

ORIGINAL ARTICLE

# Effect of aeration and material composition in soil mixture block on the removal of colored substances and chemical oxygen demand in livestock wastewater using multi-soil-layering systems

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## Abstract

To investigate the efficiency of multi-soil-layering (MSL) systems on the removal of colored substances and chemical oxygen demand (COD) from livestock wastewater, four MSL systems with different soil mixture block (SMB) compositions were constructed in four 50 cm × 10 cm × 68 cm acrylic boxes. Livestock wastewater (diluted 10-fold) with an absorbance of 0.9215 at a wavelength of 406 nm and a COD concentration of approximately 3,000 mg L<sup>-1</sup> was applied to the systems at a hydraulic loading rate (HLR) of 250 L m<sup>-2</sup> day<sup>-1</sup>. Aeration pipes were set in the water permeable layers (PL) in MSL 1–3 and in the SMB layers in MSL 4. The results showed that MSL systems could keep mean decolorization rates of 60.7–67.1% and COD removal rates of 48.8–58.0% for 6 weeks of operation. The different aeration pipe positions did not have any significant influence on the removal efficiency of the systems. However, an increase in aeration intensity from 1,000 to 2,000 L min<sup>-1</sup> per system (27.4–54.8 L min<sup>-1</sup> L<sup>-1</sup>) increased decolorization rates by 3.0–12.1%. For COD removal, both an increase in aeration intensity and temperature enhanced the removal rates by 23.0–43.3%. The addition of sawdust and iron into the SMB of MSL 1 improved the system's decolorization rate by 9.1% and COD removal rate by 12.0% compared with MSL 2 during the fifth and sixth months of operation. Interruption of MSL systems for 1 month could recover the decolorization and COD removal rates to over 50% and 80%, respectively.

**Key words:** chemical oxygen demand, colored substances, livestock wastewater, multi-soil-layering system, soil mixture block.

## INTRODUCTION

Livestock wastewater is characterized by high concentrations of organic matter, nitrogen compounds and pathogenic bacteria. Pollutants from intensive stock farming can result in significant impairment of surface water and groundwater quality by entering surface waters from non-point sources of surface runoff or point sources from, for example, a concentrated farming house. In a survey of stream water quality and underground water quality undertaken in a stock farming area in Wonju, Korea, it was found that nearly all physicochemical and

bacteriological parameters examined in the boreholes located downstream of a livestock waste disposal site were much higher than those in the background boreholes and the infiltration of livestock wastewater clearly adversely affected groundwater quality (Cho *et al.* 2000). Point and non-point source pollution caused by insufficient livestock waste management has become an increasing concern in many countries and stipulations have been made setting limits on the discharge of biological and chemical properties of livestock effluent (Knight *et al.* 2000). In Japan, the livestock feeding density is relatively high, approximately 0.48 ha NEU<sup>-1</sup> (1 NEU equals 65 kg nitrogen released by one cow per year), compared with 11.43 ha NEU<sup>-1</sup> in Australia and 3.39 ha NEU<sup>-1</sup> in the United States (Mizuma *et al.* 2002). The more concentrated livestock operations with less land per animal for manure application caused more serious problems to the surrounding environments. In 1999, the Ministry of Agriculture, Forestry and Fisheries of Japan stipulated legislation on

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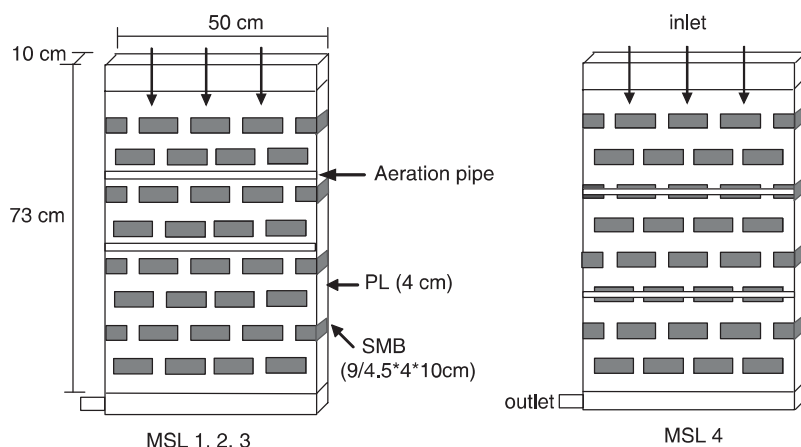
promoting livestock waste management and recycling that set strict limits on the permissible biological and chemical properties of the effluent, including biological oxygen demand (BOD), nitrogen (N) and phosphorus (P). However, most of the non-biodegradable contaminants, such as chemical oxygen demand (COD) and colored substances, could not be treated in an effective way, which not only caused environmental impairment but also prohibited the recycling of the treated water.

Previous research has shown that adsorption by soil is an effective method for livestock wastewater treatment, especially for non-biodegradable contaminants, such as colored substances and COD. Andisol was reported to be an effective natural adsorbent because of its ubiquitous presence and high content of organic matter (Chen *et al.* 2006). To continue research on the practical application of Andisol-based livestock wastewater treatment systems, we conducted a comparative study using multi-soil-layering (MSL) systems. Detailed descriptions of the MSL systems were reported by Wakatsuki *et al.* (1993) and Luanmanee *et al.* (2002). The purpose of the present study was to investigate the efficiency of Andisol-based MSL systems in removing colored substances and COD in relation to their material composition and aeration condition. We also examined the influence of temperature change on removal efficiency. Furthermore, decolorization mechanisms for livestock wastewater were explored in terms of an ion-exchange process and molecular size distribution analysis.

## MATERIAL AND METHODS

### Structure and components of the MSL systems

Fig. 1 shows the four laboratory-scale MSL system structures used in this study. The four MSL systems were packed in W50 cm × H73 cm × D10 cm acrylic boxes.



**Figure 1** Structures and components of the four multi-soil-layering (MSL) systems. The size of the MSL systems was W50 cm × H73 cm × D10 cm for wastewater treatment. The size of the soil mixture block (SMB) is shown in parentheses (W cm × H cm × D cm). PL, permeable layer.

**Table 1** Compositions and dry weight (g) of the materials in the soil mixture blocks in each of the four multi-soil-layering (MSL) systems

MSL System	Andisol (g)	Activated carbon (g)	Charcoal (g)	Sawdust (g)	Iron (g)
1	6451	922	0	922	922
2	8179	922	0	0	0
3	6566	922	922	0	0
4	6566	922	922	0	0

Each system has eight soil mixture block (SMB) layers with different material compositions arranged to form an alternative brick layer-like pattern (Table 1). Zeolite, with a diameter of 3–5 mm, was used to fill the void spaces between adjacent SMB. Aeration pipes (2.5 mm in diameter) were installed under the second and fourth SMB layers of MSL 1 to 3 and inside the third and fifth SMB layers of MSL 4 (Fig. 1). The main component of the SMB was Andisol and activated carbon (AC) based on a previous study (Chen *et al.* 2006). Andisol was obtained from the top surface soil of Mount Sanbe in Shimane, Japan, and had a total carbon content of 6.6% and effective cation exchange capacity (eCEC) of approximately 4.39 cmol<sub>c</sub> kg<sup>-1</sup>. The AC is commercially available in Japan (Enviro-Chemicals, Osaka, Japan) and is called Shirasagi M. It was made from wood and in a powdered form with a peak pore diameter of 20 Å.

### Operation of the MSL systems, laboratory and statistical analyses

To examine the removal efficiency of the systems, wastewater from a local dairy farm without any pre-treatment was applied after 10-fold dilution with an average absorbance of 0.9215 (at 406 nm) and COD concentration

of 3,000 mg L<sup>-1</sup>. The wastewater was diluted 10-fold because the systems were expected to be used after a secondary treatment, such as the activated sludge method or bio-film method, which set a target for BOD removal of approximately 90% (Japan Agriculture, Forestry and Fishery Culture Association 2004a). Although in the case of COD, the diluted wastewater might contain more degradable organic matter than in the secondary treated livestock wastewater and, thus, influence the removal efficiency to a certain degree, the experimental results are still effective in explaining the relationship between COD removal efficiency and MSL material composition and treatment conditions. The wavelength of 406 nm was selected according to a former study that found that humic substances are positively correlated with color and the absorbance of humic solutes increases with decreasing wavelength (Hautala *et al.* 2000).

The study was conducted over five periods from 1 November 2005 to 28 August 2006. After continuous operation for 8 months, the systems were interrupted for 1 month during July 2006 to check the systems' recovery process. The systems were restarted again after 1 month and ran until the end of August 2006. Over the entire study period the hydraulic loading rate (HLR) was set at 250 L m<sup>-2</sup> day<sup>-1</sup>, but aeration was applied differently to the four MSL systems in each period at amounts ranging from 1,000 to 2,000 L min<sup>-1</sup> per system (27.4–54.8 L min<sup>-1</sup> L<sup>-1</sup>) (Table 2).

Wastewater and treated water were sampled once per week for the following analyses: COD using the potassium dichromate method (American Public Health Association 1992) and color absorbance using a spectrophotometer (JASCO V-530, Tokyo, Japan) at 406 nm. Decolorization rate and COD removal rate were calculated as follows:

$$Re = (Co - Ce)/Co \times 100 \quad (1)$$

where  $Co$  and  $Ce$  refer to the absorbance of colored substances at 406 nm and the concentration of COD in the wastewater and treated water, respectively.

Statistical analysis of the data at different treatment periods was subjected to SPSS (version 12.0) with a

univariate analysis using Tukey's honestly significant difference (HSD) method (Ishimura 2001).

### **Ion exchange process and molecule size distribution analysis of the colored substances**

To clarify the decolorization mechanisms by different adsorbents, further study was conducted on the ion exchange process and molecular size distribution of the colored substances. Cation (IR120B Na AG) and anion (IRA402BL Cl AG) exchange resins from Organo-Co. (Tokyo, Japan) were used after washing with distilled water. Different amounts of resins based on dry weight (50, 100, 200, 500, 1,000 and 2,500 mg) were put into 50-mL centrifuge tubes and 30 mL of 10-fold diluted livestock wastewater was added to the tubes, which were shaken for 1 h on a horizontal shaker at 150 rpm at a temperature of 20°C. Treated solution was filtered through filter paper (5 µm) and the supernatant was taken for color absorbance measurement at 406 nm. All of the experiments were done in triplicate.

For molecular size distribution analysis in the wastewater and treated water, three water samples were taken as follows: (1) before and after treatment by Andisol (5 g 30 mL<sup>-1</sup>), AC and charcoal (50 mg 30 mL<sup>-1</sup>), respectively, with the same treatment process used in the ion exchange experiment, (2) before and after MSL system treatment after 6 months of continuous operation at the end of period 3, (3) before and after MSL treatment after 1 month of interruption at the beginning of period 5. The molecular size distribution of colored substances was analyzed using tangential ultra filtration at ambient room temperature (20–25°C). The splitting was made using Pellicon XL membranes (Millipore, Billerica, MA, USA) with cut-off sizes of 5 kDa and 50 kDa, and a peristaltic pump that gave a flow rate of 40 mL min<sup>-1</sup>. Previous to the tangential ultrafiltration, the solution was passed through a cellulose filter with a cut-off size of 1 µm to avoid a quick obstruction of the ultrafiltration membranes (Navia *et al.* 2005). Molecular size distribution of colored substances was expressed as the portion divided by the two cuts as well as the cellulose filter paper (1,000 kDa).

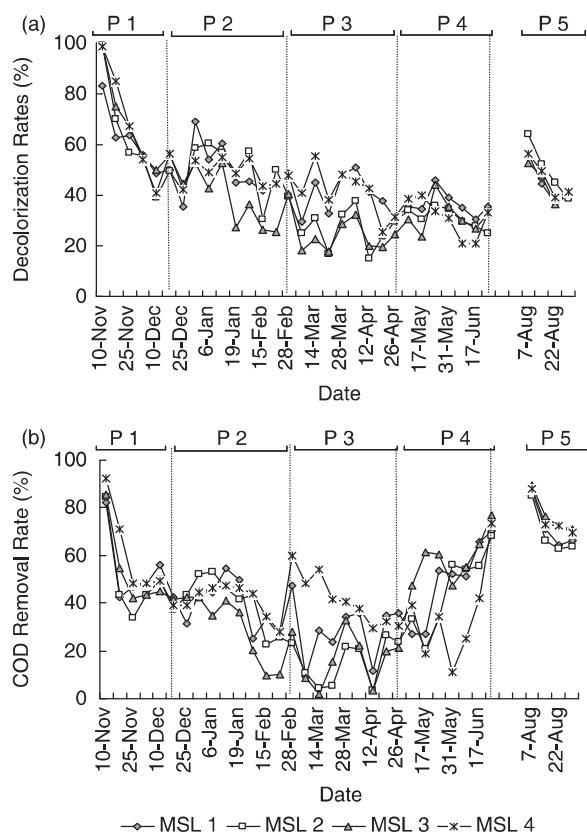
**Table 2** Experimental conditions in each period (time, hydraulic loading rate [HLR], air temperature and aeration conditions) of the four multi-soil-layering (MSL) systems

Period	Time	HLR (L m <sup>-2</sup> day <sup>-1</sup> )	Air temperature (°C)	Aeration (mL min <sup>-1</sup> per system)
1	1 Nov.–19 Dec. 2005	250	11–16	1000 (MSL 1–4)
2	20 Dec.–28 Feb. 2006	250	5–15	1000 (MSL 1–3); 2000 (MSL 4)
3	1 Mar.–27 Apr. 2006	250	9–18	1000 (MSL 1–3); 2000 (MSL 4)
4	28 Apr.–28 Jun. 2006	250	20–30	2000 (MSL 1–4)
5	1–28 Aug. 2006	250	30–35	2000 (MSL 1–4)

## RESULTS AND DISCUSSION

### Results of decolorization and COD removal rates using MSL systems over the whole experimental period

Livestock wastewater was characterized by a dark brown color and a high concentration of COD. The dark brown color comes from stercobilin, which is oxidized from urobilin as well as other undefined humic substances (Mori and Sakimoto 1999). The MSL systems showed similar trends in decolorization and COD removal rates during the first three periods (Fig. 2). At the beginning of period 1, all four of the MSL systems showed high removal efficiency over 80%. With time, the removal rates of the systems decreased gradually to approximately 40–50% after 2–3 weeks of operation and then remained stable for approximately 2 months until the middle of January 2006. After that, the removal efficiency of MSL 2 and 3 continued to decrease, whereas MSL 1 and 4 were able to maintain higher removal rates until the end of period 3.



**Figure 2** (a) Decolorization rates and (b) chemical oxygen demand (COD) removal rates in the four multi-soil-layering (MSL) systems as affected by material composition and aeration. P, period.

Statistical analysis showed that there was no significant difference among the four MSL systems in colored substances and COD removal during period 1 (Table 3). The mean decolorization rates ranged from 60.7% (MSL1) to 67.1% (MSL3) and COD removal rates ranged from 48.8% (MSL 2) to 58.0% (MSL4). With time, the difference among the four systems appeared gradually and MSL 3 showed significantly lower removal efficiency than the other three systems during period 2. The mean decolorization and COD removal rates for MSL 3 were 39.8% and 29.4%, respectively, while those for the other three systems were from 48.8–49.2% and 39.1–39.7%, respectively. The difference among the MSL systems became clearer during period 3, with MSL 1 and 4 showing significantly better results over the other two systems in decolorization (36.5–40.1% *vs* 25.5–26.4%) and COD removal efficiency (26.6–39.3% *vs* 14.6–15.6%), which was possibly because of the different composition of materials in the SMB and an aeration application effect during periods 1–3.

During period 4, the decolorization rates of MSL 1–3 increased by 3.0% (MSL 1) to 12.1% (MSL 3) and showed significantly higher removal efficiency than MSL 4. This was probably caused by the increase in aeration intensity in these three systems. However, the increase in decolorization rates (3.0–12.1%) was much smaller compared with the increase in COD removal rates (23.0–43.3%) for MSL 1–3. By the end of June 2006, COD removal rates for all the systems increased sharply to more than 60%, which might have been because of a temperature increase (approximately 30°C) at this time (Table 2). According to Navia *et al.* (2005), colored substances are poorly desorbed even at

**Table 3** Effect of material composition and aeration on mean decolorization rates and mean chemical oxygen demand (COD) removal rates in the four multi-soil-layering (MSL) systems

Period	<i>n</i>	MSL 1	MSL 2	MSL 3	MSL 4
Mean decolorization rate					
1	6	60.7% a	61.6% a	67.1% a	67.0% a
2	9	49.2% b	49.2% b	39.8% a	48.8% b
3	8	36.5% b	26.4% a	25.5% a	40.1% b
4	7	39.5% b	31.0% b	37.6% b	26.8% a
5	4	44.4% a	50.0% a	44.4% a	46.6% a
Mean COD removal rate					
1	6	50.2% a	48.8% a	52.0% a	58.0% a
2	9	39.7% b	39.1% b	29.4% a	43.3% b
3	8	26.6% ab	14.6% a	15.6% a	39.3% b
4	7	49.6% b	43.1% b	58.9% c	36.3% a
5	4	72.0% a	69.5% a	77.1% b	75.6% b

Means with the same letter in the same line within the same period are not significantly different using Tukey's honestly significant difference test ( $P < 0.05$ ).

temperatures up to 60°C, thus, decolorization rates were hardly influenced. The COD removal rates, however, could be greatly influenced by temperature change. Tahir *et al.* (1997) also indicated that in Hirakata, Japan, when the temperature was below 15°C, the efficiency of a MSL system for COD removal decreased by approximately 10% compared with when the temperature was at 18°C, while BOD and suspended solid (SS) removal rates were not affected by fluctuations in seasonal temperature.

Interruption of the operation for 1 month recovered the decolorization rates of MSL systems to over 50% and COD removal rates to over 80% at the beginning of period 5. The efficiency of the systems then decreased to the level before recovery. Navia *et al.* (2005) reported that 31% of the original capacity of Andisol was re-establishment with a reactivation process washing with 0.01 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub> solution in the case of color adsorption. Although it is difficult to compare this result with our study because of the different wastewater and regeneration processes used, it appears that the used soil demonstrated an important bioremediation capacity for adsorbed color, which could contribute to the repetitive application of the used soil for color removal. The lifetime of MSL systems to treat COD has also been reported to be semi-permanent if the accumulated organic matter could be decomposed by stopping the operation over a period of 1–2 months (Wakatsuki *et al.* 1999).

#### Effect of aeration position and intensity on decolorization and COD removal efficiency by MSL systems

As shown in Fig. 1, the positions of the aeration pipes in MSL 4 were different from those in the other three MSL systems. Aeration pipes in MSL 4 were set in the SMB layers, while those in MSL 1–3 were set in the water permeable layers (PL). During period 1, aeration was applied to all systems at a rate of 1,000 L min<sup>-1</sup> per system. Comparing MSL 3 and MSL 4, which had the same structure and material compositions, the difference in aeration position did not result in any significant difference in decolorization and COD removal efficiency. This meant that the aeration position did not have any significant influence on colored substances and COD removal under this treatment condition.

During periods 2 and 3, the aeration intensity for MSL 4 was increased to 2,000 L min<sup>-1</sup> per system, while the intensity in MSL 1–3 was still kept at a rate of 1,000 L min<sup>-1</sup> per system to investigate the aeration intensity effect on performance of MSL systems. MSL 3 showed significantly lower mean decolorization (39.8–25.5% *vs* 48.8–40.1%) and COD (29.4–15.6% *vs* 43.3–39.3%) removal rates than MSL 4 (Table 3), which showed that the aeration rate of 2,000 L min<sup>-1</sup> per system was more effective for colored substances and COD removal under

such treatment conditions. The effect of intensified aeration on the removal efficiency was reconfirmed during period 4. When the aeration amount of MSL 1–3 was increased to 2,000 L min<sup>-1</sup> per system, it resulted in an increase in both decolorization and COD removal efficiency of the systems compared with that in period 3. Effective aeration could enhance COD removal efficiency of MSL systems by enhancing microbial activity in decomposing organic matter, particularly when the systems become ineffective after a long operation time (Boonsook *et al.* 2003; Sato *et al.* 2005). In the case of colored substance removal, aside from microbial decompositions, aeration might have enhanced other physical and chemical processes by, for example, decreasing the negative charge of the adsorbents to increase the preferential adsorption of polar organic solutes (Ahmedna *et al.* 2000).

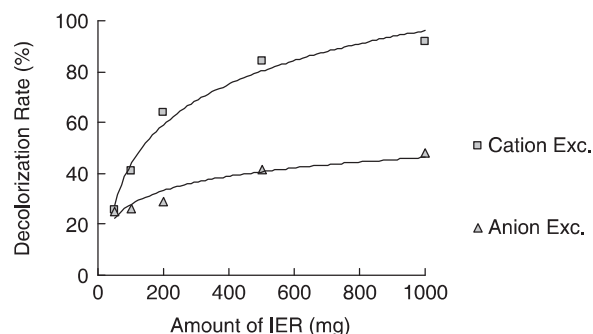
#### Effect of material composition in SMB on decolorization and COD removal efficiency by MSL systems

The material composition in the SMB of MSL 1–3 were different (Table 2). With time, significant differences appeared among the three systems, especially during period 3 (Table 3). MSL 1 showed significantly higher efficiency than MSL 2 in colored substances (36.5% *vs* 26.4%) and COD (26.6% *vs* 14.6%) removal. Aside from Andisol and AC, the SMB of MSL 1 also contained sawdust and iron. Sawdust has been reported to be an effective adsorbent because it contains a high concentration of cellulose, which irreversibly adsorbs colored substances through charge attraction and ion exchange process (Mckay *et al.* 1987). Moreover, it could also function as a carbon source for microorganisms as biological decomposition became more important with time through the accumulation of various organisms (Sato *et al.* 2005).

Iron added in MSL 1 was gradually oxidized into ferrous or ferric iron with time. The formation of cationic ions might contribute to the enhanced removal efficiency by precipitation with humic or fulvic acids. Mori and Sakimoto (1999) also reported that the addition of ferric chloride to wastewater resulted in not only the efficient removal of color, but also simultaneous reduction of COD by forming coagulation in wastewater. Colored substances in the livestock wastewater were mainly composed of negatively charged humic substances (Li *et al.* 2002). Cations formed in the SMB could precipitate more anionic colored substances from the wastewater.

#### Colored substance removal by ion exchange process and molecule size distribution analysis

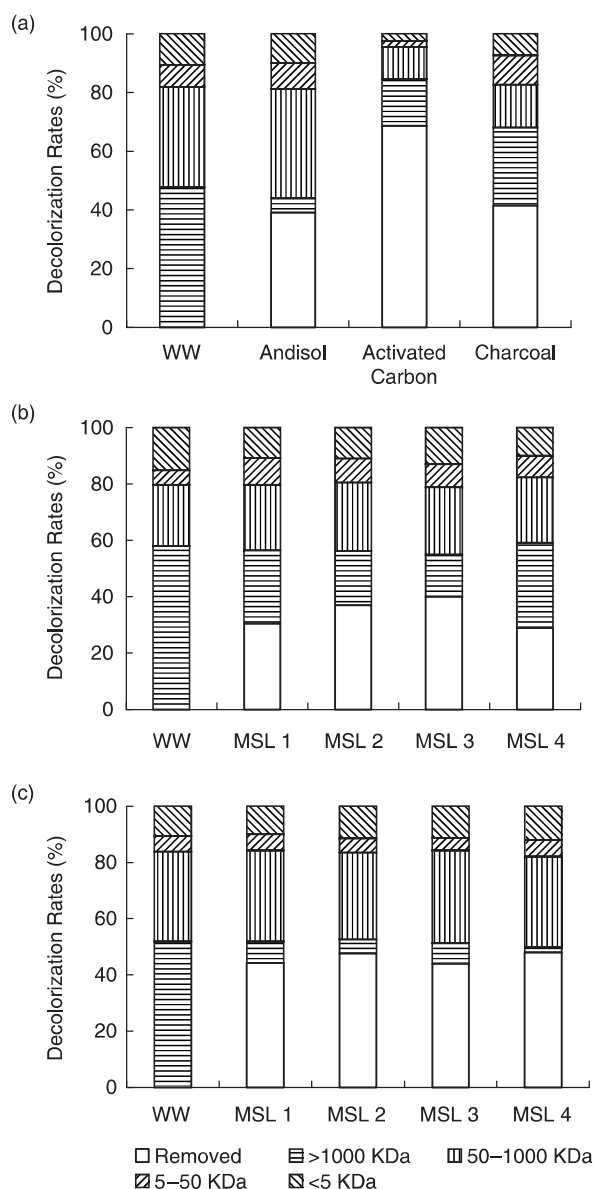
To further confirm the function of the ion exchange process, an experiment was conducted using cation and



**Figure 3** Decolorization rates by ion exchange resins (IER) at different amounts for 10-fold diluted livestock wastewater. Exc., exchange.

anion exchange resins as adsorbents. Fig. 3 shows the decolorization rates of the livestock wastewater by cation and anion exchange resins. At an amount of 50 mg, the ion exchange resins had a decolorization rate of approximately 25% and there was basically no difference between the cation and anion exchange resins. With the increase in resin amounts, decolorization rates by cation exchange resin increased sharply and reached 90% at an amount of 1,000 mg, while that of anion exchange resin was only approximately 50% at the same amount. It was expected that aside from the ion exchange process, direct adsorption onto the resins might be a possible mechanism for colored substance removal. In the present study, it was difficult to separate these two processes; however, the large difference in the decolorization rates by cation and anion exchange resins shows that aside from direct adsorption, cation exchange was the main process for colored substance removal.

The molecular size distribution analysis showed that 80% of the colored substances in the wastewater were composed of molecules > 50 kDa (Fig. 4a). Andisol and charcoal could adsorb half of the color molecules > 50 kDa, but could not adsorb those smaller than 50 kDa. This was in agreement with the report by Navia *et al.* (2005), which showed that a molecular size fraction > 30 kDa was the main fraction adsorbed onto the acidified Andisol. Molecules < 5 kDa were less adsorbed and, therefore, remained in the output effluent. The main decolorization mechanism of Andisol seems to be due to the adsorption onto organic matter, which has a great affinity with pollutants such as humic and fulvic acids. Furthermore, its clay-reactive sites, such as the Al and Fe hydroxide groups, can also be involved in fixing organic pollutants onto the soil matrix (Mora and Canales 1995). The AC showed a high decolorization rate of 68.7% as a result of its capacity to remove colored substances from small (< 5 kDa) to large (> 1,000 kDa)



**Figure 4** Molecule size distribution of colored substances in the water (a) before and after treatment with Andisol, activated carbon and charcoal with 10-fold diluted livestock wastewater, (b) before and after treatment in multi-soil-layering (MSL) systems after 6 months of operation and (c) before and after treatment in multi-soil-layering (MSL) systems after 1 month recovery. WW, waste water.

molecules. The adsorption capacity of AC was determined by its surface area, pore size distribution, and polarity of the surface (Ahmedna *et al.* 2000). Aside from its large surface area, the macropores of AC could serve as avenues for the rapid diffusion of color molecules to the smaller pores where they can be absorbed, while the

surface charge of AC was important in affecting preferential adsorption to polar organic solutes.

The molecular size distribution in the treated water in the MSL systems at the end of period 3 and beginning of period 5 showed that the main portion adsorbed and recovered was colored substances with molecules > 1,000 kDa (Figure 4b,c). As colored substances were mainly composed of non-degradable organic matter, the main decolorization mechanism at the beginning was considered to be a physicochemical reaction, such as filtration or adsorption. Because of the large specific surface area and developed pore systems of the adsorbents in MSL systems, color molecules could be easily trapped by the adsorbents in MSL systems and allowed a longer period for desorption. With time, the pores of adsorbents were fixed and filled with adsorbed molecules that were difficult to be desorbed over a short time. At this stage, the main decolorization mechanism was assumed to be processes such as ion exchange, precipitation, and reactions with soil organic matter for large molecules (> 1,000 kDa). When the pores of adsorbents were filled with pollutants, the smaller molecules could not be adsorbed and remained in the treated water. During the recovery process, large molecules (> 1,000 kDa) were assumed to be decomposed or degraded into smaller ones because the treated water of MSL 1–3 contained smaller molecules (< 1,000 kDa) than those in the wastewater. Because the recovered decolorization sites were probably on the surface of the adsorbents, when it became saturated with adsorbed colored molecules that were difficult to be decomposed or desorbed within a short time, the decolorization efficiency decreased quickly during period 5. Further study is still necessary to determine how to keep and recover the adsorption capacity of the systems for small colored substances to maintain its sustainable decolorization efficiency.

### Conclusions

The MSL systems showed quite stable and efficient treatment of colored substances and COD in the livestock wastewater. Treatment efficiency was influenced to a certain degree by MSL compositions and treatment conditions. The addition of sawdust and iron improved COD and colored substance removal by possibly enhancing biological decomposition and physio-chemical adsorption. The COD removal rate could be greatly improved through intensified aeration, higher temperature as well as interruption of the MSL operation for a period of time. For colored substance removal, the effect of temperature was not clear in the present study, while intensified aeration and a recovery period did result in higher removal efficiency.

During practical application of MSL systems, to obtain decolorized treated water as described in this study, it

needs 0.164 m<sup>2</sup> for an adult cow and 0.031 m<sup>2</sup> for an adult pig, if we assume that the average wastewater produced by a cow is 41 L day<sup>-1</sup> and that by a pig is 7.7 L day<sup>-1</sup> (Japan Agriculture, Forestry and Fishery Culture Association 2004b) when applied for secondary treated livestock wastewater. However, to control COD and coloration degree within standard or acceptable levels, further study is necessary on MSL systems as well as MSL in combination with other technologies such as ultraviolet or ozonation.

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