with time. Peaks in NO_2-N concentrations were observed in each layer of each system on Day 18 and then disappeared (Fig. 7). Nitrate nitrogen concentrations increased until 48 days from start of the experiment and

then decreased except in system II which showed no decrease in NO_3-N in the SML. Although the decrease in NO_3-N concentrations was observed in system III after Day 48, the differences in NO_3-N concentrations among systems IV–VI were small. Total nitrogen and the sum of the NH_4-N, NO_2-N and NO_3-N concentrations showed similar trends (Fig. 7). Total nitrogen was higher than the sum of the inorganic nitrogen fractions in the first and second SML and the first to fourth PLb. This difference was not found in the fifth and sixth layers. This suggests that organic forms of nitrogen gradually decomposed as water moved down through the MSL system.

Although the removal rate of nitrogen increased with an increase in the number of layers, this effect gradually decreased with an increase in the number of layers (Fig. 8). A high removal rate which was ascribed to ammonium adsorption was found on Day 3, and dropped by Day 18 due to production of NO_2-N in all of the systems. Thereafter in systems I and II, the removal rates dropped or remained around the same level. This was mainly caused by the increased flow rate in the PLb and subsequent discharging of NH_4-N and organic nitrogen (Figs 2 and 7). In systems with more than three layers, the rates gradually increased.

Total inputs and outputs of biological oxygen demand, chemical oxygen demand, total phosphorus, and total nitrogen in the multi-soil-layering systems

Table 1 shows total inputs and outputs of selected parameters in wastewater and treated water in the SML and the PLb of the different systems. The total removal rate of BOD reached 75.6 % in system II, and finally reached 98.0 % in system VI. Although the total flow volume gradually increased in the SML and decreased in the PLb toward the lower layers (Fig. 2), the output quantity of BOD in the SML was similarly low in every system. This was because BOD was significantly
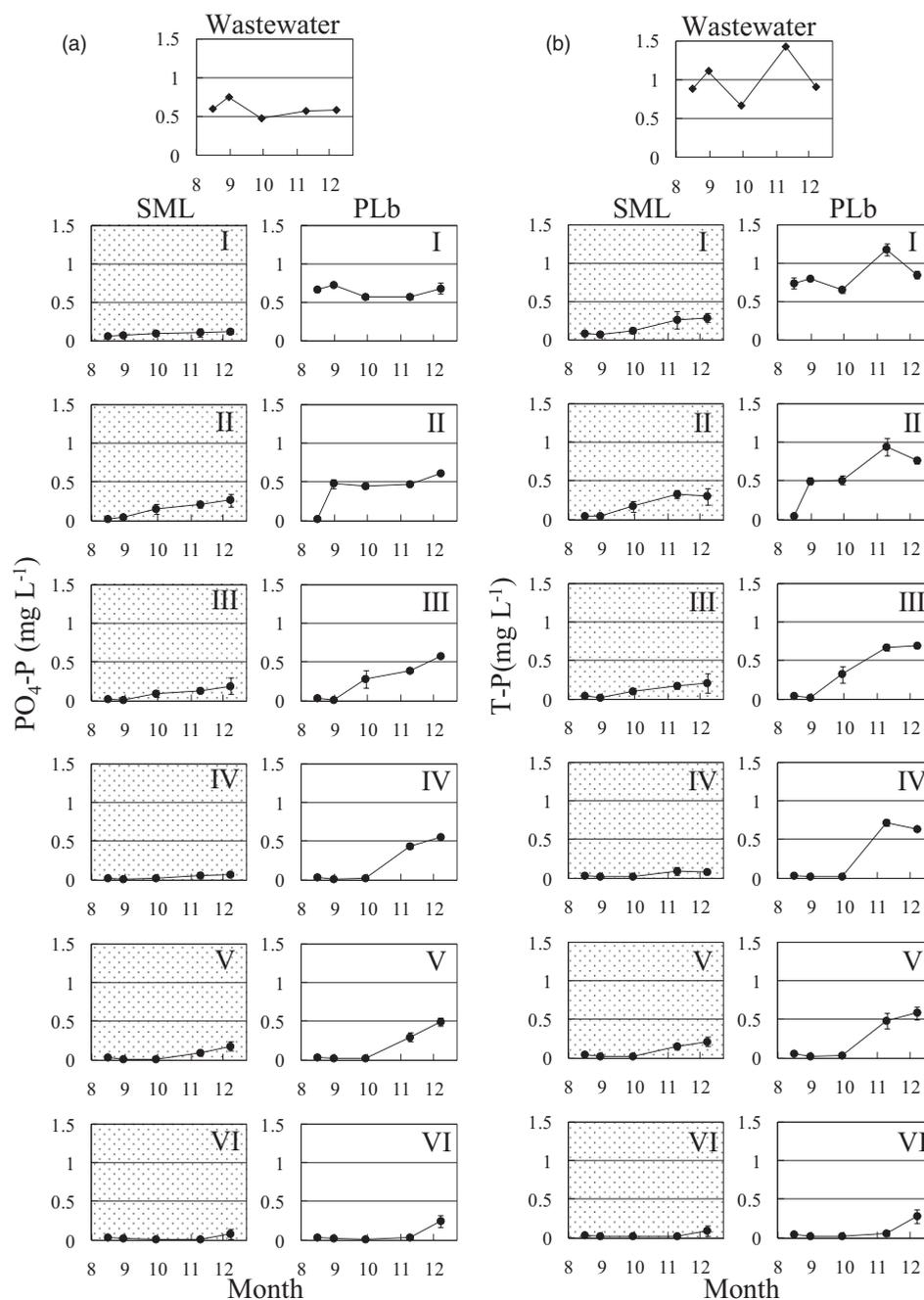
Figure 5 Changes in mean concentrations of (a) phosphate phosphorus (PO_4-P) and (b) total phosphorus (T-P) of the wastewater and treated water in each system. The error bars show the standard deviations. I–VI show the number of the system. SML, soil mixture layers; PLb, permeable layers between SML.

removed in the SML (Fig. 3). In contrast, the output quantity of BOD in the PLb decreased with increasing numbers of layers due to decreased BOD concentrations and the total flow volume in the PLb toward the lower parts of the system.

The total removal rate of COD in each system was lower than that of BOD as shown in Table 1. As for

BOD, the output quantity of COD in the PLb decreased with increasing numbers of layers. In the SML, however, the quantity increased with increasing numbers of layers mainly due to an increase in the total flow volume towards the lower layers.

The improvement in the total removal rate of T-P with increasing numbers of layers was greater than for BOD

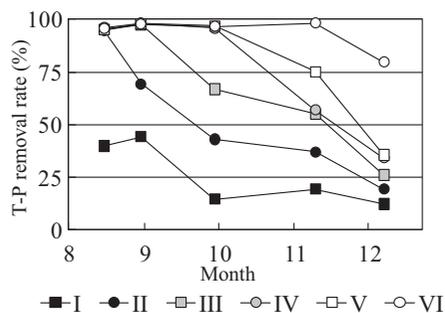**Figure 6** Changes in removal rate (%) of total phosphorus (T-P) in each system. I–VI show the number of the system.

and COD (Table 1). Phosphorus was mainly removed in the SML and the output quantity in the SML was quite small (0.10–0.25 g) in each system. The total output quantity and removal rate of T-P in each system depended to a great extent on the output quantity in the PLb (Table 1).

The total removal rate of T-N was lower than for BOD, COD and T-P, with the maximum rate being about 75% (Table 1). The output quantities of T-N in the SML and the PLb strongly depended on the total flow volume in each part of the system (Table 1, Fig. 2). The output quantity in the SML increased with increasing numbers of layers but the opposite trend was found in the PLb. The quantities in the PLb of systems I and II were especially higher because of the discharge of untreated organic nitrogen and NH_4-N (Fig. 7).

DISCUSSION

The efficiency of organic matter removal was higher in the SML than in the PLb, and the water permeability in the SML was important for organic matter removal. In the initial stages of the experiment, physicochemical reactions such as filtration and adsorption in the upper part of the system were probably the major treatment processes. It appeared that the organic matter referred to as COD and BOD was easily trapped in the SML, because of the amount of pore space, large surface area and enhanced hydrophobic properties provided by the addition of charcoal. Consequently, biological process of BOD and COD removal seemed to occur with time, in addition to the physicochemical reactions.

Although the removal rates of BOD and COD increased as the number of layers increased, the rates on Day 117 in system V were lower than those in system IV (Fig. 4). This might be because, compared to the other systems, system V had been earlier influenced by the development of anaerobic conditions and the occurrence of shortcut flow due to clogging. In the PLb of system I,

the flow rate in I-3 (Fig. 1) sharply decreased on Day 117 and partial clogging occurred (Sato *et al.* 2011). In a previous study using the same quality of wastewater, HLR, and system structure (with six layers) as in this study, an increase in the system weight was observed around Day 100 signifying clogging (Sato *et al.* 2005c). In addition, the total removal rate of nitrogen in system V was higher than that in system VI (Table 1). Denitrification might proceed more rapidly in system V due to the influence of such clogging.

The removal rate of COD was usually lower than that of BOD in each system (Fig. 4, Table 1). The results suggest that the removal of COD required more layers than were needed for the removal of BOD. This is because COD includes not only easily but also slowly decomposable organic matter, whereas BOD represents easily decomposable organic matter. Also, COD in the SML of systems IV–VI remained around the same level (Fig. 3), and the improvement in the removal rate with increasing numbers of layers decreased more than that for BOD removal (Fig. 4, Table 1). Chemical oxygen demand included easily and slowly decomposable organic matter and probably was not significantly removed to the same extent as BOD, as discussed earlier. Therefore, the ratio of slowly decomposable organic matter probably increased as water moved down the system. Furthermore, microbial decomposition of organic matter was probably greater in the upper parts of the system because nutrients such as nitrogen and phosphorus and easily decomposable organic matter were relatively rich there. These probably caused the decrease in the improvement of the treatment capacity for organic matter, especially COD, with increasing numbers of layers. Therefore, it is necessary to set an appropriate and cost-effective number of layers, depending on a planned treatment target as well as the quality and quantity of target wastewater.

The PO_4-P and T-P concentrations in the SML were lower those in the PLb (Fig. 5). Previous studies suggested that phosphorus removal is mainly based on phosphorus adsorption on active aluminum hydroxides contained in soil and also on ferric hydroxides formed from the metal irons added in the SML (Wakatsuki *et al.* 1993). In addition, PO_4-P and T-P concentrations in the PLb clearly increased with a decrease in the flow rate in the SML of each system (Figs 2 and 5). This was probably because the water which could not flow in the SML flowed into the PLb, which indicated that the permeability of the SML was also a very important factor contributing to phosphorus removal in the MSL system. In addition, concentrations in the SML of systems I–III tended to increase with time (Fig. 5). This was probably because the adsorption of phosphorus on ferric hydroxides decreased and/or the adsorbed phosphorus leached

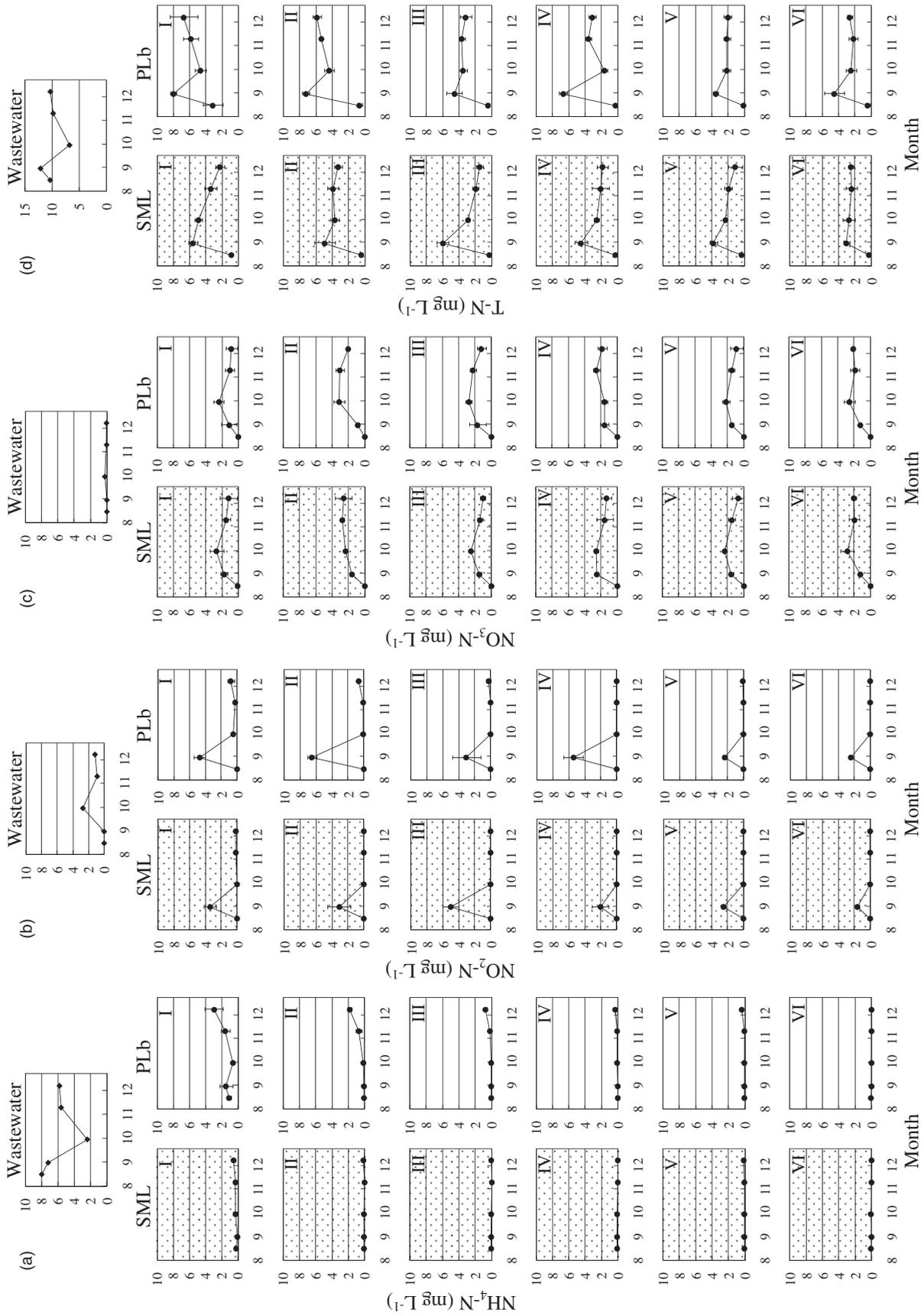**Figure 7** Changes in mean concentrations of (a) ammonium nitrogen (NH_4-N), (b) nitrite nitrogen (NO_2-N), (c) nitrate nitrogen (NO_3-N) and (d) total nitrogen (T-N) of the wastewater and treated water in each system. The error bars show the standard deviations. I–VI show the number of the system. SML, soil mixture layers; PLb, permeable layers between SML.

from the SML due to the establishment of an anaerobic condition in those SML. In the SML of systems I–III, a decrease in Eh with a decrease in the water flow rate was observed (Sato *et al.* 2011).

Compared to the removal rate of BOD and COD, the rate of T-P was more influenced by the water flow rate in the SML (Figs 2, 4 and 6). Although BOD and COD were mainly removed in the SML, it is likely that these were also removed in the PL. In contrast, it is suggested that removal of phosphorus is more dependent on the SML than is removal of organic matter. When the flow rate in the SML was higher, the removal rate of T-P was approximately 100% even in system II (Fig. 6). Also, the improvement in the total removal rate of T-P with increasing numbers of layers was greater than those of BOD and COD (Table 1). This is also because phosphorus removal was strongly influenced by the flow rate in the SML.

In the case of nitrogen removal, it appears that ammonium adsorption on soil materials and zeolite took place and that nitrification activity developed with time (Fig. 7). In the PLb of systems I and II, NH_4-N
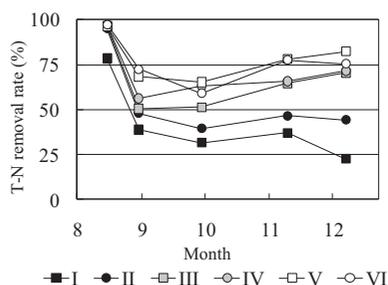
concentrations showed an increasing trend with time. This is probably because the contact efficiency of wastewater with the soil and zeolite decreased due to the increased flow rate in the PLb with time (Fig. 2). Peaks of NO_2-N concentration were observed in each part of the systems on Day 18 (Fig. 7). Nitrite is the intermediate product of transformation of NH_4 to NO_3 and disappeared after Day 18 with an increase in nitrification activity (Fig. 7). Although NO_2 is also generated in the denitrification process, concentrations were low in the latter stages of this experiment, indicating that NO_3 reduction proceeded to N_2O or N_2 in the denitrification process. Nitrate nitrogen increased until Day 48 and decreased thereafter except in system II (Fig. 7). This indicates that nitrification activity developed from the start of the experiment with denitrification occurring subsequently. In system I, a reduction in the nitrification rate probably also contributed to the decrease in NO_3-N after Day 48 because NH_4-N increased with time in this system (Fig. 7). In system II, a decrease in NO_3-N was not observed in the SML. This was probably because NH_4-N which was not removed in the first layer was transformed to NO_3-N by nitrification in the second layer. Although NO_3-N decreased as a result of denitrification after Day 48 in system III, the differences in NO_3-N concentrations among systems IV–VI were small (Fig. 7). Although a decrease in Eh with a decrease in the water flow rate was observed in the SML of systems I–III (Sato *et al.* 2011), Eh in the SML of systems IV–VI and the PLb of each part showed relatively high values. Also, more than 80% of the BOD was removed down to the third layer (Fig. 4). Therefore, it is suggested that denitrification mainly occurred in the SML of the first to third layers. In the fourth to sixth layers, in contrast, it was probably

Figure 8 Changes in removal rate (%) of total nitrogen (T-N) in each system. I–VI show the number of the system.

Table 1 Total inputs (wastewater) and outputs [treated water in soil mixture layers (SML) and permeable layers between SML (PLb) of selected parameters

			Biological oxygen demand (g)	Chemical oxygen demand (g)	Total phosphorus (g)	Total nitrogen (g)
Input	Wastewater		186.9	429.6	6.57	61.8
Output	System I	SML:PLb	2.1:88.4	11.5:219.0	0.10:4.80	3.2:35.0
		Total (removal rate)	90.6 (51.5%)	230.5 (46.3%)	4.90 (25.4%)	38.2 (38.2%)
	System II	SML:PLb	2.3:43.4	26.0:122.9	0.26:3.35	5.9:25.2
		Total (removal rate)	45.7 (75.6%)	148.9 (65.3%)	3.61 (45.0%)	31.1 (49.7%)
	System III	SML:PLb	1.8:21.9	35.1:78.4	0.19:2.27	9.5:13.3
		Total (removal rate)	23.7 (87.3%)	113.5 (73.6%)	2.46 (62.5%)	22.8 (63.1%)
	System IV	SML:PLb	1.7:13.4	31.9:52.5	0.12:1.82	9.8:10.5
		Total (removal rate)	15.1 (91.9%)	84.4 (80.4%)	1.94 (70.5%)	20.3 (67.1%)
	System V	SML:PLb	2.3:14.4	37.5:44.6	0.22:1.27	9.4:5.5
		Total (removal rate)	16.7 (91.1%)	82.1 (80.9%)	1.49 (77.3%)	14.9 (75.9%)
	System VI	SML:PLb	2.6:1.2	43.4:9.5	0.25:0.14	13.7:2.6
		Total (removal rate)	3.8 (98.0%)	53.0 (87.7%)	0.39 (94.1%)	16.3 (73.5%)

difficult for denitrification to occur because of insufficient organic matter being available as an electron donor for denitrification and relatively high values of Eh in the SML of those layers. Although ammonium adsorption reactions with the materials and nitrification were almost complete at the third layer, denitrification, which is one of the main problems for soil-based wastewater treatment systems, was a limiting factor for nitrogen removal. In addition, the improvement in nitrogen removal with increasing numbers of layers also gradually decreased due to the limit of denitrification (Fig. 8, Table 1).

In this study, the water flow rate in the SML was an essential factor for removal of organic matter, phosphorus and ammonium. This result and previous studies suggest that selection of soils with proper particle size, an appropriate mixing ratio of high permeability materials in the SML and setting of an aeration regime are important factors in maintaining a suitable flow rate in the SML. Sato et al. (2005a), in a study on direct polluted river water treatment by a MSL system, found that a blend of sandy soil, leaf mold and charcoal with a clayey alluvial soil made possible increased organic matter removal efficiency. It has also been suggested that aeration to the system is effective to prevent clogging and to enhance the decomposition of organic matter (Sato et al. 2002b, 2005b). Additionally, clogging due to the accumulation of organic matter can be reduced by halting the operation during periods of two to three months (Wakatsuki et al. 1998, 1999). This implies that alternating use of some systems would also be effective in retaining water permeability in the SML. However, excessively high permeability in the SML could cause a decrease in water treatment capacities by filtration and adsorption. Furthermore, Luanmanee et al. (2001) showed that an excess of aeration was inappropriate for nitrogen removal due to decreasing denitrification. Further study on proper materials for the SML and optimum aeration regimes is needed.

In this study, the removal processes of nitrogen were more complicated than those of BOD, COD and phosphorus. Ammonium adsorption and nitrification were almost completed at the third layer and denitrification only proceeded until the third layer. This suggests that aeration in the upper parts of the system for nitrification and the extra addition of organic matter (especially high carbon/nitrogen ratio materials) to the SML in the lower parts for denitrification are effective for simultaneous removal of organic matter, phosphorus and nitrogen. Furthermore, it might also be effective to inject dissolved easily decomposable organic matter such as methanol and part of the wastewater into the SML of the lower parts of the system during wastewater treatment.

For more quantitative evaluation of organic matter decomposition, denitrification and phosphorus adsorption, we plan to analyze the materials of the MSL system which was used in this research. To generalize the effect of different numbers of layers and to establish design guidelines for the MSL system, we plan to continue further studies on the relationship between the treatment processes in MSL systems and different wastewater quality and quantity.

CONCLUSION

This study evaluated wastewater treatment processes and mass balances from the quality and quantity of wastewater and treated water. The important findings are as follows: (1) the efficiency of organic matter removal was higher in the SML than in the PLb, and water permeability in the SML was important for organic matter removal, (2) the removal of COD needed more layers than that for reduction in BOD because COD includes not only easily but also slowly decomposable organic matter, (3) removal of phosphorus in the SML was higher than in the PLb, and the removal depended more on water permeability in the SML than organic matter, (4) ammonium adsorption and nitrification were almost completed at the third layer, but denitrification only proceeded until the third layer due to insufficient availability of organic matter as an electron donor for denitrification and relatively high Eh values in the SML below the fourth layer, and (5) it was suggested that the aeration in the upper parts of the system for nitrification and the addition of extra organic matter to the SML in the lower parts for denitrification were effective for the simultaneous removal of organic matter, phosphorus and nitrogen.

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