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ORIGINAL ARTICLE

Characteristics of wastewater treatment using a multi-soil-layering system in relation to wastewater contamination levels and hydraulic loading rates

Tsugiyuki MASUNAGA[1], Kuniaki SATO[1], Junichi MORI[1], Matsusige SHIRAHAMA[2], Hiromoto KUDO[3] and Toshiyuki WAKATSUKI[4]

[1]*Faculty of Life and Environmental Science, Shimane University, Matsue 690-8504,* [2]*Kankyo Gijutsu Co. Ltd, Suita City 564-0051,* [3]*Top Ecology Co. Ltd, Tokyo 110-0015, and* [4]*Faculty of Agriculture, Kinki University, Nara City 631-8505, Japan*

Abstract

Characterization of wastewater treatment using a multi-soil-layering (MSL) system at two levels of wastewater contamination (a low level [LWW] and a high level [HWW]) with a mean biochemical oxygen demand (BOD) of 30 and 70 mg L^{-1}, respectively, and at hydraulic loading rates (HLRs) of 500–2,000 L m^{-2} day^{-1} was conducted. Despite fluctuations in the wastewater contamination levels, the BOD and total phosphorus (T-P) concentration in the treated waters remained constantly lower than those in the wastewaters. However, the total nitrogen (T-N) concentration in the treated water was influenced by the level in the wastewater and fluctuated. In general, the two different levels of wastewater contamination did not influence the mean concentrations of the contaminants in the treated waters and the mean removal percentages in the MSL system. Mean removal percentages ranged from 90 to 96% for suspended solids (SS), from 88 to 98% for BOD, from 83 to 94% for CODcr, from 44 to 57% for T-N, and from 63 to 89% for T-P. Removal of SS, BOD, chemical oxygen demand (CODcr) and T-N was not appreciably influenced by changes in the HLR. For the removal of nitrogen, a negative correlation was observed with the oxidation–reduction potential (ORP) in the treated water, suggesting that the development of reduced conditions in the system was the key factor for the removal of T-N through the enhancement of the denitrification process. The mean removal percentage of phosphorus tended to be higher at lower HLRs. An increase in the HLR probably reduced the contact time of phosphorus and materials in the MSL system, which resulted in the decrease in the removal percentage. Removal rates (g m^{-2} d^{-1}) of BOD, CODcr, nitrogen and phosphorus were highest at the highest HLR in the HWW treatments. However, higher HLRs, especially in the HWW treatments, caused clogging of the system during the early periods of operation. Clogging was alleviated after an 8-week rest of the system. An operation cycle in which periodical resting time of the system is used to prevent or alleviate clogging could be recommended for the use of the MSL system.

Key words: clogging, hydraulic loading rate, multi-soil-layering system, wastewater contamination level, wastewater treatment.

INTRODUCTION

Wastewater treatment by soil involves physical, chemical and biological processes that have been known to be effective and that have been used in natural land treatment and soil trench systems. Refractory organic materials, such as colored materials, in livestock wastewater can also be removed effectively (Mori *et al.* 1997). Such soil (land) wastewater treatment systems cost less, but need a wider land area than modern mechanical treatment systems. Where the land area is not limited and land cost is low, soil wastewater treatment systems have often been used for on-site wastewater disposal. However, there is a limitation in the use of soils for wastewater treatment systems because of the low permeability of soils, which reduces the discharge rate of wastewater

Correspondence: T. Masunaga, Faculty of Life and Environmental Science, Shimane University, Matsue 690-8504, Japan. Email: masunaga@life.shimane-u.ac.jp
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into a treatment system. In soil wastewater treatment systems, the hydraulic loading rate (HLR) normally ranges from 10 to 40 L m^{-2} day^{-1} for traditional soil trench systems (Perkins 1989) and from 1 to 8 L m^{-2} day^{-1} for slow-rate land treatment systems (Reed *et al.* 1995) to avoid the accumulation of waste materials and the metabolic by-products that cause clogging. Variations in the chemical and biological properties of the soil types, which can influence the treatment ability of the soils, are also a possible limitation.

We should consider these factors in order to use soils appropriately for wastewater treatment systems. For example, sandy soils exhibit a relatively high permeability with infiltration rates of 1,800–3,600 L m^{-2} day^{-1}, which results in higher wastewater discharge rates, but their chemical and biological properties are not as satisfactory as those of silty or clayey soils with lower infiltration rates of 300–900 L m^{-2} day^{-1} (Perkins 1989). With these properties, sandy soils would be suitable for treatments that require higher speed rather than higher removal percentages of contaminants. It is important to select optimum soils depending on the treatment conditions required. However, the availability of soil types is, in fact, limited in the respective regions.

The multi-soil-layering (MSL) system has been developed and studied to use soils effectively for wastewater treatment through the alleviation of the above limitations of soils. A MSL system is composed of soil mixture layers and permeable layers that are arranged in a brick-layer-like pattern, and this structure keeps the water permeability high and reduces the risk of clogging of the system. Many studies have been conducted to enhance the treatment efficiency of the MSL system by changing the material composition and aeration conditions of the system as follows: effects of iron metal and organic matter addition in soil mixture layers on nitrogen and phosphorus removal have been examined by Luanmanee *et al.* (2002), Wakatsuki and Omura (1991a,b) and Wakatsuki *et al.* (1991, 1993). The influence of material types in permeable layers was also reported by Boonsook *et al.* (2003). Effects of aeration on organic matter, nitrogen and phosphorus removal were studied by Luanmanee *et al.* (2002) and Sato *et al.* (2002a). Based on these studies, it appears that changing the material composition and aeration conditions by matching specific wastewater conditions could control the treatment characteristics of MSL systems. The knowledge and experience gained from previous studies may enable the design of better MSL systems. However, there is still a need for further information on the factors that influence the treatment efficiency of MSL systems to properly design a MSL system depending on treatment conditions, such as the concentrations of the contaminants in wastewater, quality target of treated water and treatment speed (i.e. HLR). Wastewater levels of contamination and HLRs are the major factors that influence the treatment efficiency of the MSL system and also of other wastewater treatment systems. In the present study, the effects of two levels of domestic wastewater concentration and five degrees of HLRs on the treatment efficiency and the occurrence of clogging in a MSL system were examined.

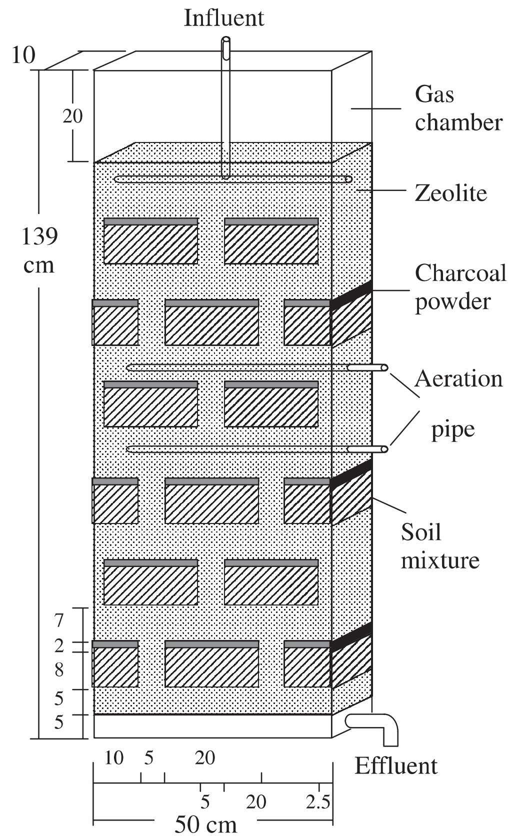

Figure 1 Structure of the multi-soil-layering system.

MATERIALS AND METHODS

Description of the MSL system

Figure 1 shows the structure of the laboratory-scale MSL system with an acrylic box measuring 139 cm in height by 50 cm in width by 10 cm in depth that was used in the present study. The MSL system was composed of soil mixture layers and zeolite layers with a diameter of 1–3 mm. The soil mixture layers consisted of volcanic ash soil rich in organic materials classified as Andisols (United States Department of Agriculture 1999), sawdust and granular iron metal at a volume ratio of 75%, 12.5% and 12.5%, respectively. These layers were set at a thickness of 8 cm, with the upper surface covered with a 2-cm thick powder charcoal layer. The layers were arranged in a brick-layer-like pattern surrounded by zeolite layers. A porous inlet pipe for wastewater was placed in the top layer of zeolite. Two aeration pipes were installed in the third and fourth zeolite layers. However, in the present study, aeration was not used during the experimental period in order to evaluate the effects of the wastewater contamination levels and HLRs on treatment efficiency and clogging in the MSL system separately from the influence of aeration. The MSL system was installed in a prefabricated house at Shimane University, and the wastewater treatment experiment was conducted from 27 July 1998 to 16 October 1999.

Wastewater level of contamination and HLR

Domestic wastewater from a housing development in which approximately 200 households lived was diluted fourfold and eightfold and used in the present study. The original wastewater was taken from a holding tank with a volume of more than 15 m^3 before treatment in the wastewater treatment plant of the housing development. The wastewater was diluted in two 1000 L storage tanks for each dilution level with well water approximately every 3 days. As for the quality of the well water, biological oxygen demand (BOD), NH_4-N and NO_2-N were not detected and the concentrations of PO_4-P, NO_3-N, Na, K, Ca, Mg and Fe were 0.11–0.48, 0.9–2.3, 71–76, 3.0–3.5, 34–127, 15.6–55.8 and 0.03 mg L^{-1}, respectively, over 3 days during the experimental period. The fluctuations and mean values of the wastewater contaminants are indicated in Fig. 2 and Table 1. In the present study, there were two levels of wastewater contamination (a low level [LWW] and a high level [HWW]). Each type of wastewater was discharged continuously into the MSL system at HLRs of 500, 1,000, 1,250, 1,500 and 2,000 L m^{-2} day^{-1} using tube pumps (IWAKI PST-1120, Asahi Techno Glass Co Ltd, Tokyo, Japan) for 24 h. Therefore, in total there were 10 treatments with a combination of five HLRs and two levels of wastewater contamination. The treatments were hereafter referred to as L-500 to L-2000 and H-500 to H-2000, indicating the wastewater levels of contamination and the HLRs, respectively.

The water content and hydraulic retention time (HRT) ranged from 18.9 to 20.8 kg and 4.8 to 18.2 h, respectively, in the MSL system under non-polluted water (well water) flow conditions before the onset of the wastewater treatment. The water content increased as HLR increased under unsaturated flow conditions. There was also an inverse relationship between HRT and HLR. Detailed aspects of the relationships between HLR and HRT in a different MSL system were described by Sato *et al.* (2005).

Sampling and analyses

Wastewater and treated water were collected at almost the same time at 2-week intervals and were analyzed for the following properties using standard methods for the examination of water and wastewater (American Public Health Association/American Water Works Association/Water Environment Federation 1995): pH using the glass electrode method (Horiba D-24, Kyoto, Japan), oxidation–reduction potential (ORP) with an Eh meter (TOA HM-14P, Tokyo, Japan), BOD using the modified Winkler method, suspended solids (SS) concentration using the filtration method, chemical oxygen demand (CODcr) using the potassium dichromate method, NH_4-N concentration using the Nessler method, NO_2-N concentration using the diazotization method, NO_3-N concentration using the diazotization method after the cadmium reduction method, and PO_4-P concentration using the ascorbic acid method. Total nitrogen (T-N) and total phosphorus (T-P) contents were determined as NO_3-N and PO_4-P after potassium peroxodisulfate digestion.

Room air and water temperatures were also recorded when the water was sampled. Treated water was collected at the outlet of the system using a plastic container and its temperature was measured.

As the sampling interval of 2 weeks was longer than that of the wastewater preparation (every 3 days), we determined whether the wastewater sampled in the present study was representative in the following way. We compared the analytical results for BOD, T-N and PO_4-P from the present study with those reported by Sato (2005), who conducted water analyses at intervals of 2–7 days from May 1999 to March 2000. Variance, mean values and coefficients of variation of the data in the two studies were compared, considering the dilution of the original domestic wastewater in the present study. There were no statistical differences ($P > 0.44$) in the variances and the means of the two datasets, and the means and coefficients of variation were comparable for all three parameters (BOD, T-N and PO_4-P). This indicated that the 14-month sampling at 2-week intervals in our present yield data was as representative as that from more frequent sampling. In addition, the results of the previous study (Sato 2005) also showed that the BOD, T-N and PO_4-P concentrations of wastewater generally fluctuated by less than 20% when the values from two consecutive samples collected within 2–7 days were compared. Based on the above, we concluded that the analytical values from sampling at 2-week intervals (Fig. 2) were representative for each treatment. Further data analyses, such as the calculation of the removal efficiency and removal rates of contaminants (Tables 1,2), were also carried out based on this assumption.

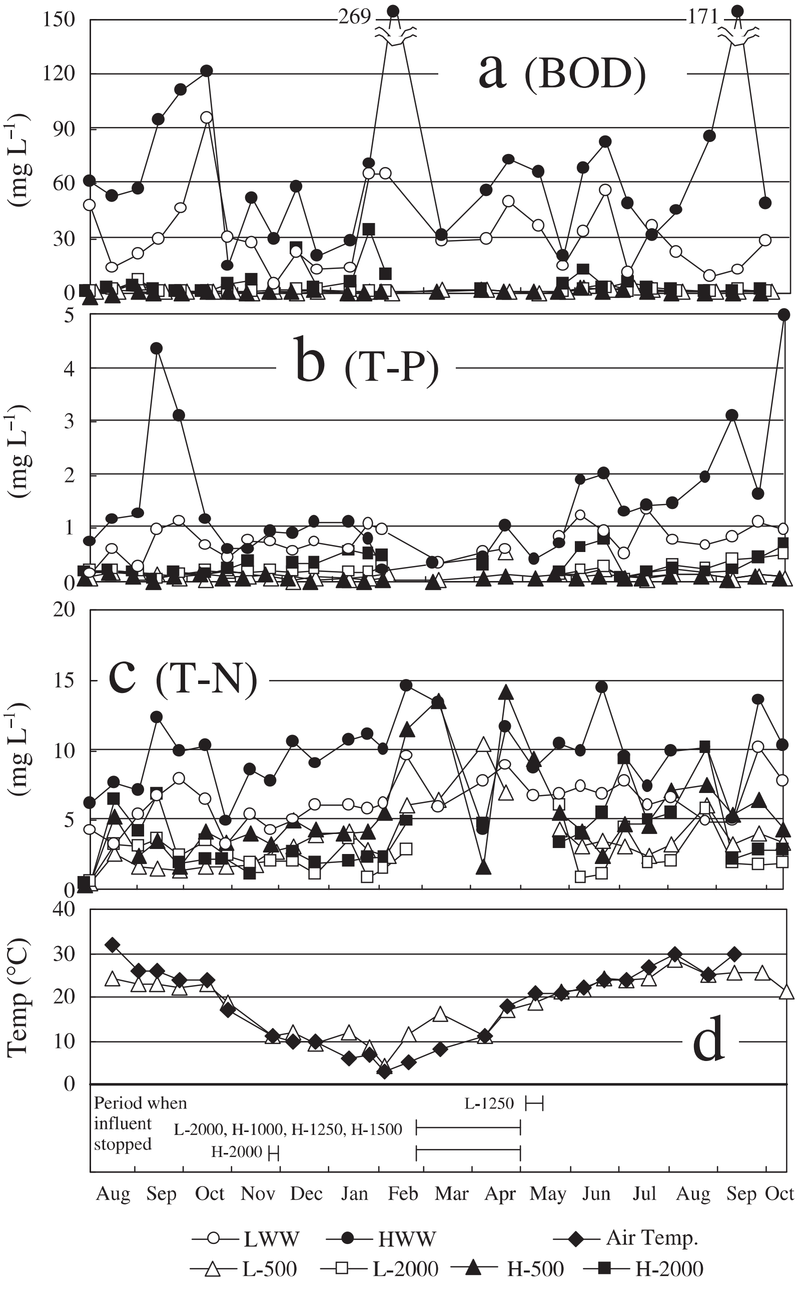

Figure 2 Changes in (a) biological oxygen demand (BOD), (b) total phosphorus (T-P) and (c) total nitrogen (T-N) concentrations in wastewater and treated L-500, L-2000, H-500 and H-2000 waters, and (d) the air and water temperatures. HWW, high level of wastewater; LWW, low level of wastewater; 500 and 2,000, hydraulic loading rates of 500 and 2,000 L m^{-2} day^{-1}, respectively.

Interruption of system operation during clogging

Some treatments caused clogging and the top layers were covered with stagnant wastewater. At that time, wastewater discharge into the clogged system was interrupted for 8 weeks for recovery. In the L-1250 and H-2000 treatments, wastewater application stopped for only 2 weeks and 1 week, respectively, in the first clogging. At that time, in the L-1250 and H-2000 treatments, the top 2-cm layer of zeolite, which represented the clogged layer, was sampled for the observation of the status of material accumulation and clogging, and was replaced with a new one. Wastewater application was then resumed when the system was no longer subjected to clogging.

Table 1 Mean level of contamination (mean ±standard deviation) and mean removal percentage† of selected parameters in wastewater and treated water during the experiment

Treatment	pH	ORP (mV)	SS (mg L^{-1})		BOD (mg L^{-1})		CODcr (mg L^{-1})		NH_4_N (mg L^{-1})	
LWW	7.41 ± 0.25	232 ± 78	27.8 ± 21.9		32.1 ± 20.9		43.9 ± 21.5		3.34 ± 1.08	
L-500	7.49 ± 0.24a	335 ± 51a	1.8 ± 2.8a	(93)	1.2 ± 1.1b	(94)	3.7 ± 3.7b	(89)	0.05 ± 0.08b	(98)
L-1000	7.51 ± 0.33a	327 ± 60a	1.9 ± 2.4a	(90)	1.3 ± 1.1b	(94)	5.5 ± 4.7b	(84)	0.05 ± 0.06b	(98)
L-1250	7.48 ± 0.24a	326 ± 60a	1.3 ± 1.8a	(91)	1.3 ± 0.8b	(94)	5.7 ± 4.4b	(83)	0.05 ± 0.07b	(98)
L-1500	7.40 ± 0.30a	321 ± 68ab	1.0 ± 1.2a	(92)	1.1 ± 0.5b	(95)	4.8 ± 4.5b	(86)	0.05 ± 0.04b	(98)
L-2000	7.37 ± 0.24a	324 ± 67ab	0.7 ± 0.8a	(94)	1.5 ± 1.3b	(91)	5.6 ± 5.2b	(85)	0.06 ± 0.05b	(98)
HWW	7.40 ± 0.18	177 ± 126	78.3 ± 75.3		69.5 ± 52.7		121.6 ± 96.7		6.61 ± 2.13	
H-500	7.47 ± 0.27a	333 ± 55a	1.7 ± 3.3a	(95)	1.1 ± 1.0b	(98)	5.0 ± 4.5b	(94)	0.05 ± 0.06b	(98)
H-1000	7.41 ± 0.21a	330 ± 61a	1.2 ± 1.9a	(96)	2.1 ± 2.9b	(94)	6.7 ± 6.3b	(92)	0.06 ± 0.06b	(98)
H-1250	7.41 ± 0.25a	302 ± 65ab	1.8 ± 2.4a	(93)	2.1 ± 2.5b	(95)	7.8 ± 6.9ab	(91)	0.08 ± 0.07b	(98)
H-1500	7.36 ± 0.26a	301 ± 86ab	2.1 ± 3.8a	(92)	2.7 ± 4.2b	(94)	9.7 ± 7.7ab	(90)	0.19 ± 0.22b	(96)
H-2000	7.33 ± 0.22a	264 ± 93b	3.3 ± 4.9a	(91)	6.2 ± 8.1a	(88)	12.1 ± 7.6a	(87)	0.41 ± 0.54a	(93)

Treatment	NO_3_N (mg L^{-1})	NO_2_N (mg L^{-1})	T-N (mg L^{-1})		PO_4_P (mg L^{-1})		T-P (mg L^{-1})	
LWW	0.9 ± 0.5	0.07 ± 0.08	6.3 ± 1.7		0.49 ± 0.12		0.76 ± 0.30	
L-500	2.1 ± 1.1b	0.01 ± 0.01b	4.0 ± 2.1abc	(44)	0.11 ± 0.05de	(74)	0.15 ± 0.11cd	(73)
L-1000	2.0 ± 1.1b	0.01 ± 0.01b	3.1 ± 1.4bc	(48)	0.13 ± 0.04cd	(72)	0.21 ± 0.19bcd	(69)
L-1250	2.0 ± 1.2b	0.01 ± 0.01b	2.9 ± 1.5bc	(51)	0.16 ± 0.06cd	(65)	0.20 ± 0.14bcd	(69)
L-1500	2.1 ± 1.0b	0.01 ± 0.01b	3.3 ± 1.8bc	(47)	0.18 ± 0.08abc	(60)	0.23 ± 0.12abc	(66)
L-2000	1.7 ± 1.1b	0.01 ± 0.01b	2.5 ± 1.4c	(56)	0.21 ± 0.09abc	(56)	0.24 ± 0.11abc	(63)
HWW	0.9 ± 0.7	0.01 ± 0.01	9.6 ± 2.7		0.73 ± 0.27		1.47 ± 1.16	
H-500	4.0 ± 2.0a	0.01 ± 0.01b	5.3 ± 3.3a	(45)	0.09 ± 0.04e	(86)	0.10 ± 0.04d	(89)
H-1000	3.2 ± 2.5ab	0.01 ± 0.01b	4.2 ± 2.6abc	(54)	0.12 ± 0.04cde	(82)	0.14 ± 0.06cd	(85)
H-1250	3.0 ± 3.1ab	0.03 ± 0.05ab	4.0 ± 2.9abc	(56)	0.17 ± 0.07bcd	(72)	0.22 ± 0.13abcd	(74
H-1500	3.5 ± 3.3ab	0.06 ± 0.20ab	4.8 ± 2.6ab	(48)	0.24 ± 0.13ab	(64)	0.31 ± 0.21ab	(71)
H-2000	2.4 ± 2.7ab	0.16 ± 0.45a	3.9 ± 2.4abc	(57)	0.25 ± 0.11a	(62)	0.34 ± 0.20a	(65)

†Mean removal percentage represents the average of the values at respective sampling times. Means within a column followed by the same letter are not significantly different according to a Tukey's test ($P > 0.05$). HWW, high level of wastewater; LWW, low level of wastewater; L(H)–500(–2,000), treatment with LWW (HWW) at a hydraulic loading rate of 500 (–2000) L m^{-2} day^{-1}. BOD, biological oxygen demand; chemical oxygen demand using the potassium dichromate method (CODcr); ORP, oxidation–reduction potential; SS, suspended solids; T-N, total nitrogen; T-P, total phosphorus.

Table 2 Mean removal rate (g m^{-2} day^{-1}) of selected contaminants†

	SS	BOD	CODcr	T-N	T-P
L-500	13	15	20	1.2	0.31
L-1000	26	31	38	3.2	0.56
L-1250	33	39	48	4.3	0.71
L-1500	40	47	59	4.6	0.81
L-2000	54	61	77	7.6	1.05
H-500	38	34	58	2.2	0.68
H-1000	77	67	115	5.5	1.32
H-1250	96	84	142	7.1	1.56
H-1500	114	100	168	7.3	1.73
H-2000	150	127	219	11.5	2.26

†The clogging period was omitted from the calculation. BOD, biological oxygen demand; chemical oxygen demand using the potassium dichromate method (CODcr); SS, suspended solids; T-N, total nitrogen; T-P, total phosphorus.

RESULTS AND DISCUSSION

Fluctuations of wastewater level of contamination and effect on treatments in the MSL system

Figure 2a–c shows the fluctuations in the concentrations of BOD, T-P and T-N in the LWW and HWW wastewaters, and L-500, L-2000, H-500 and H-2000 treated waters. Figure 2d shows the changes in the air temperature and treated water temperature.

Concentrations of BOD, T-P and T-N in the wastewaters fluctuated widely during the experimental period and ranged for LWW and HWW, respectively, as follows: 4.9–95.6 and 14.4–269.2 mg L^{-1} for BOD, 0.2–1.4 and 0.2–5.1 mg L^{-1} for T-P, and 0.5–10.5 and 0.5–14.1 mg L^{-1} for T-N. The BOD and concentrations of T-P and T-N did not appear to be largely influenced by seasonal factors, but were probably affected by other factors, such as the contents of night soil or other organic SS, in the holding tank when the original wastewater was collected. In terms of the contaminants in wastewater that are not shown in Figure 2, the CODcr and SS concentrations also displayed the same pattern as BOD, while variations in the concentrations of PO_4-P and NH_4-N were similar to those of T-P and T-N, respectively. Nitrite-N and NO_3-N concentrations in the wastewaters remained very low compared with the NH_4-N and T-N concentrations during the experimental period. Despite fluctuations in the BOD and T-P concentrations in the delivered wastewaters, the values in the treated waters remained low for all treatments. However, the T-N concentrations in treated waters appeared to be influenced by fluctuations in the T-N concentrations in the delivered wastewaters. Nitrate-N was the major component of T-N in the treated waters (Table 1). These results indicated that T-N discharged into the MSL system was converted to NO_3-N and that NO_3-N was then not fully removed from the system by denitrification, further suggesting that the denitrification process was limited for T-N removal in the MSL system. Total N concentrations in the L-500 and H-500 treatments tended to be high and were sometimes even higher than the wastewater concentrations in February to April, except at the beginning of April for the H-500 treatment, when the T-N concentration of HWW was low (Fig. 2c). An increase in the water temperature at this time probably enhanced the biological activity, including nitrification in the MSL system. Accumulation of organic matter containing nitrogen in the MSL system in winter and a higher rate of nitrate leaching from the system than the amount of T-N loaded into the system in the following warm season (March to April) were also observed in previous studies (Matsuyama *et al.* 2003; Wakatsuki *et al.* 1989). In the present study, nitrification of NH_4-N derived from the decomposition of the organic matter accumulated in the system during the winter probably occurred, and increased the NO_3-N concentrations in the L-500 and H-500 treated waters during that period. However, such a high level of nitrate leaching was not observed for the other treatments. The treatments with higher HLRs such as L-2000, H-1250, H-1500 and H-2000 caused clogging of the system during the same period when L-500 and H-500 showed a high level of nitrate leaching. The differences in the system under warm conditions were probably related to the difference in the balance of organic matter decomposition and loading in the treatments. The treatments with lower HLRs, such as L-500 and H-500, tended to show a higher rate of carbon dioxide emission (Masunaga *et al.* 2007), indicating the presence of a higher organic matter decomposition rate in these two treatments. Although warm conditions might contribute to the decomposition of organic matter in all treatments, it also enhanced biofilm development, which is responsible for clogging of the system (Rodgers *et al.* 2004). Biofilm development caused clogging at high organic matter loading rates and wastewater stagnated in the system with higher HLRs during that warm period, leading to a delay in organic matter decomposition. Detailed aspects of the clogging will be described in a later section.

Mean contamination level of treated water

Table 1 shows the mean contamination levels of wastewater and treated water. In terms of the general properties of treated water in relation to the contamination level, the pH values were near neutral for all treatments and did not show any significant differences between LWW and HWW treatments. Mean ORP values were the lowest in the H-2000 treatment. Mean ORP values of the H-1250 and H-1500 treatments, in which the third and second highest BOD and CODcr discharges were recorded among the ten treatments, also appeared to be relatively low, although statistical differences were not observed for these two treatments ($P > 0.05$). The lower ORP value probably resulted from the higher oxygen consumption associated with the higher BOD and CODcr loadings. Despite the differences in the two contamination levels of wastewater, statistical differences were observed only for the BOD and CODcr values and the NH_4-N concentration between the L-2000 and H-2000 treatments and the NO_3-N concentration between the L-500 and H-500 treatments, compared with a pair of LWW and HWW treatments at the same HLR. The differences in the wastewater contamination levels generally did not influence appreciably the mean concentrations of the contaminants in the treated waters.

Treatment efficiency of the MSL system and HLR

Table 1 shows the mean removal percentages of selected parameters in the respective treatments. Removal percentages of SS, BOD and CODcr remained as high as 80 or 90% in both LWW and HWW treatments. Removal of these contaminants was very efficient in all treatments and was not appreciably affected by changes in the HLRs.

As the HLRs increased from 500 to 2000 L m^{-2} day^{-1}, the mean concentrations of PO_4-P increased and the removal percentages decreased from 74 to 56% and 86 to 62% for the LWW and HWW treatments, respectively (Table 1). The results of the T-P concentration also showed the same trend because phosphorus mostly occurred as PO_4-P dissolved in wastewater. The mechanism of the treatment of PO_4-P in the MSL system basically involves physico-chemical adsorption onto adsorbents, such as aluminum or iron oxides, contained in soil or derived from the iron metal added to the MSL system (Wakatsuki *et al.* 1991). Contact between PO_4-P in wastewater and the adsorbents is the key factor for phosphorus removal. Higher HLRs shortened HRT, as described above and in the report of Sato *et al.* (2005b), which possibly reduced the contact time of PO_4-P in the wastewater and adsorbents in the MSL system. This probably resulted in the reduction of the removal percentage of phosphorus at higher HLRs. In terms of the relationship between HLR and phosphorus removal, T-P removal from a polluted river water in Fukuoka prefecture that contained approximately 0.6 mg P L^{-1} at a HLR of 4,000 L m^{-2} day^{-1} was approximately 60% (Unno *et al.* 2003). This value was as high as the removal percentage at 2,000 L m^{-2} day^{-1} in the present study. The depth of the MSL system in Fukuoka prefecture was 1.5 m, a value higher than that of the system used in the present study, which probably increased the contact time of phosphorus in the wastewater and the adsorbents in the system, resulting in higher T-P removal at that high HLR.

The influence of HLR on the treatment of nitrogen was different from that of the other contaminants. Ammonium-N concentration, which was the major nitrogen form in the wastewater, was efficiently reduced at rates of 93–99% in both LWW and HWW treatments. Then the concentration of NO_3-N increased and NO_3-N was the major nitrogen form in the treated waters. These results indicated that ammonium adsorption onto zeolite and the soil mixture layers and/or nitrification to NO_3-N occurred in all treatments. However, for the complete removal of nitrogen aside from ammonium adsorption, processes involving nitrification and denitrification are required in the MSL system. The system was generally kept in an aerobic state, as evidenced by the high ORP value in the treated water (Table 1), and the nitrification process was probably not limited in the MSL system. The amount of T-N loaded in the MSL system was estimated to be in the range of 2.14–10.60 g N kg^{-1} of zeolite (70.7–349.7 g N per system) during the experimental period in the present study, while the amount of T-N removed by the system ranged between 0.87 and 6.15 g N kg^{-1} of zeolite (27.7–202.1 g N per system). In contrast, the ammonium adsorption capacity was estimated to be in the range of only 0.63–3.87 g N kg^{-1} of zeolite, based on the analytical data of NH_4-N adsorption onto zeolite in different MSL systems reported in previous studies (Sato *et al.* 2005; Wakatsuki *et al.* 1990). The amount of T-N removed by the treatments exceeded the ammonium adsorption capacity in some MSL systems, although these values could not be directly compared because the experimental conditions, such as the material components and structure of the MSL systems, and the concentrations of ammonium in wastewater differed. Therefore, we speculate that nitrogen removal

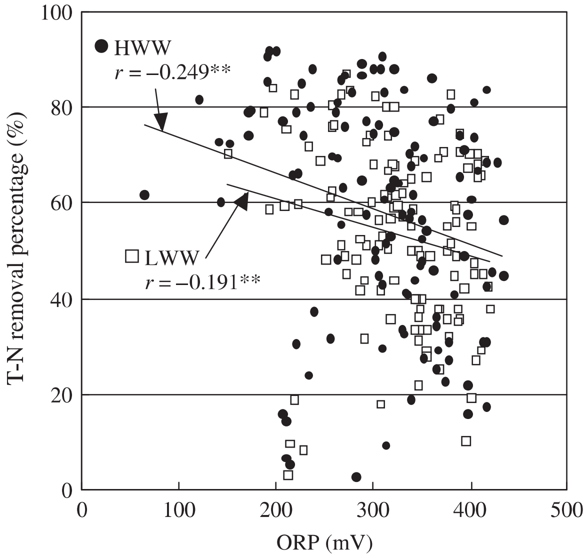

Figure 3 Relationship between the oxidation–reduction potential (ORP) and the removal percentage of total nitrogen (T-N). $^{**}P < 0.01$.

occurred through denitrification in addition to the removal by ammonium adsorption in the MSL system. The removal percentage of T-N increased in the L-2000 and H-2000 treatments, compared with that in the L-500 and H-500 treatments. The removal percentages were 44 and 56% for the L-500 and L-2000 treatments, and 45 and 57% for the H-500 and H-2000 treatments, respectively. The improvement of T-N removal was possibly because of an increase in denitrification with an increase in the HLR in the MSL system, although this assumption needs to be confirmed in future studies because statistical differences in T-N concentrations were not observed among the treatments. In general, denitrification is promoted by reduced (anaerobic) conditions and by the availability of BOD or COD as hydrogen donors. The values of the CODcr / T-N ratio ranged from 7 to 12 for LWW and HWW (Table 1), and these values were higher than the ratio (4–5) for efficient T-N removal (Isaacs and Henze 1994). This fact indicates that the levels of CODcr were suitable for T-N removal. Furthermore, a significant (1% level) negative correlation between the ORP value and the removal percentage of T-N was observed (Fig. 3). These facts support the assumption that a reduced condition developed inside the MSL system and that T-N was removed through a denitrification process. In our next study, direct measurement of ORP inside the soil mixture layers will be carried out to confirm this assumption.

Table 2 shows the removal rate (g m^{-2} day^{-1}), which corresponded to the integration of the removal percentages of contaminants (Table 1) and the HLRs for selected contaminants. Removal rates were higher in the HWW treatments and increased as HLRs increased for all contaminants, indicating that higher HLRs were more beneficial in terms of treatment efficiency of contaminants per unit system surface area. The removal rates of SS, T-N and T-P in the H-2000 treatment were 150, 11.5 and 2.26 g m^{-2} day^{-1}, respectively (Table 2). These values were equivalent to the annual application level of organic materials and various fertilizers in agricultural fields (e.g. Hiryo-Kyokai 2003).

As for the fate of the contaminants removed in the MSL system, it was reported that organic matter represented by SS, BOD and CODcr did not accumulate and probably decomposed in a MSL system used for on-site domestic wastewater treatment with aeration for 10 years at HLRs of 250–500 L m^{-2} day^{-1} (Sato *et al.* 2002b; Wakatsuki *et al.* 1991). However, in the present study, the loaded organic matter was not fully decomposed and partly accumulated in the MSL system with high HLRs, especially in the HWW treatments. This was probably because of the lack of aeration and a higher discharge of organic matter in the present study. The daily amount of BOD discharged into the MSL system in the H-2000 treatment was estimated to be fourfold as high as that into the previous MSL system (Wakatsuki *et al.* 1991). The accumulation of organic matter could be confirmed by direct observation of the system. Accumulation of the contaminants was responsible for the clogging of the MSL system. The relationship between HLR and clogging will be discussed in detail hereafter.

Clogging of the MSL system and HLR

The duration of the period of clogging is shown in Fig. 2d. In the H-2000 treatment, clogging occurred in the fourth month and at the end of the seventh month, while in the L-2000 and H-1000, H-1250 and H-1500 treatments, clogging occurred at the end of the seventh month. Clogging in the L-1250 treatment was then observed in the tenth month. In the higher HLR treatments, especially in the HWW treatments, clogging occurred earlier. Four treatments caused clogging at the end of February when the air and water temperature increased after the lowest air and water temperature was recorded in January and February. Rodgers *et al.* (2004) reported that the main mechanism of clogging of a sand filter system involved biofilm development. Organic matter accumulation when air and water temperatures were low and a biofilm developed at the time when the temperature was rising, were the possible triggers of clogging in the higher HLR treatments. Based on observations of the top 2-cm layer of zeolite sampled when clogging first occurred in the L-1250 and H-2000 treatments, it was evident that a dark-colored biofilm developed on the zeolite particles, and filled the pore spaces of the zeolite layer with accumulated SS. The clogging of the system in the present study was no longer observed after an 8-week rest of the system without aeration, which agreed with the observation of Wakatsuki *et al.* (1991). For the treatments in which clogging did not occur, such as the L-500 and H-500 treatments, biofilm development or organic matter accumulation was not as evident as in the treatments in which clogging occurred. In the treatments with low HLRs, organic matter decomposition during the warm period was higher than the loading or accumulation and, thus, clogging was prevented.

The risk of clogging should be avoided as much as possible. Setting a periodical resting time in a system operation cycle could be recommended to prevent or alleviate clogging for the use of the MSL system. If we design operational conditions including a 4-month work and 2-month rest cycle of the system for the H-2000 treatment, and a 7-month work and 2-month rest cycle for the H-1000, H-1250, H-1500 and L-2000 treatments, net annual HLRs could reach vales of 800–1,600 L m^{-2} day^{-1} for these treatments. The net removal rates calculated when the above operational cycles were adopted were the highest for the L-2000 and H-2000 treatments, respectively. These HLRs were still far higher than those of conventional soil trench systems in which normal HLRs ranged from 10 to 40 L m^{-2} day^{-1} (Perkins 1989). To reduce the risk of clogging, the removal of SS from the wastewater by pre-treatments including sedimentation, contact filtration and aeration in a settlement tank could also be effective (Sato *et al.* 2002a).

Conclusion

Despite fluctuations in the BOD and T-P concentrations in the delivered wastewaters, the concentrations of these contaminants in the treated waters remained constantly lower than those in the wastewaters during the experimental period. However, the T-N concentrations in the treated waters were influenced by those in the wastewaters and fluctuated. Two different levels of wastewater contamination generally did not influence appreciably the mean concentrations of the contaminants in the treated waters and the mean removal percentages in the MSL system. The mean removal percentage of phosphorus tended to be higher at lower HLRs. The changes in HLRs possibly affected phosphorus removal by influencing the contact time of phosphorus and adsorbents in the system. For the removal of nitrogen, apparently, denitrification was the limiting factor. Among the two factors required for the denitrification process, the amount of organic matter, such as CODcr components, appeared to be suitable under these experimental conditions. The development of reduced (anaerobic) conditions was probably the key factor in the MSL system as the system remained under aerobic conditions and a negative correlation was observed between T-N removal and ORP in the treated water (Fig. 3). Higher organic matter discharge might contribute to the development of reduced conditions in the MSL system.

Removal rates of SS, BOD, CODcr, nitrogen and phosphorus were higher at higher HLRs and were the highest in the H-2000 treatment. However, in the H-2000 treatment, the occurrence of clogging in the fourth month of operation was the earliest among the treatments. To reduce the risk of clogging, setting a periodical resting time in a system operation cycle and reduction of SS discharge into the system by pretreatments of wastewaters could be recommended for the use of the MSL system. Net removal rates in operational cycles of 7-month work and 2-month rest for the L-2000 treatment and of 4-month work and 2-month rest cycle for the H-2000 treatment were the highest among the treatments, respectively.

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